RECYCLING OF AIR POLLUTION CONTROL RESIDUES INTO ENGINEERED LIGHTWEIGHT AGGREGATES

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Abstract

The thermal treatment of Municipal Solid Waste (MSW) is continuously increasing in the UK as a sustainable practice to reduce the amount of waste that is sent to landfills and to recover energy in Energy from Waste (EfW) facilities. However, an issue associated with this practice is the volatilisation of hazardous compounds contained in the waste. These contaminants are removed from the gaseous emissions by air pollution control units before the gases are released into the environment. The solid wastes generated during this cleaning process are known as air pollution control residues (APCr) and are classified as hazardous because of their environmental impact associated with their chemical composition and leaching properties. APCr treatment before landfilling can be highly expensive, and therefore, there is a need for recycling these waste materials. The aim of this research was to investigate the viability of incorporating APCr into the manufacture of lightweight aggregates (LWA).

In the first stage, APCr samples collected from different EfW facilities across the UK were characterised for elemental composition, mineralogy and leaching properties. In the second stage, a mix composed of all samples of APCr previously characterised was prepared for the evaluation of APCr treatment. Washing with water and accelerated carbonation were optimised to reduce the leaching properties of APCr. The third stage comprised the manufacture of artificial LWA, through hot bonding and cold bonding processes at a laboratory scale. The physical and technological characteristics of the manufactured LWA were assessed for the fulfilment of the corresponding standards for LWA as well as compared with those of the commercial products Lytag[®] and Carbon8[®]. It was demonstrated that the hot-bonded LWA exhibited better technological characteristics and improved leaching properties compared with the cold-bonded LWA.

In the final stage, hot-bonded and cold-bonded LWA were incorporated into lightweight concrete (LWAC) to evaluate their performance. The LWAC made of hot-bonded LWA containing 25% washed APCr, was strong enough for applications in structural concrete exposed to different environmental conditions. Also, this LWAC formulation showed a reduction in the carbon emissions associated with its production, which represents an important environmental benefit. A future scale-up of LWA manufacture from 25% of washed APCr can significantly reduce the amount of APCr that is currently sent to landfills and increase the recycling rates of this waste material.

Declaration of originality

I hereby declare that the work presented in this PhD thesis is my own original work, and the information derived from other research works have been appropriately cited.

Ximena Chamorro Bolaños

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List of Abbreviations

APCr	Air Pollution Control Residues
BA	Bottom Ash
BI	Bloating index
BSI	Back Scattered Image
CE	Circular Economy
CFP	Carbon Footprint of a Product
CIF	Crystallographic Information Files
CIWM	Chartered Institution of Waste Management
COD	Crystallography Open Database
DEFRA	Department for Environment, Food and Rural Affairs
DOC	Dissolved Organic Carbon
EDS	Energy Dispersive X-Ray Spectroscopy
EfW	Energy from Waste
EWC	European Waste Catalogue
ESP	Electrostatic precipitator
FA	Fly Ash
HPLC	High Performance Liquid Chromatography
IAWG	International Ash Working Group
ICP/OES	Inductively Coupled Plasma/Optical Emission Spectroscopy
IED	Incineration Emissions Directive
LOI	Loss on ignition
LWA	Lightweight Aggregates
LWAC	Lightweight Aggregate Concrete
MSW	Municipal Solid Waste
Mt	Million tonnes
OPC	Ordinary Portland Cement
PDXL	Integrated X-Ray Powder Diffraction Software
POP	Persistent Organic Pollutants
rpm	Revolutions per minute
S/S	Stabilisation/Solidification
SEM	Scanning Electron Microscopy
SEI	Secondary Electron Image
TOC	Total Organic Carbon

- TDS Total Dissolved Solids
- WAC Waste Acceptance Criteria
- WfH Waste from Households
- XRD X-Ray Diffraction
- wt.% weight percentage

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1 Chapter 1 Introduction

1.1 Background

Municipal Solid Waste (MSW) comprises domestic, light industrial, commercial and institutional waste that is collected by local authorities or contracted companies (DEFRA, 2013). A report published by the World Bank (Kaza, et al., 2018), estimated that the annual production of MSW worldwide is about 2.01 billion metric tonnes and from that amount, 37% is landfilled, 33% is disposed of in open dumps, 19% is recovered by recycling and composting and 11% is thermally treated for final disposal with or without energy recovery. According to that report, management of MSW is highly expensive, accounting for about 20% of the municipal budget for lower-income countries and for about 4% in high-income countries. As a consequence, suitable disposal and treatment of waste is almost exclusively practised in wealthier countries.

Waste treatment by incineration is the process used to combust waste and recover energy in Energy from Waste (EfW) facilities. Waste incineration technology, which is associated with higher operation costs, is mainly used in high-capacity, high-income and land-constrained countries, where around 22% of their MSW is incinerated (Kaza, et al., 2018). It is calculated that per each 1.3 billion tonnes of MSW produced around the world per year, 130 million tonnes are thermally treated (Joseph, et al., 2018). From the latest available data¹, the total amount of MSW produced in the UK can be estimated at 30.8 Mt in 2018. Considering that the total EfW inputs were 11.5 Mt by 2018, the fraction of MSW treated by incineration in the UK was about 37.4%. The last report of waste management for England (DEFRA, 2023a) indicates that during the financial year 2021/2022, the local authorities managed 26.1 Mt of waste, and from that amount, 2.1 Mt of waste (8.1%) were sent to landfill, 10.8 Mt (41.4%) were recycled, and 12.4 Mt (47.5%) were incinerated. According to the same report, the regions of London, North East, South East, West Midlands and Yorkshire and the Humber sent greater amounts of MSW for incineration than for recycling, and London sent the largest proportion (64.4%) of MSW for incineration in 2021/2022 accounting for 2.3 Mt. it should be noted that the

¹ The production of municipal waste in the UK was 463 kg per capita in 2018 (Eurostat, 2023) and the UK population for the same year was 66,435,600 (Office for National Statistics, 2022).

MSW incinerators currently operating in the UK recover energy either in the form of electricity and/or heat (Tolvik Consulting Ltd, 2023).

The use of waste incineration has continuously increased over recent years in the UK, according to official data. By 2014, about 6.7 Mt of waste were incinerated in UK EfW facilities (Tolvik Consulting Ltd, 2015); then, this amount increased to 15.3 Mt by 2022 (Tolvik Consulting Ltd, 2023). Figure 1.1 compares the amounts of MSW treated in EfW plants in the UK in the last 9 years (Tolvik Consulting Ltd, 2023) with the amounts of Waste from Households (WfH) generated and recycled between 2015 and 2021 (DEFRA, 2023b). WfH is a subset of MSW which comprises domestic waste, civic amenity sites, bulky waste and other household waste. The comparison is done against WfH because this is the measure agreed upon by the UK countries to report waste recycling. It is observed that the generated WfH varied around 27 Mt while the recycled fraction of this waste has remained almost constant around 12 Mt. That is, the average recycling rate of WfH in the UK between 2015 and 2021 was 25%. The forecast trendlines show that the amounts of both generated and recycled WfH will remain around the same average values at least for the next few years. In comparison, the EfW inputs have increased between 2014 and 2022. These inputs also comprise the MSW fraction that is not WfH, such as light industrial and commercial waste. A further extrapolation suggests that by the end of 2026, the amount of waste treated in EfW sites will have exceeded 20 Mt.

It is important to note that the information presented in Figure 1.1 for recycling of WfH includes metals recovered and recycled after incineration (DEFRA, 2023b). This fraction is known as Incinerator Bottom Ash metal (IBAm) and its inclusion increased the recycling rates of WfH. For 2021, the IBAm raised the recycling of WfH by about 0.9 percentage units, which counts for 243 kt.



Figure 1.1 Waste from households generated and recycled in the UK from 2015 to 2021 (DEFRA, 2023b) and tonnage of waste incinerated in UK EfW sites from 2014 to 2022 (Tolvik Consulting Ltd, 2023). EfW inputs comprise WfH and other MSW such as light industrial and commercial waste. The dot lines show the forecast tendencies for each series.

If the tendencies described above continue, it can be predicted that thermal treatment of MSW will soon account for over 50% of all waste disposal in the UK. In addition, taxes recently introduced on landfills have made incineration a more affordable option, leading to an increase in the number of incinerators and EfW plants (Bawden, 2019). By the end of 2022, there were 57 EfW facilities in operation and 3 in late-stage commissioning across the UK (most of them located in England), accounting for a total capacity of 17.52 Mt of waste per year (Tolvik Consulting Ltd, 2023).

Waste incineration is considered a successful practice that helps to protect the environment and to reduce the greenhouse gas emissions from landfill sites (Chartered Institution of Wastes Management, 2022). In addition, from the three main waste management options (landfilling, recycling and incineration), the technologies of waste incineration bring the alternative use of waste as an energy and heat source in EfW facilities, resulting in a more sustainable option to reduce the dependence on other energy sources, such as fossil fuels (Chartered Institution of Wastes Management, 2022), as well as to reduce the amount of waste that is sent to landfill (DEFRA, 2013; Quina, et al.,

2014a). In fact, incineration of MSW can reduce the waste by 90% in volume (Hjelmar, 1996) and up to 80% in mass (Bertolini, et al., 2004). Despite the benefits of MSW incineration, there is an issue associated with the volatilisation of hazardous compounds contained in the treated waste. These contaminants are removed from the gaseous emissions by Air Pollution Control (APC) units. The solid wastes generated during this cleaning process are known as APC residues (APCr).

1.1.1 APCr production

During waste incineration, hazardous materials contained in solid waste are transferred to the flue gases. The gas phase is then, cleaned to prevent the release of pollutants into the environment. Currently, different technologies such as dry, semi-dry and wet scrubbers, fabric filters and electrostatic precipitators or a combination of them, are used to remove the particulate material from the gaseous emissions. Cleaning of gaseous emissions from EfW facilities generates two different types of solid residues, Fly Ashes (FA) and APCr. FA are fine particles originated from the waste combustion process and should be recovered separately from the gas stream, although they are sometimes mixed with APCr. APCr comprise the solid wastes generated as by-products during the cleaning process of the gaseous emissions and include all the particulate material collected after the injection of any reagent and before discharging gases to stack (Sawell, et al., 1995; Chandler, et al., 1997).

Usually, the production of APCr is from 2% to 6% of the total inputs of a waste incineration facility (DEFRA, 2013). By the end of 2021, there were 53 fully operational EfW plants in the UK and it was reported that 6 out of them generated APCr at proportions over 5% of their total inputs and only in 2 EfW sites this proportion was less than 2% (Tolvik Consulting Ltd, 2022). By 2022, with 57 EfW sites in full operation, the annual production of APCr in the UK was estimated as 3.0% of the total waste inputs (15.32 Mt) (Tolvik Consulting Ltd, 2023), which is about 459,600 tonnes of APCr. Figure 1.2 shows how the amounts of APCr generated and recycled in the UK over the last 9 years have increased, according to annual figures reported by Tolvik Consulting Ltd. This increase is due to a greater proportion of waste currently incinerated to produce energy in new emerging EfW projects and the enlargement of existing EfW capacities. However, the recycling of APCr for 2019, 2020 and 2021 shows similar values. The recycled proportion of APCr tends to be constant at around 35%. It can be said that the increasing

amounts of APCr generated every year in the UK are becoming more difficult to be managed by the existing recycling capacities.



Figure 1.2. APCr production and recycling in UK from 2014 to 2022. Based on data provided by the UK Energy from Waste Statistics annual reports (Tolvik Consulting Ltd, 2015 – 2023). Data about recycling of APCr for years 2014, 2018 and 2022 are not available.

1.1.2 APCr and the Circular Economy

The traditional industrial model follows a linear system that consists in taking resources, making and using products, and disposal of products at the end of their life. This linear economy is moving forward to a Circular Economy (CE). This system is an approach that re-designs the way the future economy works, based on three principles: design products and services out of waste and pollution; recirculate products, components and materials in use; and regenerate natural systems (Ellen McArthur Foundation, 2017). Under the principles of CE, manufactured products must be used for as long as possible extracting their maximum benefit, and then, at the end of their service life, they can be recycled to manufacture new products and materials (De Schoenmakere, et al., 2018; The Waste and Resources Action Programme, 2019). Implementing a CE policy brings environmental and social benefits. The environmental effect is in the reduction of the negative impact of a linear economy by reducing the amount of waste released to the generation of business and economic opportunities and competitiveness (The Waste and Resources

Action Programme, 2018; Ellen McArthur Foundation, 2017). The recycling of APCr into LWA can be strategically designed, considering the principles and benefits of the CE. The following paragraphs provide a further description of the CE principles (Ellen McArthur Foundation, 2017; UK Green Building Council, 2023) and how they are applied to the context of this research.

CE Principle 1: Eliminate waste and pollution. Specific actions to minimise the impact and waste include using low-impact new materials, using recycled or secondary materials and designing products out of waste. This principle is addressed by:

- a. Ensuring the end of waste: APCr is considered a hazardous waste due to its contents of leachable heavy metals and salts. The treatment of APCr is aimed at reducing or eliminating the leaching properties of this material. Then, the treated waste can be incorporated into engineered manufacturing processes to produce LWA. This way, APCr ceases to be classified as waste as LWA is produced with minimum requirements in their technological and environmental properties.
- b. Considering the generation of by-products, contaminated effluents and emissions as integral part of the developed manufacturing process. If these non-desired outcomes cannot be avoided, they must be reduced or addressed by treatment for disposal or recycling.
 - By-products: no by-product is expected to be generated during the proposed manufacturing processes of LWA.
 - Contaminated effluent (wastewater): its chemical composition must be determined for future considerations regarding its treatment and disposal. The treatment should enable the further recycling of water or recovery of materials, for example metals. This will depend on the chemical characteristics of the effluent and on the needs of the related industrial processes. If recovery and recycling are not feasible, the treated effluent must achieve the minimum requirements to be legally disposed of as treated wastewater into natural bodies of surface water or saline water.
 - Emissions: CO₂ emissions should be calculated along the different stages of the APCr treatment and LWA manufacturing processes, including material treatment, drying, pelletising and firing. These emissions are part of the embodied carbon assessment for the manufacture of LWAC, added to other emission sources,

including the transport of raw materials and production of other components of the concrete mix.

CE Principle 2: Circulate products and materials at their highest value. At the end of their life, products and materials can be used as components or raw materials of new products to remain as part of the materials closed loop. In the technical cycle, the products and materials are kept in circulation by reusing, repairing, remanufacturing and as a least option recycling. This research addresses this principle by proposing treatment options for APCr to enable it for further recycling, which means a revaluation of the waste material. Thus, the treated APCr becomes a raw material for the manufacture of artificial LWA.

CE Principle 3: Regenerate natural systems. This refers to regenerating nature by supporting natural processes and leaving more space for nature to thrive. It is addressed by:

- a. Replacing part of the clay in the LWA manufacture by washing APCr and using the washed APCr as raw material helps to reduce the depletion of the land that is used for natural clay sourcing.
- b. Treating APCr by carbonation, leads to the absorption of CO₂, reducing the carbon emissions, and as a result it would help to tackle climate change.

1.2 Aim and Objectives

Given the increasing generation of APCr from MSW incineration in the UK EfW sites, there is a need for research on the recycling options of this waste material. The aim of this project was to investigate the viability of incorporating APCr into the manufacture of LWA.

The following objectives were achieved throughout this research:

1- Characterisation of APCr coming from various EfW facilities in the UK.

APCr samples from different EfW sites throughout the UK were analysed for elemental composition, mineralogy, microstructure and leaching properties. The analytical

techniques used for these tests were ICP/OES, XRD and SEM/EDS. These analyses allowed to determine the extent of variability of APCr chemical compositions and their relationship with the technologies used in the thermal treatment of solid waste.

2- Assessment and optimisation of different treatments of APCr to enable use in the manufacture of LWA.

Two different treatments were tested for APCr: water washing and carbonation, in order to reduce or stabilise the content of heavy metals and soluble salts and to improve their leaching properties. The treatment time was optimised in both treatment options, the liquid-to-solid ratio (L/S) was optimised for the washing treatment, while the moisture content was optimised for the carbonation treatment. The APCr treated under the optimised conditions were combined with clay and fired (hot bonding) or lime as a binder (cold bonding) at different proportions to produce LWA.

3- Evaluation of the variables that affect the production of LWA from treated APCr.

The effects of production parameters such as firing temperature and APCr content on the properties of LWA were investigated. These conditions were assessed by making LWA at different proportions between 5 and 30% of APCr, and firing the green pellets at different temperatures in the range between 1,120 and 1,180 °C. For the manufacture of LWA through cold bonding, the assessed variable was the addition of lime as a binder from 20 to 30%.

4- Assessment of the performance of LWA manufactured from treated APCr, in terms of their standard mechanical and environmental properties.

The LWA made from untreated or treated APCr were analysed for their mechanical properties (bloating index (BI), particle density, water absorption, bulk and specific gravity and crushing strength), and environmental properties (mineral and elemental composition and leaching behaviour). In addition, the LWA from treated APCr were incorporated into concrete cubes, which were tested for density and 28-day compressive strength. The obtained results were compared with the reported values for commercial LWA (Lytag[®] and Carbon8[®]) and with the requirement of the applicable British Standards.

1.3 Contribution to knowledge

This thesis provides a novel contribution to the field of construction materials under the principles of sustainable consumption and production, which is the sustainable development goal No. 12 of the United Nations (United Nations, 2022). The original outcomes of this research can be summarised as follows:

- An in-depth knowledge of the chemical composition, microstructure and environmental properties of APCr coming from different EfW facilities throughout the UK.
- Implementation of a complete sample preparation process for analysis of APCr by SEM/EDS, which has not been previously described in the literature.
- Optimisation of treatment options for APCr, aimed at the reduction of their leaching properties.
- Delivering a viable recycling option for APCr in the manufacture of LWA.
- Improvement on the physical and technological properties of LWA by the incorporation of treated APCr.
- Incorporation of APCr-based LWA into the production of lightweight aggregate concrete (LWAC) and estimation of the associated carbon emissions.

1.4 Experimental Approach

In order to achieve the objectives previously described in section 1.2, a systematic literature review, laboratory work and statistical analysis were conducted. Table 1.1 shows the experimental approach of this research according to each objective.

Table 1.1. Experimental tasks according to each research objective.

APCr characterisation

- Sampling
- Dry matter content
- Loss on ignition
- Total contents of metals
- pH
- Leaching tests
- Mineralogy
- Contents of amorphous material
- Morphology and particle size
- Elemental composition

APCr treatment

- Water washing
- Optimisation of washing: liquid/solid ratio and washing time
- Accelerated carbonation
- Optimisation of carbonation: moisture and carbonation time
- Characterisation of treated APCr:
 - Leaching tests
 - Total contents of metals
 - Mineralogy
 - Contents of amorphous material
 - Changes in microstructure
 - Elemental composition

LWA production

- Hot bonding:
 - Formulation: clay, treated or untreated APCr and water
 - Firing temperature range (relationship with particle density and water absorption capacity)
- Cold bonding:
 - Formulation: lime, treated APCr and water
 - Curing time (development of strength and water resistance)
- Comparison of the LWA made from treated APCr with LWA made from untreated APCr or without APCr

LWA performance

- Physical and chemical characterization of LWA:
 - Particle density
 - Water absorption capacity
 - Bulk density
 - Crushing strength
 - Porosity (pore structure analysis)
 - Bloating index
 - Mineralogy
 - Elemental composition
 - Contents of amorphous material
 - Morphology and particle size
 - Leaching tests
- LWA performance in concrete: density and 28-days compressive strength
- Comparison between the LWA made of treated APCr and commercial LWA

2 Chapter 2 APCr and its potential use in the manufacture of LWA

2.1 Waste incineration

Waste treatment by means of incineration involves the combustion of unprepared (raw or residual) MSW, which in turn, offers the further option to recover energy by utilising the calorific value of the waste to produce heat and/or power (DEFRA, 2013). Waste incineration plants operate according to the minimum requirements of combustion temperature of 850°C and time of 2 seconds, set by the Industrial Emissions Directive (IED) to ensure that the waste is oxidised into carbon dioxide and water (The European Parliament and the Council, 2011). The non-combustible materials that are present in the waste, such as metals and glass, remain as a solid residue, known as bottom ash (BA), which also contains a small amount of residual carbon (DEFRA, 2013).

The EfW plant design and configuration may vary depending on the different technology providers, but in general, these kind of facilities comprise the following operating units: waste reception and handling, combustion chamber, energy recovery plant, emissions clean-up units for combustion gases, bottom ash handling and APCr handing (DEFRA, 2013).

Figure 2.1 schematically shows the process of waste incineration in an EfW plant. The solid waste is discharged into a storage bunker, where overhead grab cranes load the waste into a feeding hopper. From the hopper, the waste is fed onto the main combustion grate where it is burned. The furnace walls surrounding the grate have a series of pipes containing water that is continuously flowing. The heat produced during the waste incineration is used to heat up that water and turn it into steam which goes to power generation (Deltaway, 2018; Hockenos, 2021; Youcai, 2017). The thermal treatment volatilises the hazardous material contained in the solid waste, producing gaseous emissions that must be cleaned before being released into the atmosphere via the stack (Denison & Ruston, 1990). In this regard, the incineration temperature is a crucial parameter that determines the distribution of volatile elements among the different size fractions of the solid outputs (Joseph, et al., 2018). The pollutants contained in the flue gases and associated with potential damage to the environment and human health are acid gases (hydrochloric acid HCl, sulphur dioxide SO₂, fluorhydric acid HF, nitrogen monoxide NO and nitrogen dioxide NO₂), carbon dioxide CO₂, heavy metals, particulate

material and organic pollutants (dioxins and furans) (Chartered Institution of Wastes Management, 2022; Bodénan & Deniard, 2003). To perform the cleaning of flue gases, these are passed through a scrubber system, where alkaline additives, such as lime, sodium hydroxide and sodium bicarbonate, are injected. Then, these gases pass through the air pollution control system, which consists of a series of filters, where solid residues are captured and collected. These residues comprise fly ashes and APCr.



Figure 2.1 Process of waste incineration in an energy from waste plant. Modified from https://deltawayenergy.com (Accessed on 8 January 2020).

APCr can be of different varieties depending on the type of incineration and the type of flue gas cleaning technologies used in the EfW site. Therefore, a description of these technologies is provided in the following sections.

2.1.1 Incineration technologies

There are four incineration technologies that are currently used to treat MSW:

- Moving grates
- Fixed grates
- Fluidised bed
- Rotary kiln

These technologies are described in the following sections.

2.1.1.1 Grate technology using moving grates

The moving grate furnace is the most commonly used combustion system for MSW processing in the UK (DEFRA, 2013). Figure 2.2 shows a diagram of a moving grate incinerator. In this system, the waste is slowly driven through the combustion chamber by a mechanically operated series of inclined moving grate bars (Chartered Institution of Wastes Management, 2022). The incineration system is designed to enable complete combustion while the waste passes through the furnace; that is, as the waste enters the furnace, the BA is discharged to the other end of the grate (DEFRA, 2013). In addition, the combustion chamber may be equipped with auxiliary burners that help to raise the temperature in the main chamber before starting the process from cold and during shutting down the plant, ignite the refuse at the start-up, and comply with the Incineration Emissions Directive (IED) requirements aimed at reducing the flue gas emissions (Wardell Armstrong, 2018)



Figure 2.2 Moving roller grate incinerator. Source: (Igniss Energy, 2022).

2.1.1.2 Grate technology using fixed grates

In fixed grates, the waste is gradually moved by a series of rams along the combustion unit. The process is usually done in three steps, as it can be seen in Figure 2.3. Firstly, drying the waste and initial combustion; secondly, further combustion; and finally, a post-combustion that fully burns the carbon out (DEFRA, 2013; Tokyo Environmental Public Service Corporation, 2012).



Figure 2.3. Fixed grates incinerator. Modified from (Tokyo Environmental Public Service Corporation, 2012).

2.1.1.3 Fluidised bed

This technique involves a pre-sorting step of the waste, to remove heavy and noncombustible material, such as metals, followed by a mechanical reduction of the particle size of the waste (DEFRA, 2013; Chartered Institution of Wastes Management, 2022). The resulting particle size is less than 150 mm (Chartered Institution of Wastes Management, 2022). Then, the waste is sent to the furnace, which consists of a vertical chamber containing a granular bubbling bed made of an inert material like coarse sand/silica that resists high temperatures. The bed is "fluidised" by air, which can be mixed with recycled flue gas, which is vertically blown through the material at a high flow rate. Thus, the waste material is mobilised by the effect of the fluidised bed particles (DEFRA, 2013).

Figure 2.4 shows the schemes of two types of fluidised-bed technologies: a bubbling bed and a circulating bed. These systems differ in the interaction between airflow and bed material. In the bubbling bed, the airflow mobilises the bed and provides good contact with the waste, but it is not high enough to promote large amounts of solids to be released from the combustion chamber. In the circulating bed, the airflow is higher and allows the solids particles to be removed by the flue gas (DEFRA, 2013).

The technology of bed combustion is thermally more efficient than the moving grate system, however, it produces a greater amount of fly ash (Chartered Institution of Wastes Management, 2022). This fact, added to the increase in energy consumption during the pre-treatment of the waste, before the incineration, may be the reason why the use of fluidised bed for MSW incineration is lower in the UK compared to the use of moving grates.



Figure 2.4. Scheme of a fluidised-bed incinerator. Bubbling bed (left). Circulating bed (right). Modified from (SSWM, 2020).

2.1.1.4 Rotary kiln

A rotary kiln can be either a complete or a partial rotation vessel. A diagram of this technology is shown in Figure 2.5. The incineration process occurs in two stages: first, the primary combustion that takes place in the rotary kiln; and then the incineration continuous in a secondary combustion chamber. The kiln is inclined downwards from the feed entry point. The rotation moves the waste through the kiln with a tumbling effect that exposes the waste to heat and oxygen (DEFRA, 2013).



Figure 2.5. Rotary kiln furnace. Source: Modified from (Focus Technology Co., Ltd, 2022).

2.1.2 Air pollution control technologies

Waste incineration plants include air pollution control (APC) systems aimed to clean the flue gases resulting from the waste incineration before releasing these gases into the atmosphere. The APC technologies consist of filtration systems with specific designs of bag filters, adsorption and absorption/neutralisation processes. There are three different types of flue gas cleaning technologies: dry, semi-dry and wet scrubber, as described in the following sections.
2.1.2.1 Dry scrubber

Alkaline chemicals, such as hydrated lime Ca(OH)₂, are injected into the flue gas in dry form to neutralise its acidic components. Other chemicals, such as sodium bicarbonate NaHCO₃ and quick lime CaO are used to remove acid gases by adsorption (LAB, 2016; Hitachi Zosen INOVA, 2021). This is usually done before removing the fly ash from the flue gas. Then, the fly ash, reaction products and unreacted additives are retained by fabric filters. Activated carbon may be injected for the removal of volatile organics (dioxins) and metals (Hitachi Zosen INOVA, 2021). Then, the carbon is removed together with the fly ash (DEFRA, 2013). Figure 2.6 shows the flow diagram of a dry system, which comprises three stages: firstly, a flue gas conditioning to ensure optimum temperature; secondly, the injection of dry lime for neutralisation of acidic gases, and removal of mercury, heavy metals and dioxins with activated carbon; and finally, a reactivation and recirculation of residues by means of a system ActiLABTM, which allows the recycling of reagents for the final removal of pollutants (LAB, 2016).



Figure 2.6. Scheme of a dry system for flue gas treatment (SecoLABTM). Source: (LAB, 2016).

2.1.2.2 Sem-dry scrubber

In semi-dry systems, hydrated lime or quick lime is injected for the removal of acidic pollutants (LAB, 2016; Hitachi Zosen INOVA, 2021). The operation of a semi-dry system is similar to a dry-system, but the alkaline additives are mixed with water and injected as a slurry. As a result, the semi-dry process residue has lower content of unreacted lime.

Semi-dry sorption processes are used based on the principle of the circulating fluidised bed for the removal of acid gases by adsorption with lime (Hitachi Zosen INOVA, 2021). Activated carbon or coke can be injected for the removal of volatile organic pollutants. Figure 2.7 shows the scheme of a semi-dry system, in which the initial step is the spraying of lime slurry; then, activated carbon or potassium hydroxide is added to remove mercury and dioxins/furans. The dust and solid salts resulting from the neutralisation reactions are retained by the downstream fabric filters (LAB, 2016).



Figure 2.7. Scheme of a semi-dry system for flue gas treatment SemisecoLABTM. Source: (LAB, 2016).

2.1.2.3 Wet scrubber

In wet systems, the neutralisation of acidic components of the flue gases is done after removing the fly ashes. Then, the flue gas is directed towards a multistage arrangement of scrubbers (wet scrubbers), where the pollutants are captured by contact between the flue gases and water. The wastewater produced from the scrubber is treated to produce sludge and gypsum (DEFRA, 2013). The wet scrubber system is considered the most effective method for the removal of acid gases; besides it produces the lowest emissions (Hitachi Zosen INOVA, 2021). Figure 2.8 shows the scheme of a wet system based on saturation temperatures in scrubbers and absorption of acid gases. This system uses as additives limestone (sedimentary rock, composed mainly of calcium carbonate CaCO₃, in its crystalline forms of calcite and aragonite) or lime slurry (suspension of calcium hydroxide Ca(OH)₂) or sodium hydroxide NaOH, which easily combine with flue gas condensation allowing additional heat recovery (LAB, 2016).



Figure 2.8. Scheme of a wet system for flue gas treatment GraniLABTM. Source: (LAB, 2016).

2.2 Characteristics and chemical composition of APCr

The properties of APCr are influenced by the properties of the incinerated waste, which in turn, are modified by changes related to consumption habits, recycling technologies, product requirements, among other factors (Ecke, 2003a). APCr may be in the forms of solid, liquid or sludge, depending on the scrubber technology used for air pollution control (dry, semi-dry or wet) (Astrup, 2008; Chandler, et al., 1997; Amutha Rani, et al., 2008; Sabbas, et al., 2003). The colour ranges from light to dark grey (Astrup, 2008; Quina, et al., 2008b; Chandler, et al., 1997; Bogush, et al., 2015), which depends on their chemical composition and the combustion technology used to treat the original waste (Chandler, et al., 1997). Usually, APCr look like a fine-grained powder, and different particle sizes have been reported, as shown in Table 2.1. The particles with size in the order of mm in APCr result from small pieces of partially combusted paper (Dimech, et al., 2008).

Table 2.1 Particle sizes of APCr according to different studies.

Reference	Country	Particle size
(Sabbas, et al., 2003)	Non specified	1 μm to 1 mm
(Quina, et al., 2008b)	Portugal	159 to 244 µm
(Fernández Bertos, et al., 2004b)	UK	Mostly less than 300 μm
(Dimech, et al., 2008)	UK	$2 \ \mu m$ to $2 \ mm$
(Bogush, et al., 2015)	UK	2 to 150 µm

APCr composition varies substantially depending on the original waste composition, incineration process parameters and the APC system (Amutha Rani, et al., 2008). The chemical composition of APCr also differs from one region to another, however it can be comparable at some extent (DEFRA, 2013). There is an organic fraction in APCr, quantified by the total organic content (TOC) that is usually lower than 10,000 mg/kg (Wiles, 1996). This fraction comprises small amounts of volatile organic substances, namely, polychlorinated dibenzo-p-dioxins (PCDD) and polychlorinated dibenzofurans (PCDF), which are produced during the waste incineration process (Wiles, 1996). The major fraction in APCr comprises inorganic substances, which are associated with the environmental impact of this waste, due to the high contents of toxic heavy metals, such as Hg, Pb, Cr, Cd and As, and soluble salts, including sulphates and chlorides, with high leaching rates (Quina, et al., 2008b).

Appendixes I and II present an extensive review of the chemical composition of MSWI APCr as reported by different papers since 1996 up to date in different countries. Some of the revised articles refer to APCr as fly ashes (Li, et al., 2004; Song, et al., 2004; Chen, et al., 2012; Colangelo, et al., 2012; Keppert, et al., 2015; Atanes, et al., 2019; Dontriros, et al., 2020; Cao, et al., 2021; Fan, et al., 2022; Han, et al., 2022), however they describe that the materials were sampled downstream the APC devices. In other papers, the authors specify that the APCr were mixed with fly ashes (Hjelmar, 1996; Mizutani, et al., 2000; Hyks, et al., 2009; Cappai, et al., 2012).

Appendix I shows the elemental composition of APCr according to 42 studies, of which 10 were conducted in the UK. According to this review, major elements are those elements whose concentrations exceed 1,000 mg/kg. Figure 2.9 shows the concentrations of major elements for the APCr in the UK. This group includes Al, Ca, F, Fe, Cl, Mg, K, Na, P, Pb, S, Si, Ti and Zn. Also, as it was stated by Hjelmar (1996) for MSWI residues and observed in Appendix I for APCr, many of the elements are present in the form of oxides, thus, oxygen is also a major element.



Figure 2.9 Major elements detected in APCr from MSWI in the UK by different studies. A: 50,000 - 650,000 mg/kg, B: 14,000 - 50,000 mg/kg, C: up to 14,000 mg/kg.

Elements that are present at concentrations between 100 mg/kg and 1,000 mg/kg are considered minor elements. Figure 2.10 Part A shows the concentrations of minor elements in APCr from the UK. This group includes Ba, Cd, Co, Cr, Cu, Mn, Sb, Sn and Sr. Trace elements are those with concentration lower than 100 mg/kg. Figure 2.10 Part B the concentrations of trace elements detected in APCr from the UK. The most common trace elements are As, Mo, Ni and Se. Some trace elements have been detected only in the APCr from the UK, especially Se which was reported by 5 out the 10 studies (Lee, et al., 1999; Sun, et al., 2008; Gunning, et al., 2011a; Bogush, et al., 2015; Bogush, et al., 2019). The origin of elements in MSWI residues is mainly household waste of small, sealed batteries containing Pb and Ni/Ca, preserved food containing Cu, Ca and As, as well as flame-proofed products containing Sb (Hong, et al., 2000). The release of Cd and Pb from APCr, has been the key issue leading to the classification and management of this material as hazardous waste (Wiles, 1996).





Figure 2.10 Minor and trace elements detected in APCr from MSWI in the UK by different studies. A: Minor elements, B: Trace elements.

Appendix II summarises the mineral composition of APCr according to 26 studies, of which six were conducted in the UK. Most of the elements that are present in APCr constitute mineral phases, such as oxides, chlorides, sulphates and carbonates. In this review, excepting the percentages shown by Keppert, et al. (2015), the other studies do not report the concentration of each mineral phase; some of them specify their levels as either major or minor. The main crystalline phases in APCr, usually reported as major, and the phases that have been found only in APCr from the UK are listed in Table 2.2.

Reference	Phase name	Formula
Main phases found in APCr		
	Anhydrite	CaSO ₄
	Calcite	CaCO ₃
Studies referenced in Appendix II	Calcium hydroxychloride	CaOHCl
- Mineral composition of	Halite	NaCl
APCr reported by previous	Portlandite	Ca(OH) ₂
	Quartz	SiO ₂
	Sylvite	KCl
Phases detected only in the UK		
(Fernández Bertos, et al., 2004c)	Sygenite	K ₂ Ca ₂ (SO ₄)H ₂ O
	Botallackite	Cu ₂ (OH) ₃ Cl
	Calcium sulphate hydrate	CaSO ₄ •H ₂ O
	Cerussite	PbCO ₃
	Copper nickel zinc oxide	$Cu_{1.02}ZnNi_{3.27}O_{5.29}$
	Fedotovite	$K_2Cu_3O(SO_4)_3$
(Pogush at al. 2015)	Gahnite	$ZnAl_2O_4$
(Dogush, et al., 2015)	Magnesium oxide hydroxide	$Mg_3O_2(OH)_2$
	Potassium aluminium silicate	$K_{1.25}Al_{1.25}Si_{0.75}O_4$
	Sodalite	$Ca_8Al_{12}O_{24}(MoO_4)_2$
	Tenorite	CuO
	Tobermorite	$Ca_5Si_6O_{16}(OH)_2{\bullet}4H_2O$
	Zincowoodwardite	$Al_{0.375}H_6O_{2.686}S_{0.188}Zn_{0.625}$
	Apatite	Ca10(PO4)6(OH,F,Cl)2
(Regustrated 2010)	Corundum	Al_2O_3
(Dogusii, ci al., 2017)	Melilite	Ca ₂ (Mg,Al)(Al,Si) ₂ O ₇
	Willemite	Zn_2SiO_4

Table 2.2 Main crystalline phases found in APCr.

Alkaline chemicals injected during the flue-gas cleaning, such as lime CaO, portlandite $Ca(OH)_2$, sodium bicarbonate NaHCO₃ and caustic soda NaOH, are responsible for the high contents of portlandite, calcite, and lime in APCr. Due to the presence of these compounds, APCr are characterised by alkaline pH which can reach values between 12.0 and 12.6 (Sabbas, et al., 2003; Quina, et al., 2008a). The pH associated to APCr coming from dry and semi-dry APC systems is usually higher than 12, whereas the pH values of APCr from wet scrubber systems vary around 10.5 (Wiles, 1996). Sulphates are formed by the neutralisation reactions between the mentioned alkaline substances and the acid gases. The presence of chlorides in APCr is due to the high contents of polyvinyl chloride $(C_2H_3Cl)_n$ in the original waste. Sulphates and chlorides are highly soluble in water.

2.3 Management of APCr

In the UK, APCr are classified as hazardous waste with absolute entry and code 19 01 07 according to the European Waste Catalogue (EWC) (European Comission, 2013), which states that the environmental impact of APCr is associated with their chemical composition and leaching properties. Due to the fine particle size of APCr (max. 1,000 μ m), as well as their high contents of persistent organic pollutants (POP) and leachable salts and metals, this waste must be treated before being sent to landfill (European Commission, 2015; Quina, et al., 2008a; Todorovic & Ecke, 2006).

Based on a business perspective, APCr management comprises three main options: utilisation for the neutralisation of acid waste, backfilling of underground salt mines and landfilling after appropriate stabilisation (Maresca, et al., 2022). More alternative management options have been identified considering both wastes APCr and FA (Quina, et al., 2018):

- Options not aimed at recovery: backfilling or treatment followed by landfilling.
- Options aimed at recovery of secondary materials/products: detoxification treatment, for instance, washing.

The APCr management options aimed at recovery of materials and products are currently subject of research, rather than been applied at industrial scale. Some examples include manufacture of products, such as cementing materials (Ghouleh & Shao, 2018; Ashraf,

et al., 2019), practical applications like CO₂ sequestration (Cappai, et al., 2012), and recovery of metals like Pb and Cu (Yang, et al., 2013).

These options are available in different countries and can be combined depending on the local traditions and legislation (Astrup, 2008). The regulation for APCr management has been basically aimed to relieve the impact on the environment and human health through the safe disposal of waste (The Council of the European Union, 1999; Quina, et al., 2018). In contrast, the reuse of APCr is forbidden in many countries due to their high contents of heavy metals (Quina, et al., 2008a). Despite this barrier, there is an increasing interest in recycling of APCr (Huang & Chu, 2003; Ferreira, et al., 2003; Quina, et al., 2008a; Gunning, et al., 2011a; Hwang, et al., 2012; Quina, et al., 2014a; Keppert, et al., 2015; Ashraf, et al., 2019; Han, et al., 2022), as well as recovery and recycling of different materials from APCr, such as metals, salts and inert fractions (Quina, et al., 2018; Aguiar del Toro, et al., 2009).

Currently, in the UK, most of the APCr generated by MSWI plants is treated and then disposed of in hazardous landfills or in underground salt mines (Amutha Rani, et al., 2008; Keely, 2015; Veolia, 2023) and at a lesser extent, APCr is used to neutralise acid waste (Amutha Rani, et al., 2008). The backfilling of APCr into salt mines takes advantage of the chemical stability and homogeneity of this waste material, as well as the atmospheric conditions of the mine to prevent any release of pollutants into the environment (Veolia, 2023). The most used treatment applied to APCr before sending the waste to landfill is complex and consists of stabilisation/solidification (S/S) with chemical additives. This treatment leads to an augmentation of the mass and volume of the waste, which results in additional costs due to transportation and disposal of the treated waste (Li, et al., 2007). The cost of disposal of APCr in the UK ranges between £110 and £150 per tonne by 2023 (Augean, 2023). This cost is not very different from the landfill of nonhazardous waste that ranges from £112 to £127 per tonne, but it is significantly higher than the cost of landfill of inert waste which varies between £13 and £28 per tonne (Soilutions, 2023; WRAP, 2022). These estimations include disposal and landfill tax. Also, the Gate Fees 2021/22 Report (WRAP, 2022) states that the EfW operators in the UK have remarked on a potential increase in the costs of treating APCr as per changes in the legislation. Therefore, there is a need for an alternative management that allows to divert APCr from landfilling and, instead it enables this waste material for further recycling.

APCr should comply with the limit values set by the Waste Acceptance Criteria (WAC) for wastes that need to be landfilled as expressed in the Landfill Directive (The Council of the European Union, 2003). However, for APCr it results difficult to accomplish those limits because of their high contents and leaching behaviour of pollutants, mainly chlorides, sulphates and lead (Amutha Rani, et al., 2008). To deal with this issue, APCr management in the UK takes place within the 3xWAC by derogation of hazardous waste rules (Date, 2017; Deegan, 2017), which allows some landfill sites to receive and dispose of APCr with up to three times the limit values established in the WAC for hazardous waste. The 3xWAC sites were intended to be temporary until better options for APCr management were developed (Keely, 2015), however, in February 2017, Ministers of the Department for Environment, Food and Rural Affairs (DEFRA) decided not to remove the 3xWAC derogation, fact that was not well received by the industrial stakeholders, because according to their opinions, the UK Government's commitment and investment in alternative technologies aimed to reuse and recycle APCr could be reduced (Date, 2017; Deegan, 2017). In this regard, it is important to highlight the commitment from waste treatment operators and researchers in keeping the interest in developing APCr treatment technologies with the purposes of recycling.

2.4 Treatment of APCr

The variable concentrations of metals, such as lead, and salts, especially chlorides, make APCr treatment a difficult practice, hence these wastes must be carefully characterised before being treated (Quina, et al., 2008a). The main challenges for the potential reuse of APCr are focused on their high contents of heavy metals and salts, mainly chlorides, and their leaching properties, as well as the presence of organic micropollutants, such as dioxins and furans (Ferreira, et al., 2003; Aguiar del Toro, et al., 2009). In addition, it must be taken into account aspects like financial and energy costs, generation of pollutant effluent, environmental impact of the treated waste and quality control of the recovered materials.

Waste treatment techniques for APCr can be grouped into four categories according to the main principle of operation: extraction-separation, chemical stabilisation, solidification and thermal treatment (Astrup, 2008). The goals to be achieved when treating APCr must be related to the main objective of the selected technique. For example, decreasing the amount of a contaminant requires the use of a separation process,

while the reduction of its leaching availability can be achieved by using a stabilisation method, and S/S is more advisable to decrease the leaching rates (Sabbas, et al., 2003). A description of the principles and objectives of the most used techniques for the treatment of APCr is shown in the following sections.

2.4.1 Extraction-Separation technologies

Extraction-separation techniques are aimed to extract some of the contaminants from MSWI residues in order to recover those species or to obtain a better-quality residue which can be subsequently recycled (Quina, et al., 2008a). The main focus of these techniques is the removal of heavy metals and salts from APCr mainly using water or acids as extracting agents (Astrup, 2008). These two techniques will be described in the following sections.

2.4.1.1 Water washing

Treating APCr by washing using water as extracting medium produces a highly alkaline suspension with pH usually between 11 and 13 and high concentrations of salts, including chlorides and sulphates, as well as heavy metals, such as Pb, Zn, Cr and As (Astrup, 2008). This treatment option has been tested in various studies aimed to remove metals and soluble salts from APCr, resulting in an improvement of the leaching properties of the waste material. Table 2.3 summarises the water washing conditions for the treatment of APCr, reported by different studies. All the referenced studies have been performed at laboratory scale and ambient temperature and used either distilled or deionised water. In most of the studies, the APCr washing requires only one extraction step. If water washing of APCr is brought into a full-scale, the requirement of water and the extraction time should be as low as possible without detriment to the efficiency of the treatment (Abbas, et al., 2003). Also, the washing must be optimised to lessen the water consumption as much as possible, so that the management of APCr is economically feasible (Colangelo, et al., 2012).

Reference	Country	Type of washing	Final pH of the leachate	L/S ratio (L water/Kg APCr)	Extraction steps	Contact time
Rouchotas & Cheeseman (2001) cited by (Amutha Rani, et al., 2008)	UK	Washing with distilled water	NA	5, 10, 20, 40, 60 Optimum: 20	1	30 min
(Lundtorp, et al., 2002)	Denmark	Mixing with water and Fe(SO ₄) plus aeration step	11.0 – 12.5 adjusted to 10.4 – 11.00	2.7 – 3.9	4	Aeration: 20 – 50 min After pH adjustment: 30 – 60 min
(Abbas, et al., 2003)	Sweden	Washing with water	NA	1, 2	1 3	15 min 5 min each
(Chimenos, et al., 2005)	Spain	Washing with water	12.2 - 12.6	1 – 10 Optimum: 3	2 washings plus 1 rinse	0.5 to 24 h Optimum: 1 h
(Hammy, et al., 2005)	Canada	Washing with tap water plus further treatment of the leachate with peat moss	11.3 – 12.5	5	1 washing plus 3 sorption	Washing: 1 h
(Aguiar del Toro, et al., 2009)	Germany /Sweden	Washing with distilled water Full carbonation until constant pH Acidification with H ₂ SO ₄ (c) or alkalinisation with 1 or 12 M NaOH	After washing: >6 After carbonation: 6 Adjusted with acid: 3 Adjusted with base: 7	3,10, 20	1 water washing 1 CO2 bubbling 1 acidification or alkalinisation	10, 60, 120 (After pH adjustment)
(Karlfeldt Fedje, et al., 2010)	Sweden	Washing with deionised water plus a rinse step of the solid residue	12.4	50	1	24 h (Sampling at 6 and 24 h)
(Chen, et al., 2012)	Taiwan	Roasting process at different temperatures Washing with deionised water	12.18	10	1	1.5 h

Table 2.3. Conditions for APCr water washing reported by different studies.

(Colangelo, et al., 2012)	Italy	Washing with deionised water	>12	2, 3, 5, 10 Optimum: 3	1	24 h
(Chen, et al., 2013)	Taiwan	Washing with deionised water	NA	5	2	5 min
(Quina, et al., 2014a)	Portugal	Washing with distilled water	12.1	10	1	10 min
(Quina, et al., 2014b)	Portugal	Washing with distilled water and addition of at least 0.06 mL of phosphoric acid per g of APCr		10	1	0.5, 1, 5, 10, 30, 60, 480 min, and 24 h Optimum: 10 min
(Keppert, et al., 2015)	Czech Republic	Washing with pure water	NA	5, 10	1	NA
(Chen, et al., 2017)	Denmark	Washing with distilled water	NA	2, 5	1 at L/S: 2 and 5 3 at L/S: 5	5 min
(Yang, et al., 2017)	China	Washing with distilled water	Around 11.95 – 12.00 at optimum conditions	3, 10 and 50 Optimum: 3	1	2, 5, 10, 15 and 30 min 1, 2, 4 and 16 h Optimum:5 min
(Bogush, et al., 2019)	UK	Washing with deionised water	11.8	10	1	30 min
(Dontriros, et al., 2020)	Thailand	Washing with deionised water	NA	5	1	60 min
(Cao, et al., 2021)	China	Washing with deionised water plus acid washing with mixed 0.2 M HCl/0.06 M H ₂ SO ₄	11.29	2 and 10	1 at L/S: 2 1 at L/S:10	1 hr at L/S: 2 10 min at L/S:10

NA indicates that the value of the parameter was not available in the corresponding report.

The liquid/solid (L/S) ratio as a measure of the water requirement and the extracting or washing time are determining factors in the washing of APCr. In a systematic study about the effects of water washing techniques on the removal of chlorides, sulphates and heavy metals in APCr intended either for disposal or recycling, it was concluded that over 70% of chlorides and about 25% of sulphates can be removed from these wastes independently of the extracting time, but depending on the L/S ratio, while the extracting behaviour of heavy metals is highly dependent on the time factor; for that reason the reaction kinetics must be taken into account (Yang, et al., 2017). Some researchers optimised the APCr treatment in terms of the factors L/S ratio and washing time in their works. The most reported value for optimum L/S ratio is 3 (Chimenos, et al., 2005; Colangelo, et al., 2012; Yang, et al., 2017), while the optimum washing time ranges from 5 min (Yang, et al., 2017) to 1 hr (Chimenos, et al., 2005).

A number of studies that evaluated the washing of APCr with water have demonstrated that this treatment leads to important modifications in the elemental and mineralogical composition of the waste material. A single-stage washing of APCr allowed the extraction of large amounts (order of mg/g) of soluble cations, including Na, K and Ca, in the form of water-soluble species, such as CaO, CaOHCl, NaCl and KCl (Rouchotas & Cheeseman, 2001, cited by (Amutha Rani, et al., 2008)). The study conducted by Bogush, et al. (2019) showed that a single-stage washing of APCr achieved a substantial removal of Na, K and Cl in the forms of NaCl, KCl and CaOHCl, as well as Ca, while the concentration of elements such as Al, Si, Mg, Fe and potential pollutants, like Zn, As, Cd, Co, Cr, Cu, among others, increased as a consequence of the loss of dry matter during the dissolution of soluble salts. Also, the author points out that the washing of APCr changes the Pb speciation because this metal in the form of glass may be partially dissolved and turned into PbO. Cao, et al. (2021) applied a two-step washing pre-treatment to APCr achieving the removal of 24.8 to 93.1 % of Ca, Na, K and Cl. The author states that this efficiency of the washing treatment is due to most of the compounds containing the extracted metals are freely soluble and that some metals turned from their oxidation forms into exchangeable forms.

It is important to describe the efficiency of washing of APCr in terms of the removal of soluble salts, mainly chlorides, and heavy metals, also considering the aspects of washing steps, water consumption and extracting time. A significant reduction in the leachability of chlorides and heavy metals from APCr by means of a treatment consisting of two

washing steps plus a rinse step was achieved in the tests carried out by (Chimenos, et al., 2005). The removal of chlorides and sulphates from APCr by washing with a minimum amount of water was achieved with a L/S ratio as low as 3, extracting up to 8 wt% of chlorides and up to 5.8 wt% of sulphates (Colangelo, et al., 2012). The leaching of chlorides and heavy metals was significantly improved by washing with water according to the results reported by Quina, et al. (2014a). In that study, the leaching rate of chlorides, initially as high as 98,400 mg/kg was non-existent in the washed material. With respect to the leaching of metals the heavy metals, Cu, Ni and Cd were not detected in the leachate from washed APCr, while Zn and Pb leached only at 36% and 40%, respectively, of the amounts released from the raw APCr. In the research conducted by Yang, et al. (2017), a single-step washing process (L/S:3 and time: 5 min) allowed the extraction of over 70% of chlorides and almost 25% of sulphates and some heavy metals from APCr. The author remarks on the extracting behaviour of these contaminants: while the extracted amounts of Co, Cr, Fe, Ni, V and Cu increased over time; Ba and Mn increased initially but decreased afterwards; Pb and Zn gradually declined, and Cd and As were not affected by the extraction time. Dontriros, et al. (2020) compared the use of acidic and alkaline solutions and water for chlorides and sulphates removal from APCr, among other MSWI residues, concluding that the treatment with deionised water produced the highest efficiency.

In addition, the physical characteristics of the APCr can be modified by washing with water. In a study about the assessment of different leaching methods, it was found that after washing with water, the maximum particle sizes of APCr of 20-40 μ m were reduced to 10-20 μ m, whereas their specific surface area rose from 5.1 to 10.7 m²/g (Karlfeldt Fedje, et al., 2010). Comparable results were found in another research in which the average particle diameters of raw and washed APCr were 84 and 44 μ m, respectively, while the specific surfaces increased from 0.394 m²/g to 0.742 m²/g (Keppert, et al., 2015).

APCr washing is sometimes applied in the first stages of more elaborated procedures that are designed for the stabilisation of the ash, extraction of polluting substances or enabling the ash for subsequent use. In a study on the potential of a counter-current sorption process for Pb extraction from APCr, the washing of the waste with tap water prior to a treatment of the leachate with peat moss allowed an efficient recovery of Pb (Hammy, et al., 2005). A wet treatment process consisting of water washing followed by carbonation

and pH adjustment was optimised for the extraction of chlorides, Cu, Cd, Pb and Zn (Aguiar del Toro, et al., 2009). A treatment comprising the steps of roasting, washingflushing and carbonation achieved 97% of chloride removal from APCr, enabling the ash for recycling and reuse in the production of construction materials (Chen, et al., 2012). In a study of the mechanical-chemical stabilisation of APCr, the ash was pre-treated by washing with deionised water for removal of chlorides, and then, a wet milling process for 96 hours was applied to stabilise the heavy metals; the final product was incorporated into an OPC paste to induce pozzolanic reactions. The results showed that the milling treatment helped to stabilise the metals and had an activating effect on the washed APCr, enabling the residue for partial substitution of OPC in OPC paste (Chen, et al., 2013). In the research conducted by (Quina, et al., 2014a), different quantities of APCr treated by washing with water were incorporated into the production of LWA. In a study that compared the washing of APCr with different substances, including pure water, acid water coming from a flue gas absorber, diluted HCl and 0.1% (NH₄)₃PO_{4;} the washing with pure water produced the best results in terms of enabling the ash for reuse as a supplementary cementitious material in concrete (Keppert, et al., 2015). The research conducted by Chen, et al. (2017) determined that the combination of water washing, followed by electrodialytic separation and sintering, stabilised APCr and lowered the evaporation rate, mainly of Pb, during the thermal treatment of the ash. Cao, et al. (2021) developed a process consisting of two steps of water washing plus one step of acid washing (mixed 0.2 M HCl/0.06 M H₂SO₄) for the treatment of APCr to reduce their environmental risk. In this last study, the removal efficiency achieved by water washing was 93.1% for Cl, 41.4% for Na, 48.5% for K and 24.8% for Ca.

The use of chemical additives is suggested to avoid the simultaneous leaching of heavy metals, especially Pb and Zn (Quina, et al., 2008a). For example, the addition of small amounts of phosphoric acid H₃PO₄ into the water reduced the leaching of Pb, Zn, Ni and Cu from APCr (Quina, et al., 2014b). This is possible because of the formation of insoluble metal phosphates, for example Pb₅(PO₄)Cl (Mizutani, et al., 2000). The addition of reagents, such as Na₂S or NaOH to form sulphide or hydroxide sludge, when washing APCr in order to meet the effluents requirements is also recommended (Hu, 2005). In a pilot-scale experiment, it was found that mixing raw APCr with water and Fe₂(SO₄) followed by an aeration step promoted the removal of salts (Lundtorp, et al., 2002). The addition of small quantities of MgSO₄ into a counter-current batch process for APCr washing with water, decreased the leaching of heavy metals because it promoted the

formation of the corresponding insoluble hydroxides of these elements (Chimenos, et al., 2005).

Despite the multiple benefits of washing APCr with water, it is clear that this treatment generates wastewater that can be difficult to dispose of, given its high contents of chlorides and metals. Therefore, treatment options of the APCr washing effluent aimed at decontamination prior to discharge or reuse must be considered. This wastewater can be treated by means of well-established procedures like chemical precipitation, crystallisation, adsorption, membrane filtration, among others; however, these treatments can be expensive depending on the amount of wastewater generated (Colangelo, et al., 2012). If the wastewater is sent for treatment that includes procedures of evaporation and crystallisation, it is possible to recover salts like NaCl, CaCl₂, KCl and CaSO₄•2H₂O (Amutha Rani, et al., 2008; Astrup, 2008). Another option to deal with the wastewater can be its reuse as raw material in the same MSWI plant where the APCr is produced, for example, to extinguish the fire of bottom ash (Chimenos, et al., 2005). Following this last approach, Colangelo, et al. (2015) suggests evaporating the wastewater by means of the heat generated from the waste combustion and reusing the treated water in the industrial cycle.

2.4.1.2 Acid washing

A common treatment aimed to reduce the leaching of hazardous substances, including salt and heavy metals from APCr is washing the residue with acid solutions. The extracted contaminants are mainly heavy metals because these elements are more soluble in acidic conditions (Astrup, 2008). Variables such as extracting agent, pH and the L/S ratio, affect the efficiency of the acid extraction (Quina, et al., 2008a). A number of laboratory-scale studies aimed at stabilising APCr for further disposal or subsequent recycling have used acid extraction, mainly with HCl, HNO₃ and H₂SO₄ (Mizutani, et al., 1996; Van Der Bruggen, et al., 1998; Karlfeldt Fedje, et al., 2010; Dontriros, et al., 2020; Cao, et al., 2021; Hong, et al., 2000; Keppert, et al., 2015). The development of a treatment method that uses acid extraction with either 35% HCl or 65%HNO₃/35%HCl and stabilisation with NaHS at full-scale was described by Katsuura, et al. (1996). APCr can be treated in situ by extraction with the acid solutions coming from the incineration flue gas treatment. Keppert, et al. (2015) evaluated the APCr treated with acid effluent for potential use as supplementary cementitious material. Cao, et al.

(2021), treated APCr with an acid solution of mixed 0.2 M HCl and 0.06 M H_2SO_4 that simulated the incineration flue gas solutions.

Acid washing of APCr consists of making acid solutions pass through the raw ash to obtain solutions with high concentrations of targeted metals (mainly Pb and Zn) followed by further separation and recovery. The acid extraction experiments carried out on APCr by Karlfeldt Fedje, et al. (2010) showed that the use of inorganic acids is highly effective for the removal of many elements, for example, 3 M HCl, 3 M HNO₃ and 1.5 M H₂SO₄ leached from 36 to 98% of Fe, Mn, Al and Si; while the extractions with the organic acids 0.1 M formic acid, 0.1 M acetic acid, 0.1 M and 1.0 M lactic acid and 0.5 M oxalic acid had a negligible effect on the removal of the metals.

The removal of metals from APCr can be improved by adding chemical agents into the acid-extracting medium. For example, in the tests conducted by Karlfeldt Fedje, et al. (2010), the addition of the chelating agent EDTA into the acid-extracting solutions increased the removal efficiency of many elements from APCr, mainly Cu, Zn and Pb. Also, it has been demonstrated that a pre-treatment of APCr with water prior an acid extraction helps to substantially reduce the leaching properties of the ash, according to the two steps water washing and one step acid washing process developed by Cao, et al. (2021), which reduced the leaching of Cu, Zn, Cd and Pb by 80 to 100%, As by 30 to 80% and Ni up to 30%.

Although acid washing increases the removal of the metals, it does not always produce an improvement of the APCr leaching, because this property is directly determined by the mineralogy of the treated residues rather than the metals contents (Astrup, 2008). The other disadvantage of acid washing is the high requirement of acid due to the alkaline nature of the APCr (Karlfeldt Fedje, et al., 2010). In addition, the experiments conducted by Dontriros, et al. (2020) showed that extracting MSWI residues, including APC, with acidic solutions, did not change the pH of the treated ashes because the CaCO₃ contents of these materials act as a buffer.

2.4.2 Chemical stabilisation

The objective of chemical stabilisation is binding and immobilising contaminants, mainly heavy metals, in the waste matrix by using additives or binders (Astrup, 2008). During stabilisation, the geochemical stability of the waste is improved because new mineral phases with low solubility rates are formed (Taylor Eighmy, et al., 1998). In general, stabilisation of incineration waste involves a stage of extraction with a chemical substance in solution, then a series of chemical reactions and finally a dewatering process of the product, resulting in the improvement of the leaching properties of the treated waste (Astrup, 2008). This enables the waste material for recycling or landfilling.

Different substances, such as sulphates and phosphates, have been tested as stabilising agents for APCr. A treatment called Ferrox for reducing the leaching of trace elements, such as Pb and Cd, and salts from APCr by mixing with FeSO₄ solution and subsequent oxidation of the suspension was developed and evaluated at laboratory and semiindustrial scale (Christensen, et al., 2000; Lundtorp, et al., 2002; Lundtorp, et al., 2003). The efficiency of extracting APCr with a mixed FeSO₄/Fe₂(SO₄)₃ solution was assessed by Hu (2005) aimed to prevent the leaching of heavy metals from the treated ash.

In a study (Quina, et al., 2010), five different additives were assessed for reduction of heavy metals leaching from fly ash and APCr: sodium hydrosulphide hydrate (NaHS), H_3PO_4 , Na₂CO₃, sodium diethyldithiocarbamate trihydrate (C₅H₁₀NNaS₂•3H₂O) and soluble sodium silicate (Na₂O•SiO₂). The results showed that all these additives, mainly the soluble phosphates, stabilised the most concerning heavy metals, including Pb and Zn, while the leaching of Cr increased with to the addition of H₃PO₄, Na₂CO₃ and Na₂O•SiO₂. However, the effect of these subtances on elements associated with soluble salts (K, Na and Cl) was insignificant, therefore, the authors recommended a pre-washing of the ash.

Keppert, et al. (2015) tested the washing with $(NH_4)_3PO_4$ to remove soluble salts from APCr and to enable the waste to be used as a supplementary cementitious material. The Vandkvalitests Institute (VKI) treatment consisting of extraction and chemical stabilisation of APCr with water, concentrated H_3PO_4 and CO_2 was evaluated in terms of the leaching of heavy metals Cd, Cr, Cu, Pb and Zn by Heng, et al. (2018). Atanes, et al. (2019) treated APCr with a Na₂CO₃ solution as a stabilising agent achieving the

simultaneous separation of soluble salts and leaching stabilisation of heavy metals. Dontriros, et al. (2020) found that a 0.25M Na₂CO₃ solution was more efficient than water and diluted solutions of HCl and HNO₃ for the removal of sulphates.

Some organic chelating reagents have been assessed for the stabilisation of APCr, however, the stability of the products still needs to be assessed. Mizutani, et al. (2000) evaluated the use of an organic sulfide chelating agent, as well as phosphate and ferrite treatments. Hong, et al. (2000) assessed the extraction of heavy metals from APCr by means of the chelating agents ethylenediaminetetraacetic acid EDTA, nitrilotriacetic acid (NTA) and diethylenetriaminepentaacetate (DPTA) in the pH range from 2.5 to 10. Karlfeldt Fedje, et al. (2010) investigated the extraction of APCr with EDTA, NH4NO3 and NH4Cl. Chelating agents are expensive; therefore, chemical stabilisation with inorganic reagents is considered more cost-effective (Hu, 2005).

2.4.2.1 Carbonation

Carbonation is a natural phenomenon which occurs due to the exposure to air at ambient conditions of a solid material containing alkali minerals that react with the atmospheric CO_2 producing stable carbonates (Pan, et al., 2012). In accelerated carbonation the material is exposed to a high concentration of CO_2 in the presence of moisture (Sun, et al., 2008; Gunning, et al., 2010; Pan, et al., 2012), so that, the reaction takes place in only a few minutes or hours (Pan, et al., 2012). The exposure of the solid residue under a gaseous CO_2 -saturated environment promotes a quick hardening of the non-hydrated product (Fernández Bertos, et al., 2004c). The carbonation of APCr, alike other alkaline waste materials, involves a complex series of reactions, which are strongly influenced by the characteristics of the material (nature of the surface, particle size, microstructure, mineralogy and elemental composition) and by the exposure conditions (CO_2 concentration, moisture of the surface of the solid material, it is conditioned by the chemical absorption of the CO_2 (Sun, et al., 2008).

According to Zhang, et al. (2008), if the final disposal of APCr is considered, accelerated carbonation works only as a temporary stabilisation that could be useful to reduce the leaching of heavy metals during the transportation and storage of the waste. Therefore, it is more viable to consider accelerated carbonation as a technique that enables APCr for

further reuse. In fact, accelerated carbonation offers two simultaneous environmental benefits: permanent binding of CO₂ and valorisation of a waste material (Fernández Bertos, et al., 2004c; Di Maria, et al., 2020). It has been demonstrated that by means of accelerated carbonation, it is possible to modify the properties of certain solid residues, such as APCr, so that the treated residues can be reused as construction materials, for example, engineering fills and aggregates (Fernández Bertos, et al., 2004a; Fernández Bertos, et al., 2004c; Gunning, et al., 2011a; Gunning, et al., 2011b; Gunning, et al., 2015). Table 2.4 summarises the procedures and conditions used for the carbonation of APCr reported by different authors.

Reference	Country	Reactor	Procedure	Temperature (°C)	Moisture (L water/Kg APCr)	CO2 pressure (bars)	Contact time	Effects on treated ash
(Ecke, 2003a)	Sweden	Glass reaction tubes. 50% CO ₂ :50% air	A daily gas flow of 2.2 L of CO ₂ was applied through the tubes with a peristaltic pump	Ambient (20) Oven (60)	0 and 0.5	NA	4 to 40 days	Pb and Zn mobility was reduced by two orders of magnitude. Cr was immobilised by adding water, but remobilised along time. Cd mobility increased
(Fernández Bertos, et al., 2004c)	UK	Stainless steel chamber saturated with 100% CO ₂ . Relative humidity was maintained at 65% with aid of a bath containing a solution of NaCl.	APCr were mixed with water at prefixed ratio and placed into cylindrical plastic dishes (9 cm diameter). The CO_2 was continuously fed from a pressurised cylinder.	NA	0 to 0.6 Optimum: 0.2 to 0.3	3	20 min to 24 hr Optimum: 2.5 hr	Leaching of Pb and Zn significantly decreased. Leaching of chlorides decreased. Leaching of sulphates increased. APCr particles became coarser due to agglomeration.
(Todorovic & Ecke, 2006)	Sweden	Plastic bucket	A daily flow of 33 L CO ₂ /Kg material was applied through the ash layer with a peristaltic pump	Ambient	0 Moisture was provided through saturation of the gas with water	NA	25 days	SO ₄ mobility increased by 62% CI ⁻ , Cr, Se and Pb remained unaffected or increased
(Prigiobbe, et al., 2009)	Italy	Thermogravimetric system coupled with Gas chromatography	Around 44 mg of APCr were placed on a holder in a layer of 3mm and treated with different mixes of CO_2 – argon (10, 22 and 50% CO_2)	300, 350, 400, 450, 500 (Optimum: over 350°C)	0	1	2 hr	Maximum conversions of Ca(OH) ₂ and CaOHCl into CaCO ₃ were between 60 and 80%, depending on the temperature and CO ₂ concentration.
(Baciocchi, et al., 2009a; Baciocchi, et al., 2009b; Costa, 2009)	Italy	150 mL stainless steel reactor placed in a thermostatic bath.Temperature was monitored with a thermocouple.Gas humidity was maintained at 75% using a saturated NaCl solution in the reactor.	1 g of sample previously dried at 105°C was moisturised and treated with 100% CO ₂ . A control sample was prepared by the same procedure and stored in a closed container in contact with air at atmospheric pressure in the thermostatic bath	30, 40, 50	0.02; 0.1; 0.2; 0.4; 0.6	1, 3, 5, 7, 10	10 min to 48 hr	Pb, Zn and Cu mobility decreased at high pH. Cr and Sb mobility slightly changed or increased. No effects on As and soluble salts.

Table 2.4. Procedures and conditions for APCr carbonation reported by different studies.

		-		-				
(Li, et al., 2007) *	UK	Closed chamber containing 100% CO ₂ at 75% relative humidity, which was controlled with a saturated NaCl solution placed in the reactor	The sample previously dried at 105°C was moisturised and placed in the chamber. A control sample was prepared by the same procedure and stored in a closed container under ambient conditions	Ambient	0.1 to 0.8 Optimum: 0.3	3	Previous trails: 3 hr, Then: 3 days	Pb mobility decreased. Cd mobility decreased. Zn mobility was reduced or slightly increased in samples of different origin. Cl and SO ₄ release were reduced.
(Zhang, et al., 2008)	China	Two columns (5 cm diameter, 25 cm length) Column 1: accelerated carbonation. Pure CO_2 at a rate of 0.4 m ³ /h, 500g of material Column 2: Slow carbonation. Air containing 0.03% v/v CO ₂ flowing through at 0.4 m ³ /h, 500g of material	500 g of moisture sample	Ambient	0.2	NA	Accelerated: 10 hr Slow: 7 days	Manufacture of carbonated aggregates made of waste materials, which show characteristics comparable to LWA available on the market
(Gunning, et al., 2010)*	UK	Pressurised reaction vessel. Relative humidity was maintained at 75% using a saturated NaCl solution	Oven dried and then moistened samples were carbonate in a 100% CO ₂ atmosphere	NA	NA	2	72 hr	Leaching of Ba, Mo, Cr and chlorides were reduced., while leaching of As and sulphates increased. Leaching rates of Cu, Ni, Pb and Se were not significantly affected.
(Cappai, et al., 2012)	Italy	Open vessel	Blowing CO ₂ through the slurry which is continuously stirred by a magnetic stirrer. pH was monitored and CO ₂ was suspended when pH reached prefixed values between initial 13.17 (untreated APCr) and final 8.22 (carbonate APCr). Leaching was monitored at the prefixed value of pH.	Ambient (20)	2.5	NA	3 hr	Mobility of Pb, Zn, Cu, Cr and Mo decreased. Sb mobility increased. Chlorides mobility was not affected.
(Araizi, et al., 2016)	UK	Closed system consisting of: 99.9% CO ₂ chamber, a rota-meter to control the gas flow, an aspirator to humidify the incoming gas, a water bath to generate ultrasonic field and containing 10 sealed plastic tubes connected in series, and a condenser to keep a constant temperature.	0.5 g of sample was placed inside a plastic tube (capacity: 50 mL) and moistened at prefixed ratio. The moistened samples were carbonated without agitation and then sonicated without agitation.	20	0.2; 0.4; 0.6; 0.8; 1; 5; 10;25; 50 and 100 Optimum: 0.4	1	1 hr	Maximum CO ₂ uptake was 13.3%. Reactivity of samples to CO ₂ was high at low w/s ratios $(0.2 - 0.6)$, weak at intermediate ratios $(0.8 - 10)$, and it increased again at w/s ratio equal or higher than 25.

* The tested material was MSWI FA containing APCr, NA = unspecified

There are two possible ways for the treatment of alkaline residues by accelerated carbonation, according to their exposure conditions: the dry route (gas-solid) and the wet route (fluid-solid) (Lim, et al., 2010; Li & Wu, 2022). The L/S ratio determines the carbonation route, for example, Lim, et al. (2010) used the L/S ratios of 0.2 and 5 for the dry route and wet route, respectively. In the gas-solid carbonation, as shown in Figure 2.11 Part A, the CO₂ mineral capture takes place under a relative humidity condition; while in the fluid-solid carbonation, as shown in Figure 2.11 Part B, the solid material is immersed in a liquid medium in which the CO₂ is bubbled (Li & Wu, 2022). The reactivity of incineration waste with CO₂ is due to their high contents of calcium oxides, hydroxides and silicates (Jiang, et al., 2000). The chemical and physical properties of APCr make this waste a good candidate to be treated by means of carbonation (Fernández Bertos, et al., 2004b). Specifically, APCr tend to be highly reactive to CO₂ because of their high contents of calcium hydroxides (Baciocchi, et al., 2009a; Costa, 2009). These two options of carbonation were analysed and compared for APCr by varying temperature, reaction time and CO_2 saturation, obtaining a result that both ways achieved a similar maximum conversion (65%) of calcium to carbonates, which means that 1 kg of APCr may be able to capture up to 250 g of CO₂ (Baciocchi, et al., 2009b).



Figure 2.11. Accelerated carbonation setups. A. Dry route (gas-solid), B. Wet route (fluid-solid). Modified from Li & Wu (2022).

It can be said that natural carbonation is not as effective as accelerated carbonation for the stabilisation of APCr. The study conducted by Todorovic & Ecke (2006), showed that the immobilisation of critical contaminants after 25 days of carbonation was not effective for immobilising chlorides, Cr, Se and Pb in APCr, since the cumulative release of these contaminants from the carbonated APCr remained constant or increased, also the cumulative release of sulphates after carbonation increased by 62%. On the other hand, different studies have demonstrated that accelerated carbonation produces significant changes in the elemental and mineralogical composition of APCr, which results in a decrease of their leaching properties (Fernández Bertos, et al., 2004b; Cappai, et al., 2012; Baciocchi, et al., 2009a; Li, et al., 2007; Costa, 2009). By mineralogical analysis, Li, et al. (2007) confirmed the disappearance of lime, portlandite and calcium hydroxychloride, and the formation of carbonates like calcite and vaterite as a result of carbonation. Costa (2009) found that the carbonation process has the potential to convert the metal-based minerals of the raw ash into less soluble carbonates. The most important chemical and physical changes in APCr, promoted by accelerated carbonation, were investigated and described in detail by Fernández Bertos, et al. (2004c); the list of changes comprises: increase of CaCO₃ content, agglomeration of particles, formation of calcium-metal salts and metal silicate complexes, significant reduction in leaching of Pb and Zn, slight reduction in leaching of chlorides, reduction of alkalinity and increase of leaching of sulphates.

The improvement of leaching properties in carbonated APCr is directly related to the immobilisation of hazardous substances and cementation by precipitation of carbonates (Fernández Bertos, et al., 2004b; Jiang, et al., 2009). In some studies, it was demonstrated that the leaching of heavy metals, such as Pb, Zn, and Cu from APCr can be reduced by accelerated carbonation at the natural alkaline pH of the material (Cappai, et al., 2012; Baciocchi, et al., 2009a; Costa, 2009). Besides the metals mentioned before, Cappai, et al. (2012) showed that Cr and Mo leaching was reduced by carbonation at an optimal pH close to 10.5. Li, et al. (2007) found that accelerated carbonation significantly decreased the leaching of Pb by 2 to 3 orders of magnitude. However, a mobilisation effect of carbonation on Sb was observed (Baciocchi, et al., 2009a; Cappai, et al., 2012; Costa, 2009). On the other hand, accelerated carbonation did not show major effects on the leaching of other metals and soluble salts. Baciocchi, et al. (2009a) and Costa, (2009) found that Cr, As, chlorides and sulphates are only slightly affected by carbonation. The same behaviour was observed by Li, et al. (2007) in the leaching of soluble sulphates and chlorides. Furthermore, in the study carried out by Cappai, et al. (2012), chloride leaching was not modified by carbonation. To address that issue, it is suggested to combine carbonation with other treatments, like the addition of sorbents or washing treatment after the carbonation step (Cappai, et al., 2012). In addition, the extreme alkaline character of APCr (pH usually higher than 12) can be mitigated by means of carbonation (Ecke, et al., 2003b; Li, et al., 2007). The reduction in pH varies in the different studies: nearly neutral

values (Fernández Bertos, et al., 2004c), 7 to 9 according to the reactivity of the ash (Li, et al., 2007), 8.22 (Cappai, et al., 2012), among others.

Taking into account the three systems that determine the accelerated carbonation of a solid residue: the reactor, the interaction of ash particle and CO₂ and the inner part of the porous particle, Fernández Bertos, et al. (2004b) conducted a series of experiments at laboratory scale to elucidate the kinetics of accelerated carbonation of APCr focused on the solid/gas interaction and the intraparticle reactions. The results of this study introduced the basis for modellling the process at a large scale. In another study (Fernández Bertos, et al., 2004c), the authors investigated how the parameters that affect the diffusivity and reactivity of CO₂ (particle size, reaction time and water content) can influence the extent and quality of carbonation in APCr and bottom ash. The optimum values of these parameters determined for accelerated carbonation of APCr were particle size less than 212 µm, reaction time 2.5 h and water/solid ratio from 0.2 to 0.3. The factors that are commonly evaluated due to their influence on the stabilisation of APCr by carbonation, are partial pressure of CO₂, the addition of water, temperature and reaction time (Ecke, 2003a). It was found that treating APCr by carbonation reduced the leaching of the major pollutants, especially Pb and Zn, by two orders of magnitude when the factor levels were increased; although under these conditions, the leaching of Cd increased and Cr was remobilised over time (Ecke, 2003a; Ecke, et al., 2003b).

For natural carbonation, the CO₂ source is atmospheric CO₂, while for the accelerated process, it can be either pure CO₂ or any CO₂-rich gas, for example, incineration emissions (Jiang, et al., 2009). In most of the studies at the laboratory scale on accelerated carbonation, pure or high-purity CO₂ is used for the experiments (Nielsen, et al., 2017). Nevertheless, those works provide valuable information about the efficiency of carbonation for specific materials. For example, Costa (2009) optimised the accelerated carbonation of APCr achieving CO₂ uptake capacities over 25% (w/w) using a setup with 100% CO₂ gas.

There is a need for efficient and low-cost CO_2 gas sources for accelerated carbonation, such as flue gas or biogas (Nielsen, et al., 2017). Some studies have been focused on this regard. Gunning, et al. (2011a) demonstrated the use at the pilot scale of landfill gas containing 13% to 14% of CO_2 for accelerated carbonation of APCr, achieving the reduction of CO_2 in the gas stream to less than 2%. Due to the physical characteristics

and chemical composition of APCr, the waste was successfully used to regenerate the spent K_2CO_3 solutions produced by an absorption column that captured CO_2 from landfill biogas; the treated wastes showed improvement in their leaching behaviour (Baciocchi, et al., 2012; Baciocchi, et al., 2014).

The possibility of sequestering CO₂ from waste incineration flue gas by means of accelerated carbonation of APCr has been assessed in various studies. Sun, et al. (2008) reported that APCr can achieve a CO₂ intake between 8 and 12 % (w/w) from incineration flue gas. Jiang, et al. (2009) compared the CO₂ capture of fly ash under different conditions using pure CO₂ gas or simulated incineration flue gas (12% CO₂), and concluded that the maximum CO₂ intakes, given in %(w/w), were comparable when using any of both sources: 16.3 % and 13.2 % in dry ash for pure CO₂ and for simulated incineration flue gas, respectively, as well as 21.4% and 18.5% in ash added with 20% of water for pure CO₂ and for simulated incineration flue gas, respectively; although, the intake rate was fast for pure CO₂ and slow for simulated emissions. Also, the amount of APCr produced in MSWI plants is scarce compared to CO₂ emissions (around 50 g ash/Kg waste compared to 1,320 g CO₂/Kg waste), so accelerated carbonation of APCr could capture only about 1% of the total CO₂ produced in an incineration plant (Costa, 2009).

For a potential use of carbonation at large scale, it is necessary to consider a practical source of CO_2 that meets the requirements of volume or mass of residue to be treated, gas pressure and constant flow. Two options recommended by Ecke (2003) can be landfill gas and flue gas produced during the waste incineration. In both cases, the availability of the gas and its interaction with the ashes need a thorough examination.

2.4.3 Solidification

The purpose of the solidification techniques is to physically and hydraulically encapsulate and immobilise solid residues, obtaining as a result a decrease in the leaching of heavy metals and an improvement on the mechanical properties of the residues (Astrup, 2008). For APCr solidification, this waste material can be mixed with other residues to meet the WAC for landfill, for example with co-fired pulverised fuel ash (Shirley & Black, 2011), or to form "cement-like materials". At the laboratory scale, the use of additives containing micro silica was evaluated and compared with different standard types and grades of cement for immobilisation of APCr. The types and grades of cement were CEM I 42.5, CEM I 52.5, CEM II 32.5, CEM III/A 32.5, CEM III/B 32.5, CEM III/B 42.5, CEM III/C 32.5. The results showed that micro silica-based materials reduced the leaching of Pb 2 to 20 times more than cement (Geysen, et al., 2004). Nevertheless, the most common substances used to solidify APCr are water and cement.

Solidification of APCr with water is currently in use in the UK as an integral part of the multi-step process of stabilisation/solidification (S/S). Solidification was also, investigated as a stand-alone process at a laboratory scale (Todorovic, et al., 2003). The authors concluded that this technique was effective for decreasing leaching rates of Ba, Ca, S, Mg, Al, Hg, Si, Mn, Pb and Zn, while Na, K and Cl were weakly bound to the solid mass and over 88% of their total amounts was leached, as well as more than 40% of total Cr was released.

Portland cement is currently the most used binder to solidify APCr, and its focus can be either landfilling or use of the final product in the manufacture of secondary materials. The immobilisation of heavy metals with cement lies in the physical and chemical capture of these pollutants in the cement matrix (Chandler, et al., 1997). This technique has been investigated mainly with the use of Portland cement, which is also the most used practice at an industrial scale in Europe (Quina, et al., 2008a). Generally, solidification of APCr with cement involves mixing the residue with cement, water and other additives that could be other wastes or specific materials that improve the strength of the mix (Astrup, 2008). Research in this field has been mainly focused on metals binding, strength development and improvement of leaching properties (Astrup, 2008; Huang & Chu, 2003; Lampris, et al., 2011; Geysen, et al., 2004). Nevertheless, the disadvantage of APCr solidification with cement is that the solidified product tends to deteriorate due to the presence of compounds that absorb moisture, leading to reduce the effectiveness of solidification and leaching of hazardous components (Hu, 2005).

Usually, solidification processes are associated with previous stabilisation treatments. The combination of both techniques, stabilisation and solidification (S/S), for the treatment of APCr is aimed to chemically stabilise the substances that are easily leachable and to solidify the residues. The binders frequently used to treat APCr by S/S at large scale are cement and pozzolanic materials (Quina, et al., 2008a). Although S/S has been considered the best option for APCr treatment to comply waste disposal regulations

(Huang & Chu, 2003), it should be noted that these methods are not highly effective to immobilise soluble salts, such as chlorides (Quina, et al., 2008a; Lampris, et al., 2009; Lampris, et al., 2008), consequently, it is needed an additional stage for extraction-separation or monitoring over time the leaching of those substances. In addition, the S/S technique has the disadvantage of increasing the mass and volume of waste, which results in additional costs of transportation and disposal (Li, et al., 2007).

2.4.4 Thermal treatment

Thermal treatments of MSWI residues use heating at high temperatures, usually above 1,000°C (Amutha Rani, et al., 2008). By means of thermal treatment, the highly volatile metals contained in the residues can easily evaporate into a gaseous state; then, the gas can be cooled and condensed for a subsequent separation and/or recycling of the metals (Lee, et al., 1999). Usually, the metals of interest for separation/recycling are Zn, Pb, Cu and Cd (Quina, et al., 2008a). The substances that do not fully vaporise, are bonded into a more stable solid matrix through some chemical reactions that take place during thermal treatment, such as oxidation (Lee, et al., 1999). These processes induce changes in the chemical and physical properties of the waste and even encapsulation of their leachable components, stabilising the waste and making it suitable for the manufacture of construction materials (Astrup, 2008; Amutha Rani, et al., 2008). Thus, the advantages of thermal treatments can be summarised as reducing the leaching of pollutants, further decrease of the volume of residues going to landfill, and production of stable materials suitable for utilisation (Chandler, et al., 1997; Amutha Rani, et al., 2008). In addition, thermal treatment is the only option that ensures the removal of the POP existing in the waste incineration residues matrix and, to some extent reduces the mass of the waste (Ecke, et al., 2000). These treatments have been investigated for many years in different residues from waste incineration (Quina, et al., 2008a).

Application of thermal conditioning in APCr has shown a good performance for heavy metals stabilisation and the treated waste may be inserted in the production of building materials. It is possible to use the final product because thermal treatment allows the incorporation of elements of interest into the glass matrix by chemical bonding and/or encapsulation (Quina, et al., 2008a; Dimech, et al., 2008). However, the high costs of equipment, high energy consumption and secondary pollution related to the production of flue gases that contain pollutant volatile metals are aspects that need to be considered

as disadvantages of this technology (Hu, 2005; Chandler, et al., 1997). Studies about the thermal treatment of APCr are mostly focused on vitrification and sintering processes.

Vitrification is the process of melting a mixture of waste material and glass precursors (e.g. Si) in order to form an amorphous single-phase glassy product, in which the contaminants are fixed (Chandler, et al., 1997; Amutha Rani, et al., 2008). APCr were mixed with silica and alumina and melted using plasma technology to produce a glass-ceramic material with properties similar to marble and porcelain (Roether, et al., 2010). Nevertheless, vitrification is much more expensive compared with other thermal treatments, and also it requires further treatment of salt concentrates because the salt cannot be vitrified (Van Der Sloot, et al., 2000).

Sintering consists of heating the waste, without reaching the melting point, in order to get a reconfiguration of its chemical phases, obtaining a denser and less porous material (Astrup, 2008). Treating MSWI ashes by sintering allows to decontaminate the waste and to recycle the metals that are released during the process. Usually, sintering temperatures range between 900 and 1,300°C (Dimech, et al., 2008; Ashraf, et al., 2019; Zhang & Qi, 2012; Chen, et al., 2017; Ashraf, et al., 2019), but even lower temperatures can be used, for example Lee, et al. (1999) heated APCr up to 850°C with a high-efficiency processing system at pilot scale obtaining a sintered product with better quality to be reused or landfilled. In a recent study (Ashraf, et al., 2019), APCr were sintered at 1,000°C and then inserted into the production of eco-cement with only 6% of additives developing strength comparable to OPC.

2.5 Recycling options for APCr

In order to obtain a beneficial product from APCr, there are two different approaches that should be considered (Quina, et al., 2008a), the first one is applying a pre-treatment aimed to improve the properties of the residues and then manufacturing the final product. The second approach is making the final product without a pre-treatment of the precursor material. The first approach is preferred by researchers, despite the high costs that may be associated with treatment technologies. It is also important to note that recovery and utilisation of MSWI residues are derived from and related to any treatment technology, and the treatment processes that are currently used form an integral part of the utilisation solutions (Astrup, 2008). Currently, APCr treatment combines extraction-separation

technologies with S/S methods. For the treatment of APCr, it is important to remove soluble salts as well as to immobilise the leachable elements, especially when the final product is intended to be used in the manufacture of secondary materials.

Different studies have demonstrated that APCr could be recycled as a secondary material for several applications in the field of construction, as shown in Table 2.5. From the different products potentially manufactured from APCr, the LWA is a good option to develop given the demand and positive impact of this type of material in the construction industry. The incorporation of APCr into the structure of artificial LWA brings three benefits: reducing the need for landfilling of a hazardous waste, helping to reduce the extraction of natural resources and creating new value-added products. From a technological perspective, the manufacture of LWA has become a focus of special interest since the low particle and bulk densities of these materials led to reduce the load transmitted to the ground, as well as less work and effort are required for their transport (de' Gennaro, et al., 2004). Since these low densities are due to the formation of pores and voids, the technical benefits of LWA include high thermal and acoustic insulation as well as good resistance to fire (González-Corrochano, et al., 2009). Thus, the recycling LWA manufactured from APCr can become a more sustainable and cost-effective option of recycling/recovery of this waste material. The literature review about the incorporation of APCr into LWA (since it is the aim of this research) and the different applications of these materials will be detailed in the following section.

Table 2.5 Applications of APCr recycled as a secondary material in the construction industry.

Secondary material	Reference	Comments
Cementing materials known as "eco-cements"	(Huang & Chu, 2003; Ghouleh & Shao, 2018; Ashraf, et al., 2019; Bogush, et al., 2020)	The eco-cements show properties comparable to OPC. However, their binding strength needs to be developed by carbonation activation.
Supplementary cementitious material	(Keppert, et al., 2015)	Washed APCr has a fraction of particles larger than 125 μ m that must be removed. Persistent high leaching of sulphates can affect the durability of the mortar. The compressive strength of mortar was moderately reduced, but its resistance to freezing improved.
	(Dontriros, et al., 2020) (Formosa, et al., 2017)	Treated APCr can improve mortar strength Optimal mortar formulation incorporated 50% of APCr. Potential application of the mortar as road-sub-base aggregate.
	(Dimech, et al., 2008)	Glass-ceramic product obtained by sintering of washed APCr. It has high density, structural integrity and appropriate hardness to be used in construction products.
Glass-ceramics materials	(Lee, et al., 1999)	Two fractions obtained from sintering APCr One containing the vaporised and condensed heavy metals. And a major fraction that is more stable. Energy efficient process because the thermal treatment occurs at just at 850°C.
Ceramic bricks	(Zhang & Qi, 2012)	Fired bricks incorporating 25% of APCr with compressive strength from 19 to 24 MPa. The leaching of heavy metals was reduced to below 1.9% in the bricks.

	(Hwang, et al., 2012; Quina, et	Hot-bonded LWA with technological characteristics comparable to commercial
	al., 2006; Quina, et al., 2014a;	products, such as Leca [®] and Lytag [®] , and low leaching properties.
	Quina, et al., 2014b; Ayati, et al.,	
LWA	2016; Han, et al., 2022)	
	(Gunning, et al., 2009; Gunning, et al., 2015; Gunning, et al., 2011a; Gunning, et al., 2011b)	Cold-bonded LWA with high performance in different applications. Carbon negative product. Require additives apart from APCr.

2.5.1 Lightweight aggregate (LWA)

Lightweight aggregate as defined in (BS EN 13055, 2016), is a granular material of mineral origin, whose maximum value for particle density is 2 g/cm³ or for bulk density is 1.2 g/cm³. The main technical advantages of LWA are low-density, thermal and sound insulation, fire resistance, minimum water absorption, resistance against extreme pH conditions and high durability (Quina, et al., 2014a). Table 2.6 summarises the classification and applications of LWA according to their purposes and characteristics (Quina, et al., 2014a; Mahmad Nor, et al., 2016).

Table 2.6 Classification and applications of LWA.

Type of LWA	Applications	Purpose	
	Lightweight structural concrete		
Structural	Geotechnical fillers	Maintaining	the
Structural	Construction of drainage systems,	compressive str	rength
	roofs gardens and filters		
Mansory	Concrete masonry units	Reducing the d	ensity
Inculating	Ultra-lightweight concrete	Thermal	insulating
insulating	Concrete not exposed to the weather	properties	

According to their origin, there are three types of LWA (BS EN 13055, 2016):

- 1. Natural LWA: aggregates from mineral sources, that is natural rocks, for example pumice, scoria, tuff, breccias and volcanic cinders, that has been mechanically processed (crushing) (Mahmad Nor, et al., 2016).
- Manufactured (also called artificial or engineered) LWA: aggregates that are produced by means of industrial processes, such as expanding, pelletising or thermally treatment of natural products (like clay, shale and slate) or industrial products (like glass) or industrial by-products (like blast-furnace slag, FA and BA) (Chandra & Berntsson, 2002).

 By-product LWA: aggregates comprising waste material that has been treated by mechanical processing, for instance heavy metal sludge, mining residues, palm shell, paper sludge, pet bottles, sewage sludge, steel slag and marine clay (Mahmad Nor, et al., 2016).

In the production of LWA, the raw materials are mixed and then subjected to an agglomeration process. Agglomeration or pelletising is a method for upgrading fines into larger particles called pellets ("green pellets") by sticking particles together with binders through mechanical agitation (Schaafsma, et al., 1998). There are two types of production process for LWA namely hot bonding and cold bonding. LWA manufactured through the hot bonding method are those in which the green pellets are thermally treated at temperatures ranging from 1,000 to around 1,200 °C (Mahmad Nor, et al., 2016). In contrast, LWA produced through cold bonding are those in which the green pellets are cured at specific conditions of relative humidity (from 70 to 100%) and temperature (from 20 to 100°C) (Tajra, et al., 2019).

2.5.1.1 LWA manufactured through hot bonding

Aggregates produced by hot bonding are also called expanded aggregates; this is because the aggregates are produced by rapid heating at high temperatures of materials that have bloating properties (de' Gennaro, et al., 2004). There are two conditions required in the raw materials to foster suitable bloating in the aggregates (Riley, 1951):

- 1. At high temperatures, the material must produce a glassy liquid phase with high viscosity, so that it can entrap gases.
- 2. The material must contain substances that release gases at the temperature at which the glassy phase is formed.

In addition, during the cooling process, an external glassy layer is developed, making the aggregate surface impermeable, uniform and mechanically resistant (de' Gennaro, et al., 2004).

The main natural sources for the production of LWA are clays or shales (de' Gennaro, et al., 2004). In general, the major mineral constituents of clay materials include mica-illite,
chlorite, kaolinite, vermiculite and smectite, among others, while other phases called nonclay minerals are found in small quantities, for instance, silica minerals, feldspars, zeolites, carbonates, iron oxi-hydroxides and sulphates (Moore & Reynolds Jr, 1989). The presence of some of these components influences the bloating capacity of the clay; this relationship was elucidated by Riley (1951) by means of a composition diagram (Riley diagram), that considers the main oxides present in the clay: SiO₂, Al₂O₃ and fluxing (See Figure 2.12). The fluxing parameter is the sum of Fe₂O₃, CaO, MgO, Na₂O and K₂O. From these materials, the gases associated with the expansion at high temperatures can be water vapour, CO and CO₂ from the combustion of organic matter, CO₂ due to the decomposition of carbonates, O₂ and CO₂ produced from the reduction of Fe⁺³, SO_x from sulphide oxidation, as well as F and Cl from different clay minerals (Heller-Kallai, et al., 1998; de' Gennaro, et al., 2004).



Figure 2.12. Riley diagram for clay and for APCr as reported in the literature. Modified from (Quina, et al., 2014b).

Waste materials such as coal fly ash or slag (Aineto, et al., 2005) or sewage sludge (Latosinska & Zygado, 2011) can be added to the clay in order to promote bloating as they exhibit expanding properties when treated at high temperatures in oxidising

conditions. In this case, the gases causing expansion can be mainly CO and CO₂. The most widespread LWA products on the market are Leca[®] (Lightweight Expanded Clay Aggregate) that is produced from expanded clay (Leca, 2022) and Lytag[®] which is manufactured from coal fly ash (Aggregate Industries, 2022).

In accordance with the revision presented in Section 2.4.4, thermal treatment of APCr can stabilise the residue for further use in the production of ceramic materials, as are LWA. The partial replacement of natural clays in the production of LWA by the addition of APCr has been investigated by different authors at a laboratory scale (Hwang, et al., 2012; Quina, et al., 2014a; Ayati, et al., 2016; Quina, et al., 2014b; Han, et al., 2022) and the results obtained up to this time have demonstrated that this approach could be a promising alternative option for recycling APCr and producing good quality construction materials.

In a theoretical study, by means of a Riley diagram (Figure 2.12), it was found that the maximum incorporation of APCr in LWA was 5% because the main oxides compositions of these residues are distant regarding the bloating area (Quina, et al., 2006). Therefore, the amount of APCr that can be incorporated into the LWA structure is the main challenge to overcome, given the lack of bloating properties and the high leaching rates. Thus, subsequent studies have been focused on increasing the percentage of incorporation of APCr by adding bloating agents and treating the residue to improve its leaching characteristics.

It was demonstrated that LWA manufactured by sintering a mixture of reservoir sediment and MSWI FA containing APCr can be used for production of self-consolidating lightweight concrete, with good quality and corrosion resistance, as long as the maximum content of MSWI FA does not exceed 30% of the total mix (Hwang, et al., 2012). In a study at a laboratory scale, Quina, et al. (2014b) evaluated the possibility of producing LWA with commercial technological properties from natural industrial clay and APCr, concluding that the inclusion of the ash does not improve the technological characteristics of the final product, although this approach could be feasible if the percentage of APCr incorporation is low (maximum 5%) and a pre-treatment of the waste is performed. In other work (Quina, et al., 2014a), the same authors adapted the industrial production of LWA to a laboratory scale by pelletising natural clay, APCr (as-received or washed), a small quantity of oil and water, and further sintering in a muffle furnace, following the process shown in Figure 2.13. The technological characterisation of the produced aggregates revealed that, despite no technical advantages were observed in the products, APCr can be incorporated into LWA manufacture after a washing pre-treatment and at additions as low as 5% of the total mix; also the leaching of heavy metals from the LWA was not significant, meaning that its impact on the environment would be the minimum. A method to produce LWA by pelletising washed APCr mixed with sodium silicate followed by clay coating and sintering at 1,150°C was developed, resulting in a final product with acceptable technological properties and incorporation of up to 30% APCr; also, the leaching of contaminants was reduced below the EU limit values for hazardous and non-hazardous waste (Ayati, et al., 2016). In a more recent study, ultra-LWA (bulk density < 0.5 g/cm³) were produced from natural clay, APCr and the addition of silicon carbide SiC as a bloating agent with further sintering; under optimal conditions (0.3% SiC and 1,120°C for 30 min) and fixed proportions of 30% APCr and 70% clay, the final product exhibited properties meeting the Chines LWA standard, while the heavy metals leaching was significantly reduced and met the Chines MSWI FA utilisation standard (Han, et al., 2022).



Figure 2.13. Flow diagram of LWA production from APCr by hot bonding at laboratory scale (Quina, et al., 2018).

2.5.1.2 LWA manufactured trough cold bonding

The cold bonding method for the manufacture of aggregates for recycling of industrial wastes exhibits different advantages compared to hot bonding, mainly a minimum energy consumption, reduction of pollutants emissions (Tajra, et al., 2019; Gunning, et al., 2009) as well as low investment requirements (Tajra, et al., 2019). However, information about the use of MSWI residues for the manufacture of cold-bonded LWA is limited. A recent review on the production and properties of cold-bonded aggregates (Tajra, et al., 2019) shows that the most used raw material for research on the production of cold-bonded LWA is fly ash from different origins. The most commonly used binders are PC and/or lime. Only 4 out 42 studies cited in that review refer the use of MSWI residues: fly ash by (Colangelo, et al., 2015) and bottom ash by (Cioffi, et al., 2011; Tang, et al., 2017;

Tang & Brouwers, 2018), while no paper refers the use of APCr as raw material. Certainly, the incorporation of APCr into cold bonded LWA has been only reported in the UK (Gunning, et al., 2011a; Gunning, et al., 2011b). Some details of the studies that used MSWI residues for the production of cold bonded LWA are presented in Table 2.7.

Table 2.7. Use of MSWI residues and binders for cold bonded LWA production.

		0/ Down			Characteristic of the LWA		
Reference	Raw material	material	Binder	% Binder	Strength (MPa)	Water absorption (%wt)	
(Gunning, et al., 2009)	Quarry fines	50 - 90	MSWI FA MSWI BA	$\begin{array}{c} 10-50\\ 10-50 \end{array}$	0.17 (50% MSWI FA)	27.8 (50% MSWI FA)	
(Gunning, et al., 2011b)	MSWI FA/APCr	NA	Sand + Cement	NA	0 - 20	29	
(Gunning, et al., 2011a)	APCr	NA	Non-specified additives and stabilisation by ACT	NA	NA	19.6	
(Cioffi, et al., 2011)	MSWI BA	70 80 90 70 70	CEM II 42.5R CEM II 42.5R CEM II 42.5R Lime CEM II 42.5R – Lime	$30 \\ 20 \\ 10 \\ 30 \\ 10 - 20$	1.5 3.5 2.5 2.5 2.8	11 * 12 13 11 12	
(Colangelo, et al., 2015)	MSWI FA	70 70	CEMII/A-L42.5R Lime	30 30	3.8 - 5.1 2.3-3.0	9.7-11.2 * 11.9-12.9	
(Tang, et al., 2017)	MSWI BA fines Washing aggregate sludge Paper sludge ash + FA	20 - 80 5 5 - 65	CEM I 42.5 N (OPC)	10	4.4	14.8 *	
(Tang & Brouwers, 2018)	MSWI BA fines Washing aggregate sludge Paper sludge ash + FA	40 - 75 5 10 - 45	CEM I 42.5 N (OPC)	10	9.6	19 *	

*Approximate values, NA = non-reported values

An alternative way to incorporate MSWI residues in the manufacture of LWA through cold bonding is the pre-treatment of the wastes by carbonation, which takes advantage of the high tendency of thermal residues to react with CO₂ producing carbonates, as described in Section 2.4.2.1. The approach of combining carbonation with pelletisation was first proposed by Gunning, et al. (2009) for valorisation of alkaline thermal residues, producing aggregates whose properties are comparable to commercially available LWA. In that research Accelerated Carbonation Technology (ACT) was used to stabilise the residues, which were then mixed with binders, pelletised and cured to obtain the final hardened products.

Gunning, et al. (2011b) conducted some laboratory scale trials to produce cold bonded LWA from different industrial wastes, including MSWI fly ash which contained APCr; the set-up consisted of a 5 L drum pelletiser, connected to a CO₂ gas delivery system, and fed by the premixed formulation. The mix formulation was MSWI fly ash/APCr, sand, PC and water. The resulting aggregates were cured for about 48 hr in a chamber receiving a continuous flow of CO₂ gas. In a subsequent work (Gunning, et al., 2011a), the process was scaled up to an industrial level using APCr as the main raw material, pure CO₂ for stabilisation of leachable contaminants and additives to produce a mixture which was pelletised in the presence of pure CO₂ (See Figure 2.14). The leaching rates of the resulting LWA regarding to the untreated APCr were significantly reduced: Pb leaching was reduced by two orders of magnitude, resulting in lower than inert WAC limits, and Cu and Hg leaching was reduced to below detection limits. Based upon the performance of these trials, the cold bonded LWA made of APCr have been produced at an industrial scale and commercialised by the Carbon8 company (Carbon8, 2022).



Figure 2.14. Flow diagram of LWA production from APCr by cold bonding at industrial scale. Modified from Gunning, et al. (2011a).

2.6 Life Cycle Assessment (LCA) of APCr management

Literature about LCA of APCr management options is limited and the recommendations derived from these assessments are not definitive considering the differences in geographical locations, technologies and LCA methods. The LCA of five different APCr management options (landfilling without treatment, disposal after neutralisation, disposal in salt mines, ferrox treatment and asphalt production) showed that global warming, human toxicity through soil and water, and ecotoxicity in water were the categories with the highest impact during the first 100 years; after this time, the heavy metals were predicted to still remain in the residue matrix, given to its long-term leaching properties (Fruergaard & Astrup, 2007). In other research work, LCA was performed on seven APCr management options: landfilling without treatment, backfilling in salt mines, neutralisation of acid waste, filler replacement in asphalt, ferrox treatment, vitrification and melting with automobile shredder residues. The results showed that vitrification and melting processes contributed the highest impact to global warming due to high energy consumption; also, all treatment alternatives were better than landfilling without treatment in terms of ecotoxicity and human toxicity, because they diminish the leaching properties of the residue, although the thermal treatments showed slightly higher impacts associated to air emissions (Fruergaard, et al., 2010).

The environmental effects of LWA manufactured by sintering natural clay with and without APCr incorporation were evaluated by means of LCA in a recent study (Quina, et al., 2020). The results showed that the impacts in all environmental categories decreased when 3% of the natural clay was replaced by APCr, achieving the most significant reductions in ecotoxicity and human toxicity due to the avoidance of APCr landfilling. For the non-toxicity categories, the major impacts were associated with the emissions generated from the thermal treatment, independently of the presence or absence of APCr in the raw material; consequently, the APCr incorporation into LWA in these categories did not represent a significant reduction.

Maresca, et al. (2022) used LCA to investigate the environmental impact of the various APCr management technologies, focusing on the possibility of cascading management, that is, applying different consecutive treatments. The assessment of different scenarios showed that increasing the processing and cascading management of the APCr contribute to the uncertainty of the results in contrast to landfilling, therefore, this approach may not always reduce the environmental impacts. The authors remark on APCr stabilisation through carbonation followed by incorporation into the production of cement-based aggregates for use in construction as one of the management options with lower environmental impacts and smaller uncertainty in the results.

2.7 Summary

A thorough literature review was conducted, addressing the topics of waste incineration and APC technologies, APCr characterisation, treatment and recycling potential. The main highlights from the revision and their impacts on this research are summarised in the following points.

 Physical characteristics, chemical composition and environmental properties of APCr vary substantially depending on different factors, including original waste composition, waste incineration technologies, APC systems, and location of the APCr producers. To evaluate the characteristics of APCr produced in the UK, it is required that several samples are taken from different sources across the country to ensure their representativeness and to properly demonstrate their variability.

- Data about the chemical composition of MSWI APCr reported by different studies was collated in this research to have an insight into the variability and complexity of the waste. It was found that the major fraction in APCr comprises inorganic substances, which are associated with its environmental impact, due to the high contents of leachable substances containing toxic heavy metals, such as Zn, Pb, Cr, Cd and As, and soluble salts, including sulphates and chlorides. The chemical characterisation of APCr in this study was required to elucidate its extent of variability, identify the concerning substances, establish relationships with the APC technologies and propose treatment options that effectively mitigate the environmental impact of the waste.
- Currently, in the UK, APCr management comprises a treatment by stabilisation/solidification (S/S) with chemical additives and further landfilling. This treatment has the drawback of increasing the volume of the waste, which results in additional costs due to transportation and disposal of the treated waste. Also, there is a potential increase in the costs of treating APCr as per changes in the legislation. Therefore, in this study, the alternative treatment options of washing and carbonation, were proposed to enable APCr for further recycling into LWA, and thus, diverting the waste from landfilling.
- The treatment techniques for APCr can be grouped into 4 categories: extractionseparation, chemical stabilisation, solidification and thermal treatment. The principles of operation, advantages, disadvantages and possible combinations of these techniques were described. The APCr treatment and LWA manufacture proposed in this study took advantage of the benefits derived from each technique. The extractionseparation by washing with water was applied to remove leachable contaminants from APCr enabling the material for subsequent recycling. The chemical stabilisation was applied by accelerated carbonation of APCr and further solidification with lime in the cold bonding LWA manufacture. The thermal treatment was applied in the LWA manufacture through hot bonding to remove or encapsulate contaminants.
- The water requirement and the extraction or reaction time are determining factors in the treatment of APCr by washing or carbonation. These conditions have been optimised by different studies at the laboratory scale to lessen the water consumption

and the extraction or reaction time as much as possible without detriment to the efficiency of the treatments. Structured matrices, based on previous studies, were designed for the optimisation of the L/S ratio as a measure of the water requirement and the extraction time for washing with water, and % of moisture and reaction time for accelerated carbonation.

- Different studies have demonstrated that substantial modifications in the chemical and physical characteristics of APCr took place because of treating the residue by washing with water as well as by carbonation. Those changes were evaluated in this study by analysing the raw and the treated APCr for elemental composition, mineralogy, microstructure and leaching behaviour.
- The potential recycling of APCr into construction materials has been addressed by several studies. From the different products potentially manufactured from APCr, the artificial LWA is a viable option to develop given the demand and positive impact of this type of aggregates in the construction industry, given their technical advantages, such as high thermal and acoustic insulation, high strength and good resistance to fire. The benefits of incorporating APCr into LWA include reducing the need for landfilling of hazardous waste, helping to reduce the extraction of natural resources and creating new value-added products. This study aimed to evaluate the LWA manufacturing from APCr as a more sustainable and cost-effective option for recycling/recovery of this waste material.
- There are two ways for LWA production, hot bonding (sintering of pellets at 1,000 1,200 °C) and cold bonding (curing of pellets at relative humidity from 70 to 100% and temperature from 20 to 100°C). Some studies on LWA production from APCr by hot bonding have been conducted in different countries, but there is a lack of research on this method in the UK. Recycling of APCr through cold bonding has been only reported in the UK and the products are currently commercialised. In this study, both LWA manufacture routes, incorporating APCr, were tested and compared in terms of raw materials requirements, maximum APCr incorporation, optimisation of manufacturing procedures, chemical and technological properties of the final products, performance of the LWA in concrete manufacture and analysis of embodied carbon which comprised the estimation of energy consumption and reduction of pollutants emissions.

3 Chapter 3 Methodology

3.1 Sourcing, sampling and initial processing of material

3.1.1 APCr sampling and sub-sampling

The APCr samples for this research were taken from Augean's² inputs in two sampling stages: from November 2019 to March 2020 (32 samples) and from August to September 2020 (10 samples). In total, 42 APCr samples coming from 22 different EfW plants located throughout the UK were collected. These plants treat mainly MSW.

The average sample size was 667g and the total amount of collected material was 22.3 kg. The as-received samples were codified to keep the identities of the producers on anonymous basis, using the codes from APCr-01 to APCr-42. The descriptions of appearance, consistency and odour of all the APCr samples are detailed in Appendix III and some representative samples can be observed in Figure 3.1. Most of the samples were fine or grained powders with colours ranging from dark to light grey or from dark to light beige. There were some samples consisting of wet or dry lumps that could easily be broken by pressure. Particles of unburnt carbon or activated carbon added during the gas cleaning process were observed. Some samples were odourless, while others emitted a strong odour corresponding to ammonia that is one of the additives used for the neutralisation of the flue acid gases.

² Augean is a company in the UK that specialises in managing hard to handle waste, including APCr from EfW plants, by means of an appropriate waste treatment and disposal infrastructure (Augean, 2022).



Figure 3.1. Some as-received APCr samples.

The method of coning and quartering (BS 3406-1, 1986) was used to obtain a representative sub-sample from each one of the APCr samples, as follows: the total amount of sample was poured on a tray, so that it took a conical shape, then, it was gently flattened with a spatula into a "cake", which was divided into four equal parts, as shown in Figure 3.2. The two quarters in opposite positions were replaced in the sample container, while the other two were combined to make up the sub-sample. This process was replicated as many times as it was needed until collecting a sub-sample of 100 g. All sub-samples were dried at 105°C for 24 hours. Approximately 20 g of each dry sub-sample were manually ground. Then, the dry and dry-ground sub-samples were kept apart in airtight plastic tubes for further analysis. The remaining amounts of as-received samples were stored in airtight plastic buckets at ambient conditions for subsequent use.



Figure 3.2 Coning and quartering of an APCr sample.

3.1.2 Preparation of a composite sample of APCr

A composite sample, referred to APCr-mix, was prepared by mixing the 42 individual APCr samples at different proportions, as shown in Table 3.1. These proportions were determined according to the available quantity of each sample and proportional to the APCr producers' outputs. This mixture is considered representative of the APCr generated in the UK, because it comprises the outputs of 22 different EfW plants located throughout the country. APCr-mix was the sample used for all tests of APCr treatments and LWA production.

Sample A	wt% in	Sampla	wt% in	Sample	wt% in	
	APCr-mix	Sample	APCr-mix	Sample	APCr-mix	
APCr-01	2.3	APCr-15	1.2	APCr-29	1.2	
APCr-02	2.1	APCr-16	1.6	APCr-30	2.4	
APCr-03	1.9	APCr-17	1.4	APCr-31	2.3	
APCr-04	1.8	APCr-18	0.7	APCr-32	1.5	
APCr-05	1.9	APCr-19	1.1	APCr-33	2.0	
APCr-06	1.1	APCr-20	0.9	APCr-34	2.2	
APCr-07	2.4	APCr-21	0.5	APCr-35	1.3	
APCr-08	0.9	APCr-22	0.5	APCr-36	1.0	
APCr-09	1.4	APCr-23	1.9	APCr-37	2.5	
APCr-10	1.6	APCr-24	2.5	APCr-38	3.1	
APCr-11	2.5	APCr-25	3.3	APCr-39	3.4	
APCr-12	1.2	APCr-26	2.0	APCr-40	2.2	
APCr-13	1.0	APCr-27	2.6	APCr-41	3.1	
APCr-14	0.9	APCr-28	4.0	APCr-42	25.0	
Total wt%:	21.9	+	24.8	+	53.3	= 100
				Total weight	t of material:	20 Kg

Table 3.1 Mix proportions of individual samples in the composite APCr sample.

3.1.3 Clay sampling and sub-sampling

The samples of clay were taken at King's Cliffe by digging the ground. Six samples were collected, counting for a total of 50.6 Kg of wet material (See Figure 3.3).



Figure 3.3 As-received clay samples.

The clay was initially observed to describe its physical characteristics, including colour, appearance, odour, consistency, texture and presence of additional material. Table 3.2 presents a summary of these characteristics. The as-received clay lumps were manually crushed, to 7 cm pieces. Additional material, such as, stones and pieces of vegetation were removed. All the clay material was dried at 105°C during 48 h. Then, it was milled in a disc mill at a particle size less than 150 μ m. The milled clay was mixed and saved for further use in a plastic bucket, which was stored in a dry place. The total amount of dry-milled clay, referred to clay, was 37.6 Kg.

Table 3.2 Physical description of clay.

Characteristic	Description		
Colour	In general, all samples show dark colouring, including dark grey, dark beige and black. Some green sheens can be shown in portions, especially in the samples containing biggest lumps. When the medium and large lumps are crushed, it is observed a material coloured in dark green with some areas in brown. Other lumps showed beige particles.	Different colouring	
Appearance	Thick, highly moisturised powder with presence of agglomerates, small, medium and large lumps, some vegetation particles like sticks and branches. Small lumps are from 0.5 to 5 cm diameter; medium lumps are from 5 to 10 cm diameter: large lumps are from 10 to 15 cm diameter. Manual crushing of the material evidenced the presence of presence of stones (from 2 to 20 cm diameter); also, the dark black samples are wet and more difficult to be crushed into small lumps.	Large Large Lump	
Texture	Irregular, oily	div Lunps	
Consistency	Small and medium lumps are easy to break manually or with a spatula	soft lumps	
Odour	Imperceptible odour at first, but an organic matter characteristic odour is detected in some of the sample more than in others		

3.1.4 Water content and Loss on Ignition (LOI) of APCr and clay

The APCr and clay samples were analysed for water content by drying at 105°C for 24 hr (BS EN 15934, 2012). The water content was calculated as the percentage of loss of mass of water according to Equation 3.1. This parameter is used to calculate the dry matter fraction of a sample. It must be noted that volatile compounds that evaporate at 105°C or less are measured as water during this test. The sample size for analysis of water content was 100g or 50g for APCr, depending on the available amounts of as-received samples, while for clay, 100g were taken randomly from each of the six samples of clay.

Equation 3.1

Water content =
$$\frac{W_i - W_f}{W_i} \times 100$$

Where W_i is the weight of the wet sample and W_f is the weight of the sample dried at 105°C.

LOI is an estimation of the content of organic matter in a sample. It is defined as the mass fraction lost by means of incineration of a dried sample at a specified temperature (BS EN 15935, 2021). For analysis of LOI, 20 g of dry sample (APCr or clay), previously tested for water content, was fired in a muffle furnace at temperature of 550°C for 4 hours (BS EN 15935, 2021). The clay was analysed by triplicate. The LOI was calculated as a percentage of weight loss due to the thermal process according to Equation 3.2.

Equation 3.2

$$LOI = \frac{W_i - W_f}{W_i} \times 100$$

Where W_i is the weight of the dry sample before firing and W_f is the weight of the sample fired at 550°C.

The results of water content and LOI for clay are shown in Table 3.3. The results of both parameters for APCr will be reported in Chapter 4 as part of the APCr characterisation.

Clay sample	Water content %
1	17.55
2	14.90
3	19.87
4	12.73
5	16.57
6	17.74
Average	16.56 ± 2.48
Replicate	LOI %
A	3.80
В	3.80
С	3.82
Average	3.81 ± 0.01

Table 3.3 Water content and LOI of clay.

3.2 Total contents of metals and leaching tests

The landfill Waste Acceptance Criteria (WAC) sets the minimum testing requirements that need to be met for the disposal of waste in the relevant landfill. The WAC considers the composition as well as the leachate analysis of the waste to determine the degree of contamination that could potentially leach out from the material once it is landfilled (The Council of the European Union, 2003). The leaching properties of heavy metals and soluble salts from APCr can cause pollution to the soil as well as to the surrounding sources of groundwater or surface water bodies.

3.2.1 Total contents of metals

For analysis of the total contents of metals, every sample was dried overnight at 105° C. After this time, the sample was weighed into a digest vial and extracted with Aqua Regia, which is a mix of concentrated hydrochloric acid HCl (35%) and concentrated nitric acid HNO₃ (65%) at proportion 3:1, respectively) on a hot plate for 2-4 hours, according to (BS EN 13657, 2002). The objective of this pre-treatment is the oxidation of the organic

matter of the sample and the dissolution of its mineral fraction. The liquid extract was then diluted and analysed by ICP/OES for the metals listed in Table 3.4. This procedure was applied for the analysis of the total contents of metals in APCr (untreated and treated), clay and LWA.

3.2.2 Leaching tests

Leaching tests were performed to quantify the release of soluble substances from APCr to water over a long period of time, according to the granular waste compliance leaching test (BS EN 12457-2, 2002). For leaching test, around 150 g of dry sample was ground and sieved through a 4 mm aperture diameter sieve, then, 90 g of material passing the sieve were placed into a 21 polyethylene plastic bottle, in which 900 mL of deionised water were added, so that the liquid/solid ratio (L/S) was 10 L/Kg. The bottles had been previously rinsed with 0.1 M HNO₃ and deionised water. The bottle containing the mix was placed in an overhead shaker Hedolph Reax 20 (See Figure 3.4), in which the extraction was done by constant rotation at speed of 10 rpm for 24 hours.



Figure 3.4 Overhead shaker used for leaching tests and APCr washing.

After agitation, the bottles were removed from the shaker and left to stand for 15 min to allow settlement of the solid particles. Then, the liquid fraction (leachate) was passed through a 0.45 μ g pore size membrane with help of vacuum (See Figure 3.5). The filtering membrane was previously rinsed with 0.1 M HNO₃ and deionised water.



Figure 3.5 Vacuum filtering of leachate in a leaching test.

Two leachate portions of 100 mL each were kept under refrigeration for subsequent analyses, one acidified at pH lower than 2.0 for analysis of metals and other without any preservative for analysis of salts. Table 3.4 details the analytical techniques used for analysis of different parameters, which are the soluble substances regulated by the WAC. The pH and temperature were measured as soon as the leachates were obtained.

Table 3.4 Parameters and techniques for APCr characterisation according to BS EN 13657, 2002 and leachate analysis of according to BS EN 12457-2:2002.

Parameter		Technique	
Metals	Arsenic as (As)		
	Barium as (Ba)		
	Cadmium as (Cd)		
	Chromium as (Cr)		
	Copper as (Cu)		
	Mercury as (Hg)	ICP/OES (Inductively coupled plasma -	
	Molybdenum as (Mo)	optical emission spectrometry)	
	Nickel as (Ni)		
	Lead as (Pb)		
	Antimony as (Sb)		
	Selenium as (Se)		
	Zinc as (Zn)		
Soluble salts	Chloride as (Cl ⁻)	High Dorformance Liquid Chromotography	
	Fluoride as (F ⁻)	(HBI C)	
	Sulphate as (SO ₄ ²⁻)		
Total Dissolved Solids (TDS)			
рН @ 20 °С		Potentiometry	
Conductivity @ 20°C			
Dissolved Organic Carbon (COD)		Combustion analysis with Non-dispersive	
		Infrared Detection (NDIR)	

3.2.2.1 Regulatory values for leaching substances

Table 3.5 shows the maximum concentrations accepted for the different metals, salts and total dissolved solids (TDS) in the liquid fraction resulting from the leaching analysis of granular waste as per the Landfill Directive (The Council of the European Union, 2003).

Table 3.5 Leaching limit values in mg/kg as expressed in Landfill Directive for waste acceptance at landfills, calculated at 10 L/kg for total release.

Domonostores	Inert	Non-hazardous	Hazardous
Parameters	Waste	waste	waste
Arsenic, As	0.5	2	25
Barium, Ba	20	100	300
Cadmium, Cd	0.04	1	5
Chromium, Cu	0.5	10	70
Copper, Cu	2	50	100
Mercury, Hg	0.01	0.2	2
Molybdenum, Mo	0.5	10	30
Nickel, Ni	0.4	10	40
Lead, Pb	0.5	10	50
Antimony, Sb	0.06	0.7	5
Selenium, Se	0.1	0.5	7
Zinc, Zn	4	50	200
Chloride	800	15,000	25,000
Fluoride	10	150	500
Sulphate	1,000	20,000	50,000
Total Dissolved Solids	4,000	60,000	100,000
DOC	500	800	1,000

3.3 Mineral characterisation of APCr, clay and LWA

Pollutant elements that form the chemistry of APCr are associated to crystalline phases, as it was discussed in section 2.2. Crystalline phases have a periodic arrangement of their

atoms or molecular chains (Venkateshaiah, et al., 2016). X-Ray powder diffraction (XRD) is a well-established analytical technique for the study of crystalline structures and atomic spacing, which is based on irradiation of the sample with X-Rays and detection of the diffracted rays (Dutrou, 2020; Venkateshaiah, et al., 2016). The theory about X-ray generation, X-ray diffraction and its application to qualitative and quantitative analysis of minerals are briefly discussed in the following paragraphs based on specialised publications (Ewald, 1962; Moore & Reynolds Jr, 1989; Chrysochoou, 2013; He, 1954).

X-rays are a form of electromagnetic radiation with a wavelength in the range from 0.01 to 100 Å. They are produced when electrically charged particles are decelerated as they pass through an electrical field. In an X-ray tube, a high voltage is maintained across the electrodes to direct the electrons towards a metal target (anode). X-rays are produced at the point of impact and propagate in different directions. The typical X-rays used in XRD are produced in tubes with Cu targets, which generate their strongest radiation (K α) at wavelengths around 1.5 Å, which is comparable to the range of interatomic spacing in mineral phases. When a monochromatic X-ray beam hits a crystal lattice, besides absorption and other phenomena, it produces scattered X-rays with the same wavelength of the incident beam. Diffraction occurs when the scattered X-rays in a certain direction are in phase with the scattered X-rays from other atomic planes. The scattered X-rays are not evenly distributed in space, but as a function of the electron distribution in the sample. Therefore, the intensities and spatial distributions of the scattered X-rays form a specific diffraction pattern that is uniquely determined by the crystalline structure of the sample.

Bragg's law, illustrated in Figure 3.6, describes the diffraction of X-rays by a crystal. Two incident X-ray beams, which are in the same phase, strike the crystal planes with an incident angle θ and a reflection angle θ . The first X-ray is reflected from the atom in the top layer, while the second X-ray reflected from the atom in the bottom layer travels an extra distance (2dSin θ). To remain in the same phase and produce a detectable constructive interference, the extra distance travelled by the second ray must be equal to an integer number of the incident beam wavelength (n λ). Thus, the diffraction peak is observed when Bragg condition is satisfied, as per Equation 3.3. This condition allows a maximum intensity that enables the measurement of the characteristics of a crystal structure. The diffraction patterns from crystals have sharp peaks corresponding to

various crystal planes based on the Bragg's law. To satisfy this law at all crystal planes, the diffraction pattern is generated from polycrystalline materials or powder materials.

Equation 3.3

$$d(\sin\theta) = n\lambda$$

Where:

d is the distance between each adjacent crystal planes (d-spacing)

 θ is the Bragg angle at which a diffraction peak is observed

n is an integer number called order of reflection

 λ is the wavelength



Figure 3.6 Geometry of X-ray diffraction for demonstration of Bragg's law. Modified from (Encyclopædia Britannica, 2023)(Accessed 10 Jun 2023).

The method more widely accepted for structural analysis of crystalline materials is the Rietveld refinement, invented by Hugo Rietveld. This method consists of a structure refinement that obtains a least-squares fit between a calculated profile and the measured intensities at equal angular intervals on a powder diffractometer (Döbelin, 2015; Dinnebier & Kern, 2015). The integration of this method to a software allows the refinement of various profile properties, such as lattice parameters, peak width and shape and preferred orientation, to generate a calculated diffraction pattern (Rigaku Corporation, 2023). The calculated pattern elucidates different properties of the analysed sample, including accurate quantitative information.

Quantification of mineral phases in XRD is complex because the analysis accuracy is affected by the matrix effects. There are different approaches to address this analysis by XRD, such as, the single peak method, external standard, and internal standard, among others (Dinnebier & Kern, 2015; Zhou, et al., 2018). The method of the internal standard is widely used and consists of adding a mineral (internal standard) to the sample during the XRD measurement. This method is based on peak intensities ratios from the phases on the original sample to the intensity of a single reflection from the internal standard, which has been added in a known proportion to the sample (Moore & Reynolds Jr, 1989). The formula for calculation of a phase content in a sample by the method of the internal standard in XRD is as given in Equation 3.4 (Zhou, X., 2018):

Equation 3.4

$$%P = k \frac{I_p}{I_s}$$

Where:

%P is the mass percentage of phase P

k is a coefficient related to the nature of the phase P and the internal standard S, and to the amount of standard added to the sample

I_p is the X-ray diffraction intensity of the phase P

Is is the X-ray diffraction intensity of the standard S

The method of the internal standard eliminates the matrix effects because the experimental conditions become the same for the sample and the standard. Thus, the Ip/Is value remains invariable with different sample compositions and under different analysis conditions. Minerals used as internal standards usually have only a few XRD reflections and at least one conveniently located strong reflection (Zhou, et al., 2018).

On the other hand, amorphous materials are compounds without long-range order and periodicity (Hu, et al., 2017; Vessal, 1997). The internal standard method is particularly used when the crystalline and amorphous regions overlap. To define the boundaries of these regions, the areas are integrated into a diffraction profile, and profile refinement is used to elucidate the crystalline and amorphous peaks (He, 1954). The content of amorphous material can be calculated by deducting the percentage of crystalline material from 100% after the analysis of the sample that has been added with the standard.

In this research, for XRD analysis, approximately 1g of ground samples were mounted on an aluminium sample holder. The diffraction patterns were obtained in a Rigaku MiniFlex benchtop X-ray diffractometer equipped with a Cu k- α X-ray tube, which was run at voltage of 40kV and current of 15 mA. All patterns were scanned over the range $3^{\circ} < 2\theta < 70^{\circ}$ with step length 0.015° and speed 2°/min.

The mineral phases were identified by using the Integrated X-Ray Powder Diffraction Software (PDXL) in conjunction with the Crystallography Open Database (COD) (Crystallography Open Database, 2021; Grazulis, et al., 2009). For mineral characterisation of APCr, 205 Crystallographic Information Files (CIF) were downloaded from the COD website and imported to PDXL. These files were selected based on the mineral phases previously identified in APCr according to the literature review presented in Appendix II. For identification of minerals in clay and LWA, the complete COD for PDXL was installed and used.

In this research, the method of internal standard addition was used for the quantification of mineral phases and amorphous material in APCr, clay and LWA. Anatase (TiO₂) has been previously used as internal standard for APCr analysis (Bodénan & Deniard, 2003). However, in this study, anatase was detected in various APCr samples. Therefore, Rutile (TiO₂) was used as internal standard for analysis of APCr. To do this, 1.05 g of APCr was mixed with 0.45 g of rutile (addition at 30%). For analysis of clay and LWA, 0.80 g of sample was added with 0.80 g of corundum (Al₂O₃) (addition at 50%). Corundum is usually used as internal standard for analysis of clay-rich samples (Zhou, et al., 2018). The added samples were analysed by XRD, doing the identification of mineral phases with the CIF files previously obtained from each sample, and finally, the quantification was performed by the Rietveld refinement in the PDXL software. Equation 3.5 was used to calculate the weight fraction of each mineral phase in a sample.

Equation 3.5

$$\%P = \%P_m \times \frac{\%S_k}{\%S_m}$$

Where:

%P is the percentage of a phase P in the sample

 $%P_m$ is the percentage of a phase P measured in the added sample

%Sk is the percentage of the standard S added to the sample

 $\%S_m$ is the percentage of the standard S measured in the added sample

The weight of amorphous material was calculated from Equation 3.6.

Equation 3.6

$$%A = 100 - \sum_{1}^{n} \%M$$

Where:

%A is the percentage of amorphous material in the sample n is the number of mineral phases in the sample %M is the sum of the percentages of all mineral phases detected in the sample

3.4 Analysis of APCr by SEM/EDS

3.4.1 Scanning electron microscopy and energy dispersive spectroscopy (SEM/EDS)

In microscopy, the resolution and hence the quality of magnified images is subject to diffraction effects (Mehta, 2012). Diffraction refers to the bending of waves around narrow openings and obstacles. While the optical wavelengths from ultraviolet (UV) to infrared (IR) spectrum are in the range of hundreds of nm, the electron beam of energy in keV has wavelengths as short as fractions of nm. Therefore, the electron beam is more suitable for imaging due to the dependence on diffraction. Scanning Electron Microscopy is a technique widely used for the microstructural analysis of materials. A Scanning Electron Microscope (SEM) is an instrument that uses electron beams in the range of keV to produce images of objects in the range of μ m to nm with very low diffraction effects (Mehta, 2012). The application of this technique for analysis of APCr allows the assessment of structural characteristics, such as, particle size, particle shape and crystalline and amorphous structures. The combination of SEM with Energy Dispersive Spectroscopy (EDS) allows the analysis of the elemental composition.

The images in an SEM are generated by striking the specimen with a focused high energy beam of electrons that is scanned across the sample surface in a horizontal scanning pattern (Schatten, 2013). This leads to different physical interactions. The electron beam interacts with the sample surface, and the beam electrons interact with the sample atoms (Hercules & Hercules, 2018; Swapp, 2017; Schatten, 2013) These interactions are collectively known as scattering events and include backscattered electrons (BSE), secondary electrons (SE) and X-rays, among others (Goldstein, et al., 2017). SE are generated from the sample itself, yielded by the energy of the incident electron beam. These are low energy electrons with less than 50eV and mostly less than 10Ev (Stokes, 2008). BSE are the incident electrons that bounce around in the sample and then return out of it. Characteristic X-rays are emitted when an electron from an inner shell is removed from the sample by a beam interaction causing that an electron of higher energy fills the inner shell releasing characteristic energy (Schatten, 2013).

Figure 3.7 Part A shows the different interactions between the incident electron beam and the sample. Figure 3.7 Part B shows a diagram that represents the functioning of a conventional SEM. The equipment consists of an electron gun that generates the electron beams and an electromagnetic lens system that guides and focuses the beam. For conventional SEM, an electron gun with a tungsten filament cathode is used as beam electron source. Specific detectors are used to collect each type of electrons produced from the electron beam - sample interactions. The energy, intensity and location of emission of these electrons are used to construct physical images of the sample surface (Mehta, 2012). A secondary electron detector is used for conventional imaging, called Secondary Electron Imaging (SEI), the depth of field produces images that can be interpreted as three dimensional representations (Schatten, 2013). Back scattered electrons (BSE) are reflected from the sample by elastic scattering. Both Back Scattered Electron Images (BSEI) and Secondary Electron Images (SEI) reveal information about the sample surface, like coarse and fine-scale topographic features and crystal structure (Goldstein, et al., 2017). Since the focus of an electron beam can be done at the scale of nanometres, SEM provides a high level of topographical details (Hercules & Hercules, 2018). In addition, the signal intensity of BSE is related to the atomic number of the elements contained in the sample (Schatten, 2013). The analysis of the X-rays with an Energy Dispersive Spectroscopy (EDS) detector allows to obtain an elemental mapping of the sample surface to determine the content of specific chemical elements (Hercules & Hercules, 2018; Goodge, 2017).



Figure 3.7 A: Signals emitted by interactions between the incident electron beam and the sample surface. Modified from (Ezzahmouly, et al., 2019), B: Schematic diagram of a conventional SEM (Ali, 2020).

SEM analysis was used to evaluate the microstructure and elemental composition of APCr. The equipment was a Scanning Electron Microscope JSM-6460LV (JEOL), shown in Figure 3.8. Topography, morphology and particle size of APCr were assessed by means of SEI and BSEI at different magnifications. The analysis conditions were: working distance 8 mm and accelerating voltage 20kV. For elemental analysis, it was used an Xplore EDS detector (Oxford Instruments). The calibration to ensure accuracy of the results obtained was done by using a certified standard of Cobalt. Spectra acquisition was done with the SEM/EDS software AZtecLive.



Figure 3.8 Scanning Electron microscope used for analysis of morphology and elemental composition.

3.4.2 Sample preparation for elemental analysis

As per the extent of the literature reviewed on this research, there is not a reported procedure for the assessment of APCr samples by SEM/EDS; therefore, a complete sample preparation procedure was developed for this material as shown in Figure 3.9.



Figure 3.9. Main stages of APCr sample preparation for SEM/EDS analysis.

A detailed description of the procedure is given below.

Approximately 2 g of each sample was pressed to obtain a "pressed pill", which was further treated by resin impregnation. To do that, the compressed sample was placed in a rubber mounting cup, where it was covered with a mix of bisphenol-A based epoxy resin and hardener (cycloaliphatic polyamine) at proportion 2:1. The embedding of samples into the epoxy resin was performed in a KemVac vacuum impregnation unit, adjusting the inverse pressure at 340 mm Hg for the first hour and 100 mm Hg for 12 hours. This treatment removes the air from the samples and eliminates the gaps between the sample and the resin.

The cured resins containing the samples were manually polished against a silicon carbide grinding paper, grit 320, until exposing the surface of the samples. The sample surface was polished in a modular grinding and polishing system Forcipol 102 according to the three steps indicated in Table 3.6. The diamond suspensions indicated in this table were oily-based suspensions containing polycrystalline diamond particles of angular and blocky shaped grains, which provide numerous cutting facets, which in contact with a solid material produced a polished surface. Some APCr samples, particularly those containing high amounts of residues of carbon, only required polishing until step 2, because a further polishing (step 3) destroyed the compressed sample embedded in the resin.

Step	Polishing surface	Polishing solution	Time (min)
1	Carbide grinding paper (1200)	Isopropanol	10
2	Velvet pad	Diamond suspension with 9 µm of diamond particles size	45
3	Velvet pad	Diamond suspension with 3 μ m of diamond particle size	45

Table 3.6 Polishing conditions of APCr samples for analysis by SEM/EDS.

The polished samples were coated in an Agar Auto carbon coater that uses a controlled carbon-rods evaporation system to produce a transparent conductive layer of about 20nm thickness. In this equipment, a high current (more than 100 mA) was passed through two

carbon rods, under a vacuum chamber. The heat caused the carbon to evaporate and to be deposited as a thin film on the sample surface. This process was performed in a vacuum chamber at minimum reverse pressure of 125 mbar, the voltage was set at 4.2V, the current was 115 mA, and the coating time was 8 seconds. The process was repeated as necessary once the current and vacuum conditions were reached again.

The complete processing of a single sample took about 24 hours. The objective of implementing this procedure was to obtain a compressed sample that exhibits the microstructure of the APCr, which needs to be coated in order to prevent charging under the electron beam. Appendix IV summarises the challenges faced during the development of APCr sample preparation for SEM/EDS. Despite these issues, it was possible to obtain samples with smooth surfaces that resulted suitable for analysis of elemental composition by SEM/EDS as that one shown in Figure 3.10 (A).

All the APCr samples were prepared according to the developed procedure and kept under relative humidity lower than 10% inside a desiccator containing silica gel crystals.

3.4.3 Sample preparation for analysis of particle size and morphology

A piece of double-sided carbon conductive tape was attached to an SEM specimen stub; then, the stub was pressed several times against a certain amount of powdered sample, so that, the powder particles stuck on the surface of the tape. Finally, the sample was coated under the same conditions described for sample preparation for elemental analysis. Figure 3.10 (B) shows the appearance of a carbon-coated powdered sample. These samples were used for the analysis of particle size and morphology by SEM.



Figure 3.10. (A) APCr sample for elemental analysis, (B) APCr sample for morphology and particle size analysis.

3.5 APCr treatment

When a waste material, like APCr, is intended to be recycled into an aggregate substitute for building applications, the leaching of hazardous components to soils and water is a crucial environmental aspect that requires special attention (Van Der Sloot, et al., 2000). Two treatment methods were evaluated in this study, washing with water and accelerated carbonation, both aimed to reduce the leaching properties of APCr and enable this residue for further recycling into LWA. The treatments were conducted on the APCr-mix, referred to as APCr-m.

3.5.1 Water washing

3.5.1.1 Washing procedure

The general steps of APCr washing are shown in Figure 3.11. Based on the washing extraction conditions reported in the literature for the treatment of APCr at a laboratory scale, previously detailed in Table 2.3, the washing was done with deionised water in only one extraction step. The solid material and the water at the prefixed or optimum L/S ratio (see section 3.5.1.2), were placed into 2 L PE plastic bottles with PE lids, previously rinsed with 0.1 M HNO₃ and deionised water. The maximum amount of APCr washed in a bottle was 600 g. For agitation, an overhead shaker Hedolph Reax 20 (the same equipment used for leaching tests, shown in Figure 3.4) was used at a speed of 15 rpm.



Figure 3.11 Flow diagram of APCr washing.

After agitation, the bottles containing the washing mixes were removed from the shaker. Before settlement took place, the liquid component of the mix (wastewater) was passed through a 0.45 μ g pore size membrane with the help of vacuum. The filtering membrane was previously rinsed with 0.1 M HNO₃ and deionised water. The solid residue remaining on the filter, referred to as w-APCr, was transferred to a glass tray and dried overnight at 105°C. Then, the w-APCr was manually ground and saved for further analyses or use in LWA production. The analyses included mineralogical characterisation by XRD, morphology by SEM and leaching test.

3.5.1.2 Optimisation of washing conditions

The extraction efficiency of the washing process was evaluated according to the leaching reduction of chlorides, sulphates and heavy metals. The conditions that are considered to have an effect on the efficiency are the L/S ratio and extraction time. Therefore, these variables were evaluated at three different levels: L/S ratio at 2.5, 5 and 10 L of water per kg of the solid sample, and extraction time at 5, 30 and 60 minutes, according to the matrix presented in Table 3.7. The w-APCr obtained at different L/S ratios and the extraction time were tested for the leaching of metals and soluble salts according to the procedure described in 3.2.1 . The results were analysed and compared with the leaching of the untreated material to determine the percentage of leaching reduction for each group of parameters. The combination of L/S ratio and extraction time that favoured a greater reduction of APCr leaching properties was considered as the optimum conditions of a one-step water washing process. The leaching rates obtained at optimum conditions of washing were compared with the limit values for waste landfill detailed in Table 3.5.

Treatment	L/S ratio	Extraction time (min)
1	2.5	5
2	2.5	30
3	2.5	60
4	5	5
5	5	30
6	5	60
7	10	5
8	10	30
9	10	60

Table 3.7 Washing conditions assessed for treatment of APCr.

3.5.1.3 Characterisation of wastewater

The wastewater streams produced in 3.5.1.2 were characterised in order to consider its further treatment for disposal or potential recirculation. To do that, a portion of about 100 mL of the filtered liquid (wastewater) was immediately tested for pH, and a 25 mL aliquot was analysed for TDS by gravimetry. Two wastewater portions of 100 mL each were kept under refrigeration for subsequent analyses, one acidified at pH lower than 2.0 for analysis of metals by ICP/OES and the other without any preservative for analysis of salts by HPLC. The analysed parameters were the same as listed in Table 3.4 for leachate analysis.

3.5.2 Accelerated carbonation

3.5.2.1 Accelerated carbonation procedure

The general steps of APCr carbonation are shown in Figure 3.12. Based on the conditions reported for accelerated carbonation of APCr in the literature, as detailed in Table 2.4, this treatment was evaluated under ambient conditions for temperature and pressure, and a dynamic flow of CO₂. Deionised water was used to moisten the APCr samples before being treated by carbonation, and soon after this treatment the samples were dried to obtain the final product referred to as carbonated APCr (c-APCr).



Figure 3.12 Flow diagram of APCr accelerated carbonation.

All carbonation tests were carried out in a chamber which consisted of a non-hermetic closed plastic box with a capacity of 5L, as shown in Figure 3.13. A CO₂ atmosphere was created inside the chamber by supplying 99.9% pure CO₂ gas from a pressurised cylinder. The gas was passed through a flowmeter which was directly attached to the chamber (CO₂ inlet). A continuous gas flow was set to 0.1 L/min of CO₂, corresponding to the minimum graduation of the flowmeter. The CO₂ was allowed to pass for 30 sec before starting to

count the reaction time. An outlet hole in the lid of the chamber located in the opposite side of the CO₂ inlet allowed this gas to escape in order to prevent a gradual increase of pressure. Two beakers, each one containing 250 mL of a saturated NaCl solution (40 g NaCl/100 mL water) were placed inside the chamber to control the relative humidity, which was maintained at 75 ± 3 % as registered by a humidity/temperature sensor. The temperature varied as per the laboratory temperature between 20 and 25 °C. The samples were always placed on wide aperture containers or trays at layers of maximum 5 mm to allow maximum exposure of the material to the CO₂. The maximum amount of APCr treated in the chamber in a single batch was 250 g.



Figure 3.13. Carbonation chamber for APCr treatment.

3.5.2.2 Carbonation potential and reaction time

A preliminary test of carbonation of APCr was carried out at 20% and 30% of moisture during a reaction time of 2.5 hours. These operating conditions have been previously reported to be the optimum for carbonation of APCr generated in the UK, as shown in Table 2.4 (Fernández Bertos, et al., 2004c; Li, et al., 2007). The carbonation was tested by triplicate at each moisture contents and the sample size of each replicate was 10 ± 0.0001 g. The quantified variable was CO₂ uptake as the increased percentage in the mass
of the initial amount of untreated APCr, according to Equation 3.7. All the weights were measured on a scale with a precision ± 0.0001 g.

Equation 3.7

$$CO_2 uptake (wt\%) = \frac{W_{car} - W_{unt}}{W_{unt}} \times 100$$

Where:

 W_{car} = weight of carbonated sample in g W_{unt} = weight of untreated sample in g

To determine W_{car}, the carbonated sample was removed from the chamber, dried for 2 hr at 105°C, cooled inside a desiccator and finally weighed.

The results showed that 20% of moisture produced greater CO₂ uptakes in all replicates, as it can be seen in Figure 3.14. Over the period of 2.5 hours, the average CO₂ uptake in wt% at 20% moisture was 12.1 ± 0.4 %, while at 30% moisture, it was 11.0 ± 0.1 %.



Figure 3.14. Preliminary results of effect of moisture on APCr carbonation over 2.5 hr of reaction.

Therefore, the carbonation potential test of APCr was performed at 20% moisture. The objective of this test was to assess the CO_2 uptake over different periods of time to determine the optimum reaction time. To do that, 18 replicates of 10 ± 0.0001 g of

untreated APCr were exposed to CO_2 in the carbonation chamber. From these, three replicates were removed from the chamber, at the reaction times indicated in Table 3.8, until completing 9 hr of carbonation. The carbonation potential of APCr was determined for each period of time, according to Equation 3.7. A control sample was prepared in the same manner and kept under ambient conditions.

Table 3.8 Carbonation conditions for assessment of carbonation potential.

Sample size	Replicates	% Moisture	Reaction time
10 g	18 (3 per each	20	30 min, 60 min, 90 min, 2 hr, 8 hr
	reaction time)		and 9 hr

3.5.2.3 Optimisation of moisture

For optimisation of moisture, samples of 10 ± 0.0001 g of APCr-m were carbonated according to the percentages of moisture and reaction times given in Table 3.9 The moisture percentages proposed in this study (also called water additions or water/solid ratio) have been evaluated for APCr carbonation by different authors in the UK (Fernández Bertos, et al., 2004c; Li, et al., 2007; Araizi, et al., 2016). The CO₂ uptake was determined for the carbonated samples according to the procedure and calculation described in Section 3.5.2.2. The effect of the moisture over a period of 2 hours was evaluated by plotting the reaction time against the CO₂ uptake and the optimum % of moisture was the one producing the highest CO₂ uptake at 2 hours.

Sample size	Replicates*	% Moisture	Reaction time (min)			
	8	10				
10 g	8	20	15 20 45 (0 75 00 105 - 1120			
	8	30	15, 30, 45, 60, 75, 90, 105 and 120			
	8	40				

Table 3.9 Carbonation conditions for optimisation of % of moisture.

*Each replicate was evaluated at one reaction time

In an additional experiment, samples of 10 ± 0.0001 g of APCr-m were added with 10%, 20%, 30% and 40% of moisture and carbonated for 2 hours. Each moisture percentage was evaluated in triplicate. This test was carried out to determine the precision of the results CO₂ uptake at 2 hours of reaction.

3.5.2.4 Carbonation kinetics

The kinetics of carbonation reaction was evaluated over a period of 3 hours (180 min). For this test, 24 replicates of 10 ± 0.0001 g of APCr were prepared at optimum moisture and exposed to CO₂ in the carbonation chamber. To evaluate the CO₂ uptake at different reaction times, two replicates were removed from the chamber at 15, 30, 45, 60, 75, 90, 110, 135, 150, 165 and 180 min. The CO₂ uptake was determined for all the carbonated samples According to the procedure and calculation described in Section 3.5.2.2. Then, the results of CO₂ uptake were plotted against carbonation time to generate the corresponding kinetics model that described the APCr carbonation.

3.5.2.5 Effects of carbonation on chemical composition and leaching properties of APCr

To assess the stabilisation of contaminants in APCr treated by accelerated carbonation, 150 ± 0.01 g of APCr were carbonated under optimum conditions, dried at 105°C for 2 hr, cooled in a desiccator and manually ground in a mortar. The carbonated material was analysed for total contents of metals according to the procedure described in Section 3.2.19, mineralogy according to Section 3.3 and leaching according to Section 3.2.2. The results of c-APCr were compared with those of APCr-m to determine the effects of carbonation on each parameter. The results of the leaching test were also contrasted with the limit values for hazardous waste acceptance at landfill listed in Table 3.5.

3.6 Manufacturing of LWA from APCr

3.6.1 LWA from APCr by hot bonding

3.6.1.1 Procedure for manufacturing hot bonded LWA

The general procedure for the manufacture of LWA from clay and APCr through hot bonding is shown in Figure 3.15. The raw materials, either untreated APCr or w-APCr and clay were mixed with deionised water and manually pelletised into spherical particles. For this research the spherical particles were preferred to facilitate the measurements of their technological properties. Also, it is known that the LWA of spherical shape are preferred in the construction industry because they reduce the costs of concrete production requiring less cement paste to produce workable concrete mixtures than elongated or angular aggregates (Gonzalez-Corrochano, et al., 2011). The pellets were dried overnight at 100°C. The dried pellets were called green pellets, which were then fired to finally produce the LWA. Following the same procedure, LWA containing only clay were produced to compare their technological properties with those of LWA made from APCr or w-APCr.



Figure 3.15 Flow diagram of LWA production from clay and APCr through hot bonding.

3.6.1.2 Optimisation of conditions for hot bonding LWA manufacture

Batches of 500g of LWA were produced according to each formulation and firing temperature detailed in Table 3.10. The percentages of volume of water/weight of solid material (v/wt.), indicated in Table 3.10 correspond to the addition of water which produced a favourable consistency for manual pelletising. For LWA incorporating untreated or washed APCr, the water additions were higher than requirements usually reported in LWA production from waste material which range from 20 to 30 v/wt. % (Mahmad Nor, et al., 2016).

All these formulations were tested for the environmental and technological properties described in Section 3.7. The correlations between each property and the prepared LWA formulations were evaluated to determine the best condition for hot bonding LWA production.

Formulations	% APCr	% w- APCr	% Clay	% (v/wt) Water	Firing temperatures °C
LWA from clay	0	0	100	30	1,100; 1,120; 1,140; 1,160; 1,180; 1,200; 1,220 and 1,230
	5	0	95	32	
LWA from	10	0	90	34	1,100; 1,120; 1,140;
untreated	15	0	85	34	1,160; 1,180 and
APCr	20	0	80	34	1,200
	25	0	75	34	
	0	5	95	38	
	0	10	90	38	
LWA from	0	15	85	40	1,140, 1,160 and
w-APCr	0	20	80	42	1,180
	0	25	75	43	
	0	30	70	44	

Table 3.10. Formulations of LWA made of APCr or w-APCr through hot bonding.

According to the description provided by Quina, et al. (2014b), for the sintering of LWA at industrial scale, it is used a tubular rotary kiln with a tilt angle lower than 5° and comprised of two heating zones: the first zone has a length of 40 m for drying the pellets with a slow rotation and a residence time of 1.8 hours; the second zone has a length of 20 m and is for firing the pellets at temperatures around 1,170°C and residence time is between 8 and 10 min. The time during which the pellets are fired at the maximum temperature is called dwell time. In this study, this process was not simulated, but it was adapted to the laboratory conditions. The pellets were sintered in a muffle furnace SNOL 6.7/1300 over a refractory board coated with a thick layer of Al₂O₃. The temperature rate is illustrated in Figure 3.16 and the maximum firing temperatures are indicated in Table 3.10. The heating rate was defined based on a 2-stages thermal treatment used for the production of LWA from fly ash, in which the first stage is at 750°C for 10 to 15 min and the second is the expansion stage at 1,150°C – 1,175°C for 10 to 15 min (Aineto, et al., 2005). The 2-stage heating is expected to produce more porous LWA with properties similar to commercial LWA compared to a single-stage heating (Mahmad Nor, et al.,

2016). The cooling rate was divided into two segments: initially, a slow rate determined by the cooling capacity of the furnace (around 12° C/min) until 600°C; at this point, in which incandescence was not observed, the pellets were removed from the furnace and allowed to cool down until ambient temperature at about 20°C/min. After the preliminary tests were done with clay with and without addition of untreated APCr, the range of firing temperature for LWA containing w-APCr was reduced to 1,140 – 1,180°C. Firing temperatures greater than 1,180°C and additions over 30% of w-APCr resulted in melting of the final products.



Figure 3.16 Temperature rate for LWA production from clay and APCr.

3.6.2 LWA from APCr by cold bonding

3.6.2.1 Procedure for manufacturing cold-bonded LWA

The general procedure for cold-bonded LWA production is shown in Figure 3.17. The raw materials (APCr, lime and clay) were mixed with deionised water and manually pelletised. Lime was used as the binder. Clay was added at small percentages to help to obtain a consistency that favours the pelletising. The pellets were dried overnight at 40°C and then, cured in open containers by exposition to the air under laboratory ambient conditions (average temperature of 20°C and relative humidity of 57%). The curing

conditions usually reported for cold-bonded LWA production are temperature around 20°C and relative humidity around 70% (Tajra, et al., 2019). The curing time was 28 days.



Figure 3.17 Flow diagram of cold bonded LWA production.

3.6.2.2 Preliminary tests of cold bonded LWA from APCr

According to the information presented in Table 2.7 for other MSWI residues, different percentages of APCr incorporation and the use of lime as a binder were assessed for the manufacture of cold-bonded LWA. Two preliminary tests were carried out to investigate the development of resistance to direct contact with water of different LWA formulations. In these experiments, the term "water resistance" indicates the capability of individual pellets to resist a direct contact with water and a slight pressure applied manually with a glass bar. The aim of the preliminary tests was to determine a range of weight proportions of APCr and lime that could be suitable for a formal assessment of cold-bonded LWA production.

Test No. 1: Batches of 100 g of the LWA formulations detailed in Table 3.11 were produced. The constant addition of 10% of lime was based on the binder dosage used for MSWI BA (Tang, et al., 2017). The development of water resistance was monitored every 3 days during a period of 3 weeks by immersing individual pellets in 50 mL of water for 2 hours. Every test was done by triplicate. The relationship between APCr and lime additions and early, late or no development of water resistance were evaluated.

Type of		10% Lime		40% Clay			
APCr	%	%	%	%	%	%	
	APCr	Clay	Water	APCr	Lime	Water	
	10	80	36	10	50	54	
Untreated	20	70	36	20	40	47	
APCr	30	60	36	30	30	44	
(APCr-m)	40	50	38	40	20	44	
	50	40	38	50	10	40	
	10	80	36	10	50	45	
	20	70	36	20	40	44	
w-APCr	30	60	36	30	30	43	
	40	50	38	40	20	42	
	50	40	38	50	10	40	
	10	80	37	10	50	56	
	20	70	38	20	40	53	
c-w-APCr	30	60	40	30	30	47	
	40	50	42	40	20	45	
	50	40	43	50	10	45	

Table 3.11 Cold bonded LWA formulations from low incorporation of APCr and constant or variable additions of lime.

Test No.2: Batches of 100 g of the LWA formulations detailed in Table 3.12 were produced. 70% was the highest percentage of APCr that allowed pelletising. 30% was kept as the maximum addition of lime for different additions of APCr, similar to the formulations reported by other studies using lime and MSWI BA (Cioffi, et al., 2011) or lime and MSWI FA (Colangelo, et al., 2015). The development of water resistance was monitored every 3 days during a period of 4 weeks. The water resistance was examined by triplicate as previously described in Test No.1.

	Type of	Formulation	%	%	%	%
	APCr	No.	APCr	Lime	Clay	Water
		1	50	10	40	40
		2	50	20	30	43
		3	50	30	20	45
		4	60	10	30	38
	APCr-m	5	60	20	20	39
		6	60	30	10	44
		7	70	10	20	42
		8	70	20	10	43
		9	70	25	5	43
		10	50	10	40	40
		11	50	20	30	42
		12	50	30	20	45
		13	60	10	30	38
	w-APCr	14	60	20	20	38
		15	60	30	10	41
		16	70	10	20	41
		17	70	20	10	42
		18	70	25	5	52
		19	50	10	40	35
		20	50	20	30	36
		21	50	30	20	41
		22	60	10	30	35
	c-APCr	23	60	20	20	35
		24	60	30	10	42
		25	70	10	20	40
		26	70	20	10	41
		27	70	25	5	42

Table 3.12 Cold bonded LWA formulations from variable additions of APCr, lime and clay.

3.6.2.3 Optimisation of cold bonded LWA production from APCr

According to the results arising from the preliminary tests, 12 formulations were evaluated for cold-bonded LWA, as detailed in Table 3.13. Batches of 500 g of each formulation were prepared and cured for 28 days at laboratory ambient conditions. After curing, the cold bonded LWA were tested for leaching, particle density, water absorption, compressive strength and loose bulk density and voids, according to the descriptions given in Section 3.7.

Type of APCr	Formulation No.	% APCr	% Lime	% Clay	% Water	APCr/Lime
	1	60	20	20	38	3.0
	2	60	25	15	39	2.4
W-	3	60	30	10	41	2.0
APCr	4	70	20	10	42	3.5
	5	70	25	5	49	2.8
	6	70	30	0	53	2.3
	7	60	20	20	35	3.0
	8	60	25	15	38	2.4
c-	9	60	30	10	42	2.0
APCr	10	70	20	10	41	3.5
	11	70	25	5	42	2.8
	12	70	30	0	45	2.3

Table 3.13. Formulation of cold bonded LWA from w-APCr or c-APCr.

3.7 Environmental and technological properties of LWA

3.7.1 Leaching of metals and soluble salts

To analyse the leaching properties of LWA, approximately 200 g of pellets were crushed to particle size less than 4 mm. Then, 90 g of the crushed material was tested for leaching according to the granular waste compliance leaching test (BS EN 12457-2, 2002) described in Section 3.2.2. In the absence of regulation for leaching of contaminants from LWA, the results were compared with the limit values for metals, soluble salts and TDS, expressed in the Landfill Directive for waste acceptance (The Council of the European Union, 2003), which were indicated in Table 3.5. The approach of comparing the leaching properties of LWA with limit values for waste material has been previously used by different authors (Gunning, et al., 2011a; Quina, et al., 2014b; Ayati, et al., 2016) because it is taken into consideration that the raw material is a hazardous waste, whose leaching properties should be reduced as much as possible or eliminated for a further reuse. In addition, leaching properties of LWA were compared with those determined in the laboratory for Lytag[®].

3.7.2 Bloating capacity

The analysis of bloating index (BI) was performed to assess the bloating capacity of the pellets produced by hot bonding. The BI is calculated as the percentage of diameter increase due to the firing process, according to Equation *3.8* (Quina, et al., 2014a):

Equation 3.8

$$BI = \frac{D-d}{d} * 100$$

Where D is the diameter of the fired pellet and d is the diameter of the pellet before firing, both values are given in mm. The measurements of the initial and final diameters on the pellets were done with a digital micrometre with a of precision 0.001mm (see Figure 3.18). The number of replicates analysed varied from 5 to 7 per batch of LWA.



Figure 3.18 Measurement of diameter of a sintered pellet.

3.7.3 Crushing strength

The analysis of crushing strength of single pellets was done using a point load testing system (see Figure 3.19). For this analysis, it is assumed that the pellets are brittle spheres. 15 pellets were tested per batch to ensure reproducibility of the results of fracture load.



Figure 3.19 Measurement of crushing strength of a pellet in a point load system.

The crushing strength for each pellet was calculated by Equation 3.9 (González-Corrochano, et al., 2009; Li, et al., 2000):

Equation 3.9

$$S = \frac{2.8 \times P_c}{\pi X^2}$$

Where:

S is the crushing strength of a single pellet in mega Pascals (MPa)

Pc is fracture load in Newtons

X is the distance between loading points, equivalent to the sphere diameter in mm

3.7.4 Particle density and water absorption

3.7.4.1 Method of the wired basket

The method of the wire basket for analysis of particle density and water absorption in LWA according to (BS EN 1097-6, 2022) was used for hot bonded LWA during the optimisation of conditions for their manufacture. A specific gravity kit placed over an analytical scale with a precision of ± 0.0001 g was used for this analysis (see Figure 3.20). Around 20g of pellets was dried at 105°C for at least 2 hours to ensure constant mass, and allowed to cool down before starting the test. The analysis was done in triplicate for each batch of LWA. The procedure and calculations are summarised in Table 3.14.



Figure 3.20 Setup for analysis of particle density and water absorption of LWA (wire basket method).

The following procedure is described for the analysis of a single pellet. Firstly, the weight of the pellet (D) was measured. Then, the specific gravity kit was placed over the scale, including 100 mL of water in the beaker but excluding the basket, and it was set the zero

in the scale. The basket was hung in the wire and immersed into the water to determine its weight in water (C). The pellet was placed in the wire basket and immersed into the water up to about 5 cm under the surface of the water. The basket containing the pellet was moved up and down inside the water several times to allow the release of the entrapped air from the pellet. Once the air was eliminated from the pellet, the weight of the basket containing the pellet in water (B) was measured. The pellet was left and remained completely immersed in a beaker containing 100 mL of water for 24 hours. After this time, the pellet was removed from the water, its surface was dried with a tissue and finally, its weight (A) was measured.

Table 3.14 Determination of particle density and water absorption of LWA by the wire basket method (BS EN 1097-6, 2022).

Property	Definition	Calculation	Units						
Apparent particle density	Oven-dried mass of a pellet divided by the volume that it occupies in water including the volume of any internal sealed voids but excluding the volume of water accessible voids	Equation 3.10 $\frac{D}{D - (B - C)}$	g/cm ³						
Oven-dried particle density	Oven-dried mass of a pellet divided by the volume that it occupies in water including the volume of any internal sealed voids and the volume of any water accessible voids	Equation 3.11 $\frac{D}{A - (B - C)}$	g/cm ³						
Saturated and surface-dried particle density	Sum of the oven-dried mass of a pellet and the mass of water in any water accessible voids divided by the volume of any internal sealed voids and the volume of any water accessible voids	Equation 3.12 $\frac{A}{A - (B - C)}$	g/cm ³						
Water absorption	Mass of absorbed water expressed as a percentage of the oved-dried mass of the pellet.	Equation 3.13 $\frac{(A - D)}{D} \times 100$	%						
A is the mass, i	in g, of the saturated surface-dry aggregates in	air							
B is the appare	B is the apparent mass, in g, in water of the basket containing the saturated pellet								
D is the mass,	in g, of the oven-dried aggregate in air								

3.7.4.2 Method of the pycnometer

For the analysis of particle density and water absorption in hot bonded LWA and cold bonded LWA, the pycnometer method was applied according to the Annex C of (BS EN 1097-6, 2022), which specifies the determination of these properties in coarse LWA with particle diameter between a 4 and 31.5 mm. The hot bonded LWA were oven-dried at 105°C and the cold bonded LWA were oven-dried at 40°C. To determine the pycnometer volume, the pycnometer was assembled to the conical lid and filled up with water at 20 ± 2 °C until the narrow edge of the lid; then, it was placed for about 1 hr in a water bath at 20 ± 2 °C; after this period, the assemble was removed from the water bath, carefully cleaned, and weighed (M₃). Depending on the available amount of each LWA formulation, the tests were done by duplicate or triplicate. The sample size of each LWA formulation (M₄) was that one having a loose bulk volume between 500 to 600 mL, which corresponds to around half of the volume of the pycnometer, as it can be observed in Figure 3.21 Part A.



Figure 3.21 Analysis of particle density and water absorption of LWA by the method of the pycnometer. A: Soaking of LWA in the pycnometer. B: Saturated and surface-dried LWA.

The appropriate mass of LWA was placed into the pycnometer. The pycnometer was assembled to the conical lid and filled up with water at 20 ± 2 °C until the narrow edge of the lid. Immediately, a timer was started to count the soaking time. The assembly was stirred by gently rolling and tapping against the bench to remove the air entrapped in the

LWA. The assembly was placed in a water bath at 20 ± 2 °C for 24 hr. Water was added as required during the test to maintain the water level close to the narrow edge of the lid. The weight of the assembly was measured after 10 min M₂ (10 min), 1hr M₂ (1 hr), 6 hr M₂ (6 hr) and 24 hr M₂ (24 hr). Before every weight register, the assembly was gently stirred by rolling and tapping against the bench to remove entrapped air, the loss of water close to the narrow edge of the lid was replaced and the surface of the assembly was carefully cleaned.

After registering M_2 (24 hr), the conical lid was removed, and all the water was drained. The LWA were transferred to a tray covered with a sorbent tissue, as it is shown in Figure 3.21 Part B. The water on the LWA surface was removed by gently moving the tray, so that the pellets rolled along the tissue for maximum 15 sec. The weight of these saturated and surface-dried LWA was measured (M₁).

The apparent particle density, oven-dried particle density, saturated and surface-dried particle density as well as water absorption, previously defined in Table 3.14, were calculated according to the method of the pycnometer, as per the equations detailed in Table 3.15.

Table 3.15 Calculation of particle density and water absorption in LWA by the method of the pycnometer (*BS EN 1097-6, 2022*) – Annex C.

F	Property	Calculation	Units		
Apparent pa	article density, ρ_a	Equation 3.14 $\rho_w \times \frac{M_4}{M_4 - [M_2(24hr) - M_3]}$	g/cm ³		
Oven-dried ρ_P	particle density,	Equation 3.15 $ \rho_w \times \frac{M_4}{M_1 - [M_2(24hr) - M_3]} $	g/cm ³		
Saturated a particle den	and surface-dried sity, ρ_s	Equation 3.16 $\rho_w \times \frac{M_1(24hr)}{M_1 - [M_2(24hr) - M_3]}$	g/cm ³		
	At 24 hr, WA ₂₄	Equation 3.17 $\frac{M_1 - M_4}{M_4} \times 100$	%		
Water absorption	At 10 min, WA ₁₀	Equation 3.18 $WA_{24} - \frac{M_2(24\text{hr}) - M_2(10 \text{ min})}{M_4} \times 100$	%		
	At 60 min, WA ₆₀	Equation 3.19 $WA_{24} - \frac{M_2(24\text{hr}) - M_2(60 \text{ min})}{M_4} \times 100$	%		
ρ_w is the det	nsity, in g/cm ³ , of w	vater at test temperature: 0.9963 g/cm ³ at 19°C			
M_1 is the m	ass, in g, of the satu	rated surface-dry aggregates in air after 24 hr			
M ₂ is the mass, in g, of the pycnometer, conical lid, water and saturated aggregates after					
10 min M ₂ (10 min), 60 min M ₂ (60 min) or 24 hr M ₂ (24 hr)					
M ₃ is the ma	ass, in g, of the pycr	nometer, conical lid and water as calibrated at 20	$) \pm 2 \ ^{\circ}\mathrm{C}$		
M ₄ is the m	ass, in g, of the over	n-dried LWA			

3.7.5 Loose bulk density and voids

Analysis of loose bulk density and voids in LWA was conducted according to (BS EN 1097-3, 1998). The standard defines loose bulk density as the ratio between the mass of dry aggregate filling a specified container without compaction and the volume of that container, while voids are the air-filled spaces between the aggregate particles in the container. Previous analysis, the LWA were oven dried at 105°C (hot bonded LWA) or 40°C (cold bonded LWA). In this research, a cylindrical glass container was used, as shown in Figure 3.22, whose capacity, determined by calibration with water at 20 ± 2 °C, was 1.061 L (V).



Figure 3.22 Container filled up with LWA for analysis of loos bulk density and voids.

The container was filled up with the LWA sample. The surplus pellets were removed from the top of the container and the surface of the aggregate was levelled with a metallic straightedge. The filled container was weighed, and its mass was recorded (m₂). Each batch of LWA was tested in triplicate. the loose bulk density was calculated according to Equation 3.20:

Equation 3.20

$$\rho_b = \frac{m_2 - m_1}{V}$$

Where:

 ρ_b is the loose bulk density, in g/cm³

m2 is the mass of the container and the LWA sample, in g

m1 is the mass of the empty container, 228.17 g

V is the capacity of the container, 1,061 cm³

The percentage of voids v represents the volumetric proportion of voids in the container and it was calculated according to Equation 3.21:

Equation 3.21

$$v = \frac{\rho_p - \rho_b}{\rho_p} \times 100$$

Where:

v is the percentage of voids

 ρ_b is the loose bulk density, in g/cm³

 ρ_p is the oven-dried particle density determined by the method of the pycnometer according to the procedure described in Section 3.7.4.2.

3.8 Manufacture of lightweight aggregate concrete (LWAC)

3.8.1 Formulation and casting of LWAC blocks

The manufactured LWAs were incorporated into different formulations of concrete cubes according to the concrete production requirements detailed in (BS EN 206, 2013+A2, 2021). The concrete formulations, as detailed in Table 3.16, were calculated according to the absolute volume method, which assumes that the final volume of compacted concrete is equal to the sum of the absolute volumes of all its components (Neville, 1995). The quantities of cement, water, sand and LWA were calculated based on the production of one cubic metre of concrete. Appendix V - Formulation of LWAC by the absolute volume method shows all the calculations made for concrete formulations according to each LWA formulation. Concrete cubes formulated from Lytag[®] were used as control for LWAC prepared from hot-bonded LWA, whereas concrete cubes containing Carbon8[®] LWA were used as control for LWAC formulated from cold-bonded LWA.

	LV	VA Form	ulation			Concrete formulation $m = kg LWA/m^3$ concrete					
	% w- APCr	% c- APCr	% Clay	% Lime		m cement	m water	m sand	m LWA		
	0	0	100	0		268	107	250	463		
Hot-	25	0	75	0 0		268	107	280	518		
bonding	30	0	70			268	107	242	448		
	Lytag®					268	107	229	423		
	60	0	10	30		268	107	232	430		
~ 11	70	0	0	30		268	107	222	412		
Cold	0	60	10	30		268	107	259	481		
bonding	0	70	0	30		268	107	244	452		
	Carbon8®					268	107	184	340		

Table 3.16 Formulation of LWAC blocks.

Cubic polystyrene moulds of 100 mm side were used to prepare the concrete cubes, as it is shown in Figure 3.23 – Part A, meeting the shape and dimension requirements of (BS EN 121390-1, 2021). The inner walls of the moulds were previously covered with a thin film of petroleum jelly to prevent the concrete from adhering to the mould. The fresh concrete mix was poured into the mould and a small trowel was used to distribute and compact the different layers of the mix. Then, 25 strokes per layer were applied to remove pockets of entrapped air. After compaction of each layer, the sides of the mould were carefully tapped by hand until no bubbles of air were observed on the surface. The trowel was used to remove the excess concrete above the upper edge of the mould and to level the surface. All the specimens were left in the moulds for 24 hours at laboratory temperature $(18 \pm 3 \,^{\circ}\text{C})$ and covered with a plastic film to avoid dehydration. After this period, the concrete cubes were removed from the moulds (see Figure 3.23 – Part B) and properly labelled before being transferred to the curing tank.



Figure 3.23 Concrete cubes preparation. A: Fresh concrete mix in a polystyrene mould. B: Concrete cube unmoulded after 24 hr of hardening.

The specimens were cured in a polypropylene curing tank (Figure 3.24) that meets the requirements of (BS EN 121390-2, 2019). This tank is equipped with heater element, a thermostat and a temperature controller that was set of 20 ± 3 °C. A water pump was attached to the tank to ensure a continuous flow of the water. The concrete cubes were placed on a galvanised support rack, allowing the complete exposure of specimen to the water. The curing time was 28 days. During this time, the temperature and pH (7.5 – 8.0) of the water were monitored.



Figure 3.24 Curing of LWAC cubes. Set up as per requirements of the British Standards (BS EN 121390-2, 2019).

3.8.2 Testing of density and compressive strength of LWAC cubes

The concrete cubes were tested for density according to (BS EN 12390-7, 2019) after 28 days of curing. The water-saturated concrete cubes were weighed in water (at 20 °C) and in air. The density of the concrete cubes was calculated according to Equation 3.22

Equation 3.22

$$D = \frac{m_{air}}{\left(\frac{m_{air} - m_{water}}{\rho}\right)}$$

Where:

D is the density of the water-saturated concrete cube, in kg/m³ m_{air} is the mass of the concrete cube in the air, in kg m_{water} is the mass of the concrete cube in water, in kg ρ is the density of water at 20°C, taken as 998 kg/m³

The compressive strength of the concrete cubes was determined according to (BS EN 12390-3, 2019). For this analysis a Matest compression testing machine, shown in Figure 3.25, with 2,000 kN capacity was used. This equipment meets the requirements of (BS EN 12390-4, 2019) The maximum load and the maximum strength values were directly shown in the equipment display.



Figure 3.25 Analysis of compressive strength of a LWAC cube.

3.8.3 Carbon footprint of LWAC

The carbon footprint of LWAC incorporating APCr-based LWA was conducted according to the principles, requirements and guidelines for the quantification and reporting of the carbon footprint of a product (CFP) specified in the British Standard (BS EN ISO 14067, 2018). This standard is consistent with the principles of life cycle assessment (LCA) defined in (BS EN ISO 14040:2006+A1, 2020) and the requirements and guidelines of LCA stated in (BS EN ISO 14044:2006+A2, 2020).

3.9 Summary

In this chapter, the methodology followed to investigate the viability of incorporating APCr into engineered LWA was described in detail. Figure 3.26 shows the schematic summary of this methodology, which can be briefly described in 4 stages. In the first stage, APCr samples collected from different EfW facilities across the UK were characterised for elemental composition, mineralogy and leaching properties. In the second stage, a mix composed of all samples of APCr previously characterised was prepared for the assessment of washing with water and accelerated carbonation as treatment options. The third stage consisted of the manufacture of artificial LWA from treated APCr, through hot bonding and cold bonding processes at a laboratory scale. In the fourth stage, hot-bonded and cold-bonded LWAs were incorporated into lightweight concrete (LWAC) to evaluate its performance and the carbon emissions associated with its production. A number of analytical techniques, including XRD, ICP/OES, SEM and SEM/EDS, among others were used for the characterisation of raw material, treated materials and final products.



Figure 3.26 Schematic summary of the methodology for recycling of APCr into engineered LWA. Stage 1: APCr characterisation, Stage 2: APCr treatment, Stage 3: LWA manufacture, Stage 4: Performance of LWA in concrete. The laboratory techniques (shown outside the stages) were applied for characterisation of raw materials, treated materials and final products.

4 Chapter 4 Chemical characterisation of APCr

4.1 Water content

The water content of APCr depends on the APC technologies used for the flue gas cleaning, including acid-gas scrubbing systems (dry, semi-dry and wet) and fabric filters. In this study, it is known that the APCr samples originated in different EfW facilities equipped with either dry, semi-dry or wet acid-gas scrubbing systems. However, the samples were not classified according to the APC systems where they came from, due to the difficulty to access the detailed information of every facility. Figure 4.1 presents the results of water content analysed by gravimetry in 42 APCr samples.



Figure 4.1 Water content of APCr samples.

It can be seen Figure 4.1 that there is a significant variability of two orders of magnitude in the water contents of the analysed APCr samples. The calculate Coefficient of Variation (CV) was 168%. These variations were expected, given the multiple origins of the samples in terms of the acid-gas scrubbing systems adopted by different MSWI facilities. Therefore, the estimated mean value for water content, 8.3%, should not be considered reliable; instead, it is concluded that the water content of APCr samples coming from different MSWI facilities in the UK ranged from 0.0 to 53.3%. Table 4.1 shows the statistical analysis and comparison of the water contents of the analysed samples with data reported by different authors. Based on the agreement of the water contents lower than 10% with the percentages reported by other studies, it can be expected that the APCr samples with water contents not exceeding 10% came from dry and semidry scrubbing systems. Whereas, those samples with water contents greater than 10% may have an origin in wet acid-gas scrubbing systems, for which there was not available data for comparison. It is important to note that the water content of APCr may vary over time because they contain hygroscopic phases, such as Ca-based salts (Quina, et al., 2008b).

Water content %									
Min	Min Max		ST DEV	CV%	Acid-gas scrubbing				
0.0	53.3	8.3	14.0	168	Dry, semi-dry, wet				
		Compariso	n with other stu	ıdies					
Author		Country	Acid-gas scr	ubbing	Water content %				
This study		UK	Dry, semi-dr	y and wet	0.0 - 53.3				
(Sun, et al., 2	008)	UK	Semi-dry		1.79				
(Quina, et al.	, 2008b)	Portugal	Semi-dry		0.73 - 1.05				
(Hyks, et al.,	2009)	Denmark	Semi-dry		4.7				
(Cappai, et al	., 2012)	Italy	Dry		2.52				
(Bogush, et al., 2015) U		UK	Unspecified		0.42 - 2.48				
(Nikravan, et	al., 2020)	Iran	Semi-dry		0.231				

Table 4.1 Water contents of APCr, statistics and comparison with other studies.

The dry matter contents of APCr varied between 46.7% and 100% in the analysed samples. The variability of the results, as expressed by the CV of 15%, has the same explanation as the variability of the water contents. Therefore, it is not suitable to express

the dry matter content of APCr as an estimated mean value. Instead, this characteristic must be taken into account for individual samples.

4.2 Total metals contents

The total contents of the 12 metals regulated by the WAC determined in 27 APCr samples by ICP/OES are reported in Table 4.2 with the corresponding statistical analysis and comparison with values reported by four of the most recent studies conducted in the UK. There was a significant variation in metal composition among the analysed APCr samples, with coefficients of variation (CV%) ranging from 32% to 105% (except Hg which was detected in only one sample). This variation indicates that the composition of APCr changes according to the different waste composition, incineration conditions and APC systems adopted by different EfW facilities. In general, there was an agreement with other studies conducted in the UK (Gunning, et al., 2011a; Bogush, et al., 2015; Ayati, et al., 2016; Bogush, et al., 2019) for the metals detected at trace levels of concentrations (Mo and Ni), metals that were not detected in most of the samples (Hg and Se) and Sb that was detected as a minor element. In contrast, the other metals, detected as minor and major elements, were present in concentrations much higher than the values reported by the mentioned studies, especially As, Cu, Pb and Zn.

The total contents of metals for the APCr samples are also shown in Figure 4.2. During the second sampling episode, higher concentrations of Zn and Cu were observed in some of the samples, while the majority of the samples showed lower concentrations of Pb. However, no clear tendencies of increasing or decreasing total metal concentrations are observed over the time during the sampling periods. The metals with the highest concentrations and that were present in all the analysed APCr samples were Zn (1,280 to 32,200 mg/kg), Pb (122 - 10,500 mg/Kg) and Cu (115 – 3,750 mg/kg). Other metals that were detected in all the samples, but at lower concentrations were Ba (79.8 – 681 mg/kg), Cr (10.4 – 440 mg/kg) and Cd (4.4 – 316 mg/kg). Hg is not usually reported to be present in APCr. In this study, it was detected in only one sample at a concentration as low as 65.8 mg/kg. According to (Song, et al., 2004), Hg is easily converted into vapour during MSW incineration, the vapour reacts with HCl in the flue gases producing mostly HgCl₂ and this compound is further removed in the water spray tower since it is highly soluble in water.

	As	Ba	Cd	Cr	Cu	Hg	Мо	Ni	Pb	Sb	Se	Zn	Sum of total metals
APCr-02	487	311	30.9	164	981	< 0.975	6.56	53.5	4,790	148	<1.77	11,600	18,572
APCr-03	205	152	12	58.4	334	<1.03	2.85	13	2,240	82.1	<1.87	2,790	5,889
APCr-04	<4.19	535	143	79.4	507	<1.01	11.1	77.2	1,050	499	<1.84	8,860	11,762
APCr-05	<4.09	475	10.2	18.2	507	< 0.986	2.69	10.4	1,220	139	108	8,140	10,630
APCr-06	597	316	59.3	83.2	724	<1.01	6.38	12.2	10,500	352	<1.84	13,300	25,950
APCr-07	611	377	30.3	440	1,370	< 0.909	2.83	46.2	3,330	188	<1.65	5,420	11,815
APCr-08	<3.86	623	11.9	42.9	115	< 0.930	<2.28	31.8	122	<4.25	<1.69	1,280	2,227
APCr-09	331	79.8	27.8	175	499	<1.06	11.3	72.7	9,270	331	<1.92	7,630	18,428
APCr-10	<3.70	304	4.43	10.4	219	65.8	<2.19	5.69	246	114	68.4	6,670	7,708
APCr-11	332	323	31.3	173	515	< 0.926	8.52	40.3	5,050	255	<1.68	4,610	11,338
APCr-13	321	352	30.3	127	458	< 0.943	<2.31	30.0	5,550	233	<1.71	4,660	11,761
APCr-14	<4.31	580	170	83.1	500	<1.04	9.7	83.0	859	472	<1.89	8,590	11,347
APCr-15	1,120	534	92.3	210	1,400	<1.04	11.5	60.6	7,400	466	<1.89	32,200	43,494
APCr-16	288	234	25.2	123	392	< 0.938	6.8	36.6	5,260	217	<1.71	4,360	10,943
APCr-17	<4.16	579	61.4	69.1	339	<1.00	7.8	48.2	615	440	<1.82	4,300	6,459
APCr-18	286	338	37.1	79.3	461	< 0.969	14.0	17.0	6,470	278	<1.76	6,850	14,830
APCr-32	<3.85	236	124	61.8	397	< 0.928	8.3	25.5	1,300	536	<1.69	8,260	10,949
APCr-33	156	296	316	64.6	887	< 0.944	14.0	32.7	4,200	823	<1.72	18,200	24,989
APCr-34	209	257	310	60.0	867	< 0.961	10.5	27.9	5,110	871	<1.75	20,500	28,222
APCr-35	<4.12	198	118	125	698	< 0.994	9.5	117	834	490	<1.81	7,430	10,020
APCr-36	<3.78	448	149	92.4	536	< 0.912	10.3	72.0	1,510	456	<1.66	7,840	11,114
APCr-37	73.3	338	301	55.6	631	< 0.964	6.1	25.2	2,310	990	<1.75	13,500	18,230
APCr-38	<3.95	494	27.6	132	3,750	< 0.952	10.1	88.7	844	302	<1.73	2,700	8,348
APCr-39	<4.14	446	16.5	139	2,910	< 0.998	8.7	82.0	640	203	<1.81	2,350	6,795
APCr-40	<3.86	681	20.4	150	3,590	< 0.929	11.3	96.8	1,030	261	<1.69	3,370	9,211
APCr-41	234	162	238	41.0	677	<1.05	6.9	17.4	5,740	849	<1.91	21,400	29,365
APCr-42	<3.76	371	133	76.1	418	< 0.906	8.4	61.4	1,320	471	<1.65	6,670	9,529
Minimum	<3.70	79.8	4.4	10.4	115	< 0.906	<2.28	5.7	122	<4.25	<1.65	1,280	2,227
Maximum	1,120	681	316	440	3,750	65.8	14.0	117	10,500	990	108	32,200	43,494
Average	375	372	94	109	914	< 0.906	8.6	47.6	3,289	403	88.2	9,018	14,442
CV %	71	41	104	77	105	0	36	63	88	62	32	78	63
(Bogush, et al., 2019)	5.5	450	30	370	260	3.2	12	36	550	200	1.9	2,900	4,819
(Ayati, et al., 2016)	<3.82	429	116	114	611	< 0.92	11	41	1,840	543	<1.67	9,190	12,895
(Bogush, et al., 2015)	12 - 38	316 - 452	26 - 190	58 - 110	320 - 580	NA	4.8 - 15	21 - 59	500 - 2,000	170 - 510	<0.5 - 9.6	2,600 - 7,300	4,028 - 11,264
(Gunning, et al., 2011a)	<0.5 - 485	196 - 476	22 - 123.1	48 - 109.3	315 - 480	2.7 - 3.3	2.4 - 7.1	7.4 - 36	1,449 - 6,774	24.3 - 346	0.9 - 3	560 - 6,110	2,628 - 14,953

Table 4.2 Total contents of metals in APCr.

Results are expressed as mg/kg. Less than values (<) indicate results lower than the detection limits. NA: not analysed. The values are expressed with no contribution from uncertainty of measurement



Figure 4.2 Total contents of metals of APCr.

4.3 Elemental composition of APCr

In this section, 41 APCr samples were analysed for elemental composition by SEM/EDS. The EDS spectrums and SEM micrographs are presented in Appendix VI. The results of elemental composition in wt.% and their corresponding uncertainty values are detailed in Appendix VII. Figure 4.3 and Figure 4.4 show the concentrations of 17 chemical elements detected out of 25 chemical elements analysed in APCr.



 $0 \quad 5 \quad 10 \quad 15 \quad 20 \quad 25 \quad 30 \quad 35 \quad 40 \quad 45 \quad 50 \quad 55 \quad 60 \quad 65 \quad 70 \quad 75 \quad 80 \quad 85 \quad 90 \quad 95 \quad {\rm wt.\%}$

Figure 4.3 Elemental composition determined by SEM/EDS (normalised data) for samples APCr-01 to APCr-21.



0 5 10 15 20 25 30 35 40 45 50 55 60 65 70 75 80 85 90 95 $_{wt.\%}$

Figure 4.4 Elemental composition determined by SEM/EDS (normalised data) for samples APCr-22 to APCr-42.

It can be seen in Figure 4.3 and Figure 4.4, that O, Ca and Cl were detected in all the analysed APCr samples as major elements at concentrations varying from 30.4 to 88.5 % O, 3.6 to 45.0 % Ca and 1.1 to 22.1 % Cl. The variability in concentration of these elements, expressed as the CV was 26%, 31% and 54% for O, Ca and Cl, respectively. Other major element detected in all the APCr samples, although with a greater variability in content (CV of 114%) was Si at concentrations ranging from 0.1% to 22.3%. S was detected in 98% of the analysed samples, but its concentrations were significantly lower than those determined for the elements previously mentioned, varying between 0.3% and 5.9%. The alkaline metals K and Na were found in 83% and 90% of the APCr samples, respectively, at concentrations between 0.3 and 15.6% K and from 0.4 to 6.3% Na. Other elements were detected in most of the APCr samples, and their concentrations were Al 0.2 to 9.2%, Mg 0.2 to 4.4%, Fe 0.3 to 4.5%, P 0.2 to 1.1% and Zn 0.5 to 4.6%. The other elements were detected in less than the third part of the total number of samples and their concentrations were the lowest: Ba 0.5 to 2.9%, Mn 0.2 to 0.5%, Pb 1.2 to 4.4%, Sb 1.0 to 1.2% and Cu that was detected in only one sample at a concentration of 0.5%. The elements analysed by SEM/EDS but not detected in any of the APCr samples were As, Cd, Co, Cr, F, Mo, N and Ni.

Some disagreements were observed in the concentrations of Zn, Pb and Cu in some of the 26 APCr samples that were analysed by both techniques ICP/OES and SEM/EDS. Firstly, it was determined by ICP/OES that these metals were present in all 26 samples, whereas by SEM/EDS, Zn was detected in 73% of the samples, Pb in 23% and Cu in only 4%. The reasons for this difference are the low limits of detection of the technique ICP/OES (7.68x10⁻⁴ % Zn, 5.48x10⁻⁵ % Pb and 9.17x10⁻⁵ % Cu) that allow the detection of concentrations up to 5 orders of magnitude lower than the concentrations detected by SEM/EDS. Secondly, for most of the samples in which these metals were detected by SEM/EDS, their concentrations (0.5 to 4.6% Zn, 1.2 - 4.4% Pb and 0.5% Cu) were in general higher than the percentages calculated from the total concentration determined by ICP/OES (0.1 - 3.2% Zn, 0.01 - 1.0% Pb and 0.01 - 0.4% Cu). Figure 4.5 shows the comparison between the Zn and Pb contents expressed as wt % determined by ICP/OES against those determined by SEM/EDS. With only a few exceptions, the Zn concentrations obtained by both techniques were equivalent or close to each other with differences of no more than 0.6% (Figure 4.5 - Part A), indicating that the results obtained by both techniques are consistent. Concentrations of Pb determined by SEM/EDS were in all cases greater than those determined by ICP/OES (Figure 4.5). This indicates that

probably, there is an overestimation of Pb in the analysis of APCr by SEM/EDS, or the aqua regia (extracting solution) did not completely dissolve the element in the ICP/OES analysis.





Figure 4.5 Comparison of contents of metals determined by ICP/OES and by SEM/EDS, A: Comparison of Zn, B: Comparison of Pb. The error bars of SEM/EDS measurements correspond to +/- 1 standard deviation from the mean value, calculated by the EDS software.
Table 4.3 shows the complete list of chemical elements analysed in APCr by SEM/EDS and the comparison of their average and range of concentrations with the values reported by other studies conducted in the UK. It must be noted that there was not available data obtained by SEM/EDS for comparison, and in the referenced studies, XRF and/or ICP/OES were the analytical techniques. In general, the elemental composition of APCr determined in this study was in good agreement with the results previously found by other authors either expressed as metals or as oxides.

% of samples Elemental composition reported by other studies in the UK									in the UK			
Element	Average wt%	SD	CV%	Min	Max	in which the element was detected	(Sun, et al., 2008) ^a	(Dimech, et al., 2008) ^a	Dimech, et al., (Bogush, et al., 2008) ^a 2015) ^b		(Bogush, et al., 2019) ^a	(Bogush, et al., 2020) ^c
Al	2.0	2.1	104	0.2	9.2	90	12.12 as Al ₂ O ₃	2.8 as Al ₂ O ₃	0.9 - 2.9	0.8 as Al_2O_3	2.20	8.3 as Al ₂ O ₃
As	< 0.1	0	0	< 0.1	< 0.1	0						
Ba	1.2	0.7	59	0.5	2.9	27						
Ca	29.4	9.1	31	3.6	45	100	36.27 as CaO	42 as CaO	22.4 - 32	65.0 as CaO	35.0	47 as CaO
Cd	< 0.1	0	0	< 0.1	< 0.1	0						
Cl	10.0	5.4	54	1.1	22.1	100		21 as Cl ⁻		9.8	7.20	7.2 as Cl ⁻
Co	< 0.1	0	0	< 0.1	< 0.1	0						
Cr	< 0.1	0	0	< 0.1	< 0.1	0		0.03 as Cr ₂ O ₃		<dl as="" cr<sub="">20₃</dl>		
Cu	0.5	0.0	0	0.5	0.5	2		0.40 as CuO				
F	< 0.5	0	0	< 0.5	< 0.5	0		0.13				
Fe	1.1	1.0	88	0.3	4.5	63	1.05 as Fe ₂ O ₃	0.47 as Fe ₂ O ₃		0.8 as Fe ₂ O ₃	0.76	2.2 as Fe ₂ O ₃
K	2.5	3.2	132	0.3	15.6	83	2.03 as K ₂ O	1.87 as K ₂ O	0.92 - 3.5	2.0 as K ₂ O	1.00	2.5 as K ₂ O
Mg	0.9	0.8	86	0.2	4.4	76	2.5 as MgO	0.05 as MgO	0.52 - 0.84	1.2 as MgO	0.78	1.3 as MgO
Mn	0.4	0.2	40	0.2	0.5	10	0.04 as MnO			0.2 as MnO		
Мо	< 0.3	0	0	< 0.3	< 0.3	0						
Ν	<3.7	0	0	<3.7	<3.7	0						
Na	1.3	1.0	79	0.4	6.3	90	7.81 as Na ₂ O	2.68 as Na ₂ O	1.2 - 3.5			3.2 as Na ₂ O
Ni	< 0.1	0	0	< 0.1	< 0.1	0					36.0	
0	44.9	11.8	26	30.4	88.5	100						
Р	0.5	0.2	50	0.2	1.1	68		0.78 as P ₂ O ₅	0.2 - 0.64	0.2 as P ₂ O ₅	0.37	1.7 as P ₂ O ₅
Pb	2.1	1.3	60	1.2	4.4	17		0.1 as PbO				
S	3.1	1.3	41	0.3	5.9	98	6.29 as SO ₃	5.66 as SO ₃		12.0 as SO ₃	1.30	3.9 as SO ₄ -2
Sb	1.1	0.1	13	1	1.2	5						
Si	4.4	5.0	114	0.1	22.3	100	30 as SiO ₂	7.14 as SiO ₂	< 0.04 - 0.92	2.5 as SiO ₂	3.70	7.9 as SiO ₂
Zn	1.2	0.9	77	0.5	4.6	59		0.87 as ZnO	0.26 - 0.73	2.5 as ZnO	0.29	

Table 4.3 Elemental composition of APCr determined by SEM/EDS and comparison with values reported by other studies in the UK.

All the results are expressed as wt%

Analysis techniques: ^a XRF, ^b ICP/OES, ^c Ion chromatography for Cl as Cl⁻ and S as SO₄⁻² and XRF for the other elements Less than values (<) indicate results lower than the detection limits. Highlighted values indicate greater agreement with the current study

4.4 Leaching tests

The results of leaching tests (BS EN 12457-2, 2002) of heavy metals for 26 APCr samples are reported in Table 4.4 with the corresponding statistical analysis and comparison with the hazardous Waste Acceptance Criteria WAC limit values

4.4.1 Leaching of heavy metals

Figure 4.6 also summarises the leaching rates of heavy metals from APCr.

									mg/kg								pH at	Conductivity at
	As	Ba	Cd	Cr	Cu	Hg	Mo	Ni	Pb	Sb	Se	Zn	Chlorides	Fluorides	Sulphates	TDS	20°C	20°C µS/cm
APCr-02	<1.582	79.6	< 0.071	2.4	4.0	<1.158	1.6	< 0.325	1,391.8	< 0.858	<1.008	58.2	84,999.4	28.4	16,765.1	194,300	12.54	28,700
APCr-03	<1.582	15.8	< 0.071	12.8	1.3	<1.158	3.2	< 0.325	778.9	< 0.858	<1.008	70.9	82,693.1	22.8	16,751.3	191,100	12.93	29,870
APCr-04	<1.582	41.6	< 0.071	1.9	8.6	<1.158	2.3	< 0.325	267.7	< 0.858	<1.008	123.7	139,672.1	42.0	10,574.0	267,700	13.08	40,620
APCr-05	<1.582	71.5	< 0.071	< 0.201	< 0.712	<1.158	0.5	< 0.325	144.4	< 0.858	<1.008	74.6	225,929.6	34.1	7,115.1	401,300	12.42	49,870
APCr-06	<1.582	59.5	< 0.071	6.6	7.3	<1.158	1.8	< 0.325	2,206.6	< 0.858	<1.008	184.9	167,608.8	31.4	10,497.1	331,300	12.58	43,740
APCr-07	<1.582	14.3	< 0.071	36.3	< 0.712	<1.158	0.9	< 0.325	288.1	< 0.858	<1.008	11.8	66,343.5	<20.3	17,659.1	158,800	12.54	24,580
APCr-08	<1.582	2.9	< 0.071	2.7	< 0.712	<1.158	3.3	< 0.325	<2.209	< 0.858	<1.008	21.3	9,755.7	<20.3	27,566.2	104,200	12.86	20,030
APCr-09	<1.582	4.2	< 0.071	0.7	< 0.712	<1.158	2.2	< 0.325	43.9	< 0.858	<1.008	<2.483	41,602.2	27.8	27,465.0	123,200	11.01	15,800
APCr-10	<1.582	44.6	< 0.071	0.9	< 0.712	<1.158	0.2	< 0.325	7.0	< 0.858	2.3	19.3	193,735.8	25.0	4,336.0	297,000	12.35	43,090
APCr-11	<1.582	13.8	< 0.071	12.4	< 0.712	<1.158	2.0	< 0.325	632.3	< 0.858	<1.008	17.2	45,308.5	24.1	15,116.3	130,500	12.57	20,780
APCr-13	<1.582	14.5	< 0.071	10.2	0.8	<1.158	2.1	< 0.325	882.2	< 0.858	<1.008	35.1	78,466.0	24.8	15,802.8	172,600	12.69	27,650
APCr-14	<1.582	21.0	< 0.071	1.0	2.2	<1.158	1.0	< 0.325	43.5	< 0.858	<1.008	26.1	124,091.3	29.0	12,025.6	229,300	12.62	35,490
APCr-15	<1.582	0.5	< 0.071	1.2	< 0.712	<1.158	0.2	< 0.325	<2.209	< 0.858	<1.008	<2.483	<51.0	<20.3	14,080.8	20,000	9.13	1,875
APCr-16	<1.582	13.2	< 0.071	15.8	< 0.712	<1.158	3.2	< 0.325	915.3	< 0.858	<1.008	19.1	77,819.5	<20.3	20,858.9	191,500	12.58	29,410
APCr-17	<1.582	34.5	< 0.071	0.4	0.8	<1.158	0.4	< 0.325	11.6	< 0.858	<1.008	5.8	97,286.7	<20.3	2,614.2	171,900	12.07	27,880
APCr-18	<1.582	6.5	< 0.071	3.4	< 0.712	<1.158	2.2	< 0.325	558.9	< 0.858	<1.008	19.5	17,654.0	<20.3	11,672.2	60,600	12.74	11,940
APCr-33	<1.582	48.7	< 0.071	0.6	5.6	<1.158	1.6	< 0.325	1,355.3	< 0.858	<1.008	57.9	167,456.7	43.9	17,565.2	423,400	11.68	54,350
APCr-34	<1.582	50.9	< 0.071	0.4	8.4	<1.158	1.2	< 0.325	2,007.8	< 0.858	<1.008	55.5	259,726.3	30.7	11,738.3	458,300	11.67	59,060
APCr-35	<1.582	1.4	< 0.071	1.7	< 0.712	<1.158	1.6	< 0.325	<2.209	< 0.858	<1.008	<2.483	30,002.0	<20.3	15,589.2	80,000	11.21	10,050
APCr-36	<1.582	25.5	< 0.071	< 0.201	< 0.712	<1.158	2.2	< 0.325	250.1	< 0.858	<1.008	50.3	148,925.9	33.1	20,210.4	279,400	12.49	29,430
APCr-37	<1.582	53.3	0.1	< 0.201	5.8	<1.158	1.2	< 0.325	943.4	< 0.858	<1.008	76.7	261,783.2	29.6	17,962.7	487,700	12.29	60,440
APCr-38	<1.582	279.5	< 0.071	< 0.201	< 0.712	<1.158	1.3	< 0.325	<2.209	< 0.858	<1.008	<2.483	93,580.9	<20.3	177.0	160,700	11.43	22,750
APCr-39	<1.582	256.0	< 0.071	< 0.201	< 0.712	<1.158	1.6	< 0.325	<2.209	< 0.858	<1.008	<2.483	96,394.2	<20.3	173.0	163,000	11.38	22,620
APCr-40	<1.582	295.5	< 0.071	< 0.201	< 0.712	<1.158	0.9	< 0.325	<2.209	< 0.858	<1.008	2.7	73,619.9	<20.3	53.0	139,400	12.25	21,800
APCr-41	<1.582	53.1	0.1	0.3	11.4	<1.158	1.0	< 0.325	2,569.5	< 0.858	<1.008	73.5	330,275.4	37.5	6,914.0	485,900	12.22	60,960
APCr-42	<1.582	58.5	< 0.071	< 0.201	0.7	<1.158	1.5	< 0.325	318.1	< 0.858	<1.008	122.2	151,631.2	34.1	16,602.7	289,900	12.47	40,340
Minimum	<1.582	0.5	< 0.071	< 0.201	< 0.712	<1.158	0.2	< 0.325	<2.209	< 0.858	<1.008	<2.483	<51.0	<20.3	53.0	20,000	9.13	1,875
Maximum	<1.582	295.5	0.1	36.3	11.4	<1.158	3.3	< 0.325	2,569.5	< 0.858	2.3	184.9	330,275.4	43.9	27,566.2	487,700	13.08	60,960
Average	<1.583	60.0	0.1	5.9	4.7	<1.158	1.6	< 0.325	780.8	< 0.858	<1.008	53.6	122,654.5	31.1	12,995.6	231,269	12.15	32,043
Coefficient of	0	130	0	150	77	0	54	0	00	0	0	86	67	20	58	57	7	50
variation %	0	139	0	150	//	0	54	0	27	0	0	00	07	20	50	57	/	50
Limit values for	r waste acc	eptance a	ıt hazardoı	ıs waste la	ndfill													
	25	300	5	70	100	2	30	40	50	5	7	200	25,000	50	50,000	100,000	-	-

Table 4.4 Results of leaching analysis on APCr (BS EN 12457-2, 2002).

Less than values (<) indicate that the results are less than the limits of detection

The values are expressed with no contribution from uncertainty of measurement The highlighted values exceed the limits for waste acceptance at hazardous waste landfills



Figure 4.6 Leaching of heavy metals from APCr according to BS 12457-2.

The environmental concern of APCr is due to the leaching of Ba, Cr, Cu, Mo, Pb and Zn, from which Ba and Mo were released from all the analysed APCr samples. Similar to the total metal contents, the leaching values of metals showed a considerable variability with CV% ranging from 54% to 150%.

Pb was the most concerning heavy metal because it showed the highest leaching rates (ranging from 7.0 to 2,569.5 mg/kg) and because it was the only metal whose leachates failed the regulation of WAC for hazardous waste landfill. As it is detailed in Table 4.4, for the batch of 26 analysed APCr samples, 77% of the samples released Pb and 62% exceeded the WAC limit value of 50 mg/kg for hazardous waste landfill. The high leachability of Pb is mainly due to chloride complexation (Hjelmar, 1996), but also, as explained by (Quina, et al., 2008b), is promoted by the alkaline pH (higher than 12 for most of the samples) generated when the APCr contact with water.

Despite the total contents of Zn being much higher than those of Pb, the leachability of Zn was low (max. 184.9 mg/kg), which is associated with the behaviour of this element at a high pH (Ecke, et al., 2002). Nevertheless, Zn as well as Ba, deserve attention due to their leaching rates as can be seen in Figure 4.6. Although both elements met the WAC for hazardous waste, the maximum leachable fraction of Zn, 184.9 mg/kg, is very close to the limit value of 200 mg/kg, and the three highest leachate concentrations of Ba (279.5, 256.0 and 295.5 mg/kg) are very close to its limit value of 300 mg/kg.

The metals As, Hg, Ni and Sb were not detected in any of the analysed leachates. Se and Cd were released from only one and two APCr samples, respectively, with leachate concentrations well under the limit values for hazardous waste. Table 4.4 and Figure 4.6 show that Cr, Cu and Mo never exceeded the regulatory of WAC limit values for hazardous waste.

The total contents and the amounts released during the leaching tests for Ba, Cr, Mo, Pb, Cu and Zn, were compared. Figure 4.7 shows the box plots of the leaching percentages for each of these metals. If the outliers (atypical data) are ignored, it can be seen that the maximum leaching rates, were 45% of Pb, 33% of Ba, 32% of Mo, 13% of Cr, 1.4% of Zn and 1% of Cu. Despite the high variability of the data regarding to total metal contents and leaching rates, shown in Table 4.2 and Table 4.4, respectively, the mean values of leaching percentage are within the interquartile ranges defined for each metal. Thus, it



can be said that for 75% of the APCr samples, the mean values of leaching percentages are 14% of Pb, 15% of Ba, 20% of Mo, 4% of Cr, 0.6% of Zn and 0.3% of Cu.

Figure 4.7 Box plots showing the leaching percentages of heavy metals in APCr. A) Ba, Cr, Mo and Pb B) Cu and Zn.

4.4.2 Leaching of soluble salts

A significant difference between the leaching rates of chlorides, sulphates and fluorides was observed as shown in Figure 4.8. Chlorides and sulphates were leached out from all of the analysed APCr samples, while 62% of them released fluorides. 23 out of the 25 APCr samples exceeded the hazardous WAC limit value of 25,000 mg/kg for chlorides. The leaching fractions of chlorides varied from 9,755.7 to 330,275.4 mg/kg, indicating that the chlorides represent a crucial parameter to be controlled in a subsequent APCr treatment aimed at disposal or recycling. In LWA incorporating APCr, the leaching of chlorides towards the concrete can cause corrosion of the reinforcement, which in turn would expand producing cracking of the concrete structure (Shraddhu, 2023).

The leachate concentrations of sulphates which ranged between 53.0 and 27,566.2 mg/kg were well under the limit value of 50,000 mg/kg for hazardous waste. Despite meeting the regulation for hazardous waste acceptance at landfill, the high leaching rates of sulphates should be carefully addressed in the treatment to enable APCr for recycling into LWA to avoid the chemical attack of these salts on concrete.

For the samples that released fluorides, the leachate concentrations of these salts varied from 22.8 to 43.9 mg/kg, meeting the WAC for hazardous waste that set the limit for these substances at a maximum of 50 mg/kg. These leaching rates are low and might be reasonably reduced or even eliminated to enable the material for a subsequent recycling.



Figure 4.8 Leaching of soluble salts from APCr according to BS 12457-2.

4.4.3 Total dissolved solids and conductivity

Figure 4.9 shows the conductivity values of the APCr leachates. It is clear that conductivity is directly related to Total Dissolved Solids (TDS). The TDS values varied between 20,000 and 487,700 and 88% of the analysed samples, exceeding the WAC limit value for hazardous waste of 100,000 mg/kg. The high contents of TDS in the APCr eluates are mainly due to the leaching of the soluble chlorides, such as NaCl and KCl, sulphates and some Ca-based salts.



Figure 4.9 Relationship between conductivity and TDS in the APCr leachates.

4.4.4 pH

Figure 4.10 shows the box plot of the pH values of the leachates obtained from the APCr samples. It can be seen that the pH was always highly alkaline, with values ranging from 11.01 to 13.08 (only one sample with a pH of 9.13 was an outlier). The average pH was 12.15 ± 0.8 . The high pH values of the leachate solutions demonstrate the alkaline nature of the APCr due to their high content of substances, such as hydroxides and lime, which are added during the flue gas cleaning process.



Figure 4.10 Box plot showing the pH of the APCr leachates.

4.5 Estimation of organic fraction by Total Organic Carbon (TOC), Dissolved Organic Carbon (DOC) and Loss on ignition (LOI)

The values determined for TOC, DOC and LOI in APCr are reported in Table 4.5 with the corresponding statistical analysis and comparison with the WAC limit values for hazardous waste. These parameters provide an estimation of the contents of organic matter in the different APCr samples.

	TOC	LOI at 550°C	DOC *
	%	%	mg/kg
APCr-02	5.3	7.4	<300
APCr-03	<1.64	6.6	424
APCr-04	<1.75	6.3	<300
APCr-05	4.7	0.4	<300
APCr-06	<1.9	6.2	653
APCr-07	<1.67	4.9	<300
APCr-08	<1.67	11.4	<300
APCr-09	<1.79	4.6	412
APCr-10	1.9	5.3	<300
APCr-11	<1.86	7.2	<300
APCr-13	<1.64	4.8	<300
APCr-14	<1.61	3.3	<300
APCr-15	7.1	15.1	<300
APCr-16	1.76	7.1	<300
APCr-17	1.62	5.5	<300
APCr-18	<1.64	13.5	<300
APCr-33	2.3	1.6	182
APCr-34	<1.94	3.6	<100
APCr-35	<1.75	7.0	<100
APCr-36	<1.67	2.0	<100
APCr-37	<1.66	2.5	<100
APCr-38	<1.57	0.9	<100
APCr-39	<1.72	3.5	<100
APCr-40	<1.6	2.5	<100
APCr-41	<1.93	4.4	<100
APCr-42	<1.9	0.0	<100
Minimum	<1.61	0.0	<100
Maximum	7.1	15.1	653
Average	3.5	5.3	418
CV %	61	70	46
Limit value for h	azardous waste	as expressed in Landfill	Directive
	6	10	1.000

Table 4.5 TOC, LOI and DOC in APCr.

* DOC was determined in the eluate obtained from the leaching test

According to the data reported in Table 4.5, TOC was detected in 7 out of the 26 APCr samples with a maximum concentration of 7.1%, while the highest rate of LOI at 550°C was 15.1%. The maximum values for both TOC and LOI exceeded the WAC for hazardous waste, 6% and 10% respectively. Also, there is significant variability in the results, given the high CV% for both parameters. Therefore, the box plots for TOC and for LOI were generated for further analysis of the data, as it is shown in Figure 4.11. When detected, the TOC concentrations ranged from 1.6% to 7.1%, this maximum value

is not an outlier and exceeds the limit of WAC for hazardous waste of 6%. For LOI at 550°C, it can be said that the proportion of this parameter varied from 0% to 15.1% in the analysed APC samples; although the highest values are outliers, the maximum accepted data (11.4%) still exceeds the WAC for hazardous waste of 10%. It can be concluded that both TOC and LOI are parameters of interest for the characterisation and treatment for subsequent recycling of the analysed APCr. While the detection of TOC in some of the APCr samples indicates the eventual presence of organic substances, such as dioxins and furans, produced during the waste combustion, the LOI is related to the presence of organic matter and the activated carbon added during the flue gas cleaning process.



Figure 4.11 Box plots for the estimation of organic fraction in APCr. A: TOC, B: LOI.

Despite the levels of TOC and LOI eventually exceeding the regulatory limits for hazardous WAC, the leachable organic fraction, indicated by the DOC in the leaching eluates, was detected in only 4 out of the 26 analysed APCr samples at rates between 182 and 653 mg/kg, values that accomplished the regulatory limit of 1,000 mg/kg. The absence of DOC in the majority of the eluates, suggests that the organic fraction of APCr, characterised by dioxins and furans among other contaminants, strongly remains in the solid phase. In fact, (Robinson, et al., 2004) who reported levels of DOC from 94 to 448 mg/kg in APCr from the UK, found that dioxins and furans were not detected in APCr eluates. This is advantageous for a subsequent recycling of APCr in LWA because it can be considered that the leaching of the organic fraction is not a concerning parameter.

4.6 Mineralogy of APCr

4.6.1 Qualitative analysis of mineral phases

The qualitative analysis of the mineral composition of APCr was determined by the identification of the different peaks in the XRD patterns of 42 APCr samples, according to the COD database. Figure 4.12 and Figure 4.13 show the XRD patterns with the identification of the main peaks. It must be clarified that in these graphs only the highest peaks have been labelled. The overlapping of peaks created a difficulty for mineral phase identification; therefore, the candidate phases were carefully contrasted with the elemental composition and leaching results of the APCr to ensure the appropriate mineral characterisation of the residues.



Figure 4.12 XRD patterns showing identification of some peaks for samples APCr-01 to APCr-21.



Figure 4.13 XRD patterns showing identification of some peaks for samples APCr-22 to APCr-42.

The complete list of mineral phases identified in APCr is presented in Table 4.6. The most common phases were Ca-based minerals, including calcite (CaCO₃), anhydrite (CaSO₄), calcium hydroxychloride (CaOHCl), portlandite (Ca(OH)₂), soluble salts including halite (NaCl) and sylvite (KCl), and quartz (SiO₂). This mineral composition is in good agreement with previous studies conducted in the UK by the authors referenced in Table 4.6. Other mineral phases commonly detected in the analysed APCr samples included bassanite (CaSO₄•0.5H₂O), clinotobermorite (Ca₅H₈O₂₁Si₆), magnesium oxide hydroxide (Mg₃O₂(OH)₂) and zincowoodwardite (Al_{0.375}H₆O_{2.686}S_{0.188}Zn_{0.625}). These last two phases as well as bottalockite (Cu₂(OH)₃Cl), cerussite (PbCO₃) and tobermorite (Ca_{2.25}H_{3.5}O₁₀Si₃), have been reported only in APCr from different EfW facilities in the UK by (Bogush, et al., 2015).

It should be noted that 21 crystalline phases were detected for the first time in this study as present in different APCr samples, as it is detailed in Table 4.6. This is due to the sampling stage comprised of a cross-section of several EfW facilities. From these phases, it is worth mentioning that the most common were the silicon enriched minerals clinotobermorite $Ca_5H_8O_{21}Si_6$ and kumdykolite NaAlSi₃O₈ which were detected respectively in 26% and 19% of the total number of the analysed APCr samples.

Some of the identified mineral phases were enriched with the heavy metals that were previously identified in the leaching analysis: Pb was present in anglesite (PbSO₄), cerussite (PbCO₃), litharge (PbO) and tetrapotassium lead oxide (K₄PbO₄); Zn was present in potassium tetrachloro zincate (K₂ZnCl₄), zinc chloride (ZnCl₂), zinc oxysulphate (Zn₃O(SO₄)₂), zincite (ZnO) and zincowoodwordite (Al_{0.375}H₆O_{2.686}S_{0.188}Zn_{0.625}); Cd was present in the form of monteponite (CdO), and nickel was present as nickel ferrite (NiFe₂O₄).

Table 4.6 Mineralogy of APCr.

		Number	% of	Dhagag	Min	eral pl stu	hases ro idies in	eported the UI	by oth K	er
Mineral phases	Chemical formula	of samples that contain the phase	samples that contain the phase	reported in APCr for the first time	(Fernández Bertos, et al., 2004c)	(Sun, et al., 2008)	(Dimech, et al., 2008)	(Bogush, et al., 2015)	(Araizi, et al., 2016)	(Bogush, et al., 2019)
Akermanite	Ca ₂ MgSi ₂ O ₇	5	12	х						
Albite	NaAlSi ₃ O ₈	2	5	х						
Anatase	TiO ₂	8	19	х						
Anglesite	PbSO ₄	2	5	х						
Anhydrite	CaSO ₄	26	62		х	Х	х	Х	х	Х
Anorthite	CaAl ₂ Si ₂ O ₈	1	2							
Arcanite	K_2SO_4	5	12							
Bassanite	CaSO ₄ •0.5H ₂ O	10	24							
Bottalockite	Cu ₂ (OH) ₃ Cl	4	10					Х		
Calcite	CaCO	37	88		x	x	x	х	x	x
Calcium Hydroxychloride	CaOHCl	28	67		A	x	x	x	x	x
Cerussite	PhCO ₂	3	7				А	x	А	11
Chloranatite	Ca-(PO_)-Cl	3	7	v				21		
Clinotobermorite	Ca-HoOn Sic	11	26	v						
Dipotassium oxide	K.O	2	5	A V						
Ettringite	K_2O	2	7	A V						
Cablanita	$Ca_{6}Al_{2}(304)_{3}(011)_{12}^{-2}011_{2}^{-2}0$	2	7	х				v		v
Genienite	$Ca_2AI(AISIO_7)$	3	2		х			л		А
Grossite	$CaAl_4O_7$	1 5	2	х						
Gypsum	$CaSO_4 \bullet 2H_2O$	20	12					37		37
Halite	NaCl	32	/6		х		х	Х	х	Х
Hematite	Fe_2O_3	2	5							
Kumdykolite	NaAlS ₁₃ O ₈	-	17	х						
Larnite	$Ca_2S_1O_4$	5	12	х						
Lime	CaO	5	12			Х		Х	х	Х
Litharge	РЬО	3	7							
Magnesium oxide hydroxide	$Mg_3O_2(OH)_2$	10	24					Х		
Monteponite	CdO	2	5	х						
Nickel ferrite	NiFe ₂ O ₄	2	5	х						
Periclase	MgO	1	2			Х				
Perovskite	CaTiO ₃	5	12	х						
Phosphorous pentoxide	P_2O_5	2	5	х						
Portlandite	Ca(OH) ₂	25	60		Х	Х	х	Х	х	
Potassium tetrachloro zincate	K_2ZnCl_4	7	17							
Pseudowollastonite	CaSiO ₃	2	5	х						
Quartz	SiO_2	23	55		Х	Х		Х		Х
Rutile	TiO ₂	1	2	х						
Sanidine	KAlSi ₃ O ₈	5	12	х						
Sylvite	KCl	33	79		Х	Х	х	Х		Х
Tetrapotassium lead oxide	K ₄ PbO ₄	1	2	х						
Tobermorite	Ca _{2.25} H _{3.5} O ₁₀ Si ₃	7	17					Х		
Whitlockite	$Ca_3(PO_4)_2$	4	10							
Zinc chloride	ZnCl ₂	7	17	х						
Zinc oxysulphate	$Zn_3O(SO_4)_2$	2	5	х						
Zincite	ZnO	4	10							
Zincowoodwardite	Al _{0.375} H ₆ O _{2.686} S _{0.188} Zn _{0.625}	16	38					Х		

Highlighted x indicate phases that have been identified only in the UK

The Ca-based mineral phases present in APCr are the result of the reactions that take place during the neutralisation of the acid gases (such as hydrochloric acid HCl, sulphur dioxide SO₂ and carbon dioxide CO₂), for which alkaline agents, including portlandite Ca(OH)₂ and lime CaO, are added into the flue gas. Calcite CaCO₃ was the most common phase, it was present in 88% of the analysed samples. This is the product of the neutralisation of carbon dioxide with portlandite, according to Equation 4.1 (Fan, et al., 2022).

Equation 4.1 $Ca(OH)_2 + CO_2 \rightarrow CaCO_3 + H_2O$

Calcium hydroxychloride CaOHCl is the first product to form when hydrochloric acid is absorbed by lime CaO in hot flue gases, according to the mechanism described from Equation 4.2 to Equation 4.5 (Partanen, et al., 2005). In agreement with previous studies on the mineral composition of APCr, referenced in Appendix II, calcium hydroxychloride was detected in the majority of the samples, while calcium chloride CaCl₂ was not detected in any of the samples. It is likely that hydrochloric acid was completely neutralised by lime, so that, there was not a remaining amount of acid to promote the reaction in Equation 4.3, and consequently Equation 4.4 and Equation 4.5 did not take place. It is also possible that calcium hydroxychloride is stable enough to not react with an excess of hydrochloric acid as per Equation 4.3. Therefore, the formation of calcium hydroxychloride from lime, which takes place in dry and semi-dry scrubbing systems, can be attributed to the reaction described in Equation 4.2.

Equation 4.2 CaO $_{(s)}$ + HCl $_{(g)} \rightleftharpoons$ CaOHCl $_{(s)}$

Equation 4.3 CaOHCl (s) + HCl (g) \Leftarrow CaCl₂•H₂O (s)

Equation 4.4 CaCl₂•H₂O (s) \Leftarrow CaCl₂(s) + H₂O (g)

Equation 4.5 CaCl₂•H₂O _(s) + O_{2 (g)} \rightleftharpoons Ca(ClO)_{2 (s)} + H₂O _(g) 143 The formation of calcium hydroxychloride from the interaction of portlandite Ca(OH)₂ with gaseous HCl was described by (Jozewicz & Gullett, 1995) according to Equation 4.6, 4.7 and 4.8. Equation 4.8 proceeds when the reaction is incomplete and involves the neutralisation of only one hydroxide group (Bodénan & Deniard, 2003). This incomplete reaction can be more likely to occur than the complete reaction described in Equation 4.6, as per their standard enthalpies of neutralisation ΔH_n calculated by (Jozewicz & Gullett, 1995) from laboratory experiments (-25.787 and -17.754 kJ/mol for Equation 4.6 and Equation 4.8, respectively). Consequently, in the APCr samples analysed in this research, the formation of calcium hydroxychloride from portlandite can be attributed mainly to the reaction described in Equation 4.8. In any case, if CaCl₂ was formed as per Equation 4.6, it completely reacted with portlandite as indicated in Equation 4.7 increasing the yield of calcium hydroxychloride.

Equation 4.6 $Ca(OH)_2 + 2HCl \rightarrow CaCl_2 + H_2O$

Equation 4.7 $Ca(OH)_2 + CaCl_2 \rightarrow 2CaOHCl$

Equation 4.8 $Ca(OH)_2 + HCl \rightarrow CaOHCl + H_2O$

Anhydrite CaSO₄ is one of the products of the neutralisation of sulphur dioxide, according to the reaction described in Equation 4.9 (Bodénan & Deniard, 2003; Fan, et al., 2022). This reaction particularly occurs in dry and semi-dry scrubbing systems. Whereas, the hydrated form Gypsum CaSO₄•2H₂O is produced in wet scrubbing systems, according to Equation 4.10. The aging of APCr may cause the partial conversion of gypsum into bassanite CaSO₄•0.5H₂O or into anhydrite, as per Equation 4.11 (Bayuseno & Schmahl, 2011; Priharyoto, 2006).

Equation 4.9 $Ca(OH)_2 + SO_2 + \frac{1}{2}O_2 \rightarrow CaSO_4 + 2H_2O_4$

Equation 4.10 $Ca(OH)_2 + SO_2 + \frac{1}{2}O_2 \rightarrow CaSO_4 \cdot 2H_2O$ Equation 4.11 CaSO₄•2H₂O \rightarrow CaSO₄ + 2H₂O

The excess of alkaline chemicals added for neutralisation of flue gases becomes part of APCr. Nevertheless, lime reacts with water in semi-dry scrubbing systems to form portlandite according to Equation 4.12 (Fan, et al., 2022). This indicates that usually, lime is completely consumed during the flue gas cleaning, and explains why lime was detected in only 12% of the analysed APCr samples, whereas portlandite was present in 60% of them.

Equation 4.12 CaO + H₂O \rightarrow Ca(OH)₂

Halite comes from the original compositions of the MSW, but there is also a fraction of this phase that is produced from the neutralisation of hydrochloric acid with sodium bicarbonate NaHCO₃ according to the reaction described in Equation 4.13 (Bodénan & Deniard, 2003). This reaction takes place in dry-scrubbing systems, which use sodium bicarbonate as alkaline additive.

Equation 4.13 NaHCO₃ + HCl \rightarrow NaCl + CO₂ + 2H₂O

4.6.2 Quantitative analysis of mineral phases by the Rietveld method

Table 4.7 summarises the quantification of APCr crystalline phases by the Rietveld method in terms of minimum and maximum wt. %. registered for the batch of 42 APCr samples. The complete mineral characterisation of each sample is presented in detail in Appendix VIII. The most abundant mineral phases were anhydrite, calcite, calcium hydroxychloride, portlandite, halite, sylvite and quartz. These phases showed the highest values, ranging from 15 wt.% to 44 wt.%. Other Ca-based phases, including bassanite, clinotobermorite and tobermorite showed maximum concentrations around 17 wt.%.

Mineral	wt.%						
Name	Formula	Miı	nimu	ım	Maximum		
Akermanite	Ca ₂ MgSi ₂ O ₇	2.21	±	0.19	7.90	±	0.27
Albite	NaAlSi ₃ O ₈	0.30	±	0.30	6.54	±	1.78
Anatase	TiO ₂	0.13	±	0.13	3.34	±	0.84
Anglesite	PbSO ₄	0.14	±	0.14	0.56	±	0.24
Anhydrite	CaSO ₄	0.13	±	0.05	14.59	±	0.68
Anorthite	$CaAl_2Si_2O_8$	1.78	±	0.10	1.78	±	0.10
Arcanite	K_2SO_4	0.11	±	0.02	7.59	±	0.46
Bassanite	$CaSO_4 \bullet 0.5 H_2 O$	1.26	±	0.17	16.69	±	0.49
Bottalockite	Cu ₂ (OH) ₃ Cl	0.02	±	0.01	9.00	±	1.27
Calcite	CaCO ₃	1.34	±	0.67	18.33	±	0.46
Calcium Hydroxychloride	CaOHCl	1.48	±	0.18	43.48	±	0.44
Cerussite	PbCO ₃	0.01	±	0.00	1.37	±	0.07
Chlorapatite	Ca ₅ (PO ₄) ₃ Cl	0.19	±	0.01	7.52	±	0.70
Clinotobermorite	$Ca_5H_8O_{21}Si_6$	0.37	±	0.05	16.71	±	0.75
Dipotassium oxide	K ₂ O	0.82	±	0.15	0.97	±	0.24
Ettringite	Ca ₆ Al ₂ (SO ₄) ₃ (OH) ₁₂ •26H ₂ O	0.11	±	0.11	9.55	±	0.63
Gehlenite	$Ca_2Al(AlSiO_7)$	0.87	±	0.77	7.31	±	0.86
Grossite	CaAl ₄ O ₇	3.05	±	0.20	3.05	±	0.20
Gypsum	CaSO ₄ •2H ₂ O	0.12	±	0.01	6.56	±	0.57
Halite	NaCl	0.38	±	0.45	14.99	±	0.55
Hematite	Fe ₂ O ₃	0.36	±	0.36	1.31	±	0.50
Kumdykolite	NaAlSi ₃ O ₈	0.20	±	0.20	4.20	±	0.96
Larnite	Ca_2SiO_4	1.20	±	0.07	21.73	±	1.00
Lime	CaO	0.44	±	0.18	1.45	±	0.19
Litharge	РЬО	0.07	±	0.02	0.82	±	0.11
Magnesium oxide hydroxide	$Mg_3O_2(OH)_2$	0.01	±	0.00	8.34	±	0.69
Monteponite	CdO	0.07	±	0.02	0.21	±	0.02
Nickel ferrite	NiFe ₂ O ₄	0.41	±	0.23	0.90	±	0.11
Periclase	MgO	0.72	±	0.07	0.72	±	0.07
Perovskite	CaTiO ₃	0.10	±	0.02	13.79	±	0.59
Phosphorous pentoxide	P_2O_5	0.83	±	0.21	1.64	±	0.11
Portlandite	Ca(OH) ₂	0.87	±	0.24	30.36	±	0.68
Potassium tetrachloro zincate	K_2ZnCl_4	0.18	±	0.05	2.71	±	0.91
Pseudowollastonite	CaSiO ₃	0.86	±	0.19	2.13	±	0.34
Quartz	SiO ₂	0.01	±	0.01	19.46	±	0.46
Rutile	TiO ₂	13.60	±	1.69	13.60	±	1.69
Sanidine	KAlSi ₃ O ₈	0.01	±	0.00	7.94	±	0.95
Sylvite	KCl	0.87	±	0.10	22.62	±	0.53
Tetrapotassium lead oxide	K ₄ PbO ₄	0.66	±	0.14	0.66	±	0.14
Tobermorite	$Ca_{2.25}H_{3.5}O_{10}Si_3$	0.50	±	0.06	16.55	±	1.36
Whitlockite	$Ca_3(PO_4)_2$	1.84	±	0.46	12.24	±	1.19
Zinc chloride	ZnCl ₂	0.06	±	0.19	6.61	±	0.81
Zinc oxysulphate	$Zn_3O(SO_4)_2$	0.04	±	0.00	0.42	±	0.04
Zincite	ZnO	0.08	±	0.03	0.71	±	0.09
Zincowoodwardite	Al _{0.375} H ₆ O _{2.686} S _{0.188} Zn _{0.625}	0.01	±	0.00	5.82	±	0.87

Table 4.7. Mineral	phases of APCr sam	ples determined b	y XRD analysis.

The formation of calcium hydroxychloride is favoured by the solid-gas interactions in dry and semi-dry scrubbing systems, rather than the liquid-gas interactions in wet scrubbing processes (Bodénan & Deniard, 2003). Certainly, in APCr samples with the highest water contents reported in Table 4.1 (APCr-14 to APCr-18, APCr-31 and APCr-35 with water contents from 21.6% to 53.3%) which were assumed to come from wet scrubbing systems, calcium hydroxychloride was not detected (see Appendix VIII). The high contents of Cl-bearing mineral phases, including calcium hydroxychloride, halite and sylvite were in agreement with the high leaching rates determined for these soluble salts.

4.6.3 Estimation of the amorphous fraction

The diffractograms of some of the APCr samples revealed the presence of amorphous material, given the deformations (humps) in the background mainly between 20 °2 θ and 40 °2 θ and the presence of some broadened peaks. For example, it can be seen in Figure 4.14 that APCr-23 had a low content of amorphous material because the background of its diffractogram did not deform, while both APCr-02 and APCr-30 exhibited humps around 30°2 θ and 27 °2 θ , respectively. The amorphous content may be non-crystalline or poorly crystalline material (Bogush, et al., 2015; Bodénan & Deniard, 2003).



Figure 4.14. Comparison of diffractograms of APCr samples according to their contents of amorphous material.

The quantification of the total contents of crystalline and amorphous material for each APCr sample is detailed in Appendix VIII. The summary of the results can be observed in Figure 4.15. The total content of crystalline phase in each APCr sample was calculated as the sum of wt.% of all mineral phases quantified in the sample. The values ranged from 13.53 ± 0.58 to 76.73 ± 1.99 wt.% of crystalline material. The percentage of amorphous material was calculated by deducting the wt.% of crystalline material from 100%, obtaining values that varied between 23.27 ± 1.99 % and 86.47 ± 0.58 %.

0	% 10%	20%	30%	40%	50%	6	0%	70%	80%	90%	100%
APCr-01	20.37%	H-I								79.6	3%
APCr-02	39.39%			н						60.6	1%
APCr-03	37.99%			H-1						62.0	1%
APCr-04	52.14%				F					47.8	6%
APCr-05	59.91%					F				40.0	9%
APCr-06	50.40%									49.6	0%
APCr-07	45.65%				H					54.3	5%
APCr-08	46.47%				H-1					53.5	3%
APCr-09	52.76%				ŀ					47.2	4%
APCr-10	70.91%									29.0	9% 1
APCr-11	60.05%					F				39.9	5%
APCr-12	60.92%						H			39.0	8%
APCr-13	27.00%		н							73.0	0%
APCr-14	53.36%					H				46.6	4%
APCr-15	42.22%			H						57.7	8%
APCr-16	52.44%				ł					47.5	6%
APCr-17	54.04%					H-I				45.9	6%
APCr-18	58.84%					F				41.1	6%
APCr-19	60.52%						H			39.4	8% 1
APCr-20	69.06%							H		30.9	4%
APCr-21	43.52%			H	-					56.4	8% 1
APCr-22	38.54%			H						61.4	6%
APCr-23	71.93%							н		28.0	7%
APCr-24	51.59%				I	н				48.4	1%
APCr-25	58.05%					н				41.9	5%
APCr-26	31.34%		H							68.6	6%
APCr-27	22.84%	H								77.1	6%
APCr-28	41.00%			H-I						59.0	0%
APCr-29	38.12%			н						61.8	8% 1
APCr-30	13.53% н									86.4	7% (
APCr-31	58.05%						-			41.9	5%
APCr-32	76.73%							I		23.2	7% I
APCr-33	60.32%						н			39.6	8% 1
APCr-34	72.02%							H		27.9	8% 1
APCr-35	43.73%			F	H					56.2	7%
APCr-36	53.92%					н				46.0	8%
APCr-37	49.30%				н					50.7	0% 1
APCr-38	59.55%					H				40.4	5%
APCr-39	36.02%		H							63.9	8%
APCr-40	60.47%						н			39.5	3% 1
APCr-41	39.01%			н						60.9	9%
APCr-42	40.86%			н						59.1	4%
		■ % (Crystaline	e material	• • •	6 Amor	phous r	naterial			

Figure 4.15 Crystalline and amorphous fractions in APCr.

4.7 Microstructure and local elemental composition

The microstructure and the local elemental composition of APCr were investigated by SEM/EDS. Appendix IX shows the SEM micrographs and their corresponding SEM/EDS spectra for 42 APCr samples. It was found that APCr consist of a fine-grained material in which particles with different shapes and sizes are spread. The main particle shapes that were observed include spheres, cubes, dipyramids, prismatic particles and hexagonal particles. Also, pieces of unburnt organic matter, carbon and metals were found. These particle structures will be described in the following sections.

4.7.1 Fine-grained material

APCr mainly consisted of fine-grained material (Figure 4.16 A) with particle sizes ranging from 0.1 to 1 μ m. The material was mostly composed of O, Ca and Cl, with presence of S, K, Mg, Na, Si, Zn, Al, Fe, Cu and P. Although unshaped particles were the dominant structures in fine material, some spheres, cubes and other shapes were also observed. It is known that this fine material in APCr contains the different forms of the mineral phase CaOHCl, such as blocks of about 10 μ m, and very fine spheres of no more than 1 μ m (Bogush, et al., 2015). For example, Figure 4.16 B shows some blocks of layers of about 1 μ m each. Due to its lightness, the fine-grained material adheres to the surface of bigger particles, covering their structure.



Figure 4.16 SEM images of APCr: (A) Fine-grained material from APCr-42, (B) Blocks of fine-grained material from APCr-36.

4.7.2 Spherical particles

Plain spherical particles (Figure 4.17 A), with sizes ranging from 10 to 130 μ m, were mostly composed of O, Si and Ca, with presence of moderate concentrations (1 – 5 %) of Fe, Al, Na, K, Cl, Zn, S, Mg, as well as small quantities (less than 1%) of Pb, Ti, Cu, P and Mn. It has been reported that spherical particles in APCr are aluminosilicates (Bogush, et al., 2015). In the APCr samples analysed in this study, these spheres are specifically calcium aluminosilicates according to the SEM/EDS local map spectra.

Spheres with holes (Figure 4.17 B), with sizes ranging from 55 to 62 μ m, showed the same elemental composition as the plain spheres but were generally enriched in K and contained fine-grained impurities of Ca that accumulated in their holes.



Figure 4.17 SEM images of Spherical particles of APCr: (A) Plain spherical particle from APCr-08, (B) Holey sphere from APCr-08.

Hemispheres (Figure 4.18 A and B) with sizes from 14 to 86 µm were observed in some of the APCr samples. These particles showed in general the same elemental composition of plain and holey spheres: mostly O, Si and Ca, with moderate concentrations of Fe, Al, Na, K, Cl, Zn, S, Mg, and small quantities of Pb, Ti, Cu, P and Mn. Therefore, it can be said that they were likely crushed spheres. Hemispheres exhibited better the microstructure and composition of spheres. They showed a solid thick structure mainly composed of O, Ca and S (Figure 4.18 A) or O, Si, Ca and Fe (Figure 4.18 B), and a non-concentric hollowed area that can be shallow (Figure 4.18 A) or deep (Figure 4.18 B) where fine-grained impurities, mainly of Ca, were deposited.



Figure 4.18 SEM images and spectrum of hemispheres in APCr: (A) Hollow hemisphere from APCr-13, (B) Hollow hemisphere from APCr-02.

Hollow spheres, named cenospheres (Figure 4.19 A and B) were found in APCr, with sizes between 11 and 14 μ m, and composed mainly of O, Ca and Si, with moderate amounts of Fe, Al and Mg, and at lesser extent P, S, Mn, Cl and Na. In agreement with this composition, it is known that the shells of cenospheres consist of aluminosilicate phases (Ranjbar & Kuenzel, 2017). Cenospheres were perfectly rounded spheres with a hole (Figure 4.19 A) or flattened spheres with a hole (Figure 4.19 B). Flattened cenospheres were always covered by fine-grained material containing mostly Ca. Cenospheres that result from waste incineration have special properties, including, being lightweight (as they are filled with air or inert gases), having high thermal stability and high strength (Ranjbar & Kuenzel, 2017). These properties are particularly important for the potential use of APCr in LWA manufacture.



Figure 4.19 SEM images of spherical particles in APCr: (A) Hollow sphere from APCr-18, (B) Flattened hollow sphere from APCr-27.

The lowest size spheres (1 to 10 μ m) were found in APCr among the fine-grained material, as shown in Figure 4.20 A and B. These particles were mainly composed of O, Ca and Cl, with some amounts of K and Na. Given their main components and abundance in the majority of the APCr samples, these particles are likely one of the forms of calcium hydroxychloride CaOHCl.



Figure 4.20 SEM images of spherical particle in APCr: (A) Sphere from APCr-33, (B) Spheres among fine grained material from APCr-16.

4.7.3 Crystal forms and aggregates

Cubes like those shown in Figure 4.21 were found in APCr samples with sizes around 5 μ m. These particles were composed of Na and Cl and correspond to the crystalline phase of halite. Due to their small sizes and brightness similar to the grained material, it was difficult to acquire images of these particles in the samples, despite it was known that halite was present in over 75% of them.



Figure 4.21 SEM images of cubic particles in APCr: Cubes among fine grained material from APCr-05.

Groups of octahedral bipyramids (Figure 4.22) were observed in a few APCr samples with sizes around 6 μ m. Given their shape and their elemental composition consisting of K and Cl, these particles are one of the crystalline forms of sylvite, KCl. These bipyramids exhibited brightness and size similar to the fine-grained material. It was difficult to observe them, although it was known that almost 85% of the APCr samples contained sylvite.



Figure 4.22 SEM images of APCr: (A) Octagonal bipyramid from APCr-34.

Tabular particles (Figure 4.23) of size around 30 μ m were found in APCr. They were mainly composed of O, Ca and S. These particles are one of the crystalline forms of gypsum, CaSO₄•2H₂O.



Figure 4.23 SEM Images of APCr: Rhombohedral particle from APCr-13.

Hexagonal clusters (Figure 4.24) with sizes around 13 μ m were found in some APCr samples and were mainly composed of Ca, O and C. These particles are one of the forms of calcite. Although there are different crystal shapes of calcite, those were not observed under the analysis conditions.



Figure 4.24 SEM images of APCr: Hexagonal particle from APCr-17.

Hexagonal particles were found in APCr, rarely as isolated particles (Figure 4.25 A) and more commonly forming part of needle-like aggregates (Figure 4.25 B). These particles consisted mainly of Ca, Cl and O and are likely one of the forms of calcium hydroxychloride.



Figure 4.25 SEM images of APCr: (A) Hexagonal particle from APCr-04, (B) Needlelike aggregate from APCr-01.

4.7.4 Metallic particles

In some of the APCr samples, spherical shells with diameters around 15 μ m (Figure 4.26) or pieces of broken shells, whose composition was mostly Fe, were found throughout the grained material. This type of spherical particles, called ferrospheres, are commonly found in incineration residues, like fly ash. At the waste incineration temperatures, Febased wires, filaments and meshes from electronic and electrical devices, can be evaporated and then condensed into microspheres. In proportion to their contents of Fe, the ferrospheres can be composed of ferro-oxides (over 75% Fe), aluminosilicate-bearing ferro-oxides (50-75% Fe), high ferriferous aluminosilicates (25-50% Fe) and ferro-aluminosilicates (less than 25% Fe) (Zhao, et al., 2006). In the analysed APCr samples, the composition of ferrospheres was around 80% Fe, so that, they can be associated with the presence of ferro-oxides, for instance, with the mineral phase hematite Fe₂O₃.



Figure 4.26 SEM images of APCr: Metallic shell from APCr-11.

In one of the APCr samples, a laminar particle with a size of around 34 μ m was found among the fine-grained material (Figure 4.27). It consisted of 82% Ti with small amounts of O and impurities of Ca, Zn, K, Al, Fe, Cl, S and Na. This particle was likely of rutile, TiO₂.



Figure 4.27 SEM images of APCr: Laminar piece from APCr-06.

Several fragments of aluminium were found in different APCr samples, such as crushed laminas (Figure 4.28 A) or fibres (Figure 4.28 B). These particles are likely unburnt aluminium foil coming from the waste. The fine-grained material, mainly composed of O, Ca and Cl, deposited on the surface of the laminar pieces of Al, specifically in the cracks, while the Al fibres appeared tangled between aggregates of fine material.



Figure 4.28 SEM images of APCr: (A) Crushed laminas of aluminium foil from APCr-10, (B) Fibres of aluminium foil from APCr-38.

4.7.5 Unburnt organic matter

Laminar structures of unburnt organic matter with wrinkled surface (Figure 4.29 A) or porous surface (Figure 4.29 B) were observed in most of the APCr samples. These particles are probably residues of food or vegetation that commonly form part of MSW. The elements associated to these residues were Na, P, Si, Zn, Al, Mg and Fe. The finegrained material was deposited in the pores and wrinkles of the unburnt particles.



Figure 4.29 SEM images of APCr: (A) Unburnt organic matter from APCr-28, (B) Unburnt organic matter from APCr-32.

5 Chapter 5 Treatment of APCr

In this chapter, the methods of washing with water and carbonation were investigated for treatment of APCr. The experiments aimed to optimise the conditions of both methods to improve the characteristics and enable the APCr for further recycling into the manufacture of LWA. The treated residues were tested for elemental composition, mineralogy, microstructure and leaching properties.

5.1 Water washing

5.1.1 Optimisation of water washing of APCr

The washing of APCr was assessed at three different extraction times and L/S ratios. The washing conditions and leaching results for APCr-m before and after the treatment and the comparison with the regulatory values for hazardous waste acceptance at landfill are presented in Table 5.1.

BS 12457-2, LS = 10 (mg/kg)													
	Washed APCr												
	L/S ratio \rightarrow		2.5			5			value for				
Ti	me (min) \rightarrow	5	30	60	5	30	60	5	30	60	hazardous		
	APCr-m	T1	T2	T3	T4	T5	T6	Τ7	T8	T9	WAC		
As	<1.582	<1.582	<1.582	<1.582	<1.582	<1.582	<1.582	<1.582	<1.582	<1.582	25		
Ba	12.5148	7.8	5.7	5.6	7.0	6.1	5.8	6.6	6.3	6.0	300		
Cd	0.0932	0.1	< 0.071	0.1	< 0.071	< 0.071	< 0.071	< 0.071	< 0.071	< 0.071	5		
Cr	3.2208	2.3	3.0	2.6	1.9	2.9	3.4	2.2	3.3	3.0	70		
Cu	0.727	< 0.712	< 0.712	< 0.712	< 0.712	< 0.712	< 0.712	< 0.712	< 0.712	< 0.712	100		
Hg	<1.158	<1.158	<1.158	<1.158	<1.158	<1.158	<1.158	<1.158	<1.158	<1.158	2		
Mo	2.1436	2.0	1.8	1.8	2.0	1.9	1.9	2.0	2.0	1.8	30		
Ni	< 0.325	< 0.325	< 0.325	< 0.325	< 0.325	< 0.325	< 0.325	< 0.325	< 0.325	< 0.325	40		
Pb	193.025	62.5	61.3	61.7	74.6	67.7	68.6	63.5	71.3	62.6	50		
Sb	< 0.858	< 0.858	< 0.858	< 0.858	< 0.858	< 0.858	< 0.858	< 0.858	< 0.858	< 0.858	5		
Se	<1.008	<1.008	<1.008	<1.008	<1.008	<1.008	<1.008	<1.008	<1.008	<1.008	7		
Zn	42.3476	12.4	16.1	13.9	18.6	15.4	16.1	15.4	20	19	200		
Chlorides	114,102.9	34,767.2	21,617.3	14,153.5	34,126.8	20,528.3	12,007.3	32,809.5	20,440.3	12,517.4	25,000		
Fluorides	<20.3	<20.3	<20.3	<20.3	<20.3	<20.3	<20.3	<20.3	<20.3	<20.3	500		
Sulphates	15,230.3	12,703.4	12,245.7	9,142.8	15,078.2	13,102.8	10,722.7	14,548.6	12,612.7	9,139.8	50,000		
TDS	219,600	97,500	72,400	56,900	98,800	74,200	58,300	97,900	73,200	55,400	100,000		
DOC	<100	109	<100	<100	<100	<100	<100	111	<100	<100	1,000		
					BS 12457-2	, LS/10							
pH at 20°C	12.51	12.61	12.63	12.70	12.60	12.63	12.70	12.60	12.65	12.65	-		
Conductivity at 20°C (µS/cm)	32,680	15,520	12,600	10,730	16,090	12,440	10,360	15,380	12,610	10,580	-		

Table 5.1 Leaching results of APCr-m before and after washing at different washing conditions.
Given the differences in leaching rates of APCr-m before and after washing, it can be seen that, independently of the L/S ratio and the extraction time, the washing treatment reduced the leaching of the metals Ba, Cr, Mo, Pb and Zn, and the salts chlorides and sulphates. The stabilisation of these metals and soluble salts caused significant reductions of the TDS concentration and in the conductivity of the leachates. Pb, chlorides and TDS exceeded their corresponding regulatory values for leaching in the untreated APCr-m. After washing, at any of the evaluated conditions, the leaching of Pb did not meet the required maximum value of 50 mg/kg, but the L/S ratio 2.5 produced leaching rates of Pb closer to this limit. The leaching of chlorides from w-APCr only met the limit value of 25,000 mg/kg when the extraction times were 30 or 60 min, achieving lower leaching rates at 60 min of washing. The concentrations of TDS in the leachates from w-APCr were reduced to values lower than the limit of 100,000 mg/kg at any of the tested washing conditions, but this reduction was enhanced as the extraction time increased, so that the lowest levels of TDS were achieved at 60 min of washing. It could be said that washing APCr at a L/S ratio 2.5 and extraction times between 30 to 60 min ensures that the leaching rates of the washed material comply with the regulatory limit values for the parameters considered by the hazardous WAC, with the only exception of Pb.

The metals Cd and Cu were detected in the leachates of the untreated APCr-m at concentrations too close to the detection limits, and they were not detected in the majority of the leachates of w-APCr. The elements As, Hg, Ni, Sb and Se, as well as fluorides were not released from APCr-m before washing; consequently, these parameters were not detected in the leachates of w-APCr. The pH values of the leachates of w-APCr slightly increased with respect to the pH of the leachate generated from the untreated residue, however, these pH increments were only around 1% in average; so that, the washing treatment did not have a significant effect on the pH of the APCr leachate.

The efficiency of APCr washing was estimated from the reduction in the leaching rates of the most impacted parameters Ba, Cr, Mo, Pb, Zn, chlorides and sulphates. Figure 5.1 and Figure 5.2 show the results of optimisation of APCr washing, where "decrease %" means percentage of leaching reduction of each metal or salt; thus, this value increases as the efficiency of the treatment increases.



Figure 5.1. Metals leaching reduction achieved by washing at different extraction times and L/S ratios.



Figure 5.2. Leaching reduction of soluble salts achieved by washing at different extraction times and L/S ratios.

From Figure 5.1 it can be seen that in any of the tested washing conditions, Pb and Zn showed the highest leaching reductions, from 61% to 68% Pb and from 51% to 71% Zn, without any tendency regarding to the extraction time, but with percentages slightly higher at the L/S ratio of 2.5. Very close to these efficiencies was the leaching reduction of Ba, which ranged between 38% and 55%, and it was favoured by the increment of the extraction time. The leaching decrease of Cr ranged between 0% to 41% and it was clearly promoted by an extraction time as short as just 5 min at any of the evaluated L/S ratios. The lowest leaching reduction was observed for Mo with efficiencies from 7% to 16% and it was enhanced by the longer extraction times.

Figure 5.2 shows that the decrease in leaching of soluble salts was greater for chlorides than for sulphates. For chlorides, the leaching reduction rates, that ranged from 70% to 89%, were equivalent at the same extraction times and different L/S ratios, and they were favoured by the increase in the extraction time. The leaching decrease of sulphates varied from 1% to 40% and was favoured by the longer extraction times; also, the efficiencies at 30 and 60 minutes of extraction time were equivalent when the L/S ratios were 2.5 and 10.

From the information presented in Figure 5.1 and Figure 5.2, it is clear that if the extraction time is kept constant, the L/S ratio does not have a significant effect on the washing efficiency because for both metals and soluble salts, no general tendency was observed regarding this variable. Therefore, the lowest L/S ratio (2.5) is preferred for APCr washing, giving priority to the use of the minimum amount of water in the process. On the other hand, at a constant L/S ratio, the efficiency of APCr washing increases as the extraction time increases. This tendency is especially clear in the leaching of Ba and Mo (Figure 5.1) and chlorides and sulphates (Figure 5.2). In consequence, 60 minutes, which is a moderate time for treatment by washing was selected as the optimum extraction time of 60 min, enable the use of a low amount of water and the washing of the residue in a time as short as possible without compromising the efficiency of the treatment; thus, the APCr washing under these conditions may be economically feasible if this procedure is replicated at full-scale.

5.1.2 Changes on APCr due to washing treatment

5.1.2.1 Decrease in APCr mass

In addition to the solid material that dissolved into the water during the washing of APCr, a fraction of solids was transferred into the water although they did not dissolve but remained suspended (suspended solids). As a consequence, there was a mass loss of APCr that should be taken into account for the mass balance in further use of w-APCr. Figure 5.3 shows the % decrease in mass of APCr-m due to washing at the conditions evaluated during the optimisation of the treatment and the comparison with values reported by other studies.



Figure 5.3 Mass loss of APCr due to washing and comparison with other studies.

From the results in Figure 5.3, it can be said that the extraction time did not have a major influence on the mass loss of APCr-m during the washing because similar percentages of mass decrease were obtained at the same L/S ratio. Nevertheless, the mass loss of APCr-m was greater as the L/S ratio increased. This tendency was in accordance with the high % of mass decrease reported by Chen, et al. (2012), applying a L/S ratio of 10 l/kg, which can be comparable with the results and conditions of T7, T8 and T9 of this study. The fractions of APCr-m lost by washing at an L/S ratio of 2.5 l/kg were in agreement with the percentage reported by Bogush, et al. (2019) for washing of APCr from the UK at an

L/S ratio equal 10 l/kg. This is another indication that for stabilisation of leaching properties by washing, it is not required to increment the amount of water further than the optimum L/S ratio. The lowest mass loss was achieved at the optimum conditions of washing (T3), with a 20.6% of mass decrease, which indicates that per each kg of APCr treated by washing, around 0.8 kg of w-APCr can be recovered.

5.1.2.2 Total contents of metals

The total contents of the 12 metals regulated by the WAC were analysed for APC-m before and after washing at optimum conditions. Figure 5.4 shows the results for 10 of these metals. Hg and Se were not included in the graph because they were not detected either in APCr-m or in w-APCr.



Figure 5.4 Effect of washing on total contents of metals of APCr.

The concentrations of metals either at trace, minor of major levels, increased in w-APCr. The reason is that the soluble fraction extracted during the washing caused a mass loss, mainly represented by the solubilisation of chlorides and sulphates. As a consequence, the proportion of each metal respect to the solid matrix of APCr increased. Comparing the contents of metals of APCr-m with w-APCr, it can be seen that the greater effect of re-concentration was for Cu, which was a minor element in untreated sample (concentration lower than 1,000 mg/kg) and became a major element in the washed

sample. After washing of APCr-m, 6 out the 10 metals shown in Figure 5.4, increased their concentrations by 15% to 26%; also, considering the similarity between the percentages of increase for the most abundant metals, 20% for Pb and 21% for Zn, it could be concluded that the APCr washing causes an increment of at about 20% in the concentration of heavy metals in the washed material.

5.1.2.3 Mineralogy

The XRD patterns of APCr-m before and after washing at optimum conditions are presented in Figure 5.5.



Figure 5.5 XRD patterns and identification of the main mineral phases in APCr-m before washing (APCr-m) and after washing (w-APCr) under optimum conditions.

Chlorides and sulphates in APCr are considered water-soluble, also, as described in Section 4.7 these substances are usually deposited on the surface of the bigger particles, so that they should be easily removed by washing. Therefore, the changes in the mineralogy of APCr due to washing can be described from the variations of the identification peaks of chlorides and sulphates. The diffractograms shown in Figure 5.5 revealed that calcium hydroxychloride and sylvite were removed from APCr-m by washing because the main peaks for identification of these phases were absent for calcium hydroxychloride and barely detected for sylvite in the diffractogram of w-APCr. The dissolution of these phases in water is theoretically described in Equation 5.1 for calcium hydroxychloride and Equation 5.2 for sylvite. The production of portlandite according to Equation 5.1 could explain the increase in the intensity of the peaks for portlandite in w-APCr. While portlandite remained in the solid material, calcium chloride dissolved into water.

Equation 5.1 $2CaOHCl_{(s)} \rightarrow Ca(OH)_{2(s)} + CaCl_{2(aq)}$

Equation 5.2 $KCl_{(S)} \rightarrow KCl_{(aq)}$

A moderate reduction in the intensity of the peaks for halite in the pattern of w-APCr is observed, indicating a partial removal of this phase. The dissolution of halite in water is given in Equation 5.3. Zincowoodwardite exhibited pronounced peaks in the diffractogram of APCr-m, but these signals were significantly reduced in the pattern for w-APCr, indicating a high effect of removal by the washing treatment for this phase. The two forms of calcium sulphates detected in APCr-m, anhydrite and gypsum, were likely dissolved into water according to Equation 5.4 and Equation 5.5, respectively. The peak for gypsum and the minor peak for anhydrite, which were hardly detected in APCr-m, increased in intensity in w-APCr, probably as a consequence of the removal of soluble salts that allowed to differentiate these signals from the background noise in the XRD patterns.

Equation 5.3 NaCl_(s) \rightarrow NaCl_(aq) Equation 5.4 $CaSO_{4(s)} \rightarrow CaSO_{4(aq)}$

Equation 5.5 CaSO₄•2H₂O_(s) \rightleftharpoons CaSO_{4(aq)}

The main identification peaks of calcite, clinotobermorite, portlandite and quartz remained invariable after the washing of APCr-m, indicating that probably the washing did not have a significant effect on the removal of these mineral phases, or a reconcentration effect caused by the loss of dry matter during the dissolution of the most water-soluble substances.

Figure 5.6 presents the results of mineral composition and total contents of mineral and amorphous material of APCr-m and w-APCr. It should be noted that this quantitative analysis is based on the comparison of the main mineral phases initially detected in APCr-m and their changes in w-APCr. Some other phases were present in the analysed material at lower concentrations.



Figure 5.6 Contents of mineral phases and total contents of crystalline and amorphous material in APCr before (APCr-m) and after(w-APCr) washing. The error bars correspond to +/- 1 Standard Deviation from the mean value.

The major effects of the washing treatment are observed in the complete removal of calcium hydroxychloride and the removal of most of the content of sylvite. Although it is known that part of the halite was removed by washing, the concentration of this phase was statistically equivalent in APCr-m and w-APCr. This can be a re-concentration effect associated with the loss of dry matter during the dissolution of soluble substances, but it is also an indication that the washing treatment at an L/S ratio of 2.5 and extraction time of 60 min was not effective enough for the removal of halite. Considering that the chlorides fraction in APCr mostly comprises calcium hydroxychloride, sylvite and halite, it can be concluded that the remaining content of chlorides in the washed material is mostly halite.

From Figure 5.6, it is clear that the hydrated form of calcium sulphate (gypsum) was mostly removed from APCr-m by washing, while the sulphates that remained in the washed material were due to the presence of anhydrite, which in addition had an effect of re-concentration similar to halite. In both cases, halite and sulphates, it is necessary to further research the removal of these minerals from APCr by washing, evaluating the different factors that may influence their solubility, for example sequential extractions and temperature. An option for the removal of sulphates that can be investigated is the treatment with solutions of sodium carbonate Na₂CO₃, since this salt combines with CaSO₄ to form the more water-soluble sodium sulphate Na₂SO₄ (Dontriros, et al., 2020).

The initial concentrations of portlandite, quartz and zincowoodwardite were low in APCrm and further reduced by washing in w-APCr. While calcite and clinotobermorite were detected at higher concentrations in w-APCr likely because of the re-concentration effect due to the dry mass loss. The total effect of washing in the mineralogy of APCr can be observed in the reduction of the fraction of crystalline material from 40% in the untreated APCr-m to 37% in the w-APCr.

5.1.2.4 Morphology and microstructure

The appearance of APCr-m before and after washing treatment is shown in Figure 5.7. The raw APCr-m was a light grey fine powder with the presence of some black particles of carbon. The washed and dried material, w-APCr, was a light grey, coarse-grained material whose particles grouped in moderately hard lumps. For further use of w-APCr, the material needs to be firstly ground to powder.



Figure 5.7 Appearance of raw and washed APCr. A: raw APCr-m, B: washed under optimised conditions and dried APCr-m (w-APCr).

APCr-m was the combination of 42 APCr samples, whose microstructures were described in Section 4.7 and individually detailed in Appendix IX. As shown in the SEM image in Figure 5.8 Part A, the surface of APCr-m was dominated by spherical particles of different sizes surrounded and covered by irregular agglomerates of fine-grained material, while the different crystal shapes were mostly hidden among the fine-grained material, the bigger spherical particles and the amorphous material. The SEM image shown in Figure 5.8 Part B, shows that there was much less fine-grained material in w-APCr, so that the spherical particles and fragments of some crystalline phases were more noticeable. The fine-grained material missing in w-APCr was mostly chlorides. Also, there were less spherical particles and specifically bigger spheres as the one observed in Figure 5.8 Part A (around 50 μ m) were hardly found in w-APCr. As discussed in Section 4.7.2, these spheres contained elements such Ca, Si, Mg, K and Al, which likely formed part of complex molecules of soluble salts.



Figure 5.8 SEM images of raw and washed APCr. A: raw APCr-m, B: w-APCr.

Figure 5.9 shows the SEM images and their corresponding EDS map spectrums for APCrm and w-APCr. The contents of Cl and K are easily observable in the EDS map of APCrm (Part A), but extremely low on the surface of w-APCr (Part B). This confirms that most of the chlorine and potassium, in the form of chlorides were removed from APCr-m during the washing treatment. In addition, the content of calcium greatly increased in w-APCr, as shown by the corresponding layered image and the EDS spectrum, which shows an increment of about 25 times in the signal intensity for this element, with respect to APCr-m. The concentration of calcium increased in w-APCr due to the removal of soluble substances and because more stable Ca-based phases, mainly calcite, remained in the solid matrix.



Figure 5.9 SEM images and EDS map spectrums of raw and washed APCr. A: APCr, B: w-APCr.

5.1.3 Characterisation of the wastewater produced from APCr washing

During the APCr washing, it is produced a residual liquid (wastewater) enriched mainly with heavy metals and salts. The chemical characterisation of this liquid obtained from the APCr washing under optimum conditions is presented in Table 5.2. For washing APCr at an industrial scale, the wastewater must be handled without causing environmental impact and in way that results economically feasible. This effluent must be treated for a final discharge or for recirculation. Given the characteristics of the wastewater shown in

Table 5.2., especially the contents of salts and TDS, a suitable option to treat this effluent can be evaporation which allows the removal of chlorides and sulphates, diminishing the TDS contents and conductivity enabling the water for subsequent reuse. The treated water may be recycled into the washing treatment of APCr depending on its final quality parameters.

Table 5.2 Characterisation of wastewater produced from APCr washing at L/S ratio 2.5 and extraction time of 60 minutes.

Metals (mg/l)											
As	Ba	Cd	Cr	Cu	Hg	Мо	Ni	Pb	Sb	Se	Zn
<0.158	2.59	0.01	0.21	0.11	<0.116	< 0.152	< 0.033	27.98	<0.086	<0.101	4.44
Soluble material (mg/l)							− nH at 20°C		Conductivity at		
Cl		F-	SO4 ⁻²		TDS		DOC	pir at 20 C		20°C (μS/cm)	
11,140.08		<2.03	1,575.44		23,940		13	12.56		32,150	

5.2 Accelerated carbonation

Accelerated carbonation was assessed in APCr by means of exposure of the material to a high concentration of CO_2 in the presence of moisture. This treatment leads to the absorption of CO_2 and production of stable carbonates in APCr, enabling this residue for subsequent recycling. To better understand the characteristics of the APCr carbonation, it is required to comprehend the mechanism of this reaction. Figure 5.10 illustrates the carbonation mechanism through 7 steps (Maries, 1985) over the three phases (gas-liquid-solid) that converge on the surface of the solid material, according to the following description.

- 1. The CO₂ gas penetrates into the solid matrix of APCr through their pores.
- The gas CO₂ dissolves in the pore water becoming CO_{2(aq)}. It represents the adsorption of CO₂.
- The hydration of CO_{2(aq)} produces H₂CO₃ lowering the pH. This can occur on the surface of the water layer that covers the phases that are prone to carbonation, such as Ca(OH)₂. This stage and the CO₂ adsorption (step 3) are the slowest steps of carbonation (Sun, et al., 2008).

- 4. The ionisation of H₂CO₃ (product of the CO_{2(aq)} hydration) generates the CO₃⁻², which is directly involved in the carbonation reactions.
- 5. When the APCr (of alkaline nature) contact with water that contains CO₃⁻² ions and has a low pH, the solid and liquid phases are not in equilibrium. To achieve the equilibrium, some Ca-based minerals, like Ca(OH)₂ from APCr dissolve into the liquid from the surface of the particles. The rate of this dissolution is faster than the reactions taking place in steps 3, 4 and 5, because APCr has been previously mixed with water, so that Ca⁺² and OH⁻ are already present in the liquid phase (Sun, et al., 2008).
- 6. The nucleation is the early formation of the crystalline form of calcite.
- The calcite (nuclei) precipitates on the surface of the APCr particles, increasing their mass.



Figure 5.10 Mechanism of accelerated carbonation of solid waste residues based on the mechanism of reaction between cementitious materials and CO₂ described by Maries, A (1985).

5.2.1 Carbonation potential

The carbonation potential of APCr was evaluated over a period of 9 hours to determine the extent of the reaction with respect to the reaction time. Figure 5.11 shows that the CO₂ uptake of APCr-m is directly proportional to the exposition time. During the first two hours of exposition, APCr-m increased its original weight by 13.3% due to the capture of CO_2 . This percentage was exactly the maximum CO_2 uptake achieved by Araizi, et al. (2016) in APCr carbonation enhanced by sonication (details of this experiment were provided in Table 2.4). To facilitate the estimation of the CO₂ uptake from 2 to 9 hours, it was assumed that the CO₂ uptake was linearly related to the reaction time. That relationship can be described by the tendency line and the linear equation shown in Figure 5.11. This equation suggests that from the second hour of reaction, the weight gain of APCr-m was no more than 0.49% of its initial weight every hour. Therefore, it can be stated that the exposition of APCr to CO₂ during the accelerated carbonation treatment requires at least 2 hours to achieve a CO_2 uptake of around 13% of its original weight, while longer periods of time will not cause significant increments of CO₂ consumption. In fact, the experiment showed that when carbonating APCr for 9 hours, 80% of the reaction occurs during the first 2 hours. For that reason, 2 hours was selected as the optimum reaction time for accelerated carbonation of APCr. This contact time is as short as the time used by Prigiobbe, et al. (2009) in a thermogravimetric system. Also, 2 hours is lower than the reaction time previously used for APCr carbonation in closed chambers, such as, 2.5 hr, optimised for carbonation of APCr in the UK by Fernández Bertos, et al. (2004c), and 3 hr used by Li, et al. (2007) and Cappai, et al. (2012).



Figure 5.11. Carbonation potential of APCr. 80% of the CO_2 uptake occurs during the first 2 hours of reaction. A linear relationship between time and CO_2 uptake is assumed from 2 to 9 hours to facilitate the estimation of the CO_2 uptake per hour over this period.

The control sample of APCr-m, which was left outside the chamber for 9 hours under ambient conditions of the laboratory did not increase in weight due to CO₂ uptake. APCr may have been carbonated during handling at ambient conditions of temperature and pressure (natural carbonation) or even during the storage (although the samples were stored in airtight containers). It is advisable to assess the CO₂ uptake in fresh samples and compare the results with aged samples.

According to the mechanism of the reaction shown in Figure 5.10, it is known that both the rate and extent of the APCr carbonation are influenced by different factors, whose effects may change as the carbonation proceeds. These factors, identified by Pan, et al. (2012) are listed and commented for this research in Table 5.3.

Table 5.3 Factors that affect the rate and extent of APCr carbonation.

Factor	Comments				
Transportation-controlled steps	It is known that the diffusion of CO ₂ is 10,000				
of the carbonation mechanism,	times lower in water than in the air.				
e.g. diffusion of CO ₂ and Ca ⁺²	Transport of Ca^{+2} in the solid depends on the % of				
to and from reaction sites	moisture.				
Boundary layer effects, e.g.	Saturation in the solid phase due to precipitation				
diffusion across precipitate	of new-formed phases decelerate or stops the CO_2				
coatings on particles	absorption.				
Dissolution of Ca(OH) ₂ on the	Given the mineralogy of APCr-m, the dissolution				
particle surface	of other Ca-based mineral phases, such as				
	CaOHCl, must be considered. Also, phases				
	containing Mg^{+2} and Zn^{+2} can dissolve, and these				
	ions may take part in the carbonation reaction.				
Pore blockage	An excess of water may block the pores of the				
	APCr decreasing the carbonation rate				
Precipitate coating	It increases the mass on the APCr surface. A				
	dynamic process could be advisable.				

5.2.2 Effect and optimisation of moisture

The water addition has an important effect on the carbonation rate of APCr. According to the mechanism described in Figure 5.10, it is needed a certain amount of water absorbed into the pores of the solid material during the carbonation reaction because this water hydrates and solvates the CO_2 and dissolves the Ca^{+2} ions promoting the CaCO₃ formation. Nevertheless, an excess of water obstructs the pores of the solid, making CO_2 diffusion more difficult and slower (Sun, et al., 2008). As a consequence, an optimum water addition must be determined for APCr carbonation.

The effect of the water addition on the APCr carbonation was evaluated over the optimum reaction time of 2 hours. The results of CO_2 uptake by APCr-m determined for 10%, 20%, 30% and 40% of moisture are shown in Figure 5.12.



Figure 5.12 Effect of water addition on APCr carbonation.

Figure 5.12 shows that the rates of carbonation before completing the first hour of reaction were higher in the APCr samples with 10% of moisture. This is because a low content of water facilitates the permeation of CO_2 into the pores of the solid material. In contrast, 40% of moisture caused a certain blocking of the pores, inhibiting the carbonation reaction particularly during the first 45 min. At the reaction time of 1 hour, all samples exhibited similar CO_2 uptakes, from 8.1% to 8.7% of their initial weights, independently of their contents of water. After this time, the carbonation rate decreased for the samples with 10% of moisture because the mobility of the ions involved in the

reaction (Ca⁺² and CO₃⁻²) was more difficult due to the reduced amount of water absorbed in the solid material. It can be seen that from 75 min until 2 hr of reaction, the CO₂ uptakes increased for the samples added with 20% and 30% of water. Therefore, the optimum percentage of moisture for APCr carbonation is between 20% and 30%.

Figure 5.13 shows the CO₂ uptakes of different samples of APCr-m added with the percentages of moisture under evaluation and carbonated for 2 hr. Each CO₂ uptake is shown with its corresponding error interval (+/- 1 SD for 3 replicates). It is clear that, when the moisture was 20%, the APCr samples achieved the highest CO₂ uptakes with an average of 12.1%. The error interval of this value does not overlap with any of the intervals of CO₂ consumed by samples with other additions of water. That is, the CO₂ uptake of APCr added with 20% of water, was higher and statistically different from the CO₂ uptake of APCr added with 10%, 30% or 40% of water. Therefore, 20% was determined as the optimum percentage of moisture for carbonation of APCr.



Figure 5.13 Optimisation of moisture on APCr carbonation. The error bars correspond to +/- 1 Standard Deviation from the mean value.

It should be noted that in this last experiment (Figure 5.13), the percentages of CO_2 uptake were higher than those presented in Figure 5.12 at 2 hr of carbonation. The reason is that for the generation of the data in Figure 5.12, the carbonation chamber was opened every 15 min to remove the samples, causing a decrease of the CO_2 saturation; while all the samples tested for the optimisation of moisture in Figure 5.13 were removed from the chamber just when completing the reaction time of 2 hours.

5.2.3 Carbonation kinetics

The steps of the carbonation mechanism shown in Figure 5.10 are consecutive, so that if the speed of any of them changes, it affects the kinetics of the reaction, but considering the complex structure of APCr, the reaction is probably influenced by more than one step (Sun, et al., 2008). Figure 5.14 shows the progression of APCr-m carbonation at optimum % of moisture (20%) over a period of 3 hr.



Figure 5.14. Kinetics of APCr carbonation. The error bars of each CO_2 uptake measurement correspond to +/- 1 SD for 2 replicates.

It is observed that the APCr carbonation rate was very fast during the first 15 min of the reaction, with almost 40% of the full carbonation potential, which represented an increment of 0.3% of CO₂ consumption per min. This is because the optimum percentage of moisture provided the right amount of pore water to hydrate and solvate the CO₂ and to dissolve the Ca⁺² ions, without blocking the pores of the solid. Then, the reaction rate decreased to about 0.06% of weight increment per minute, achieving a CO₂ uptake of 10.5% at the optimum reaction time of 2 hours. The velocity of the carbonation decreases because as the reaction proceeds, the CaCO₃ crystals, formed as a result of the carbonation, precipitate on the solid surface and obstruct the pores, reducing the speed of CO₂ diffusion into the pores. This effect is illustrated in Figure 5.15 in accordance with the description of the carbonation as a reaction controlled by the diffusion of CO₂ (Bin Shafique, et al., 1998): the gas diffuses into the solid structure of APCr producing a

growing ring of carbonated material (CaCO₃) that surrounds an inner zone of unreacted (uncarbonated) material.



Figure 5.15 Carbonation of APCr controlled by the diffusion of CO₂.

After 2 hours, the CO₂ uptake did not show significant increments, instead, it exhibited fluctuations around 10%, which indicates that beyond the optimum reaction time, the APCr carbonation tends to stop, although not all the species susceptible to carbonation have completed the reaction. The fluctuations and the increase in the uncertainty of the measurements (error intervals) after 2 hours of carbonation may be attributed to the physical changes of the APCr particles, including enlargement of particle size, reduction of porosity and agglomeration of carbonation products.

5.2.4 Changes on APCr due to carbonation

5.2.4.1 Total contents of metals

It is important to note that the contents in the mass of metals do not change due to the carbonation treatment, but the mass ratio (concentration) of each metal declines after carbonation because the total mass of the treated material increases as a result of the CO_2 capture. Figure 5.16 presents the total concentrations of the metals regulated by the WAC in the untreated APCr-m and in c-APCr, as well as the percentages of concentration reduction for each metal. These results were obtained from the carbonation tests carried out at optimum conditions (20% moisture, 2 hr reaction), of which it was confirmed that the CO_2 capture was around 12% of the initial mass of untreated APCr-m. The elements Hg and Se are not reported because they were not detected in APCr-m and c-APCr. The reduction in total concentrations of the metals by carbonation of APCr-m, ranged between 47% to 69%. The total concentrations of the most abundant metals in APCr-m, Zn and Pb, were reduced by 61% and 62%, respectively; followed by Ba and Cu, whose concentrations declined by 47%.



Figure 5.16 Effect of carbonation on the total metal's contents of APCr.

5.2.4.2 Leaching behaviour

The results of leaching tests for APCr-m and c-APCr are shown in Table 5.4.

Table 5.4 Leaching results of APCr-m before and after carbonation at optimum conditions (20% moisture, 2 hr reaction).

BS 12457-2, LS = 10 (mg/kg)							
	APCr-m	c-APCr	Limit value for hazardous WAC				
As	<1.582	<1.582	25				
Ba	12.5148	7.9	300				
Cd	0.0932	0.75	5				
Cr	3.2208	1.3	70				
Cu	0.727	0.071	100				
Hg	<1.158	<1.158	2				
Mo	2.1436	2.4	30				
Ni	< 0.325	< 0.325	40				
Pb	193.025	0.072	50				
Sb	< 0.858	< 0.858	5				
Se	<1.008	<1.008	7				
Zn	42.3476	0.25	200				
Chlorides	114,102.9	103,551.3	25,000				
Fluorides	<20.3	31.1	500				
Sulphates	15,230.3	10,127.2	50,000				
TDS	219,600	215,600	100,000				
DOC	<100	117	1,000				
BS 12457-2, LS/10							
pH at 20°C	12.51	8.37	-				
Conductivity at 20°C (µS/cm)	32,680	26,470	-				

It can be seen that the carbonation of APCr-m reduced the leaching of Ba, Cr, Cu, Pb, Zn, chlorides and sulphates from c-APCr. The stabilisation of these metals and soluble salts favoured the decrease in the TDS concentration and the conductivity of the leachate of c-APCr. The leaching reduction of metals, particularly the most abundant metals, Pb and Zn, was in agreement with other studies on APCr carbonation (Ecke, 2003a; Fernández Bertos, et al., 2004c; Baciocchi, et al., 2009a; Baciocchi, et al., 2009b; Costa, 2009; Li,

et al., 2007; Gunning, et al., 2010; Cappai, et al., 2012). In APCr-m, the leaching rates of Pb, chlorides and TDS did not comply with the regulatory values for hazardous waste acceptance at landfills. After carbonation, the leaching of Pb decreased to a value just detectable, meeting the required limit of 50 mg/kg. The leaching rates of chlorides and TDS from c-APCr were far above their guidance limits of 25,000 and 100,000 mg/kg, respectively.

On the other hand, the release of Cd and Mo incremented due to carbonation. Although the leaching rates of both elements from c-APCr were below the limit values for hazardous waste acceptance, the mobilisation effect on these metals due to APCr carbonation must be taken into account. The leaching of Cd from c-APCr was about 7 times higher than from APCr-m. This increment was similar to the value reported by Ecke, (2003a), who found that the leaching of Cd from APCr increased around 10 times after carbonation. Regarding Mo, it should be noted that the leaching rate of this metal in APCr-m was low (2.2 mg/kg) and its percentage of increase in c-APCr was also low (12%). In addition, the increment in the leaching of Mo is against the effect of demobilisation of Mo due to carbonation reported by Cappai, et al. (2012). In the absence of more data available for comparison, there is not enough evidence to state that the APCr carbonation increases the leaching of Mo and the change in the leaching rate of this metal should be further investigated.

Fluorides and the organic fraction DOC were not present in the leachate from the untreated APCr-m but were detected in the leachate from c-APCr. This could be a mobilisation effect due to the carbonation treatment, similar to the leaching increase of Cd. Nevertheless, it can be attributed to the statistical error caused by analytical variability, since the leachate concentrations for both parameters, fluorides and COD, were close to their limits of detection, 20.3 and 100 mg/kg, respectively. The elements As, Hg, Ni, Sb and Se were not released from APCr-m before carbonation; consistently, these parameters were not detected in the leachate of c-APCr.

The carbonation of APCr had a significant effect on the pH of the leachate. The pH dropped from a highly alkaline value (12.51) before the carbonation of APCr-m to a moderate alkaline value (8.37) in the carbonated residue. Such a reduction in pH by carbonation of APCr has been previously reported by other studies (Ecke, 2003a; Fernández Bertos, et al., 2004c). This reduction in the pH is important because the pH

upon carbonation has an influence on the mobility of metals in APCr (Fernández Bertos, et al., 2004c). The immobilisation of metals in APCr treated by carbonation has been attributed to the changes in the pH of the material occurring as the CO₂ capture proceeds (Astrup, et al., 2006).

The efficiency of APCr carbonation was evaluated according to the metals and salts for which the treatment reduced the leaching rates. Figure 5.17 shows the percentages of leaching reduction for each parameter after the carbonation of APCr-m. The decrease percentage in the graph increases as the efficiency of the treatment increases.



Figure 5.17 Effect of APCr carbonation on the leaching reduction of metals and soluble salts.

At optimum conditions of APCr carbonation, the efficiency of the treatment for reducing the leaching of metals ranged from 37% to 100%. The greatest mobility reductions were achieved for the metals of major concern, Pb and Zn, whose leaching rates were diminished by 100% and 99%, respectively. The leaching reductions for the other metals followed the order Cu > Cr > Ba and were inversely proportional to their leaching rates in the untreated APCr-m reported in Table 5.4. Regarding the soluble salts, the decrease of leaching was much lower compared with the reductions achieved for metals. Particularly the reduction of leaching of chlorides by carbonation was not satisfactory because these salts were the most concerning leachable substance in the untreated material. Despite the immobilisation effect of carbonation on the metals and salts shown

in Figure 5.17, the reduction in the leachate concentration of TDS for c-APCr was only 2%. It is concluded that the carbonation of APCr was significantly efficient for the immobilisation of the metals Pb, Zn, Cu, Cr and Ba, while it was not efficient for the immobilisation of chlorides and sulphates. For further use of c-APCr, it should be considered a complementary treatment or strategy to deal with the leachable salts that remain in the solid matrix of the treated residue.

5.2.4.3 Mineralogy

XRD analysis of APCr-m, before and after carbonation were performed to assess the changes in the mineralogy of the material. Figure 5.18 presents the XRD patterns for APCr-m and c-APCr.



Figure 5.18 XRD patterns and identification of the main mineral phases in APCr-m before carbonation (APCr-m) and after carbonation (c-APCr) under optimum conditions.

The most significant changes that are observed in the XRD patterns include the intensity rise of the peaks corresponding to CaCO₃ and the disappearance of Ca(OH)₂ and CaOHCl in c-APCr. This indicates that the conversion into CaCO₃ took place by full carbonation of Ca(OH)₂ and CaOHCl according to Equation 5.6 and Equation 5.7, respectively. These equations represent the overall carbonation reactions, which follow the mechanism shown in Figure 5.7.Equation 5.6

Equation 5.6 $Ca(OH)_{2 (aq)} + CO_{2(aq)} \rightarrow CaCO_{3(s)} + H_2O_{(l)} \quad \Delta H = -113 \text{ kJ/mol } CO_2$

Equation 5.7 $2CaOHCl_{(aq)} + CO_{2(aq)} \rightarrow CaCl_{2(aq)} + CaCO_{3(s)} + H_2O_{(l)}$

Besides CaCO₃, the carbonation of CaOHCl produces calcium chloride CaCl₂, although peaks corresponding to this phase were not detected in the diffractogram of c-APCr. Sun, et al. (2008) suggested that probably, the CaCl₂ produced by carbonation reacted with the KCl from the uncarbonated sample to form chlorocalcite KCaCl₃, according to Equation 5.8.

Equation 5.8 $CaCl_{2(aq)} + KCl_{(aq)} \rightarrow KCaCl_{3(s)}$

It can be also observed in Figure 5.18 the disappearance of the peaks corresponding to clinotobermorite, magnesium oxide hydroxide and zincowoodwardite $Al_{0.375}H_6O_{2.686}S_{0.188}Zn_{0.625}$ in the diffractogram of c-APCr, indicating a full reaction of these phases with CO₂, which increases the carbonation efficiency. The carbonation of clinotobermorite may follow the carbonation reaction of a calcium silicate hydrate (C-S-H) described by Bin Shafique, et al. (1998) as per Equation 5.9. Therefore, the balanced equation for the carbonation of clinotobermorite is given by Equation 5.10.

Equation 5.9 C-S-H_(aq) + CO_{2(aq)} \rightarrow CaCO_{3(s)} + SiO₂ • nH₂O_(s) + H₂O_(l)

Equation 5.10 (5CaO • $6SiO_2 • 4H_2O)_{(aq)} + 5CO_{2(aq)} \rightarrow 5CaCO_{3(s)} + 6(SiO_2 • 0.5H_2O)_{(s)} + H_2O_{(l)}$ The carbonation of magnesium oxide hydroxide can be explained by the dissolution of the mineral according to Equation 5.11 and the subsequent reaction of magnesium oxide MgO with CO_2 described in Equation 5.12. The final product magnesium carbonate MgCO₃ stabilises the heavy metal Mg. Since the amount of magnesium oxide hydroxide in the untreated APCr-m was low, the magnesium carbonate produced through carbonation should be at a trace amount, and for that reason, peaks for this product were not observed in the XRD pattern of c-APCr.

Equation 5.11 $Mg_3O_2(OH)_{2(aq)} \rightarrow 3MgO_{(aq)} + H_2O_{(l)}$

Equation 5.12 $MgO_{(aq)} + CO_{2(aq)} \rightarrow MgCO_{3(s)} \Delta H = -118 \text{ kJ/mol } CO_2$

The carbonation reaction of zincowoodwardite is difficult to propose given the complex formulation of this phase ($Al_{0.375}H_6O_{2.686}S_{0.188}Zn_{0.625}$), of which it is needed further research on its chemistry. Part of the stabilisation of Zn described in 5.2.4.2 can be related to the carbonation of this phase.

It is clear that calcite was the major product of the APCr-m carbonation, but additionally, cerussite PbCO₃ was detected in c-APCr. This can be attributed to the carbonation of Pb⁺² produced by the dissolution of anglesite PbSO₄ and litharge PbO, according to Equation 5.13. Although the peaks of anglesite and litharge were not evidenced in the diffractogram of APCr-m, it is known that these phases were present in the sample at trace concentrations, because they had been detected in some of the individual APCr samples that constituted APCr-m. Considering that Pb was the metal with the highest leaching rate in APCr-m (Section 5.2.4.2) and was completely immobilised after the carbonation of the residue, the leaching reduction of this element is related not only to the pH variation of the treated material due to carbonation, but also is determined by the formation of a stable phase. This result is consistent with the stabilisation of soluble Pb in APCr achieved by Jiang, et al. (2009) through the formation of stable phases including PbCO₃ upon carbonation of the residue.

Equation 5.13 $Pb^{+2}_{(aq)} + CO_3^{-2}_{(aq)} \rightarrow PbCO_{3(s)}$ Figure 5.19 shows the mineral composition and the total contents of mineral and amorphous material of APCr-m and c-APCr. It must be noted that the CO₂ uptake increased the total mass of the untreated APCr-m by 12% of its initial mass. These results are aimed to compare the concentrations of the major phases initially detected in APCr-m and their variations due to the carbonation of the residue. Some other phases were present in APCr-m and in c-APCr at minor or trace concentrations and are not part of this quantitative analysis.



Figure 5.19 Mineral contents of APCr before and after carbonation. The error bars correspond to +/- 1 Standard Deviation from the mean value.

The carbonation treatment significantly changed the mineralogy of APCr-m. Several phases that were present in APCr-m before the treatment, were not detected in the carbonated material. Through the CO₂ capture, the Ca-based mineral phases: calcium

hydroxychloride, portlandite and clinotobermorite were completely converted into stable phases, mainly calcite, which increased in concentration from 5.3 wt% in APCr-m to 14.0 wt% in c-APCr. The minerals magnesium oxide hydroxide and zincowoodwardite were also absent in c-APCr. Their reaction products were not detected in the XRD pattern of c-APCr, probably because they were produced at concentrations below the detection limits. It is also possible that some of those products formed part of the amorphous phase of the c-APCr. Cerussite, which was not present in the untreated APCr-m, was detected in c-APCr, which confirms the stabilisation of Pb as per Equation 5.13. The increment in the concentration of quartz in c-APCr was probably due to the detection of the second product of the carbonation of clinotobermorite shown in Equation 5.10 (SiO₂ • 0.5H₂O).

The concentration of anhydrite decreased by more than 50% and gypsum was not detected in c-APCr. The participation of these phases in the carbonation is unlikely as the low amount of water required in the process would not favour their dissolution to release Ca^{+2} ions. Thus, the reduction in the concentration of anhydrite and gypsum in c-APCr is more likely due to the mass increment of the residue upon carbonation. The concentration of halite was slightly reduced in c-APCr, which is due to the increase in mass of the residue upon carbonation. The concentration of sylvite had a further reduction (about 45%) after the carbonation treatment. This can be explained by the possible reaction of sylvite with the carbonation product of calcium hydroxychloride as per Equation 5.8.

The overall effect of carbonation in the mineralogy of APCr was the reduction of the crystalline fraction from 40% in the untreated APCr-m to 32% in c-APCr due to the conversion of some phases into more stable forms mainly calcite.

5.2.4.4 Morphology and microstructure

Figure 5.20 shows the appearance of the untreated APCr-m and the c-APCr. The raw APCr-m was a light grey, fine powder with the presence of some black pieces of carbon; whereas c-APCr was a darker material, coarser due to agglomeration and with the presence of small and slightly hard lumps.



Figure 5.20 Morphology of raw APCr-m before and after carbonation treatment. A: untreated APCr-m, B: carbonated under optimum conditions and dried APCr-m (c-APCr).

SEM images of APCr-m and c-APCr are shown in Figure 5.21. It is clear that the untreated APCr-m was mostly fine-grained material surrounding the bigger particles, while c-APCr had a larger grain size with a reduced pore space due to the agglomeration effect of the carbonation treatment. Consequently, the carbonated sample exhibited a more regular and rounded-like morphology, which is typical of calcium carbonate agglomerated particles. The agglomeration effect is consistent with the observations described by other studies (Baciocchi, et al., 2009a; Fernández Bertos, et al., 2004c; Jiang, et al., 2009).



Figure 5.21 SEM images of raw and carbonated APCr. A: raw APCr-m, B: c-APCr

Figure 5.22 shows the SEM image and EDS map of a coarse grain formed by carbonation in c-APCr. The surface area is mostly composed of Ca, which is due to the precipitation of CaCO₃, and small amounts of Pb, likely due to the precipitation of cerussite (PbCO₃). In the periphery, it can be seen the presence of chlorine that could be part of calcium hydroxychloride particles which were not carbonated due to the blocking of the pores of the solid.



Figure 5.22 SEM image and EDS spectrum of c-APCr.

6 Chapter 6 Production and characterisation of LWA manufactured from APCr

The aim of this chapter was to investigate the suitability of producing LWA from clay and APCr. Two different routes of LWA manufacturing were tested, hot bonding and cold bonding. The products were characterised for structure, technological properties, mineralogy and leaching behaviour. The effects of different factors typical of each manufacturing route on the LWA characteristics were assessed.

6.1 Hot-bonded LWA

For manufacturing of LWA throughout hot bonding, clay and APCr were pelletised and thermally treated over a specific firing temperature range, as described in Chapter 3. The effects of the APCr incorporation and the firing temperature were evaluated on the characteristics of the final products.

6.1.1 Structural characteristics of hot-bonded LWA

In this section, the effects of the APCr incorporation and the firing temperature were evaluated on the morphology, external appearance and internal structure of hot-bonded LWA. The structural characteristics of the LWA containing APCr were compared with the LWA made exclusively of clay.

6.1.1.1 Morphology and external appearance

The external appearance of the different formulations of LWA produced from clay and incorporating increasing amounts of untreated APCr-m or w-APCr are presented in Figure 6.1.

	% ADC:	Firing temperatures °C										
	70 AFCI	1,100	1,120	1,140	1,160	1,180	1,200	1,220	1,230			
LWA from clay	0				0							
LWA from untreated APCr	5											
	10	CC										
	15											
	20											
	25						0 je					
LWA from w-APCr	5											
	10						_	\bigcirc				
	15					6		11 mm				
	20						_	Approx. diameter				
	25											
	30											

Figure 6.1 External and internal appearance of hot-bonded LWA made from clay and APCr.

From Figure 6.1 it can be seen that the external surfaces of the LWA without incorporation of APCr and fired from 1,100 to 1,140°C were smooth with minimal defects. However, these formulations exhibited cracking from 1,160°C and more irregular and porous outer surfaces from 1,180°C. Excessive expansion (density $< 1g/cm^3$) of the particles without addition of APCr was observed at the 2 highest firing temperatures (1,220 and 1,230 °C). With the first addition of APCr-m (5%), the LWA did not show cracking in their outer surfaces, although they partially melted from 1,220°C. Therefore, the subsequent tests were done only up to the firing temperature of 1,200°C.

Considering the LWA made of only clay as the reference products, it can be observed that incorporating increasing amounts of untreated APCr-m led to the loss of the even texture of the LWA surface. The irregularities were likely caused by the release of gas from the APCr during the firing process, since these residues have been found to lose weight between 1,100 to 1,170°C (Quina, et al., 2014b).

On the other hand, the addition of untreated APCr to the LWA formulations led to a detrimental effect on the resistance to the firing temperature. Incorporating 10 and 15 % of APCr-m caused the LWA melted at 1,200°C, and increasing the addition of the residue to 20 and 25%, caused the melting of the pellets from 1,180°C. By comparing the incorporation of untreated and washed APCr into the LWA, it is clear that adding w-APCr had a positive effect on the smoothness of the aggregate's surfaces. However, a rough surface texture might increase the adhesion between the binding material and the LWA (Adhikary, et al., 2022). The insertion of w-APCr also favoured the resistance of the pellets to the firing temperature. This enabled the production of LWA containing up to 30% of w-APCr at firing temperatures up to 1,180°C with no risk of melting.

6.1.1.2 Internal structure and bloating capacity

Figure 6.1 in Section 6.1.1.1 shows the pictures of fracture surface of LWA revealing their internal structure. The LWA made of 100% clay and fired from 1,100 to 1,160°C consisted of a dense glassy outer shell encapsulating a pore system (core). The thickness of the dense outer shell increased, and the diameter of the core decreased when 5% of untreated APCr was incorporated into the LWA formulation. With further additions of untreated APCr, dense internal structures without differentiated external layers were observed. These structures were denser at firing temperatures of 1,100 and 1,120°C but
developed bigger pores from 1,140°C. Having a pore system in the core of the LWA is important because this type of structure is quite related to their physical properties (density, water absorption and crushing strength); therefore, the formulations of LWA with w-APCr were only fired from 1,140 to 1,180°C. At the firing temperatures 1,140, 1,160 and 1,180°C, the LWA incorporating w-APCr exhibited internal pore structures with no well-differentiated external shells.

Figure 6.2 shows the internal structure, observed by digital microscopy, of a LWA made of clay and fired at 1,160°C (Part A) and the LWA containing different additions of w-APCr fired at 1,180°C (Part B). The LWA made exclusively of clay exhibited a pore structure with unconnected pores increasing in size from the inner part of the core to the zone close to the surface and a thick, non-porous outer shell. The incorporation of w-APCr into the LWA significantly changed the appearance of their internal structure. Adding 5% or 10% of w-APCr originated LWA with structures mostly uniform with just a few noticeable pores in the areas adjacent to the surface. Additions from 15% to 30% of w-APCr were effective to increase the number and the size of the pores in the internal pore system of the LWA. The pores had different shapes and were unconnected and randomly distributed throughout the cross-sections of the particles. Besides, these formulations exhibited internal structures with a glossy appearance, which evidenced that vitrification took place not only on the surface but also across the internal core of the LWA.





Figure 6.2 Digital microscopy images of internal structure of LWA. A: 100% clay, fired at 1,160°C, B: 5% to 30% w-APCr, fired at 1,180°C.

The internal pore structure of LWA is developed by the heating of the raw materials to the point of early fusion, in which the gases expand within the pyroplastic mass producing an expansion that is retained upon cooling (ESCSI, 2007). This phenomenon is commonly known as bloating. As described in Section 2.5.1.1, the bloating requires some specific conditions. Firstly, at high temperatures, the material must produce a glassy liquid phase with appropriate viscosity that entraps the released gases. Secondly, the material should contain substances that release gases at the temperature at which the glassy phase is formed. During the firing of LWA, the release of gases is mainly caused by evaporation of the remaining moisture, oxidation of organic matter at high temperature (LOI) and decarbonisation of CaCO₃ (González-Corrochano, et al., 2009). The gases responsible for bloating in hot-bonded LWA production from clay and APCr include CO₂, CO, H₂O, O₂, SO₂, HF and HCl, among others (Quina, et al., 2014b). They can be generated from the clay as well as from the w-APCr. Some reactions that may be involved in the release of gases during the LWA sintering are proposed in Table 6.1.

Origin	Reaction	Temperature °C		
	Equation 6.1	IOI		
Organic matter mainly from	$C + O_{2(g)} \rightarrow CO_{2(g)} + H_2O_{(g)}$	temperature		
clay	Equation 6.2			
	$C + O_{2(g)} \rightarrow CO_{2(g)} + CO_{(g)}$	550, 800		
Calcite from clay and w-APCr	Equation 6.3	700 - 770 *		
Calcule from elay and w-Art Cr	$CaCO_{3(s)} \rightarrow CaO_{(s)} + CO_{2(g)}$	700 - 770		
Portlandite from clay and w-	Equation 6.4	400 440 *		
APCr	$Ca(OH)_{2(s)} \rightarrow CaO_{(s)} + H_2O_{(g)}$	400 - 440		
Calcium hydroxychloride from	Equation 6.5	465 - 600 *		
w-APCr	$CaOHCl_{(s)} \rightarrow CaO_{(s)} + H_2O_{(g)}$	-000 - 000		

Table 6.1 Chemical reactions probably taking place during the bloating of LWA produced from clay and w-APCr.

* Temperature ranges of thermal decomposition determined in APCr (Bodénan & Deniard, 2003).

Figure 6.3 shows the bloating index (BI) expressed as the volume change of the pellets due to the firing at different temperatures for LWA incorporating untreated APCr (Part A) and w-APCr (Part B). The calculations and statistics of the results are presented in Appendix X.



Figure 6.3. Bloating index of A: LWA incorporating untreated APCr, B: LWA incorporating w-APCr. The error bars correspond to +/- 1 Standard Deviation from the mean value.

In Figure 6.3 A, it can be seen that the bloating of the LWA made exclusively of clay was not significant at the lowest tested firing temperatures (maximum BI was 1.6%), but it was more noticeable from 1,160°C with a BI of almost 5% and increased as the firing temperature increased achieving a maximum BI of 40% when the pellets were fired at 1,220°C. The error intervals of BI for LWA made of clay became wider as this percentage incremented, because the expansion due to the firing deformed to a certain extent the original rounded shape of the pellets, making the measurement of the particle's diameter less precise. The incorporation of 5% of APCr-m into the LWA did not produce expansion at firing temperatures between 1,120 and 1,160°C, only slightly expanded the pellets at 1,100 and 1,180°C (BI no greater than 1.8%) and caused a maximum expansion at 1,200°C with a BI of 14%. By comparing the expansion achieved by the LWA containing only clay with those containing 5% of APCr-m, it is observed that this incorporation of untreated APCr, reduced the expansion of the pellets by 88% and 57% at the firing temperatures 1,180°C and 1,200°C, respectively. Greater additions of APCr-m did not cause or just caused negligible expansion of the LWA when the pellets were fired at 1,100°C, while these additions did not cause any signal of expansion at 1,120°C or over.

Figure 6.3 B shows that the incorporation of 5% to 30% of w-APCr into the LWA did not produce any increment in the volume of the pellets within the firing temperature range 1,140 - 1,180°C. This behaviour as well as the results previously described for the additions of APCr-m were in accordance with the lack of expansion properties of untreated and washed APCr determined by Quina, et. al (2014b) according to the oxides composition of these residues represented in a Riley diagram (see Figure 2.12).

Given the negative percentages of BI shown in graphs A and B in Figure 6.3, it is concluded that the incorporation of APCr, either untreated or washed, into the LWA not only inhibited the clay expansion but also caused a reduction in the volume of the pellets after the firing treatment. Such a reduction was up to 9% of the original volume of the green pellets. Despite the incorporation of w-APCr did not cause an increment in the final volume of the LWA, it favoured the development of the internal pore structure, as previously discussed.

6.1.2 Technological properties of hot-bonded LWA

In this section, the effects of APCr incorporation and firing temperature were evaluated on the technological properties of hot-bonded LWA. Table 6.2 presents the list of the LWA that were manufactured by hot bonding for testing of the technological properties and the codification regarding to their contents of APCr. The results were compared with the technological properties of Lytag® determined in the laboratory and reported in Table 6.3.

	% APCr	Code
LWA from clay	0	100%Clay
	5	5%APCr-m
	10	10%APCr-m
LWA from APCr-m	15	15%APCr-m
	20	20%APCr-m
	25	25%APCr-m
	5	5%w-APCr
	10	10%w-APCr
I WA from w APCr	15	15%w-APCr
LWA IIOIII W-AI CI	20	20%w-APCr
	25	25%w-APCr
	30	30%w-APCr

Table 6.2 Codification of LWA produced by hot bonding.

Table 6.3 Technological properties of Lytag® measured in laboratory.

Property	Units	Value
Oven-dried particle density	g/cm ³	1.47 ± 0.02^{-a}
oven-uneu partiele density	g/cm	1.46 ± 0.01^{b}
Saturated and surface-dried particle density	a/cm ³	1.61 ± 0.02 ^a
Saturated and surface-dired particle density	g/cm	1.63 ± 0.02^{b}
Apparent particle density	a/am ³	1.72 ± 0.02 ^a
Apparent particle density	g/cm	1.76 ± 0.02^{b}
Water absorption (at 24 hr)	0/2	9.63 ± 0.81^{a}
water absorption (at 24 m)	70	$11.24\pm0.21~^{b}$
Loose bulk density	g/cm ³	0.84 ± 0.01
Voids	%	42.4 ± 0.7
Crushing strength	MPa	3.23 ± 1.03

^a Method of the wired basket (BS EN 1097-6, 2022)

^b Method of the pycnometer (BS EN 1097-6, 2022)

 \pm values indicate one standard deviation of 3 replicates

6.1.2.1 Particle density

The effect of the firing temperature on the particle density was evaluated for the LWA produced from APCr-m or w-APCr. To do that, the variation of the particle density determined by the wire basket method (BS EN 1097-6, 2022) in hot-bonded LWA was assessed in regards to the firing temperature. The calculations and statistical analysis of the results are reported in Appendix XI.

6.1.2.1.1 Particle density of LWA containing APCr-m

Figure 6.4 shows the variation of the oven-dried (A), saturated and surface-dried (B) and apparent (C) particle densities of the LWA made from APCr-m and fired from 1,100 to 1,200 °C. The desirable oven-dried particle density for LWA should be maximum 2 g/cm³. Figure 6.4 A shows that the LWA 100%Clay met this requirement over the firing temperature range 1,100 – 1,200°C with a maximum value of 1.60 g/cm³ at 1,100°C and a minimum of 1.02 g/cm³ at 1,180°C. The graph shows that the incorporation of 5% of

APCr-m slightly increased the oven-dried particle density. With greater additions of APCr-m, the oven-dried particle density decreased at the lower firing temperatures (1,100 and 1,120°C), but densification was observed when the firing temperature varied from 1,140 to 1,180°C. The effect of the firing temperature on the saturated and surface-dried particle density of LWA containing APCr-m (Figure 6.4 C) followed the same tendencies described for the oven-dried particle density with just small increments in the density values. The particle densities (oven-dried, apparent and saturated/surface-dried) of 100%Clay fired over the range 1,100 to 1,160°C were slightly higher or statistically equivalent to the values determined for Lytag® (see Table 6.3), while greater firing temperatures reduced the densities to values lower than those determined for this reference product. The addition of APCr-m generated LWA with densities higher than the densities of Lytag® within the firing temperature range of 1,100 to 1,180° (except for the apparent particle density of LWA containing 5% of APCr-m and fired at 1,180°C). The apparent particle density of LWA made of only clay in the firing temperature range between 1,100 and 1,200°C, as shown in Figure 6.4 C, ranged from 1.13 g/cm³ at 1,180°C to 1.92 g/cm³ at 1,120°C. When APCr-m was added to the formulations of the LWA, the apparent particle density increased, particularly with additions of 5, 10 and 15% of APCr up to values around 2.2 g/cm³ at firing temperatures between 1,100 to 1,180°C.



Figure 6.4 Effect of the firing temperature on the particle density (determined by the method of the wire basket (BS EN 1097-6, 2022)) of hot-bonded incorporating APCr-m. A: Oven-dried particle density, B: Saturated and surface-dried particle density, C: Apparent particle density. The error bars correspond to +/- 1 Standard Deviation from the mean value.

6.1.2.1.2 Particle density of LWA containing w-APCr

Figure 6.5 shows the variation of the oven-dried (A), saturated and surface-dried (B) and apparent (C) particle densities of the LWA incorporating w-APCr and fired at 1,140, 1,160 and 1,180 °C. When different amounts of w-APCr were added to the formulation of LWA, the oven-dried particle density remained almost constant at a firing temperature of 1,140°C, but increased when the temperature was 1,160 or 1,180°C. The incorporation of 5 to 30% of w-APCr into LWA fired at 1,180°C increased the oven-dried particle density by 70% to 95% with respect to the LWA 100%Clay. In the firing temperature range from 1,140 to 1,180°C, the oven-dried particle density values for LWA containing 5-30 % of w-APCr ranged between 1.62 to 1.99 g/cm³, meeting the requirement of not exceeding 2 g/cm³ to be considered LWA. The saturated and surface-dried particle density of LWA incorporating 5 - 30% of w-APCr varied between 1.82 and 2.03 g/cm³ over the firing temperature range of 1,140 to 1,180°C. The incorporation of w-APCr incremented the apparent particle density of the LWA up to values between 1.9 to 2.2 g/cm³ at firing temperatures from 1,140 to 1,180°C. In this range, the firing temperature did not significantly affect the apparent particle density of the LWA containing w-APCr. However, it is noticeable that from the formulations with the two highest additions of w-APCr, 25% at any of the three assessed temperatures and 30% at 1,180°C had apparent particle densities lower than 2 g/cm³. All the formulations of LWA incorporating w-APCr exhibited particle densities (oven-dried, saturated and surface-dried and apparent) higher than the values determined for Lytag® within the firing temperature range of 1,140 to 1,180°C. These results indicate that the LWA incorporating untreated or w-APCr were heavier than the LWA that contains fly ashes (Lytag®). It must be noted that from the formulations containing w-APCr, the greater additions, 25% and 30%, produced LWA with saturated and surface-dried and apparent particle densities more comparable to the corresponding reference values of Lytag®.



Figure 6.5 Effect of the firing temperature on the particle density (determined by the method of the wire basket (BS EN 1097-6, 2022))of hot-bonded LWA made from w-APCr. A: Oven-dried particle density, B: Saturated and surface-dried particle density, C: Apparent particle density. The error bars correspond to +/- 1 Standard Deviation from the mean value.

6.1.2.2 Particle density of LWA fired at 1,180°C

Two batches of LWA, 25%w-APCr and 30%w-APCr fired at 1,180°C, were produced and analysed for particle density according to the method of the pycnometer (BS EN 1097-6, 2022). These formulations were selected because they will be subsequently used for concrete casting (Chapter 7). The calculations and statistical analysis are shown in Appendix XII. The results were compared with Lytag® and are summarised in Figure 6.6.



Figure 6.6 Particle density (determined by the method of the pycnometer (BS EN 1097-6, 2022)) of LWA fired at 1,180°C and comparison with Lytag®. The error bars correspond to +/- 1 Standard Deviation from the mean value.

The particle densities (oven-dried, saturated & surface dried and apparent) of the LWA made exclusively of clay were only slightly higher than the values determined for Lytag[®]. With the incorporation of 25% of w-APCr, the 3 evaluated densities increased further, and the major change was observed in the oven-dried particle density $(1.89 \pm 0.01 \text{ g/cm}^3)$, which was 29% higher than the value determined for Lytag $(1.46 \pm 0.01 \text{ g/cm}^3)$. The saturated and surface dried and apparent densities of LWA containing 30% of w-APCr increased with respect to the LWA without the addition of the washed residue, but the oven-dried particle density dropped, even so, its value $(1.57 \pm 0.00 \text{ g/cm}^3)$ was slightly higher than the reference value of Lytag. Despite the increments in density, both formulations, 25%w-APCr and 30%w-APCr, sintered at 1,180°C, showed oven-dried

particle densities lower than 2.0 g/cm³, meeting the requirement for being considered LWA. There are no guidance values for the other two densities, but considering 2.0 g/cm³ as a reference, it is observed that for LWA incorporating 25%w-APCr and 30%w-APCr, the saturated & surface dried particle density was under the limit, while the apparent density exceeded the limit value only by 3 to 5%. Therefore, these densities were also satisfactory for LWA.

6.1.2.3 Loose bulk density and voids

The LWA without incorporation of APCr as well as those incorporating 25% and 30% of w-APCr and fired at 1,180°C were analysed for loose bulk density and percentage of voids according to (BS EN 1097-3, 1998). The calculations and statistical analysis are shown in Appendix XIII. The results are summarised in Figure 6.7.



Figure 6.7 Loose bulk density and voids of hot-bonded LWA fired at 1,180°C and comparison with Lytag®. The error bars correspond to +/- 1 Standard Deviation from the mean value.

Figure 6.7 Part A shows that the LWA containing w-APCr showed loose bulk density values higher than those determined for the LWA without the addition of the washed residue and for Lytag[®]. This means that the incorporation of w-APCr induces an increase in the mass of the LWA per volume unit. The loose bulk density for LWA incorporating 25% and 30% of w-APCr were 1.10 ± 0.01 g/cm³ and 0.91 ± 0.01 g/cm³, respectively. Both formulations met the requirement of not exceeding 1.2 g/cm³ of loose bulk density to be considered LWA.

In Figure 6.7 Part B, it is noticeable the overlapping of the error intervals defined by one standard deviation with respect to the average value of the percentages of voids for the evaluated LWA. Two or more average values can be considered statistically equivalent to others in proportion to the degree of overlapping of their error intervals. Therefore, it can be said that the LWA incorporating 30% of w-APCr had a percentage of voids (42.3%) statistically equivalent to the percentages determined for LWA without the addition of this residue and for Lytag®. Whereas the percentage of voids for LWA containing 25% of w-APCr were lower and statistically different from the other formulations. The percentages of voids were consistent with the bulk density values shown in Figure 6.7 Part A since the more densified formulation (LWA with 25% of w-APCr) contributes more mass and leaves fewer empty spaces in a determined volume.

6.1.2.4 Water absorption

The variation of the water absorption in LWA due to different additions of APCr-m or w-APCr was evaluated over a determined firing temperature range. The calculations and statistics of the results are presented in Appendix XI.

Figure 6.8 shows that the average water absorption of 100%Clay over the firing temperature interval 1,100 - 1,200°C ranged between 7.4% at 1,120°C and 10.4% at 1,160°C. For 5%APCr-m, the water absorption slightly increased at the 3 lowest firing temperatures reaching a maximum of 12.3% at 1,120°C and dropped to 4.5% at 1,160°C and 1.6% at both 1,180°C and 1,200°C. For greater APCr-m additions, it is observed that at the two lowest firing temperatures 1,100°C and 1,120°C, the water absorption increased considerably as the addition of the residue increased. Whereas at 1,140°C, 1,160°C and 1,180°C, the water absorption dropped to values that tended to remain constant over the evaluated range of APCr-m additions.



Figure 6.8. Effect of the firing temperature on the water absorption (wire basket method) of LWA incorporating APCr-m. The error bars correspond to +/- 1 Standard Deviation from the mean value.

Figure 6.9 shows that the average water absorption of 100%Clay varied from 9.34% to 10.42% over the firing temperature interval 1,160 - 1,180°C. With the incorporation of 5% to 30% of w-APCr into the LWA, it is observed a general tendency, according to which the water absorption decreased as the firing temperature increased. The reason is that a higher temperature favours a greater degree of sintering and a surface vitrification, which in turn prevents open porosity. At a constant firing temperature, the water absorption tended to decrease as the incorporation increased from 5% to 25% of w-APCr. Exceptions to this tendency were 5%w-APCr LWA fired at 1,140°C and 10%w-APCr LWA fired at 1,160°C. There is a point of inflexion in 25%w-APCr at the 3 evaluated temperatures, and an increment in the water absorption was observed for 30%w-APCr. This point was particularly marked at 1,140°C. This behaviour indicates that the minimum water absorption of LWA from w-APCr can be achieved with the incorporation of 25% of w-APCr and a firing temperature of 1,180°C. In this case, the minimum water absorption was as low as 1.0%. The LWA 30%w-APCr showed very low water absorption percentages at 1,160°C and 1,180°C but showed the highest water absorption among all the formulations containing w-APCr (14.5%) when the pellets were fired at 1,140°C. This fact suggests that the high water absorption was the result of a lack of sintering due to a low firing temperature and it was not the consequence of the addition of w-APCr.



Figure 6.9 Effect of the firing temperature on the water absorption (determined by the wire basket method) of LWA incorporating w-APCr. The error bars correspond to +/- 1 Standard Deviation from the mean value.

The variations in water absorption can be explained by the degree of vitrification that the LWA exhibited according to the APCr-m or w-APCr incorporation and the firing temperatures. The vitrification of the outer surfaces of LWA has an important role in reducing the water absorption (Gonzalez-Corrochano, et al., 2011). The surface of 100%Clay fired from 1,100°C to 1,180°C were not vitrified. With the incorporation of APCr-m, only the formulations of LWA containing 10% to 25% of the residue and fired at 1,160°C and those containing 10% or 15% of the residue and fired at 1,180°C exhibited glossy vitrified surfaces. In fact, these formulations showed the lowest percentages of water absorption (<1.5%) from the results shown in Figure 6.8. Likewise, the LWA incorporating at least 20% of w-APCr and fired at 1,160°C and 1,180°C showed vitrified surfaces and had the lowest percentages of water absorption (maximum 5.8%) as shown in Figure 6.9.

There is not a regulatory value for water absorption of LWA, so that, the value determined for Lytag® $(9.63 \pm 0.81 \text{ g/cm}^3)$ was considered as a reference for this study. From Figure 6.8 A, the water absorption of 100%Clay was lower than or very close to Lytag® over the evaluated firing temperature range. The LWA containing 5% to 25% of APCr-m and fired at 1,100°C or 1,120°C, as well as 5%APCr-m fired at 1,140°C showed higher water absorption than Lytag®. The LWA incorporating 10% to 25% of APCr-m fired at a

1,140°C, 5% to 25% fired at 1,160°C, 5% to 15% of APCr-m fired at 1,180°C, and 5% of APCr-m fired and 1,120°C were all under the reference value of Lytag®. From Figure 6.8 B, the LWA containing 5% to 30% of w-APCr fired at 1,140°C, except 25%w-APCr exceeded the water absorption of Lytag®. While the LWA containing 5% to 30% of w-APCr, excepting 10%w-APCr fired at 1,160°C, were below the reference water absorption of Lytag®.

The LWA formulations 100%Clay, 25%w-APCr and 30%w-APCr were produced at a firing temperature of 1,180°C and tested for water absorption according to the pycnometer method. This is the method recommended by the British Standard (BS EN 1097-6, 2022) for LWA, and it is performed over a batch of aggregates rather than in a single particle. The calculations and statistical analysis are presented in Appendix XII. Figure 6.10 summarises the results and comparison with Lytag®.



Figure 6.10 Water absorption (determined by the method of the pycnometer (BS EN 1097-6, 2022))of hot-bonded LWA fired at 1,180°C and comparison with Lytag®. The error bars correspond to +/- 1 Standard Deviation from the mean value.

Figure 6.10 shows that the water absorption in 24 hours of soaking was comparable between 100%Clay (4.7%) and 25%w-APCr (4.4%), and both percentages were well below the reference value determined for Lytag® (11.4%). Whereas the water absorption at 24 hours for 30%w-APCr was the highest among the tested formulations (16.2%) exceeding the value of Lytag®. It is observed that for Lytag® as well as for the evaluated LWA formulations 100%Clay, 25%w-APCr and 30%w-APCr fired at 1,180°C, the water

absorption increased as the soaking time increased. For the LWA containing w-APCr, similarly to Lytag®, most of the water absorption took place during the first 10 min of the test. In only 10 min, 25%w-APCr and 30%w-APCr absorbed, respectively, about 70% and 85% of the total amount of water absorbed in 24 hours.

The high percentage of water absorption achieved by 30%w-APCr in this test was not the value expected, considering that this formulation only absorbed 1.9% of water in 24 hours as per the wire basket method, reported in Figure 6.8. The reason for this discrepancy could be attributed to a lack of sintering of the batch of 30%w-APCr that was analysed by the pycnometer method. A low degree of sintering increased the open porosity and consequently incremented the water absorption of the LWA. Probably, during the production of this batch, the furnace was not operating at the set firing temperature (1,180°C) but at a lower temperature that altered the proper sintering process. Figure 6.11 shows the appearance of the LWA 25%w-APCr (A) and 30%w-APCr (B) fired at 1,180°C that were used for this test. It can be seen that 25%w-APCr mostly showed a glossy vitrified surface, while 30%w-APCr did not exhibit such vitrification. The appearance of 30%w-APCr did not match with the one shown by the batches of the same formulation produced for other tests and also shown in Figure 6.1.



Figure 6.11 Hot-bonded LWA fired at 1,180°C and digital microscopy images showing degree of sintering.

6.1.2.5 Crushing strength

The effect of the firing temperature on the crushing strength of individual pellets of LWA incorporating APCr-m or w-APCr was evaluated. The results were also compared with the crushing strength determined in the laboratory for Lytag®. The calculations and statistical analysis of the results are reported in Appendix XIV.

Figure 6.12 shows the variation of the crushing strength of LWA from 100% of clay and from LWA containing additions from 5% to 25% of APCr-m and fired over the range of 1,120°C to 1,180°C. The wide error intervals (defined by one standard deviation with respect to the average value) of the crushing strength measurements were due to a lack of uniformity in the strength of individual pellets and a deviation from a perfect sphere shape. Despite this, the average values clearly show the tendencies. For 100%Clay, the average crushing strength was higher at 1,120°C (4.47 MPa) and it showed slightly lower values (about 2.7 MPa) from 1,140° to 1,180°C. With the incorporation of 5% to 25% of APCr-m, the crushing strength, in general, decreased at the firing temperatures 1,120°, 1,140°C and 1,160°C. Exceptions to this tendency were the crushing strengths registered for 10%APCr-m fired at 1,160°C and 15%APCr-m fired at 1,120°C, which were 6.02 and 5.31 MPa, respectively, both values exceeding the crushing strength of 100%Clay. The three formulations evaluated at 1,180°C exhibited crushing strengths higher than the values determined for 100%Clay. At this temperature, 5%APCr-m and 15%APCr-m had crushing strengths around 5.5 MPa, while 10%APCr-m exhibited the highest crushing strength (10.87MPa) from all the batches of LWA with the incorporation of APCr-m. The five formulations of LWA with the highest crushing strengths (15%APCr-m fired at 1,140°C, 10%APCr-m fired at 1,160°, and 5%APCr-m, 10%APCr-m and 15%APCr-m fired at 1,180°C) exceeded the crushing strength of 3.23 MPa determined for Lytag[®].



Figure 6.12. Effect of the firing temperature on the crushing strength of LWA incorporating APCr-m. The error bars correspond to +/- 1 Standard Deviation from the mean value.

Figure 6.13 presents the variation of the crushing strength in LWA due to the incorporation of 5% to 30% of w-APCr over a firing temperature interval between 1,140°C and 1,180°C. The crushing strength of 100%Clay increased as the firing temperature increased, from 2.51 MPa at 1,140°C to 2.91 MPa at 1,180°C. The incorporation of 5% to 30% of w-APCr and the firing at 1,140°C had a detrimental effect on the crushing strength of the LWA, reducing its value up to 0.71 MPa for 30%w-APCr. The other formulations with crushing strength lower than 100%Clay were 5%w-APCr, 10%-APCr, 15%w-APCr and 30%w-APCr fired at 1,160°C and 5%w-APCr, 10%-APCr and 30% w-APCr fired at 1,180°C. The crushing strength of 20%w-APCr fired at 1,160°C was comparable to 100%Clay. The highest values of crushing strength ranged around 4.0 MPa, they were obtained for 25%w-APCr fired at 1,160°C, and 15%w-APCr, 20%w-APCr and 25%w-APCr fired at 1,180°C. These LWA were stronger than Lytag®, whose average crushing strength was 3.23 MPa.

It is worth noticing that most of the error intervals of the crushing strength average values of LWA containing w-APCr had a width of about 1 MPa. By comparison of these intervals with the wider intervals observed in Figure 6.12 for LWA made from APCr-m,

it can be said that the incorporation of w-APCr favours the uniformity of the single pellets in a batch of LWA in terms of crushing strength.



Figure 6.13 Effect of the firing temperature on the crushing strength of LWA incorporating w-APCr. The error bars correspond to +/- 1 Standard Deviation from the mean value.

6.1.3 Leaching properties of hot-bonded LWA

The leaching behaviour of the hot-bonded LWA is affected by the leaching properties of the raw materials (clay and APCr-m or w-APCr). Evidently, the contribution from each raw material will depend on the different percentages of APCr incorporation and it will be modified by the thermal treatment. These materials were analysed for the leaching compliance test (BS EN 12457-2, 2002) to evaluate their contribution to the leachate concentrations of the LWA. The results for APCr-m and w-APCr were previously reported in Table 5.1. The results for w-APCr correspond to the washing treatment T₃ (optimum conditions L/S: 2.5, time: 60 min). Figure 6.14 summarises the leachate concentrations of heavy metals (A) and soluble salts (B) for clay, APCr-m and w-APCr. It can be seen from Figure 6.14 A that there was not any leaching of metals from the clay (the release of Ba is negligible), while the high leaching rates of Pb from APCr-m and Ba from APCr-m and w-APCr, although moderate, could also affect the leaching properties

of the final products. Figure 6.14 B shows that APCr-m and w-APCr with leaching rates of chlorides at 5 and 3 orders of magnitude of mg/kg, respectively, determine the leaching of chlorides in LWA, while the contribution from clay (67.6 mg/kg) is negligible. The leaching rates of sulphates from clay and w-APCr were at 3 orders of magnitude of mg/kg and from APCr-m was at 4 orders of magnitude of mg/kg, so that, the contribution of the 3 raw materials should be considered for the LWA production.



Figure 6.14. Leaching from clay, APCr-m and w-APCr. A: heavy metals, B: soluble salts.

The leaching properties of hot-bonded LWA produced from APCr-m or w-APCr and fired over the range of 1,140° to 1,180°C were evaluated. The results were compared with the leaching rates determined in the laboratory for Lytag[®] and with the limit values for waste acceptance at hazardous, non-hazardous and inert landfills as expressed in the Landfill

Directive (The Council of the European Union, 2003). The analysis of the results will be discussed in the following sections.

6.1.3.1 Leaching properties of LWA incorporating APCr-m

Table 6.4 presents the results of the leaching tests for 100%Clay and for LWA incorporating 5% to 25% of APCr-m fired over the interval 1,140°C to 1,180°C. The results obtained for Lytag® and the WAC limit values are shown for comparison. The findings will be discussed in the following sections.

1,140°C								1,160°C							0°C		Limits for WAC			
	100% Clay	5% APCr- m	10% APCr- m	15% APCr- m	20% APCr- m	25% APCr- m	100% Clay	5% APCr- m	10% APCr- m	15% APCr- m	20% APCr- m	25% APCr- m	100% Clay	5% APCr- m	10% APCr- m	15% APCr- m	Lytag	Hazardous	Non- hazardous	Inert
BS 12457-2, LS = 10 (mg/kg)																				
As	<1.582	<1.582	<1.582	<1.582	<1.582	<1.582	<1.582	<1.582	<1.582	<1.582	<1.582	<1.582	<1.582	<1.582	<1.582	<1.582	1.6	25	2	1
Ba	0.3	0.8	1.0	1.7	1.3	2.4	0.5	0.7	0.8	1.0	1.6	1.5	0.8	0.3	0.6	1.3	< 0.231	300	100	20
Cd	< 0.071	< 0.071	< 0.071	< 0.071	< 0.071	< 0.071	< 0.071	< 0.071	< 0.071	< 0.071	< 0.071	< 0.071	< 0.071	< 0.071	< 0.071	< 0.071	< 0.071	5	1	0
Cr	< 0.201	0.3	0.3	0.4	< 0.201	< 0.201	< 0.201	< 0.201	< 0.201	< 0.201	< 0.201	0.3	< 0.201	< 0.201	0.3	< 0.201	< 0.201	70	10	1
Cu	< 0.712	< 0.712	< 0.712	< 0.712	< 0.712	< 0.712	< 0.712	< 0.712	< 0.712	< 0.712	< 0.712	< 0.712	< 0.712	< 0.712	< 0.712	< 0.712	< 0.712	100	50	2
Hg	<1.158	<1.158	<1.158	<1.158	<1.158	<1.158	<1.158	<1.158	<1.158	<1.158	<1.158	<1.158	<1.158	<1.158	<1.158	<1.158	<1.158	2	0	0
Mo	<1.522	<1.522	<1.522	<1.522	<1.522	<1.522	<1.522	<1.522	<1.522	<1.522	<1.522	<1.522	<1.522	<1.522	<1.522	<1.522	<1.522	30	10	1
Ni	< 0.325	< 0.325	< 0.325	< 0.325	< 0.325	< 0.325	< 0.325	< 0.325	< 0.325	< 0.325	< 0.325	< 0.325	< 0.325	< 0.325	< 0.325	< 0.325	< 0.325	40	10	0
Pb	<2.209	<2.209	<2.209	<2.209	<2.209	<2.209	<2.209	<2.209	<2.209	<2.209	<2.209	<2.209	<2.209	<2.209	<2.209	<2.209	<2.209	50	10	1
Sb	< 0.858	< 0.858	< 0.858	< 0.858	< 0.858	< 0.858	< 0.858	< 0.858	< 0.858	< 0.858	< 0.858	< 0.858	< 0.858	< 0.858	< 0.858	< 0.858	< 0.858	5	1	0
Se	<1.008	<1.008	<1.008	<1.008	<1.008	<1.008	<1.008	<1.008	<1.008	<1.008	<1.008	<1.008	<1.008	<1.008	<1.008	<1.008	<1.008	7	1	0
Zn	<2.483	<2.483	<2.483	<2.483	<2.483	<2.483	<2.483	<2.483	<2.483	<2.483	<2.483	<2.483	<2.483	<2.483	<2.483	3.7	<2.483	200	50	4
Cl-	<51.0	<51.0	<51.0	<51.0	1,211.7	2,836.1	<51.0	<51.0	<51.0	86.3	402.3	922.7	<51.0	<51.0	67.3	111.8	<51.0	25,000	15,000	800
F-	<20.3	<20.3	<20.3	<20.3	<20.3	<20.3	<20.3	<20.3	<20.3	<20.3	<20.3	<20.3	<20.3	<20.3	<20.3	<20.3	<20.3	500	150	10
SO_4^{-2}	6,277	7,130	6,741	6,675	5,992	5,879	5,350	5,546	4,895	9,935	4,892	3,457	4,548	2,654	3,392	7,793	437	50,000	20,000	1,000
TDS	9,000	8700	7,800	8,400	9,200	12,400	5,800	3,900	4,400	12,100	8,700	6300	3,600	1,600	1,800	11,100	1,100	100,000	60,000	4,000
DOC	<100	<100	<100	<100	<100	<100	<100	<100	<100	<100	<100	<100	<100	<100	<100	<100	<100	1,000	800	500
BS 12	457-2, I	LS/10					•						•							
pH at	20°C																			
	11.59	10.13	8.26	7.31	8.10	7.65	11.55	9.97	8.27	7.55	7.95	10.97	10.34	9.55	9.18	8.19	9.16			
Cond	uctivity a	at 20°C (μS/cm)																	
	1,517	1136	1,057	1,051	1,367	1,810	1,381	934	842	1,503	1,216	1075	886	533	664	1,396	137			
C 11	1 . 1	1. 1. 1	1 .1		1	1														

Table 6.4. Results of leaching test (BS EN 12457-2, 2002) on hot-bonded LWA incorporating APCr-m and comparison with Lytag® and WAC limits.

Cells are highlighted where the parameter was detected. < values indicate the limits of detection.

6.1.3.1.1 Leaching of heavy metals from LWA incorporating APCr-m

Table 6.4 shows that from the analysed metals, only Ba was detected in the leachates from all the LWA formulations at the 3 evaluated firing temperatures, including 100% Clay and the LWA with the incorporation of APCr-m. The leaching rates of Ba for LWA containing APCr-m ranged between 0.3 mg/kg (5%APCr-m fired at 1,180°C) and 2.4 mg/kg (25%APCr-m fired at 1,140°C), values that were well below the inert WAC limit of 20 mg/kg. Cr was released from 5%APCr-m, 10%APCr-m and 15%APCr-m fired at 1,140°C, 25%APCr-m fired at 1,160°C and 10%APCr-m fired at 1,180°C. From these formulations, the maximum leaching of Cr was 0.4 mg/kg from 15%APCr-m fired at 1,140°C, and the others released 0.3 mg/kg so that they all were under the inert WAC limit of 1.0 mg/kg. Zn was detected only in the leachate from 25%APCr-m fired at 1,180°C with a leaching rate equal to 3.7 mg/kg, being under the inert WAC limit of 4 mg/kg for this metal. Apart from Ba, Cr and Zn, none of the other 9 metals regulated for the WAC was detected in the leachates of the LWA with or without incorporation of APCr-m. The presence of Ba in all the LWA, and Cr and Zn in some of them, differed from Lytag[®], which did not release metals excepting As.

Figure 6.15 shows the relationship between the firing temperature with the release of Ba and Cr in the LWA containing increasing additions of APCr-m. For 100%Clay, the leaching of Ba increased as the firing temperature increased. After the incorporation of APCr-m, the release of Ba was reduced by increasing the firing temperature, except for 20%APCr-m. Despite the control by the firing temperature, the leaching rate of Ba kept rising with the increasing additions of APCr-m. Since 15%APCr-m was the LWA with the maximum addition of APCr-m fired at 1,180°C, it can be said that this formulation ensures the right control of leaching of Ba. On the other hand, Cr was not released from 100%Clay at any of the evaluated firing temperatures. In the LWA containing APCr-m that released Cr, this leaching occurred particularly at the lowest firing temperature 1,140°C without variation regarding the APCr-m addition.



Figure 6.15 Effect of the APCr-m addition and the firing temperature on the leaching of heavy metals from hot-bonded LWA.

6.1.3.1.2 Leaching of soluble salts and TDS from LWA incorporating APCr-m

According to the results shown in Table 6.4, sulphates were leached out from all the manufactured LWA at the 3 evaluated firing temperatures, including 100%Clay and those incorporating 5% to 25% of APCr-m. Among the LWA containing APCr-m, the leaching of sulphates ranged from 2,654 mg/kg (5%APCr-m fired at 1,180°C) to 9,935 mg/kg (15%APCr-m fired at 1,160°C). This leaching interval was much higher than Lytag®, which released sulphates at a rate of 437 mg/kg. The leaching of sulphates for 100%Clay and for LWA containing 5% to 25% of APCr-m exceeded the inert WAC limit of 1,000 mg/kg but were below the non-hazardous WAC limit of 20,000 mg/kg. Chlorides did not leach out from any of the LWA 100%Clay, but they were released from 20%APCr-m and 25%APCr-m fired at 1,140°C, 15%APCr-m, 20%APCr-m and 25%APCr-m fired at 1,160°C, and 10%APCr-m and 15%APCr-m fired at 1,180°C. The leaching rates of chlorides varied between 67.3 mg/kg (10%APCr-m fired at 1,180°C) and 2,836 mg/kg (25%APCr-m fired at 1,140°C). These LWA differed from Lytag[®], which did not release chlorides. The LWA 20%APCr-m and 25%APCr-m fired at 1,140°C, and 25%APCr-m fired at 1,160°C, exceeded the inert WAC limit of 800 mg/kg for chlorides, but were well below the non-hazardous WAC of 15,000 mg/kg for these salts. The other formulations

releasing chlorides were below the inert WAC limit. There was no leaching of fluorides either from the manufactured LWA or from Lytag[®].

Figure 6.16 shows the relationship between the firing temperature and the APCr-m addition with the leaching of soluble salts and TDS in hot-bonded LWA. The high leaching rates of sulphates (Figure 6.16 Part A) from all the manufactured LWA and the leaching of chlorides (Figure 6.16 Part B) from some of the LWA containing APCr-m explain the high leachate concentrations of TDS (Figure 6.16 Part C). The leaching of sulphates represented more than 50% of the total TDS leaching concentrations. For 100% Clay and for LWA containing APCr-m, the leaching rates of soluble salts, both sulphates and chlorides and TDS decreased as the firing temperature increased. An exception to this tendency was observed in 15%APCr-m, for which the lowest leaching rates of sulphates and TDS were achieved at 1,140°C. The greatest reductions in leaching of sulphates and TDS were achieved by 5%APCr-m and 10%APCr-m, fired at 1,180°C, becoming closer to the leaching rates of Lytag[®]. This fact indicates that for LWA with low contents of APCr-m, the leaching of sulphates, and consequently of TDS, can be effectively controlled by increasing the firing temperature. However, the LWA with greater additions of APCr-m (20%APCr-m and 25%PCr-m) did not tolerate the firing treatment at a temperature higher than 1,160°C; consequently, for these formulations it was not possible a further reduction of the sulphates and TDS leaching rates. On the other hand, it is observed that in LWA with different contents of APCr-m, the leaching rates of sulphates were comparable to 100%Clay when the same firing temperature was used, particularly 1,140°C. This means that apart from the sulphates leaching out from the additions of APCr-m, the contribution to the leaching of these salts from the clay should be taken into consideration.



Figure 6.16 Effect of APCr-m addition and firing temperature on the leaching of soluble salts from hot-bonded LWA. A: Leaching of sulphates, B: Leaching of chlorides, C: Leaching of TDS.

6.1.3.1.3 pH of leachates from LWA incorporating APCr-m

Figure 6.17 shows the effect of the firing temperature on the pH of the leachates from the LWA incorporating APCr-m. The leachates of 100%Clay exhibited a highly alkaline character (pH above 10). The 3 evaluated firing temperatures reduced the pH of the leachates. Greater reductions were observed for the additions from 10% to 25% of APCr-m (excepting 25%APCr-m fired at 1,160°C) up to values lower than the reference of Lytag® and close to the neutral pH 7.



Figure 6.17 Effect of APCr-m incorporation and firing temperature on the pH of the leachates from hot-bonded LWA.

6.1.3.2 Leaching properties of LWA incorporating w-APCr

Table 6.5 presents the results of the leaching tests for LWA incorporating 5% to 30% of w-APCr fired over the interval 1,140°C to 1,180°C. The results obtained for, Lytag® and the WAC limit values are shown for comparison. The findings will be discussed in the following sections.

1,140°C							1,160°C								1,18	30°C		Limits for WAC				
	5%	10%	15%	20%	25%	30%	5%	10%	15%	20%	25%	30%	5%	10%	15%	20%	25%	30%	Lytag	Hazardous	Non-	Inert
	APCr	APCr	APCr	APCr	APCr	APCr	APCr	APCr	APCr	APCr	APCr	APCr	APCr	APCr	APCr	APCr	APCr	APCr		Tiazardous	hazardous	mert
BS 12457-2 LS = 10 (mg/kg)																						
As	<1.582	<1.582	<1.582	<1.582	<1.582	<1.582	<1.582	<1.582	<1.582	<1.582	<1.582	<1.582	<1.582	<1.582	<1.582	<1.582	<1.582	<1.582	1.6	25	2	1
Ba	1.7	1.2	0.8	0.8	1.6	0.6	1.6	1.0	1.0	1.0	0.4	0.6	1.5	1.1	1.6	2.1	1.2	1.0	<0.231	300	100	20
Cd	< 0.071	< 0.071	< 0.071	< 0.071	< 0.071	< 0.071	< 0.071	< 0.071	< 0.071	< 0.071	< 0.071	< 0.071	< 0.071	< 0.071	< 0.071	< 0.071	< 0.071	< 0.071	< 0.071	5	1	0
Cr	0.7	0.9	1.2	1.0	0.6	0.8	0.5	1.0	1.2	0.9	1.0	0.5	0.4	0.8	0.7	0.8	0.6	0.4	< 0.201	70	10	1
Cu	< 0.712	< 0.712	< 0.712	< 0.712	< 0.712	< 0.712	< 0.712	< 0.712	< 0.712	< 0.712	< 0.712	< 0.712	< 0.712	< 0.712	< 0.712	< 0.712	< 0.712	< 0.712	<0.712	100	50	2
Hg	<1.158	<1.158	<1.158	<1.158	<1.158	<1.158	<1.158	<1.158	<1.158	<1.158	<1.158	<1.158	<1.158	<1.158	<1.158	<1.158	<1.158	<1.158	<1.158	2	0	0
Mo	<1.522	<1.522	<1.522	<1.522	<1.522	<1.522	<1.522	<1.522	<1.522	<1.522	<1.522	<1.522	<1.522	<1.522	<1.522	<1.522	<1.522	<1.522	<1.522	30	10	1
Ni	< 0.325	< 0.325	< 0.325	< 0.325	< 0.325	< 0.325	< 0.325	< 0.325	< 0.325	< 0.325	< 0.325	< 0.325	< 0.325	< 0.325	< 0.325	< 0.325	< 0.325	< 0.325	< 0.325	40	10	0
Pb	<2.209	<2.209	<2.209	<2.209	<2.209	<2.209	<2.209	<2.209	<2.209	<2.209	<2.209	<2.209	<2.209	<2.209	<2.209	<2.209	<2.209	<2.209	<2.209	50	10	1
Sb	< 0.858	< 0.858	< 0.858	<0.858	< 0.858	< 0.858	<0.858	< 0.858	< 0.858	< 0.858	< 0.858	< 0.858	< 0.858	< 0.858	< 0.858	< 0.858	< 0.858	< 0.858	< 0.858	5	1	0
Se	<1.008	<1.008	<1.008	<1.008	<1.008	<1.008	<1.008	<1.008	<1.008	<1.008	<1.008	<1.008	<1.008	<1.008	<1.008	<1.008	<1.008	<1.008	<1.008	7	1	0
Zn	<2.483	<2.483	<2.483	<2.483	<2.483	<2.483	<2.483	<2.483	<2.483	<2.483	<2.483	<2.483	<2.483	<2.483	<2.483	<2.483	<2.483	<2.483	<2.483	200	50	4
Cl-	56.2	60.7	<51	<51	<51	<51	65.4	<51	<51	<51	<51	<51	<51	<51	<51	<51	53.2	58.7	<51.0	25,000	15,000	800
F-	<20.3	<20.3	<20.3	<20.3	<20.3	<20.3	<20.3	<20.3	<20.3	<20.3	<20.3	<20.3	<20.3	<20.3	<20.3	<20.3	<20.3	<20.3	<20.3	500	150	10
SO_4^{-2}	12,573	12,248	12,042	9,155	7,840	10,392	10,014	11,336	10,124	10,182	10,424	7,248	9,465	8,331	7,421	6,907	6,498	5,762	437	50,000	20,000	1,000
TDS	19,000	18,900	18,200	13,600	8,800	16,200	16,200	17,300	15,800	14,300	16,100	10,700	14,700	13,600	12,500	10,700	9,600	9,000	1,100	100,000	60,000	4,000
DOC	169	<100	<100	<100	<100	169	<100	<100	<100	<100	<100	<100	<100	<100	<100	168	<100	<100	<100	1,000	800	500
BS 12	457-2 L	LS/10																				
pH at	20°C																					
	10.82	10.63	9.84	9.21	9.04	9.36	10.52	10.38	9.99	8.50	8.24	9.15	10.32	8.52	7.85	7.91	8.11	7.75	9.160			
Condu	uctivity	at 20°C	(µS/cm	I)																		
	1,759	1767	1,680	1,308	925	1,505	1,533	1,644	1469	1,372	1,371	1,079	1,296	1,179	1,181	1,040	976	933	137			
Calla	ara hiah	lightad	for the	dataataa	Inorom	atore																

Table 6.5 Results of leaching test (BS EN 12457-2, 2002) on hot-bonded LWA incorporating w-APCr and comparison with Lytag® and WAC limits.

Cells are highlighted for the detected parameters.

< values indicate the limits of detection.

6.1.3.2.1 Leaching of heavy metals from LWA incorporating w-APCr

Table 6.5 shows that from the analysed metals, only Ba and Cr were detected in the leachates from all the LWA incorporating w-APCr at the 3 evaluated firing temperatures. The leaching rates of Ba varied between 0.8 mg/kg (15%w-APCr and 20%w-APCr fired at 1,140°C) and 2.1 mg/kg (20%w-APCr fired at 1,180°C), values that were below the inert WAC limit of 20 mg/kg. The leaching rates of Cr varied from 0.4 mg/kg (5%w-APCr and 30%w-APCr fired at 1,180°C) to 1.2 mg/kg (15%w-APCr fired at 1,140°C and 1,160°C). Apart from the maximum value of this interval, the leaching rates of Cr were below the inert WAC limit of 1.0 mg/kg. The leaching of Ba and Cr from all the LWA incorporating w-APCr differed from Lytag®, which did not release metals except As.

Figure 6.18 shows the effect of the firing temperature on the leaching of Ba and Cr from the LWA containing increasing additions of w-APCr. The release of Ba ions (Figure 6.18 Part A) was controlled by the firing temperature. For the LWA 15%w-APCr, 20%w-APCr and 30%w-APCr, the leaching of Ba increased as the firing temperature increased. The same tendency was observed for 100%Clay. However, a reduction of the leaching of Ba by increasing the firing temperature was observed in the formulations 5%w-APCr, 10%w-APCr and 25%w-APCr. The leaching of Cr (Figure 6.18 Part B) was in general reduced by increasing the firing temperature. The formulations 10%w-APCr and 25%w-APCr fired at 1,160°C were the only exceptions for this tendency.



Figure 6.18. Leaching of Barium and Chromium from LWA incorporating w-APCr. A: Leaching of Ba, B: Leaching of Cr.

6.1.3.2.2 Leaching of soluble salts from LWA incorporating w-APCr

According to the results shown in Table 6.5, sulphates leached out from all the LWA containing w-APCr at the 3 evaluated firing temperatures. The leaching rates of sulphates ranged between 5,762 mg/kg (30%w-APCr fired at 1,180°C) and 12,573 mg/kg (5%APCr-m fired at 1,140°C). This leaching interval was much higher than Lytag® (437

mg/kg). It also exceeded the inert WAC limit (1,000 mg/kg), although it was under the non-hazardous WAC limit (20,000 mg/kg). Chlorides were released from 5%w-APCr and 10%w-APCr fired at 1,140°C, 5%w-APCr fired at 1,160°C, and 25%w-APCr and 30%w-APCr fired at 1,180°C. The leaching rates of chlorides were very low, varying from 53.2 mg/kg (25%w-APCr fired at 1,180°C) to 65.4 mg/kg (5%w-APCr fired at 1,140°C). These LWA differed from Lytag[®], from which there was not leaching of chlorides. Also, these leaching rates of chlorides were well below the inert WAC of 800 mg/kg. None of the LWA incorporating w-APCr leached out fluorides.

Figure 6.19 shows the effect of the firing temperature on the leaching of sulphates and TDS from LWA incorporating 5% to 30% of w-APCr. Chlorides were excluded from this graph because their leaching rates were negligible. The high leaching rates of sulphates (Figure 6.19 Part A) from all the manufactured LWA containing w-APCr represented more than 50% of the total TDS leaching concentrations (Figure 6.19 Part B). For all the LWA incorporating w-APCr, the leaching rates of both sulphates and TDS were generally reduced by increasing the firing temperature. The greatest reductions in leaching of sulphates and TDS were achieved by the LWA 25%w-APCr and 30%w-APCr fired at 1,180°C. For both formulations, the leaching rates of sulphates were comparable to 100%Clay fired at any of the 3 evaluated temperatures (no more than 6,500 mg/kg). Nevertheless, the leaching of sulphates and TDS from all the LWA containing w-APCr were much higher than the leaching rates determined for Lytag® (437 mg/kg for sulphates and 1,100 mg/kg for TDS). As previously shown in Figure 6.14, the clay had an initial leaching of sulphates of 6,351 mg/kg, which was not significantly reduced by the firing of the LWA 100%Clay, as can be seen in Figure 6.19, Consequently, the persistent release of sulphates from the LWA was due to both clay and the w-APCr.



Figure 6.19. Leaching of sulphates and TDS from LWA incorporating w-APCr. A: Leaching of sulphates, B: Leaching of TDS.

The main problem associated with the sulphates leaching from building materials is the sulphate attack on concrete. There are two forms of sulphate attack, the conventional and the thaumatise form (Building Research Establishment, 2008). In the conventional attack, sulphates and water react with the tricalcium aluminate from the OPC to form ettringite (3CaO·Al₂O₃·3CaSO₄·31H₂O) (Bai, 2016). This mineral has a solid volume greater than the reactants and usually grows as needle-shaped crystals that in group can produce an internal stress (Building Research Establishment, 2008). This leads to expansion and

fracture of the concrete structure (Bai, 2016). In the thaumatise form of sulphate attack, the reaction product is a calcium silicate carbonate sulphate hydrate called thaumatise $(CaSiO_3 \cdot CaCO_3 \cdot CaSO_4 \cdot 15H_2O)$. This causes the softening and loss of concrete strength as well as some expansion (Building Research Establishment, 2008).

6.1.3.2.3 pH of leachates from LWA incorporating w-APCr

From Table 6.5 it is observed that the pH values of the leachates from LWA incorporating 5% to 30% of w-APCr ranged from 7.75 (30%w-APCr-m fired at 1,180°C) to 10.82 (5%w-APCr fired at 1,140°C). This interval comprised the pH value of the leachate produced by Lytag[®] (9.16). Figure 6.20 shows the effect of the firing temperature on the pH of the leachates generated from the LWA incorporating w-APCr. It is observed that at the same addition of w-APCr, the pH values of the leachates were reduced by increasing the firing temperature. The only exception to this tendency was 15%w-APCr fired at 1,160°C. At a constant firing temperature, the pH decreased as the w-APCr incorporation decreased. The firing temperature of 1,180°C favours the reduction on the pH up to values closer to the neutrality.



Figure 6.20 Effect of the firing temperature on the pH of the leachates produced by LWA incorporating w-APCr.

6.1.4 Mineralogy of hot-bonded LWA

The LWA 25%APCr-m fired at 1,160°C and 25%w-APCr fired at 1,180°C were assessed in their mineralogy and compared with 100%Clay fired at 1,180°C. These formulations were selected because they exhibited the optimum technological characteristics, more favourable leaching properties and allowed high incorporation of APCr. Figure 6.21 shows the XRD patterns with markers at the main identification peaks for the mineral phases detected in the mentioned LWA and in the raw clay. Table 6.6 presents the results of the quantitative analysis for these LWA formulations.


Figure 6.21 XRD patterns showing the identification of the main peaks for hot-bonded LWA: 100%Clay and 25%w-APCr fired at 1,180°C and 25%APCr-m fired at 1,160°C.

Table 6.6 Quantification of mineral phases and estimation of amorphous material content by Rietveld analysis for hot-bonded LWA: 100%Clay and 25%w-APCr fired at 1,180°C and 25%APCr-m fired at 1,160°C.

Mineral	Chemical formula	wt %					
phase	Chemical formula	100%Clay	25%APCr-m	25%w-APCr			
Quartz	SiO ₂	32.39 ± 0.25	21.81 ± 0.33	16.72 ± 0.17			
Anorthite	$CaAl_2Si_2O_8$	20.34 ± 0.33	-	-			
Bytownite	A17.76Ca3.44Na0.56O32Si8.24	3.44 ± 0.47	-	-			
Vanthoffite	MgNa ₆ (SO ₄) ₄	0.29 ± 0.02	-	-			
Albite	NaAlSi ₃ O ₈	-	15.69 ± 0.84	9.44 ± 0.26			
Anhydrite	CaSO ₄	-	1.39 ± 0.69	4.22 ± 0.20			
Sanidine	KAlSi ₃ O ₈	-	-	0.59 ± 0.13			
	% Crystalline material	56.47 ± 0.63	38.89 ± 1.14	30.97 ± 0.39			
	% Amorphous material	43.53 ± 0.63	61.11 ± 1.14	69.03 ± 0.39			

 \pm values indicate one standard deviation.

6.1.4.1 Mineralogy of 25%APCr-m fired at 1,160°C

Figure 6.21 shows that quartz was the main phase identified in 25%APCr-m, showing peaks with intensities only slightly lower than those observed for 100%Clay. The identity peaks for quartz were spread from 20 °2 θ to 68 °2 θ , and the highest intensities were observed at 26.62 °2 θ , 20.84 °2 θ and 50.11 °2 θ . The presence of quartz in the LWA was mainly due to the presence of the phase in the clay and a small contribution from APCr-m (mineralogy of APCr-m is reported in Figure 5.6.). The transitions of quartz derived from the thermal treatment of the LWA can be explained according to the description provided by (Götze, et al., 2021). In both raw materials, clay and APCr-m, the form of quartz was the trigonal low-temperature α -quartz, which is stable at ambient conditions. The second phase detected in 25%APCr-m was albite, for which the main peaks were located at 27.99 °2 θ and 22.01 °2 θ . Albite was present in 25%APCr-m, probably as a product of the thermal decomposition of phases such as muscovite from the clay and silicates, like clinotobermorite from APCr-m. The third phase detected in 25%APCr-m was anhydrite, for which the main peak was at 25.48 °2 θ .

Anhydrite in 25%APCr-m may be the product of the dehydration of gypsum from the clay or from APCr-m. Also, the content of anhydrite in APCr-m may contribute to the amount of this phase in the final LWA because this phase is thermally stable at the firing temperature 1,160°C. Despite the similarity between the patterns of 100%Clay and 25%APCr-m, the qualitative analysis showed that the phases formed due to the firing in 100%Clay (bytownite, vanthoffite and anorthite) were not formed in 25%APCr-m. It should be noted that the peaks at 21.99 °2 θ and 27.93 °2 θ identify vanthoffite and anorthite, respectively for 100%Clay, while the very close peaks at 22.01°2 θ and 28.06°2 θ identify albite in 25%APCr-m.

Table 6.6 shows that quartz and albite were the most abundant mineral phases in 25%APCr-m, representing 21.81wt.% and 15.69 wt%, respectively, of the total mineral content of this LWA. Anhydrite, although with a concentration as low as 1.39 wt.%, would be responsible for the sulphates leaching from 25%APCr-m, since this is the only sulphate-bearing phase forming part of its mineralogy. The total content of crystalline material of 25%APCr-m (38.89 wt.%) was noticeably lower than the content determined for 100%Clay (56.47 wt.%). Apart from anhydrite and albite, none of the other main phases of APCr-m (such as calcite, calcium hydroxychloride, halite, sylvite, among others) was detected in the LWA 25%APCr-m. This indicates that the majority of the mineral compounds from APCr-m were effectively removed or transformed into the more stable phase albite by the firing of 25%APCr-m at 1,160°C. However, the mineralogy of 25%APCr-m does not explain the leaching of chlorides (reported in Table 6.4). Therefore, further research is required to find the origin of the leaching of chlorides and to diminish the leaching of sulphates caused by anhydrite.

6.1.4.2 Mineralogy of 25%w-APCr fired at 1,180°C

Figure 6.21 shows that quartz, albite, anhydrite and sanidine were identified as the phases forming the mineralogy of 25%w-APCr. The intensities of the peaks identifying quartz, albite and anhydrite were lower than those observed for 100%Clay and for 25%APCr-m. Peaks identifying albite were noticeable in the pattern of 25%w-APCr, located at 27.96 °20 and 22.04 °20. The presence of albite in 25%w-APCr could be attributed to the thermal decomposition of muscovite from the clay and clinotobermorite from w-APCr. Anhydrite was detected in 25%w-APCr. Anhydrite in this LWA may be the product of the dehydration of gypsum from the clay and the contribution in the content of this phase

from w-APCr. The pattern for identification of sanidine in 25%w-APCr was very similar to albite with some distinctive peaks, for instance, a peak at 21.14 °20. The peaks at 21.99 °20 and 27.93 °20 identify vanthoffite and anorthite, respectively for 100%Clay, while the very close peaks at 22.05 °20 and 27.96 °20 identify albite in 25%w-APCr.

Table 6.6 shows that quartz and albite were the most abundant mineral phases in 25%wAPCr, with the concentration of 16.72 wt.% and 9.44 wt.%, respectively. Anhydrite, although with a concentration of 4.22 wt.% may be responsible of the sulphates leaching from 25%w-APCr because this is the only sulphate-bearing phase in the mineral content of these LWA. Sanidine was determined to be present at a concentration as low as 0.59 wt.%. The total content of crystalline material of 25%w-APCr (30.93 wt.%) was noticeably lower than the crystalline content determined for 100%Clay (56.47 wt.%) and for 25%APCr-m (38.89 wt.%). Apart from anhydrite, none of the other phases of w-APCr (such as calcite, calcium hydroxychloride, halite and sylvite, among others) was detected in the LWA 25%w-APCr. It can be said that most of the mineral phases from w-APCr were effectively removed or transformed into the more stable phases albite and sanidine through the firing of the LWA 25%w-APCr at 1,180°C. As a consequence of these transformations, the leaching of chlorides was reduced to a negligible level in the final product (as reported in Table 6.5), while the leaching of sulphates was moderately controlled. Therefore, the formulation 25%w-APCr can be a suitable option to produce LWA with very low leaching properties.

6.2 Cold-bonded LWA

For manufacturing of LWA throughout cold bonding, APCr, lime and clay were pelletised and cured at ambient conditions, as described in Chapter 3. The effects of the APCr incorporation and the APCr/Lime ratio were evaluated on the characteristics of the final products. A preliminary analysis, presented in detail in Appendix XV, was conducted to assess the suitability of incorporating APCr-m, w-APCr, c-APCr and c-w-APCr (carbonated and washed APCr) into cold-bonded LWA. The results obtained from the preliminary experiments suggested that LWA formulations containing 60% to 70% of w-APCr or c-APCr with lime additions between 20% and 30% should be evaluated for coldbonded LWA manufacture. While APCr-m and c-w-APCr were not suitable for further evaluation of recycling into cold-bonded LWA under the assessed conditions. Based on this reasoning, only w-APCr and c-APCr were evaluated for recycling into cold-bonded LWA according to the formulation presented in Table 6.7.

Type of APCr	% APCr	% Lime	APCr/Lime	Code
	60	20	3	60w-APCr/20
	60	25	2.4	60w-APCr/25
	60	30	2	60w-APCr/30
w-APCr	70	20	3.5	70w-APCr/20
	70	25	2.8	70w-APCr/25
	70	30	2.3	70w-APCr/30
	60	20	3	60c-APCr/20
	60	25	2.4	60c-APCr/25
	60	30	2	60c-APCr/30
C-AFCI	70	20	3.5	70c-APCr/20
	70	25	2.8	70c-APCr/25
	70	30	2.3	70c-APCr/30

Table 6.7 Codification of cold-bonded LWA.

6.2.1 Visual appearance of cold-bonded LWA from w-APCr and c-APCr

Cold-bonded LWA incorporating 60% and 70% of w-APCr or c-APCr with lime additions of 20%, 25% and 30% were manufactured for subsequent analysis. The external appearance of these LWAs after 28 days of curing at ambient conditions can be observed in Figure 6.22 (cold-bonded LWA from w-APCr) and Figure 6.23 (cold-bonded LWA from c-APCr). The single pellets were about 11 – 14 mm in size, spherical in shape and with a rough surface texture. The LWA from w-APCr had a light grey colour, while the LWA from c-APCr were dark grey due to the dark colour acquired by the APCr after carbonation. The LWA incorporating 60% of c-APCr exhibited more irregular and brittle surfaces.

60% w-APCr / 20% Lime



70% w-APCr / 20% Lime

60% w-APCr / 25% Lime



70% w-APCr / 25% Lime

60% w-APCr / 30% Lime



70% w-APCr / 30% Lime





Figure 6.22 Cold-bonded LWA from w-APCr.



Figure 6.23 Cold-bonded LWA from c-APCr and digital microscopy images of a pellet surface and its cross section.

6.2.2 Technological properties of cold-bonded LWA

In this section, the effects of the APCr incorporation and lime addition were evaluated on the technological properties of cold-bonded LWA. The results were compared with the technological properties of Carbon8[®] determined in the laboratory and reported in Table 6.8.

Property	Units	Value
Oven-dried particle density ^a	g/cm ³	1.55 ± 0.01
Saturated and surface-dried particle density ^a	g/cm ³	1.93 ± 0.01
Apparent particle density ^a	g/cm ³	2.49 ± 0.02
Water absorption (at 24 hr) ^a	%	24.4 ± 0.04
Loose bulk density	g/cm ³	0.64 ± 0.01
Voids	%	58.7 ± 0.07
Crushing strength	MPa	1.56 ± 0.42

Table 6.8 Technological properties of Carbon8[®] measured in the laboratory.

^a Method of the pycnometer (BS EN 1097-6, 2022)

 \pm values indicate one standard deviation of 3 replicates for all the properties, except for crushing strength which is the result of 15 replicates.

6.2.2.1 Particle density

The cold-bonded LWA formulations were analysed for particle density by the method of the pycnometer as detailed in Section 3.7.4.2. The calculations and statistical analyses are presented in Appendix XVI.

Figure 6.24 shows the particle density values (oven-dried, saturated and surface-dried and apparent) determined for cold-bonded LWA incorporating 60% and 70% of w-APCr. It is observed that the variation in APCr and lime contents did not affect the particle density, since the values ranged from 1.49 g/cm³ for 70w-APCr/30 to 1.60 g/cm³ for 60w-APCr/20. This interval was under the specification of 2.0 g/cm³, therefore, the tested formulations can be considered as LWA. Also, the values were comparable with the oven-dried particle density of Carbon8[®]. The saturated and surface-dried particle density varied between 1.86 g/cm³ for 70w-APCr/30 to 1.93 70w-APCr/30 for 60w-APCr/20. The maximum value of this interval was equal to the saturated & surface-dried particle density

of Carbon8[®], while the other formulations exhibited values slightly lower. The apparent particle density varied between 2.34 g/cm³ for 70w-APCr/20 and 2.40 g/cm³ for 60w-APCr/20. All the LWA had an apparent particle density lower than Carbon8[®]. In general, the oven-dried particle density for the cold-bonded LWA incorporating w-APCr decreased as the percentage of w-APCr increased from 60 to 70% and the addition of lime increased from 20% to 30%.



Figure 6.24 Particle density of cold-bonded LWA incorporating w-APCr. The error bars correspond to +/- 1 Standard Deviation from the mean value.

Figure 6.25 shows the particle density values (oven-dried, saturated & surface-dried and apparent) for cold-bonded LWA incorporating 60% and 70% of c-APCr. The oven-dried particle density ranged from 1.65 g/cm³ for 70c-APCr/20 to 1.93 g/cm³ for 60c-APCr/30. All the tested formulations had oven-dried particle densities higher than the value determined for Carbon8[®] (1.55 g/cm³). Nevertheless, all of them were below the specification of 2 g/cm³, therefore, the tested formulations can be considered as LWA. The saturated and surface-dried particle density varied between 1.85 g/cm³ for 70c-APCr/20 to 2.02 g/cm³ for 60c-APCr/30. The density values of this interval were comparable with the saturated and surface-dried particle density of Carbon8[®]. The apparent particle density ranged between 2.03 g/cm³ and 70c-APCr/25 and 2.12 g/cm³ for

60c-APCr/20. This variation is low because no bloating mechanism occurs during cold bonding, compared to the hot bonding. All the LWA had an apparent particle density significantly lower than Carbon8[®]. In general, the oven-dried particle density for the cold-bonded LWA incorporating c-APCr decreased as the percentage of c-APCr increased from 60% to 70% and the addition of lime increased from 20% to 30%.



Figure 6.25 Particle density of cold-bonded LWA incorporating c-APCr. The error bars correspond to +/- 1 Standard Deviation from the mean value.

6.2.2.2 Loose bulk density and voids

The cold-bonded LWA were analysed for bulk density and voids according to the procedure described in Section 3.7.5. The calculations and statistical analysis are presented in Appendix XVII.

Figure 6.26 shows the bulk density (A) and percentage of voids (B) of the cold-bonded LWA incorporating 60% and 70% of w-APCr. The bulk density for the assessed formulations ranged from 0.81 g/cm³ for 70w-APCr/30 to 0.86 g/cm³ for 60w-APCr/20 and 60w-APCr/30. This interval comprised values about 30% higher than the bulk density determined for Carbon8[®]. However, they were under the specification of 1.2 g/cm³ for bulk density, therefore the manufactured cold-bonded formulations

incorporating w-APCr are considered LWA. The voids percentages of these LWA varied between 45.0% for 60w-APCr/25 and 46.4% for 60w-APCr/20. These values were about 20% lower than the voids percentage of Carbon8[®]. The variation in w-APCr incorporation and lime addition did not affect the bulk density and the voids percentage for cold-bonded LWA.





Figure 6.27 shows the bulk density (A) and percentage of voids (B) of the cold-bonded LWA incorporating 60% and 70% of c-APCr. The bulk density for the assessed formulations ranged from 0.86 g/cm³ for 70c-APCr/20 to 0.99 g/cm³ for 60c-APCr/25 and 60c-APCr/30. The values in this interval were on average 47% higher than the bulk density of Carbon8[®]. However, they were under the specification of 1.2 g/cm³ for bulk density. At the same c-APCr content, the bulk density values were observed at the highest incorporation of c-APCr. The voids percentages of the LWA ranged between 47.0% for 60c-APCr/20 and 49.9% for 70c-APCr/30. These values were about 18% lower than the voids percentage of Carbon8[®]. Although with only slight changes, the voids percentage increased as the c-APCr incorporation and the lime addition increased.





6.2.2.3 Water absorption

The cold-bonded LWA were analysed for water absorption by the method of the pycnometer described in Section 3.7.4.2. The calculations and statistical analysis are presented in Appendix XVI. The water absorption reflects the porosity and the degree of compaction of the LWA.

Figure 6.28 shows the water absorption at 10 min, 60 min and 24 hr of the cold-bonded LWA incorporating 60% and 70% of w-APCr. The water absorption in 24 hours of soaking ranged between 20.8% for 60w-APCr/20 and 25.0% for 70w-APCr/30. Only the formulation 70w-APCr/30 exceeded the water absorption at 24 hr determined for Carbon8® (24.4%). It is observed that most of the water absorption took place during the first 10 min of the test. During this period, the cold-bonded LWA absorbed about 90% of the total amount of water absorbed in 24hr. The water absorption at 10 min, 60 min and 24 hr for the cold-bonded LWA increased with the increase in w-APCr incorporation and with the lime addition.



Figure 6.28 Water absorption of cold-bonded LWA incorporating w-APCr. The error bars correspond to +/- 1 Standard Deviation from the mean value.

Figure 6.29 shows the water absorption at 10 min, 60 min and 24 hr of the cold-bonded LWA incorporating 60% and 70% of c-APCr. The water absorption in 24 hours of soaking ranged between 3.9% for 70c-APCr/25 and 12.0% for 70c-APCr/20. This interval was much lower than the water absorption at 24 hr determined for Carbon8[®] (24.4%). It is observed that most of the water absorption took place during the first 10 min of the soaking time. During this period, the cold-bonded LWA absorbed 60% to 80% of the total amount of water absorbed in 24hr. For both incorporations, 60% and 70% of c-APCr, adding 25% or 30% of lime significantly reduced the water absorption of the LWA to values no greater than 5.7% at 24 hr.



Figure 6.29 Water absorption of cold-bonded LWA incorporating c-APCr. The error bars correspond to +/- 1 Standard Deviation from the mean value.

6.2.2.4 Crushing strength

The cold-bonded LWA were analysed for crushing strength according to the procedure described in Section 3.7.3. The calculations and statistical analysis are shown in Appendix XVIII.

Figure 6.30 shows the crushing strength of the cold-bonded LWA incorporating 60% and 70% of w-APCr. The crushing strength ranged from 0.38 MPa for 60w-APCr/25 to 0.46

MPa for 60w-APCr/20. This interval was on average 73% lower than the crushing strength determined for Carbon8[®]. It is observed that the variation in w-APCr incorporation and the lime addition did not affect the crushing strength of the evaluated cold-bonded LWAs.



Figure 6.30 Crushing strength of cold-bonded LWA incorporating w-APCr. The error bars correspond to +/- 1 Standard Deviation from the mean value.

Figure 6.31 shows the crushing strength of the cold-bonded LWA incorporating 60% and 70% of c-APCr. Although all the formulations had crushing strengths lower than the value determined for Carbon8[®], the LWA containing 60% of c-APCr exhibited a better performance compared with those containing 70% of c-APCr. For the LWA containing 60% of c-APCr, at least the formulation 60c-APCr/30 with the highest crushing strength of the series of batches (1.41 MPa) was comparable with Carbon8[®], and it could become stronger if the error interval is considered. For both c-APCr incorporations, the crushing strength increases as the lime addition increased. By comparing these results with the results shown for LWA containing w-APCr (Figure 6.30), it can be concluded that for the development of crushing strength in cold-bonded LWA, it is more suitable to incorporate c-APCr into their structure.



Figure 6.31 Crushing strength of cold-bonded LWA incorporating c-APCr. The error bars correspond to +/- 1 Standard Deviation from the mean value.

6.2.3 Leaching properties of cold-bonded LWA

The leaching properties of cold-bonded LWA produced from w-APCr and c-APCr were evaluated. The results are shown in Table 6.9. The leaching rates determined in the laboratory for Carbon8[®] and the limit values for waste acceptance at hazardous, non-hazardous and inert landfills as expressed in the Landfill Directive (The Council of the European Union, 2003) are shown for comparison. The findings will be discussed in the following sections.

			LWA from	n w-APCr					LWA from	n c-APCr				Lim	its for WAC	
	60w-	60w-	60w-	70w-	70w-	70w-	60c-	60c-	60c-	70c-	70c-	70c-	Carbon8®	Hazardous	Non-	Inert
	APCr/20	APCr/25	APCr/30	APCr/20	APCr/25	APCr/30	APCr/20	APCr/25	APCr/30	APCr/20	APCr/25	APCr/30		1102010000	hazardous	
BS 124	457-2 LS =	10 (mg/kg)												l		
As	<1.582	<1.582	<1.582	<1.582	<1.582	<1.582	<1.582	<1.582	<1.582	<1.582	<1.582	<1.582	<1.582	25	2	1
Ba	2.8	4.2	3.4	2.8	2.7	7.6	8.6	7.8	9.3	8.5	11.0	12.0	1.8	300	100	20
Cd	< 0.071	< 0.071	< 0.071	< 0.071	< 0.071	< 0.071	< 0.071	< 0.071	< 0.071	< 0.071	< 0.071	< 0.071	< 0.071	5	1	0
Cr	2.3	1.4	1.2	1.9	1.5	1.8	1.5	1.4	1.2	1.6	1.5	1.3	0.220	70	10	1
Cu	< 0.712	< 0.712	< 0.712	< 0.712	< 0.712	< 0.712	< 0.712	< 0.712	< 0.712	< 0.712	< 0.712	< 0.712	< 0.712	100	50	2
Hg	<1.158	<1.158	<1.158	<1.158	<1.158	<1.158	<1.158	<1.158	<1.158	<1.158	<1.158	<1.158	<1.158	2	0	0
Mo	1.6	<1.522	<1.522	1.7	<1.522	<1.522	<1.522	<1.522	<1.522	<1.522	<1.522	<1.522	<1.522	30	10	1
Ni	< 0.325	< 0.325	< 0.325	< 0.325	< 0.325	< 0.325	< 0.325	< 0.325	< 0.325	< 0.325	< 0.325	< 0.325	< 0.325	40	10	0
Pb	<2.209	18.0	19.0	<2.209	21.0	33.0	<2.209	<2.209	3.5	<2.209	<2.209	<2.209	<2.209	50	10	1
Sb	< 0.858	< 0.858	< 0.858	< 0.858	< 0.858	< 0.858	< 0.858	< 0.858	< 0.858	< 0.858	< 0.858	< 0.858	1.500	5	1	0
Se	<1.008	<1.008	<1.008	<1.008	<1.008	<1.008	<1.008	<1.008	<1.008	<1.008	<1.008	<1.008	<1.008	7	1	0
Zn	<2.483	5.9	5.5	<2.483	6.5	8.1	<2.483	<2.483	<2.483	<2.483	<2.483	<2.483	<2.483	200	50	4
Cl-	24,479	23,626	22,696	27,761	25,954	26,489	37,684	67,922	50,300	57,812	45,705	40,552	9,401	25,000	15,000	800
F-	<20.3	<20.3	<20.3	<20.3	<20.3	<20.3	<20.3	29.3	25.1	28.5	26.3	<20.3	<20.3	500	150	10
SO4 ⁻²	10,451	9,389	8,917	10,628	10,011	10,599	5,888	6,199	4,060	5,552	3,747	3,357	5,612	50,000	20,000	1,000
TDS	61,600	72,100	71900	66,700	78,100	96,400	116,200	111,600	113,000	129,700	124,500	120,800	26,300	100,000	60,000	4,000
DOC	199	146	133	111	113	<100	127	<100	<100	<100	<100	<100	<100	1,000	800	500
BS 124	457-2 LS/10	1														
pH at 2	20°C															
	11.70	13.01	13.03	11.90	12.92	13.01	12.06	12.34	12.56	11.75	12.30	12.39	9.360			
Condu	ctivity at 20 ^c	°C														
	7,783	13,180	12980	8,751	13,170	15,460	15,740	16,100	17,370	17,090	17,690	17,590	3606			

Table 6.9 Results of leaching analysis (BS EN 12457-2, 2002) on cold-bonded LWA incorporating w-APCr and c-APCr and comparison with Carbon8® and with WAC.

Cells are highlighted for the detected parameters.

6.2.3.1 Leaching properties of cold-bonded LWA incorporating w-APCr

6.2.3.1.1 Leaching of heavy metals

Table 6.9 shows that the metals Ba and Cr leached out from all the cold-bonded LWA incorporating w-APCr. The leaching rates of Ba ranged between 2.7 mg/kg (70w-APCr/25) and 7.6 mg/kg (70w-APCr/30). These values were higher than the leaching of Ba from Carbon8[®] (1.8 mg/kg) and were below the inert WAC limit of 20 mg/kg. The leaching rates of Cr were under the non-hazardous WAC limit of 10 mg/kg. Other released metals were Mo, Pb and Zn, while none of them were released from Carbon8[®]. When released, Mo and Zn were below their non-hazardous WAC limits, 10 and 50 mg/kg, respectively, and Pb was below the hazardous WAC limit of 50 mg/kg.

Figure 6.32 summarises the leaching rates of heavy metals from cold-bonded LWA containing w-APCr. The leaching of Pb and Zn increased as the incorporation of w-APCr increased from 60% to 70%. At the same w-APCr incorporation, the leaching of Pb and Zn increased as the additions of lime increased from 25% to 30%, while no leaching of these metals was observed when 20% of lime was added to the LWA formulations. There are no tendencies with respect to the incorporation of w-APCr or lime additions and the leaching rates of Ba, Cr and Mo.



Figure 6.32 Leaching of heavy metals from cold-bonded LWA incorporating w-APCr.

6.2.3.1.2 Leaching of soluble salts and TDS

Table 6.9 shows that soluble chlorides and sulphates were leached out from all the coldbonded LWA made from w-APCr. The leaching of chlorides ranged between 22,696 mg/kg (60w-APCr/30) and 27,761 mg/kg (70w-APCr/20). These high leachable concentrations were not expected, because the leaching of chlorides had been reduced by the washing treatment under optimum conditions to 14,153 mg/kg in w-APCr-m (Section 5.1.1). Probably, the removal of chlorides in the batch of w-APCr produced for coldbonded LWA manufacture was not properly achieved, leaving a higher leachable fraction of these salts. This leaching interval for chlorides was much higher than the leaching of chlorides observed from Carbon8[®] (9,401 mg/kg). The LWA incorporating 60% of w-APCr had leaching rates of chlorides below the hazardous WAC limit (25,000 mg/kg), but the LWA with 70% of w-APCr exceeded this reference value. The leaching of sulphates ranged from 8,917 mg/kg (60w-APCr/30) to 10,628 mg/kg (70w-APCr/20). This interval was higher than the sulphates leaching from Carbon8[®] (5,612 mg/kg) and below the non-hazardous WAC (20,000 mg/kg). The leaching rates of TDS ranged between 61,600 mg/kg (60w-APCr/20) and 96,400 mg/kg (70w-APCr/30). This interval was significantly higher than the TDS leaching from Carbon8[®], but still lower than the hazardous WAC limit (100,000 mg/kg).

Figure 6.33 summarises the leaching rates of soluble salts (A) and TDS (B) from coldbonded LWA incorporating w-APCr. It is observed that in the evaluated formulations of LWA, the leaching of chlorides and sulphates increased as the incorporation of w-APCr increased from 60% to 70%. At the same w-APCr content, the leaching rates of chlorides and sulphates were reduced by increasing the lime addition. This tendency is particularly clear in the LWA incorporating 60% of w-APCr. Nevertheless, these reductions were not significant compared with the high leaching rates of both chlorides and sulphates. The leaching of TDS increased as the w-APCr incorporation and the lime addition increased in the cold-bonded LWA. The TDS leaching from the assessed LWA would comprise not only chlorides and sulphates, but also other substances associated with the metals Pb, Zn and the others discussed in Section 6.2.3.1.1.



Figure 6.33 Leaching of soluble salts from cold-bonded LWA incorporating w-APCr. A: Chlorides and sulphates, B: TDS.

6.2.3.1.3 pH

Table 6.9 shows that the pH of the leachates generated from cold-bonded LWA incorporating w-APCr was highly alkaline with values ranging between 11.70 (60w-APCr/20) and 13.03 (60w-APCr/30). This pH interval was higher than the pH of the leachate produced by Carbon8[®]. Figure 6.34 shows that in the evaluated LWA, the pH slightly increased as the w-APCr incorporation and the lime addition increased.



Figure 6.34 pH of leachates from cold-bonded LWA incorporating w-APCr.

6.2.3.2 Leaching properties of cold-bonded LWA incorporating c-APCr

6.2.3.2.1 Leaching of heavy metals

Table 6.9 shows that the metals Ba and Cr leached out from all the cold-bonded LWA incorporating c-APCr. Ba was released at rates between 8.5 mg/kg (70c-APCr/20) and 11.0 mg/kg (70c-APCr/25). These values were higher than the leaching of Ba from Carbon8[®] (1.8 mg/kg) and were below the inert WAC limit of 20 mg/kg. The leaching rates of Cr ranged between 1.2 mg/kg (60c-APCr/30) and 1.6 mg/kg (70c-APCr/20). This interval was higher than the leaching of Cr determined for Carbon8[®] and lower than the non-hazardous WAC limit of 10 mg/kg. None of the other metals were released from the

cold-bonded LWA containing c-APCr, except for Pb that was detected in the leachate from 60c-APCr/30 (3.5 mg/kg).

Figure 6.35 summarises the leaching rates of Ba and Cr from cold-bonded LWA containing c-APCr. The leaching of Ba increased as the incorporation of c-APCr increased from 60% to 70%. At the same c-APCr incorporation, the leaching of Ba increased as the additions of lime increased. It is observed that the leaching of Cr increased as the c-APCr incorporation increased, but it decreased with the increments in the lime addition.



Figure 6.35 Leaching of heavy metals from cold-bonded LWA incorporating c-APCr.

6.2.3.2.2 Leaching of soluble salts and TDS

Table 6.9 shows that soluble chlorides and sulphates were leached out from all the coldbonded LWA made from c-APCr. The leaching of chlorides ranged between 37,684 mg/kg (60c-APCr/20) and 67,922 mg/kg (60c-APCr/25). These leaching rates indicate a stabilisation of the chlorides through the cold bonding and dilution by the lime addition, considering that the c-APCr released 103,551 mg/kg of chlorides (Section 5.2.4.2). Nevertheless, the leaching interval for chlorides was much higher than the leaching of chlorides from Carbon8[®] (9,401 mg/kg) and exceeded by far the hazardous WAC limit of 25,000 mg/kg. The leaching of sulphates ranged from 3,357 mg/kg (70c-APCr/30) to 6,199 mg/kg (60c-APCr/25). This interval comprised the leaching rate of sulphates from Carbon8[®] (5,612 mg/kg) and it was below the non-hazardous WAC (20,000 mg/kg). Fluorides were released from the cold-bonded LWA incorporating c-APCr, except from 60c-APCr20 and 70c-APCr/30. The leaching rates of fluorides were between 25.1 mg/kg (60c-APCr/30) and 29.3 mg/kg (60c-APCr/25). These values indicate that there was no stabilisation of fluorides through the cold bonding since the c-APCr had released fluorides at a rate of 31 mg/kg. When released, fluorides were below the non-hazardous WAC limit. The leaching rates of TDS ranged between 111,600 mg/kg (60c-APCr/20) and 129,700 mg/kg (70c-APCr/20). This interval was significantly higher than the TDS leaching from Carbon8[®] (26,300 mg/kg) and exceeded the hazardous WAC limit (100,000 mg/kg).

Figure 6.36 summarises the leaching rates of soluble salts (A) and TDS (B) from coldbonded LWA incorporating c-APCr. The fluorides were excluded from Figure 6.36 A because their leaching rates were considered low in comparison with the leaching of chlorides and sulphates. The leaching of chlorides and sulphates varied randomly with respect to the c-APCr incorporation. This could be an effect of dilution caused by the addition of lime. At 70% of APCr incorporation, it is observed that the leaching of chlorides and sulphates significantly decreased as the lime addition increased. The leaching rates of sulphates were comparable to the leaching of these salts from Carbon8[®]. A general tendency, particularly clear in the LWA incorporating 70% of c-APCr indicates that the leaching of TDS was reduced by increasing the lime addition.



Figure 6.36 Leaching of soluble salts and TDS from cold-bonded LWA incorporating c-APCr.

6.2.3.2.3 pH

Table 6.9 shows that the pH of the leachates generated from cold-bonded LWA incorporating c-APCr was highly alkaline with values ranging from 11.75 (70c-APCr/20) to 12.56 (60c-APCr/30). This pH interval was higher than the pH of the leachate produced by Carbon8[®] (9.36). Figure 6.37 shows that in the cold-bonded LWA, the pH slightly increased as the c-APCr incorporation and the lime addition increased. This behaviour was expected given the use of lime as the binder.



Figure 6.37 pH of leachates from cold-bonded LWA incorporating c-APCr.

6.2.4 Mineralogy of cold-bonded LWA

Samples of 60w-APCr/30 and 60c-APCr/30 LWA were analysed for mineralogy. These formulations were selected because they exhibited the optimum technological characteristics and more favourable leaching properties. Figure 6.38 presents the XRD patterns with markers at the main identification peaks for the mineral phases detected in the mentioned LWA. Table 6.10 presents the results of the quantitative analysis for these LWA formulations.



Figure 6.38 XRD patterns showing the identification of the main peaks for cold-bonded LWA: 60c-APCr/30 and 60w-APCr/30.

		wt. %			
Mineral phase	Chemical formula	60w-APCr/30	60c-APCr/30		
Albite	NaAlSi ₃ O ₈	-	5.16 ± 1.17		
Anhydrite	CaSO ₄	0.72 ± 0.06	-		
Calcite	CaCO ₃	31.04 ± 0.26	21.68 ± 0.67		
Calcium carbide	CaC_2	2.68 ± 0.13	-		
Cotunnite	PbCl ₂	0.25 ± 0.02	-		
Ettringite	$Ca_6Al_2(SO_4)_3(OH)_{12}\bullet 26H_2O$	0.95 ± 0.07	3.32 ± 0.27		
Gypsum	CaSO ₄ •2H ₂ O	4.52 ± 0.22	4.28 ± 0.45		
Quartz	SiO ₂	4.13 ± 0.12	5.75 ± 0.31		
Sanidine	KAlSi ₃ O ₈	-	5.46 ± 0.60		
Wollastonite	Vollastonite CaSiO ₃		1.92 ± 0.44		
	% Crystalline material	44.29 ± 0.39	47.57 ± 2.12		
% Amorphous material		55.71 ± 0.39	52.43 ± 2.12		

Table 6.10 Quantification of mineral phases and estimation of amorphous material content by Rietveld analysis for cold-bonded LWA: 60w-APCr/30 and 60c-APCr/30.

 \pm values indicate one standard deviation

6.2.4.1 Mineralogy of 60w-APCr/30

Figure 6.38 shows that calcite was the main phase identified in 60w-APCr/30, showing peaks of high intensity. The high-intensity peaks for calcite were observed at 29.36 °20, 23.03 °20, 35.94 °20 and 43.12 °20. The presence of calcite was due to the high content of this phase in the w-APCr (as reported in Figure 5.6) added to the calcite resulting from the hydration of lime during the pelletising process and further carbonation of portlandite during the curing of the pellets. Quartz was also identified by high-intensity peaks at 26.60 °20 and 20.84 °20. The presence of quartz could be associated with the contribution from the 10% of clay that was incorporated in the formulation of 60w-APCr-30 and a minimum contribution from the w-APCr. Anhydrite and gypsum were identified, although with low-intensity peaks. Gypsum may be the result of the hydration of anhydrite that was originally in w-APCr. The other phases identified were ettringite, calcium carbide and cotunnite. These phases may have been produced during the curing of the pellets.

Table 6.10 shows that calcite was the most abundant mineral phase in 60w-APCr/30, representing 31.04 wt.% of the total mineral content of this LWA. Gypsum, anhydrite, and ettringite, although present at low concentrations, would be responsible for the sulphates leaching from 60w-APCr/30. However, the mineralogy of these LWA does not explain the leaching of chlorides (reported in Figure 6.33), because the only chloride detected was cotunnite at a concentration as low as 0.25 wt.%. Therefore, is required further research to find the origin and to control the leaching of chlorides and sulphates.

6.2.4.2 Mineralogy of 60c-APCr/30

Figure 6.38 shows that calcite was the main phase identified in 60c-APCr/30, with peaks distributed from 23 °20 to 65 °20. The peaks with higher intensities for calcite were observed at 29.53 °20, 23.16 °20, 36.13 °20, 39.59 °20 and 43.35 °20. The presence of calcite was due to the high content of this phase in c-APCr (as reported in Figure 5.19) besides the calcite formed from the carbonation of portlandite during the curing of the pellets, which in turn was the product of the hydration of lime during the pelletising process. Quartz was identified by peaks of moderate intensity distributed from 20 °20 to 68 °20. The peaks with greater intensity for quartz are observed at 26.79 °20 and 20.93 °20. The content of quartz in 60c-APCr/30 came from the c-APCr as well as from the 10% of clay that complemented the formulation of this LWA. Gypsum was identified, probably as the product of the hydration of anhydrite that was originally present in c-APCr.

Table 6.10 shows that the most abundant mineral phases in 60-cAPCr/30, was calcite with a concentration as high as 21.68 wt.% of the total mineral content of this LWA. Gypsum, and ettringite would be responsible for the sulphates leaching from 60c-APCr/30. The high leaching of chlorides (reported in Figure 6.36), could not be explained by the mineralogy of these LWA, because no chloride-based mineral phases were detected at a quantifiable amount. Therefore, is required further research to find the origin and to control the leaching of chlorides.

6.3 Summary

To investigate the suitability of producing LWA from APCr, two different routes of LWA manufacturing were tested, hot bonding and cold bonding. Several formulations obtained from these manufacturing processes were characterised for microstructure, technological properties, mineralogy and leaching behaviour. The effects of different factors typical of each manufacturing route were assessed on the LWA characteristics. Figure 6.39 shows a schematic summary and comparison of the LWA production at optimised conditions by the hot and cold bonding routes and the characterisation of the best final products, 25%w-APCr (hot-bonded LWA incorporating 25% of w-APCr) and 60c-APCr/30 (cold-bonded LWA incorporating 60% of c-APCr with 30% of lime binder).

The leaching properties of 25%w-APCr were satisfactory, with the leaching of sulphates and TDS below the non-hazardous WAC and Ba, Cr and chlorides below the inert WAC. The formulation 60c-APCr/30 showed high leaching rates of chlorides exceeding the hazardous WAC, while the releasing of Pb and sulphates were below the non-hazardous WAC and Ba, Cr and fluorides were below the inert WAC. The releasing of chlorides from 60c-APCr/30 could not be explained by the mineralogy of this formulation since no chloride-based mineral phases were detected. The releasing of sulphates was mainly associated to the presence of anhydrite in 25%w-APCr and gypsum in 60c-APCr/30. Both 25%w-APCr and 60c-APCr/30 met the requirements to be considered LWA as per their oven-dried particle density (maximum 2.0 g/cm³) and loose bulk density (maximum 1.2 g/ cm³). In both formulations, there was a great reduction in water absorption at 24 hours compared to commercial brands Lytag[®] and Carbon8[®]. The crushing strength of the hot and cold bonded LWA were comparable with the values determined for Lytag[®] and Carbon8[®], respectively; but the crushing strength of 25%w-APCr was significantly higher than that of 60c-APCr/30.

It was concluded that the hot-bonded LWA 25%w-APCr showed higher strength, better environmental properties, and their manufacture required a moderate length of time of maximum two days. In comparison, the cold-bonded LWA 60c-APCr/30 had lower strength, high leaching rates of chlorides, and required a curing period of 28 days. However, the second formulation allowed a greater incorporation of APCr. Therefore, the performance in concrete of both formulations should be further investigated to determine their possible applications. In addition, the production of these LWA should be evaluated in terms of energy consumption, carbon footprint and economic viability of a future scaleup and commercialisation.



Leaching properties

		Chlorides and TDS
	Hazardous WAC	
	Non-hazardous WAC	
Sulphates and TDS		Pb and sulphates
	Inert WAC	
Ba, Cr and chlorides		Ba, Cr and fluorides

Mineralogy						
 Stable quartz and albite were the most abundant mineral phases. Anhydrite, although with a lower concentration may be responsible of the sulphates leaching. 	 Stable calcite resulting from the c-APCr was the most abundant mineral phase. Gypsum and ettringite would be responsible of the sulphates leaching. No chloride-based mineral phases were detected at a quantifiable amount. 					

25% w-APCr	Lytag®	Technological properties	Carbon8®	60% c-APCr/30
1.89 ± 0.01	1.46 ± 0.01	Oven-dried particle density (g/cm ³)	1.55 ± 0.01	1.93 ± 0.01
1.10 ± 0.01	0.84 ± 0.01	Loose bulk density (g/cm ³)	0.64 ± 0.01	0.99 ± 0.00
4.40 ± 0.11	11.24 ± 0.21	Water absorption at 24 hr (%)	24.4 ± 0.04	4.40 ± 0.10
3.76 ± 1.00	3.23 ± 1.03	Crushing strength (MPa)	1.56 ± 0.42	1.41 ± 0.42

Figure 6.39 Schematic summary of the production and characterisation of APCr-based LWA through hot bonding and cold bonding.

7 Chapter 7 Concrete testing and estimation of costs of LWA manufacture

In this chapter, the hot-bonded and cold-bonded LWA with optimal technological properties were incorporated into different formulations of concrete. The concrete cubes were tested for compressive strength and density after 28 days of curing. In addition, an estimation of the embodied carbon associated to the industrial production of LWAC was conducted, based on the materials and procedures analysed in this study.

7.1 Compressive strength and density of LWAC

The calculations of compressive strength and density for the LWAC cubes after 28 days of curing are detailed in Appendix XIX.

Figure 7.1 shows the compressive strength and density values of LWAC made of hotbonded LWA fired at 1,180°C. The results for LWAC formulated from Lytag[®] are shown for comparison. The LWAC cubes made of 25%w-APCr had a compressive strength of 48.3 MPa, which was 25% higher than the cubes made of Lytag[®] and 33% higher than the cubes using 100%Clay LWA. The LWAC cubes incorporating 30%w-APCr in their structure exhibited a compressive strength of 30.6 MPa, that was 21% lower than the cubes made of Lytag[®] and 15% lower than the cubes made of 100%Clay. These results were in agreement with the high crushing strength of the LWA 25%w-APCr and the low crushing strength of 30%w-APCr, as discussed in Section 6.1.2.5. The LWAC cubes formulated from LWA containing w-APCr were denser than the LWAC cubes made of 100%Clay LWA and those made of Lytag[®]. According to (BS EN 206, 2013+A2, 2021), the compressive strength class for LWAC incorporating 25%w-APCr is LC40/44, while the LWAC incorporating 30%w-APCr is LC30/33. This classification determines the possible applications of the LWAC.



Figure 7.1 Compressive strength and density of LWAC formulated from hot-bonded LWA. The error bars correspond to +/- 1 Standard Deviation from the mean value.

Figure 7.2 shows the compressive strength and density values of LWAC cubes made from cold-bonded LWA. The results were compared with LWAC formulated from Carbon8[®]. All the LWAC cubes made of cold-bonded LWA based on w-APCr or c-APCr had compressive strength values of on average 19% lower than the LWAC made of Carbon8[®]. It is observed that the LWA incorporating 60% of w-APCr or c-APCr produced LWAC cubes with higher compressive strength (no less than 20 MPa). These results were in agreement with the tendencies of the crushing strengths of the cold-bonded LWA, as discussed in Section 6.2.2.4. On the other hand, all the LWAC cubes formulated from cold-bonded LWA containing w-APCr or c-APCr were denser than the LWAC cubes made from Carbon8[®].



Figure 7.2 Compressive strength and density of LWAC formulated from cold-bonded LWA. The error bars correspond to +/- 1 Standard Deviation from the mean value.

The failures caused by the compressive strength testing on the LWAC cubes made of hotbonded LWA and cold-bonded LWA are shown in Figure 7.3 and Figure 7.4, respectively.

The cubes exhibited a satisfactory fracture shape, according to (BS EN 12390-3, 2019). All four exposed faces were fractured in a similar manner and minimal damage was observed on the faces in contact with the compression loads. 25% w-APCr

30% w-APCr











Figure 7.3 LWAC cubes made of hot-bonded LWA after 28 days of casting, showing the failures caused by the compressive strength test and examples of patterns of satisfactory and unsatisfactory failures (BS EN 12390-3, 2019).

60w-APCr/30

70w-APCr/30



60c-APCr/30



70c-APCr/30



Carbon8®



Figure 7.4 LWAC cubes made of cold-bonded LWA after 28 days of casting, showing the failures caused by the compressive strength test.

Table 7.1 presents the classification of the different LWAC formulations based on their densities and compressive strengths according to (BS EN 206, 2013+A2, 2021). The density of LWAC depends on the type and grading of the LWA, and the mix proportions (The Concrete Society, 2022). According to their density values, the evaluated LWAC formulations incorporating either hot-bonded or cold-bonded LWA can be classified as structural lightweight concrete. In addition, Table 7.1 shows the possible applications of the LWAC formulations according to their exposure classification based on compressive strength (BS 8500-1:2015+A2, 2019). The exposure classes suggest the intensity of the environmental and/or chemical conditions to which the concrete can be subjected. The LWAC formulations made of 25%w-APCr and the one made of 30%w-APCr LWA were suitable for applications in which concrete with reinforcement is exposed to air or moisture (XC). The LWAC made of 25%w-APCr LWA was also suitable for applications where concrete with reinforcement can be in direct contact with water containing chlorides, except seawater (XD), and seawater or airborne salt from seawater (XS). The LWAC formulations made of cold-bonded LWA were suitable for applications where concrete without reinforcement can be used in environments free of corrosion or chemical attack (X0).
Table 7.1 LWAC classification according to density and compressive strength (BS EN 206, 2013+A2, 2021) and possible applications according to exposure class (BS 8500-1:2015+A2:2019.

			LWA inco	rporation		
	25%w-	30%w-APCr	60w-	70w-	60c-	70c-
	APCr	50700 711 01	APCr/30	APCr/30	APCr/30	APCr/30
Density class	D2,0	D2,0	D2,0	D2,0	D2,0	D2,0
Compressive strength class	LC40/44 LC25/28 LC16/18 LC16		LC16/18	LC16/18	LC16/18	
	XC3/4					
Exposure class	XD1		X0			
for at least 50	XD2	$\mathbf{V}\mathbf{C}2\mathbf{V}\mathbf{C}2/\mathbf{A}$		\mathbf{v}_0	\mathbf{v}_0	\mathbf{v}_0
compressive	XD3	AC2 AC5/4		$\Lambda 0$	$\Lambda 0$	Λ0
strength class	th class XS1					
	XS2					
	XC3/4					
Exposure class for at least 100	XD1					
years, as per	XD2	XC2 XC3/4	X0	X0	X0	X0
compressive strength class	XD3					
	XS1					

Exposure class	Description	Examples					
X0	All exposures except freeze/thaw, abrasion or chemical attack	Concrete without reinforcement or embedded metal. Concrete inside buildings with very low air humidity.					
XC2	Wet, rarely dry	Concrete surfaces exposed to long-term water contact. Foundations.					
XC3/4	Moderate humidity	Concrete inside buildings with moderate to high air humidity, External concrete sheltered from rain.					
	Cyclic wet and dry	Concrete surfaces exposed to water contact, not within CX2					
XD1	Moderate humidity	Concrete surfaces exposed to airborne chlorides					
XD2	Wet, rarely dry	Swimming pools, concrete exposed to industrial waters containing chlorides.					
XD3	Cyclic wet and dry	Parts of bridges exposed to spray containing chlorides, pavements, car park slabs.					
XS1	Exposed to airborne salt but not in direct contact with sea water	Structures near to or on the coast.					
XS2	Surfaces completely submerged and remaining saturated	Concrete surfaces below mid-tide level.					

7.2 Carbon footprint of LWAC

Life Cycle Assessment (LCA) is an account of all resources and materials consumption and emissions related to a specific product, considering all upstream and downstream processes (Astrup, 2008). In an LCA, the environmental impact of a product is evaluated according to different categories that can be classified into two groups (BS EN ISO 14040:2006+A1, 2020):

- Non-toxicity categories: include climate change, acidification, nutrient enrichment, photochemical ozone formation and stratospheric ozone depletion.
- Toxicity categories: ecotoxicity (in water and soil) and human toxicity (through air, water and soil).

Carbon footprint of a product (CFP), as defined in (BS EN ISO 14067, 2018), is the sum of greenhouse gas (GHG) emissions and removals in a product system. The CFP is expressed in CO₂ equivalents and is based on an LCA evaluating only the impact category of climate change. In the construction industry, carbon emissions have two sources, operational emissions and embodied emissions (RICS, 2017). The operational emissions arise from the energy consumption in the daily running of built assets. The embodied emissions, also called embodied carbon, come from the production, acquisition and installation of materials and components that constitute a built structure. According to the United Nations' 2022 Global Status Report (United Nations Environment Programme, 2022), the building and construction sector contributes 37% of all carbon emissions worldwide. This report stresses that most of this contribution, 28%, comes from operational emissions and the other 9% is from embodied carbon. However, a previous report from the same organisation (United Nations Environment Programme, 2022), revealed that more than 50% of the construction industry professionals admitted that they do not estimate embodied carbon in their projects. Consequently, the construction sector has so far prioritised the reduction of operational emissions but has only partially addressed the embodied carbon.

If the manufacture of construction materials, such as LWA, is adopted as an option for APCr recycling, the embodied carbon associated with the whole process of LWAC

production can be estimated. The aspects addressed in this context include (Astrup, 2008; BS EN ISO 14040:2006+A1, 2020):

- Treatment: APCr must always be stabilised or treated to reduce the leaching of potential contaminants.
- Transportation: APCr are carried from the producers EfW to Augean facilities.
- Energy: Drying of materials and comparative analysis between the high energy consumption of hot-bonding and cold-bonding for LWA manufacture.

Table 7.2 presents a framework designed for the evaluation of the embodied carbon associated with the production of LWAC incorporating w-APCr or c-APCr, based on an LCA framework. The functional unit was arbitrarily set as 1 tonne (1,000 kg) of LWAC, considering that this should be the minimum amount of material expected to be produced in a certain time at an industrial scale. For Scenario 1, the incorporation of LWA 25%w-APCr fired at 1,180°C was selected because it exhibited the optimum technological properties, more favourable leaching characteristics and highest strength in the concrete testing. Likewise, for Scenario 2, the formulation evaluated was 60c-APCr/30. The assumptions were associated mainly with the fact that the industrial scale processes were estimated from laboratory scale set-ups. The handling of the wastewater resulting from APCr washing was avoided because the treatment and disposal of this effluent was out of the extent of the present work. Nevertheless, some options for treatment and recycling of this wastewater were discussed in Section 5.1.3. The data quality refers to the statistical analysis detailed in Appendixes X to XVII, which supports the data originated in the characterisation of the raw materials for LWA manufacture. The environmental impact is focused only on the category of global warming; in consequence, the environmental assessment is limited to the estimation of embodied carbon.

	LCA stages		Scenario 1	Scenario 2			
	Functional unit		Production of 1 tonne of LWAC	Production of 1 tonne of LWAC			
			Clay extraction, clay grinding, APCr washing, APCr drying,	Clay extraction, clay grinding, APCr accelerated carbonation,			
			mixing/pelletising, green pellets drying, sintering, cooling,	APCr drying, mixing/pelletising, green pellets drying, LWA			
1.Goal and	System boundaries		packing, sand acquisition, cement acquisition, water	curing, cooling, packing, sand acquisition, cement acquisition,			
scope	scope		acquisition, concrete mixing, concrete curing, transport of	water acquisition, concrete mixing, concrete curing, transport			
			raw APCr	of raw APCr			
	Time horizon		100 years	100 years			
	Indirect or avoided	processes	Handling of wastewater	Handling of wastewater			
	Data collection		Sections: 5.1 (APCr washing), 6.1 (hot-bonded LWA	Sections: 5.1 (APCr carbonation) and 6.2 (cold-bonded LWA			
	Data conection		production) and 7.1 (concrete testing)	production) and 7.1 (concrete testing)			
2 Inventory	Innuta		Materials: clay, APCr, water, sand, cement	Materials: clay, APCr, lime, water, sand, cement			
2. Inventory	mputs		Resources: Energy	Resources: Energy			
anarysis	Outputs		LWAC	LWAC			
	Data quality		Precision of data (reproducibility and uncertainty),	Precision of data (reproducibility and uncertainty),			
	Data quality		representativeness, consistency of results	representativeness, consistency of results			
	Impact categories	Global	Emissions to air	Emissions to air			
3.Impact	to evaluate warming						
assessment	ssessment Emiss		CO ₂ emissions	CO ₂ emissions			
		to air	Regulation for CO ₂ emissions	Regulation for CO ₂ emissions			
4.Interpretation of results			Contrast between environmental advantages and disadvantages of using hot-bonded or cold-bonded LWA for LWAC				

Table 7.2 Framework for estimation of embodied carbon in the manufacture of LWAC incorporating APCr.

In LWA manufacturing, the processes of major energy consumption are pelletising, pellet drying and pellet firing. The energy inputs of these processes were calculated and transformed into the corresponding carbon emissions expressed as kg CO₂/kg material. The conversion of energy into released carbon was done based on the carbon emissions generated by the UK power stations per each kWh generated, according to the Greenhouse gas reporting: conversion factors (Department for Energy Security and Net Zero and Department for Business, Energy and Industrial Strategy, 2022). The conversion factor is 0.28307 kg of CO₂ generated from each kWh consumed. This factor also includes the gases methane CH₄ and nitrous oxide N₂O, that are converted into their carbon dioxide equivalents. That is, the factor denotes kg CO₂ eq. per kWh. The estimation of embodied carbon for pelletising, pellet drying, and pellet firing are presented in Table 7.3.

For pelletising, the energy consumption of a ring die pellet machine was considered (Vukimirovic, et al., 2010).

For pellet drying, the latent heat of water evaporation was considered as the energy consumption in a convection oven at 40°C for drying of cold-bonded LWA and at 100°C for drying of hot-bonded LWA. The latent heat of evaporation is the energy that must be supplied to turn 1 kg of moisture into vapour (kJ/kg water evaporated) (Kemp, 2012). Assuming an efficiency of at least 90% in the drying process, the amount of water expected to be removed was calculated from the water added to each LWA as per formulations detailed in Table 3.10 and Table 3.13. The water loss due to friction during pelletising was considered negligible.

For pellet firing, the equipment most commonly used in LWA manufacture is the rotary kiln. This is usually a direct-fired kiln, in which the raw materials and the combustion products are in direct contact with each other (Ebben & Carlson, 2023). Thus, the kiln contents constitute a chemical system of combustion. The energy requirement of the kiln is the enthalpy change of the combustion reaction that was calculated for the system according to Equation 7.1.

Equation 7.1

$$Q = mC\Delta T$$

Where:

Q is the energy required for the temperature rise in the kiln in J

m is the mass of the kiln contents in kg

C is the specific heat capacity of the kiln contents in kJ/kg•°C. The specific heat capacity is the heat required to raise the temperature of 1 kg of a substance by one degree. For w-APCr, it was used the specific heat capacity of fly ash (Bentz, et al., 2011). For the mix 75%Clay/25%w-APCr, the specific heat capacity was calculated according to the weight fraction of each component.

 ΔT is the temperature change: 1,180°C (firing temperature) – 20°C (initial temperature)

Table 7.3 Calculation of emissions from the processes of pelletising, drying and sintering of LWA.

Energy consumption (kWh/ton)	Reference	Energy consumption (kWh/kg)	Embodied carbon (kg CO ₂ /kg material)
42.5	(Vukimirovic, et al., 2010)	0.04250	0.01203

Calculation of embodied carbon from pelletising 1 kg of green pellets

Calculation of embodied carbon from drying 1 kg of green pellets

	Latent heat of evaporation (Energy consumption) (kJ/kg water)	Reference	Mass of water evaporated (kg)	Energy consumption (J)	Energy consumption (kWh)	Embo (kg CO	died carbon 2/kg material)
At 40°C	2,400	(Kemp, 2012)	0.378	907.20	0.00025	0.00025 0.00	
At 100°C	At 100°C 2,256		0.387	873.07	0.00024	0	.00007
Calculation of e	embodied carbon from si	ntering					
Raw mate	erial Mass m (kg)	Specific heat capacity C (J/kg•°C)	Reference	ΔΤ	Q (J)	Q (kWh)	Embodied carbon (kg CO ₂ /kg material)
Clay	1	878	(Evans, P., 2016)	1,160	1,018,480	0.28291	0.08008
w-APCr	1	0.73 ^a	(Bentz, et al., 2011)	1,160	846.8	0.00024	0.00007
75%Clay/25%w	v-APCr 1	659	Calculated in this study	1,160	764,072	0.21224	0.06008

^a Specific heat capacity of Fly ash

Table 7.4 presents the collected data and the calculations of the embodied carbon of LWA, for the cradle-to-gate-stages (A1, A2 and A3) of a whole-life carbon assessment of LWAC production, according to (EN 15978, 2011). The embodied carbon from APCr due to extraction and supply (A1) was 0 for both LWA 100%Clay and 25%w-APCr, because it is a residue intended for recycling. The carbon emissions due to extraction and supply of clay, lime and water were taken from the from ICE Database Version 3.0. launched in 2019 (Circular Ecology, 2023). For LWA 25%w- APCr, the emissions from the water supply were calculated from the water consumption due to pelletising and the APCr washing. The pelletising required 0.43 l of water per kg of raw materials mix. The APCr washing required 0.787 l of water per 0.25 kg w-APCr, considering the optimum L/S ratio of 2.5 (Section 5.1.1) and the mass loss of APCr due to washing of 20.6% (Section 5.1.2.1). For LWA 60c-APCr/30, the emissions from APCr were negative and corresponded to 60% of the maximum carbonation potential of 13% as determined in Section 5.2.1. The emissions from clay, lime and water were estimated based on the weight fraction of each substance in the LWA formulation (Table 3.10 and Table 3.13.).

For the estimation of the carbon emissions due to the transport of raw materials (A2), it was assumed that the manufacturing plant would be located in the Augean's facilities at Peterborough. The distance between any EfW site producing APCr and Augean's plant was assumed at 300 km that is the national average distance (RICS, 2017). The distance between the origin place of the clay used for this study, King's Cliffe, and Augean's facilities at Peterborough was estimated to be around 8 km. The carbon emissions associated with both distances were calculated based on the EU+UK baseline data provided by the Implementing Decision (EU)2021/781 (European Commission, Directorate-General for Climate Action, 2021). That document shows the average specific CO₂ emissions of all new heavy-duty vehicles (lorries) registered from 1 July 2019 to 30 June 2020. The general average was calculated as 50.77 g CO₂/tonne•km. The contribution from the transport of each raw material was calculated according to their weight fraction in the LWA formulation.

The emissions due to manufacturing and fabrication of LWA (A3) are the emissions determined in Table 7.3 for each process. The total embodied carbon generated from manufacturing of each LWA formulations was calculated as the sum of the emissions due to stages A1, A2 and A3.

Table 7.4 Calculation of embodied carbon in the production of 1 kg of LWA.

Information for cradle-to-gate stages of a whole life carbon assessment for LWAC production

	-	-									
	A1				A	A3					
	Raw material extraction and supply ^a Transport to n					ort to manufacturing plant ^b Manufacturing and fabrication ^c					cation ^c
Source of emission	APCr	Clay	Lime	Water	APCr: EfW plant to Augean facilities: kg CO ₂ /kg•300km	Clay: King's Cliffe to Augean facilities: kg CO ₂ /kg•8 km	Pelletising	Drying at 40°C	Drying at 100°C	Clay firing	75%Clay/25%w-APCr firing
Embodied carbon (kg CO ₂ /kg material)	0	0.255	0.78	0.00034	0.015231	0.00041	0.01203	0.00007	0.00007	0.08008	0.06008

a Data for clay, lime and water were taken from ICE Database Version 3.0.

b Data calculated from baseline data provided by the Commission Implementing Decision (EU)2021/781

c Data taken from calculations in Table 7.3

Calculation of embodied carbor	(kg CO ₂ /kg	material) on LWA	over the cradle to	gate stages
culculation of emoculea earoor	(ng coging	material) on E	over the endance to	Sale stages

	A1				A2		A3					
	Raw m	aterial extr	action an	d supply	Transport to manufacturing plant			Total embodied				
LWA formulation	APCr	Clay	Lime	Water	APCr: EfW plant to Augean facilities: kg CO ₂ /kg•300km	Clay: King's Cliffe to Augean facilities: kg CO ₂ /kg•8 km	Pelletising	Drying at 40°C	Drying at 100°C	Clay firing	75%Clay/ 25%w-APCr firing	carbon (kg CO ₂ /kg material)
100%Clay	-	0.255	-	0.00010	-	0.00041	0.01203	-	0.00007	0.08008	-	0.34769
25%w-APCr	-	0.19125	-	0.00042	0.00381	0.00030	0.01203	-	0.00007	-	0.06008	0.26796
60c-APCr/30	-0.078	0.0255	0.234	0.00014	0.00914	0.00004	0.01203	0.00007	-	-	-	0.20293

^d The embodied carbon for expanded clay LWA in ICE Database (2019) Version 3.0. was 0.39321 kg CO₂/kg LWA

The calculations of embodied carbon for LWAC manufacturing for Scenario 1 and Scenario 2 are shown in Table 7.5. The contribution from each raw material was estimated based on their weight fraction in the concrete mix determined by the absolute volume method (Appendix V - Formulation of LWAC by the absolute volume method). The emissions associated with the use of LWA 100%Clay and Lytag[®] were also calculated for comparison with Scenario 1. It was not possible to calculate the embodied carbon for LWAC made of Carbon8[®] due to the lack of information about this product. It is known that this LWA is a carbon-negative material (Carbon8, 2022), but specific data was not available.

The values in Table 7.5 show that the greater contributions to the carbon emissions in the production of LWAC came from the cement and the LWA. Also, the hot-bonded LWA had a greater contribution to these emissions than the cold-bonded LWA. This is because of the higher energy consumption during the manufacturing of hot-bonded LWA and the carbon-negative extraction and supply of APC for cold-bonded LWA.

Raw material	Embodied carbon (kg CO ₂ /kg material)	Reference	Scenario 1 LWAC from 25%w-APCr	Scenario 2 LWAC from 60c-APCr/30	LWAC from 100% Clay	LWAC from Lytag [®]
CEM I OPC	0.912 ICE Database Version 3.0.		0.2098	0.2098	0.2098	0.2098
Sand	0.007	ICE Database Version 3.0.	0.0013	0.00119	0.00112	0.0011
Water	0.00034	ICE Database Version 3.0.	0.00004	0.00004	0.00004	0.00004
LWA						
25%w-APCr	0.26796	Calculated in this study	0.12594	-	-	-
60c-APCr/30	0.20293	Calculated in this study	-	0.09740	-	-
100%Clay	0.34769	Calculated in this study	-	-	0.17037	-
Lytag [®]	0.249	(Aggregate Industries, 2022)	-	-	-	0.1245
	Total embe	odied carbon (kg CO ₂ /kg concrete)	0.3370	0.3084	0.3813	0.3353
	Total embodie	ed carbon (kg CO ₂ /tonne concrete)	337.0	308.4	381.3	335.3

Embodied carbon (kg CO₂/kg material) from LWAC

Table 7.5 Calculation of embodied carbon of LWAC incorporating APCr.

Figure 7.5 summarises the values of embodied carbon determined for the production of 1 tonne of LWAC with incorporation of LWA containing APCr. It is clear that this incorporation reduced the carbon emissions compared with LWAC made of LWA 100%Clay. For Scenario 1, the production of LWAC from hot-bonded LWA 25%w-APCr exhibited a carbon emission 12% lower than the production of LWAC from LWAC from LWA 100%Clay and comparable to the production of LWAC from Lytag[®]. Scenario 2 exhibited the greatest reductions in embodied carbon for LWAC manufacturing. The use of cold-bonded LWA 60c-APCr/30 generated carbon emissions 19% lower than the use of LWA 100%Clay, 8% lower than the use of Lytag[®] and almost 9% lower than the incorporation of 25%w-APCr. It can be concluded that the carbon emissions for both scenarios were satisfactory and can help to reduce the total carbon footprint associated to a whole-life carbon assessment for LWAC production.



Figure 7.5 Embodied carbon of LWAC made of LWA incorporating APCr.

7.3 Estimation of cost of LWA manufacture

An estimation of the costs of producing LWA from treated APCr was conducted, considering the 3 main stages of the production process: raw materials supply, transport and manufacturing. It should be noted that this assessment is only an indicative estimation of costs aimed at highlighting the economic benefit of diverting APCr from landfill to

recycling. Aspects such as packaging, storage, quality control and distribution of the final product are not integrated to this cost analysis. In addition, the cost of carbonation of APCr was not estimated. In a potential scale-up of that treatment, it is expected to incorporate CO_2 generated as waste from other industrial processes to keep the circular economy approach. Thus, at the extent of this research, the cost of acquiring and incorporating CO_2 into the LWA production is unknown.

Table 7.6 shows the costs associated with the 3 stages of the LWA production and the estimation of the costs of producing 1 tonne of each formulation 100%Clay, 25%w-APCr and 60c-APCr/30. The cost of raw APCr was set at 0 because this is the waste material intended for recycling. For the other raw materials supply (clay, lime and water), their current commercial prices for 1 tonne of each material were considered (Statista, 2023; Lincolnshire Lime, 2023; AquaSwitch, 2023a). The costs of transport of materials were calculated from a haulage estimation (Returnloads, 2023) and considering the distances as defined in Section 7.2. and Table 7.4. The cost of water included the water used for APCr treatment and the water required for pelletising. The cost of each manufacturing stage (pelletising, drying and firing) was calculated from the energy consumption in kWh reported in Table 7.3 and the average business electricity rate, which corresponds to 0.2524 £/kWh in 2023 for a company consuming more than 100,000 kWh (AquaSwitch, 2023b). For the estimation of the costs of producing 1 tonne of LWA, the contribution from each material was calculated according to their weight fraction in each LWA formulation. The same approach was applied to calculate the costs of materials transport. The total investment in manufacturing each LWA formulation was calculated as the sum of the costs due to the 3 production stages.

It can be seen in Table 7.6 that the cost of producing 1 tonne of hot-bonded LWA was reduced from 98 \pounds /tonne when using only clay as raw material (LWA 100%Clay) to 83 \pounds /tonne when w-APCr was incorporated into the formulation (LWA 25%w-APCr). This reduction, equivalent to 15%, was mainly due to the decline in energy consumption associated with the replacement of clay by w-APCr. On the other hand, the production of 1 tonne of cold-bonded LWA (60c-APCr/30) showed the highest cost (134 \pounds /ton) due to the requirement of lime as raw material which represented almost 83% of the total investment.

Table 7.0 Estimation of costs of manufacturing 1 tonne of EWA	Table 7	7.6	Estimation	of cost	s of ma	anufacti	uring 1	tonne	of LWA
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	Costs of materials, transport and manufacturing of LWA											
		Raw mat	erials supp	ly	Transport to manu	Manufacturing ^e				ng ^e		
	APCr	Clay ^a	Lime ^b	Water ^c	APCr: EfW plant to Augean facilities: (300 km)	Clay: King's Cliffe to Augean facilities: (8 km)	Pelletising	Drying at 40°C	Drying at 100°C	Clay firing	75%Clay/25%APCr firing	
Cost per tonne (£)	0	15	375	1.781	15.6	0.4	10.73	0.06	0.06	71.41	53.57	

^a Average price of clay used in construction was about 17 UDS (15 £) per tonne in 2022 (Statista, 2023).

^b Commercial price of lime for application in construction works (Lincolnshire Lime, 2023).

^c Regional commercial water rate for Augean's Peterborough facility, period 2023-2024 (AquaSwitch, 2023a)

^d Price per mile (1.6 km) is £1.50 per tonne of material when using a truck with capacity of 7.5 tonnes (Returnloads, 2023)

^e The costs were calculated from the kWh consumed by each process (Table 7.3) and the average business electricity rate, which is 0.2524 £/kWh for annual consumption greater than 100,000 kWh in 2023 (AquaSwitch, 2023b)

Calculation of costs (L) of manufacturing 1 tonne of LWA												
	Raw material supply			ly	Transport to manufacturing plant			T (1) ()				
LWA formulation	APCr	Clay	Lime	Water	APCr: EfW plant to Augean facilities (300 km)	Clay: King's Cliffe to Augean facilities (8 km)	Pelletising	Drying at 40°C	Drying at 100°C	Clay firing	75%Clay/ 25%APCr firing	fotal cost of producing 1 tonne of LWA (£)
100%Clay	-	15	-	0.534	-	0.4	10.73	-	0.06	71.41	-	98
25%w-APCr	-	11	-	2.167	4.9	0.3	10.73	-	0.06	-	53.57	83
60c-APCr/30	-	1.5	112.5	0.748	9.4	0.04	10.73	0.06	-	-	-	134

Calculation of costs (£) of manufacturing 1 tonne of LWA

The traditional APCr management in the UK by stabilisation/solidification (S/S) and landfilling is expensive, as discussed in Section 2.3, with prices ranging from 110 to 150 £/tonne. Considering LWA production as an alternative management for APCr, the costs for the management of 1 tonne of APCr can be calculated as shown in Table 7.7. The estimated costs were 264 £/tonne of APCr for hot bonding and 252 £/tonne of APCr for cold bonding. The difference in cost between both management options is due to the incorporation percentage of APCr in each LWA formulation. Also, both management ways were more expensive than the typical treatment by S/S and landfilling of APCr. Therefore, the economic benefit should be considered from the valorisation of this waste material, which is only possible through the adoption of LWA manufacture as management option. Assuming that hot-bonded and cold-bonded LWA were sold at the same price, the values in the last column of Table 7.7 show that the valorisation of APCr would be much higher from the commercialisation of the hot-bonded LWA rather than from the cold-bonded LWA, because of the difference in incorporation percentage of APCr. The prices that are currently paid for treatment and landfilling of APCr (up to 150 £/tonne) added to the landfill tax of hazardous waste (102.1 £/tonne) could partially cover the costs of treating this waste by washing and manufacturing LWA through hot bonding. In addition, considering that the hot-bonded LWA 25%w-APCr showed better technological properties and performance in concrete compared with the cold-bonded LWA 60c-APCr/30, it can be concluded that it is more cost-effective to adopt the production of the hot-bonded LWA 25%w-APCr as an alternative management to diver the landfilling of APCr.

APCr Management	Cost of management £/tonne APCr	Valorisation	
		Price of LWA £/tonne	Price of APCr £/tonne
Production of LWA 25%w-APCr through hot bonding	264 ª	312 °	1,248
Production of LWA 60c-APCr/30 through cold bonding	252 ª	312 °	520
Typical stabilisation/solidification and landfilling	$110 - 150^{\text{b}}$		

Table 7.7 Estimation of cost of APCr management and valorisation of the material.

^a Values calculated from the total costs of producing 1 tonne of LWA (Table 7.6)

^b Estimation for 2023 in the UK (Augean, 2023)

^c Average value of the prices of two commercial brands of LWA. Lytag[®] No fines lightweight floor and roof screed basecoat costs £367.2 per tonne (Specialist Aggregates, 2023) and Argex[®] expanded clay costs £232.16 per 900 kg (£256.8 per tonne) (Day aggregates, 2023).

8 Conclusions, Contribution to Knowledge and Recommendations

8.1 Conclusions

In this research, APCr from different EfW facilities across the UK were investigated for recycling into the manufacture of LWA. Representative samples of APCr were analysed for elemental and mineral composition, leaching properties and microstructure. Washing and accelerated carbonation were optimised for the treatment of APCr aimed to improve the leaching properties of this waste material. The manufacture of LWA from APCr through hot bonding and cold bonding was investigated. The technological characteristics, mineralogy and leaching properties of LWA with different incorporations of APCr were determined. Hot-bonded and cold-bonded LWA with optimal technological and leaching properties were manufactured and incorporated into different formulations of LWAC. The compressive strength, density and possible applications of the manufactured LWAC specimens were evaluated. The embodied carbon associated to the production of 1 tonne of LWAC incorporating APCr-based LWA was estimated to determine the environmental impact of the evaluated option for recycling of APCr. The following are the main conclusions that arise from this study:

- 1. The elemental composition and leaching properties, as regulated by the WAC for hazardous waste, were determined for 27 APCr samples coming from 22 different UK EfW. The environmental concern of APCr was associated with the leaching of heavy metals, including Ba, Cr, Cu, Mo, Pb and Zn. Pb was the most concerning element because it showed the highest leaching rates that exceeded the hazardous WAC. In terms of soluble salts, chlorides and sulphates represented the main environmental concern of APCr due to their high leaching rates. For most of the samples, the leachable fractions of chlorides exceeded the hazardous WAC.
- 2. The mineral characterisation by XRD of 42 APCr samples coming from 22 different UK EfW facilities revealed that APCr was mostly composed of Ca-based phases. These phases included Anhydrite (CaSO₄), Calcite (CaCO₃), Calcium hydroxychloride (CaOHCl) and Portlandite (Ca(OH)₂). Other major phases included Halite (NaCl) and Sylvite (KCl), as well as Quartz (SiO₂). The mineral fraction of APCr, directly related to their leaching properties, due to the presence of chlorides and sulphates, ranged from 13.53±0.58 to 76.73±1.99 wt.%.

- 3. The treatment of APCr by washing with water reduced the leaching rates of metals and soluble salts from the waste material. The optimisation of this treatment showed that when the extraction time is kept constant, the L/S ratio does not significantly affect the washing efficiency. Whereas at a constant L/S ratio, the efficiency of the washing increases as the extraction time increases. The optimum conditions for washing of APCr were determined as an L/S ratio of 2.5 and an extraction time of 60 min. At these washing conditions, the leaching rates from APCr were reduced by 55% Ba, 19% Cr, 16% Cu, 68% Pb, 67% Zn, 88% chlorides and 40% sulphates.
- 4. The accelerated carbonation of APCr by exposition to a closed environment saturated with CO₂, stabilised and reduced the leaching properties of the waste material due to the production of stable carbonates, mainly calcite (CaCO₃). At a continuous flow of 0.1 L/min of CO₂ at atmospheric pressure, the optimum conditions for APCr carbonation were 20% moisture and 2 hr of reaction, achieving a maximum CO₂ uptake of around 12% of the initial mass of APCr. The leaching rates from carbonated APCr were reduced by 37% Ba, 60% Cr, 90% Cu, 100% Pb, 99% Zn, 9% chlorides and 34% sulphates.
- 5. In hot-bonded LWA production, the partial replacement of clay by APCr, either untreated or washed, inhibited the clay expansion and caused a reduction of up to 9% of the original volume of the pellets after the firing treatment. Despite this, the incorporations of 10% to 25% of APCr-m and firing temperature of 1,140 1,160°C, as well as 15 30% of w-APCr and firing temperature of 1,140 to 1,180°C favoured the development of an internal pore structure that positively influenced the physical and technological properties of the LWA.
- 6. The leaching properties of hot-bonded LWA were satisfactory. When APCr-m was mixed with clay and fired, Ba and sulphates leached out from all the manufactured LWA at rates between 0.3 and 2.4 mg/kg of Ba and between 2,654 and 9,935 mg/kg of sulphates, while the other metals and salts did not leach or were detected at negligible concentrations. All the LWA incorporating w-APCr released Ba from 0.8 to 2.1 mg/kg, Cr from 0.4 to 1.2 mg/kg and sulphates from 5,762 to 12,573 mg/kg, while some formulations released chlorides at negligible concentrations and none of the other metals and soluble salts was leached. The persistent leaching of sulphates should be further investigated.

- 7. The hot-bonded LWA 25%w-APCr fired at 1,180°C exhibited physical and technological properties comparable to Lytag[®]. For this LWA formulation, the particle density was 1.89 g/cm³, the loose bulk density was 1.10 g/cm³, the water absorption at 24 hours was 4.4% (lower than Lytag[®]) and the crushing strength was 3.76 MPa (higher than Lytag[®]). Compared with the LWA without the incorporation of APCr, the microstructure of 25%w-APCr exhibited an increase in the number and size of the pores in the internal pore system. In addition, these LWA exhibited vitrification on the surface and across the internal pore structure. Given the mentioned characteristics, LWA 25%w-APCr can be suitable for applications in structural lightweight concrete production.
- 8. Cold bonding of APCr (carbonated or washed) and lime (binder) led to the production of LWA with the incorporation of 60% to 70% of APCr. For this type of LWA, the physical and technological characteristics were influenced by the weight fraction of both APCr and lime. For LWA incorporating c-APCr, the technological characteristics were to a certain extent comparable to Carbon8[®]. The leaching properties associated with heavy metals, such as Ba and Cr were acceptable. However, the leaching of soluble salts was not sufficiently controlled, and particularly the high leaching rates of chlorides can represent an environmental concern. The leaching rates of chlorides from LWA incorporating w-APCr or c-APCr were up to 27,761 mg/kg and 67,922 mg/kg, respectively. The mineralogy of these aggregates did not explain the leaching of chlorides. Consequently, further research aimed to improve the leaching properties of cold-bonded LWA should be addressed.
- 9. In the manufacture of LWA through cold bonding, the LWA incorporating c-APCr exhibited some advantages compared to LWA containing w-APCr, including lower water absorption, lower leaching of sulphates and higher crushing strength. From these LWA, the formulation 60c-APCr/30 exhibited the following technological properties that were more consistent with Carbon8[®]: particle density of 1.93 g/cm³, loose bulk density of 0.99 g/cm³, 4.4% water absorption, 48.5% of voids and crushing strength of 1.41 MPa. Taking into account these characteristics, the LWA 60c-APCr/30 should be further investigated for applications in concrete production.
- 10. The LWAC cubes incorporating hot-bonded LWA 25%w-APCr had a compressive strength of 48.3 MPa, which was 25% higher than the cubes made of Lytag[®] and 33%

higher than the cubes formulated from LWA without incorporation of APCr. These results were in line with the high crushing strength of 25%w-APCr. According to the exposure classification based on the compressive strength, the LWAC made of 25%w-APCr is suitable for applications in which concrete with reinforcement is exposed to air or moisture, direct contact with water containing chlorides (except seawater) and seawater or airborne salt from seawater. The LWAC formulated from cold-bonded LWA 60c-APCr/30 had a compressive strength of 20.3 MPa, which was 15% lower than the LWAC formulated from Carbon8[®]. According to the exposure classification based on the compressive strength, this LWAC is suitable for applications in which concrete without reinforcement can be exposed to environments free of corrosion or chemical attack.

11. The embodied carbon to produce 1 tonne of LWAC incorporating hot-bonded LWA 25%w-APCr was estimated as 337 kg of CO₂. These emissions were 12% lower than the production of LWAC from LWA 100%Clay and comparable to the production of LWAC from Lytag[®]. To produce 1 tonne of LWAC from cold-bonded LWA 60c-APCr/30, the embodied carbon was calculated as 308 kg of CO₂, which was almost 9% lower than the LWAC with incorporation of the hot-bonded LWA 25%w-APCr. The greatest sources of carbon emissions derived from the LWA production were the extraction and supply of clay and the firing process for hot-bonded LWA, and the supply of lime for cold-bonded LWA. The carbon emissions for both scenarios were satisfactory and can help to reduce the total carbon footprint associated with a whole-life carbon assessment for LWAC production.

8.2 Contribution to knowledge

In accordance with the summary of contributions listed in Section 1.3, the following are the novel aspects addressed in this research:

- 1. An in-depth characterisation of APCr samples coming from 22 different EfW facilities across the UK. This characterisation included elemental composition, mineralogy, leaching properties, microstructure, and physical properties.
- Development and implementation of a complete sample preparation process through resin impregnation for analysis of APCr by SEM/EDS, which has not been previously described in the literature. This sample preparation allowed the quantitative analysis of elemental composition of APCr.
- 3. Optimisation of washing with water for treatment of APCr aimed at the reduction of the leaching properties. The optimum conditions allowed to use the minimum amount of water and to perform the procedure in a moderate extraction time. The application of the optimised treatment enabled the APCr for subsequent recycling into LWA.
- 4. Production of hot-bonded LWA incorporating up to 30% of w-APCr. While previous studies only managed to integrate up to 10% of APCr into LWA, the high percentages of incorporation achieved in this research can significantly increase the recycling rates of this challenging hazardous waste. In addition, the hot-bonded LWA exhibited technological properties comparable to or even better than a commercial product. The characterisation of these LWA comprised mineral composition, microstructure of the internal pore system, bloating capacity, leaching properties and technological properties including particle density, loose bulk density, percentage of voids, water absorption and crushing strength.
- 5. Production of LWAC from hot-bonded LWA that incorporated 25% of w-APCr, achieving compressive strength higher than the LWAC made from a commercial product of reference. This LWAC was suitable for applications in concrete with reinforcement exposed to air or moisture, direct contact with water containing chlorides (except seawater) and seawater or airborne salt from seawater.

6. Estimation of the embodied carbon associated with the production of LWAC made from hot-bonded or from cold-bonded LWA incorporating APCr. The carbon emissions were comparable to the use of commercial LWA and could help to reduce the total carbon footprint of a whole-life carbon assessment for LWAC production.

8.3 Recommendations

The technological characteristics of the APCr-based LWA must be considered for taking a decision on which option would be more viable for a pilot or industrial scaling of the LWA manufacturing. The recycling of APCr into LWA through a hot bonding, according to the flow diagram shown in Figure 8.1, is recommended as a viable option based on the laboratory scale development.



Figure 8.1 Flow diagram illustrating the manufacturing of LWA incorporating 25% of w-APCr and the associated carbon emissions.

In regards to the manufacture of LWA 25%w-APCr, the following recommendations need to be considered:

- 1. Implementation of APCr washing at a pilot scale is required in line with the conditions optimised at a laboratory scale. The set up would include a mixing tank with speed and time control, coupled to a filtration system. The feasibility of the treatment will be subject to factors, such as, the type of equipment used for the extraction and operational capacity. In addition, the knowledge of the energy input of the washing process will allow an assessment of the energy balance of the LWA manufacturing process and a more accurate estimation of the embodied carbon. The APCr washing can be improved by implementing a system of treatment and recycling of the generated effluent.
- 2. Additional research on the removal or stabilisation of sulphates in APCr is recommended. For example, the treatment with solutions of Na₂CO₃ during the washing to increase the removal of sulphates by the formation of the more water-soluble Na₂SO₄. Thus, the mobility of these salts can be further reduced in the LWA. It is also advisable to investigate the possible effects of sulphates on the characteristics of the LWAC with the incorporation of w-APCr. In this regard, it is required to consider the sulphate attack occurrence, which leads to the formation of ettringite, that destructively expands generating fracture of the concrete structure, or thaumasite, that produces softening and loss of concrete strength.
- 3. The production of LWA 25%w-APCr at a pilot scale is recommended. The set up would include, among other equipment, a disc pelletiser and a tubular rotary kiln. The physical characteristics and the technological properties of the LWA must be reevaluated in the scaled-up process because the operational conditions may change. The pelletising method will affect aspects like morphology, surface texture and particle uniformity of the LWA. Both, the drying and firing processes, taking place in the kiln, will be affected by the temperature rate, while the reactions caused by the firing will be also subject to the aeration generated by the rotary system. The energy consumption due to drying and firing will be integrated in only one process unit. The emissions from the rotary kiln, including particulate matter (PM), volatile organic compounds (VOCs), heavy metals, chlorides and CO₂, among others, must be quantified to support the technical and environmental viability of the process.

the APCr washing and the hot bonding LWA manufacturing have been optimised at pilot scale, the complete process can be upgraded to a full-scale level.

- 4. The properties and performance of LWAC incorporating LWA 25%w-APCr requires further research, including aspects such as resistance to water penetration, resistance to abrasion, durability, reaction to fire, chloride content, etc., in accordance with the British Standard (BS EN 206, 2013+A2, 2021). The conformity with the specifications for concrete will support the viability of the production of the hotbonded LWA at a large scale. Leaching tests on concrete are not considered in the British Standards, but this analysis can be performed according to procedures previously developed for research purposes or according to international standards. The main aim of the leaching test in the LWAC containing the LWA 25%w-APCr will be evaluating the possible release of sulphates, considering the remaining leachable amount of these salts in the LWA formulation.
- 5. It is recommended to perform an environmental impact assessment of the LWAC manufactured from LWA 25%w-APCr by means of the LCA approach according to the British Standard (BS EN ISO 14040:2006+A1, 2020). In this study, the emissions derived from the LWAC manufacture, which form part of the non-toxicity categories, were calculated. As discussed in Section 7.2, for a complete LCA, all non-toxicity categories (climate change, acidification, nutrient enrichment, photochemical ozone formation and stratospheric ozone depletion) and toxicity categories (ecotoxicity and human toxicity) must be evaluated. The design of an LCA framework setting the scope, inventory analysis, impact assessment and interpretation of the results will be required. Figure 8.2 presents a diagram of the expected life cycle of the LWAC manufactured from LWA 25%w-APCr, showing the materials and processes to be considered in the LCA. It can be seen that the environmental impact management for this product has been approached from the raw materials acquisition to the concrete production, use, demolition and waste recycling.



Figure 8.2 Diagram of the life cycle of LWAC manufactured from hot-bonded LWA incorporating w-APCr. The embodied emissions for LWAC production were calculated in this study. At the end of its lifetime, the LWAC as part of the demolition waste can be sent to reprocessing and subsequently incorporated into the production of new concrete.

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Reference	(Hjelmar, 1996)	(Mizutani, et al., 1996)	(Alba, et al., 1997)	(Lee, et al., 1999)	(Mizutani, et al., 2000)	(Lundtorp, et al., 2002)
Country of origin	The Netherlands and Denmark	Japan	Spain	UK	Japan	Denmark
Acid-gas scrubbing	D: Dry/semi-dry W: Wet	D: Dry W: Wet S: Semi-dry	Semi-dry + ESP	Dry	Dry	Semi-dry
Elements (n	ng/Kg)					
Al	D/S: 17,000 – 46,000 W: 71,000 – 81,000		70,750 as Al ₂ O ₃	14,000		
Ag	D/S: 14 – 60 W. 53					
As	D/S: 40 – 260 W: 130 – 190		<50	19.6		195
В						
Ba	D/S: 310 - 1,400 W: 330 - 1,900			288.75		795
Be	D/S: 0.5 – 0.9 W: 1.5 – 1.9					
Bi						
Br						
С						
Ca	D/S: 170,000 – 290,000 W: 93,000 – 110,000	D: 210,000 – 370,000 W: 105,000 – 120,000 S: 330,000	277,000 as CaO	343,800		253,000
Ce						
	D/S: 140 - 300	D: 74 – 76				
Cd	W: 220 – 270	W: 109 – 410 S: 27	132		74	214
Cl	D/S: 92,000 - 220,000 W: 48,000 - 71,000			195,200		135,000
Co	D/S: 4 – 15 W: 14 – 22			15.8		13
Cr	D/S: 150 – 570 W: 390 – 660		276	163.1		335
Cs						
Cu	D/S: 440 – 1,100 W: 1,000 – 1,400	D: 410 – 570 W: 500 – 2,600 S: 1,900	465	546.35	570	979
F						
Fe	D/S: 3,600 – 18,000 W: 15,000 – 18,000		14,400 as Fe ₂ O ₃	4,900		11,000
Ga						
Ge						
H						
Hf						
Hg	D/S: 9.3 – 44 W: 38 – 390		2.89			13
I						
ln						
K	D/S: 27,000 – 40,000 W: 35,000 – 58,000		43,550 as K ₂ O	12,800	38,000	30,000
La						
Li						
Mg	D/S: 7,100 – 12,000 W: 18,000 – 23,000		19,100 as MgO	4,150		
Mn	D/S: 300 – 700 W: 1,400 – 2,400		600 as MnO	350		

Appendix I – Elemental composition of APCr reported by previous studies. Part A

Мо	D/S: 9.3 - 20 W: 20 - 38			21.4		14
Na	D/S: 12,000 – 19,000		41 200 as Na-O	8 100	29.000	32,000
INd	W: 28,000 - 33,000		41,200 as 11a20	8,100	29,000	52,000
Nb						
Nd						
Ni	D/S: 20 - 63		51.5	28.9		50
0	W: 67 – 110					
	D/S: 1.700 – 4.600					
Р	W: 6.000 – 7.400		15,700 as P ₂ O ₅	2,300		
	D/S: 4.000 – 6.500	D: 1,400 – 1,900				
Pb	W: 5.900 – 8.300	W: 2.900 – 10.000	3,117	3.012	1.400	5,150
		S: 1,300	-,:	-,	-,	-,
Pd						
Pr						
Rb						
c	D/S: 8,000 - 18,000		81 800 as 50	20,800		24,000
3	W: 11,000 – 26,000		81,800 as SO ₃	50,800		34,000
Sb						
Sc						
Sa	D/S: 8.2 - 16			6.05		
30	W: 12			0.03		
c:	D/S: 57,000 - 98,000		150,000	22 100		65 000
51	W: 120,000		150,000	23,100		05,000
Sm						
Sn	D/S: 620 - 780			1 553		410
SII	W: 1,000			1,555		410
Sr	D/S: 400 - 500			196.9		384
51	W: 200			170.5		
Та						
Th						
Ti	D/S: 1,500 – 5,100		10,300 as TiO ₂	2,750		
Tm	W: 5,300 – 8,400					
U						
	D/S: 26 - 62					
V	W: 62					
W						
Y						
Yb						
	D/S: 12,000 – 19,000	D: 5,500 - 6,000				
Zn	W: 20,000 – 23,000	W: 12,900 – 29,000	11,392	5,508	6,000	22,200
		S: 2,200	<i>,</i>			
Zr						

Appendix I - Elemental composition of APCr reported by previous studies. Part B

Reference	(Bodénan & Deniard, 2003)	(Abbas, et al., 2003; Karlfeldt Fedje, et al., 2010)	(Geysen, et al., 2004)	(He, et al., 2004)	(Li, et al., 2004)
Country of origin	Germany, Belgium, Spain, Italy, France and Portugal	Sweden	Belgium	China	China
Acid-gas scrubbing	D: Dry S: Semi-dry	Dry	Semi-dry	Semi-dry	Semi-dry
Al	D: <1,000 - 14,000 S: 1,000 - 46,000	22,200	4,500 - 8,630		39,000 as Al ₂ O ₃
Ag		<1			
As		80			130.7
В					
Ba		770			
Be		1			
Bi					
Br					
С					
Ca	D: 279,000 – 389,000 S: 237,000 – 370,000	363,00	262,700 - 348,000		139,000 as CaO
Ce					
Cd		90	120 - 200	44.9 - 65.8	289.7
Cl	D: 75,000 - 232,000 S: 86,000 - 283,000				124,700
Co		20			
Cr		190	80 - 120	255 - 350	366.2
Cs					
Cu		5,400	430 - 550	561 - 770	1,286
F					
Fe	D: 4,000 – 22,000 S: 2,000 – 22,000	5,602			25,800 as Fe ₂ O ₃
Ga					
Ge					
H					
Hf					
Hg		3			0.435
I In					
K	D: 1,000 – 23,000 S: 2,000 – 24,000	22,800	20,000 - 25,300		87,700 as K ₂ O
La					
Li					
Mg	D: 8,000 - 10,000 S: 5,000 - 17,000	10,100	3,600 - 5,400		31,600 as MgO
Mn	D: 300 – 1,200 S: 200 – 700	574			1,200 as MnO

Мо		10			
Na	D: <1,500 - 11,000 S: 4,000 - 16,000	32,000	19,200 - 24,600		140,000 as Na ₂ O
Nb					
Nd					
Ni		30	10-60	88.1 - 136	74.85
0					
Р	D: <200 - 4,100 S: 200 - 9,200	4,000			28,100 as P ₂ O ₅
Рb		5,730	3,050 - 4,840	972 - 2,480	4,451
Pd					
Pr					
Rb					
S	D: 16,300 – 33,000 S: 16,000 – 31,300	7,000			153,600 as SO ₃
Sb					
Sc					
Se					
Si	D: 2,000 – 24,000 S: 6,000 – 87,000	32,700			85,700 as SiO ₂
Sm					
Sn		20			
Sr		500			
Та					
Th					
Ti	D: 300 – 2,900 S: 400 – 7,200	1,860			7,600 as TiO_2
Tm					
U					
V		10			
W					
Y					
Yb					
Zn		5,780	6,490 - 12,280	3,610 - 4,940	5,622
Zr		40			

Part C

Reference	(Song, et al., 2004)	(Robinson, et al., 2004)	(Hu, 2005)	(Chimenos, et al., 2005)	(Todorovic & Ecke, 2006)
Country of origin	South Korea	UK	Taiwan	Spain	Sweden
Acid-gas scrubbing	Semi-dry	Unspecified	Unspecified	Semi-dry	Dry
Elements (mg/Kg	g)				
Al		17,000 - 22,000	7,000 - 15,000	51,400 as Al ₂ O ₃	
Ag					2.00
As				8.8	309
B	22 082 41 08	(2		506.0	
Ba	55.985 - 41.98	03		500.9	
Bi				14.1	
Br				14.1	
C					
Ca	65.206.959 - 117.000.0	260,000	237.000 - 300.000	301,400 as CaO	
Ce	,	,		12.9	
Cd	15.690 - 190.162	53 - 130	101 - 269	166.6	49.7
Cl	203,200 - 317,200		286,000 - 357,000		
Со	1.874 - 3.275			11.5	34.5
Cr	169.070 - 183.303	67 - 83	174 – 3,213	119	835
Cs				6.2	
Cu	601.880 - 406.700	300 - 900	754 – 1,484	462.2	7,160
F					
Fe	761.848 - 2,006.000	3,800 - 7,300	5,000 - 16,000	8,700 as Fe ₂ O ₃	
Ga				4.7	
Ge					
Ηα	11 443 - 48 445			87	
I	11.443 - 40.445			0.7	
In					
K	15.300 - 105.700 as K ₂ O	14.000 - 29.000	16.000 - 27.000	65.200 as K ₂ O	
La				7.3	
Li				15.9	
Mg	6681.664 - 8,851.0	3,900 - 6,200		20,800 as MgO	
Mn	315.437 - 580.300	253 - 400		500 as MnO	
Мо		40		10.4	27
Na	6,178 - 37,122.575	14,000 - 19,000	8,000 - 13,000	62,200 as Na ₂ O	
Nb				1.4	
Nd				6.1	
N1		19 – 45		40.7	166
<u> </u>	12,000, 14,200, P.O.			14.700 D.O.	
P Dh	$12,000 - 14,200 \text{ as } P_2O_5$	1 200 2 500	2 224 4 720	$14,700 \text{ as } P_2O_5$	2 210
PO	234.000 - 2,033.389	1,300 - 3,300	2,234 - 4,729	4,031.00	5,510
Pr					
Rh				77.5	
S	66.900 - 79.800 as SO ₃		48.000 - 81.000	11.5	33,300
Sb	75.420 -157.768	86 - 260	10,000 01,000	256.6	326
Sc					
Se		<8			
Si	47,300 - 58,300 asSiO ₂		12,000 - 63,000	106,000 as SiO ₂	
Sm					
Sn	366.700 - 767.147	710		545.4	
Sr	108.978 - 138.300			482.5	
Ta				2.0	
Th	11 700 10 100 700			0.000 50	
	$11,700 - 19,400 \text{ as } 11O_2$			8,300 as T1O ₂	
Im TT					
V	1166 6002	14		10.9	
W	+.100 - 0,995	10		40.8	
V				5.4	
Yb					
Zn	9,036.000 -12.813.437	3,600 - 11.000	4.866 - 11.472	9.978.00	10.100
Zr			,,	58.3	

Part D

Reference	(Sun, et al., 2008)	(Dimech, et al., 2008)	(Quina, et al., 2008b)	(Zhang, et al., 2008)	(Hyks, et al., 2009)
Country of origin	UK	UK	Portugal	China	Denmark
Acid-gas scrubbing	Semi-dry	Unspecified	Semi-dry	Semi-dry	Semi-dry
Elements (n	ng/Kg)				
Al	121,200 as Al ₂ O ₃	28,000 as Al ₂ O ₃	29,900 - 40,000		183,000
Ag					
As	210.66	-		71	80
B	210.46	7,000 as B_2O_3			(20)
Ba	319.46				620
Be	0.97				
B1 Dr	2 061 60				000
DI C	2,001.00	21 300	28 400 65 500		900
Ca	362 700 as CaO	420,000 as CaO	26,400 = 05,500 nd = 361,600	300.000	331.000
Ce	58.90	420,000 us CuO	iid 501,000	500,000	551,000
Cd	50.50		65 - 87	57	100
Cl				94,000	173,000
Co	14.16				
Cr		300 as Cr ₂ O ₃	188 - 259	450	200
Cs					
Cu	534.36	400 as CuO	456 - 647	980	500
F					
Fe	10,500 as Fe ₂ O ₃	4,700 as Fe ₂ O ₃	9,620 - 16,100	30,000	9,700
Ga	21.56				
Ge			6 000 10 000		
H			6,800 - 18,900		
			nd 16.21	20	
пg т			nd – 10.51	39	
In					
K	20 300 as K ₂ O	18 700 as K ₂ O	24 200 - 30 200	32,000	17 100
La	45.04	10,700 us 1120	21,200 50,200	52,000	17,100
Li					
Mg	25,000 as MgO	500 as MgO		14,000	7,800
Mn	400 as MnO			1,000	
Mo	0.94				9
Na	78,100 as Na ₂ O	26,800 as Na ₂ O	27,800 - 33,100	29,000	16,000
Nb	2.16				
Nd	50.00			120	
N1	70.32		1 100 400	130	37.5
<u> </u>		7 800 D O	nd – 192,400		2 400
P Dh	2.026	$1,800 \text{ as } P_2 O_5$	1 952 2 409	2.620	3,400
PO	5,020	1,000 as PDO	1,635 - 2,408	2,020	2,100
Pr					
Rb	23.8				
S	62.900 as SO3	56.600 as SO3	11.900 - 18.900		35,000
Sb	,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,	00,000			340
Sc					
Se	0.04				
Si	300,000 as SiO ₂	71,400 as SiO ₂	45,400 - 83,100		63,000
Sm					
Sn					
Sr	473.98				500
Ta	11.50				
Th	5 200 T.O	5 (00 T'O			
11 T	5,300 as 110 ₂	5,600 as 11O ₂			
I M					
V					
W	53.88				
V	4 86				
Yh	00				
Zn	7.521	8.700 as ZnO		5.400	9.100
Zr	111.34	.,,	4,308 - 6,367	2,.30	,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,

Appendix I -	- Elemental co	mposition of	APCr repor	rted by	previous studies.
11		1			1

Part E

Reference	(Gunning, et al., 2011a)	(Cappai, et al., 2012)	(De Boom & Degrez, 2012)	(Chen, et al., 2012)	(Colangelo, et al., 2012)	(Chen, et al., 2013)
Country of origin	UK	Italy	Belgium	Taiwan	Italy	Taiwan
Acid-gas scrubbing	Unspecified	Dry	Dry, wet and semi- dry	Unspecified	Unspecified	Semi-dry
Elements (mg	/Kg)		~			
Al		6,625	2,400 - 7,400	46,000	12,000 - 27,000	3,600
Ag	<0.5 - 485				2.1-5.9	
B	010 100				2.11 0.0	
Ba	196 - 476		230 - 950		112 - 227	
Be			nd 70			
Br			190 - 5,200			
С			,			
Ca		308,069	52,000 - 583,000	150,000	165,000 - 270,000	309,000
Ce	22 - 123 1	112	nd	300	65 - 217	218
Cl	22 125.1	112	16,000 - 395,000	86,000	05 217	210
Co						
Cr	48 - 109.3	74.04	70 - 260	500	85-412	187
Cu	315 - 480	441	110 - 8,400	1,100	815-6,220	129
F				44.000		
Fe		1,946	1,900 - 17,000	11,000	9,450 - 12,000	2,400
Ge						
Н						
Hf	27.22					
Hg I	2.7 - 3.3					
In						
K		41,178	2,500 - 37,000	40,000	11,300 - 24,000	46,000
La						
Mg		29,243	4,000 - 13,000	16,000	1,240 - 8,500	5,000
Mn		170	140 - 530			
Mo	2.4 - 7.1	8.28	2 100 120 000	42 200	15 000 110 000	41.9
Nb		20,233	5,100 - 120,000	42,200	15,000 - 119,000	42,400
Nd						
Ni	7.4 - 36	86.87	nd - 60	475.000	117 – 163	
P			900 - 2 100	475,000		
Pb	1,449 - 6,774	2,176	400 - 3,600	1,600	6,580 - 17,110	3,900
Pd						
Pr Ph						
S			6,600 - 68,000	12,000		
Sb	24.3 - 346	434	330 - 8,130	,		609
Sc	0.0.2					
Si	0.9 - 3	30.002	7.500 - 27.000	103.000	97.000 - 130.000	10.800
Sm			.,			.,
Sn			nd - 220			
Sr Ta			140 - 340			
Th						
Ti			260 - 1,400			800
Tm						
V						
W						
Y						
Y b Zn	560 - 6 110	4 372	820 - 13 000	8 900	6 230 - 9 100	11 300
Zr	200 0,110	-1,572	10	0,700	5,250 9,100	11,500

Part F

Reference	(Bogush, et al., 2015)	(Kepper, et al., 2015)	(Araizi, et al., 2016)	(Ayati, et al., 2016)	(Li, et al., 2017)	(Yang, et al., 2017)
Country of origin	UK	Czech Republic	UK	UK	Hong Kong	China
Acid-gas scrubbing	Unspecified	Unspecified	Unspecified	Unspecified	Wet	Semi-dry
Elements (m	ig/Kg)					
Al	9,000 - 29,000	92,000 as Al ₂ O ₃	8,000 as Al ₂ O ₃		104,200 as Al ₂ O ₃	12,000 as Al ₂ O ₃
Ag	5.5 – 15					
As	12 - 38	<dl< td=""><td></td><td><3.82</td><td></td><td>76</td></dl<>		<3.82		76
В						
Ba	316 - 452	317		429		655
Be D;	0.26 - 0.43					
Br	9,1 - 110					
C						
Ca	224,000 - 320,000	239,000 as CaO	650,000 as CaO	303,000	60,900 as CaO	418,000 as CaO
Ce	8.6 - 12	,	,	,	,	,
Cd	26 - 190	62		116		70
Cl		112,000	98,000	199,000	27,200	166,000
Co	10 - 26	<dl< td=""><td></td><td></td><td></td><td>7</td></dl<>				7
Cr	58-110	131.4	nd Cr ₂ 0 ₃	114		26
Cs	0.86 - 4.1	204.6		(11	700 C O	216
Cu E	320 - 580	204.6		611	700 as CuO	316
г Fe	6 000 - 21 000	26,000 as Fe ₂ O ₂	8 000 as Fe ₂ O ₂		67 400 as FeaOa	4 935
Ga	4.2 - 5.8	20,000 as 1 C ₂ O ₃	0,000 as 1 C ₂ O ₃		07,400 as 1 0203	т,755
Ge	0.19 - 0.66					
Н						
Hf	0.03 - 0.87					
Hg				< 0.92		
Ι						
In	0.72 - 13					
K	9,200 - 35,000	66,000 as K ₂ O	20,000 as K ₂ O	39,300	16,800 as K_2O	
La	5.1-12					
Mg	5 200 - 8 400	18 000 as MgO	12 000 as MgO		24 500 as MgO	17.000 as MgO
Mn	270 - 760	266.4	2,000 as MnO		2 1,000 us 101g0	269
Мо	4.8 - 15	<dl< td=""><td>,</td><td>11</td><td></td><td></td></dl<>	,	11		
Na	12,000 - 35,000	94,000 as Na ₂ O		37,800	138,200 as Na ₂ O	73,000 as Na ₂ O
Nb	<0.7					
Nd	3.6 - 6.9					
Ni	21 - 59	17.2		41		120
0	2 000 (100	12.000 D.O.	2.000 P.O		00.000 D .O	1000 D.O.
P	2,000 - 6,400	$13,000 \text{ as } P_2O_5$	$2,000 \text{ as } P_2O_5$	1.840	99,800 as P ₂ O ₅	$4,000 \text{ as } P_2O_5$
Pd	-2,000 <0.28	998.4		1,040		042
Pr	1.0 - 1.9					
Rb	15-60					
S		115,000 as SO3	120,000 as SO ₃		235,300 as SO3	42,000 as SO3
Sb	170 - 510	218.4		543		
Sc	1.8 - 3.4					
Se	< 0.5 - 9.6			<1.67		
Si	<400-9,200	156,000 as SiO ₂	25,000 as SiO ₂		221,300 as SiO ₂	31,000 as SiO ₂
Sm	0.68 - 1.2	202.4				
Sn Sr	150 - 570 310 450	302.4				
SI Ta	510 - 450 <0.03 - 0.186					
Th	0.93 - 2 1					
Ti	1,100 - 2.200		19,500 as TiO ₂		2,700 as TiO ₂	
Tm	0.043 - 0.060		- ,		,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,	
U	0.63 - 1.4					
V	12 - 36	22.2				141

W	< 6-17					
Y	4.8 - 7.3					
Yb	0.33 - 0.46					
Zn	2,600 - 7,300	5,912.80	25,000 as ZnO	9,190	1,000 as ZnO	3,263
Zr	10 - 37					

Part G

Reference	(Ashraf, et al., 2019)	(Atanes, et al., 2019)	(Bogush, et al., 2019)	(Bogush, et al., 2020)	(Nikravan, et al., 2020)
Country of origin	Canada	Spain	UK	UK	Iran
Acid-gas scrubbing	Unspecified	Unspecified	Unspecified	Unspecified	Semi-dry
Elements (mg	g/Kg)				
Al	1,700 - 14,300 as Al ₂ O ₃	22,300	22,000	83,000 as Al ₂ O ₃	2,800 as Al ₂ O ₃
Ag			13		
As		<25	5.5		
В					
Ba		412	450		
Be					
Bi		<5	16		
Br			530		
С	19,700 - 40,600				
Ca	411,100 - 554,700 as CaO	330,000	350,000	470,000 as CaO	42,000 as CaO
Ce			11		
Cd		60	30		
Cl	58,500 - 173,000		72,000		191,000
Co		9.2	16		
Cr		242	370		180 as Cr ₂ O ₃
Cs					
Cu		330	260		1,500 as CuO
F					
Fe	1,500 - 4,200 as Fe ₂ O ₃	7,900	7,600	22,000 as Fe ₂ O ₃	4,500 as Fe ₂ O ₃
Ga					
Ge			< 0.8		
Н					
Hf			8.8		
Hg			3.2		
Ι			2.5		
In					
K	2,000 - 4,900 as K ₂ O	31,000	10,000	25,000 as K ₂ O	88,800 as K2O
La			8.1		
Li					
Mg	10,600 - 48,600 as MgO	9,400	7,800	13,000 as MgO	6,100 as MgO
Mn	100 - 300 as MnO	315	600		180 as MnO
Mo		8.3	12		
Na	4,100 - 9,100 as Na ₂ O	8,400	12,000	32,000 as Na ₂ O	92,900 as Na ₂ O
Nb					
Nd			7.5		
Ni		93	36		
0					
Р	1,300 - 2,600 as P ₂ O ₅	5,005	3,700	17,000 as P ₂ O ₅	3,000 as P ₂ O ₅
Pb		1,026	550		3,000 as PbO
Pd			0.9		
Pr			2.3		
Rb			21		
S	4,300 - 107,500	13,800	13,000		55,000 as SO ₃
Sb		372	200		
Sc			1.6		
Se			1.9		
Si	17,900 as SiO ₂	42,000	37,000	79,000 as SiO ₂	9,800 as SiO ₂
Sm			1.4		
Sn		351	160		
Sr		361	480		
Та					
Th			6.1		
Ti	300 - 900 as TiO ₂	4,000	6,700		2,600 as TiO ₂
Tm			0.07		
U			42		
V			25		
W		8.2			
Y			11		
Yb			0.64		
Zn		449	2,900		12,200 as ZnO
Zr		73	110		

Appendix I -	Elemental	composition of	of APCr reported	by	previous studies.
11		1	1	~	1

Part H

Reference	(Dontriros, et al., 2020)	(Cao, et al., 2021)	(Fan, et al., 2022)	(Han, et al., 2022)
Country of origin	Thailand	China	China	China
Acid-gas scrubbing	Semi-dry	Semi-dry	Semi-dry	Semi-dry
Elements (mg/	Kg)			
Al	8,500 as Al ₂ O ₃		9,500 - 133,500 as Al ₂ O ₃	$7,900 \text{ as Al}_2O_3$
Ag	nd			
B	IIU			
Ba			1.200 - 2.600 as BaO	
Be			1,200 2,000 us Buo	
Bi				
Br				
С				
Ca	321,000 as CaO	368,200	285,000 - 452,500 as CaO	422,800 as CaO
Ce				
Cd	304 as CdO			
Cl	293,000	107,800	63,000 - 227,500	211,900
Co	01.0 0.0			
Cr C-	91.9 as Cr ₂ O ₃			
Cs Cu	687 as CuO		1 200 - 2 000 as CuO	
E	087 as CuO		1,200 - 3,900 as CuO	
Fe	5 780 as FeaOa		7 500 - 79 700 as FeaOa	12 900 as FeaOa
Ga	5,700 d5 1 0203		7,500 77,700 051 0203	12,900 d3 1 0203
Ge				
Н				
Hf				
Hg				
Ι				
In				
K	65,200 as K ₂ O	22,700	21,200 - 81,500 as K ₂ O	70,500 as K ₂ O
La				
L1	14 100 M-O		0.500 - 20.500 M-O	22 100 M-O
Mg	14,100 as MgO		9,500 - 30,500 as MgO	23,100 as MgO
Mo			500 - 2,100 as MIIO	
Na	139 000 as Na ₂ O	22 000	15 300 - 202 300 as Na ₂ O	87 500 as Na2O
Nb	159,000 us 11020	22,000	10,500 202,500 451420	07,000 ab 11a20
Nd				
Ni				
0		376,200		
Р	7,240 as P ₂ O ₅		3,000 - 74,700 as P ₂ O ₅	
Pb	1,300 as PbO		1,100 - 4,300 as PbO	
Pd				
Pr				
Kb	40.200 az SO	10 000	40.200 00.500 as SO	57 600 ag SQ
Sh	40,500 as 503	18,800	49,500 - 90,500 as 503	57,000 as 503
Sc				
Se				
Si	27,000 as SiO ₂	43,500	21,200 - 203,300 as SiO ₂	49,700 as SiO ₂
Sm				
Sn				
Sr				
Ta				
Th				
Ti T	3,690 as TiO ₂		4,400 - 13,900 as TiO ₂	
Im				
U				
V				
Yh				
Zn	6.000 as ZnO		8,900 - 12,100 as ZnO	16.100 as ZnO
Zr	3,000 43 2110		0,700 12,100 43 2110	10,100 45 2110

Appendix II – Mineral composition of APCr reported by previous studies. Part A

Reference	(Alba, et al., 1997)	(Bodénan & Deniard, 2003)	(Abbas, et al., 2003; Karlfeldt Fedje, et al., 2010)	(Fernández Bertos, et al., 2004c)	(Geysen, et al., 2004)	(He, et al., 2004)
Country	Spain	Germany, Spain, Belgium, Italy, France, Portugal	Sweden	UK	Belgium	China
Acid-gas scrubbing	Semi-dry + ESP	Dry, Semi-dry	Dry	Unspecified	Semi-dry	Semi-dry
Mineral phases (X: detected, M: major,m: nino	r, %: wt%)		-			
Aluminium phosphate AlPO ₄						
Anhydrite CaSO4	Х	М	Х	Х	Х	Х
Anorthite CaAl ₂ Si ₂ O ₈						
Apatite Ca ₁₀ (PO ₄) ₆ (OH,F,Cl) ₂						
Arcanite K ₂ SO ₄						
Bassanite CaSO ₄ *0.5H ₂ O	Х					
Botallackite Cu ₂ (OH) ₃ Cl						
Cadmium lead oxide Cd ₂ PbO ₄						
Calcite CaCO ₃	Х	М	Х	Х	Х	Х
Calcium Chlorite Hydroxide Ca(OCl)2.2Ca(OH)2						
Calcium hydroxychloride CaOHCl	Х	М	Х		Х	
Calcium sulphate hydrate CaSO4*H2O						
Cerussite PbCO ₃						
Copper nickel zinc oxide Cu _{1.02} ZnNi _{3.27} O _{5.29}						
Corundum Al ₂ O ₃						
Cristobalite SiO ₂						
Fedotovite K ₂ Cu ₃ O(SO ₄) ₃						
Feldspar KAlSi ₃ O ₈ – NaAlSi ₃ O ₈ – CaAl ₂ Si ₂ O ₈				X (K, Na)		
Gahnite ZnAl ₂ O ₄						
Gehlenite Ca ₂ Al(AlSiO ₇)			Х	Х		Х
Gismondine CaAl ₂ Si ₂ O ₈ *4H ₂ O	Х					
Gypsum CaSO ₄ *2H ₂ O						
Halite NaCl	М	М	Х	Х	Х	Х
Hematite Fe ₂ O ₃						
Lime CaO						Х
Litharge PbO						Х
Magnesium oxide hydroxide Mg ₃ O ₂ (OH) ₂						
Melilite Ca ₂ (Mg,Al)(Al,Si) ₂ O ₇						
Muskovite (KF) ₂ (Al ₂ O ₃) ₃ (SiO ₂) ₆ (H ₂ O)						
Periclase MgO			Х			
Portlandite Ca(OH) ₂		М	Х	Х	Х	
Potassium aluminiumsilicate K _{1.25} Al _{1.25} Si _{0.75} O ₄						
Potassium Calcium Aluminium Carbonate Silicate Sulfabate (Ca K) (Si Al) Ou (SO (CO))						
Potassium tetrachlorozincate K ₂ ZnCl ₄						
Quartz SiO ₂	Х	М	Х	Х	Х	Х
Sodalite Ca ₈ Al ₁₂ O ₂₄ (MoO ₄) ₂						
Sygenite K ₂ Ca ₂ (SO ₄)H ₂ O				Х		
Sylvite KCl	М	М	Х	Х	Х	Х
Thenardite Na ₂ SO ₄				Х		
Tenorite CuO						
Tobermorite Ca ₅ Si ₆ O ₁₆ (OH) ₂ *4H ₂ O						
Willemite Zn ₂ SiO ₄						
Whitlockite Ca ₃ (PO ₄) ₂						
Zincite ZnO						
Zincowoodwardite Al _{0.375} H ₆ O _{2.686} S _{0.188} Zn _{0.625}						
Al ₂ SiO ₅						Х
Ca ₃ Al ₂ O ₆			X			
CaAl ₂ SiO ₂ O ₈						X
CaCl ₂ *2H ₂ O						
CaCl ₂ *Ca(OH) ₂ *H ₂ O						
Ca(ClO) ₂ *H ₂ O						
Cu ₂ CO ₄						X
KCaCl ₃			Х			
SiCl ₄						
Zn ₂ SiO ₄						Х

Appendix II - Mineral composition of APCr reported by previous studies. Part B

Reference	(Li, et al., 2004)	(Hu, 2005)	(Chimenos, et al., 2005)	(Sun, et al., 2008)	(Dimech, et al., 2008)	(Quina, et al., 2008b)
Country	China	Taiwan	Spain	UK	UK	Portugal
Acid-gas scrubbing	Semi-dry	Unspecified	Semi-dry	Semi-dry	Unspecified	Semi-dry
Mineral phases (X: detected, M: major,m: nino	r, %: wt%)			•	•	
Aluminium phosphate AlPO ₄						
Anhydrite CaSO4		Х	m	Х	М	Х
Anorthite CaAl ₂ Si ₂ O ₈						
Apatite Ca10(PO4)6(OH,F,Cl)2						
Arcanite K ₂ SO ₄						
Bassanite CaSO ₄ *0.5H ₂ O						
Botallackite Cu ₂ (OH) ₃ Cl						
Cadmium lead oxide Cd ₂ PbO ₄						
Calcite CaCO ₃	m	Х	m	Х	М	Х
Calcium Chlorite Hydroxide Ca(OCl)2.2Ca(OH)2						
Calcium hydroxychloride CaOHCl		Х		Х	М	Х
Calcium sulphate hydrate CaSO4*H2O						
Cerussite PbCO ₃						
Copper nickel zinc oxide Cu _{1.02} ZnNi _{3.27} O _{5.29}						
Corundum Al ₂ O ₃						
Cristobalite SiO ₂	m					
Fedotovite K ₂ Cu ₃ O(SO ₄) ₃						
Feldspar KAlSi ₃ O ₈ – NaAlSi ₃ O ₈ – CaAl ₂ Si ₂ O ₈						
Gahnite ZnAl ₂ O ₄						
Gehlenite Ca ₂ Al(AlSiO ₇)						Х
Gismondine CaAl ₂ Si ₂ O ₈ *4H ₂ O						
Gypsum CaSO4*2H2O						
Halite NaCl	М	Х	М		М	Х
Hematite Fe ₂ O ₃						х
Lime CaO				x		
Litharge PbO		Х				
Magnesium oxide hydroxide Mg ₃ O ₂ (OH) ₂						
Melilite Ca2(Mg,Al)(Al,Si)2O7						
Muskovite (KF) ₂ (Al ₂ O ₃) ₃ (SiO ₂) ₆ (H ₂ O)						
Periclase MgO				Х		
Portlandite Ca(OH) ₂		Х		X	М	Х
Potassium aluminiumsilicate K ₁ 25Al ₁ 25Si ₀ 75O ₄						
Potassium Calcium Aluminium Carbonate						
Silicate Sulfphate (Ca,K) ₄ (Si,Al) ₅ O ₁₁ (SO ₄ ,CO ₃)						
Potassium tetrachlorozincate K2ZnCl4						
Quartz SiO ₂				Х		Х
Sodalite Ca ₈ Al ₁₂ O ₂₄ (MoO ₄) ₂						
Sygenite K ₂ Ca ₂ (SO ₄)H ₂ O						
Sylvite KCl	М	Х	М	Х	М	Х
Thenardite Na ₂ SO ₄						
Tenorite CuO						
Tobermorite Ca ₅ Si ₆ O ₁₆ (OH) ₂ *4H ₂ O						
Willemite Zn ₂ SiO ₄						
Whitlockite Ca ₃ (PO ₄) ₂						
Zincite ZnO		Х				
Zincowoodwardite Al _{0.375} H ₆ O _{2.686} S _{0.188} Zn _{0.625}						
Al ₂ SiO ₅						
Ca ₃ Al ₂ O ₆						
CaAl ₂ SiO ₂ O ₈						
CaCl ₂ *2H ₂ O						
CaCl ₂ *Ca(OH) ₂ *H ₂ O		Х				
Ca(ClO) ₂ *H ₂ O		Х				
Cu ₂ CO ₄						
KCaCl ₃						
SiCl ₄						
Zn ₂ SiO ₄						

Appendix II - Mineral composition of APCr reported by previous studies. Part C

Reference	(Cappai, et al., 2012)	(Chen, et al., 2012)	(Bogush, et al., 2015)	(Kepper, et al., 2015)	(Araizi, et al., 2016)	(Li, et al., 2017)	(Yang, et al., 2017)
Country	Italy	Taiwan	UK	Czech Rep.	UK	Hong Kong	China
Acid-gas scrubbing	Dry	Unspecified	Unspecified	Unspecified	Unspecified	Wet	Semi-dry
Mineral phases (X: detected, M: major,m: nine	or, %: wt%)	•	•	•		•	
Aluminium phosphate AlPO ₄						Х	
Anhydrite CaSO ₄	Х		Х	7.0%	Х		Х
Anorthite CaAl ₂ Si ₂ O ₈				11.7%			
Apatite Ca10(PO4)6(OH,F,Cl)2							
Arcanite K ₂ SO ₄				3.8%			
Bassanite CaSO ₄ *0.5H ₂ O							
Botallackite Cu ₂ (OH) ₃ Cl			Х				
Cadmium lead oxide Cd ₂ PbO ₄							
Calcite CaCO ₃	m	М	Х	6.0%	Х		
Calcium Chlorite Hydroxide							
Calcium hydroxychloride CaOHCl	М	Х	Х		Х		х
Calcium sulphate hydrate CaSO4*H2O			X				
Cerussite PbCO ₃			X				
Copper nickel zinc oxide Cu ₁ 02ZnNi ₃ 27O ₅ 29			X				
Corundum Al ₂ O ₃							
Cristobalite SiO ₂							
Fedotovite K ₂ Cu ₃ O(SO ₄) ₃			Х				
Feldspar KAlSi ₃ O ₈ – NaAlSi ₃ O ₈ – CaAl ₂ Si ₂ O ₈						М	
Gahnite ZnAl ₂ O ₄			Х				
Gehlenite Ca ₂ Al(AlSiO ₇)			Х	10.8%			
Gismondine CaAl ₂ Si ₂ O ₈ *4H ₂ O							
Gypsum CaSO ₄ *2H ₂ O							
Halite NaCl	М	М	Х	3.2%	Х		М
Hematite Fe ₂ O ₃						М	
Lime CaO		М	Х		Х		
Litharge PbO							
Magnesium oxide hydroxide Mg ₃ O ₂ (OH) ₂			Х				
Melilite Ca2(Mg,Al)(Al,Si)2O7							
Muskovite (KF)2(Al2O3)3(SiO2)6(H2O)				2.30%			
Periclase MgO							
Portlandite Ca(OH) ₂	М	М	Х		Х		Х
Potassium aluminiumsilicate K1.25Al1.25Si0.75O4			Х				
Potassium Calcium Aluminium Carbonate Silicate Sulfphate (Ca,K)4(Si,Al)5O11(SO4,CO3)							
Potassium tetrachlorozincate K ₂ ZnCl ₄							
Quartz SiO ₂			Х	8.5%		М	М
Sodalite Ca ₈ Al ₁₂ O ₂₄ (MoO ₄) ₂			Х				
Sygenite K ₂ Ca ₂ (SO ₄)H ₂ O							
Sylvite KCl	Х	М	Х	2.3%			М
Thenardite Na ₂ SO ₄	М						
Tenorite CuO			Х				
Tobermorite Ca ₅ Si ₆ O ₁₆ (OH) ₂ *4H ₂ O			Х				
Willemite Zn ₂ SiO ₄							
Whitlockite Ca ₃ (PO ₄) ₂						М	
Zincite ZnO							
Zincowoodwardite Al _{0.375} H ₆ O _{2.686} S _{0.188} Zn _{0.625}			Х				
Al ₂ SiO ₅							
Ca ₃ Al ₂ O ₆							
CaAl ₂ SiO ₂ O ₈							
CaCl ₂ *2H ₂ O		Х					
CaCl ₂ *Ca(OH) ₂ *H ₂ O							
Ca(ClO)2*H2O							
Cu ₂ CO ₄							
KCaCl ₃							
SiCl ₄							
Zn ₂ SiO ₄							

Appendix II - Mineral composition of APCr reported by previous studies. Part D

Reference	(Atanes, et al., 2019)	(Bogush, et al., 2019)	(Nikravan, et al., 2020)	(Dontriros, et al., 2020)	(Cao, et al., 2021)	(Fan, et al., 2022)	(Han, et al., 2022)
Country	Spain	UK	Iran	Thailand	China	China	China
Acid-gas scrubbing	Unspecified	Unspecified	Semi-dry	Semi-dry	Semi-dry	Semi-dry	Semi-dry
Mineral phases (X: detected, M: major,m: nine	or, %: wt%)						
Aluminium phosphate AlPO ₄							
Anhydrite CaSO ₄	Х	М		М	М	Х	М
Anorthite CaAl ₂ Si ₂ O ₈							
Apatite Ca10(PO4)6(OH,F,Cl)2		m					
Arcanite K ₂ SO ₄							
Bassanite CaSO ₄ *0.5H ₂ O							
Botallackite Cu ₂ (OH) ₃ Cl							
Cadmium lead oxide Cd ₂ PbO ₄			Х				
Calcite CaCO ₃	Х	М		Х	М	М	
Calcium Chlorite Hydroxide Ca(OCl) _{2.2} Ca(OH) ₂			Х				
Calcium hydroxychloride CaOHCl	X	М		М	Х	М	М
Calcium sulphate hydrate CaSO ₄ *H ₂ O							
Cerussite PbCO ₃							
Copper nickel zinc oxide Cu _{1.02} ZnNi _{3.27} O _{5.29}							
Corundum Al ₂ O ₃		М					
Cristobalite SiO ₂							
Fedotovite K ₂ Cu ₃ O(SO ₄) ₃							
$Feldspar \ KAlSi_3O_8 - NaAlSi_3O_8 - CaAl_2Si_2O_8$							
Gahnite ZnAl ₂ O ₄							
Gehlenite Ca ₂ Al(AlSiO ₇)		М				Х	
Gismondine CaAl ₂ Si ₂ O ₈ *4H ₂ O							
Gypsum CaSO4*2H2O						М	
Halite NaCl	Х	М	М	М	М	М	М
Hematite Fe ₂ O ₃						Х	
Lime CaO		М					
Litharge PbO							
Magnesium oxide hydroxide Mg ₃ O ₂ (OH) ₂							
Melilite Ca2(Mg,Al)(Al,Si)2O7		М					
Muskovite (KF) ₂ (Al ₂ O ₃) ₃ (SiO ₂) ₆ (H ₂ O)							
Periclase MgO							
Portlandite Ca(OH) ₂	Х					Х	
Potassium aluminiumsilicate K _{1.25} Al _{1.25} Si _{0.75} O ₄							
Potassium Calcium Aluminium Carbonate Silicate Sulfphate (Ca,K)4(Si,Al)5O11(SO4,CO3)			х				
Potassium tetrachlorozincate K2ZnCl4			Х				
Quartz SiO ₂		М			Х	Х	
Sodalite Ca ₈ Al ₁₂ O ₂₄ (MoO ₄) ₂							
Sygenite K ₂ Ca ₂ (SO ₄)H ₂ O							
Sylvite KCl	Х	М	М	М	М	М	М
Thenardite Na ₂ SO ₄							
Tenorite CuO							
Tobermorite Ca ₅ Si ₆ O ₁₆ (OH) ₂ *4H ₂ O							
Willemite Zn ₂ SiO ₄		m					
Whitlockite Ca ₃ (PO ₄) ₂							
Zincite ZnO							
Zincowoodwardite Al _{0.375} H ₆ O _{2.686} S _{0.188} Zn _{0.625}							
Al ₂ SiO ₅							
CaAl2SIU2U8							
$C_{a}(C_{1}O) = *H_{2}O$							
SiCL						v	
			v			Λ	
21120104	1		л	1		1	

Appendix III – Visual description of APCr samples

Part A

APCr-01	APCr-02
Light grey, fine powder, strong and irritating smell (ammonia), no moisture	Dark grey, fine powd presence of white fine particl odourless, no moisture
APCr-03	APCr-04
Light grey, fine powder, strong smell (ammonia), no moisture	Light grey, fine powder, presence of black fine particles (charcoal), odourless, no moisture
APCr-05	APCr-06
Medium grey, fine powder, presence of white fine particles, strong smell (ammonia), no moisture	Beige, fine powder, strong and irritating smell (ammonia), no moisture
APCr-07	APCr-08
Medium grey, fine powder, strong smell (ammonia), odourless, no moisture	Dark grey powder mixed with coarse-grained charcoal, quite dusty, odourless, no moisture
APCr-09	APCr-10
Brown-grey, fine powder, presence of white fine particles, strong and irritating smell (ammonia), apparently moistured	Light grey, fine powder, presence of various lumps at different sizes that can be easily broken by pressure, odourless, probably certain moisture causes formation of lumps
APCr-11	APCr-12
Light grey-beige, fine powder, presence of various lumps at different sizes that can be easily broken by pressure, slight odour of ammonia, probably certain moisture causes formation of lumps	Light grey, fine powder, presence of small lumps that can be broken easily by pressure, presence of a few fine black particles (probably carbon), odourless, no moisture
APCr-13	APCr-14
Light grey, fine powder, odourless, no moisture	Medium grey-beige, fine powder, presence of lumps at different sizes that can easily be broken by pressure, odourless, presence of a few fine granules of coal, evidently moistured
APCr-15	APCr-16
Dark grey, wet lumps that can be destroyed by pressure, odourless	Light grey, fine powder, slight odour of ammonia, presence of small and medium lumps that can be broken easily by pressure, no moisture

Appendix III – Visual description of APCr samples

Part B

APCr-17	APCr-18
Medium grey, fine powder, presence of lumps at different sizes that can easily be broken by pressure, odourless, evidently moistured	Beige, wet lumps that can easily be broken by pressure
APCr-19	APCr-20
Light grey, fine powder, presence of black fine granules (coal), odourless, no moisture	Medium grey, fine powder, presence of black fine particles, odourless, no moisture
APCr-21	APCr-22
Light grey, fine powder, odourless, no moisture	Medium grey-beige, fine powder, odourless, no moisture
APCr-23	APCr-24
Medium grey, fine powder, presence of a few black fine particles, odourless, no moisture	Medium grey, fine powder, presence of black fine particles, odourless, no moisture
APCr-25	APCr-26
Beige, fine powder, presence of a few black fine particles, odourless, no moisture	Medium grey, fine powder, presence of black fine particles, odourless, no moisture
APCr-27	APCr-28
Dark beige, fine powder, presence of small lumps that can be easily be broken, presence of small crystal (colours blue, yellow green, white), no moisture	Dark grey, fine powder, lumps at different sizes that can be broken by pressure, presence of white fine particles, evidently moisture
APCr-29	APCr-30
Light grey-beige, fine powder, presence of various lumps at different sizes that can be easily broken by pressure, slight odour of ammonia, probably certain moisture causes formation of lumps	Medium grey fine powder, presence of lumps of different sizes that be can be broken easily by pressure, presence of small white lumps that can be broken easily by pressure, presence of a small solid residue (maybe plastic or paper), slight odour of ammonia, no moisture

Appendix III – Visual description of APCr samples

Part C

ADC 21	A.D.C. 22	
APCr-31	APCr-32	
White fine powder, presence of lumps of different sizes (small and medium lumps can be broken easily, bigger lumps are much harder), odourless, no moisture	Light grey-beige, fine powder, presence of small and medium lumps that can be broken easily by pressure, presence of a few black small particles (carbon), slight odour of ammonia, no moisture.	
APCr-33	APCr-34	
Medium grey, fine powder, odourless, no apparent moisture	Medium grey, fine powder, presence of a few black particles of activated carbon, odourless, no apparent moisture	
APCr-35	APCr-36	
Dark beige, powder like sand, presence of small and medium lumps that can be broken easily by pressure, presence of fine white particles as well as a few black particles (maybe carbon), it could have moisture	Light beige, fine powder, odourless, no apparent moisture	
APCr-37	APCr-38	
Medium grey, fine powder, presence of a few black particles of activated carbon, presence of fine white particles all over the sample, odourless, no apparent moisture	Beige, fine powder, presence of a few black particles (carbon), odourless, no apparent moisture]
APCr-39	 APCr-40	
Beige, fine powder, presence of white fine particles all over the sample, presence of a few black particles of carbon, odourless, no apparent moisture	Medium grey, fine powder, odourless, no apparent moisture	
APCr-41	APCr-42	
Beige, fine powder, presence of white fine particles all over the sample, presence of a few black particles of carbon, odourless, no apparent moisture	Light beige, fine powder, presence of black particles of carbon odourless, no apparent moisture	

Appendix IV – Challenges in the sample preparation for analysis of APCr analysis by SEM/EDS

Stage	Challenge	Alternative solution
Compressing of samples	 Samples containing carbon were more difficult to be compressed. Samples containing more carbon than ashes cannot be compressed. In some samples, the material kept coming out after compressing. 	
Polishing	 Samples with high contents of carbon released this material easily, then, these samples were more difficult to polish. The exact amount of diamond suspension required for polishing should be defined. The velvet fibres from the polishing pads came out and adhered to the sample surface. It may be necessary a drying step after the polishing process. It may be necessary a drying step after the polishing process. Digital microscopy image of a velvet fibre adhered to the polished sample surface. 	 Polishing with sandpaper required a continuous addition of isopropyl alcohol. Polished samples were observed under the digital microscope after each polishing step, so that, if velvet fibres appeared embedded into their microstructure, it was applied a cleaning step with isopropanol in ultrasound
Carbon coating	 Despite some samples being coated three times, there was a charging effect when they were observed under the SEM. Sometimes, the current does not pass through the carbon rods, and the whole process of coating need to be restarted. 	The coating process was repeated several times until obtaining a dark appearance of the sample surface. As a result, the real thickness of the carbon layer is unknown.

Appendix V - Formulation of LWAC by the absolute volume method

Part A

Absolute volume formula										
m water		m cement		m sand		m LWA		m air		1 m 3
d water	+	d cement	+	d sand	+	d LWA	+	d air	=	1 m

m water 0.4 _ m cement m water =0.4 m cement m LWA m sand 0.65 = +d sand d LWA m air = 0.01 d air 0.4 m cement m cement ++ 0.65 +0.01 1 = d water d cement d cement (Determined in laboratory) 1,150.39 = d water = 1,000 0.4 m cement m cement 0.01 +0.65 +1 = 1,000 +1,150.39 $\left(\frac{0.4}{1,000} + \frac{1}{1,150.39}\right)$ = 1 - 0.65 - 0.01m cement \times 0.34 m cement x 0.001269 = m cement 267.9 m water = 107.1 7 m sand 0.54 = m LWA = 13 m sand = 0.54 m LWA1,553.44 (Determined in laboratory) d sand =

m=mass (kg), d=bulk density (kg/m³)

Appendix V - Formulation of LWAC by the absolute volume method

Part B

Calculat	ion of n	n Sand and n	n LV	WA (Replace the	bul	k density of L	WA ii	n the gro	een cel	1)
water		cement		sand		LWA		air		
107.1	1	267.9		0.54 m LWA		m LWA		0.01	_	1
1,000	Ŧ	1,150	т	1,553	+	1,102.07	Ŧ	0.01	_	I

0.54 m LWA	m LWA = 0.65	
1,553.44	+ 1102.07 $-$ 0.03	
$m LWA \times \left(\frac{1}{1}\right)$	$\frac{0.54}{,553.44} + \frac{1}{dLWA} = 0.65$	
M LWA	x 0.001255 = 0.65	
m LWA	= 518	

107.1	+	267.9	+	m sand	- + -	517.9	- +	0.01	=	1
1,000		1,150.39		1,553.44		1,102.07				
m sand 1,553.44	- =	0.18								

m sand
$$=$$
 280

water		cement		sand		LWA		air			LWA Formulation
107 1,000	- +	268 1,150.39	- +	280 1,553.44	- +	518 1,102.07	- +	0.01	=	1.00	25% w-APCr, 75% clay
107 1,000	+	268 1,150.39	+	242 1,553.44	-+	448 906.57	- +	0.01	=	1.00	30% w-APCr, 70% clay
107 1,000	- +	268 1,150.39	- +	232 1,553.44	-+	430 859.67	- +	0.01	=	1.00	60% w-APCr, 10% clay, 30% lime
107 1,000	- +	268 1,150.39	- +	222 1,553.44	- +	412 812.34	- +	0.01	=	1.00	70% w-APCr, 30% lime
107 1,000	- +	268 1,150.39	- +	259 1,553.44	-+	481 994.78	- +	0.01	=	1.00	60% c-APCr, 10% clay, 30% lime
107 1,000	- +	268 1,150.39	- +	244 1,553.44	- +	452 916.53	- +	0.01	=	1.00	70% c-APCr, 30% lime
107 1,000	- +	268 1,150.39	- +	250 1,553.44	- +	463 946.73	- +	0.01	=	1.00	100% Clay
107 1,000	- +	268 1,150.39	- +	229 1,553.44	- +	423 841.24	- +	0.01	=	1.00	Lytag®
107 1,000	- +	268 1,150.39	- +	184 1,553.44	- +	340 639.78	- +	0.01	=	1.00	Carbon8®



Appendix VI – SEM/EDS spectra of APCr samples for elemental composition












































































-														
Element	APCr-01	APCr-02	APCr-03	APCr-04	APCr-05	APCr-06	APCr-07	APCr-09	APCr-10	APCr-11	APCr-12	APCr-13	APCr-14	APCr-15
Al	0.9 ± 0.1	1.7 ± 0.1	< 0.1	1.6 ± 0.1	< 0.1	0.3 ± 0.0	1.5 ± 0.1	4.8 ± 0.1	< 0.1	1.2 ± 0.1	0.2 ± 0.0	1.2 ± 0.1	2.8 ± 0.1	2.1 ± 0.1
As	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1
Ba	< 0.1	0.9 ± 0.2	< 0.1	0.7 ± 0.2	< 0.1	< 0.1	1.3 ± 0.2	2.9 ± 0.2	< 0.1	0.6 ± 0.2	< 0.1	2.1 ± 0.2	< 0.1	1.0 ± 0.2
Ca	45.0 ± 0.6	27.8 ± 0.3	3.6 ± 0.2	35.0 ± 0.4	10.9 ± 0.1	32.7 ± 0.4	35.1 ± 0.4	20.1 ± 0.3	35.5 ± 0.4	15.2 ± 0.2	33.3 ± 0.4	36.0 ± 0.4	$29.9{\pm}~0.2$	28.7 ± 0.3
Cd	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1
Cl	10.5 ± 0.2	10.9 ± 0.1	1.7 ± 0.2	10.1 ± 0.2	11.3 ± 0.1	15.5 ± 0.2	4.4 ± 0.1	2.4 ± 0.1	21.4 ± 0.2	2.4 ± 0.1	22.1 ± 0.3	7.3 ± 0.1	10.5 ± 0.1	1.1 ± 0.1
Со	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1
Cr	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1
Cu	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1
F	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5
Fe	0.4 ± 0.1	1.6 ± 0.1	< 0.1	1.2 ± 0.1	< 0.1	0.3 ± 0.1	0.8 ± 0.1	4.5 ± 0.2	< 0.1	< 0.1	< 0.1	1.1 ± 0.1	0.3 ± 0.1	1.8 ± 0.1
K	0.6 ± 0.1	2.2 ± 0.1	< 0.1	2.7 ± 0.1	< 0.1	1.0 ± 0.1	0.7 ± 0.0	2.4 ± 0.1	< 0.1	0.3 ± 0.1	< 0.1	2.6 ± 0.1	2.6 ± 0.1	0.4 ± 0.1
Mg	0.4 ± 0.1	0.7 ± 0.1	< 0.1	1.0 ± 0.1	< 0.1	< 0.1	0.9 ± 0.1	1.7 ± 0.1	< 0.1	0.8 ± 0.1	< 0.1	0.7 ± 0.1	0.9 ± 0.1	1.3 ± 0.1
Mn	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	0.5 ± 0.1	< 0.1	< 0.1	< 0.1	0.2 ± 0.1	< 0.1	< 0.1
Мо	< 0.3	< 0.3	< 0.3	< 0.3	< 0.3	< 0.3	< 0.3	< 0.3	< 0.3	< 0.3	< 0.3	< 0.3	< 0.3	< 0.3
Ν	<3.7	<3.7	<3.7	<3.7	<3.7	<3.7	<3.7	<3.7	<3.7	<3.7	<3.7	<3.7	<3.7	<3.7
Na	1.6 ± 0.1	1.3 ± 0.1	2.4 ± 0.6	1.4 ± 0.1	1.9 ± 0.2	0.5 ± 0.1	0.6 ± 0.1	1.4 ± 0.1	0.4 ± 0.1	< 0.1	< 0.1	1.1 ± 0.1	2.8 ± 0.1	1.0 ± 0.1
Ni	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1
0	36.6 ± 0.8	40.8 ± 0.6	88.5 ± 0.7	38.2 ± 0.7	75.1 ± 0.2	42.2 ± 0.6	46.1 ± 0.6	30.4 ± 0.6	41.7 ± 0.6	73.6 ± 0.4	40.5 ± 0.6	37.7 ± 0.6	42.4 ± 0.4	45.8 ± 0.6
Р	< 0.1	0.3 ± 0.0	< 0.1	0.5 ± 0.1	< 0.1	< 0.1	0.3 ± 0.0	0.8 ± 0.1	< 0.1	0.2 ± 0.1	< 0.1	0.3 ± 0.0	0.6 ± 0.1	0.4 ± 0.1
Pb	< 0.2	< 0.2	< 0.2	< 0.2	< 0.2	4.4 ± 0.3	1.2 ± 0.3	3.3 ± 0.4	< 0.2	< 0.2	< 0.2	1.5 ± 0.3	< 0.2	1.2 ± 0.3
S	1.6 ± 0.1	4.0 ± 0.1	2.1 ± 0.3	2.6 ± 0.1	0.3 ± 0.1	2.1 ± 0.1	2.7 ± 0.1	4.2 ± 0.2	< 0.1	3.3 ± 0.1	2.1 ± 0.1	4.2 ± 0.1	2.7 ± 0.1	3.4 ± 0.1
Sb	< 0.2	< 0.2	< 0.2	< 0.2	< 0.2	< 0.2	< 0.2	< 0.2	1.0 ± 0.3	< 0.2	1.2 ± 0.3	< 0.2	< 0.2	< 0.2
Si	1.9 ± 0.1	5.7 ± 0.1	1.8 ± 0.2	4.2 ± 0.1	0.4 ± 0.1	0.4 ± 0.0	3.8 ± 0.1	18.2 ± 0.2	0.1 ± 0.0	2.4 ± 0.1	0.7 ± 0.0	3.3 ± 0.1	3.9 ± 0.1	7.3 ± 0.1
Zn	0.6 ± 0.2	2.0 ± 0.2	< 0.1	0.8 ± 0.2	< 0.1	0.6 ± 0.1	0.5 ± 0.1	2.3 ± 0.2	< 0.1	< 0.1	< 0.1	0.7 ± 0.1	0.7 ± 0.1	4.6 ± 0.2
Total	100	100	100	100	100	100	100	100	100	100	100	100	100	100

Appendix VII - Elemental composition of APCr analysed by SEM/EDS

Part A

 $\hline Elemental \ concentrations \ are \ expressed \ in \ wt.\%. \ The \pm values \ correspond \ to \ one \ standard \ deviation \ of \ the \ mean \ value, \ calculated \ by \ the \ software.$

Element	APCr-16	APCr-17	APCr-18	APCr-19	APCr-20	APCr-21	APCr-22	APCr-23	APCr-24	APCr-25	APCr-26	APCr-27	APCr-28	APCr-29
Al	1.6 ± 0.1	2.6 ± 0.1	0.5 ± 0.1	0.3 ± 0.1	1.3 ± 0.1	0.3 ± 0.0	1.3 ± 0.1	1.2 ± 0.1	0.2 ± 0.0	1.0 ± 0.1	0.3 ± 0.0	1.8 ± 0.1	2.0 ± 0.1	0.4 ± 0.1
As	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1
Ba	< 0.1	0.9 ± 0.2	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	0.5 ± 0.1	1.0 ± 0.2	< 0.1
Ca	34.5 ± 0.3	35.3 ± 0.4	15.3 ± 0.2	28.0 ± 0.2	24.8 ± 0.2	31.5 ± 0.3	35.7 ± 0.3	33.4 ± 0.3	30.9 ± 0.2	38.1 ± 0.3	26.7 ± 0.2	11.0 ± 0.1	29.0 ± 0.3	41.1 ± 0.3
Cd	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1
Cl	6.7 ± 0.1	5.8 ± 0.1	2.0 ± 0.1	10.1 ± 0.1	13.8 ± 0.1	21.4 ± 0.2	9.4 ± 0.1	13.3 ± 0.1	11.6 ± 0.1	10.5 ± 0.1	9.9 ± 0.1	4.5 ± 0.1	14.2 ± 0.2	7.8 ± 0.1
Со	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1
Cr	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1
Cu	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1
F	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5
Fe	1.3 ± 0.1	0.4 ± 0.1	< 0.1	< 0.1	0.3 ± 0.1	< 0.1	0.7 ± 0.1	0.9 ± 0.1	< 0.1	0.4 ± 0.1	< 0.1	0.8 ± 0.1	0.8 ± 0.1	< 0.1
K	3.0 ± 0.1	1.3 ± 0.1	< 0.1	15.6 ± 0.2	0.8 ± 0.0	1.6 ± 0.1	0.6 ± 0.0	2.5 ± 0.1	11.1 ± 0.1	1.6 ± 0.1	9.5 ± 0.1	1.9 ± 0.1	1.7 ± 0.1	3.2 ± 0.1
Mg	0.8 ± 0.1	0.9 ± 0.1	< 0.1	< 0.1	0.4 ± 0.1	< 0.1	0.5 ± 0.1	0.7 ± 0.1	< 0.1	0.9 ± 0.1	0.3 ± 0.1	1.2 ± 0.1	0.6 ± 0.1	0.2 ± 0.1
Mn	0.3 ± 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1
Мо	< 0.3	< 0.3	< 0.3	< 0.3	< 0.3	< 0.3	< 0.3	< 0.3	< 0.3	< 0.3	< 0.3	< 0.3	< 0.3	< 0.3
Ν	<3.7	<3.7	<3.7	<3.7	<3.7	<3.7	<3.7	<3.7	<3.7	<3.7	<3.7	<3.7	<3.7	<3.7
Na	1.0 ± 0.1	1.0 ± 0.1	< 0.1	0.9 ± 0.1	0.6 ± 0.1	< 0.1	0.8 ± 0.1	1.9 ± 0.1	0.6 ± 0.1	1.3 ± 0.1	0.6 ± 0.1	6.3 ± 0.1	1.8 ± 0.1	0.9 ± 0.1
Ni	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1
0	41.7 ± 0.4	44.6 ± 0.6	76.5 ± 0.3	37.5 ± 0.4	48.0 ± 0.3	43.4 ± 0.6	44.7 ± 0.4	$39.3 \pm \! 0.4$	37.8 ± 0.4	40.0 ± 0.4	45.0 ± 0.4	44.5 ± 0.5	38.1 ± 0.6	40.8 ± 0.4
Р	0.3 ± 0.0	0.7 ± 0.1	< 0.1	0.5 ± 0.0	0.7 ± 0.0	< 0.1	< 0.1	0.3 ± 0.0	1.1 ± 0.1	0.3 ± 0.0	1.0 ± 0.1	0.4 ± 0.0	0.4 ± 0.0	< 0.1
Pb	< 0.2	< 0.2	< 0.2	< 0.2	< 0.2	< 0.2	< 0.2	< 0.2	< 0.2	< 0.2	< 0.2	1.7 ± 0.3	< 0.2	< 0.2
S	5.2 ± 0.1	2.3 ± 0.1	4.4 ± 0.1	5.9 ± 0.1	3.4 ± 0.1	1.1 ± 0.1	3.4 ± 0.1	3.0 ± 0.1	4.9 ± 0.1	2.6 ± 0.1	4.3 ± 0.1	4.0 ± 0.1	4.1 ± 0.1	4.6 ± 0.1
Sb	< 0.2	< 0.2	< 0.2	< 0.2	< 0.2	< 0.2	< 0.2	< 0.2	< 0.2	< 0.2	< 0.2	< 0.2	< 0.2	< 0.2
Si	3.1 ± 0.1	3.6 ± 0.1	1.3 ± 0.1	1.4 ± 0.1	5.8 ± 0.1	0.6 ± 0.0	2.9 ± 0.1	2.4 ± 0.1	1.9 ± 0.1	2.1 ± 0.1	2.3 ± 0.1	20.1 ± 0.2	4.0 ± 0.1	1.0 ± 0.1
Zn	0.6 ± 0.1	0.6 ± 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	1.0 ± 0.2	< 0.1	1.1 ± 0.1	< 0.1	1.2 ± 0.1	2.2 ± 0.2	< 0.1
Total	100	100	100	100	100	100	100	100	100	100	100	100	100	100

Appendix VII - Elemental composition of APCr analysed by SEM/EDS

Part B

Elemental concentrations are expressed in wt.%. The \pm values correspond to one standard deviation of the mean value, calculated by the software.

Element	APCr-30	APCr-31	APCr-32	APCr-33	APCr-34	APCr-35	APCr-36	APCr-37	APCr-38	APCr-39	APCr-40	APCr-41	APCr-42
Al	9.2 ± 0.1	< 0.1	0.6 ± 0.1	0.8 ± 0.1	0.9 ± 0.1	3.6 ± 0.1	1.0 ± 0.1	2.0 ± 0.1	5.2 ± 0.1	6.9 ± 0.1	0.7 ± 0.1	4.8 ± 0.1	6.3 ± 0.1
As	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1
Ba	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	1.5 ± 0.2	< 0.1
Ca	12.2 ± 0.1	42.9 ± 0.3	39.9 ± 0.3	32.4 ± 0.2	36.8 ± 0.3	24.7 ± 0.2	29.7 ± 0.2	29.1 ± 0.2	33.7 ± 0.2	25.2 ± 0.2	30.5 ± 0.2	33.2 ± 0.4	31.4 ± 0.2
Cd	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1
Cl	1.1 ± 0.1	12.1 ± 0.1	13.7 ± 0.1	16.8 ± 0.2	9.9 ± 0.1	4.7 ± 0.1	13.5 ± 0.1	12.8 ± 0.1	7.8 ± 0.1	9.2 ± 0.1	16.0 ± 0.2	9.3 ± 0.1	10.6 ± 0.1
Со	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1
Cr	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1
Cu	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	0.5 ± 0.1	< 0.1
F	<0.5	<0.5	< 0.5	<0.5	< 0.5	< 0.5	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5
Fe	3.6 ± 0.1	< 0.1	< 0.1	< 0.1	< 0.1	1.3 ± 0.1	0.4 ± 0.1	1.1 ± 0.1	1.3 ± 0.1	1.2 ± 0.1	0.4 ± 0.1	1.2 ± 0.1	0.7 ± 0.1
K	1.0 ± 0.1	< 0.1	1.0 ± 0.1	2.6 ± 0.1	0.9 ± 0.1	2.0 ± 0.1	1.0 ± 0.1	1.4 ± 0.1	0.9 ± 0.1	1.4 ± 0.1	0.5 ± 0.1	0.9 ± 0.1	0.8 ± 0.1
Mg	4.4 ± 0.1	< 0.1	0.3 ± 0.1	0.4 ± 0.1	0.3 ± 0.1	1.9 ± 0.1	0.4 ± 0.1	0.5 ± 0.1	1.3 ± 0.1	1.3 ± 0.1	0.3 ± 0.1	1.2 ± 0.1	0.8 ± 0.1
Mn	0.5 ± 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1
Мо	<0.3	< 0.3	< 0.3	< 0.3	< 0.3	<0.3	< 0.3	< 0.3	< 0.3	<0.3	<0.3	<0.3	< 0.3
Ν	<3.7	<3.7	<3.7	<3.7	<3.7	<3.7	<3.7	<3.7	<3.7	<3.7	<3.7	<3.7	<3.7
Na	3.0 ± 0.1	1.1 ± 0.1	0.8 ± 0.1	1.5 ± 0.1	0.5 ± 0.1	1.1 ± 0.1	1.5 ± 0.1	1.2 ± 0.1	0.9 ± 0.1	1.5 ± 0.1	0.4 ± 0.1	0.9 ± 0.1	0.6 ± 0.1
Ni	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1
0	41.6 ± 0.3	42.9 ± 0.4	38.6 ± 0.4	38.2 ± 0.4	42.2 ± 0.4	45.8 ± 0.3	46.1 ± 0.4	42.1 ± 0.4	40.8 ± 0.4	43.5 ± 0.3	43.3 ± 0.4	36.2 ± 0.7	39.0 ± 0.4
Р	< 0.1	< 0.1	0.3 ± 0.1	< 0.1	0.2 ± 0.0	1.0 ± 0.1	0.3 ± 0.1	0.5 ± 0.0	0.5 ± 0.1	0.5 ± 0.0	0.3 ± 0.0	0.5 ± 0.1	0.7 ± 0.1
Pb	< 0.2	<0.2	< 0.2	<0.2	1.3 ± 0.3	<0.2	<0.2	<0.2	<0.2	< 0.2	< 0.2	<0.2	<0.2
S	1.0 ± 0.1	0.5 ± 0.0	3.1 ± 0.1	3.6 ± 0.1	3.4 ± 0.1	4.7 ± 0.1	2.7 ± 0.1	3.2 ± 0.1	2.3 ± 0.1	1.9 ± 0.1	3.9 ± 0.1	2.0 ± 0.1	3.4 ± 0.1
Sb	<0.2	<0.2	< 0.2	< 0.2	< 0.2	<0.2	<0.2	<0.2	<0.2	< 0.2	< 0.2	<0.2	<0.2
Si	22.3 ± 0.2	0.5 ± 0.0	1.3 ± 0.1	2.3 ± 0.1	1.8 ± 0.1	8.0 ± 0.1	2.9 ± 0.1	5.0 ± 0.1	5.3 ± 0.1	7.4 ± 0.1	3.0 ± 0.1	7.1 ± 0.1	4.9 ± 0.1
Zn	< 0.1	< 0.1	0.5 ± 0.2	1.3 ± 0.2	1.8 ± 0.2	1.2 ± 0.2	0.6 ± 0.1	1.1 ± 0.2	< 0.1	< 0.1	0.9 ± 0.2	0.7 ± 0.2	0.7 ± 0.2
Total	100	100	100	100	100	100	100	100	100	100	100	100	100

Appendix VII - Elemental composition of APCr analysed by SEM/EDS

Part C

Elemental concentrations are expressed in wt.%. The ± values correspond to one standard deviation of the mean value, calculated by the software.

Appendix VIII - Content of mineral phases and amorphous material in APCr

Part A

	APCr-01	APCr-02	APCr-03	APCr-04	APCr-05	APCr-06	APCr-07	APCr-08	APCr-09	APCr-10	APCr-11	APCr-12	APCr-13	APCr-14
Akermanite		7.90 ± 0.27							6.11 ± 0.40					
Albite	0.30 ± 0.05					6.54 ± 1.78								
Anatase		1.89 ± 0.33	0.16 ± 0.65							1.00 ± 0.10				
Anglesite			0.56 ± 0.24						0.14 ± 0.14					
Anhydrite				3.99 ± 1.05	2.47 ± 0.31	$6.44 \hspace{0.2cm} \pm \hspace{0.2cm} 0.49$	14.34 ± 0.92				14.59 ± 0.68		3.08 ± 0.14	2.50 ± 0.20
Anorthite														
Arcanite								7.59 ± 0.46						
Bassanite			9.12 ± 0.76						10.59 ± 0.51				6.78 ± 0.21	9.34 ± 0.42
Bottalockite										9.00 ± 1.27		0.02 ± 0.01		
Calcite	7.38 ± 0.77	3.54 ± 0.25	9.04 ± 0.68	6.84 ± 0.40			11.51 ± 0.48	$13.48 \ \pm \ 0.33$	1.34 ± 0.67	$4.22 \hspace{0.2cm} \pm \hspace{0.2cm} 0.35$	$10.48 \ \pm \ 0.46$	3.01 ± 0.21		8.09 ± 0.41
Calcium Hydroxychloride	5.74 ± 0.70	8.72 ± 0.20		9.98 ± 0.43	35.97 ± 0.86	$12.70 \hspace{0.2cm} \pm \hspace{0.2cm} 0.48$	4.52 ± 0.28			$27.81 \ \pm \ 0.82$		$32.90 \ \pm \ 0.58$	2.47 ± 0.09	
Cerussite														
Chlorapatite										7.52 ± 0.70				
Clinotobermorite	$0.37 \hspace{0.2cm} \pm \hspace{0.2cm} 0.05$			6.84 ± 1.24			6.73 ± 0.80					3.01 ± 0.31	3.64 ± 0.21	$13.39\ \pm\ 1.18$
Dipotassium oxide			0.97 ± 0.24											
Ettringite												9.55 ± 0.63		
Gehlenite			6.78 ± 0.98											
Grossite														
Gypsum														0.39 ± 0.58
Halite	3.28 ± 0.26	1.55 ± 0.09	2.34 ± 0.25	5.99 ± 0.22	3.69 ± 0.15		0.89 ± 0.18	2.95 ± 0.27		3.78 ± 0.14			1.67 ± 0.08	4.99 ± 0.18
Hematite				0.36 ± 0.10										
Kumdykolite		0.99 ± 0.25						0.80 ± 1.16	4.20 ± 0.96					
Larnite							1.20 ± 0.07		6.30 ± 0.40					
Lime				0.51 ± 0.86			0.44 ± 0.18							
Litharge					0.11 ± 0.00			0.82 ± 0.11						
Magnesium oxide hydroxide										0.39 ± 0.09	2.06 ± 0.21	0.23 ± 0.01	1.33 ± 0.21	
Monteponite		0.07 ± 0.02							0.21 ± 0.02					
Nickel ferrite									0.90 ± 0.11					
Periclase														
Perovskite														
Phosphorous pentoxide										1.64 ± 0.11				
Portlandite	2.78 ± 0.63		3.87 ± 0.33	10.74 ± 0.36							5.15 ± 0.22	7.24 ± 0.22	4.69 ± 0.12	9.15 ± 0.25
Potassium tetrachloro zincate	0.28 ± 0.10	2.71 ± 0.91		2.19 ± 0.38		0.56 ± 0.28						0.47 ± 0.14		
Pseudowollastonite									0.86 ± 0.19					
Quartz	0.16 ± 0.04	9.79 ± 0.28	0.78 ± 0.12	0.57 ± 0.19			4.90 ± 0.18	$19.46 \ \pm \ 0.46$	12.98 ± 0.45		3.08 ± 0.31		0.65 ± 0.07	2.41 ± 0.29
Rutile														
Sanidine						7.94 ± 0.95					1.75 ± 0.31	0.21 ± 0.62		
Sylvite		2.23 ± 0.09	3.95 ± 0.26	4.13 ± 0.16		4.26 ± 0.20	1.12 ± 0.06	0.87 ± 0.10	3.31 ± 0.15		2.72 ± 0.14		2.59 ± 0.09	3.11 ± 0.13
Tetrapotassium lead oxide														
Tobermorite					16.55 ± 1.36	11.48 ± 0.71		0.50 ± 0.06		3.42 ± 1.48	15.62 ± 1.47			
Whitlockite										5.24 ± 0.47				
Zinc chloride	0.06 ± 0.19									6.61 ± 0.81	1.61 ± 0.14	4.25 ± 0.42		
Zinc oxysulphate														
Zincite			0.40 ± 0.10			0.49 ± 0.02								
Zincowoodwardite					1.13 ± 0.72				5.82 ± 0.87	0.26 ± 0.14	2.98 ± 0.42	0.03 ± 0.01	0.10 ± 0.10	
% Crystalline Material	20.37 ± 1.27	39.39 ± 1.12	37.99 ± 1.67	52.14 ± 2.03	59.91 ± 1.80	50.40 ± 2.27	45.65 ± 1.38	46.47 ± 1.41	52.76 ± 1.73	70.91 ± 2.46	60.05 ± 1.82	60.92 ± 1.23	27.00 ± 0.45	53.36 ± 1.52
% Amorphous Material	79.63 ± 1.27	60.61 ± 1.12	62.01 ± 1.67	47.86 ± 2.03	40.09 ± 1.80	49.60 ± 2.27	54.35 ± 1.38	53.53 ± 1.41	47.24 ± 1.73	29.09 ± 2.46	39.95 ± 1.82	39.08 ± 1.23	73.00 ± 0.45	46.64 ± 1.52
No. of mineral phases	9	10	11	11	6	8	9	8	12	12	10	11	10	9

Mineral phases concentrations are expressed in wt%. The uncertainty of each value is given as one standard deviation.

The standard deviation of the mean value was calculated according to the propagated error.

Appendix VIII - Content of mineral phases and amorphous material in APCr

Part B

	APCr-15	APCr-16	APCr-17	APCr-18	APCr-19	APCr-20	APCr-21	APCr-22	APCr-23	APCr-24	APCr-25	APCr-26	APCr-27	APCr-28
Akermanite														
Albite														
Anatase					0.83 ± 0.83	2.48 ± 0.19					1.12 ± 0.17		0.13 ± 0.13	
Anglesite														
Anhydrite	1.96 ± 0.26	3.44 ± 0.29			1.24 ± 0.31	5.15 ± 0.46		4.89 ± 1.64	2.98 ± 0.13			0.13 ± 0.05	6.31 ± 0.14	9.98 ± 0.31
Anorthite														
Arcanite					0.41 ± 0.52					3.94 ± 0.17		2.81 ± 0.42		
Bassanite	7.06 ± 0.44	6.97 ± 0.31		16.69 ± 0.49								1.26 ± 0.17	4.47 ± 0.20	
Bottalockite		0.23 ± 0.02												
Calcite	15.73 ± 0.57	11.55 ± 0.42	$18.33 \ \pm \ 0.46$	3.77 ± 0.72	5.06 ± 0.32	5.04 ± 0.68	12.53 ± 0.48	7.82 ± 0.69	9.42 ± 0.24	3.59 ± 0.28	5.24 ± 0.30		1.45 ± 0.20	4.53 ± 0.19
Calcium Hydroxychloride						27.64 ± 0.89	11.41 ± 0.32	9.78 ± 0.43	22.18 ± 0.29	4.21 ± 0.15	17.75 ± 0.24	1.48 ± 0.18		3.19 ± 0.26
Cerussite	0.94 ± 0.09						1.37 ± 0.07							
Chlorapatite	2.13 ± 0.43													
Clinotobermorite				4.78 ± 1.22				3.18 ± 0.90						
Dipotassium oxide	0.82 ± 0.15													
Ettringite														
Gehlenite	7.31 ± 0.86													
Grossite		3.05 ± 0.20												
Gypsum			1.75 ± 0.39											
Halite		2.90 ± 0.12	3.52 ± 0.13			14.99 ± 0.55	8.29 ± 0.29	2.69 ± 0.18	6.39 ± 0.92		5.76 ± 0.13		5.63 ± 0.10	4.28 ± 0.13
Hematite											1.31 ± 0.50			
Kumdykolite		1.43 ± 0.19		2.85 ± 0.31							0.20 ± 0.20			
Larnite														
Lime									0.56 ± 0.15					
Litharge												0.07 ± 0.02		
Magnesium oxide hydroxide		3.91 ± 0.48	8.34 ± 0.69					3.91 ± 0.74						0.01 ± 0.00
Monteponite														
Nickel ferrite														
Periclase														
Perovskite			0.10 ± 0.02						13.79 ± 0.59					
Phosphorous pentoxide														
Portlandite		3.72 ± 0.14	12.31 ± 0.26	27.89 ± 0.69	30.36 ± 0.68				11.71 ± 0.20	19.16 ± 0.19	23.39 ± 0.26	5.17 ± 0.63		
Potassium tetrachloro zincate												0.18 ± 0.05		1.97 ± 0.16
Pseudowollastonite	2.13 ± 0.34													
Quartz	4.15 ± 0.16	2.12 ± 0.10	2.73 ± 0.15	2.85 ± 0.61					$0.31 \hspace{0.2cm} \pm \hspace{0.2cm} 0.01$		0.05 ± 0.01			2.30 ± 0.16
Rutile												13.60 ± 1.69		
Sanidine														
Sylvite		3.46 ± 0.13	1.14 ± 0.08		22.62 ± 0.53	8.62 ± 0.38		2.93 ± 0.66	4.61 ± 0.09	20.69 ± 0.16	3.23 ± 0.09	6.65 ± 0.87	4.14 ± 0.07	2.31 ± 0.09
Tetrapotassium lead oxide		0.66 ± 0.14												
Tobermorite		7.54 ± 1.15												
Whitlockite							6.39 ± 0.53							12.24 ± 1.19
Zinc chloride			5.82 ± 0.88				0.94 ± 0.16						0.29 ± 0.05	
Zinc oxysulphate													0.42 ± 0.04	
Zincite														
Zincowoodwardite		1.46 ± 0.10				5.15 ± 1.68	2.59 ± 1.04	3.34 ± 0.34						0.19 ± 1.19
% Crystalline Material	42.22 ± 1.29	52.44 ± 1.44	54.04 ± 1.31	58.84 ± 1.79	60.52 ± 1.37	69.06 ± 2.18	43.52 ± 1.34	38.54 ± 2.30	71.93 ± 1.19	51.59 ± 0.44	58.05 ± 0.75	31.34 ± 2.06	22.84 ± 0.37	41.00 ± 1.30
% Amorphous Material	57.78 ± 1.29	47.56 ± 1.44	45.96 ± 1.31	41.16 ± 1.79	39.48 ± 1.37	30.94 ± 2.18	56.48 ± 1.34	61.46 ± 2.30	28.07 ± 1.19	48.41 ± 0.44	41.95 ± 0.75	68.66 ± 2.06	77.16 ± 0.37	59.00 ± 1.30
No. of mineral phases	9	14	9	6	6	7	7	8	9	5	9	9	8	10

Mineral phases concentrations are expressed in wt%. The uncertainty of each value is given as one standard deviation.

Appendix VIII - Content of mineral phases and amorphous material in APCr

Part C

	APCr-29	APCr-30	APCr-31	APCr-32	APCr-33	APCr-34	APCr-35	APCr-36	APCr-37	APCr-38	APCr-39	APCr-40	APCr-41	APCr-42
Akermanite		3.55 ± 0.12					4.59 ± 0.52			2.21 ± 0.19				
Albite														
Anatase				3.34 ± 0.84										
Anglesite														
Anhydrite	2.51 ± 0.16				1.45 ± 0.62	11.03 ± 0.81		1.96 ± 0.13	5.90 ± 0.38	2.87 ± 0.31	0.79 ± 1.58	1.71 ± 0.06	3.11 ± 0.41	4.60 ± 0.19
Anorthite		1.78 ± 0.10												
Arcanite												0.11 ± 0.02		
Bassanite							13.77 ± 0.29							
Bottalockite	0.59 ± 0.11													
Calcite		3.21 ± 0.11	9.38 ± 0.56	$6.80 \hspace{0.2cm} \pm \hspace{0.2cm} 0.26$	3.41 ± 0.21	1.61 ± 0.35	9.61 ± 0.28	3.00 ± 0.29	14.75 ± 0.67	9.84 ± 0.35	12.68 ± 0.51	1.45 ± 0.31	11.78 ± 0.27	5.52 ± 0.28
Calcium Hydroxychloride			22.39 ± 0.84	17.55 ± 0.34	34.91 ± 0.40	40.90 ± 0.64		35.02 ± 0.50	12.07 ± 0.32	11.89 ± 0.37	5.67 ± 0.28	43.48 ± 0.44	10.71 ± 0.27	10.97 ± 0.41
Cerussite								0.01 ± 0.00						
Chlorapatite											0.19 ± 0.01			
Clinotobermorite	$0.68 \hspace{0.2cm} \pm \hspace{0.2cm} 0.06$			16.71 ± 0.75							5.04 ± 0.57			
Dipotassium oxide														
Ettringite		0.11 ± 0.11	2.42 ± 0.51											
Gehlenite								0.87 ± 0.77						
Grossite														
Gypsum	6.56 ± 0.57						2.60 ± 0.52						0.12 ± 0.01	
Halite	3.96 ± 0.13	0.38 ± 0.45	2.18 ± 0.12	4.32 ± 0.96	6.67 ± 0.12	6.19 ± 0.14		6.09 ± 0.13	6.82 ± 0.18	2.60 ± 0.11	2.58 ± 0.12	6.73 ± 0.15	3.23 ± 0.10	3.11 ± 0.12
Hematite														
Kumdykolite		3.21 ± 0.17												
Larnite				21.73 ± 1.00						12.41 ± 1.15		2.48 ± 0.31		
Lime								1.45 ± 0.19						1.11 ± 0.12
Litharge														
Magnesium oxide hydroxide	1.38 ± 0.41			3.22 ± 0.24										
Monteponite														
Nickel ferrite		0.41 ± 0.23												
Periclase														0.72 ± 0.07
Perovskite										7.59 ± 1.03		0.24 ± 0.02		3.10 ± 0.26
Phosphorous pentoxide					0.83 ± 0.21									
Portlandite	16.53 ± 0.31		14.52 ± 0.55		6.53 ± 0.18	4.60 ± 0.24			5.71 ± 0.74	4.44 ± 0.20	0.87 ± 0.24	1.52 ± 0.13	9.16 ± 0.42	3.85 ± 0.43
Potassium tetrachloro zincate														
Pseudowollastonite														
Quartz							12.21 ± 0.29	0.09 ± 0.02		3.50 ± 0.18	5.51 ± 0.28	0.01 ± 0.01		3.72 ± 0.17
Rutile														
Sanidine	1.54 ± 0.32			0.01 ± 0.00	C #4				4.0.5 4.0		1 0 0 1 0 1 0		0.04	
Sylvite	4.32 ± 0.10			2.76 ± 0.72	6.54 ± 0.10	5.85 ± 0.12	0.96 ± 0.09	5.34 ± 0.11	4.05 ± 0.18	2.19 ± 0.82	1.98 ± 0.13	2.74 ± 0.20	0.91 ± 0.09	4.15 ± 0.15
Tetrapotassium lead oxide														
Tobermorite			6.05 ± 2.02			1.01 . 0.16								
Whitlockite						1.84 ± 0.46								
Zinc chloride														
Zinc oxysulphate		0.04 ± 0.00												
Zincite	0.06 + 0.01	0.02 + 0.07	1.12	0.27 + 0.12				0.08 ± 0.03			0.71 ± 0.09			
Zincowoodwardite	0.06 ± 0.01	0.83 ± 0.07	1.12 ± 0.09	0.27 ± 0.12				0.01 ± 0.00	1					
% Crystalline Material	38.12 ± 0.87	13.53 ± 0.58	58.05 ± 2.39	76.73 ± 1.99	60.32 ± 0.83	72.02 ± 1.22	43.73 ± 0.89	53.92 ± 1.01	49.30 ± 1.14	59.55 ± 1.88	36.02 ± 1.82	60.47 ± 0.69	39.01 ± 0.71	40.86 ± 0.79
% Amorphous Material	61.88 ± 0.87	86.47 ± 0.58	41.95 ± 2.39	23.27 ± 1.99	39.68 ± 0.83	27.98 ± 1.22	56.27 ± 0.89	46.08 ± 1.01	50.70 ± 1.14	40.45 ± 1.88	63.98 ± 1.82	39.53 ± 0.69	60.99 ± 0.71	59.14 ± 0.79
No. of mineral phases	10	9	7	10	7	7	6	11	6	10	10	10	7	10

Mineral phases concentrations are expressed in wt%. The uncertainty of each value is given as one standard deviation.



Appendix IX – SEM micrographs and SEM/EDS spectra of APCr samples





Foil piece with fine grained material



Hexagonal particle









Metallic particle



Needle-like particles

APCr-06









Cubes



Sphere





Prismatic particle



Laminar crushed particles







APCr-13



Tabular particle



Spheres at different sizes





Holey sphere





Spheres, carbon pieces, amorphous material





Sphere



Small spheres











Hollow cenosphere



Spheres at different sizes









Sphere particle surrounded by fine grained material



Metallic piece

















APCr-31



Spheres at different sizes





Pieces of unburnt organic matter surrounded by fine-grained material





Sphere surrounded by fine grained material



Piece of unburnt organic matter











Spheres surrounded by fine grained material



Needle-like particles aggregate









Hollow sphere







Blocks of fine material



Aggregate of grained material











Spheres at different sizes among fine grained material







Spheres among fine grained material



Fine grained material



Appendix X – Calculation of bloating index of hot-bonded LWA Part A

		100%	Clay		5% APCr-m						
Firing	D _{ini}	$\mathrm{D}_{\mathrm{fin}}$	BI	BI	%	D _{ini}	$\mathrm{D}_{\mathrm{fin}}$	BI	BI	%	
temp. °C	(mm)	(mm)	%	Ave	SD	(mm)	(mm)	%	Ave	SD	
1,100	13.101	13.253	1.2	0.9	0.3	12.508	12.690	1.5	1.8	0.8	
	12.791	12.919	1.0			13.093	13.264	1.3			
	11.804	11.925	1.0			12.279	12.658	3.1			
	11.683	11.755	0.6			12.393	12.616	1.8			
	13.310	13.372	0.5			12.671	12.827	1.2			
1,120	13.748	13.920	1.3	1.5	0.7	13.375	13.265	-0.8	-2.2	2.6	
	13.135	13.311	1.3			13.233	12.204	-7.8			
	12.859	13.180	2.5			12.965	12.901	-0.5			
	13.643	13.871	1.7			13.123	12.853	-2.1			
	12.770	12.853	0.6			13.190	13.179	-0.1			
						13.206	12.879	-2.5			
						13.039	12.780	-2.0			
1,140	12.363	12.662	2.4	1.6	0.6	13.158	13.066	-0.7	-1.1	0.7	
	12.792	12.958	1.3			12.809	12.586	-1.7			
	13.365	13.517	1.1			13.974	13.761	-1.5			
	12.962	13.151	1.5			12.895	12.886	-0.1			
						12.861	12.687	-1.4			
1,160	13.308	13.632	2.4	4.6	2.4	12.107	12.235	1.1	-0.5	1.8	
	13.193	13.615	3.2			12.481	12.545	0.5			
	12.766	13.775	7.9			12.431	12.038	-3.2			
	12.699	13.340	5.0			12.341	12.149	-1.6			
						13.092	13.149	0.4			
1,180	13.160	15.193	15.4	15.6	1.4	12.422	12.689	2.1	1.6	0.4	
	13.593	15.582	14.6			13.122	13.350	1.7			
	12.514	14.712	17.6			12.651	12.837	1.5			
	12.078	13.858	14.7			12.505	12.635	1.0			
						12.896	13.104	1.6			
1,200	13.679	17.632	28.9	32.7	6.9	12.603	14.142	12.2	13.9	4.6	
	13.410	18.444	37.5			13.424	14.750	9.9			
	14.043	17.267	23.0			13.727	15.043	9.6			
	12.972	18.161	40.0			12.726	15.151	19.1			
	12.409	16.639	34.1			12.728	15.087	18.5			
1,220	13.855	20.427	47.4	40.3	8.1						
	14.150	18.748	32.5								
	13.569	17.886	31.8								
	13.374	18.7930	40.5								
	13.535	20.184	49.1								
Appendix X – Calculation of bloating index of hot-bonded LWA Part B

		10% A	PCr-m				15% AI	PCr-m		
Firing	D _{ini}		BI	BI	%	D _{ini}		BI	BI	%
temp. °C	(mm)	$D_{\rm fin}(\rm mm)$	%	Ave	SD	(mm)	$D_{fin}(mm)$	%	Ave	SD
1,100	13.103	13.128	0.2	-1.7	2.3	13.620	13.626	0.0	0.1	1.3
	13.194	13.103	-0.7			13.499	13.251	-1.8		
	12.887	12.876	-0.1			13.527	13.517	-0.1		
	13.109	12.648	-3.5			13.677	13.657	-0.1		
	13.298	12.508	-5.9			13.516	13.800	2.1		
	13.896	13.673	-1.6			13.179	13.361	1.4		
	13.623	13.620	0.0			13.960	13.879	-0.6		
1,120	12.711	12.314	-3.1	-2.1	1.2	13.190	12.575	-4.7	-1.7	2.0
	13.065	12.576	-3.7			13.836	13.777	-0.4		
	13.344	13.298	-0.3			13.436	13.022	-3.1		
	12.900	12.543	-2.8			13.392	13.342	-0.4		
	13.506	13.340	-1.2			13.265	13.298	0.2		
	13.187	13.019	-1.3			13.027	13.021	0.0		
	13.374	13.110	-2.0			14.197	13.735	-3.3		
1,140	13.574	12.742	-6.1	-5.8	3.3	13.507	13.207	-2.2	-5.2	2.8
	12.482	12.513	0.2			13.074	12.042	-7.9		
	12.973	12.455	-4.0			12.810	12.656	-1.2		
	13.400	12.544	-6.4			12.963	11.963	-7.7		
	12.849	11.507	-10.4			13.420	12.353	-8.0		
	13.223	12.371	-6.4			13.656	13.008	-4.7		
	13.256	12.240	-7.7			13.496	12.854	-4.8		
1,160	13.440	12.231	-9.0	-8.5	1.2	13.397	12.948	-3.4	-3.1	1.5
	13.308	12.162	-8.6			12.803	12.598	-1.6		
	13.267	11.885	-10.4			13.737	13.293	-3.2		
	13.056	12.179	-6.7			12.975	12.800	-1.3		
	13.164	11.972	-9.1			13.533	12.926	-4.5		
	13.200	12.247	-7.2			13.389	13.055	-2.5		
	13.144	12.039	-8.4			13.026	12.322	-5.4		
1,180	12.788	12.175	-4.8	-5.1	1.3	13.124	12.833	-2.2	-2.3	2.2
	12.596	12.003	-4.7			13.380	13.625	1.8		
	12.858	11.962	-7.0			13.141	13.049	-0.7		
	13.820	13.004	-5.9			13.503	12.979	-3.9		
	13.687	13.327	-2.6			13.003	12.533	-3.6		
	13.326	12.586	-5.6			13.470	12.981	-3.6		
	13.022	12.352	-5.1			13.613	13.040	-4.2		

Appendix X – Calculation of bloating index of hot-bonded LWA Part C

		20% AI	PCr-m				25% AI	PCr-m		
Firing	D _{ini}	\mathbf{D} ()	BI	BI	%	D _{ini}	\mathbf{D} ()	BI	BI	%
temp. °C	(mm)	$D_{fin}(mm)$	%	Ave	SD	(mm)	$D_{fin}(mm)$	%	Ave	SD
1,100	12.950	13.204	2.0	1.0	1.7	12.564	12.601	0.3	0.3	0.6
	13.104	13.175	0.5			12.998	13.081	0.6		
	13.329	13.419	0.7			13.223	13.184	-0.3		
	12.833	13.231	3.1			13.442	13.468	0.2		
	13.061	13.245	1.4			13.352	13.519	1.3		
	13.196	12.888	-2.3			13.283	13.197	-0.6		
	13.194	13.403	1.6			13.522	13.614	0.7		
1,120	13.094	12.962	-1.0	-1.7	0.8	13.585	13.610	0.2	-0.1	1.7
	13.055	12.903	-1.2			13.039	13.155	0.9		
	12.632	12.439	-1.5			12.774	12.670	-0.8		
	13.261	12.889	-2.8			13.178	13.262	0.6		
	13.119	12.801	-2.4			13.208	13.259	0.4		
	12.814	12.543	-2.1			12.514	12.720	1.6		
	13.086	12.972	-0.9			13.381	12.921	-3.4		
1,140	13.393	13.251	-1.1	-3.8	1.3	13.567	13.590	0.2	-0.8	2.3
	13.892	13.427	-3.3			13.161	13.173	0.1		
	12.918	12.448	-3.6			12.538	12.840	2.4		
	12.744	12.159	-4.6			12.956	12.872	-0.6		
	12.949	12.438	-3.9			13.145	12.850	-2.2		
	12.555	11.968	-4.7			12.880	12.326	-4.3		
	12.915	12.270	-5.0							
1,160	13.182	12.304	-6.7	-3.9	1.9	13.745	12.906	-6.1	-3.7	3.4
	13.319	12.946	-2.8			13.267	13.092	-1.3		
	13.150	12.425	-5.5			12.990	13.363	Part	ially me	elted
	13.453	13.042	-3.1			13.315	14.293			
	13.775	13.549	-1.6			13.134	13.801			
	13.083	12.558	-4.0			12.985	13.757			
						13.286	13.508			

Appendix X – Calculation of bloating index of hot-bonded LWA Part D

		5% w-A	APCr				10% w-	APCr		
Firing	$\mathbf{D}_{\mathrm{ini}}$	D_{π} (mm)	BI	BI	%	$\mathbf{D}_{\mathrm{ini}}$	$D_{\pi}(mm)$	BI	BI	%
temp. °C	(mm)	$D_{fin}(IIIII)$	%	Ave	SD	(mm)	$D_{\rm fin}(\rm IIIIII)$	%	Ave	SD
1,140	12.231	11.965	-2.2	-0.8	1.2	11.691	11.492	-1.7	-1.6	0.7
	11.378	11.193	-1.6			11.273	10.992	-2.5		
	12.153	12.118	-0.3			12.268	12.079	-1.5		
	11.766	11.677	-0.8			11.787	11.748	-0.3		
	11.348	11.541	1.7			11.788	11.482	-2.6		
	11.986	11.828	-1.3			11.852	11.657	-1.6		
	11.682	11.635	-0.4			11.578	11.466	-1.0		
	11.779	11.594	-1.6			11.501	11.311	-1.7		
1,160	11.860	11.678	-1.5	-2.4	3.2	11.714	11.282	-3.7	-2.2	1.2
	11.751	11.785	0.3			11.604	11.378	-1.9		
	12.200	12.107	-0.8			11.320	11.043	-2.4		
	12.162	11.096	-8.8			11.652	11.601	-0.4		
	11.674	11.478	-1.7			11.246	11.164	-0.7		
	11.579	11.323	-2.2			11.769	11.545	-1.9		
						11.719	11.359	-3.1		
						11.268	10.899	-3.3		
1,180	11.692	11.321	-3.2	-2.0	1.4	11.858	11.242	-5.2	-3.1	1.8
	11.709	11.392	-2.7			11.343	11.075	-2.4		
	12.116	11.715	-3.3			11.808	11.451	-3.0		
	11.556	11.491	-0.6			11.825	11.632	-1.6		
	11.094	10.829	-2.4			12.076	11.754	-2.7		
	11.355	11.451	0.8			12.086	11.875	-1.7		
	11.323	11.135	-1.7			11.744	11.543	-1.7		
	11.657	11.348	-2.7			11.320	10.595	-6.4		

Appendix X – Calculation of bloating index of hot-bonded LWA Part E

		15% w-	APCr				20% w	-APCr		
Firing	D _{ini}	D. (mm)	DI 0/	BI	%	D _{ini}	D. (mm)	DI 0/	BI	%
temp. ^o C	(mm)	$D_{\rm fin}(\rm mm)$	BI %	Ave	SD	(mm)	$D_{fin}(mm)$	BI %0	Ave	SD
1,140	11.953	11.247	-5.9	-2.3	2.1	11.479	10.637	-7.3	-3.8	2.3
	11.248	10.871	-3.4			12.027	11.711	-2.6		
	10.746	10.715	-0.3			11.085	10.704	-3.4		
	11.724	11.608	-1.0			11.176	10.926	-2.2		
	11.353	11.274	-0.7			11.389	11.229	-1.4		
	11.323	11.270	-0.5			11.659	11.249	-3.5		
	11.032	10.842	-1.7			11.789	11.483	-2.6		
	11.945	11.380	-4.7			11.725	10.866	-7.3		
1,160	11.745	10.512	-10.5	-7.2	3.1	11.658	10.253	-12.1	-8.8	2.2
	11.412	10.351	-9.3			11.317	10.229	-9.6		
	11.273	10.468	-7.1			11.776	10.906	-7.4		
	11.695	10.368	-11.3			11.761	10.788	-8.3		
	11.922	11.427	-4.2			11.757	11.015	-6.3		
	12.284	11.935	-2.8			11.351	10.395	-8.4		
	11.544	10.962	-5.0			11.778	11.008	-6.5		
	11.404	10.566	-7.3			11.281	9.974	-11.6		
1,180	11.657	10.692	-8.3	-7.5	2.0	11.456	10.378	-9.4	-8.9	2.2
	10.797	9.737	-9.8			11.812	10.665	-9.7		
	10.942	10.144	-7.3			11.229	10.628	-5.4		
	10.809	10.198	-5.7			11.604	10.413	-10.3		
	11.395	10.727	-5.9			11.405	10.356	-9.2		
	10.857	10.289	-5.2			11.613	10.423	-10.2		
	11.160	9.954	-10.8			11.363	10.061	-11.5		
	10.957	10.188	-7.0			11.119	10.474	-5.8		

Appendix X – Calculation of bloating index of hot-bonded LWA Part F

		25% w-	APCr				30% w-	APCr		
Firing	D _{ini}	\mathbf{D} ()		BI	%	D _{ini}	\mathbf{D} ()	DI 0/	BI	%
temp. ^o C	(mm)	$D_{fin}(mm)$	BI %	Ave	SD	(mm)	$D_{fin}(mm)$	BI %	Ave	SD
1,140	12.265	11.322	-7.7	-3.2	2.5	13.051	12.529	-4.0	-2.2	1.1
	12.372	12.234	-1.1			12.123	11.750	-3.1		
	11.732	11.508	-1.9			11.979	11.852	-1.1		
	12.166	11.725	-3.6			12.369	11.996	-3.0		
	11.311	11.079	-2.1			12.211	12.004	-1.7		
	11.826	11.756	-0.6			11.965	11.775	-1.6		
	12.321	11.995	-2.6			11.920	11.653	-2.2		
	12.182	11.423	-6.2			11.352	11.258	-0.8		
1,160	12.425	11.185	-10.0	-5.7	2.5	11.329	10.771	-4.9	-3.2	1.2
	11.971	11.035	-7.8			11.845	11.648	-1.7		
	11.831	11.562	-2.3			12.301	12.017	-2.3		
	12.270	11.888	-3.1			11.643	11.326	-2.7		
	12.330	11.571	-6.2			12.119	11.788	-2.7		
	12.287	11.696	-4.8			12.303	11.907	-3.2		
	12.140	11.426	-5.9			12.463	11.873	-4.7		
	11.215	10.589	-5.6							
1,180	11.942	11.305	-5.3	-7.5	3.9	11.984	11.969	-0.1	-5.6	2.4
	11.928	11.336	-5.0			12.443	11.747	-5.6		
	11.383	10.822	-4.9			11.788	11.035	-6.4		
	11.227	11.014	-1.9			12.179	11.414	-6.3		
	12.154	11.242	-7.5			11.850	11.264	-4.9		
	12.448	11.164	-10.3			12.122	11.378	-6.1		
	12.394	10.704	-13.6			11.596	10.778	-7.1		
	12.363	10.967	-11.3			12.290	11.277	-8.2		

T T T T	Firing	W pellet in	W basket in	Wpellet+basket	W saturated	Oven dri	ed Particle der	nsity	Saturated a	nd surface-di	ried	Appare	ent particle den	sity	Wate	absorption	%
LWA	temp	aır (g)	water (g)	in water (g)	pellet (g)		(g/cm ³)	GD	Particle of	lensity (g/cm	²)		(g/cm ³)	a D			(D)
	°C	d	С	b	а		Average	SD		Average	SD		Average	SD		Average	SD
100%	1,100	1.6592	3.5692	4.3667	1.8187	1.62	1.60	0.02	1.78	1.75	0.03	1.93	1.89	0.03	9.61	9.73	0.13
Clay		1.9567	3.5692	4.4850	2.1498	1.59			1.74			1.88			9.87		
		2.1875	3.5692	4.5845	2.4000	1.58			1.73			1.87			9.71		
	1,120	1.8228	3.5692	4.4435	2.0012	1.62	1.62	0.01	1.78	1.78	0.00	1.92	1.92	0.00	9.79	9.58	0.19
		1.8683	3.5692	4.4670	2.0465	1.63			1.78			1.93			9.54		
		1.9621	3.5692	4.5110	2.1466	1.63			1.78			1.92			9.40		
	1,140	1.6966	3.5692	4.3753	1.8527	1.62	1.61	0.01	1.77	1.76	0.01	1.91	1.90	0.01	9.20	9.34	0.16
		1.8460	3.5692	4.4346	2.0215	1.60			1.75			1.88			9.51		
		1.8664	3.5692	4.4578	2.0402	1.62			1.77			1.91			9.31		
	1,160	1.6557	3.5692	4.3185	1.8243	1.54	1.47	0.06	1.70	1.62	0.07	1.83	1.73	0.08	10.18	10.42	0.23
		2.1103	3.5692	4.4320	2.3348	1.43			1.59			1.69			10.64		
		1.9228	3.5692	4.3446	2.1238	1.43			1.58			1.68			10.45		
	1,180	2.1447	3.5692	3.7785	2.3588	1.00	1.02	0.02	1.10	1.12	0.02	1.11	1.13	0.02	9.98	9.88	0.58
		2.0458	3.5692	3.8248	2.2352	1.03			1.13			1.14			9.26		
		2.0219	3.5692	3.8260	2.2323	1.02			1.13			1.15			10.41		
	1,200	1.9238	3.5692	4.1585	2.0710	1.30	1.18	0.18	1.40	1.26	0.19	1.44	1.29	0.21	7.65	7.44	0.36
		1.6548	3.5692	3.6493	1.7812	0.97			1.05			1.05			7.64		
		1.9151	3.5692	4.0980	2.0497	1.26			1.35			1.38			7.03		
5% APCr-	1,100	1.8098	3.5692	4.3878	1.9670	1.58	1.62	0.04	1.71	1.78	0.06	1.83	1.94	0.10	8.69	10.15	1.28
m		1.7449	3.5692	4.4417	1.9324	1.65			1.82			2.00			10.75		
		1.6528	3.5692	4.3934	1.8351	1.63			1.82			1.99			11.03		
	1,120	1.7798	3.5692	4.5211	1.9707	1.75	1.72	0.02	1.93	1.94	0.02	2.15	2.19	0.06	10.73	12.26	1.67
		1.8739	3.5692	4.6150	2.1371	1.72			1.96			2.26			14.05		
		1.9264	3.5692	4.6007	2.1575	1.71			1.92			2.15			12.00		
	1,140	1.7560	3.5692	4.5241	1.9509	1.76	1.77	0.05	1.96	1.96	0.02	2.19	2.18	0.02	11.10	10.58	2.01
	,	1.6396	3.5692	4.4568	1.8410	1.72			1.93			2.18			12.28		
		1.5061	3.5692	4.3759	1.6321	1.82			1.98			2.15			8.37		
	1.160	1.5638	3.5692	4.3653	1.6269	1.88	1.88	0.01	1.96	1.97	0.01	2.04	2.06	0.03	4.04	4.53	1.05
	,	1.6032	3.5692	4,4063	1.6951	1.87			1.98			2.09			5.73		
		1.6033	3.5692	4.3886	1.6645	1.90			1.97			2.05			3.82		
	1,180	1.7285	3.5692	4.2643	1.7564	1.63	1.63	0.02	1.65	1.66	0.02	1.67	1.68	0.02	1.61	1.57	0.11
	-,	1.8815	3.5692	4.3391	1.9087	1.65			1.68			1.69			1.45		
		1.7368	3.5692	4.2599	1.7653	1.62			1.64			1.66			1.64		
	1.200	1.7820	3.5692	4.0375	1.8152	1.32	1.34	0.05	1.35	1.36	0.05	1.36	1.37	0.05	1.86	1.59	0.26
	1,200	1.6837	3.5692	3.9768	1.7100	1.29	1.5 .	5.00	1.31	1.00	0.00	1.32	1.0 /	0.00	1.56	1.07	0.20
		1.6650	3 5692	4 0645	1 6873	1.29			1.51			1.32			1 34		
		1.0050	5.5072	UU-J	1.0075	1.10			1.74			1.72			1.57		

Appendix XI – Calculation of particle density and water absorption of hot-bonded LWA by the method of the wired basket (BS EN 1097-6, 2022) Part A

LWA	Firing temp	W pellet in air (g)	W basket in water (g)	Wpellet+basket in water (g)	W saturated pellet (g)	Over der	n dried Partionsity (g/cm ³)	cle)	Saturated Particle	and surface- density (g/c	-dried m ³)	Apparen	nt particle de (g/cm ³)	ensity	Wate	r absorptior	1 %
	Ĵ	d	С	b	а		Average	SD		Average	SD		Average	SD		Average	SD
10%		1.6505	3.5690	4.4655	1.8995	1.65	1.62	0.02	1.89	1.88	0.01	2.19	2.19	0.00	15.09	15.91	0.94
APCr-m	1,100	2.0316	3.5690	4.6705	2.3507	1.63			1.88			2.18			15.71		
		1.6870	3.5690	4.4864	1.9728	1.60			1.87			2.19			16.94		
		1.6747	3.5690	4.4740	1.9205	1.65	1.69	0.04	1.89	1.92	0.02	2.18	2.18	0.02	14.68	13.20	1.39
	1,120	1.9329	3.5690	4.6258	2.1843	1.71			1.94			2.21			13.01		
		1.9404	3.5690	4.6110	2.1718	1.72			1.92			2.16			11.93		
		1.7574	3.5690	4.5013	1.8153	1.99	1.96	0.09	2.06	2.05	0.05	2.13	2.15	0.02	3.29	4.65	2.24
	1,140	1.8449	3.5690	4.5637	1.9080	2.02			2.09			2.17			3.42		
		1.7206	3.5690	4.4873	1.8451	1.86			1.99			2.14			7.24		
		1.7596	3.5690	4.4819	1.7806	2.03	2.05	0.02	2.05	2.08	0.02	2.08	2.10	0.02	1.19	1.19	0.01
	1,160	1.9066	3.5690	4.5794	1.9289	2.08			2.10			2.13			1.17		
		1.7564	3.5690	4.4905	1.7774	2.05			2.08			2.10			1.20		
		1.8063	3.5690	4.4708	1.8287	1.95	1.98	0.03	1.97	2.00	0.02	2.00	2.03	0.02	1.24	1.18	0.08
	1,180	1.7218	3.5690	4.4474	1.7406	2.00			2.02			2.04			1.09		
		1.9391	3.5690	4.5564	1.9625	1.99			2.01			2.04			1.21		
15%		1.7179	3.5690	4.4626	2.0548	1.48	1.49	0.02	1.77	1.77	0.01	2.08	2.07	0.01	19.61	18.97	0.83
APCr-m	1,100	1.6200	3.5690	4.4060	1.9321	1.48			1.76			2.07			19.27		
		1.6457	3.5690	4.4193	1.9426	1.51			1.78			2.07			18.04		
		1.6845	3.5690	4.4067	1.9137	1.57	1.61	0.07	1.78	1.82	0.04	1.99	2.03	0.04	13.61	12.73	2.88
	1,120	1.7637	3.5690	4.4616	1.9316	1.70			1.86			2.02			9.52		
		1.7462	3.5690	4.4723	2.0095	1.58			1.82			2.07			15.08		
		1.7906	3.5690	4.5202	1.8366	2.02	1.91	0.13	2.07	1.97	0.11	2.13	2.04	0.10	2.57	3.53	1.41
	1,140	1.7222	3.5690	4.4508	1.7715	1.94			1.99			2.05			2.86		
		1.7691	3.5690	4.4249	1.8602	1.76			1.85			1.94			5.15		
		1.7796	3.5690	4.4204	1.8020	1.87	1.89	0.03	1.90	1.91	0.03	1.92	1.94	0.04	1.26	1.30	0.11
	1,160	1.8462	3.5690	4.4513	1.8687	1.87			1.89			1.92			1.22		
		1.5564	3.5690	4.3396	1.5787	1.93			1.95			1.98			1.43		
		1.7495	3.5690	4.3523	1.7680	1.78	1.90	0.10	1.80	1.92	0.11	1.81	1.93	0.11	1.06	1.05	0.03
	1,180	2.1747	3.5690	4.6641	2.1981	1.97			1.99			2.01			1.08		
		1.8941	3.5690	4.5050	1.9134	1.94			1.96			1.98			1.02		

Appendix XI - Calculation of particle density and water absorption of hot-bonded LWA by the method of the wired basket (BS EN 1097-6, 2022) Part B

LWA	Firing temp °C	W pellet in air (g)	W basket in water (g)	Wpellet+basket in water (g)	W saturated pellet (g)	Over der	n dried Parti nsity (g/cm ³	cle)	Satura dried	ated and surf Particle den (g/cm ³)	àce- sity	Appare	nt particle d (g/cm ³)	ensity	Water	absorption	%
		d	С	b	а		Average	SD		Average	SD		Average	SD		Average	SD
20%	1,100	1.6455	3.5690	4.4248	2.0415	1.39	1.39	0.01	1.72	1.71	0.02	2.08	2.06	0.04	24.07	23.54	0.68
APCr-m		1.6269	3.5690	4.3875	1.9974	1.38			1.69			2.01			22.77		
		1.6453	3.5690	4.4254	2.0364	1.39			1.73			2.09			23.77		
	1,120	1.5412	3.5690	4.2895	1.7482	1.50	1.49	0.01	1.70	1.70	0.02	1.88	1.89	0.05	13.43	14.25	1.44
		1.5282	3.5690	4.2741	1.7331	1.49			1.69			1.86			13.41		
		1.4342	3.5690	4.2663	1.6624	1.49			1.72			1.95			15.91		
	1,140	1.8938	3.5690	4.4402	1.9963	1.68	1.70	0.10	1.77	1.78	0.08	1.85	1.86	0.06	5.41	5.03	1.72
		1.4704	3.5690	4.2726	1.5168	1.81			1.87			1.92			3.16		
		1.7332	3.5690	4.3382	1.8464	1.61			1.71			1.80			6.53		
	1,160	1.6174	3.5690	4.3402	1.6368	1.87	1.92	0.05	1.89	1.94	0.05	1.91	1.96	0.05	1.20	1.16	0.23
		1.6008	3.5690	4.3689	1.6155	1.96			1.98			2.00			0.92		
		1.6094	3.5690	4.3642	1.6315	1.92			1.95			1.98			1.37		
25%	1,100	1.5684	3.5690	4.3929	1.9715	1.37	1.37	0.01	1.72	1.72	0.00	2.11	2.09	0.03	25.70	24.86	1.08
APCr-m		1.6183	3.5690	4.4150	2.0265	1.37			1.72			2.10			25.22		
		1.6015	3.5690	4.3918	1.9801	1.38			1.71			2.06			23.64		
	1,120	1.5771	3.5690	4.2505	1.8137	1.39	1.39	0.01	1.60	1.61	0.01	1.76	1.79	0.03	15.00	16.28	1.40
		1.3873	3.5690	4.1977	1.6340	1.38			1.63			1.83			17.78		
		1.5535	3.5690	4.2538	1.8027	1.39			1.61			1.79			16.04		
	1,140	1.5417	3.5690	4.2382	1.5866	1.68	1.59	0.13	1.73	1.69	0.05	1.77	1.77	0.00	2.91	3.72	1.14
		1.5734	3.5690	4.2503	1.6445	1.63			1.71			1.76			4.52		
		1.6593	3.5690	4.2882	1.8690	1.44			1.63			1.77			12.64		
	1,160	1.6233	3.5690	4.2749	1.6365	1.74	1.71	0.08	1.76	1.73	0.09	1.77	1.75	0.10	0.81	1.45	0.88
		1.6047	3.5690	4.1993	1.6220	1.62			1.64			1.65			1.08		
		1.6640	3.5690	4.3323	1.7049	1.77			1.81			1.85			2.46		

Appendix XI - Calculation of particle density and water absorption of hot-bonded LWA by the method of the wired basket (BS EN 1097-6, 2022) Part C

Shaded values were refused

LWA	Firing temp °C	W pellet in air (g)	W basket in water (g)	Wpellet+basket in water (g)	W saturated pellet (g)	Ove: de	n dried Parti nsity (g/cm ³	cle)	Satura dried	ated and surf Particle den (g/cm ³)	ace- sity	Appare	nt particle d (g/cm ³)	ensity	Water	absorption	%
	-	d	С	b	а		Average	SD		Average	SD		Average	SD		Average	SD
5% w-	1,140	1.1145	3.5664	4.1605	1.2605	1.67	1.67	0.01	1.89	1.90	0.01	2.14	2.16	0.01	13.10	13.39	0.49
APCr		1.2287	3.5664	4.2276	1.4002	1.66			1.89			2.17			13.96		
		1.2858	3.5664	4.2578	1.4543	1.69			1.91			2.16			13.10		
	1,160	1.1962	3.5664	4.1992	1.3055	1.78	1.76	0.03	1.94	1.93	0.01	2.12	2.12	0.02	9.14	9.59	1.32
		1.3432	3.5664	4.2797	1.4919	1.73			1.92			2.13			11.07		
		1.1685	3.5664	4.1791	1.2685	1.78			1.93			2.10			8.56		
	1,180	1.4249	3.5664	4.3035	1.5604	1.73	1.74	0.01	1.90	1.91	0.01	2.07	2.09	0.02	9.51	9.54	0.08
		1.3195	3.5664	4.2531	1.4466	1.74			1.90			2.09			9.63		
		1.2456	3.5664	4.2198	1.3638	1.75			1.92			2.10			9.49		
10% w-	1,140	1.1712	3.5664	4.2110	1.3361	1.69	1.74	0.10	1.93	1.94	0.03	2.22	2.18	0.06	14.08	14.18	0.15
APCr		1.1086	3.5664	4.1511	1.1843	1.85			1.98			2.12			6.83		
		1.2815	3.5664	4.2627	1.4646	1.67			1.91			2.19			14.29		
	1,160	1.2100	3.5664	4.2248	1.3425	1.77	1.78	0.09	1.96	1.96	0.04	2.19	2.17	0.03	10.95	11.99	1.47
		1.2869	3.5664	4.2619	1.4545	1.70			1.92			2.18			13.02		
		1.2458	3.5664	4.2291	1.3254	1.88			2.00			2.14			6.39		
	1,180	1.3641	3.5664	4.2895	1.4931	1.77	1.79	0.04	1.94	1.95	0.02	2.13	2.14	0.02	9.46	8.96	1.47
		1.3145	3.5664	4.2610	1.4105	1.84			1.97			2.12			7.30		
		1.1880	3.5664	4.2048	1.3082	1.77			1.95			2.16			10.12		
15% w-	1,140	1.0772	3.5664	4.1528	1.2028	1.75	1.70	0.04	1.95	1.92	0.03	2.19	2.18	0.03	11.66	12.99	1.49
APCr		1.1599	3.5664	4.1861	1.3075	1.69			1.90			2.15			12.73		
		1.0172	3.5664	4.1204	1.1657	1.66			1.91			2.20			14.60		
	1,160	1.1159	3.5664	4.1688	1.1967	1.88	1.91	0.10	2.01	2.03	0.05	2.17	2.16	0.02	7.24	7.40	0.22
		1.0022	3.5664	4.0998	1.0779	1.84			1.98			2.14			7.55		
		1.1135	3.5664	4.1643	1.1488	2.02			2.09			2.16			3.17		
	1,180	1.0242	3.5664	4.1144	1.1335	1.75	1.83	0.10	1.94	1.98	0.05	2.15	2.14	0.02	10.67	7.96	3.28
		1.0072	3.5664	4.1047	1.0967	1.80			1.96			2.15			8.89		
		1.1360	3.5664	4.1664	1.1850	1.94			2.03			2.12			4.31		

Appendix XI - Calculation of particle density and water absorption of hot-bonded LWA by the method of the wired basket (BS EN 1097-6, 2022) Part D

Shaded values were refused

* ** *	Firing	W pellet in	W basket in	Wpellet+basket	W saturated	Oven dr	ied Particle d	lensity	Saturate	d and surface	e-dried	Appare	ent particle de	nsity	Wate	absorption	%
LWA	temp °C	air (g)	water (g)	in water (g)	pellet (g)		(g/cm ²)	SD	Partici	e density (g/	cm ²)		(g/cm ²)	SD		Averege	SD
20% w-	1 1/10	1 2017	3 5611	4 2046	a 1 3353	1.74	1 71	0.03	1.03	1 QO	0.02	2.15	2 13	0.02	11.12	11.62	0.54
ΔPCr	1,140	0.0078	3 5611	4.0302	1.0184	1.74	1./1	0.05	1.95	1.70	0.02	2.15	2.15	0.02	12 12	11.02	0.54
ni ci		1.0491	2 5611	4.0392	1.0104	1.00			1.00			2.11 2.11			12.10		
	1 160	1.0401	2 5611	4.1155	1.1091	1.70	1.96	0.02	1.90	1.07	0.02	2.11	2.08	0.04	5.49	5 70	0.86
	1,100	1.0393	2 5611	4.1027	1.11/4	1.04	1.00	0.02	1.94	1.97	0.02	2.05	2.08	0.04	5.10	5.19	0.80
		1.0011	2 5611	4.1123	1.1134	1.00			1.98			2.08			5.12		
	1 1 2 0	1.0003	2 5611	4.0922	1.0743	1.65	1.00	0.09	2.00	2.01	0.08	2.12	2.02	0.08	0.70	1 17	0.12
	1,180	1.2132	2.5011	4.1742	1.2238	1.90	1.99	0.08	2.00	2.01	0.08	2.02	2.05	0.08	1.04	1.1/	0.12
		1.0780	3.3011	4.1302	1.0910	2.07			2.09			2.12			1.21		
250/	1 1 4 0	1.10/3	3.3011	4.1041	1.1214	1.91	1.70	0.17	1.94	1.07	0.12	1.90	1.05	0.10	1.27	()5	0.00
23% W-	1,140	1.2920	3.3011	4.1545	1.3003	1.0/	1./9	0.17	1.//	1.8/	0.13	1.85	1.95	0.10	5.//	0.23	0.08
APCr		1.2250	3.5611	4.1906	1.2450	1.99			2.02			2.06			1.63		
	1.1.0	1.1504	3.5611	4.11/4	1.2278	1./1	1.00	0.07	1.83	1.01	0.00	1.94	1.05	0.00	6./3	1.70	0.40
	1,160	1.2967	3.5611	4.2256	1.3258	1.96	1.88	0.07	2.00	1.91	0.08	2.05	1.95	0.09	2.24	1./8	0.48
		1.2767	3.5611	4.1651	1.2932	1.85			1.88			1.90			1.29		
		1.0268	3.5611	4.0439	1.0452	1.83			1.86			1.89			1.79		
	1,180	1.2622	3.5611	4.1509	1.2744	1.84	1.84	0.02	1.86	1.86	0.02	1.88	1.88	0.02	0.97	0.98	0.02
		1.0647	3.5611	4.0657	1.0754	1.87			1.88			1.90			1.00		
		1.0494	3.5611	4.0452	1.0595	1.82			1.84			1.86			0.96		
30% w-	1,140	1.4320	3.5611	4.3057	1.6748	1.54	1.61	0.12	1.80	1.84	0.08	2.08	2.10	0.05	16.96	14.46	3.33
APCr		1.0215	3.5611	4.1075	1.1306	1.75			1.94			2.15			10.68		
		1.1744	3.5611	4.1636	1.3594	1.55			1.80			2.05			15.75		
	1,160	1.1650	3.5611	4.1281	1.1828	1.89	1.83	0.07	1.92	1.89	0.03	1.95	1.94	0.02	1.53	3.24	2.26
		1.3765	3.5611	4.2360	1.4564	1.76			1.86			1.96			5.80		
		1.0134	3.5611	4.0465	1.0376	1.84			1.88			1.92			2.39		
	1,180	1.1815	3.5611	4.1641	1.2156	1.93	1.81	0.11	1.98	1.84	0.13	2.04	1.87	0.15	2.89	1.86	0.89
		1.2020	3.5611	4.1064	1.2171	1.79			1.81			1.83			1.26		
		1.2838	3.5611	4.1113	1.3023	1.71			1.73			1.75			1.44		
Lyt	ag	1.5315	3.5598	4.1899	1.6887	1.45	1.47	0.02	1.60	1.61	0.02	1.70	1.72	0.02	10.26	9.63	0.81
		1.4971	3.5598	4.1819	1.6276	1.49			1.62			1.71			8.72		
		1.5928	3.5598	4.2369	1.7504	1.48			1.63			1.74			9.89		

Appendix XI - Calculation of particle density and water absorption of hot-bonded LWA by the method of the wired basket (BS EN 1097-6, 2022) Part E

Shaded value was refused

LWA	W oven-dried LWA (g)	W pycnometer + lid + water (g)	W pycnometer + lid + water + saturated LWA after 10 min (g)	W pycnometer + lid + water + saturated LWA after 60 min (g)	W pycnometer + lid + water + saturated LWA after 24 hr (g)	W saturated surface- dried LWA after 24 hr (g)
	M4	M ₃	M ₂	M_2	M ₂	M_1
100% Clay	500.19	1,596.99	1,804.11	1,807.19	1,816.48	523.51
100% Clay	500.50	1,598.72	1,804.57	1,807.49	1,816.96	523.94
250/ ADC.	600.20	1,596.99	1,900.03	1,901.24	1,907.24	626.31
23% w-APCr	600.30	1,598.72	1,900.09	1,902.18	1,908.62	627.37
200/ ADC.	500.00	1,598.10	1,850.00	1,853.99	1,862.48	580.82
50% w-APCr	500.30	1,597.55	1,850.06	1,853.01	1,862.19	581.58
Lyrta an	450.39	1,598.10	1,780.89	1,781.23	1,791.08	501.22
Lytag®	450.57	1,597.55	1,782.55	1,784.29	1,795.47	502.77

Appendix XII – Calculation of particle density and water absorption of hot-bonded LWA by the method of the pycnometer (BS EN 1097-6, 2022)

LWA	Ove	n dried Part density g/cm ³	icle	Satura dried	nted and sur Particle der g/cm ³	face- nsity	Apj	parent partie density g/cm ³	cle	Water a	bsorption at 1 %	0 min	Water a	bsorption at 6 %	60 min	Water a	ibsorption at %	24 hr
		Average	SD		Average	SD		Average	SD		Average	SD		Average	SD		Average	SD
100% Clay	1.64	1.64	0.01	1.72	1.71	0.01	1.78	1.77	0.01	2.19	2.20	0.01	2.80	2.80	0.01	4.66 4.68	4.67	0.01
	1.05			1.71			2.06			3.15			3 35			4 35		
25% w-APCr	1.88	1.89	0.01	1.97	1.97	0.00	2.06	2.06	0.00	3.09	3.12	0.04	3.44	3.39	0.06	4.51	4.43	0.11
20% w ADCr	1.57	1.57	0.00	1.83	1.83	0.00	2.11	2 1 1	0.00	13.67	13 74	0.11	14.47	14 44	0.04	16.16	16.21	0.06
5070 w-AI CI	1.57	1.57	0.00	1.83	1.65	0.00	2.12	2.11	0.00	13.82	13./4	0.11	14.41	14.44	0.04	16.25	10.21	0.00
Lytag®	1.46 1.47	1.46	0.01	1.62 1.64	1.63	0.02	1.74 1.78	1.76	0.02	9.02 8.72	8.87	0.22	9.10 9.10	9.10	0.00	11.29 11.59	11.44	0.21

Mass of Mass o		Volume of container V	Oven-dried particle density (Method of	W container + sample M ₂ (g)	Loose	bulk density (g/cm ³)	/ ρ _b	Loose bulk density pb		Voids v %	
	(g)	(cm ³)	pycnometer ρ_p (g/cm ³)			Average	SD	(g/L)*		Average	SD
1000/	228.17	1,061		1,234.52	0.95				42.2		
100% Clay	228.17	1,061	1.64	1,232.25	0.95	0.95	0.00	946.7	42.3	42.3	0.1
Clay	228.17	1,061		1,231.18	0.95				42.4		
25% m	228.17	1,061		1,389.67	1.09				42.1		
ΔPCr	228.17	1,061	1.89	1,401.27	1.11	1.10	0.01	1,102.1	41.5	41.7	0.3
AICI	228.17	1,061		1,401.47	1.11				41.5		
2004 m	228.17	1,061		1,189.97	0.91				42.3		
30% w-	228.17	1,061	1.57	1,203.87	0.92	0.91	0.01	906.6	41.4	42.3	0.8
AICI	228.17	1,061		1,176.27	0.89				43.1		
	228.17	1,061		1,125.63	0.85				42.1		
Lytag®	228.17	1,061	1.46	1,107.66	0.83	0.84	0.01	841.2	43.2	42.4	0.7
	228.17	1,061		1,128.90	0.85				41.9		

Appendix XIII – Calculation of loose bulk density and voids of hot-bonded LWA fired at 1,180°C

* Loose bulk density expressed in g/L is used for concrete formulation

Appendix XIV – Calculation of crushing strength of hot-bonded LWA	ł
Part A	

		100% C	Clay / 1,120	5% APCr-m / 1,120°C				
Sample No.	Fracture load (kN)	Fracture load (N)	Sphere Diameter (mm)	Compressive strength, MPa (N/mm ²)	Fracture load (kN)	Fracture load (N)	Sphere Diameter (mm)	Compressive strength, MPa (N/mm ²)
1	0.55	550	10	4.90	0.45	450	12	2.79
2	0.51	510	11	3.76	0.34	340	11	2.50
3	0.42	420	11	3.09	0.25	250	11	1.84
4	0.61	610	11	4.49	0.62	620	12	3.84
5	0.63	630	12	3.90	0.51	510	12	3.16
6	0.5	500	10	4.46	0.45	450	12	2.79
7	0.56	560	11	4.12	0.10	100	11	0.74
8	0.81	810	11	5.97	0.23	230	12	1.42
9	0.81	810	11	5.97	0.47	470	12	2.91
10	0.61	610	11	4.49	0.48	480	12	2.97
11	0.66	660	11	4.86	0.34	340	11	2.50
12	0.70	700	11	5.16	0.40	400	10	3.57
13	0.41	410	12	2.54	0.28	280	11	2.06
14	0.56	560	11	4.12				
15	0.7	700	11	5.16				
			Average	4.47			Average	2.54
			SD	0.94			SD	0.86
			Min	2.54			Min	0.74
			Max	5.97			Max	3 84
				0.77			1116678	5.01
		10% AP(Cr-m / 1,12	0°C		15% APC	Cr-m / 1,120	р°С
Sample No.	Fracture load (kN)	10% APC Fracture load (N)	Cr-m / 1,12 Sphere Diameter (mm)	0°C Compressive strength, MPa (N/mm ²)	Fracture load (kN)	15% APC Fracture load (N)	Cr-m / 1,120 Sphere Diameter (mm)	Compressive strength, MPa (N/mm ²)
Sample No.	Fracture load (kN) 0.2	10% APC Fracture load (N) 200	Cr-m / 1,12 Sphere Diameter (mm) 11	0°C Compressive strength, MPa (N/mm ²) 1.47	Fracture load (kN) 1.07	15% APC Fracture load (N) 1070	Cr-m / 1,120 Sphere Diameter (mm) 12	°C Compressive strength, MPa (N/mm ²) 6.62
Sample No.	Fracture load (kN) 0.2 0.31	10% APC Fracture load (N) 200 310	Cr-m / 1,12 Sphere Diameter (mm) 11 11	0°C Compressive strength, MPa (N/mm ²) 1.47 2.28	Fracture load (kN) 1.07 0.18	15% APC Fracture load (N) 1070 180	Cr-m / 1,120 Sphere Diameter (mm) 12 11	P°C Compressive strength, MPa (N/mm ²) 6.62 1.33
Sample No.	Fracture load (kN) 0.2 0.31 0.31	10% APC Fracture load (N) 200 310 310	Cr-m / 1,12 Sphere Diameter (mm) 11 11 12	0°C Compressive strength, MPa (N/mm ²) 1.47 2.28 1.92	Fracture load (kN) 1.07 0.18 0.55	15% APC Fracture load (N) 1070 180 550	Cr-m / 1,120 Sphere Diameter (mm) 12 11 11	Compressive strength, MPa (N/mm ²) 6.62 1.33 4.05
Sample No.	Fracture load (kN) 0.2 0.31 0.31 0.31	10% APC Fracture load (N) 200 310 310 310	Cr-m / 1,12 Sphere Diameter (mm) 11 11 12 11	0°C Compressive strength, MPa (N/mm ²) 1.47 2.28 1.92 2.28	Fracture load (kN) 1.07 0.18 0.55 0.51	15% APC Fracture load (N) 1070 180 550 510	Cr-m / 1,120 Sphere Diameter (mm) 12 11 11 11 10	Compressive strength, MPa (N/mm ²) 6.62 1.33 4.05 4.55
Sample No. 1 2 3 4 5	Fracture load (kN) 0.2 0.31 0.31 0.31 0.28	10% APC Fracture load (N) 200 310 310 310 280	Cr-m / 1,12 Sphere Diameter (mm) 11 11 12 11 11 11	0°C Compressive strength, MPa (N/mm ²) 1.47 2.28 1.92 2.28 2.06	Fracture load (kN) 1.07 0.18 0.55 0.51 0.28	15% APC Fracture load (N) 1070 180 550 510 280	Cr-m / 1,120 Sphere Diameter (mm) 12 11 11 10 11	Compressive strength, MPa (N/mm ²) 6.62 1.33 4.05 4.55 2.06
Sample No. 1 2 3 4 5 6	Fracture load (kN) 0.2 0.31 0.31 0.31 0.28 0.15	10% APC Fracture load (N) 200 310 310 310 280 150	Cr-m / 1,12 Sphere Diameter (mm) 11 11 12 11 11 11 11 11	0°C Compressive strength, MPa (N/mm ²) 1.47 2.28 1.92 2.28 2.06 1.10	Fracture load (kN) 1.07 0.18 0.55 0.51 0.28 0.31	15% APC Fracture load (N) 1070 180 550 510 280 310	Cr-m / 1,120 Sphere Diameter (mm) 12 11 11 10 11 11	Compressive strength, MPa (N/mm ²) 6.62 1.33 4.05 4.55 2.06 2.28
Sample No. 1 2 3 4 5 6 7	Fracture load (kN) 0.2 0.31 0.31 0.31 0.28 0.15 0.15	10% APC Fracture load (N) 200 310 310 310 280 150 150	Cr-m / 1,12 Sphere Diameter (mm) 11 11 12 11 11 11 11 11 11 11	0°C Compressive strength, MPa (N/mm ²) 1.47 2.28 1.92 2.28 2.06 1.10 1.10 1.10	Fracture load (kN) 1.07 0.18 0.55 0.51 0.28 0.31 0.25	15% APC Fracture load (N) 1070 180 550 510 280 310 250	Cr-m / 1,120 Sphere Diameter (mm) 12 11 11 10 11 11 11 10	Compressive strength, MPa (N/mm ²) 6.62 1.33 4.05 4.55 2.06 2.28 2.23
Sample No. 1 2 3 4 5 6 7 8	Fracture load (kN) 0.2 0.31 0.31 0.31 0.28 0.15 0.15 0.15 0.52	10% APC Fracture load (N) 200 310 310 310 280 150 150 150 520	Cr-m / 1,12 Sphere Diameter (mm) 11 11 12 11 11 11 11 11 11 11	0°C Compressive strength, MPa (N/mm ²) 1.47 2.28 1.92 2.28 2.06 1.10 1.10 1.10 3.83	Fracture load (kN) 1.07 0.18 0.55 0.51 0.28 0.31 0.25 0.34	15% APC Fracture load (N) 1070 180 550 510 280 310 250 340	Cr-m / 1,120 Sphere Diameter (mm) 12 11 11 10 11 11 10 11	Compressive strength, MPa (N/mm ²) 6.62 1.33 4.05 4.55 2.06 2.28 2.23 2.50
Sample No. 1 2 3 4 5 6 7 8 9	Fracture load (kN) 0.2 0.31 0.31 0.31 0.31 0.28 0.15 0.15 0.15 0.52 0.44	10% APC Fracture load (N) 200 310 310 310 280 150 150 150 520 440	Cr-m / 1,12 Sphere Diameter (mm) 11 11 12 11 11 11 11 11 11 11	0°C Compressive strength, MPa (N/mm ²) 1.47 2.28 1.92 2.28 2.06 1.10 1.10 1.10 3.83 3.92	Fracture load (kN) 1.07 0.18 0.55 0.51 0.28 0.31 0.25 0.34 1.41	15% APC Fracture load (N) 1070 180 550 510 280 310 250 340 1410	Cr-m / 1,120 Sphere Diameter (mm) 12 11 11 10 11 11 10 11 13	P°C Compressive strength, MPa (N/mm ²) 6.62 1.33 4.05 4.55 2.06 2.28 2.23 2.50 7.44
Sample No. 1 2 3 4 5 6 7 8 9 10	Fracture load (kN) 0.2 0.31 0.31 0.31 0.28 0.15 0.15 0.15 0.52 0.44 0.31	10% APC Fracture load (N) 200 310 310 310 280 150 150 150 520 440 310	Cr-m / 1,12 Sphere Diameter (mm) 11 11 12 11 11 11 11 11 11 11	0°C Compressive strength, MPa (N/mm ²) 1.47 2.28 1.92 2.28 2.06 1.10 1.10 3.83 3.92 2.28	Fracture load (kN) 1.07 0.18 0.55 0.51 0.28 0.31 0.25 0.34 1.41 0.50	15% APC Fracture load (N) 1070 180 550 510 280 310 250 340 1410 500	Cr-m / 1,120 Sphere Diameter (mm) 12 11 11 10 11 11 10 11 13 11	P°C Compressive strength, MPa (N/mm ²) 6.62 1.33 4.05 4.55 2.06 2.28 2.23 2.50 7.44 3.68
Sample No. 1 2 3 4 5 6 7 8 9 10 11	Fracture load (kN) 0.2 0.31 0.31 0.31 0.28 0.15 0.15 0.52 0.44 0.31 0.14	10% APC Fracture load (N) 200 310 310 310 280 150 150 150 520 440 310 140	Cr-m / 1,12 Sphere Diameter (mm) 11 11 12 11 11 11 11 11 11 11	0°C Compressive strength, MPa (N/mm ²) 1.47 2.28 1.92 2.28 2.06 1.10 1.10 3.83 3.92 2.28 1.03	Fracture load (kN) 1.07 0.18 0.55 0.51 0.28 0.31 0.25 0.34 1.41 0.50 0.34	15% APC Fracture load (N) 1070 180 550 510 280 310 250 340 1410 500 340	Image: Normal System Cr-m / 1,120 Sphere Diameter (mm) 12 11 10 11 10 11 10 11 13 11 13 11	P°C Compressive strength, MPa (N/mm ²) 6.62 1.33 4.05 4.55 2.06 2.28 2.23 2.50 7.44 3.68 2.50
Sample No. 1 2 3 4 5 6 7 8 9 10 11 12	Fracture load (kN) 0.2 0.31 0.31 0.31 0.28 0.15 0.15 0.52 0.44 0.31 0.14 0.11	10% APC Fracture load (N) 200 310 310 310 280 150 150 150 520 440 310 140 110	Cr-m / 1,12 Sphere Diameter (mm) 11 11 12 11 11 11 11 11 11 11	0°C Compressive strength, MPa (N/mm ²) 1.47 2.28 1.92 2.28 2.06 1.10 1.10 3.83 3.92 2.28 1.03 0.81	Fracture load (kN) 1.07 0.18 0.55 0.51 0.28 0.31 0.25 0.34 1.41 0.50 0.34 0.97	15% APC Fracture load (N) 1070 180 550 510 280 310 250 340 1410 500 340 970	Image: Cr-m / 1,120 Sphere Diameter (mm) 12 11 10 11 10 11 10 11 10 11 13 11 11 13 11 11	Stor Compressive strength, MPa (N/mm²) 6.62 1.33 4.05 4.55 2.06 2.28 2.23 2.50 7.44 3.68 2.50 7.14
Sample No. 1 2 3 4 5 6 7 8 9 10 11 12 13	Fracture load (kN) 0.2 0.31 0.31 0.31 0.31 0.28 0.15 0.15 0.52 0.44 0.31 0.14 0.11 0.38	10% APC Fracture load (N) 200 310 310 310 280 150 150 150 520 440 310 140 110 380	Cr-m / 1,12 Sphere Diameter (mm) 11 11 12 11 11 11 11 11 10 11 11 11 11	0°C Compressive strength, MPa (N/mm ²) 1.47 2.28 1.92 2.28 2.06 1.10 1.10 3.83 3.92 2.28 1.03 0.81 2.80	Fracture load (kN) 1.07 0.18 0.55 0.51 0.28 0.31 0.25 0.34 1.41 0.50 0.34 0.97	15% APC Fracture load (N) 1070 180 550 510 280 310 250 340 1410 500 340 970	Cr-m / 1,120 Sphere Diameter (mm) 12 11 10 11 10 11 13 11 11	Stor Compressive strength, MPa (N/mm²) 6.62 1.33 4.05 4.55 2.06 2.28 2.23 2.50 7.44 3.68 2.50 7.14
Sample No. 1 2 3 4 5 6 7 8 9 10 11 12 13 14	Fracture load (kN) 0.2 0.31 0.31 0.31 0.28 0.15 0.15 0.52 0.44 0.31 0.14 0.11 0.38 0.31	10% APC Fracture load (N) 200 310 310 310 280 150 150 520 440 310 140 110 380 310	Cr-m / 1,12 Sphere Diameter (mm) 11 11 12 11 11 11 11 11 11 11	0°C Compressive strength, MPa (N/mm ²) 1.47 2.28 1.92 2.28 2.06 1.10 1.10 1.10 3.83 3.92 2.28 1.03 0.81 2.80 1.92	Fracture load (kN) 1.07 0.18 0.55 0.51 0.28 0.31 0.25 0.34 1.41 0.50 0.34 0.97	15% APC Fracture load (N) 1070 180 550 510 280 310 250 340 1410 500 340 970	Image: Normal System Sphere Diameter (mm) 12 11 10 11 10 11 10 11 10 11 10 11 10 11 13 11 11 11	PC Compressive strength, MPa (N/mm ²) 6.62 1.33 4.05 4.55 2.06 2.28 2.23 2.50 7.44 3.68 2.50 7.14
Sample No. 1 2 3 4 5 6 7 8 9 10 11 12 13 14 15	Fracture load (kN) 0.2 0.31 0.31 0.31 0.28 0.15 0.15 0.52 0.44 0.31 0.14 0.11 0.38 0.31 0.34	10% APC Fracture load (N) 200 310 310 310 280 150 150 520 440 310 140 110 380 310 340	Cr-m / 1,12 Sphere Diameter (mm) 11 11 12 11 11 11 11 11 11 11	0°C Compressive strength, MPa (N/mm ²) 1.47 2.28 1.92 2.28 2.06 1.10 1.10 1.10 3.83 3.92 2.28 1.03 0.81 2.80 1.92 2.10	Fracture load (kN) 1.07 0.18 0.55 0.51 0.28 0.31 0.25 0.34 1.41 0.50 0.34 0.97	15% APC Fracture load (N) 1070 180 550 510 280 310 250 340 1410 500 340 970	Image: Note of the second se	PC Compressive strength, MPa (N/mm ²) 6.62 1.33 4.05 4.55 2.06 2.28 2.23 2.50 7.44 3.68 2.50 7.14
Sample No. 1 2 3 4 5 6 7 8 9 10 11 12 13 14 15	Fracture load (kN) 0.2 0.31 0.31 0.31 0.31 0.28 0.15 0.15 0.52 0.44 0.31 0.14 0.11 0.38 0.31 0.34	10% APC Fracture load (N) 200 310 310 310 280 150 150 520 440 310 140 110 380 310 340	Cr-m / 1,12 Sphere Diameter (mm) 11 11 12 11 11 11 11 11 11 11	0°C Compressive strength, MPa (N/mm ²) 1.47 2.28 1.92 2.28 2.06 1.10 1.10 1.10 3.83 3.92 2.28 1.03 0.81 2.80 1.92 2.10 2.06	Fracture load (kN) 1.07 0.18 0.55 0.51 0.28 0.31 0.25 0.34 1.41 0.50 0.34 0.97	15% APC Fracture load (N) 1070 180 550 510 280 310 250 340 1410 500 340 970	Cr-m / 1,120 Sphere Diameter (mm) 12 11 11 10 11 11 10 11 13 11 11 11 11 11 11	Sici Compressive strength, MPa (N/mm²) 6.62 1.33 4.05 4.55 2.06 2.28 2.23 2.50 7.44 3.68 2.50 7.14
Sample No. 1 2 3 4 5 6 7 8 9 10 11 12 13 14 15	Fracture load (kN) 0.2 0.31 0.31 0.31 0.31 0.28 0.15 0.15 0.52 0.44 0.31 0.14 0.11 0.38 0.31 0.34	10% APC Fracture load (N) 200 310 310 280 150 150 150 520 440 310 140 110 380 310 340	Cr-m / 1,12 Sphere Diameter (mm) 11 11 12 11 11 11 11 11 11 11	0°C Compressive strength, MPa (N/mm ²) 1.47 2.28 1.92 2.28 2.06 1.10 1.10 1.10 3.83 3.92 2.28 1.03 0.81 2.80 1.92 2.10 2.06 0.93	Fracture load (kN) 1.07 0.18 0.55 0.51 0.28 0.31 0.25 0.34 1.41 0.50 0.34 0.97	15% APC Fracture load (N) 1070 180 550 510 280 310 250 340 1410 500 340 970	Cr-m / 1,120 Sphere Diameter (mm) 12 11 11 10 11 11 10 11 11 13 11 11 11 11 11 11 11 5 5 5 5 5	PC Compressive strength, MPa (N/mm ²) 6.62 1.33 4.05 4.55 2.06 2.28 2.23 2.50 7.44 3.68 2.50 7.14 3.87 2.14
Sample No. 1 2 3 4 5 6 7 8 9 10 11 12 13 14 15	Fracture load (kN) 0.2 0.31 0.31 0.31 0.28 0.15 0.15 0.15 0.52 0.44 0.31 0.14 0.11 0.38 0.31 0.34	10% APC Fracture load (N) 200 310 310 280 150 150 520 440 310 140 110 380 310 340	Cr-m / 1,12 Sphere Diameter (mm) 11 11 12 11 11 11 11 11 11 11	0°C Compressive strength, MPa (N/mm ²) 1.47 2.28 1.92 2.28 2.06 1.10 1.10 1.10 3.83 3.92 2.28 1.03 0.81 2.80 1.92 2.10 2.06 0.93 0.81	Fracture load (kN) 1.07 0.18 0.55 0.51 0.28 0.31 0.25 0.34 1.41 0.50 0.34 0.97	15% APC Fracture load (N) 1070 180 550 510 280 310 250 340 1410 500 340 970	Cr-m / 1,120 Sphere Diameter (mm) 12 11 11 10 11 11 10 11 11 13 11 11 11 11 11 11 11 11	Sici °C Compressive strength, MPa (N/mm²) 6.62 1.33 4.05 4.55 2.06 2.28 2.23 2.50 7.44 3.68 2.50 7.14 3.87 2.14 1.33

Appendix XIV – Calculation of crushing strength of hot-bonded LW	Ά
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Part B

		20% APG	Cr-m / 1,12	0°C		25% APC	Cr-m / 1,120	°C
Sample No.	Fracture load (kN)	Fracture load (N)	Sphere Diameter (mm)	Compressive strength, MPa (N/mm ²)	Fracture load (kN)	Fracture load (N)	Sphere Diameter (mm)	Compressive strength, MPa (N/mm ²)
1	0.47	470	11	3.46	0.23	230	10	2.05
2	0.21	210	11	1.55	0.20	200	10	1.78
3	0.22	220	12	1.36	0.14	140	10	1.25
4	0.69	690	11	5.08	0.14	140	10	1.25
5	0.75	750	11	5.52	0.12	120	10	1.07
6	0.25	250	11	1.84	0.17	170	10	1.52
7	0.20	200	12	1.24	0.25	250	10	2.23
8	0.83	830	11	6.11	0.23	230	10	2.05
9	0.26	260	10	2.32	0.12	120	11	0.88
10	0.10	100	11	0.74	0.18	180	10	1.60
11	0.42	420	11	3.09	0.17	170	10	1.52
12	0.22	220	11	1.62	0.23	230	10	2.05
13	0.14	140	11	1.03	0.14	140	10	1.25
14 15					0.08	80	11	0.59
			Average	2.69			Average	1.51
			SD	1.83			SD	0.49
			Min	0.74			Min	0.59
			Max	6.11			Max	2.23
		100% C	Clay / 1,140	°C		5% APC	r-m / 1.140°	°C
			. ,					-
Sample No.	Fracture load (kN)	Fracture load (N)	Sphere Diameter (mm)	Compressive strength, MPa (N/mm ²)	Fracture load (kN)	Fracture load (N)	Sphere Diameter (mm)	Compressive strength, MPa (N/mm ²)
Sample No.	Fracture load (kN) 0.34	Fracture load (N) 340	Sphere Diameter (mm)	Compressive strength, MPa (N/mm ²) 2.50	Fracture load (kN) 0.15	Fracture load (N)	Sphere Diameter (mm)	Compressive strength, MPa (N/mm ²) 1.10
Sample No.	Fracture load (kN) 0.34 0.53	Fracture load (N) 340 530	Sphere Diameter (mm) 11 12	Compressive strength, MPa (N/mm ²) 2.50 3.28	Fracture load (kN) 0.15 0.34	Fracture load (N) 150 340	Sphere Diameter (mm) 11 12	Compressive strength, MPa (N/mm ²) 1.10 2.10
Sample No.	Fracture load (kN) 0.34 0.53 0.25	Fracture load (N) 340 530 250	Sphere Diameter (mm) 11 12 11	Compressive strength, MPa (N/mm ²) 2.50 3.28 1.84	Fracture load (kN) 0.15 0.34 0.25	Fracture load (N) 150 340 250	Sphere Diameter (mm) 11 12 12	Compressive strength, MPa (N/mm ²) 1.10 2.10 1.55
Sample No.	Fracture load (kN) 0.34 0.53 0.25 0.45	Fracture load (N) 340 530 250 450	Sphere Diameter (mm) 11 12 11 13	Compressive strength, MPa (N/mm ²) 2.50 3.28 1.84 2.37	Fracture load (kN) 0.15 0.34 0.25 0.34	Fracture load (N) 150 340 250 340	Sphere Diameter (mm) 11 12 12 12 12	Compressive strength, MPa (N/mm ²) 1.10 2.10 1.55 2.10
Sample No. 1 2 3 4 5	Fracture load (kN) 0.34 0.53 0.25 0.45 0.17	Fracture load (N) 340 530 250 450 170	Sphere Diameter (mm) 11 12 11 13 11	Compressive strength, MPa (N/mm ²) 2.50 3.28 1.84 2.37 1.25	Fracture load (kN) 0.15 0.34 0.25 0.34 0.23	Fracture load (N) 150 340 250 340 230	Sphere Diameter (mm) 11 12 12 12 12 12 12	Compressive strength, MPa (N/mm ²) 1.10 2.10 1.55 2.10 1.42
Sample No. 1 2 3 4 5 6	Fracture load (kN) 0.34 0.53 0.25 0.45 0.17 0.29	Fracture load (N) 340 530 250 450 170 290	Sphere Diameter (mm) 11 12 11 13 11 12 12	Compressive strength, MPa (N/mm ²) 2.50 3.28 1.84 2.37 1.25 1.79	Fracture load (kN) 0.15 0.34 0.25 0.34 0.23 0.28	Fracture load (N) 150 340 250 340 230 280	Sphere Diameter (mm) 11 12 12 12 12 12 12 12	Compressive strength, MPa (N/mm ²) 1.10 2.10 1.55 2.10 1.42 1.73
Sample No. 1 2 3 4 5 6 7	Fracture load (kN) 0.34 0.53 0.25 0.45 0.17 0.29 0.37	Fracture load (N) 340 530 250 450 170 290 370	Sphere Sphere Diameter (mm) 11 12 11 13 11 12 12 12	Compressive strength, MPa (N/mm ²) 2.50 3.28 1.84 2.37 1.25 1.79 2.29	Fracture load (kN) 0.15 0.34 0.25 0.34 0.23 0.28 0.28 0.34	Fracture load (N) 150 340 250 340 230 280 340	Sphere Diameter (mm) 11 12 12 12 12 12 12 12 12 12 12 12 12 12 12 12 12 12	Compressive strength, MPa (N/mm ²) 1.10 2.10 1.55 2.10 1.42 1.73 2.10
Sample No. 1 2 3 4 5 6 7 8	Fracture load (kN) 0.34 0.53 0.25 0.45 0.45 0.17 0.29 0.37 0.50	Fracture load (N) 340 530 250 450 170 290 370 500	Sphere Diameter (mm) 11 12 11 12 11 13 11 12 13 13	Compressive strength, MPa (N/mm ²) 2.50 3.28 1.84 2.37 1.25 1.79 2.29 2.64	Fracture load (kN) 0.15 0.34 0.25 0.34 0.23 0.28 0.34 0.25	Fracture load (N) 150 340 250 340 230 280 340 250	Sphere Diameter (mm) 11 12 12 12 12 12 12 11	Compressive strength, MPa (N/mm ²) 1.10 2.10 1.55 2.10 1.42 1.73 2.10 1.84
Sample No. 1 2 3 4 5 6 7 8 9	Fracture load (kN) 0.34 0.53 0.25 0.45 0.17 0.29 0.37 0.50 0.41	Fracture load (N) 340 530 250 450 170 290 370 500 410	Sphere Diameter (mm) 11 12 11 12 11 12 13 12 13 10	Compressive strength, MPa (N/mm ²) 2.50 3.28 1.84 2.37 1.25 1.79 2.29 2.64 3.65	Fracture load (kN) 0.15 0.34 0.25 0.34 0.23 0.28 0.34 0.25 0.31	Fracture load (N) 150 340 250 340 230 280 340 250 310	Sphere Diameter (mm) 11 12	Compressive strength, MPa (N/mm ²) 1.10 2.10 1.55 2.10 1.42 1.73 2.10 1.84 1.92
Sample No. 1 2 3 4 5 6 7 8 9 10	Fracture load (kN) 0.34 0.53 0.25 0.45 0.17 0.29 0.37 0.50 0.41 0.40	Fracture load (N) 340 530 250 450 170 290 370 500 410 400	Sphere Diameter (mm) 11 12 11 12 11 12 13 11 12 13 10 11	Compressive strength, MPa (N/mm ²) 2.50 3.28 1.84 2.37 1.25 1.79 2.29 2.64 3.65 2.95	Fracture load (kN) 0.15 0.34 0.25 0.34 0.23 0.28 0.34 0.25 0.34 0.25 0.31 0.26	Fracture load (N) 150 340 250 340 230 280 340 250 340 230 280 340 250 340 260	Sphere Diameter (mm) 11 12 12 12 12 12 12 12 12 12 12 12 11 12 12 12 11 12 11 12 11	Compressive strength, MPa (N/mm ²) 1.10 2.10 1.55 2.10 1.42 1.73 2.10 1.84 1.92 1.92
Sample No. 1 2 3 4 5 6 7 8 9 10 11	Fracture load (kN) 0.34 0.53 0.25 0.45 0.45 0.45 0.17 0.29 0.37 0.50 0.41 0.40 0.28	Fracture load (N) 340 530 250 450 170 290 370 500 410 400 280	Sphere Diameter (mm) 11 12 11 12 13 11 12 13 10 11 12	Compressive strength, MPa (N/mm ²) 2.50 3.28 1.84 2.37 1.25 1.79 2.29 2.64 3.65 2.95 1.73	Fracture load (kN) 0.15 0.34 0.25 0.34 0.23 0.28 0.34 0.25 0.31 0.26 0.22	Fracture load (N) 150 340 250 340 230 280 340 250 310 260 220	Sphere Diameter (mm) 11 12 11 12	Compressive strength, MPa (N/mm ²) 1.10 2.10 1.55 2.10 1.42 1.73 2.10 1.84 1.92 1.92 1.36
Sample No. 1 2 3 4 5 6 7 8 9 10 11 12	Fracture load (kN) 0.34 0.53 0.25 0.45 0.17 0.29 0.37 0.50 0.41 0.28 0.35	Fracture load (N) 340 530 250 450 170 290 370 500 410 400 280 350	Sphere Diameter (mm) 11 12 11 12 13 11 12 13 10 11 12 13 10 11 12 13 10 11 12 13 10 11 12 11	Compressive strength, MPa (N/mm²) 2.50 3.28 1.84 2.37 1.25 1.79 2.29 2.64 3.65 2.95 1.73 2.58	Fracture load (kN) 0.15 0.34 0.25 0.34 0.23 0.28 0.34 0.25 0.31 0.26 0.22 0.31	Fracture load (N) 150 340 250 340 230 280 340 250 310 260 310 310	Sphere Diameter (mm) 11 12 12 12 12 12 12 12 12 12 12 12 11 12 11 12 11 12 11 12 11 12 11	Compressive strength, MPa (N/mm ²) 1.10 2.10 1.55 2.10 1.42 1.73 2.10 1.84 1.92 1.92 1.36 2.28
Sample No. 1 2 3 4 5 6 7 8 9 10 11 12 13	Fracture load (kN) 0.34 0.53 0.25 0.45 0.17 0.29 0.37 0.50 0.41 0.40 0.28 0.35 0.34	Fracture load (N) 340 530 250 450 170 290 370 500 410 400 280 350 340	Sphere Diameter (mm) 11 12 11 12 13 11 12 13 10 11 12 13 10 11 12 13 10 11 12	Compressive strength, MPa (N/mm ²) 2.50 3.28 1.84 2.37 1.25 1.79 2.29 2.64 3.65 2.95 1.73 2.58 2.10	Fracture load (kN) 0.15 0.34 0.25 0.34 0.23 0.28 0.34 0.25 0.31 0.26 0.22 0.31 0.34	Fracture load (N) 150 340 250 340 230 280 340 250 310 260 310 340	Sphere Diameter (mm) 11 12 12 12 12 12 12 12 12 12 12 12 12 12 12 11 12 11 12 11 12 11 12 11 12	Compressive strength, MPa (N/mm ²) 1.10 2.10 1.55 2.10 1.42 1.73 2.10 1.84 1.92 1.92 1.92 1.36 2.28 2.10
Sample No. 1 2 3 4 5 6 7 8 9 10 11 12 13 14	Fracture load (kN) 0.34 0.53 0.25 0.45 0.45 0.17 0.29 0.37 0.50 0.41 0.40 0.28 0.35 0.34 0.42	Fracture load (N) 340 530 250 450 170 290 370 500 410 400 280 350 340 420	Sphere Diameter (mm) 11 12 11 12 13 11 12 13 10 11 12 13 10 11 12 13 10 11 12 11 12 11 12 11 12 11 12 11 12 11 12 11 12 11 12 11 12 10	Compressive strength, MPa (N/mm²) 2.50 3.28 1.84 2.37 1.25 1.79 2.29 2.64 3.65 2.95 1.73 2.58 2.10 3.74	Fracture load (kN) 0.15 0.34 0.25 0.34 0.23 0.28 0.34 0.25 0.31 0.26 0.22 0.31 0.34 0.34 0.28	Fracture load (N) 150 340 250 340 230 280 340 250 310 260 310 340 220 310 280	Sphere Diameter (mm) 11 12 12 12 12 12 12 12 12 11 12 12 11 12 12 11 12 11 12 11 12 11 12 11 12	Compressive strength, MPa (N/mm ²) 1.10 2.10 1.55 2.10 1.42 1.73 2.10 1.84 1.92 1.92 1.92 1.36 2.28 2.10 1.73
Sample No. 1 2 3 4 5 6 7 8 9 10 11 12 13 14 15	Fracture load (kN) 0.34 0.53 0.25 0.45 0.17 0.29 0.37 0.50 0.41 0.40 0.28 0.35 0.34 0.42 0.40	Fracture load (N) 340 530 250 450 170 290 370 500 410 400 280 350 340 420 400	Sphere Diameter (mm) 11 12 11 12 13 11 12 13 10 11 12 13 10 11 12 13 10 11 12 11 12 11 12 11	Compressive strength, MPa (N/mm²) 2.50 3.28 1.84 2.37 1.25 1.79 2.29 2.64 3.65 2.95 1.73 2.58 2.10 3.74 2.95	Fracture load (kN) 0.15 0.34 0.25 0.34 0.23 0.28 0.34 0.25 0.31 0.26 0.22 0.31 0.34 0.28 0.34 0.28 0.31	Fracture load (N) 150 340 250 340 230 280 340 250 340 230 280 340 250 310 260 210 310 340 280 310	Sphere Diameter (mm) 11 12 12 12 12 12 12 12 12 12 12 12 11 12 11 12 11 12 11 12 11 12 11 12 11 12 11	Compressive strength, MPa (N/mm ²) 1.10 2.10 1.55 2.10 1.42 1.73 2.10 1.84 1.92 1.92 1.92 1.36 2.28 2.10 1.73 2.28
Sample No. 1 2 3 4 5 6 7 8 9 10 11 12 13 14 15	Fracture load (kN) 0.34 0.53 0.25 0.45 0.45 0.17 0.29 0.37 0.50 0.41 0.40 0.28 0.35 0.34 0.42 0.40	Fracture load (N) 340 530 250 450 170 290 370 500 410 400 280 350 340 420 400	Sphere Diameter (mm) 11 12 11 12 13 11 12 13 10 11 12 13 10 11 12 13 10 11 12 11 12 10 11 12 10 11 Average	Compressive strength, MPa (N/mm ²) 2.50 3.28 1.84 2.37 1.25 1.79 2.29 2.64 3.65 2.95 1.73 2.58 2.10 3.74 2.95 2.51	Fracture load (kN) 0.15 0.34 0.25 0.34 0.23 0.28 0.34 0.25 0.31 0.26 0.22 0.31 0.34 0.28 0.34 0.28 0.31	Fracture load (N) 150 340 250 340 230 280 340 250 340 230 280 340 250 310 260 310 340 280 310 340 280 310	Sphere Diameter (mm) 11 12 12 12 12 12 12 12 12 12 12 12 12 12 11 12 11 12 11 12 11 12 11 12 11 12 11 12 11 12 11 12 11 Average	Compressive strength, MPa (N/mm²) 1.10 2.10 1.55 2.10 1.42 1.73 2.10 1.84 1.92 1.36 2.28 2.10 1.73 2.10
Sample No. 1 2 3 4 5 6 7 8 9 10 11 12 13 14 15	Fracture load (kN) 0.34 0.53 0.25 0.45 0.17 0.29 0.37 0.50 0.41 0.40 0.28 0.35 0.34 0.42 0.40	Fracture load (N) 340 530 250 450 170 290 370 500 410 400 280 350 340 420 400	Sphere Diameter (mm) 11 12 11 12 13 11 12 13 10 11 12 13 10 11 12 13 10 11 12 11 12 11 12 11 12 13 10 11 Average SD	Compressive strength, MPa (N/mm²) 2.50 3.28 1.84 2.37 1.25 1.79 2.29 2.64 3.65 2.95 1.73 2.58 2.10 3.74 2.95 2.51 0.72	Fracture load (kN) 0.15 0.34 0.25 0.34 0.23 0.28 0.34 0.25 0.31 0.26 0.22 0.31 0.26 0.22 0.31 0.34 0.28 0.34	Fracture load (N) 150 340 250 340 230 280 340 250 340 230 280 340 250 310 260 220 310 340 280 310	Sphere Diameter (mm) 11 12 12 12 12 12 12 12 12 11 12 12 11 12 11 12 11 12 11 12 11 12 11 12 11 12 11 12 13 14 15 16 17 18 19 11 Average SD	Compressive strength, MPa (N/mm²) 1.10 2.10 1.55 2.10 1.42 1.73 2.10 1.84 1.92 1.36 2.28 2.10 1.73 2.10
Sample No. 1 2 3 4 5 6 7 8 9 10 11 12 13 14 15	Fracture load (kN) 0.34 0.53 0.25 0.45 0.17 0.29 0.37 0.50 0.41 0.40 0.28 0.35 0.34 0.41	Fracture load (N) 340 530 250 450 170 290 370 500 410 400 280 350 340 420 400	Sphere Diameter (mm) 11 12 11 12 13 11 12 13 10 11 12 13 10 11 12 13 10 11 12 13 10 11 Average SD Min	Compressive strength, MPa (N/mm ²) 2.50 3.28 1.84 2.37 1.25 1.79 2.29 2.64 3.65 2.95 1.73 2.58 2.10 3.74 2.95 2.51 0.72 1.25	Fracture load (kN) 0.15 0.34 0.25 0.34 0.23 0.28 0.34 0.25 0.31 0.26 0.31 0.34 0.31 0.34	Fracture load (N) 150 340 250 340 230 280 340 250 340 230 280 340 250 310 260 220 310 340 280 310	Sphere Diameter (mm) 11 12 12 12 12 12 12 12 12 12 12 12 12 12 11 12 11 12 11 12 11 12 11 12 11 12 11 12 13 14 15 16 17 18 19 11 Average SD Min	Compressive strength, MPa (N/mm ²) 1.10 2.10 1.55 2.10 1.42 1.73 2.10 1.84 1.92 1.36 2.28 2.10 1.73 2.10

Appendix XIV – Calculation of crushing strength of hot-bonded LW	Ά
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Part C

		10% AP	Cr-m / 1,14	0°C		15% APC	Cr-m / 1,140	°C
Sample No.	Fracture load (kN)	Fracture load (N)	Sphere Diameter (mm)	Compressive strength, MPa (N/mm²)	Fracture load (kN)	Fracture load (N)	Sphere Diameter (mm)	Compressive strength, MPa (N/mm²)
1	0.26	260	11	1.92	0.93	930	11	6.85
2	0.32	320	11	2.36	0.63	630	11	4.64
3	0.15	150	11	1.10	0.42	420	10	3.74
4	0.25	250	11	1.84	0.31	310	10	2.76
5	0.31	310	11	2.28	0.46	460	10	4.10
6	0.31	310	11	2.28	0.64	640	11	4.71
7	0.23	230	11	1.69	0.59	590	11	4.35
8	0.27	270	11	1.99	0.87	870	10	7.75
9	0.12	120	10	1.07	0.77	770	10	6.86
10	0.23	230	11	1.69	0.74	740	10	6.60
11	0.14	140	10	1.25	0.93	930	10	8.29
12	0.14	140	10	1.25	0.37	370	9	4.07
13	0.20	200	10	1.78	1.10	1100	12	6.81
14	0.34	340	11	2.50	0.32	320	10	2.85
15	0.27	270	11	1.99	1			
			Average	1.80			Average	5.31
			SD	0.46			SD	1.82
			Min	1.07			Min	2.76
			Max	2.50			Max	8.29
		20% AP	C r-m / 1,14	0°C		25% APC	Cr-m / 1,140	°C
Sample No.	Fracture load (kN)	20% AP Fracture load (N)	Cr-m / 1,14 Sphere Diameter (mm)	0°C Compressive strength, MPa (N/mm ²)	Fracture load (kN)	25% APC Fracture load (N)	Cr-m / 1,140 Sphere Diameter (mm)	°C Compressive strength, MPa (N/mm ²)
Sample No.	Fracture load (kN) 0.22	20% AP Fracture load (N) 220	Cr-m / 1,14 Sphere Diameter (mm) 9	0°C Compressive strength, MPa (N/mm ²) 2.42	Fracture load (kN) 0.10	25% APC Fracture load (N) 100	Cr-m / 1,140 Sphere Diameter (mm) 10	°C Compressive strength, MPa (N/mm ²) 0.89
Sample No.	Fracture load (kN) 0.22 0.22	20% AP Fracture load (N) 220 220	Cr-m / 1,14 Sphere Diameter (mm) 9 10	0°C Compressive strength, MPa (N/mm ²) 2.42 1.96	Fracture load (kN) 0.10 0.20	25% APC Fracture load (N) 100 200	Cr-m / 1,140 Sphere Diameter (mm) 10 12	°C Compressive strength, MPa (N/mm ²) 0.89 1.24
Sample No.	Fracture load (kN) 0.22 0.22 0.2	20% AP Fracture load (N) 220 220 200	Cr-m / 1,14 Sphere Diameter (mm) 9 10 11	0°C Compressive strength, MPa (N/mm ²) 2.42 1.96 1.47	Fracture load (kN) 0.10 0.20 0.20	25% APC Fracture load (N) 100 200 200	Cr-m / 1,140 Sphere Diameter (mm) 10 12 12	°C Compressive strength, MPa (N/mm ²) 0.89 1.24 1.24
Sample No. 1 2 3 4	Fracture load (kN) 0.22 0.22 0.2 0.2 0.31	20% APC Fracture load (N) 220 220 200 310	Cr-m / 1,14 Sphere Diameter (mm) 9 10 11 10	0°C Compressive strength, MPa (N/mm ²) 2.42 1.96 1.47 2.76	Fracture load (kN) 0.10 0.20 0.20 0.15	25% APC Fracture load (N) 100 200 200 150	Cr-m / 1,140 Sphere Diameter (mm) 10 12 12 12 12	°C Compressive strength, MPa (N/mm ²) 0.89 1.24 1.24 1.24 0.93
Sample No. 1 2 3 4 5	Fracture load (kN) 0.22 0.22 0.2 0.31 0.32	20% APC Fracture load (N) 220 200 310 320	Cr-m / 1,14 Sphere Diameter (mm) 9 10 11 10 11	0°C Compressive strength, MPa (N/mm ²) 2.42 1.96 1.47 2.76 2.36	Fracture load (kN) 0.10 0.20 0.20 0.15 0.14	25% APC Fracture load (N) 100 200 200 150 140	Cr-m / 1,140 Sphere Diameter (mm) 10 12 12 12 12 12 13	°C Compressive strength, MPa (N/mm ²) 0.89 1.24 1.24 1.24 0.93 0.74
Sample No. 1 2 3 4 5 6	Fracture load (kN) 0.22 0.22 0.22 0.2 0.31 0.32 0.56	20% APC Fracture load (N) 220 220 200 310 320 560	Cr-m / 1,14 Sphere Diameter (mm) 9 10 11 10 11 10 11 11	0°C Compressive strength, MPa (N/mm ²) 2.42 1.96 1.47 2.76 2.36 4.12	Fracture load (kN) 0.10 0.20 0.20 0.15 0.14 0.21	25% APC Fracture load (N) 100 200 200 150 140 210	Cr-m / 1,140 Sphere Diameter (mm) 10 12 12 12 12 12 13 12	°C Compressive strength, MPa (N/mm ²) 0.89 1.24 1.24 0.93 0.74 1.30
Sample No. 1 2 3 4 5 6 7	Fracture load (kN) 0.22 0.22 0.2 0.2 0.31 0.32 0.56 0.28	20% APC Fracture load (N) 220 220 200 310 320 560 280	Cr-m / 1,14 Sphere Diameter (mm) 9 10 11 10 11 10 11 11 11	0°C Compressive strength, MPa (N/mm ²) 2.42 1.96 1.47 2.76 2.36 4.12 2.06	Fracture load (kN) 0.10 0.20 0.20 0.15 0.14 0.21 0.11	25% APC Fracture load (N) 100 200 200 150 140 210 110	Cr-m / 1,140 Sphere Diameter (mm) 10 12 12 12 12 12 13 12 13 12 11	°C Compressive strength, MPa (N/mm ²) 0.89 1.24 1.24 0.93 0.74 1.30 0.81
Sample No. 1 2 3 4 5 6 7 8	Fracture load (kN) 0.22 0.22 0.2 0.31 0.32 0.56 0.28 0.31	20% APC Fracture load (N) 220 220 200 310 320 560 280 310	Cr-m / 1,14 Sphere Diameter (mm) 9 10 11 10 11 11 11 11 11 11 10	0°C Compressive strength, MPa (N/mm ²) 2.42 1.96 1.47 2.76 2.36 4.12 2.06 2.76	Fracture load (kN) 0.10 0.20 0.20 0.15 0.14 0.21 0.11 0.10	25% APC Fracture load (N) 100 200 200 150 140 210 110 100	Cr-m / 1,140 Sphere Diameter (mm) 10 12 12 12 12 13 12 13 12 11 11	°C Compressive strength, MPa (N/mm ²) 0.89 1.24 1.24 0.93 0.74 1.30 0.81 0.74
Sample No. 1 2 3 4 5 6 7 8 9	Fracture load (kN) 0.22 0.22 0.2 0.31 0.32 0.56 0.28 0.31 0.21	20% APC Fracture load (N) 220 220 200 310 320 560 280 310 210	Cr-m / 1,14 Sphere Diameter (mm) 9 10 11 10 11 11 11 10 10 10 10	0°C Compressive strength, MPa (N/mm ²) 2.42 1.96 1.47 2.76 2.36 4.12 2.06 2.76 1.87	Fracture load (kN) 0.10 0.20 0.20 0.15 0.14 0.21 0.11 0.10 0.05	25% APC Fracture load (N) 100 200 200 150 140 210 110 100 50	Cr-m / 1,140 Sphere Diameter (mm) 10 12 12 12 13 12 13 12 11 11 11	°C Compressive strength, MPa (N/mm ²) 0.89 1.24 1.24 0.93 0.74 1.30 0.81 0.74 0.37
Sample No. 1 2 3 4 5 6 7 8 9 10	Fracture load (kN) 0.22 0.22 0.2 0.31 0.32 0.56 0.28 0.31 0.21 0.21 0.34	20% AP Fracture load (N) 220 220 200 310 320 560 280 310 210 340	Cr-m / 1,14 Sphere Diameter (mm) 9 10 11 10 11 11 11 10 10 10 10	0°C Compressive strength, MPa (N/mm ²) 2.42 1.96 1.47 2.76 2.36 4.12 2.06 2.76 1.87 3.03	Fracture load (kN) 0.10 0.20 0.20 0.15 0.14 0.21 0.11 0.10 0.05 0.07	25% APC Fracture load (N) 100 200 200 150 140 210 110 100 50 70	Cr-m / 1,140 Sphere Diameter (mm) 10 12 12 12 12 13 12 11 11 11 11	°C Compressive strength, MPa (N/mm ²) 0.89 1.24 1.24 0.93 0.74 1.30 0.81 0.74 0.37 0.52
Sample No. 1 2 3 4 5 6 7 8 9 10 11	Fracture load (kN) 0.22 0.22 0.2 0.31 0.32 0.56 0.28 0.31 0.21 0.34 0.35	20% APC Fracture load (N) 220 200 310 320 560 280 310 210 340 350	Cr-m / 1,14 Sphere Diameter (mm) 9 10 11 10 11 11 11 10 10 10 10	0°C Compressive strength, MPa (N/mm ²) 2.42 1.96 1.47 2.76 2.36 4.12 2.06 2.76 1.87 3.03 3.12	Fracture load (kN) 0.10 0.20 0.20 0.15 0.14 0.21 0.11 0.10 0.05 0.07 0.07	25% APC Fracture load (N) 100 200 200 150 140 210 110 100 50 70 70 70	Cr-m / 1,140 Sphere Diameter (mm) 10 12 12 12 12 13 12 11 11 11 11 10 10 12 12 12 13 12 11 11 11 10 12 12 13 12 11 11 11 11 11 11 11 11 11	°C Compressive strength, MPa (N/mm ²) 0.89 1.24 1.24 0.93 0.74 1.30 0.81 0.74 0.37 0.52 0.62
Sample No. 1 2 3 4 5 6 7 8 9 10 11 12	Fracture load (kN) 0.22 0.22 0.22 0.22 0.31 0.32 0.56 0.28 0.31 0.21 0.34 0.35 0.22	20% AP Fracture load (N) 220 200 310 320 560 280 310 210 340 350 220	Cr-m / 1,14 Sphere Diameter (mm) 9 10 11 10 11 11 11 11 10 10 10	0°C Compressive strength, MPa (N/mm ²) 2.42 1.96 1.47 2.76 2.36 4.12 2.06 2.76 1.87 3.03 3.12 2.42	Fracture load (kN) 0.10 0.20 0.20 0.15 0.14 0.21 0.11 0.10 0.05 0.07 0.07 0.11	25% APC Fracture load (N) 100 200 200 150 140 210 110 100 50 70 70 110	Cr-m / 1,140 Sphere Diameter (mm) 10 12 12 12 13 12 11 11 11 11 10 11 10 11	°C Compressive strength, MPa (N/mm ²) 0.89 1.24 1.24 0.93 0.74 1.30 0.81 0.74 0.37 0.52 0.62 0.81
Sample No. 1 2 3 4 5 6 7 8 9 10 11 12 13	Fracture load (kN) 0.22 0.22 0.2 0.2 0.31 0.32 0.56 0.28 0.31 0.21 0.34 0.35 0.22 0.20	20% AP Fracture load (N) 220 200 310 320 560 280 310 210 340 350 220 200	Cr-m / 1,14 Sphere Diameter (mm) 9 10 11 10 11 11 11 11 10 10 10	0°C Compressive strength, MPa (N/mm ²) 2.42 1.96 1.47 2.76 2.36 4.12 2.06 2.76 1.87 3.03 3.12 2.42 1.78	Fracture load (kN) 0.10 0.20 0.15 0.14 0.21 0.11 0.10 0.05 0.07 0.07 0.11 0.07	25% APC Fracture load (N) 100 200 200 150 140 210 110 100 50 70 70 110 70	Cr-m / 1,140 Sphere Diameter (mm) 10 12 12 12 12 13 12 11 11 11 10 11 11 10 11 11	°C Compressive strength, MPa (N/mm ²) 0.89 1.24 1.24 0.93 0.74 1.30 0.81 0.74 0.37 0.52 0.62 0.81 0.52
Sample No. 1 2 3 4 5 6 7 8 9 10 11 12 13 14	Fracture load (kN) 0.22 0.22 0.2 0.2 0.31 0.32 0.56 0.28 0.31 0.21 0.34 0.35 0.22 0.20 0.18	20% AP Fracture load (N) 220 200 310 320 560 280 310 210 340 350 220 200 180	Cr-m / 1,14 Sphere Diameter (mm) 9 10 11 10 11 11 11 11 10 10 10	0°C Compressive strength, MPa (N/mm ²) 2.42 1.96 1.47 2.76 2.36 4.12 2.06 2.76 1.87 3.03 3.12 2.42 1.78 1.60	Fracture load (kN) 0.10 0.20 0.15 0.14 0.11 0.11 0.10 0.05 0.07 0.07 0.11 0.07 0.14	25% APC Fracture load (N) 100 200 200 150 140 210 110 100 50 70 70 110 70 140 40	Sphere Diameter (mm) 10 12 12 12 13 12 13 12 13 11 11 11 11 11 11 11 11 11 11 11 11 11 11	°C Compressive strength, MPa (N/mm ²) 0.89 1.24 1.24 0.93 0.74 1.30 0.81 0.74 0.37 0.52 0.62 0.81 0.52 1.03
Sample No. 1 2 3 4 5 6 7 8 9 10 11 12 13 14 15	Fracture load (kN) 0.22 0.22 0.22 0.2 0.31 0.32 0.56 0.28 0.31 0.21 0.34 0.35 0.22 0.20 0.18 0.14	20% APC Fracture load (N) 220 220 200 310 320 560 280 310 210 340 350 220 200 180 140	Cr-m / 1,14 Sphere Diameter (mm) 9 10 11 10 11 11 11 10 10 10 10	0°C Compressive strength, MPa (N/mm ²) 2.42 1.96 1.47 2.76 2.36 4.12 2.06 2.76 1.87 3.03 3.12 2.42 1.78 1.60 1.25	Fracture load (kN) 0.10 0.20 0.20 0.15 0.14 0.21 0.11 0.10 0.05 0.07 0.07 0.07 0.11 0.07 0.14 0.07	25% APC Fracture load (N) 100 200 200 150 140 210 110 100 50 70 110 70 140 70 140 70 140 70	Sphere Diameter (mm) 10 12 12 12 13 12 13 11	°C Compressive strength, MPa (N/mm ²) 0.89 1.24 1.24 0.93 0.74 1.30 0.81 0.74 0.37 0.52 0.62 0.81 0.52 1.03 0.62
Sample No. 1 2 3 4 5 6 7 8 9 10 11 12 13 14 15	Fracture load (kN) 0.22 0.22 0.2 0.31 0.32 0.56 0.28 0.31 0.21 0.34 0.35 0.22 0.20 0.18 0.14	20% AP Fracture load (N) 220 220 200 310 320 560 280 310 210 340 350 220 200 180 140	Cr-m / 1,14 Sphere Diameter (mm) 9 10 11 10 11 11 10 10 10 10 10	0°C Compressive strength, MPa (N/mm ²) 2.42 1.96 1.47 2.76 2.36 4.12 2.06 2.76 1.87 3.03 3.12 2.42 1.78 1.60 1.25 2.33	Fracture load (kN) 0.10 0.20 0.20 0.15 0.14 0.21 0.11 0.10 0.05 0.07 0.07 0.07 0.11 0.07 0.14 0.07	25% APC Fracture load (N) 100 200 200 150 140 210 110 100 50 70 70 110 70 140 70 140 70	Sphere Diameter (mm) 10 12 12 12 13 12 13 11 11 11 11 11 11 11 11 11 10 11 10 Average	°C Compressive strength, MPa (N/mm ²) 0.89 1.24 1.24 0.93 0.74 1.30 0.81 0.74 0.37 0.52 0.62 0.81 0.52 1.03 0.62 0.82
Sample No. 1 2 3 4 5 6 7 8 9 10 11 12 13 14 15	Fracture load (kN) 0.22 0.22 0.2 0.31 0.32 0.56 0.28 0.31 0.21 0.34 0.35 0.22 0.20 0.18 0.14	20% AP Fracture load (N) 220 220 200 310 320 560 280 310 210 340 350 220 200 180 140	Cr-m / 1,14 Sphere Diameter (mm) 9 10 11 10 11 11 10 10 10 10 10	0°C Compressive strength, MPa (N/mm ²) 2.42 1.96 1.47 2.76 2.36 4.12 2.06 2.76 1.87 3.03 3.12 2.42 1.78 1.60 1.25 2.33 0.75	Fracture load (kN) 0.10 0.20 0.20 0.15 0.14 0.21 0.11 0.10 0.05 0.07 0.07 0.07 0.07 0.11 0.07 0.14 0.07	25% APC Fracture load (N) 100 200 200 150 140 210 110 100 50 70 70 110 70 140 70 140 70	Sphere Diameter (mm) 10 12 12 12 13 12 13 12 13 10 11 11 11 11 11 10 Average SD	°C Compressive strength, MPa (N/mm ²) 0.89 1.24 1.24 0.93 0.74 1.30 0.81 0.74 0.37 0.52 0.62 0.81 0.52 1.03 0.62 0.82 0.28
Sample No. 1 2 3 4 5 6 7 8 9 10 11 12 13 14 15	Fracture load (kN) 0.22 0.22 0.2 0.31 0.32 0.56 0.28 0.31 0.21 0.34 0.35 0.22 0.20 0.18 0.14	20% AP Fracture load (N) 220 220 200 310 320 560 280 310 210 340 350 220 200 180 140	Cr-m / 1,14 Sphere Diameter (mm) 9 10 11 10 11 11 11 10 10 10 10	0°C Compressive strength, MPa (N/mm ²) 2.42 1.96 1.47 2.76 2.36 4.12 2.06 2.76 1.87 3.03 3.12 2.42 1.78 1.60 1.25 2.33 0.75 1.25	Fracture load (kN) 0.10 0.20 0.20 0.15 0.14 0.21 0.11 0.10 0.05 0.07 0.07 0.07 0.11 0.07 0.14 0.07	25% APC Fracture load (N) 100 200 200 150 140 210 110 100 50 70 70 110 70 140 70 140 70	Sphere Diameter (mm) 10 12 12 12 13 12 13 12 13 10 12 13 12 13 12 13 10 11 11 10 Average SD Min	℃C Compressive strength, MPa (N/mm ²) 0.89 1.24 1.24 0.93 0.74 1.30 0.81 0.74 0.37 0.52 0.62 0.81 0.52 0.62 0.82 0.37

Appendix XIV – Calculation	of crushing strength	of hot-bonded LWA
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Part D

	100% Clay / 1,160°C					5% APCr-m / 1,160°C			
Sample No.	Fracture load (kN)	Fracture load (N)	Sphere Diameter (mm)	Compressive strength, MPa (N/mm ²)	Fracture load (kN)	Fracture load (N)	Sphere Diameter (mm)	Compressive strength, MPa (N/mm²)	
1	0.37	370	12	2.29	0.34	340	12	2.10	
2	0.47	470	11	3.46	0.53	530	11	3.90	
3	0.44	440	12	2.72	0.35	350	11	2.58	
4	0.37	370	13	1.95	0.82	820	12	5.08	
5	0.37	370	12	2.29	0.71	710	12	4.39	
6	0.29	290	12	1.79	0.32	320	11	2.36	
7	0.42	420	12	2.60	0.61	610	12	3.78	
8	0.64	640	12	3.96	0.40	400	11	2.95	
9	0.47	470	11	3.46	0.34	340	12	2.10	
10	0.26	260	11	1.92	0.68	680	11	5.01	
11	0.42	420	12	2.60	0.29	290	12	1.79	
12	0.72	720	12	4.46	0.31	310	12	1.92	
13	0.45	450	12	2.79	0.66	660	11	4.86	
14	0.29	290	11	2.14					
15	0.45	450	12	2.79					
			Average	2.75			Average	3.29	
			SD	0.78			SD	1.25	
			Min	1.79			Min	1.79	
			Max	4.46			Max	5.08	
								0.00	
		10% AP	Cr-m / 1,16	0°C		15% APC	Cr-m / 1,160	0°C	
Sample No.	Fracture load (kN)	10% AP Fracture load (N)	Cr-m / 1,16 Sphere Diameter (mm)	0°C Compressive strength, MPa (N/mm ²)	Fracture load (kN)	15% APC Fracture load (N)	Cr-m / 1,160 Sphere Diameter (mm)	Compressive strength, MPa (N/mm ²)	
Sample No.	Fracture load (kN) 0.34	10% AP Fracture load (N) 340	Cr-m / 1,16 Sphere Diameter (mm) 11	0°C Compressive strength, MPa (N/mm ²) 2.50	Fracture load (kN) 0.47	15% APC Fracture load (N) 470	Cr-m / 1,160 Sphere Diameter (mm) 11	Compressive strength, MPa (N/mm ²) 3.46	
Sample No.	Fracture load (kN) 0.34 0.37	10% AP Fracture load (N) 340 370	Cr-m / 1,16 Sphere Diameter (mm) 11 10	0°C Compressive strength, MPa (N/mm ²) 2.50 3.30	Fracture load (kN) 0.47 0.72	15% APC Fracture load (N) 470 720	Cr-m / 1,160 Sphere Diameter (mm) 11 11	Compressive strength, MPa (N/mm ²) 3.46 5.30	
Sample No.	Fracture load (kN) 0.34 0.37 0.84	10% AP Fracture load (N) 340 370 840	Cr-m / 1,16 Sphere Diameter (mm) 11 10 10	0°C Compressive strength, MPa (N/mm ²) 2.50 3.30 7.49	Fracture load (kN) 0.47 0.72 0.28	15% APC Fracture load (N) 470 720 280	Cr-m / 1,160 Sphere Diameter (mm) 11 11 12	Compressive strength, MPa (N/mm ²) 3.46 5.30 1.73	
Sample No.	Fracture load (kN) 0.34 0.37 0.84 0.43	10% APC Fracture load (N) 340 370 840 430	Cr-m / 1,16 Sphere Diameter (mm) 11 10 10 11	0°C Compressive strength, MPa (N/mm ²) 2.50 3.30 7.49 3.17	Fracture load (kN) 0.47 0.72 0.28 0.47	15% APC Fracture load (N) 470 720 280 470	Cr-m / 1,160 Sphere Diameter (mm) 11 11 12 10	Compressive strength, MPa (N/mm ²) 3.46 5.30 1.73 4.19	
Sample No. 1 2 3 4 5	Fracture load (kN) 0.34 0.37 0.84 0.43 1.05	10% AP Fracture load (N) 340 370 840 430 1050	Cr-m / 1,16 Sphere Diameter (mm) 11 10 10 11 11 10	0°C Compressive strength, MPa (N/mm ²) 2.50 3.30 7.49 3.17 9.36	Fracture load (kN) 0.47 0.72 0.28 0.47 0.14	15% APC Fracture load (N) 470 720 280 470 140	Cr-m / 1,160 Sphere Diameter (mm) 11 11 12 10 10	Compressive strength, MPa (N/mm ²) 3.46 5.30 1.73 4.19 1.25	
Sample No. 1 2 3 4 5 6	Fracture load (kN) 0.34 0.37 0.84 0.43 1.05 0.57	10% AP Fracture load (N) 340 370 840 430 1050 570	Cr-m / 1,16 Sphere Diameter (mm) 11 10 10 11 10 10 10	0°C Compressive strength, MPa (N/mm ²) 2.50 3.30 7.49 3.17 9.36 5.08	Fracture load (kN) 0.47 0.72 0.28 0.47 0.14 0.31	15% APC Fracture load (N) 470 720 280 470 140 310	Cr-m / 1,160 Sphere Diameter (mm) 11 11 12 10 10 10 10	Compressive strength, MPa (N/mm ²) 3.46 5.30 1.73 4.19 1.25 2.76	
Sample No. 1 2 3 4 5 6 7	Fracture load (kN) 0.34 0.37 0.84 0.43 1.05 0.57 1.00	10% APC Fracture load (N) 340 370 840 430 1050 570 1000	Cr-m / 1,16 Sphere Diameter (mm) 11 10 10 11 10 10 10 10	0°C Compressive strength, MPa (N/mm ²) 2.50 3.30 7.49 3.17 9.36 5.08 8.91	Fracture load (kN) 0.47 0.72 0.28 0.47 0.14 0.31 0.32	15% APC Fracture load (N) 470 720 280 470 140 310 320	Sphere Diameter (mm) 11 12 10 10 10 10 10 10	Compressive strength, MPa (N/mm²) 3.46 5.30 1.73 4.19 1.25 2.76 2.85	
Sample No. 1 2 3 4 5 6 7 8	Fracture load (kN) 0.34 0.37 0.84 0.43 1.05 0.57 1.00 1.13	10% APC Fracture load (N) 340 370 840 430 1050 570 1000 1130	Cr-m / 1,16 Sphere Diameter (mm) 11 10 10 11 10 10 10 10 11	0°C Compressive strength, MPa (N/mm ²) 2.50 3.30 7.49 3.17 9.36 5.08 8.91 8.32	Fracture load (kN) 0.47 0.72 0.28 0.47 0.14 0.31 0.32 0.23	15% APC Fracture load (N) 470 720 280 470 140 310 320 230	Sphere Diameter (mm) 11 12 10 10 10 10 10 10 10 10 10 10 10	Compressive strength, MPa (N/mm²) 3.46 5.30 1.73 4.19 1.25 2.76 2.85 2.05	
Sample No. 1 2 3 4 5 6 7 8 9	Fracture load (kN) 0.34 0.37 0.84 0.43 1.05 0.57 1.00 1.13 0.57	10% APC Fracture load (N) 340 370 840 430 1050 570 1000 1130 570	Cr-m / 1,16 Sphere Diameter (mm) 11 10 10 11 10 10 10 11 10 10	0°C Compressive strength, MPa (N/mm ²) 2.50 3.30 7.49 3.17 9.36 5.08 8.91 8.32 5.08	Fracture load (kN) 0.47 0.72 0.28 0.47 0.14 0.31 0.32 0.23 0.40	15% APC Fracture load (N) 470 720 280 470 140 310 320 230 400	Er-m / 1,160 Sphere Diameter (mm) 11 12 10 10 10 10 10 10 10 10 10 10	Compressive strength, MPa (N/mm²) 3.46 5.30 1.73 4.19 1.25 2.76 2.85 2.05 3.57	
Sample No. 1 2 3 4 5 6 7 8 9 10	Fracture load (kN) 0.34 0.37 0.84 0.43 1.05 0.57 1.00 1.13 0.57 0.40	10% APC Fracture load (N) 340 370 840 430 1050 570 1000 1130 570 400	Cr-m / 1,16 Sphere Diameter (mm) 11 10 10 11 10 10 10 10 10 10	0°C Compressive strength, MPa (N/mm ²) 2.50 3.30 7.49 3.17 9.36 5.08 8.91 8.32 5.08 4.40	Fracture load (kN) 0.47 0.72 0.28 0.47 0.14 0.31 0.32 0.23 0.40 0.25	15% APC Fracture load (N) 470 720 280 470 140 310 320 230 400 250	Er-m / 1,160 Sphere Diameter (mm) 11 12 10 10 10 10 10 10 10 10 10 10	Compressive strength, MPa (N/mm²) 3.46 5.30 1.73 4.19 1.25 2.76 2.85 2.05 3.57 2.23	
Sample No. 1 2 3 4 5 6 7 8 9 10 11	Fracture load (kN) 0.34 0.37 0.84 0.43 1.05 0.57 1.00 1.13 0.57 0.40 0.53	10% AP Fracture load (N) 340 370 840 430 1050 570 1000 1130 570 400 530	Cr-m / 1,16 Sphere Diameter (mm) 11 10 10 11 10 10 10 10 10 11 10 9 10	0°C Compressive strength, MPa (N/mm ²) 2.50 3.30 7.49 3.17 9.36 5.08 8.91 8.32 5.08 4.40 4.72	Fracture load (kN) 0.47 0.72 0.28 0.47 0.14 0.31 0.32 0.23 0.40 0.25 0.45	15% APC Fracture load (N) 470 720 280 470 140 310 320 230 400 250 450	Cr-m / 1,160 Sphere Diameter (mm) 11 12 10 10 10 10 10 10 10 10 10 10	Compressive strength, MPa (N/mm²) 3.46 5.30 1.73 4.19 1.25 2.76 2.85 2.05 3.57 2.23 3.31	
Sample No. 1 2 3 4 5 6 7 8 9 10 11 12	Fracture load (kN) 0.34 0.37 0.84 0.43 1.05 0.57 1.00 1.13 0.57 0.40 0.53 0.84	10% AP Fracture load (N) 340 370 840 430 1050 570 1000 1130 570 400 530 840	Cr-m / 1,16 Sphere Diameter (mm) 11 10 10 11 10 10 10 10 10 10	0°C Compressive strength, MPa (N/mm ²) 2.50 3.30 7.49 3.17 9.36 5.08 8.91 8.32 5.08 4.40 4.72 7.49	Fracture load (kN) 0.47 0.72 0.28 0.47 0.14 0.31 0.32 0.23 0.40 0.25 0.45 0.41	15% APC Fracture load (N) 470 720 280 470 140 310 320 230 400 250 450 410	Cr-m / 1,160 Sphere Diameter (mm) 11 12 10 10 10 10 10 10 10 10 10 10	Compressive strength, MPa (N/mm²) 3.46 5.30 1.73 4.19 1.25 2.76 2.85 2.05 3.57 2.23 3.31 3.02	
Sample No. 1 2 3 4 5 6 7 8 9 10 11 12 13	Fracture load (kN) 0.34 0.37 0.84 0.43 1.05 0.57 1.00 1.13 0.57 0.40 0.53 0.84 0.95	10% AP Fracture load (N) 340 370 840 430 1050 570 1000 1130 570 400 530 840 950	Cr-m / 1,16 Sphere Diameter (mm) 11 10 10 11 10 10 10 10 10 10	0°C Compressive strength, MPa (N/mm ²) 2.50 3.30 7.49 3.17 9.36 5.08 8.91 8.32 5.08 4.40 4.72 7.49 8.47	Fracture load (kN) 0.47 0.72 0.28 0.47 0.14 0.31 0.32 0.23 0.40 0.25 0.45 0.41 0.20	15% APC Fracture load (N) 470 720 280 470 140 310 320 230 400 250 450 410 200	Cr-m / 1,160 Sphere Diameter (mm) 11 11 12 10 10 10 10 10 10 10 10 10 10	Compressive strength, MPa (N/mm²) 3.46 5.30 1.73 4.19 1.25 2.76 2.85 2.05 3.57 2.23 3.31 3.02 1.47	
Sample No. 1 2 3 4 5 6 7 8 9 10 11 12 13 14 15	Fracture load (kN) 0.34 0.37 0.84 0.43 1.05 0.57 1.00 1.13 0.57 0.40 0.53 0.84 0.95	10% AP Fracture load (N) 340 370 840 430 1050 570 1000 1130 570 400 530 840 950	Cr-m / 1,16 Sphere Diameter (mm) 11 10 10 11 10 10 10 11 10 9 10 10 10 10 11 10 10 10 10 10	0°C Compressive strength, MPa (N/mm ²) 2.50 3.30 7.49 3.17 9.36 5.08 8.91 8.32 5.08 4.40 4.72 7.49 8.47	Fracture load (kN) 0.47 0.72 0.28 0.47 0.14 0.31 0.32 0.23 0.40 0.25 0.45 0.45 0.41 0.20 0.28	15% APC Fracture load (N) 470 720 280 470 140 310 320 230 400 250 450 410 200 280	Cr-m / 1,160 Sphere Diameter (mm) 11 11 12 10 10 10 10 10 10 10 10 10 10	Compressive strength, MPa (N/mm²) 3.46 5.30 1.73 4.19 1.25 2.76 2.85 2.05 3.57 2.23 3.31 3.02 1.47 2.50	
Sample No. 1 2 3 4 5 6 7 8 9 10 11 12 13 14 15	Fracture load (kN) 0.34 0.37 0.84 0.43 1.05 0.57 1.00 1.13 0.57 0.40 0.53 0.84 0.95	10% AP Fracture load (N) 340 370 840 430 1050 570 1000 1130 570 400 530 840 950	Cr-m / 1,16 Sphere Diameter (mm) 11 10 10 11 10 10 10 10 10 10	0°C Compressive strength, MPa (N/mm ²) 2.50 3.30 7.49 3.17 9.36 5.08 8.91 8.32 5.08 4.40 4.72 7.49 8.47 6.02	Fracture load (kN) 0.47 0.72 0.28 0.47 0.14 0.31 0.32 0.23 0.40 0.25 0.45 0.41 0.20 0.28	15% APC Fracture load (N) 470 720 280 470 140 310 320 230 400 250 450 410 200 280	Cr-m / 1,160 Sphere Diameter (mm) 11 12 10 11 11 11 11 10 10 10 10 10 11 11 11 11 11 11 11 11 11 11 12 13 14 <td>P°C Compressive strength, MPa (N/mm²) 3.46 5.30 1.73 4.19 1.25 2.76 2.85 2.05 3.57 2.23 3.31 3.02 1.47 2.50 2.84</td>	P°C Compressive strength, MPa (N/mm ²) 3.46 5.30 1.73 4.19 1.25 2.76 2.85 2.05 3.57 2.23 3.31 3.02 1.47 2.50 2.84	
Sample No. 1 2 3 4 5 6 7 8 9 10 11 12 13 14 15	Fracture load (kN) 0.34 0.37 0.84 0.43 1.05 0.57 1.00 1.13 0.57 0.40 0.53 0.84 0.95	10% AP Fracture load (N) 340 370 840 430 1050 570 1000 1130 570 400 530 840 950	Cr-m / 1,16 Sphere Diameter (mm) 11 10 10 11 10 10 10 10 10 10	0°C Compressive strength, MPa (N/mm ²) 2.50 3.30 7.49 3.17 9.36 5.08 8.91 8.32 5.08 4.40 4.72 7.49 8.47 6.02 2.40	Fracture load (kN) 0.47 0.72 0.28 0.47 0.14 0.31 0.32 0.23 0.40 0.25 0.45 0.41 0.20 0.28	15% APC Fracture load (N) 470 720 280 470 140 310 320 230 400 250 450 410 200 280	Image: Cr-m / 1,160 Sphere Diameter (mm) 11 11 12 10 11 11 11 11 10 10 10 10 11 11 11 11 11 10 10 10 10 10 11 11 <td< td=""><td>Compressive strength, MPa (N/mm²) 3.46 5.30 1.73 4.19 1.25 2.76 2.85 2.05 3.57 2.23 3.31 3.02 1.47 2.50</td></td<>	Compressive strength, MPa (N/mm²) 3.46 5.30 1.73 4.19 1.25 2.76 2.85 2.05 3.57 2.23 3.31 3.02 1.47 2.50	
Sample No. 1 2 3 4 5 6 7 8 9 10 11 12 13 14 15	Fracture load (kN) 0.34 0.37 0.84 0.43 1.05 0.57 1.00 1.13 0.57 0.40 0.53 0.84 0.95	10% AP Fracture load (N) 340 370 840 430 1050 570 1000 1130 570 400 530 840 950	Cr-m / 1,16 Sphere Diameter (mm) 11 10 10 11 10 10 10 10 10 10	0°C Compressive strength, MPa (N/mm ²) 2.50 3.30 7.49 3.17 9.36 5.08 8.91 8.32 5.08 4.40 4.72 7.49 8.47 6.02 2.40 2.50	Fracture load (kN) 0.47 0.72 0.28 0.47 0.14 0.31 0.32 0.23 0.40 0.25 0.45 0.41 0.20 0.28	15% APC Fracture load (N) 470 720 280 470 140 310 320 230 400 250 450 410 200 280	Image: Cr-m / 1,160 Sphere Diameter (mm) 11 12 10 11 11 11 11 11 11 10 10 10 11 11 11 11 11 11 11 10 10 10 11 11 10 <td>Compressive strength, MPa (N/mm²) 3.46 5.30 1.73 4.19 1.25 2.76 2.85 2.05 3.57 2.23 3.31 3.02 1.47 2.50</td>	Compressive strength, MPa (N/mm²) 3.46 5.30 1.73 4.19 1.25 2.76 2.85 2.05 3.57 2.23 3.31 3.02 1.47 2.50	

Appendix XIV – Calculation of crushing strength of hot-bonded LW.

Part E

		20% APC	C r-m / 1,16	0°C		Ι	Lytag	
Sample No.	Fracture load (kN)	Fracture load (N)	Sphere Diameter (mm)	Compressive strength, MPa (N/mm²)	Fracture load (kN)	Fracture load (N)	Sphere Diameter (mm)	Compressive strength, MPa (N/mm²)
1	0.29	290	11	2.14	0.25	250	11	1.84
2	0.15	150	10	1.34	0.45	450	10	4.01
3	0.28	280	10	2.50	0.50	500	11	3.68
4	0.14	140	11	1.03	0.69	690	12	4.27
5	0.25	250	10	2.23	0.72	720	12	4.46
6	0.14	140	10	1.25	0.19	190	10	1.69
7	0.20	200	10	1.78	0.51	510	11	3.76
8	0.25	250	11	1.84	0.26	260	11	1.92
9	0.20	200	9	2.20	0.44	440	11	3.24
10	0.15	150	9	1.65	0.52	520	12	3.22
11	0.20	200	8	2.79	0.31	310	11	2.28
12	0.38	380	11	2.80	0.57	570	11	4.20
13	0.29	290	11	2.14	0.20	200	10	1.78
14	0.29	290	11	2.14	0.56	560	11	4.12
15	0.48	480	11	3.54	0.63	630	12	3.90
			Average	2.09			Average	3.23
			SD	0.66			SD	1.03
			Min	1.03			Min	1.69
			Max	3.54			Max	4.46
		100% C	clay / 1,180°	°C		5% APC	r-m / 1,180°	°C

Sample No.	Fracture load (kN)	Fracture load (N)	Sphere Diameter (mm)	Compressive strength, MPa (N/mm²)	Fracture load (kN)	Fracture load (N)	Sphere Diameter (mm)	Compressive strength, MPa (N/mm ²)
1	0.5	500	12	3.09	0.52	520	11	3.83
2	0.62	620	12	3.84	0.51	510	11	3.76
3	0.37	370	11	2.73	0.74	740	12	4.58
4	0.28	280	13	1.48	0.78	780	12	4.83
5	0.38	380	12	2.35	0.68	680	12	4.21
6	0.25	250	12	1.55	0.90	900	12	5.57
7	0.31	310	12	1.92	0.98	980	10	8.73
8	0.61	610	12	3.78	1.56	1560	12	9.66
9	0.61	610	12	3.78	0.97	970	11	7.14
10	0.52	520	12	3.22	0.93	930	11	6.85
11	0.48	480	12	2.97	0.29	290	9	3.19
12	0.68	680	12	4.21	0.37	370	9	4.07
13	0.68	680	13	3.59	0.82	820	11	6.04
14	0.42	420	13	2.21				
15	0.40	400	11	2.95				
			Average	2.91			Average	5.57
			SD	0.86			SD	2.01
			Min	1.48			Min	3.19
			Max	4.21			Max	9.66

Appendix XIV – Calculation of crushing strength of hot-bonded LW	Ά
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Part F

		10% APC	Cr-m / 1,18	0°C		15% APC	Cr-m / 1,180	°C
Sample No.	Fracture load (kN)	Fracture load (N)	Sphere Diameter (mm)	Compressive strength, MPa (N/mm²)	Fracture load (kN)	Fracture load (N)	Sphere Diameter (mm)	Compressive strength, MPa (N/mm²)
1	1.36	1360	11	10.02	0.47	470	11	3.46
2	0.95	950	10	8.47	1.02	1020	12	6.31
3	1.38	1380	10	12.30	0.50	500	11	3.68
4	1.16	1160	10	10.34	0.46	460	9	5.06
5	0.85	850	9	9.35	0.75	750	10	6.68
6	1.75	1750	10	15.60	0.95	950	10	8.47
7	1.25	1250	9	13.75	0.85	850	10	7.58
8	0.85	850	9	9.35	0.50	500	11	3.68
9	1.19	1190	10	10.61	0.87	870	11	6.41
10	1.02	1020	10	9.09	0.50	500	9	5.50
11	1.14	1140	10	10.16	1.12	1120	11	8.25
12	0.88	880	9	9.68	0.58	580	10	5.17
13	1.07	1070	10	9.54	0.78	780	10	6.95
14	1.57	1570	10	13.99	0.84	840	10	7.49
15					0.40	400	10	3.57
			Average	10.87			Average	5.88
			SD	2.16			SD	1.74
			Min	8.47			Min	3.46
			Max	15.60			Max	8.47
5% w-APCr / 1,140°C								
		5% w-A	PCr / 1,140	0°C		10% w-A	PCr / 1,140	°C
Sample No.	Fracture load (kN)	5% w-A Fracture load (N)	<u>PCr / 1,14(</u> Sphere Diameter (mm)	P°C Compressive strength, MPa (N/mm ²)	Fracture load (kN)	10% w-A Fracture load (N)	PCr / 1,140 Sphere Diameter (mm)	°C Compressive strength, MPa (N/mm ²)
Sample No.	Fracture load (kN) 0.17	5% w-A Fracture load (N) 170	PCr / 1,140 Sphere Diameter (mm) 11	Compressive strength, MPa (N/mm ²) 1.25	Fracture load (kN) 0.17	10% w-A Fracture load (N) 170	PCr / 1,140 Sphere Diameter (mm) 11	°C Compressive strength, MPa (N/mm ²) 1.25
Sample No.	Fracture load (kN) 0.17 0.28	5% w-A Fracture load (N) 170 280	PCr / 1,140 Sphere Diameter (mm) 11 11	Compressive strength, MPa (N/mm ²) 1.25 2.06	Fracture load (kN) 0.17 0.17	10% w-A Fracture load (N) 170 170	PCr / 1,140 Sphere Diameter (mm) 11 11	°C Compressive strength, MPa (N/mm ²) 1.25 1.25
Sample No.	Fracture load (kN) 0.17 0.28 0.18	5% w-A Fracture load (N) 170 280 180	PCr / 1,140 Sphere Diameter (mm) 11 11 12	Compressive strength, MPa (N/mm ²) 1.25 2.06 1.11	Fracture load (kN) 0.17 0.17 0.20	10% w-A Fracture load (N) 170 170 200	PCr / 1,140 Sphere Diameter (mm) 11 11 11	°C Compressive strength, MPa (N/mm ²) 1.25 1.25 1.47
Sample No. 1 2 3 4	Fracture load (kN) 0.17 0.28 0.18 0.28	5% w-A Fracture load (N) 170 280 180 280	PCr / 1,140 Sphere Diameter (mm) 11 11 12 11	P°C Compressive strength, MPa (N/mm ²) 1.25 2.06 1.11 2.06	Fracture load (kN) 0.17 0.17 0.20 0.22	10% w-A Fracture load (N) 170 170 200 220	PCr / 1,140 Sphere Diameter (mm) 11 11 11 11	°C Compressive strength, MPa (N/mm ²) 1.25 1.25 1.47 1.62
Sample No. 1 2 3 4 5	Fracture load (kN) 0.17 0.28 0.18 0.28 0.28 0.37	5% w-A Fracture load (N) 170 280 180 280 370	PCr / 1,140 Sphere Diameter (mm) 11 11 12 11 12 11 12	P°C Compressive strength, MPa (N/mm ²) 1.25 2.06 1.11 2.06 2.29	Fracture load (kN) 0.17 0.20 0.22 0.20	10% w-A Fracture load (N) 170 170 200 220 200	PCr / 1,140 Sphere Diameter (mm) 11 11 11 11 11 11	°C Compressive strength, MPa (N/mm ²) 1.25 1.25 1.47 1.62 1.47
Sample No. 1 2 3 4 5 6	Fracture load (kN) 0.17 0.28 0.18 0.28 0.37 0.17	5% w-A Fracture load (N) 170 280 180 280 370 170	PCr / 1,140 Sphere Diameter (mm) 11 11 12 11 12 11 12 11	P°C Compressive strength, MPa (N/mm ²) 1.25 2.06 1.11 2.06 2.29 1.25	Fracture load (kN) 0.17 0.20 0.22 0.20 0.20 0.20	10% w-A Fracture load (N) 170 170 200 220 200 200 200	PCr / 1,140 Sphere Diameter (mm) 11 11 11 11 11 11 11 11	°C Compressive strength, MPa (N/mm ²) 1.25 1.25 1.47 1.62 1.47 1.62 1.47 1.78
Sample No. 1 2 3 4 5 6 7	Fracture load (kN) 0.17 0.28 0.18 0.28 0.37 0.17 0.20	5% w-A Fracture load (N) 170 280 180 280 370 170 200	PCr / 1,140 Sphere Diameter (mm) 11 11 12 11 12 11 12 11 12 11 10	P°C Compressive strength, MPa (N/mm ²) 1.25 2.06 1.11 2.06 2.29 1.25 1.78	Fracture load (kN) 0.17 0.20 0.22 0.20 0.20 0.20 0.10	10% w-A Fracture load (N) 170 170 200 220 200 200 200 100	PCr / 1,140 Sphere Diameter (mm) 11 11 11 11 11 11 10 11	°C Compressive strength, MPa (N/mm ²) 1.25 1.25 1.47 1.62 1.47 1.62 1.47 1.78 0.74
Sample No. 1 2 3 4 5 6 7 8	Fracture load (kN) 0.17 0.28 0.18 0.28 0.37 0.17 0.20 0.17	5% w-A Fracture load (N) 170 280 180 280 370 170 200 170	PCr / 1,140 Sphere Diameter (mm) 11 11 12 11 12 11 12 11 10 11	P°C Compressive strength, MPa (N/mm ²) 1.25 2.06 1.11 2.06 2.29 1.25 1.78 1.25	Fracture load (kN) 0.17 0.20 0.22 0.20 0.20 0.20 0.10 0.20	10% w-A Fracture load (N) 170 170 200 220 200 200 100 200	PCr / 1,140 Sphere Diameter (mm) 11 11 11 11 11 11 10 11 11 11	°C Compressive strength, MPa (N/mm ²) 1.25 1.25 1.47 1.62 1.47 1.78 0.74 1.47
Sample No. 1 2 3 4 5 6 7 8 9	Fracture load (kN) 0.17 0.28 0.18 0.28 0.37 0.17 0.20 0.17 0.23	5% w-A Fracture load (N) 170 280 180 280 370 170 200 170 230	PCr / 1,140 Sphere Diameter (mm) 11 12 11 12 11 12 11 10 11 11 11 12 11 11	Compressive strength, MPa (N/mm ²) 1.25 2.06 1.11 2.06 2.29 1.25 1.78 1.25 1.78 1.25 1.69	Fracture load (kN) 0.17 0.20 0.22 0.20 0.20 0.20 0.10 0.20 0.17	10% w-A Fracture load (N) 170 170 200 220 200 200 100 200 100 200 170	PCr / 1,140 Sphere Diameter (mm) 11 11 11 11 11 10 11 11 11 11	°C Compressive strength, MPa (N/mm ²) 1.25 1.25 1.47 1.62 1.47 1.62 1.47 1.78 0.74 1.47 1.25
Sample No. 1 2 3 4 5 6 7 8 9 10	Fracture load (kN) 0.17 0.28 0.18 0.28 0.37 0.17 0.20 0.17 0.23 0.22	5% w-A Fracture load (N) 170 280 180 280 370 170 200 170 230 220	PCr / 1,140 Sphere Diameter (mm) 11 11 12 11 12 11 12 11 10 11 11 11 11 11 11 11 11	P°C Compressive strength, MPa (N/mm ²) 1.25 2.06 1.11 2.06 2.29 1.25 1.78 1.25 1.78 1.25 1.69 1.62	Fracture load (kN) 0.17 0.20 0.22 0.20 0.20 0.20 0.10 0.20 0.17 0.12	10% w-A Fracture load (N) 170 200 220 200 200 200 100 200 170 120	PCr / 1,140 Sphere Diameter (mm) 11 11 11 11 11 11 10 11 11 11 11 11 11	°C Compressive strength, MPa (N/mm ²) 1.25 1.25 1.47 1.62 1.47 1.78 0.74 1.47 1.78 0.74 1.47 1.25 1.07
Sample No. 1 2 3 4 5 6 7 8 9 10 11	Fracture load (kN) 0.17 0.28 0.18 0.28 0.37 0.17 0.20 0.17 0.20 0.17 0.23 0.22 0.18	5% w-A Fracture load (N) 170 280 180 280 370 170 200 170 230 220 180	PCr / 1,140 Sphere Diameter (mm) 11 11 12 11 12 11 12 11 10 11 11 11 11 11 11 11 11	P°C Compressive strength, MPa (N/mm ²) 1.25 2.06 1.11 2.06 2.29 1.25 1.78 1.25 1.69 1.62 1.33	Fracture load (kN) 0.17 0.20 0.22 0.20 0.20 0.20 0.10 0.20 0.17 0.12 0.20	10% w-A Fracture load (N) 170 200 220 200 200 100 200 170 120 200	PCr / 1,140 Sphere Diameter (mm) 11 11 11 11 11 10 11 11 11 10 11	°C Compressive strength, MPa (N/mm ²) 1.25 1.25 1.47 1.62 1.47 1.62 1.47 1.78 0.74 1.47 1.25 1.07 1.47
Sample No. 1 2 3 4 5 6 7 8 9 10 11 12	Fracture load (kN) 0.17 0.28 0.18 0.28 0.37 0.17 0.20 0.17 0.20 0.17 0.23 0.22 0.18 0.18	5% w-A Fracture load (N) 170 280 180 280 370 170 200 170 230 220 180 180	PCr / 1,140 Sphere Diameter (mm) 11 11 12 11 12 11 12 11 10 11 11 11 11 11 11 11 11	P°C Compressive strength, MPa (N/mm ²) 1.25 2.06 1.11 2.06 2.29 1.25 1.78 1.25 1.69 1.62 1.33 1.33	Fracture load (kN) 0.17 0.20 0.22 0.20 0.20 0.20 0.10 0.20 0.17 0.12 0.20 0.14	10% w-A Fracture load (N) 170 200 220 200 200 100 200 100 200 170 120 200 140	PCr / 1,140 Sphere Diameter (mm) 11 11 11 11 11 10 11 11 10 11 11 10 11 11	°C Compressive strength, MPa (N/mm ²) 1.25 1.25 1.47 1.62 1.47 1.62 1.47 1.78 0.74 1.47 1.25 1.07 1.47 1.25 1.07 1.47 1.25 1.07 1.47 1.03
Sample No. 1 2 3 4 5 6 7 8 9 10 11 12 13	Fracture load (kN) 0.17 0.28 0.18 0.28 0.37 0.17 0.20 0.17 0.20 0.17 0.23 0.22 0.18 0.18 0.18 0.21	5% w-A Fracture load (N) 170 280 180 280 370 170 200 170 200 170 230 220 180 180 180 210	PCr / 1,140 Sphere Diameter (mm) 11 11 12 11 12 11 12 11 10 11 11 11 11 11 11 11 11	P°C Compressive strength, MPa (N/mm ²) 1.25 2.06 1.11 2.06 2.29 1.25 1.78 1.25 1.69 1.62 1.33 1.33 1.55	Fracture load (kN) 0.17 0.20 0.22 0.20 0.20 0.20 0.10 0.20 0.17 0.12 0.20 0.14 0.15	10% w-A Fracture load (N) 170 200 220 200 200 100 200 100 200 170 120 200 140 150	PCr / 1,140 Sphere Diameter (mm) 11 11 11 11 11 10 11 11 11 10 11 11 11	°C Compressive strength, MPa (N/mm ²) 1.25 1.25 1.47 1.62 1.47 1.62 1.47 1.78 0.74 1.47 1.25 1.07 1.47 1.25 1.07 1.47 1.25 1.07 1.47 1.25 1.07 1.47 1.25 1.07 1.47 1.25 1.07 1.47 1.25 1.07 1.47 1.25 1.07 1.25 1.07 1.17 1.02 1.17 1.02 1.17 1.17 1.02 1.17 1.17 1.02 1.17 1.17 1.02 1.17 1.17 1.02 1.17 1.17 1.02 1.17 1.17 1.02 1.17 1.17 1.02 1.17 1.17 1.02 1.17 1.17 1.02 1.17 1.17 1.02 1.17 1.103 1.10
Sample No. 1 2 3 4 5 6 7 8 9 10 11 12 13 14	Fracture load (kN) 0.17 0.28 0.18 0.28 0.37 0.17 0.20 0.17 0.20 0.17 0.23 0.22 0.18 0.18 0.18 0.21 0.21	5% w-A Fracture load (N) 170 280 180 280 370 170 200 170 230 220 180 180 180 210 210	PCr / 1,140 Sphere Diameter (mm) 11 12 11 12 11 12 11 12 11 10 11 11 11 11 11 11 11 11	P°C Compressive strength, MPa (N/mm ²) 1.25 2.06 1.11 2.06 2.29 1.25 1.78 1.25 1.78 1.25 1.69 1.62 1.33 1.33 1.55 1.55	Fracture load (kN) 0.17 0.20 0.22 0.20 0.20 0.20 0.10 0.20 0.17 0.12 0.20 0.14 0.15 0.15	10% w-A Fracture load (N) 170 200 220 200 200 100 200 100 200 170 120 200 140 150 150	PCr / 1,140 Sphere Diameter (mm) 11 11 11 11 11 11 10 11 11 11 11 11 11	°C Compressive strength, MPa (N/mm ²) 1.25 1.25 1.47 1.62 1.47 1.62 1.47 1.78 0.74 1.47 1.25 1.07 1.47 1.25 1.07 1.47 1.25 1.07 1.47 1.25 1.07 1.47 1.25 1.07 1.47 1.25 1.07 1.47 1.25 1.07 1.47 1.25 1.07 1.47 1.25 1.07 1.47 1.25 1.07 1.47 1.25 1.07 1.47 1.25 1.47 1.78 0.74 1.47 1.25 1.07 1.47 1.25 1.07 1.47 1.25 1.47 1.78 0.74 1.47 1.62 1.47 1.25 1.07 1.47 1.25 1.07 1.47 1.25 1.47 1.47 1.78 0.74 1.47 1.47 1.25 1.07 1.47 1.47 1.25 1.07 1.47 1.47 1.47 1.47 1.25 1.07 1.47 1.47 1.47 1.47 1.25 1.07 1.47 1.03 1.10 1.10
Sample No. 1 2 3 4 5 6 7 8 9 10 11 12 13 14 15	Fracture load (kN) 0.17 0.28 0.18 0.28 0.37 0.17 0.20 0.17 0.20 0.17 0.23 0.22 0.18 0.18 0.21 0.21 0.18	5% w-A Fracture load (N) 170 280 180 280 370 170 200 170 200 170 230 220 180 180 180 210 210 180	PCr / 1,140 Sphere Diameter (mm) 11 12 11 12 11 12 11 12 11 10 11 11 11 11 11 11 11 11	P°C Compressive strength, MPa (N/mm ²) 1.25 2.06 1.11 2.06 2.29 1.25 1.78 1.25 1.69 1.62 1.33 1.33 1.55 1.55 1.33	Fracture load (kN) 0.17 0.17 0.20 0.20 0.20 0.10 0.20 0.110 0.20 0.10 0.20 0.110 0.20 0.110 0.20 0.15 0.15 0.17	10% w-A Fracture load (N) 170 200 220 200 200 100 200 100 200 170 120 200 140 150 150 150 170	PCr / 1,140 Sphere Diameter (mm) 11 11 11 11 11 11 11 11 11 11 11 11 11	°C Compressive strength, MPa (N/mm ²) 1.25 1.25 1.47 1.62 1.47 1.62 1.47 1.78 0.74 1.47 1.25 1.07 1.47 1.25 1.07 1.47 1.25 1.07 1.47 1.25 1.07 1.47 1.25 1.25 1.47 1.52 1.07 1.47 1.52 1.07 1.47 1.52 1.07 1.47 1.52 1.07 1.47 1.52 1.07 1.47 1.52 1.07 1.47 1.52 1.07 1.47 1.52 1.07 1.47 1.52 1.07 1.47 1.52 1.07 1.47 1.52 1.07 1.47 1.52 1.07 1.47 1.52 1.07 1.47 1.52 1.07 1.47 1.52 1.07 1.52 1.07 1.52 1.07 1.52 1.07 1.55 1.55
Sample No. 1 2 3 4 5 6 7 8 9 10 11 12 13 14 15	Fracture load (kN) 0.17 0.28 0.18 0.28 0.37 0.17 0.20 0.17 0.20 0.17 0.23 0.22 0.18 0.21 0.21 0.18	5% w-A Fracture load (N) 170 280 180 280 370 170 200 170 200 170 230 220 180 180 210 210 180	PCr / 1,140 Sphere Diameter (mm) 11 12 11 12 11 12 11 12 11 10 11 11 11 11 11 11 11 11	P°C Compressive strength, MPa (N/mm ²) 1.25 2.06 1.11 2.06 2.29 1.25 1.78 1.25 1.69 1.62 1.33 1.55 1.55 1.33 1.56	Fracture load (kN) 0.17 0.17 0.20 0.20 0.20 0.10 0.20 0.17 0.10 0.12 0.10 0.117 0.12 0.12 0.15 0.15 0.17	10% w-A Fracture load (N) 170 170 200 200 200 100 200 100 200 170 120 200 140 150 150 150 170	PCr / 1,140 Sphere Diameter (mm) 11 11 11 11 11 11 11 11 11 11 11 11 11	°C Compressive strength, MPa (N/mm ²) 1.25 1.25 1.47 1.62 1.47 1.62 1.47 1.78 0.74 1.47 1.25 1.07 1.47 1.25 1.07 1.47 1.25 1.07 1.47 1.25 1.25 1.29
Sample No. 1 2 3 4 5 6 7 8 9 10 11 12 13 14 15	Fracture load (kN) 0.17 0.28 0.18 0.28 0.37 0.17 0.20 0.17 0.23 0.22 0.18 0.22 0.18 0.21 0.21 0.18	5% w-A Fracture load (N) 170 280 180 280 370 170 200 170 230 220 180 180 210 210 180	PCr / 1,140 Sphere Diameter (mm) 11 12 11 12 11 12 11 10 11 11 11 11 11 11 11 11	Compressive strength, MPa (N/mm²) 1.25 2.06 1.11 2.06 1.11 2.06 1.11 2.06 1.11 2.06 1.25 1.62 1.33 1.55 1.55 1.33 1.55 1.33 1.56 0.35	Fracture load (kN) 0.17 0.20 0.22 0.20 0.20 0.10 0.20 0.17 0.12 0.20 0.17 0.12 0.20 0.14 0.15 0.15 0.17	10% w-A Fracture load (N) 170 170 200 220 200 100 200 170 120 200 170 120 200 140 150 150 150	PCr / 1,140 Sphere Diameter (mm) 11 11 11 11 11 11 11 11 11 11 11 11 11	°C Compressive strength, MPa (N/mm ²) 1.25 1.25 1.47 1.62 1.47 1.62 1.47 1.78 0.74 1.47 1.25 1.07 1.47 1.25 1.07 1.47 1.25 1.07 1.47 1.25 1.25 1.25 1.25 1.25 1.25 1.25 1.25 1.25 1.25 1.47 1.62 1.47 1.62 1.47 1.62 1.47 1.62 1.47 1.62 1.47 1.62 1.47 1.62 1.47 1.62 1.47 1.25 1.25 1.25 1.25 1.47 1.62 1.47 1.62 1.47 1.62 1.47 1.62 1.47 1.62 1.47 1.62 1.47 1.62 1.47 1.62 1.47 1.25 1.25 1.25 1.25 1.47 1.62 1.47 1.62 1.47 1.62 1.47 1.55 1.07 1.47 1.25 1.07 1.47 1.25 1.07 1.47 1.03 1.10 1.10 1.25 1.25 1.25 1.07 1.47 1.25 1.07 1.47 1.03 1.10 1.25 1.25 1.25 1.25 1.07 1.47 1.25 1.07 1.47 1.25 1.07 1.47 1.03 1.10 1.25 1.29 0.26
Sample No. 1 2 3 4 5 6 7 8 9 10 11 12 13 14 15	Fracture load (kN) 0.17 0.28 0.18 0.28 0.37 0.17 0.20 0.17 0.23 0.22 0.18 0.22 0.18 0.21 0.21 0.18	5% w-A Fracture load (N) 170 280 180 280 370 170 200 170 230 220 180 180 210 210 180	PCr / 1,140 Sphere Diameter (mm) 11 11 12 11 12 11 12 11 10 11 11 11 11 11 11 11 11	Compressive strength, MPa (N/mm²) 1.25 2.06 1.11 2.06 1.11 2.06 1.11 2.06 1.11 2.06 1.25 1.78 1.25 1.69 1.62 1.33 1.55 1.33 1.55 1.33 1.56 0.35 1.11	Fracture load (kN) 0.17 0.20 0.22 0.20 0.20 0.10 0.20 0.17 0.12 0.20 0.17 0.12 0.20 0.14 0.15 0.15 0.17	10% w-A Fracture load (N) 170 200 220 200 100 200 100 200 170 120 200 140 150 150 150 170	PCr / 1,140 Sphere Diameter (mm) 11 11 11 11 11 11 11 11 11 1	°C Compressive strength, MPa (N/mm ²) 1.25 1.25 1.47 1.62 1.47 1.62 1.47 1.78 0.74 1.47 1.25 1.07 1.47 1.25 1.07 1.47 1.25 1.07 1.47 1.25 1.25 1.25 1.25 1.47 1.62 1.47 1.55 1.07 1.47 1.25 1.07 1.47 1.25 1.07 1.47 1.25 1.07 1.47 1.62 1.47 1.25 1.07 1.47 1.25 1.07 1.47 1.25 1.07 1.47 1.03 1.10 1.25 1.29 0.26 0.74

Appendix XIV – Calculation	of crushing strength	of hot-bonded LWA
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Part G

	15% w-APCr / 1,140°C					20% w-APCr / 1,140°C			
Sample No.	Fracture load (kN)	Fracture load (N)	Sphere Diameter (mm)	Compressive strength, MPa (N/mm ²)	Fracture load (kN)	Fracture load (N)	Sphere Diameter (mm)	Compressive strength, MPa (N/mm ²)	
1	0.21	210	10	1.87	0.10	100	10	0.89	
2	0.15	150	11	1.10	0.21	210	9	2.31	
3	0.17	170	10	1.52	0.07	70	10	0.62	
4	0.11	110	11	0.81	0.25	250	10	2.23	
5	0.11	110	11	0.81	0.18	180	10	1.60	
6	0.17	170	10	1.52	0.08	80	10	0.71	
7	0.28	280	11	2.06	0.17	170	10	1.52	
8	0.28	280	11	2.06	0.15	150	10	1.34	
9	0.20	200	11	1.47	0.21	210	10	1.87	
10	0.17	170	10	1.52	0.21	210	10	1.87	
11	0.15	150	10	1.34	0.22	220	9	2.42	
12	0.11	110	11	0.81	0.17	170	11	1.25	
13	0.23	230	11	1.69	0.17	170	10	1.52	
14	0.21	210	10	1.87	0.20	200	10	1.78	
15	0.20	200	10	1.78					
			Average	1.48			Average	1.57	
			SD	0.43			SD	0.57	
			Min	0.81			Min	0.62	
			Max	2.06			Max	2.42	

	25% w-APCr / 1,140°C					30% w-APCr / 1,140°C			
Sample No.	Fracture load (kN)	Fracture load (N)	Sphere Diameter (mm)	Compressive strength, MPa (N/mm ²)	Fracture load (kN)	Fracture load (N)	Sphere Diameter (mm)	Compressive strength, MPa (N/mm²)	
1	0.14	140	11	1.03	0.17	170	11	1.25	
2	0.14	140	11	1.03	0.14	140	11	1.03	
3	0.1	100	10	0.89	0.15	150	11	1.10	
4	0.17	170	10	1.52	0.11	110	11	0.81	
5	0.11	110	10	0.98	0.12	120	12	0.74	
6	0.29	290	11	2.14	0.12	120	11	0.88	
7	0.15	150	10	1.34	0.12	120	12	0.74	
8	0.26	260	10	2.32	0.08	80	12	0.50	
9	0.10	100	10	0.89	0.02	20	10	0.18	
10	0.14	140	10	1.25	0.08	80	11	0.59	
11	0.21	210	11	1.55	0.05	50	11	0.37	
12	0.26	260	11	1.92	0.07	70	10	0.62	
13	0.15	150	11	1.10	0.10	100	11	0.74	
14	0.14	140	11	1.03	0.07	70	11	0.52	
15	0.10	100	10	0.89	0.07	70	11	0.52	
			Average	1.32			Average	0.71	
			SD	0.47			SD	0.29	
			Min	0.89			Min	0.18	
			Max	2.32			Max	1.25	

Appendix XIV – Calculation of crushing strength of hot-bonded LWA

Part H

		5% w-A	PCr / 1,160)°C	10% w-APCr / 1,160°C			
Sample No.	Fracture load (kN)	Fracture load (N)	Sphere Diameter (mm)	Compressive strength, MPa (N/mm ²)	Fracture load (kN)	Fracture load (N)	Sphere Diameter (mm)	Compressive strength, MPa (N/mm ²)
1	0.10	100	11	0.74	0.07	70	11	0.52
2	0.15	150	11	1.10	0.08	80	10	0.71
3	0.08	80	10	0.71	0.10	100	11	0.74
4	0.14	140	11	1.03	0.12	120	11	0.88
5	0.14	140	11	1.03	0.10	100	11	0.74
6	0.10	100	11	0.74	0.07	70	11	0.52
7	0.10	100	11	0.74	0.07	70	11	0.52
8	0.12	120	11	0.88	0.07	70	11	0.52
9	0.08	80	11	0.59	0.07	70	11	0.52
10	0.15	150	11	1.10	0.10	100	11	0.74
11	0.14	140	11	1.03	0.08	80	11	0.59
12	0.17	170	11	1.25	0.10	100	11	0.74
13	0.14	140	11	1.03	0.07	70	11	0.52
14	0.10	100	10	0.89	0.07	70	11	0.52
15	0.10	100	11	0.74	0.07	70	11	0.52
			Average	0.91			Average	0.62
			SD	0.19			SD	0.13
			Min	0.59			Min	0.52
			Max	1.25			Max	0.88
159/ w ADC / 1 160°C						200/ w A	PCr / 1 160	°C

		15% w-A	APCr / 1,16	0°C	20% w-APCr / 1,160°C			
Sample No.	Fracture load (kN)	Fracture load (N)	Sphere Diameter (mm)	Compressive strength, MPa (N/mm ²)	Fracture load (kN)	Fracture load (N)	Sphere Diameter (mm)	Compressive strength, MPa (N/mm²)
1	0.17	170	10	1.52	0.20	200	10	1.78
2	0.20	200	10	1.78	0.26	260	10	2.32
3	0.17	170	10	1.52	0.20	200	10	1.78
4	0.38	380	10	3.39	0.26	260	10	2.32
5	0.18	180	10	1.60	0.23	230	10	2.05
6	0.25	250	11	1.84	0.37	370	10	3.30
7	0.26	260	10	2.32	0.41	410	10	3.65
8	0.18	180	10	1.60	0.21	210	10	1.87
9	0.17	170	11	1.25	0.37	370	10	3.30
10	0.28	280	10	2.50	0.25	250	10	2.23
11	0.15	150	11	1.10	0.26	260	9	2.86
12	0.17	170	11	1.25	0.28	280	9	3.08
13	0.31	310	10	2.76	0.27	270	10	2.41
14	0.14	140	10	1.25	0.25	250	10	2.23
15	0.17	170	10	1.52	0.25	250	9	2.75
			Average	1.81			Average	2.53
			SD	0.65			SD	0.60
			Min	1.10			Min	1.78
			Max	3.39			Max	3.65

Appendix XIV – Calculation	of crushing strength	of hot-bonded LWA
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Part I

	25% w-APCr / 1,160°C					30% w-APCr / 1,160°C			
Sample No.	Fracture load (kN)	Fracture load (N)	Sphere Diameter (mm)	Compressive strength, MPa (N/mm ²)	Fracture load (kN)	Fracture load (N)	Sphere Diameter (mm)	Compressive strength, MPa (N/mm ²)	
1	0.37	370	9	4.07	0.20	200	10	1.78	
2	0.47	470	10	4.19	0.26	260	10	2.32	
3	0.51	510	11	3.76	0.20	200	10	1.78	
4	0.53	530	10	4.72	0.26	260	10	2.32	
5	0.37	370	9	4.07	0.17	170	10	1.52	
6	0.57	570	10	5.08	0.18	180	10	1.60	
7	0.56	560	11	4.12	0.23	230	10	2.05	
8	0.31	310	9	3.41	0.17	170	10	1.52	
9	0.56	560	11	4.12	0.37	370	10	3.30	
10	0.50	500	11	3.68	0.15	150	10	1.34	
11	0.35	350	9	3.85	0.21	210	10	1.87	
12	0.52	520	10	4.63	0.19	190	10	1.69	
13	0.59	590	11	4.35	0.23	230	9	2.53	
14	0.45	450	10	4.01	0.15	150	9	1.65	
15	0.40	400	9	4.40	0.21	210	10	1.87	
			Average	4.17			Average	1.94	
			SD	0.43			SD	0.50	
			Min	3.41			Min	1.34	
			Max	5.08			Max	3 30	
			1716628	5.00			1 11 4A	5.50	
		5% w-A	PCr / 1,180	0°C		10% w-A	PCr / 1,180	°C	
Sample No.	Fracture load (kN)	5% w-A Fracture load (N)	PCr / 1,180 Sphere Diameter (mm))°C Compressive strength, MPa (N/mm ²)	Fracture load (kN)	10% w-A Fracture load (N)	PCr / 1,180 Sphere Diameter (mm)	°C Compressive strength, MPa (N/mm ²)	
Sample No.	Fracture load (kN) 0.17	5% w-A Fracture load (N) 170	PCr / 1,180 Sphere Diameter (mm) 12	P°C Compressive strength, MPa (N/mm ²) 1.05	Fracture load (kN) 0.22	10% w-A Fracture load (N) 220	PCr / 1,180 Sphere Diameter (mm) 10	°C Compressive strength, MPa (N/mm ²) 1.96	
Sample No.	Fracture load (kN) 0.17 0.35	5% w-A Fracture load (N) 170 350	PCr / 1,180 Sphere Diameter (mm) 12 11	P°C Compressive strength, MPa (N/mm ²) 1.05 2.58	Fracture load (kN) 0.22 0.22	10% w-A Fracture load (N) 220 220	PCr / 1,180 Sphere Diameter (mm) 10 11	°C Compressive strength, MPa (N/mm ²) 1.96 1.62	
Sample No. 1 2 3	Fracture load (kN) 0.17 0.35 0.14	5% w-A Fracture load (N) 170 350 140	PCr / 1,180 Sphere Diameter (mm) 12 11 11	P°C Compressive strength, MPa (N/mm ²) 1.05 2.58 1.03	Fracture load (kN) 0.22 0.22 0.25	10% w-A Fracture load (N) 220 220 250	PCr / 1,180 Sphere Diameter (mm) 10 11 11	°C Compressive strength, MPa (N/mm ²) 1.96 1.62 1.84	
Sample No.	Fracture load (kN) 0.17 0.35 0.14 0.31	5% w-A Fracture load (N) 170 350 140 310	PCr / 1,180 Sphere Diameter (mm) 12 11 11 11 10	S.06 O°C Compressive strength, MPa (N/mm ²) 1.05 2.58 1.03 2.76	Fracture load (kN) 0.22 0.22 0.25 0.32	10% w-A Fracture load (N) 220 220 250 320	PCr / 1,180 Sphere Diameter (mm) 10 11 11 11 10	*C Compressive strength, MPa (N/mm ²) 1.96 1.62 1.84 2.85	
Sample No. 1 2 3 4 5	Fracture load (kN) 0.17 0.35 0.14 0.31 0.11	5% w-A Fracture load (N) 170 350 140 310 110	PCr / 1,180 Sphere Diameter (mm) 12 11 11 10 11	S.00 Strength, MPa (N/mm²) 1.05 2.58 1.03 2.76 0.81	Fracture load (kN) 0.22 0.22 0.25 0.32 0.37	10% w-A Fracture load (N) 220 220 250 320 370	PCr / 1,180 Sphere Diameter (mm) 10 11 11 11 10 11	*C Compressive strength, MPa (N/mm ²) 1.96 1.62 1.84 2.85 2.73	
Sample No. 1 2 3 4 5 6	Fracture load (kN) 0.17 0.35 0.14 0.31 0.11 0.15	5% w-A Fracture load (N) 170 350 140 310 110 150	PCr / 1,180 Sphere Diameter (mm) 12 11 11 10 11 10 11 12	S.00 Strength, MPa (N/mm²) 1.05 2.58 1.03 2.76 0.81 0.93	Fracture load (kN) 0.22 0.22 0.25 0.32 0.37 0.15	10% w-A Fracture load (N) 220 220 250 320 370 150	PCr / 1,180 Sphere Diameter (mm) 10 11 11 10 11 10 11	°C Compressive strength, MPa (N/mm ²) 1.96 1.62 1.84 2.85 2.73 1.10	
Sample No. 1 2 3 4 5 6 7	Fracture load (kN) 0.17 0.35 0.14 0.31 0.11 0.15 0.11	5% w-A Fracture load (N) 170 350 140 310 110 150 110	PCr / 1,180 Sphere Diameter (mm) 12 11 11 10 11 12 11 12 11	S.00 Strength, MPa (N/mm²) 1.05 2.58 1.03 2.76 0.81 0.93 0.81	Fracture load (kN) 0.22 0.22 0.25 0.32 0.37 0.15 0.14	10% w-A Fracture load (N) 220 220 250 320 370 150 140	PCr / 1,180 Sphere Diameter (mm) 10 11 11 10 11 11 11 11	°C Compressive strength, MPa (N/mm ²) 1.96 1.62 1.84 2.85 2.73 1.10 1.03	
Sample No. 1 2 3 4 5 6 7 8	Fracture load (kN) 0.17 0.35 0.14 0.31 0.11 0.15 0.11 0.10	5% w-A Fracture load (N) 170 350 140 310 110 150 110 100	PCr / 1,180 Sphere Diameter (mm) 12 11 11 10 11 12 11 12 11 12 11	S.00 °C Compressive strength, MPa (N/mm²) 1.05 2.58 1.03 2.76 0.81 0.93 0.81 0.74	Fracture load (kN) 0.22 0.22 0.25 0.32 0.37 0.15 0.14 0.12	10% w-A Fracture load (N) 220 220 250 320 370 150 140 120	PCr / 1,180 Sphere Diameter (mm) 10 11 11 10 11 11 11 11 11	°C Compressive strength, MPa (N/mm ²) 1.96 1.62 1.84 2.85 2.73 1.10 1.03 0.88	
Sample No. 1 2 3 4 5 6 7 8 9	Fracture load (kN) 0.17 0.35 0.14 0.31 0.11 0.15 0.11 0.10 0.17	5% w-A Fracture load (N) 170 350 140 310 110 150 110 100 170	PCr / 1,180 Sphere Diameter (mm) 12 11 11 10 11 12 11 12 11 11 11 10	P°C Compressive strength, MPa (N/mm ²) 1.05 2.58 1.03 2.76 0.81 0.93 0.81 0.74 1.52	Fracture load (kN) 0.22 0.22 0.25 0.32 0.37 0.15 0.14 0.12 0.14	10% w-A Fracture load (N) 220 220 250 320 370 150 140 120 140	PCr / 1,180 Sphere Diameter (mm) 10 11 11 11 11 11 11 11 11 11	°C Compressive strength, MPa (N/mm ²) 1.96 1.62 1.84 2.85 2.73 1.10 1.03 0.88 1.03	
Sample No. 1 2 3 4 5 6 7 8 9 10	Fracture load (kN) 0.17 0.35 0.14 0.31 0.11 0.15 0.11 0.10 0.17 0.14	5% w-A Fracture load (N) 170 350 140 310 110 150 110 100 170 140	PCr / 1,180 Sphere Diameter (mm) 12 11 11 10 11 12 11 12 11 11 10 11	S.00 Compressive strength, MPa (N/mm²) 1.05 2.58 1.03 2.76 0.81 0.93 0.81 0.74 1.52 1.03	Fracture load (kN) 0.22 0.22 0.25 0.32 0.37 0.15 0.14 0.12 0.14 0.20	10% w-A Fracture load (N) 220 220 250 320 370 150 140 120 140 200	PCr / 1,180 Sphere Diameter (mm) 10 11 11 10 11 11 11 11 11 11	°C Compressive strength, MPa (N/mm ²) 1.96 1.62 1.84 2.85 2.73 1.10 1.03 0.88 1.03 1.78	
Sample No. 1 2 3 4 5 6 7 8 9 10 11	Fracture load (kN) 0.17 0.35 0.14 0.31 0.11 0.15 0.11 0.10 0.17 0.14 0.14	5% w-A Fracture load (N) 170 350 140 310 110 150 110 100 170 140 140 140 140	PCr / 1,180 Sphere Diameter (mm) 12 11 11 10 11 12 11 11 10 11 11 10 11 11 10 11 11	S.08 Compressive strength, MPa (N/mm²) 1.05 2.58 1.03 2.76 0.81 0.93 0.81 0.74 1.52 1.03 1.03	Fracture load (kN) 0.22 0.22 0.25 0.32 0.37 0.15 0.14 0.12 0.14 0.12 0.14 0.20 0.46	10% w-A Fracture load (N) 220 220 250 320 370 150 140 120 140 200 460	PCr / 1,180 Sphere Diameter (mm) 10 11 11 10 11 11 11 11 11 11	°C Compressive strength, MPa (N/mm ²) 1.96 1.62 1.84 2.85 2.73 1.10 1.03 0.88 1.03 1.78 3.39	
Sample No. 1 2 3 4 5 6 7 8 9 10 11 12	Fracture load (kN) 0.17 0.35 0.14 0.31 0.11 0.15 0.11 0.10 0.17 0.14 0.14 0.14	5% w-A Fracture load (N) 170 350 140 310 110 150 110 100 170 140 140 140 140 140	PCr / 1,180 Sphere Diameter (mm) 12 11 11 10 11 12 11 11 10 11 11 10 11 11 11 11	P°C Compressive strength, MPa (N/mm ²) 1.05 2.58 1.03 2.76 0.81 0.93 0.81 0.74 1.52 1.03 1.03 1.03	Fracture load (kN) 0.22 0.22 0.25 0.32 0.37 0.15 0.14 0.12 0.14 0.20 0.46 0.28	10% w-A Fracture load (N) 220 250 320 370 150 140 120 140 200 460 280	PCr / 1,180 Sphere Diameter (mm) 10 11 11 11 11 11 11 11 11 11	°C Compressive strength, MPa (N/mm ²) 1.96 1.62 1.84 2.85 2.73 1.10 1.03 0.88 1.03 1.78 3.39 2.06	
Sample No. 1 2 3 4 5 6 7 8 9 10 11 12 13	Fracture load (kN) 0.17 0.35 0.14 0.31 0.11 0.15 0.11 0.10 0.17 0.14 0.14 0.14 0.14 0.20	5% w-A Fracture load (N) 170 350 140 310 110 150 110 150 110 100 170 140 140 140 140 200	PCr / 1,180 Sphere Diameter (mm) 12 11 11 10 11 12 11 11 10 11 11 10 11 11 10 11 11	S.00 Compressive strength, MPa (N/mm²) 1.05 2.58 1.03 2.76 0.81 0.93 0.81 0.74 1.52 1.03 1.03 1.78	Fracture load (kN) 0.22 0.22 0.25 0.32 0.37 0.15 0.14 0.12 0.14 0.20 0.46 0.28 0.28	10% w-A Fracture load (N) 220 220 250 320 370 150 140 120 140 200 460 280 280	PCr / 1,180 Sphere Diameter (mm) 10 11 11 10 11 11 11 11 11 11 11 11 10 11 11	°C Compressive strength, MPa (N/mm ²) 1.96 1.62 1.84 2.85 2.73 1.10 1.03 0.88 1.03 1.78 3.39 2.06 2.50	
Sample No. 1 2 3 4 5 6 7 8 9 10 11 12 13 14	Fracture load (kN) 0.17 0.35 0.14 0.31 0.11 0.15 0.11 0.15 0.11 0.10 0.17 0.14 0.14 0.14 0.14 0.20 0.20	5% w-A Fracture load (N) 170 350 140 310 110 150 110 150 110 100 170 140 140 140 140 200 200	PCr / 1,180 Sphere Diameter (mm) 12 11 11 10 11 12 11 11 10 11 11 10 11 11 10 11 11	S.00 Compressive strength, MPa (N/mm²) 1.05 2.58 1.03 2.76 0.81 0.93 0.81 0.74 1.52 1.03 1.03 1.78 1.24	Fracture load (kN) 0.22 0.22 0.25 0.32 0.37 0.15 0.14 0.12 0.14 0.20 0.46 0.28 0.28 0.28 0.14	10% w-A Fracture load (N) 220 220 250 320 370 150 140 120 140 200 460 280 280 280 140	PCr / 1,180 Sphere Diameter (mm) 10 11 11 10 11 11 11 11 11 11	°C Compressive strength, MPa (N/mm ²) 1.96 1.62 1.84 2.85 2.73 1.10 1.03 0.88 1.03 1.78 3.39 2.06 2.50 1.03	
Sample No. 1 2 3 4 5 6 7 8 9 10 11 12 13 14 15	Fracture load (kN) 0.17 0.35 0.14 0.31 0.11 0.15 0.11 0.15 0.11 0.10 0.17 0.14 0.14 0.14 0.20 0.20 0.10	5% w-A Fracture load (N) 170 350 140 310 110 150 110 100 170 140 140 140 140 200 200 100	PCr / 1,180 Sphere Diameter (mm) 12 11 11 10 11 12 11 11 10 11 11 10 11 11 10 11 11	S.08 Compressive strength, MPa (N/mm²) 1.05 2.58 1.03 2.76 0.81 0.93 0.81 0.74 1.52 1.03 1.03 1.78 1.24 0.74	Fracture load (kN) 0.22 0.22 0.25 0.32 0.37 0.15 0.14 0.12 0.14 0.20 0.46 0.28 0.28 0.14 0.28	10% w-A Fracture load (N) 220 220 250 320 370 150 140 120 140 200 460 280 280 140 340	PCr / 1,180 Sphere Diameter (mm) 10 11 11 10 11 11 11 11 11 10 11 11	°C Compressive strength, MPa (N/mm ²) 1.96 1.62 1.84 2.85 2.73 1.10 1.03 0.88 1.03 1.78 3.39 2.06 2.50	
Sample No. 1 2 3 4 5 6 7 8 9 10 11 12 13 14 15	Fracture load (kN) 0.17 0.35 0.14 0.31 0.11 0.15 0.11 0.15 0.11 0.10 0.17 0.14 0.14 0.14 0.20 0.20 0.10	5% w-A Fracture load (N) 170 350 140 310 110 150 110 150 110 100 170 140 140 140 200 200 100	PCr / 1,180 Sphere Diameter (mm) 12 11 11 10 11 12 11 11 10 11 11 10 11 11 10 11 11	P°C Compressive strength, MPa (N/mm ²) 1.05 2.58 1.03 2.76 0.81 0.93 0.81 0.74 1.52 1.03 1.03 1.03 1.03 1.03 1.78 1.24 0.74 1.27	Fracture load (kN) 0.22 0.22 0.25 0.32 0.37 0.15 0.14 0.12 0.14 0.20 0.46 0.28 0.28 0.28 0.14 0.34	10% w-A Fracture load (N) 220 250 320 370 150 140 120 140 200 460 280 280 280 140 340	PCr / 1,180 Sphere Diameter (mm) 10 11 11 10 11 11 11 11 10 11 11	°C Compressive strength, MPa (N/mm ²) 1.96 1.62 1.84 2.85 2.73 1.10 1.03 0.88 1.03 1.78 3.39 2.06 2.50 1.03 2.50 1.89	
Sample No. 1 2 3 4 5 6 7 8 9 10 11 12 13 14 15	Fracture load (kN) 0.17 0.35 0.14 0.31 0.11 0.15 0.11 0.10 0.17 0.14 0.14 0.14 0.20 0.20 0.10	5% w-A Fracture load (N) 170 350 140 310 110 150 110 100 170 140 140 140 140 200 200 100	PCr / 1,180 Sphere Diameter (mm) 12 11 11 10 11 12 11 11 10 11 11 10 11 11 10 11 11	S.00 Compressive strength, MPa (N/mm²) 1.05 2.58 1.03 2.76 0.81 0.93 0.81 0.74 1.52 1.03 1.03 1.78 1.24 0.74 1.27 0.64	Fracture load (kN) 0.22 0.22 0.25 0.32 0.37 0.15 0.14 0.12 0.14 0.20 0.46 0.28 0.28 0.14 0.34	10% w-A Fracture load (N) 220 250 320 370 150 140 120 140 200 460 280 280 140 340	PCr / 1,180 Sphere Diameter (mm) 10 11 11 10 11 11 11 11 10 11 11	°C Compressive strength, MPa (N/mm ²) 1.96 1.62 1.84 2.85 2.73 1.10 1.03 0.88 1.03 1.78 3.39 2.06 2.50 1.03 2.50 1.89 0.78	
Sample No. 1 2 3 4 5 6 7 8 9 10 11 12 13 14 15	Fracture load (kN) 0.17 0.35 0.14 0.31 0.11 0.15 0.11 0.10 0.17 0.14 0.14 0.14 0.20 0.20 0.10	5% w-A Fracture load (N) 170 350 140 310 110 150 110 150 110 100 170 140 140 140 140 200 200 100	PCr / 1,180 Sphere Diameter (mm) 12 11 11 10 11 12 11 11 10 11 11 10 11 11 10 11 11	S.00 Compressive strength, MPa (N/mm²) 1.05 2.58 1.03 2.76 0.81 0.93 0.81 0.74 1.52 1.03 1.03 1.03 1.03 1.03 1.03 1.03 1.78 1.24 0.74 1.27 0.64 0.74	Fracture load (kN) 0.22 0.22 0.25 0.32 0.37 0.15 0.14 0.12 0.14 0.20 0.46 0.28 0.28 0.28 0.14 0.34	10% w-A Fracture load (N) 220 250 320 370 150 140 120 140 200 460 280 280 140 340	PCr / 1,180 Sphere Diameter (mm) 10 11 11 11 11 11 11 11 10 11 11	S.50 °C Compressive strength, MPa (N/mm ²) 1.96 1.62 1.84 2.85 2.73 1.10 1.03 0.88 1.03 1.78 3.39 2.06 2.50 1.89 0.78 0.88	

Appendix XIV – Calculation	of crushing strength	of hot-bonded LWA
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Part J

		15% w-A	APCr / 1,18	0°C	20% w-APCr / 1,180°C			
Sample No.	Fracture load (kN)	Fracture load (N)	Sphere Diameter (mm)	Compressive strength, MPa (N/mm ²)	Fracture load (kN)	Fracture load (N)	Sphere Diameter (mm)	Compressive strength, MPa (N/mm ²)
1	0.34	340	9	3.74	0.37	370	10	3.30
2	0.45	450	10	4.01	0.50	500	9	5.50
3	0.50	500	10	4.46	0.46	460	10	4.10
4	0.46	460	10	4.10	0.52	520	10	4.63
5	0.38	380	10	3.39	0.35	350	10	3.12
6	0.43	430	10	3.83	0.37	370	10	3.30
7	0.34	340	9	3.74	0.52	520	10	4.63
8	0.41	410	10	3.65	0.40	400	9	4.40
9	0.34	340	10	3.03	0.52	520	10	4.63
10	0.45	450	10	4.01	0.50	500	10	4.46
11	0.56	560	10	4.99	0.48	480	10	4.28
12	0.51	510	10	4.55	0.52	520	11	3.83
13	0.56	560	11	4.12	0.42	420	9	4.62
14	0.36	360	10	3.21	0.45	450	10	4.01
15	0.52	520	10	4.63	0.39	390	9	4.29
			Average	3.96			Average	4.21
			SD	0.54			SD	0.63
			Min	3.03			Min	3.12
			Max	4.99			Max	5.50

	25% w-APCr / 1,180°C					30% w-APCr / 1,180°C			
Sample No.	Fracture load (kN)	Fracture load (N)	Sphere Diameter (mm)	Compressive strength, MPa (N/mm ²)	Fracture load (kN)	Fracture load (N)	Sphere Diameter (mm)	Compressive strength, MPa (N/mm ²)	
1	0.52	520	10	4.63	0.15	150	10	1.34	
2	0.59	590	11	4.35	0.14	140	11	1.03	
3	0.44	440	10	3.92	0.17	170	11	1.25	
4	0.44	440	11	3.24	0.15	150	11	1.10	
5	0.48	480	11	3.54	0.14	140	11	1.03	
6	0.87	870	11	6.41	0.26	260	11	1.92	
7	0.28	280	10	2.50	0.15	150	10	1.34	
8	0.44	440	10	3.92	0.20	200	10	1.78	
9	0.42	420	10	3.74	0.20	200	10	1.78	
10	0.20	200	9	2.20	0.20	200	10	1.78	
11	0.40	400	10	3.57	0.14	140	10	1.25	
12	0.41	410	10	3.65	0.14	140	10	1.25	
13	0.44	440	10	3.92	0.15	150	11	1.10	
14	0.42	420	11	3.09	0.17	170	10	1.52	
15					0.15	150	11	1.10	
			Average	3.76			Average	1.37	
			SD	1.00			SD	0.31	
			Min	2.20			Min	1.03	
			Max	6.41			Max	1.92	

Appendix XV - Preliminary tests of cold bonded LWA produced from w-APCr or c-APCr or c-w-APCr

The development of hardness of cold-bonded LWA incorporating APCr-m, w-APCr, c-APCr or c-w-APCr (carbonated and washed), was qualitatively assessed in different formulations over a period of 3 to 4 weeks. To do that, the degree of resistance of the pellets to direct contact with water was tested by immersing individual pellets in water and applying a slight pressure with a glass bar. The degree of resistance exhibited by the pellets to this simple test is referred as "water resistance" in this study. The aim of these preliminary tests was to define a range of APCr incorporation for the LWA formulations that were subsequently manufactured and characterised.

Test 1. Cold-bonded LWA with APCr incorporation up to 50%

Table A shows the water resistance on different days (D₁, D₃, etc) for individual pellets of cold-bonded LWA containing 10% to 50% of APCr and a constant addition of 10% of lime. The observations indicate that when the addition of lime was kept constant and low, the LWA incorporating up to 40% of APCr-m or w-APCr only developed water resistance by the end of the second and the third weeks of monitoring, respectively. Whereas those LWA incorporating 50% of APCr-m or w-APCr had a better performance because they started developing water resistance from the third day of curing. All the formulations incorporating c-w-APCr developed water resistance from the second week (D₉) of curing, however, this characteristic disappeared by the end of the third week for the lower incorporations (10, 20 and 30%). It was concluded that the addition of 10% of lime was not enough to achieve a proper binding in LWA formulations containing between 10% and 50% of APCr-m, w-APCr or c-w-APCr.

A DC r	$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	Water resistance						
AICI	70 AI CI	70 Linic	D_1	D3	D ₆	D9	D14	D ₂₁
	10	10	Х	Х	Х	Х	0	0
	20	10	Х	Х	Х	Х	0	0
APCr-m	30	10	Х	Х	Х	Х	0	0
	40	10	Х	Х	Х	Х	0	0
	50	10	Х	0	0	0	0	0
	10	10	Х	Х	Х	Х	Х	Х
	20	10	Х	Х	Х	Х	Х	0
w-APCr	30	10	Х	Х	Х	Х	Х	0
	40	10	Х	Х	Х	Х	Х	0
	50	10	Х	0	0	0	\checkmark	0
	10	10	Х	Х	Х	0	0	Х
	20	10	Х	Х	Х	Ο	0	Х
c-w-APCr	30	10	Х	Х	Х	0	0	Х
	40	10	Х	Х	Х	0	0	0
	50	10	Х	Х	Х	0	0	0

Table A. Development of water resistance of individual pellets of cold-bonded LWA incorporation up to 50% of APCr-m, w-APCr or c-w-APCr and constant addition of lime.

x The pellets collapsed in water in just a few minutes (no more than 3 min).

O After 2 hours of immersion in water, the pellets did not collapse by themselves, but they were easily crushed by applying pressure with a glass bar.

 \checkmark After 2 hours of immersion in water, the pellets did not collapse and resisted the pressure applied with a glass bar.

Table B shows the water resistance on different days for individual pellets of cold-bonded LWA containing up to 50% of APCr and variable additions of lime. The pellets incorporating 10% to 40% of APCr-m or w-APCr developed water resistance during the first week of curing and exhibited water resistance by the end of the second week. By comparing these results with those reported in Table A, it can be said that the additional calcium oxide content in LWA incorporating 10% to 40% of w-APCr reduced by one week the time required for developing water resistance. On the contrary, the LWA containing 50% of APCr-m or w-APCr showed water resistance from the third day of

curing. These two formulations had been already assessed in the previous test (Table A) and showed the same behaviour. Therefore, the development of water resistance was not only influenced by the addition of lime but also was favoured by a higher incorporation of APCr-m or w-APCr.

Table B. Development of water resistance of individual pellets of cold-bonded LWA incorporation up to 50% of APCr-m, w-APCr or c-w-APCr and variable additions of lime.

APCr	% ADCr	% I ime	Water resistance						
AFCI	70 AFCI	/0 LIIIIC _	D3	D_6	D14	D ₂₁			
	10	50	Х	Х	0	0			
	20	40	Х	Х	Х	0			
APCr-m	30	30	Х	Х	Ο	0			
	40	20	Х	Х	Ο	0			
	50	10	Ο	Ο	0	Ο			
	10	50	Х	Х	0	0			
	20	40	Х	Х	Ο	0			
w-APCr	30	30	Х	Х	Ο	0			
	40	20	Х	Х	Ο	0			
	50	10	Ο	Ο	0	Ο			
	10	50	Х	Х	Х	0			
	20	40	Х	Х	Х	0			
c-w-APCr	30	30	Х	Х	Х	0			
	40	20	Х	Х	Х	0			
	50	10	Х	Х	Ο	Ο			

X The pellets collapsed in water in just a few minutes (no more than 3 min).

O After 2 hours of immersion in water, the pellets did not collapse by themselves, but they were easily crushed by applying pressure with a glass bar.

On the other hand, Table B shows that the LWA incorporating 10% to 40% of c-w-APCr and additions of lime greater than 10%, only developed water resistance by the end of the third week. The time required by the LWA containing c-w-APCr, either with constant or variable additions of lime, is always longer than the time required by the LWA incorporating APCr-m or w-APCr. This indicates that the combined carbonation/washing treatment should not be considered for APCr before incorporation into cold-bonded

LWA, because the washing removes part of the calcium content. Nevertheless, the treatment of carbonation can still be evaluated for this aim. Despite the LWA evaluated in Test 1 developed a certain water resistance over the time, these results were not satisfactory because most of the pellets were easily crushed by applying pressure with a glass bar. The results of this test suggested that greater additions of APCr-m and w-APCr must be evaluated.

Test 2. Cold-bonded LWA from high incorporation of APCr

Table C shows the development of water resistance in single pellets of LWA with high incorporations of APCr-m, w-APCr and c-APCr. All the evaluated formulations developed a certain water resistance since the beginning of the curing process. However, only the LWA incorporating 60% and 70% of c-APCr or w-APCr with lime additions from 20 to 30%, achieved a satisfactory binding that allowed them to remain in the water and to resist the pressure applied with a glass bar. This degree of binding was not achieved by the LWA incorporating APCr-m. That is, APCr without a pre-treatment was not suitable for recycling into cold-bonded LWA under the assessed conditions.

Table C. Development of water resistance of individual pellets of cold-bonded LWA incorporating 50%, 60% and 70% of APCr-m, w-APCr or c-APCr and variable additions of lime.

		0/ Lime				V	Water res	sistance			
APCI	70 APCI	76 LIIIIe	D_3	D_6	D9	D ₁₂	D15	D ₁₈	D ₂₁	D ₂₄	D ₂₈
	50	10	0	0	0	0	0	0	0	0	0
	50	20	0	0	0	0	0	Ο	0	0	Ο
	50	30	0	0	0	0	0	0	0	0	0
	60	10	0	0	0	0	0	0	0	0	0
APCr-m	60	20	0	0	0	0	0	Ο	0	0	Ο
	60	30	0	\checkmark	0	\checkmark	\checkmark	0	0	0	0
	70	10	0	0	0	0	0	0	0	0	0
	70	20	0	0	0	0	\checkmark	\checkmark	0	0	Ο
	70	25	0	0	0	0	0	0	0	0	0
	50	10	0	0	0	0	0	0	0	0	0
	50	20	\checkmark	0	0	0	0	\checkmark	0	0	Ο
	50	30	\checkmark	\checkmark	\checkmark	0	\checkmark	0	\checkmark	0	0
	60	10	0	0	0	0	0	Ο	0	0	0
w-APCr	60	20	0	\checkmark	0	0	0	\checkmark	\checkmark	\checkmark	\checkmark
	60	30	\checkmark	\checkmark	\checkmark	\checkmark	\checkmark	\checkmark	\checkmark	\checkmark	\checkmark
	70	10	✓	0	✓	0	0	\checkmark	0	0	0
	70	20	\checkmark	\checkmark	0	0	0	\checkmark	\checkmark	\checkmark	\checkmark
	70	25	\checkmark	\checkmark	\checkmark	\checkmark	\checkmark	\checkmark	\checkmark	\checkmark	\checkmark
	50	10	0	0	0	0	0	0	0	0	0
	50	20	0	0	0	0	0	Ο	0	0	Ο
	50	30	0	\checkmark	\checkmark	0	0	\checkmark	0	0	Ο
	60	10	0	0	0	0	0	0	0	0	0
c-APCr	60	20	\checkmark	\checkmark	\checkmark	0	\checkmark	\checkmark	\checkmark	\checkmark	\checkmark
	60	30	0	0	0	0	0	\checkmark	\checkmark	\checkmark	\checkmark
	70	10	0	0	0	0	0	0	0	0	0
	70	20	0	\checkmark	0	0	\checkmark	\checkmark	\checkmark	\checkmark	\checkmark
	70	25	✓	✓	\checkmark	0	\checkmark	\checkmark	\checkmark	\checkmark	\checkmark

O After 2 hours of immersion in water, the pellets did not dissolve by themselves, but they were easily crushed by applying pressure with a glass bar.

 \checkmark After 2 hours of immersion in water, the pellets did not dissolve and resisted the pressure applied with a glass bar.

Appendix XVI – Calculation of particle density and water absorption of cold-bonded LWA by the method of the pycnometer (BS EN 1097-6, 2022) Part A

LWA	W oven dried LWA (g)	W pycnometer+lid+water (g)	W pycnometer+lid+water+saturated LWA after 10 min (g)	W pycnometer+lid+water+saturated LWA after 60 min (g)	W pycnometer+lid+water+saturated LWA after 24 hr (g)	W saturated surface-dried LWA after 24 hr (g)
	M4	M ₃	$M_2(10 min)$	$M_2(60 min)$	$M_2(24 min)$	M_1
60w-APCr/20	500.73	1,596.99	1,879.29	1,882.10	1,889.86	605.06
60m ADCr/25	470.20	1,596.99	1,842.61	1,863.42	1,870.87	579.88
00w-Ar Ci/25	470.29	1,598.72	1,861.16	1,864.56	1,871.38	579.67
$60m \Lambda DC_{\pi}/20$	470.13	1,598.10	1,866.03	1,865.49	1,872.24	573.20
00w-AFCI/30	470.47	1,597.55	1,861.79	1,864.66	1,872.70	575.02
70w-APCr/20	450.53	1,598.72	1,845.91	1,849.14	1,857.17	553.95
70m ADC -/25	470.54	1,596.99	1,859.52	1,862.06	1,869.76	580.54
/0w-AFCI/23	470.43	1,598.72	1,860.53	1,863.27	1,871.05	580.29
$70_{\rm W}$ ADC $/20$	450.45	1,598.10	1,850.65	1,850.56	1,859.11	563.04
/0w-APCI/50	450.67	1,597.55	1,849.54	1,852.26	1,859.60	563.33
60c-APCr/20	500.57	1,598.10	1,852.92	1,857.49	1,863.77	553.75
60a ADC $r/25$	550.55	1,596.99	1,876.03	1,884.14	1,888.53	578.94
00C-AFCI/25	550.65	1,598.72	1,878.15	1,884.80	1,888.68	578.98
60a ADC $r/20$	545.18	1,598.10	1,877.30	1,882.63	1,885.56	569.37
00C-AFCI/30	545.33	1,597.55	1,878.41	1,883.20	1,886.37	568.76
70c-APCr/20	480.60	1,597.55	1,832.29	1,836.12	1,845.26	538.24
70_{\circ} ADC $u/25$	530.60	1,596.99	1,859.65	1,864.04	1,867.72	551.33
/0C-AFCI/23	530.50	1,598.72	1,861.29	1,865.23	1,868.74	550.80
70_{\circ} APCr/20	480.01	1,598.10	1,837.53	1,838.94	1,843.17	507.29
/00-AFCI/30	480.34	1,597.55	1,837.23	1,839.30	1,844.27	508.24
Carbon	500.23	1,596.99	1,873.35	1,896.10	1,898.24	622.15
Carbono®	500.13	1,598.72	1,873.47	1,895.89	1,898.02	622.34

Appendix XVI – Calculation of particle density and water absorption of cold-bonded LWA by the method of the pycnometer (BS EN 1097-6, 2022) Part B

	Ove	en-dried par density	ticle	Satur dried	Caturated and surface- dried particle density Apparent particle density Water absorption		boorption at 10 min Water absorption at 60 min			at 60	Water absorption at 24 hr							
LWA		(g/cm3)			(g/cm3)			(g/cm3)			%			%			%	
		Average	SD		Average	SD		Average	SD		Average	SD		Average	SD		Average	SD
60w-APCr/20	1.60	1.60	0.00	1.93	1.93	0.00	2.40	2.40	0.00	18.72	18.7	0.00	19.29	19.3	0.00	20.84	20.8	0.00
60w-APCr/25	1.53 1.53	1.53	0.00	1.89 1.88	1.88	0.00	2.39 2.37	2.38	0.01	17.32 21.08	19.2	2.66	21.74 21.81	21.8	0.05	23.33 23.26	23.3	0.05
60w-APCr/30	1.57 1.56	1.56	0.00	1.91 1.91	1.91	0.00	2.39 2.40	2.39	0.01	20.60 19.90	20.3	0.49	20.49 20.51	20.5	0.02	21.92 22.22	22.1	0.21
70w-APCr/20	1.52	1.52	0.00	1.87	1.87	0.00	2.34	2.34	0.00	20.46	20.5	0.00	21.17	21.2	0.00	22.96	23.0	0.00
70w-APCr/25	1.52 1.52	1.52	0.00	1.88 1.88	1.88	0.00	2.37 2.37	2.37	0.00	21.20 21.12	21.2	0.06	21.74 21.70	21.7	0.03	23.38 23.35	23.4	0.02
70w-APCr/30	1.49 1.49	1.49	0.00	1.86 1.86	1.86	0.00	2.37 2.38	2.37	0.01	23.12 22.77	22.9	0.25	23.10 23.37	23.2	0.19	25.00 25.00	25.0	0.00
60c-APCr/20	1.73	1.73	0.00	1.92	1.92	0.00	2.12	2.12	0.00	8.46	8.5	0.00	9.37	9.4	0.00	10.62	10.6	0.00
60c-APCr/25	1.91 1.90	1.90	0.01	2.01 2.00	2.00	0.01	2.12 2.10	2.11	0.01	2.89 3.23	3.1	0.24	4.36 4.44	4.4	0.06	5.16 5.14	5.2	0.01
60c-APCr/30	1.93 1.94	1.93	0.01	2.01 2.02	2.02	0.01	2.11 2.12	2.11	0.01	2.92 2.84	2.9	0.06	3.90 3.72	3.8	0.13	4.44 4.30	4.4	0.10
70c-APCr/20	1.65	1.65	0.00	1.85	1.85	0.00	2.06	2.06	0.00	9.29	9.3	0.00	10.09	10.1	0.00	11.99	12.0	0.00
70c-APCr/25	1.88 1.88	1.88	0.00	1.96 1.95	1.96	0.00	2.03 2.03	2.03	0.00	2.39 2.42	2.4	0.03	3.21 3.16	3.2	0.03	3.91 3.83	3.9	0.06
70c-APCr/30	1.82 1.83	1.83	0.00	1.93 1.94	1.93	0.01	2.04 2.05	2.04	0.01	4.51 4.34	4.4	0.12	4.80 4.77	4.8	0.02	5.68 5.81	5.7	0.09
Carbon8®	1.55 1.54	1.55	0.01	1.93 1.92	1.93	0.01	2.50 2.48	2.49	0.02	19.40 19.53	19.5	0.09	23.94 24.01	24.0	0.05	24.37 24.44	24.4	0.04

Appendix XVII – Calculation of loose bulk density and voids of cold-bonded LWA Part A

LWA	Mass of empty container M ₁	Volume of container V	Oven-dried particle density (Method of pycnometer ρ _p	W container + sample M ₂ (g)	Loose bulk	density ρ_b (g/cm ³)	Loose bulk density pb		Voids v %	
	(g)	(cm ³)	(g/cm^3)			Average	SD	(g/L)*		Average	SD
$60m$ ADC $\pi/20$	173.21	731	1.60	806.94	0.87	0.86	0.01	857.0	45.8	16.1	0.0
00w-AFCI/20	173.21	731	1.00	791.87	0.85	0.80	0.01	837.0	47.1	40.4	0.9
	228.17	1,061		1,116.68	0.84				45.3		
60w-APCr/25	228.17	1,061	1.53	1,124.50	0.84	0.84	0.00	841.9	44.8	45.0	0.3
	228.17	1,061		1,123.20	0.84				44.9		
	228.17	1,061		1,128.50	0.85				45.6		
60w-APCr/30	228.17	1,061	1.56	1,144.57	0.86	0.86	0.01	859.7	44.6	44.9	0.6
	228.17	1,061		1,147.77	0.87				44.4		
70 A DC=/20	173.21	731	1.50	771.37	0.82	0.92	0.00	0171	46.1	16.2	0.1
/0w-APCr/20	173.21	731	1.32	769.04	0.82	0.82	0.00	817.1	46.4	40.2	0.1
	228.17	1,061		1,111.53	0.83				45.2		
70w-APCr/25	228.17	1,061	1.52	1,113.46	0.83	0.83	0.00	832.7	45.1	45.2	0.1
	228.17	1,061		1,110.08	0.83				45.3		
70 A DC=/20	228.17	1,061	1.40	1,089.10	0.81	0.01	0.00	912.2	45.5	15 5	0.1
/0w-APCr/30	228.17	1,061	1.49	1,091.03	0.81	0.81	0.00	812.3	45.4	43.3	0.1

* Loose bulk density expressed in g/L is used for concrete formulation

Appendix XVII – Calculation of loose bulk density and voids of cold-bonded LWA Part B

LWA	Mass of empty container M1	Volume of container V	Oven-dried particle density (Method of pycnometer on	W container + sample M ₂ (g)	Loose bulk	density ρ_b (g/cm ³)	Loose bulk density pb		Voids v %	
	(g)	(cm ³)	(g/cm ³)			Average	SD	(g/L)*		Average	SD
$60a \ ADC \pi/20$	173.21	731	1 72	846.52	0.92	0.02	0.01	0174	46.7	47.0	0.2
00C-AFCI/20	173.21	731	1.75	840.58	0.91	0.92	0.01	917.4	47.2	47.0	0.5
$60a$ ADC $\pi/25$	228.17	1,061	1.00	1,271.54	0.98	0.00	0.01	087.2	48.2	18.0	0.2
00C-AFCI/25	228.17	1,061	1.90	1,279.54	0.99	0.99	0.01	987.2	47.8	40.0	0.5
$60a$ ADC $\pi/20$	228.17	1,061	1.02	1,283.39	0.99	0.00	0.00	004.8	48.5	10 5	0.0
00 C- APCI/30	228.17	1,061	1.95	1,283.88	1.00	0.99 0.0		994.8	48.4	48.3	0.0
70_{\circ} ADC $\pi/20$	173.21	731	1.65	797.78	0.85	0.96	0.01	8676	48.2	177	0.7
/0 C- APCI/20	173.21	731	1.03	809.11	0.87	0.80	0.01	802.0	47.3	4/./	0.7
70_{\circ} ADC $\pi/25$	228.17	1,061	1 00	1,264.74	0.98	0.07	0.00	074 5	48.0	10.2	0.2
/0C-APCI/23	228.17	1,061	1.00	1,259.55	0.97	0.97	0.00	974.3	48.3	40.2	0.2
70_{\circ} ADC $\pi/20$	228.17	1,061	1 02	1,202.04	0.92	0.02	0.00	016.5	49.8	40.0	0.1
/0 C- APCI/30	228.17	1,061	1.65	1,199.18	0.92	0.92	0.00	910.5	50.0	49.9	0.1
	228.17	1,061		897.43	0.63				59.3		
Carbon8®	228.17	1,061	1.55	904.85	0.64	0.64	0.01	639.8	58.9	58.7	0.7
	228.17	1,061		918.65	0.65				58.0		

* Loose bulk density expressed in g/L is used for concrete formulation

Appendix XVIII – Calculation of crushing strength of cold-bonded LWA Part A

60w-APCr /20					60w-APCr /25					
Sample No.	Fracture load (kN)	Fracture load (N)	Sphere Diameter (mm)	Compressive strength, MPa (N/mm ²)	Fracture load (kN)	Fracture load (N)	Sphere Diameter (mm)	Compressive strength, MPa (N/mm ²)		
1	0.07	70	13	0.37	0.12	120	13	0.63		
2	0.11	110	13	0.58	0.12	120	14	0.55		
3	0.08	80	12	0.50	0.07	70	12	0.43		
4	0.11	110	13	0.58	0.07	70	14	0.32		
5	0.10	100	13	0.53	0.05	50	13	0.26		
6	0.10	100	14	0.45	0.07	70	12	0.43		
7	0.07	70	13	0.37	0.07	70	13	0.37		
8	0.10	100	13	0.53	0.07	70	13	0.37		
9	0.07	70	13	0.37	0.07	70	13	0.37		
10	0.08	80	13	0.42	0.08	80	13	0.42		
11	0.11	110	13	0.58	0.07	70	13	0.37		
12	0.08	80	13	0.42	0.05	50	13	0.26		
13	0.11	110	14	0.50	0.05	50	13	0.26		
14	0.07	70	13	0.37	0.05	50	13	0.26		
15	0.07	70	13	0.37	0.07	70	13	0.37		
			Average	0.46			Average	0.38		
			SD	0.08			SD	0.11		
			Min	0.37			Min	0.26		
			Max	0.58			Max	0.63		
		60	A DC /20		l	70	A DC-/20			

		60w-	-APCr /30		70w-APCr/20					
Sample No.	Fracture load (kN)	Fracture load (N)	Sphere Diameter (mm)	Compressive strength, MPa (N/mm²)	Fracture load (kN)	Fracture load (N)	Sphere Diameter (mm)	Compressive strength, MPa (N/mm²)		
1	0.05	50	12	0.31	0.08	80	13	0.42		
2	0.07	70	12	0.43	0.07	70	13	0.37		
3	0.05	50	13	0.26	0.07	70	13	0.37		
4	0.1	100	12	0.62	0.07	70	13	0.37		
5	0.07	70	13	0.37	0.07	70	12	0.43		
6	0.07	70	12	0.43	0.07	70	13	0.37		
7	0.07	70	13	0.37	0.07	70	13	0.37		
8	0.07	70	12	0.43	0.07	70	13	0.37		
9	0.12	120	13	0.63	0.08	80	12	0.50		
10	0.05	50	13	0.26	0.07	70	13	0.37		
11	0.07	70	13	0.37	0.07	70	12	0.43		
12	0.07	70	12	0.43	0.10	100	13	0.53		
13	0.07	70	12	0.43	0.11	110	14	0.50		
14	0.14	140	12	0.87	0.07	70	13	0.37		
15	0.05	50	12	0.31	0.08	80	13	0.42		
			Average	0.44			Average	0.41		
			SD	0.16			SD	0.06		
			Min	0.26			Min	0.37		
			Max	0.87			Max	0.53		

Appendix XVIII – Calculation of crushing strength of cold-bonded LWA

Part B

		70w	-APCr/25		70w-APCr/30					
Sample No.	Fracture load (kN)	Fracture load (N)	Sphere Diameter (mm)	Compressive strength, MPa (N/mm²)	Fracture load (kN)	Fracture load (N)	Sphere Diameter (mm)	Compressive strength, MPa (N/mm²)		
1	0.07	70	14	0.32	0.10	100	14	0.45		
2	0.07	70	13	0.37	0.07	70	12	0.43		
3	0.07	70	12	0.43	0.07	70	12	0.43		
4	0.17	170	13	0.90	0.07	70	14	0.32		
5	0.05	50	12	0.31	0.07	70	13	0.37		
6	0.07	70	12	0.43	0.10	100	13	0.53		
7	0.07	70	13	0.37	0.10	100	13	0.53		
8	0.07	70	13	0.37	0.07	70	13	0.37		
9	0.05	50	12	0.31	0.07	70	13	0.37		
10	0.05	50	12	0.31	0.07	70	13	0.37		
11	0.07	70	13	0.37	0.07	70	13	0.37		
12	0.10	100	12	0.62	0.07	70	13	0.37		
13	0.05	50	12	0.31	0.10	100	13	0.53		
14	0.12	120	12	0.74	0.07	70	13	0.37		
15	0.07	70	12	0.43	0.07	70	13	0.37		
			Average	0.44			Average	0.41		
			SD	0.18			SD	0.07		
			Min	0.31			Min	0.32		
			Max	0.90			Max	0.53		
		(0)			1	<u></u>				

60c-APCr /20					60c-APCr /25					
Sample No.	Fracture load (kN)	Fracture load (N)	Sphere Diameter (mm)	Compressive strength, MPa (N/mm ²)	Fracture load (kN)	Fracture load (N)	Sphere Diameter (mm)	Compressive strength, MPa (N/mm ²)		
1	0.22	220	12	1.36	0.28	280	13	1.48		
2	0.17	170	12	1.05	0.34	340	14	1.55		
3	0.18	180	13	0.95	0.14	140	13	0.74		
4	0.20	200	13	1.05	0.21	210	13	1.11		
5	0.17	170	13	0.90	0.25	250	13	1.32		
6	0.12	120	13	0.63	0.20	200	14	0.91		
7	0.23	230	12	1.42	0.11	110	13	0.58		
8	0.20	200	13	1.05	0.25	250	12	1.55		
9	0.22	220	13	1.16	0.20	200	12	1.24		
10	0.20	200	13	1.05	0.26	260	14	1.18		
11	0.18	180	12	1.11	0.26	260	14	1.18		
12	0.12	120	12	0.74	0.25	250	12	1.55		
13	0.10	100	12	0.62	0.20	200	12	1.24		
14	0.20	200	12	1.24	0.28	280	14	1.27		
15	0.21	210	13	1.11	0.26	260	14	1.18		
			Average	1.03			Average	1.20		
			SD	0.24			SD	0.29		
			Min	0.62			Min	0.58		
			Max	1.42			Max	1.55		

Appendix XVIII – Calculation of crushing strength of cold-bonded LWA

Part C

		60c-	APCr /30		70c-APCr/20					
Sample No.	Fracture load (kN)	Fracture load (N)	Sphere Diameter (mm)	Compressive strength, MPa (N/mm²)	Fracture load (kN)	Fracture load (N)	Sphere Diameter (mm)	Compressive strength, MPa (N/mm²)		
1	0.26	260	14	1.18	0.14	140	14	0.64		
2	0.34	340	13	1.79	0.08	80	11	0.59		
3	0.22	220	13	1.16	0.15	150	13	0.79		
4	0.25	250	13	1.32	0.17	170	13	0.90		
5	0.31	310	13	1.63	0.05	50	12	0.31		
6	0.22	220	13	1.16	0.05	50	12	0.31		
7	0.21	210	14	0.95	0.07	70	13	0.37		
8	0.25	250	12	1.55	0.11	110	12	0.68		
9	0.21	210	14	0.95	0.17	170	13	0.90		
10	0.25	250	13	1.32	0.07	70	13	0.37		
11	0.50	500	13	2.64	0.17	170	13	0.90		
12	0.20	200	13	1.05	0.10	100	12	0.62		
13	0.26	260	13	1.37	0.17	170	13	0.90		
14	0.28	280	13	1.48	0.12	120	13	0.63		
15	0.25	250	12	1.55	0.07	70	12	0.43		
			Average	1.41			Average	0.62		
			ŠD	0.42			ŠĎ	0.22		
			Min	0.95			Min	0.31		
			Max	2.64			Max	0.90		

		70c	-APCr/25		70c-APCr/30						
Sample No.	Fracture load (kN)	Fracture load (N)	Sphere Diameter (mm)	Compressive strength, MPa (N/mm ²)	Fracture load (kN)	Fracture load (N)	Sphere Diameter (mm)	Compressive strength, MPa (N/mm ²)			
1	0.14	140	13	0.74	0.25	250	14	1.14			
2	0.20	200	13	1.05	0.21	210	14	0.95			
3	0.22	220	13	1.16	0.11	110	12	0.68			
4	0.21	210	13	1.11	0.22	220	13	1.16			
5	0.25	250	13	1.32	0.26	260	14	1.18			
6	0.20	200	12	1.24	0.20	200	13	1.05			
7	0.11	110	12	0.68	0.25	250	14	1.14			
8	0.10	100	13	0.53	0.20	200	13	1.05			
9	0.14	140	13	0.74	0.21	210	14	0.95			
10	0.21	210	13	1.11	0.23	230	13	1.21			
11	0.21	210	13	1.11	0.20	200	12	1.24			
12	0.20	200	13	1.05	0.20	200	13	1.05			
13	0.17	170	13	0.90	0.15	150	13	0.79			
14	0.21	210	14	0.95	0.10	100	12	0.62			
15	0.20	200	13	1.05	0.25	250	13	1.32			
			Average	0.98			Average	1.04			
			SD	0.22			SD	0.20			
			Min	0.53			Min	0.62			
			Max	1.32			Max	1.32			

Appendix XVII -	- Calculation	of crushing	strength	of cold-bo	nded LWA
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Part D

	Carbon8®										
Sample No.	Fracture load (kN)	Fracture load (N)	Sphere Diameter (mm)	Compressive strength, MPa (N/mm²)							
1	0.34	340	12	2.10							
2	0.37	370	13	1.95							
3	0.29	290	11	2.14							
4	0.17	170	11	1.25							
5	0.18	180	10	1.60							
6	0.52	520	14	2.36							
7	0.29	290	14	1.32							
8	0.22	220	13	1.16							
9	0.17	170	12	1.05							
10	0.25	250	14	1.14							
11	0.26	260	12	1.61							
12	0.28	280	12	1.73							
13	0.2	200	12	1.24							
14	0.23	230	12	1.42							
15	0.17	170	11	1.25							
			Average	1.56							
			SD	0.42							
			Min	1.05							
			Max	2.36							

		Maximum load		Compressive strength			Density									
	Formulation	Rep.	Max. Load (kN)	Average	SD	Max. Strength (Mpa)	Average	Std. Dev.	Wair (g)	Wwater (g)	Wair (kg)	Wwater (kg)	V (m ³)	D (kg/m ³)	Average	Std. Dev.
	Lytag®	1	428.604	385.861	60.448	42.860	38.586 6.	6.044	1,895.2	890.8	1.90	0.89	0.0010	1,883	1,897	10
		2	343.118			34.312		0.044	1,898.6	906.6	1.90	0.91	0.0010	1,910		19
	100%Clay 2	1	342.477	362.25	27.970	34.248	36.226	2.797	1,925.1	948.9	1.93	0.95	0.0010	1,968	1,961	10
ding		2	382.032			38.203			1,936.4	947.4	1.94	0.95	0.0010	1,954		10
Bone	25% w-APCr	1	462.815	482.96	28.495	46.282	48.297	2.849	2,080.9	1,093.9	2.08	1.09	0.0010	2,104	2,097	10
Hot		2	503.113			50.311			2,045.6	1,068.5	2.05	1.07	0.0010	2,089		10
	30% w-APCr	1	295.635	300.510	6.894	29.563	30.555	1.001	1,970.9	991.3	1.97	0.99	0.0010	2,008	2,003	
		2	305.384			30.538			1,985.3	994.4	1.99	0.99	0.0010	2,000		4
_		3	315-648			31.565			1,995.8	1,000.8	2.00	1.00	0.0010	2,002		
	Carbon8 [®]	1	238.910	238.91	0	23.891	23.891	0	1,709.4	721.0	1.71	0.72	0.0010	1,726	1,726	0
	60w-APCr/30	1	204.447	205.01	0.800	20.445	20.502	0.080	2,028.6	1,049.2	2.03	1.05	0.0010	2,067	2,062	7
guibi		2	205.579			20.558			1,880.1	968.3	1.88	0.97	0.0009	2,058		7
Cold bon	70w-APCr/30	1	170.009	172 74	3.865	17.001	17 275	75 0.387	2,011.9	1,004.0	2.01	1.00	0.0010	1,992	2,015	22
		2	175.475	1/2.74		17.548	17.275		1,536.0	784.0	1.54	0.78	0.0008	2,038		55
	60c-APCr/30	1	203.282	203.282	0	20.328	20.328	0	2,058.0	1,039.3	2.06	1.04	0.0010	2,016	2,016	0
	70c-APCr/30	2	192.057	192.057	0	19.206	19.206	0	1,996.7	992.8	2.00	0.99	0.0010	1,985	1,985	0

Appendix XIX – Calculation of compressive strength and density of LWAC $\,$