Influence of Geopolymerization Factors on Sustainable Production of Pelletized Fly Ash Based Aggregates Admixed with Bentonite, Lime and GGBS

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Highlights

- Three novel fly ash based pelletized aggregates were produced.
- Na_2O content was found to be the most influential parameter of the geopolymerization process.
- Polymerization of pelletized aggregates are analyzed through TGA and FTIR.
- Relationships between individual pellet strength and quantified amount of N-A-S-H/C-A-S-H were found to be linear.

Abstract

This experimental research investigates the influence of geopolymerization factors such as Na_2O dosages, water and mineral admixture (bentonite-BT, burnt lime-BL and ground granulated blast furnace slag-GGBS) on physio-mechanical properties of the pelletized fly ash (FA) based aggregates. Taguchi's L_9 orthogonal array was adopted to design the mixing ratios for three kinds of fly ash-based aggregates (in the combination of FA-BT, FA-BL and FA-GGBS). The advancement of the degree of geopolymerization of the produced aggregates were characterized using thermogravimetric analysis (TGA) and Fourier transform infrared spectroscopy (FTIR). Morphological characteristics of fly ash based aggregates were also studied using scanning electron microscope (SEM). Further, Grey relational analysis was carried out to identify the most influential response indices in the production of pelletized fly ash based aggregates. Obtained results for physio-mechanical characteristics of the aggregates indicated that with BL, aggregate impact value, aggregate crushing value and individual pellet strength of FABL aggregates found to be superior than GGBS and BT aggregates. However, substitution of GGBS has enhanced the pelletization efficiency and water absorption of FA-GGBS aggregates. TGA results indicated that quantified amount of hydration products i.e., N-A-S-H/C-A-S-H for fly ash based aggregates intensified with increase in Na_2O and mineral admixture dosages leading to denser and more compact microstructure. The results strongly suggest the existence of a linear relationship between the quantified amount of N-A-S-H/C-A-S-H and individual pellet strength of FA-based aggregate. Further experiments should be conducted to verify this trend for a much larger sample population and different curing regimes. FTIR spectrum showed the strong and broadened bands of Si-O terminal for all types of aggregates representing the conversion of unreacted minerals to chains of aluminosilicates gel (geopolymerized hydration product). Further, it can also be inferred from Grey relational analysis that among all other factors Na_2O content impacts significantly on the engineering properties of produced fly ash based aggregates.

Keywords: Fly ash, Geopolymerization, Pellets, Sustainability, Admixtures, Response Indices, Grey Relational Analysis, TGA, FTIR

1 Introduction

Cement production is responsible for 8-9% of all anthropogenic CO_2 emissions (Brinkman and Miller 2021). Hence, specific importance is being given to the inclusion of various industrial by-products and industrial wastes, such as fly ash, ground granulated blast furnace slag, and silica fume, as a substitute for cement and concrete production. Utilization of these by-products helps in reducing the excessive usage of ordinary portland cement (OPC) and the rising CO_2 emissions associated with its production (Mannan and Neglo 2010). One of the significant ingredients of concrete, i.e., aggregates, consists of about 70-80% of the volume of concrete (Oktay et al. 2015). It can be noted that aggregates are classified into various groups based on the type of raw materials that were used for production: first, a group of aggregates that are existent because of their natural origin from porous rocks; second, a group of aggregates that are existent due to natural origin from thermally processed rocks; and third group of aggregates that are produced artificially (categorically termed as artificial or synthetic aggregates) by using thermally processed industrial wastes or by-products like fly ash, bottom ash, rice husk ash, blast furnace slag (Bui et al. 2012a; Gesoğlu et al. 2012; Hwang and Tran 2015; Somayaji 1985). As per the report issued by Freedonia Group (Freedonia-Group 2016), the worldwide demand for aggregates reached 51.7 billion metric tonnes in the year 2019, on an average annual growth rate of 5.2 %. Another report stated that, in 2018, the global market for construction aggregates was valued at approximately \$ 360 billion, which is likely to increase to more than \$ 490 billion by 2025, at a multiple-yearly growth rate of 4.6% between 2019 and 2025 (Zion 2019). In India, it is reported that, in 2020, the demand for construction aggregates was reached up to 5 billion (MT) (Freedonia-Group, 2016). Consequently, with the increase in urbanization, finding sources for these large amounts of construction aggregates has led to the exhaustion of natural aggregates. In order to overcome this alarming context, the development of artificial aggregates using various wastes and industrial by-products (can be called synthetic aggregates) that can be used as an alternative to natural aggregates has drawn the attention of the global research community (Ayati et al. 2018; Baykal and Döven 2000; Colangelo et al. 2012; Franus et al. 2016; Narattha and Chaipanich 2018; Tang et al. 2017). Furthermore, it is reported that the production of artificial aggregates will solve a variety of environmental issues, including (i) the preservation of natural resources, (ii) use alternative cements such as calcined clay limestone (Scrivener et al. 2018) or Portland limestone cements (Gupta et al. 2020) (iii) performing mix design optimization (Park et al. 2008; Teichmann and

Schmidt 2004; Wille et al. 2011) using the Taguchi method (Dave et al. 2021; Joshaghani et al. 2015), or artificial intelligence techniques (Bhuva and Bhogayata 2022; Fan et al. 2021; Golafshani et al. 2021; Mahjoubi et al. 2023; Sadrossadat et al. 2022; Ziolkowski et al. 2021) or both combining these two (Tavares et al. 2022). However, as per 'standard specification for lightweight aggregates for structural concrete' ASTM C 330 (ASTM 2017), aggregates can be produced by adopting techniques like pelletizing and expanding with the utilisation of materials like fly ash, shale/slate, blast furnace slag, and clay.

In general, artificial aggregates were being produced by adopting a well-known metallurgical process called agglomeration, where finer particles are converted into fresh agglomerates (often called pellets) of varied shapes and sizes (Nor et al. 2016). Literature reports that the whole process of pelletization is influenced by several factors such as (i) fineness of raw materials, (ii) water content, (iii) type, nature, and dosage of binding agent, and (iv) duration of pelletization (Gomathi and Sivakumar 2014; Kockal and Ozturan 2011; Manikandan and Ramamurthy 2007; Priyadharshini et al. 2011; Ramamurthy and Harikrishnan 2006). It is understood from past literature that production of aggregates using class-C fly ash was stronger and more stable as compared to those produced using class-F fly ash (Bijen 1986a; Manikandan and Ramamurthy 2008). However, it is reported that utilisation of low calcium fly ash (i.e., ASTM class-F fly ash) is found to be more beneficial compared to that of high calcium fly ash (i.e., ASTM class-C fly ash) (ASTM 2018). This could be attributed to the existence of calcium-rich fly ash interferes with the process of polymerization, resulting in flash setting and also altering the microstructure (Zhao et al. 2019). It is also reported that the existence of silica (Si) and alumina (Al) rich precursors such as class-F fly ash undergoes a glass transition phase leading to the fusion of Si-O-Al chains (aluminosilicate gel), which aids in improving the hardened properties of aggregates (Bui et al. 2012a). Apart from class-C and class-F fly ash, there has been an attempt to utilise off-spec fly ash from landfills to produce lightweight aggregates. Lo et al. investigated the production of lightweight aggregates using high carbon fly ash, so-called off-spec fly ash, that can minimise the environmental pollution caused by its uneven disposal (Lo et al. 2016). However, it is reported that post-processing techniques like cold bonding, sintering, and autoclaving are necessary for the practical application of off-spec fly ash-based aggregates in concrete (Bijen 1986b). Various researchers state have that regardless of the positive aspects of class-F fly ash in the process of geopolymerization, fly ash added mixes showed a slow rate of strength gain (Sumer 2012), delayed setting time (Nath et al. 2015), possessing less specific gravity, lightweight as per EN 13055-1 (DIN 2015), and with lower individual pellet strength values (Chi et al. 2003). It is for these reasons that researchers started adding several admixtures such as ground granulated blast furnace slag (Bui et al. 2012b), lime (Reddy et al. 2016; Videla and Martinez 2002), clay binders (Geetha and Ramamurthy 2010; Ramamurthy and Harikrishnan 2006), bentonite (Gomathi and Sivakumar 2012; Manikandan and Ramamurthy 2009) and alternative binders like alkaline activators (Geetha and Ramamurthy 2013; Shivaprasad and Das 2018; Terzić et al. 2015) in the production of fly ash based aggregates (especially with class-F fly ash) for enhancing the engineering properties of the produced aggregates (Wasserman and Bentur 1997; Yang et al. 2011). With the inclusion of clay binders in the production of fly ash based geopolymer aggregates, it was observed that the aggregate crushing value of the produced aggregates increased was found to be limiting upto 30 % (Geetha and Ramamurthy 2011). Another study stated the utilization of ground granulated blast furnace slag in the production of aggregates exhibited superior strength, higher particle density, and crushing strength and concluded its beneficial usage compared to those produced with cement and fly ash (Bui et al. 2012b). The researcher stated that this could be attributed to the material reactivity of raw materials, which is exhibited by the amount of reactive SiO_2. Geopolymer-based fly ash aggregates produced with the addition of alkali activators (combination of sodium silicate and sodium hydroxide) were found to have improved the characteristic properties (Bui et al. 2012a; Shivaprasad and Das 2018).

It is observed from the literature that along with the chemical composition of mineral admixtures, alkali activators play an influential role in initiating the hydrolysis on the surfaces of raw materials (Al-Si materials) in the geopolymerization process (Rangan et al. 2005). Some of the influential factors are a type of alkali activator (Davidovits 1989; Hardjito et al. 2004; Khale and Chaudhary 2007; Rangan et al. 2005), alkali solution concentration (Görhan and Kürklü 2014; Hardjito et al. 2008; Khale and Chaudhary 2007; Komljenović et al. 2010; Mustafa Al Bakri et al. 2012; Patankar et al. 2014; Rattanasak and Chindaprasirt 2009) and ratio of binder to alkali (Abdul Rahim et al. 2014; Fernández-Jiménez and Palomo 2005; Rahmiati et al. 2015). (Chindaprasirt et al. 2012) stated that an increase in the molar ratio in the geopolymer mix increased its compressive strength. Hence, in order to understand the complexity of the relative influence of geopolymerization factors (mineral as well as chemical) on the properties of produced aggregates, the concept of experimental design was adopted by several researchers in the past (Cavazzuti 2013; Krishnan and

Purushothaman 2017; Montgomery 2017; Soudki et al. 2001). It is reported that by using Taguchi's experimental design methodology, a suitable understanding can be developed between the evaluated responses and considered factors through the response indices (Shivaprasad and Das 2018). Further, for obtaining a clear understanding of the response indices altogether, the researchers adopted grey relational analysis as it benefitted by converting a multi-objective problem into a single objective function (Sahoo et al. 2017; Shivaprasad and Das 2018).

As understood from the available literature stated in previous sections, the inclusion of additives like bentonite (BT), burnt lime (BL), and ground granulated blast furnace slag (GGBS) combined with a combination of alkali activators were found to be limited and focused on replacing fly ash partially either with these additives or other kinds of waste. This study is aimed at producing three kinds of novel pelletized fly ash based coarse aggregates with utilisation of fly ash (that is hundred percent) with the incorporation of BT, BL, and GGBS as binding media and a sustainable step can be taken in maximising the recycling of wastes by fulfilling the underlying need for coarse aggregates. The relative influence of geopolymerization factors and admixture additions was investigated by adopting Taguchi's orthogonal array experimental design. The characteristic properties of produced fly ash-based geopolymer aggregates were measured by carrying out a series of tests applicable to natural aggregates. Furthermore, scanning electron microscopic (SEM) analysis was conducted to understand the morphological characteristics of three different kinds of produced aggregates. Micro-scaled analysis was also conducted using thermogravimetric analysis (TGA) and Fourier transform infrared spectroscopy (FTIR).

1.1 Research significance

Sustainable usage of industrial by-products in the production of blended cement has formulated the idea of the sustainable production and development of construction materials. As per the current scenario, the consumption of artificially recreated materials using industrial by-products in cement and concrete has drawn considerable attention. The utilisation of natural aggregates in concrete production is increasing day by day, making it one of the scarce resources in the near future. Hence, there is a need for alternative building materials for construction industries which can be a substitution for natural aggregates. Producing such alternative materials by utilising the maximum number of industrial by-products such as fly ash with additive admixtures (mineral and chemical) is the need of the hour that can be achieved through a retrospective approach. Since curing regime plays a vital role it is also essential to make the production process less energy intensive and in this scenario production of pelletized fly ash based aggregates through ambient curing pays a vital contribution.

2. Materials and Methodology

2.1. Materials

The materials which are used in this present investigation comprise fly ash (FA), bentonite (BT), burnt lime (BL), and ground granulated blast furnace slag (GGBS). In this study, class-F fly confirmed to IS 3812 (part 1) – 2003 (IS: 3812, 2003) was used. The physical and chemical characteristics of FA, BT, BL and GGBS were analyzed and the pertaining results are presented in Table 1.

2.2. Alkaline activators

Laboratory grade NaOH pellets with 97% purity and Na_2SiO_3 solution (8.0% Na_2O; 26.5% SiO_2; 65.5% H_2O by mass) serve as alkaline activators in this experimental investigation. The preparation of alkaline solutions consists of mainly two stages. First, NaOH pellets were dissolved in distilled water to produce a NaOH solution which is followed by the mixing of Na_2SiO_3 with the NaOH solution in required proportions with respect to different mixes. The prepared alkaline solution was stored in an airtight container and was left to cool for 24 hours prior to its usage (Shivaprasad and Das 2018).

3. Experimental Program

3.1. Mix proportions design

Fly ash-based aggregates were produced using three different mineral admixtures such as bentonite (BT), burnt lime (BL), and ground granulated blast furnace slag (GGBS). Here, for designing the mix proportions for the production of fly ash based aggregates, a series of steps were followed that are listed below;

1) selection of experimental parameters and their respective levels of variation in the production of fly ash-based aggregates.
2) selection of suitable orthogonal arrays generated by Taguchi's experimental design methodology followed by appropriate framing of selected experimental parameters and their variation levels.

3) performing the production of fly ash-based aggregates admixed with mineral admixtures that are BT, BL, and GGBS as per the mixes designed using Taguchi's experimental design methodology.
4) evaluating the engineering properties of the produced aggregates by carrying out a series of tests on the produced aggregates (FA-BT, FA-BL, and FA-GGBS).
5) calculation of response indices individually for the obtained results of the produced aggregates with the help of statistical software.
6) Adopting grey relational analysis method for analysing the obtained results all together and identifying the most influential experimental parameter among the selected ones.

As understood from the available literature, two factors that are connected to the strength and efficiency of geopolymerization are Na_2O and water content (Harikrishnan and Ramamurthy 2006; Ramamurthy and Harikrishnan 2006; Shivaprasad and Das 2018). Along with two factors, an additional factor that was considered in this experimental investigation was dosages of three different kinds of admixtures, i.e., BT, BL, and GGBS. Hence, dosages of Na_2O, water and mineral admixtures collectively serve as experimental parameters for designing the production methodology of fly ash-based aggregates. In this study, for every experimental parameter, three levels of variation were selected. According to full factorial design methodology, a total of 27 (that is 3^3) experimental combinations are needed for evaluating the influence of every individual parameter, which could potentially become tedious and uneconomical. For this reason, in this experimental investigation, the Taguchi's experimental design methodology was adopted for evaluating the influence of different levels of selected experimental parameters on the properties of produced aggregates (FA-BT, FA-BL, and FA-GGBS) with the help of a smaller number of experiments. Orthogonal array, i.e., L9 (3^3) developed by Taguchi (Montgomery 2017), was used in this experimental study in order to represent a full factorial experiment. Preliminary trial studies were carried out to determine the combination of suitable ranges for Na_2O dosages (3, 4, 5 and 6 %), water content dosages (19, 20, 21 and 22%) and additive admixtures (5, 10, 15 and 20% by weight of fly ash) for producing FA-BT, FA-BL, and FA-GGBS aggregates. A solution with a high Na_2O content was found to have a high strength in the resulting geopolymers (Fernández-Jiménez and Palomo 2005). Moreover, as the concentration of Na_2O in the alkaline solution increases, it becomes too cohesive and difficult for its usage in the production of fly ash-based aggregates. Also, a high dosage of water and additive admixtures resulted in the formation of large, unevenly shaped agglomerates. Based on these preliminary observations, the different levels of experimental parameters used in Taguchi's experimental design methodology is presented in Table 2, where the dosage of Na_2O content in the production of FA-BT and FA-GGBS aggregates was within the range of 3-5% of the combined mass of precursor material and 4-6% of the combined mass of the precursor material for the producing FA-BL aggregates. The water content was varied in the range of 19-21% and the same was maintained for production of FA-BT, FA-BL, and FA-GGBS aggregates. The set of mixes in the production of FA–BT, FA–BL and FA– GGBS as per L_9 orthogonal arrays available in Taguchi's experimental design methodology is presented in Table 3. It is to be noted that the ratio of alkaline solution to fly ash was maintained at 0.3 with respect to water and Na_2O content in the alkaline solution for producing FA-BT, FA-BL and FA-GGBS aggregates (Shivaprasad and Das 2018).

Subsequently, grey relational analysis was adopted for analysing the results that will be obtained by carrying out a series of tests as applicable to conventional aggregates, like aggregate impact value, aggregate crushing value, water absorption, and individual pellet strength (described in detail in the subsequent section) on FA-BT, FA-BL, and FA-GGBS aggregates (as per Table 3). This method will help in obtaining an unbiased analysis for determining the order of influence of the selected experimental parameters in the produced aggregates (Sahoo et al. 2017).

Firstly, the results of various tests as mentioned above for FA-BT, FA-BL, and FA-GGBS aggregates are transformed into a normalised value using the following equations.

For smaller-is-better, the formula to transform x_i (j) to x_i^* (j) is,

$$x_i^*(j) = \frac{\max\limits_j x_i(j) - x_i(j)}{\max\limits_j x_i(j) - \min\limits_j x_i(j)} \quad (1)$$

For larger-is-better transformation, x_i (j) can be transformed to x_i^* (j), the formula is,

$$x_i^*(j) = \frac{x_i(j) - \min\limits_j x_i(j)}{\max\limits_j x_i(j) - \min\limits_j x_i(j)} \quad (2)$$

It is to be noted that properties like aggregate impact, crushing value and water absorption are principally, the lower the obtained value is, the better as per IS 2386 (part 4)-1963 (Bureau of Indian Standards (BIS) 1963). However, for individual crushing strength of pellets, the higher is the better.

Second, the grey relational coefficient, ξ_i (k) from the normalised values is calculated by using the following formula.

$$\xi i(k) = \frac{\Delta_{min} + \xi\Delta_{max}}{\Delta_{0i}(k) + \xi\Delta_{max}} \quad (3)$$

where, Δ_{0i} is the is the deviation sequence of the reference sequence and the comparability sequence and $\Delta_{0i} = \| x_0 (k) - x_i (k) \|$

where, where x_0 (k) implies the reference sequence and xi (k) termed as comparability sequence. Δ_{min} and Δ_{max} are the minimum and maximum values of the absolute differences (Δ_{0i}) of all comparing sequences. ξ is a distinguishing coefficient ($0 \leq \xi \leq 1$) and in the present study, $\xi = 0.5$ is taken (Sahoo et al. 2017).

Finally, the grey relational grade (GRG) is calculated by summing up the weighted grey relational coefficients corresponding to the responses.

$$\gamma_i = \frac{1}{n}\sum_{k=1}^{n} \xi i(k) \quad (4)$$

where, *n* = number of process responses considered in this study.

It is to be noted that a higher grey relational grade indicates a stronger relational degree between the ideal sequence and the given sequence (Sahoo et al. 2017). The sequential steps followed in Taguchi's experimental design methodology and grey relational analysis are represented in the form of the flowchart depicted in Fig. 1.

3.2.Production of FA-BT, FA-BL and FA-GGBS aggregates

The production of pelletized FA-BT, FA-BL, and FA-GGBS aggregates was carried out using a laboratory scale fabricated type disc pelletizer (diameter: 450 mm and internal depth: 100 mm) was used to produce the aggregates in conjunction with the agglomeration technique. In this study, the angle of inclination and duration of pelletization are fixed at 45° and 15 minutes, respectively (Shivaprasad and Das 2018, 2021). The sequential steps which were followed for the pelletization process include (Sharath and Das 2021) The sequential steps which were followed for the pelletization process include (Sharath and Das 2021) (i) thorough mixing of all lump-free material to be pelletized, that is FA with three admixtures, BT, BL, and GGBS (as per the experimental set of mixes presented in Table 3 for producing FA-BT, FA-BL, and FA-GGBS aggregates, respectively). (ii) placing the mixtures in the disc pelletizer; (iii) spraying the prepared alkali solution over the mixtures within 3 minutes of the pelletization process. The produced FA-BT, FA-BL, and FA-GGBS aggregates were cured at ambient temperature conditions (28 ± 2 °C) for three curing ages i.e., 14, 28 and 100 days.

3.3. Testing and analysis of FA-BT, FA-BL and FA-GGBS aggregates

3.3.1. Efficiency of pelletization and particle size distribution

Pelletization efficiency is calculated as the percentage weight of the produced aggregates of sizes greater than 4.75 mm (retention of aggregates on IS sieve no 480) against the total weight of produced aggregates as shown below in the form of an equation (Eq.5).

$$Pelletization\ efficiency\ (\%) = \frac{Weight\ of\ aggregates\ retained\ on\ IS\ sieve\ no\ 480}{Total\ weight\ of\ produced\ aggregates} \qquad (5)$$

The particle size distribution of FA-BT, FA-BL, and FA-GGBS aggregates was determined by using a standard set of sieves as per the procedure given in Bureau of Indian Standards - BIS 383:2016 (IS:383-2016).

3.3.2. Specific gravity and water absorption of aggregates

Specific gravity and water absorption tests for all produced aggregates were carried out as per the procedure given in Indian Standards - IS 383:2016 (IS:383-2016).

3.3.3. Aggregate impact and crushing value

The aggregate impact value is the determination of measure of the resistance to any application of sudden impact or shock and it gives a relative measurement of the resistance of aggregates subjected to crushing by the gradual application of a compressive load. The test samples from produced aggregates (FA-BT, FA-BL, and FA-GGBS) comprising sizes within the range 10-12.5 mm and 10-6.3 mm were used for conducting aggregate impact and crushing tests, respectively. Both the tests were carried out as per the procedure given in Indian Standards-IS 383:2016 (IS:383-2016) and 2386:1963-part 4 (IS 2386:1963-part 4).

3.3.4. Individual crushing strength of pellets

Individual crushing strength of pellets is measure of determining the fracture toughness of the aggregate test samples that fractures under a constantly applied stress on them (Abdullah et al. 2020; Gomathi and Sivakumar 2015; Shahane and Patel 2022; Shivaprasad and Das 2018, 2021). In this experimental study, the strength of the produced FA-BT, FA-BL, and FA-GGBS aggregates was determined by measuring the crushing strength of each individual aggregate in such a way that the aggregate (which needs to be tested) is loaded diametrically between two parallel plates. In order to fit this kind of arrangement, the California bearing ratio (CBR) apparatus was used. By analysing the stress imposed, it can be inferred that when a spherical pellet undergoes compression between two diametrically opposed points, the crushing strength (σ) (Gomathi and Sivakumar 2012) is calculated by the equation (Eq. 6).

$$\sigma = \frac{2.8 \times P}{\pi \times x^2} \tag{6}$$

where, x is the distance between the two loading points or the size of the aggregate samples selected for the test and P is fracture load for the sample. To determine the average crushing strength of individual pellets, a minimum of 20 pellets with diameters ranging from 6 to 18 mm were tested from FA-BT, FA-BL, and FA-GGBS aggregates (Shivaprasad and Das 2018). The selection of 6-18 mm sized pellets for carrying out the individual crushing strength of pellets was adopted in order to obtain a consistent strength data for the produced aggregates.

3.3.5. Scanning electron microscopy studies

The spherical shaped pellets which got broken into two halves obtained from the individual pellet strength test were gold sputtered for morphological characterization with the help of a scanning electron microscope. Morphological images were taken through a scanning electron microscope (JEOL, JSM-638OLA) in secondary electron mode and the same is discussed in the results and discussion section.

3.3.6. Thermogravimetric analysis (TGA) studies

Thermogravimetric analysis (TGA) helps in determining the various polymerization phases formed as a function of temperature, and at the same time, derivative of thermogravimetric (DTG) curve also helps in signifying those characteristic temperature boundaries of the phases formed. It is for this purpose that the thermogravimetric analysis was performed for the produced FA-BT, FA-BL, and FA-GGBS aggregates.

Thermogravimetric analysis was carried out using a TG/DTA analyzer from Rigaku TG-DTA 8122. Pelletized aggregates at the age of 100 days cured were crushed and samples passing through a 75 µm IS sieve are subjected to heating within the temperature boundaries of 25-850 ˚C, at a heating rate of 10 ˚C/min in a nitrogen purge environment (purge rate of 20 ml/min).

3.3.7. Fourier transform infrared spectroscopy (FTIR) analysis

Fourier transform infrared spectroscopy (FTIR) analysis helps in the identification of organic, inorganic, and polymeric materials with the utilisation of infrared light by scanning any given samples. Also, by using the FTIR technique, the occurrence of various functional groups presents in a given sample can be recognized. It is for this purpose that the FTIR analysis was performed for the produced FA-BT, FA-BL, and FA-GGBS aggregates.

A Bruker (Alpha II) instrument was used for the FTIR analysis, with a wavenumber range of 4000 to 500 cm^{-1} and a resolution of 2 cm^{-1}. FTIR spectra were obtained for powdered geopolymerized pellet samples (passing through a 75 µm sieve). On the basis of the obtained FTIR spectrum variation, functional groups were identified and analysed for different aggregate samples.

4. Results and Discussion

Individual characteristics such as particle size distribution, specific gravity, water absorption, aggregate impact and crushing value, individual crushing strength of pellets, and production efficiency were used as a basis for evaluating the performance of fly ash-based aggregates. The relevant results are reported in the subsequent sections, and suitable observations with discussions are made.

4.1. Specific gravity

The average specific gravity measured for FA-BT, FA-BL, and FA-GGBS aggregates was found to be within the range of 2.0-2.2. The prescribed limits for specific gravity of produced aggregates as per Indian standard IS 383:2016 (IS:383-2016), table 7 are 2.0-3.2. The obtained values for specific gravity of the produced FA-BT, FA-BL and FA-GGBS aggregates hence comply with the prescribed limits as per the Indian standards.

4.2.Influence of factors on properties of produced aggregates

In order to understand the influence of the governing factors (dosage levels of Na_2O, water and admixture addition contents) on the properties of produced aggregates, it is very much essential to calculate the response index values. In order to do so, Minitab software (version 20.2) was used and the procedure employed is described in the following section.

4.2.1. Calculation of response indices

Test results of aggregate impact and crushing value, individual crushing strength of pellets, and water absorption of FA-BT, FA-BL, and FA-GGBS aggregates are presented in Figures 2, 3, and 4 respectively. Assessment of the response index for each governing factor was carried out by calculating the mean at curing ages of 14, 28, and 100 days for the produced FA-BT, FA-BL, and FA-GGBS aggregates. For instance, factor I'1 was tested for FA-BT, FA-BL and FA-GGBS aggregates, i.e., FABT 1 to FABT 3, FABL 1 to FABL 3 and FAGGBS 1 to FAGGBS 3. Hence, the response index for factor I'1 for at 14 days of curing will be the mean of obtained values for trials FABT 1 to FABT 3, FABL 1 to FABL 3 and FAGGBS 1 to FAGGBS 3. Likewise, the response index for all other governing factors is calculated for other curing periods. Since lower aggregate impact, crushing and water absorption values are the requirements for FA-BT, FA-BL, and FA-GGBS aggregates as per IS 383-2016 (IS:383-2016), the 'smaller-the-better' criteria was chosen, whereas, for individual crushing strength of pellets, higher values are essential, hence 'higher-the-better' criteria was selected for this property.

4.2.2 Aggregate impact and crushing value of the produced aggregates

It can be noticed from the presented Fig. 2(a-b) that the engineering properties of mix FABT 9 (marked in red) were found to be the best performing as compared to those of all the other FA-BT aggregates. This can be attributed to the highest dosage of Na_2O content, i.e., 5% present in the mix proportion and bentonite (BT) content of 10%. However, FABT 3 (marked in blue) fared the poorest of all. It is important to note that FABT 3 consists of the lowest dosage of Na_2O content, i.e., 3%, and the highest dosage of bentonite content (BT), i.e., 15%.

From the presented Fig. 3(a-b), it can be noticed that the engineering properties of mix FABL 5 (marked in red) are found to be the best performing in comparison with all other FA-BL aggregates. This can be associated to highest dosage of Na_2O content, i.e., 5% present in the mix proportion and water content of 20%. However, FABL 1 (marked in blue) performed poorest among all, which consists of the lowest dosages of Na_2O and water content, i.e., 4% and 19%, respectively.

By observing the presented Fig. 4(a-b) for engineering properties of FA-GGBS aggregates, the mix FAGGBS 5 (marked in red) has performed best in comparison with all other FAGGBS aggregates, which is attributed to the highest dosages of Na_2O and GGBS content, i.e., 5% and 15%, respectively. The mix FAGGBS 1 (marked in blue) has performed the lowest of all. This mix is associated with the lowest dosages of Na_2O and GGBS content, i.e., 3% and 5%, respectively.

For aggregate impact and crushing value, the response indices of all governing factors associated with FA-BT, FA-BL, and FA-GGBS aggregates were determined and plotted in Figs. 5(a-b), 6(a-b), and 7(a-b), respectively. It can be noticed from the figures that among the three governing factors, water content dosage is found have the least effect on aggregate impact and crushing value of FA-BT and FA-GGBS aggregates. However, for FA-BL aggregates, it is found that dosage of BL has the least effect on aggregate impact and crushing value among the three governing factors for FA-BL aggregates. From the figures, it can be understood that the dosage of Na_2O is found to be directly proportional to the aggregate impact and crushing value of FA-BT, FA-BL, and FA-GGBS aggregates.

It can also be observed that 10 % of BT and 15% of GGBS were found to be the optimum for both aggregate impact and crushing value of FA-BT and FA-GGBS aggregates, whereas, it is 20% of water content which is found to be optimum for FA-BL aggregates.

As per the prescribed limits specified by Indian standards IS 383-2016 (IS:383-2016) and 2386:1963-part 4 (IS 2386:1963-part 4), the aggregate impact and crushing values for produced aggregates should not exceed 45 and 30 %, respectively, provided they are used for concreting purposes. The aggregate impact and crushing values obtained for produced FA-BT, FA-BL, and FA-GGBS aggregates were hence found to be adhering to the prescribed limits as per the Indian standards.

4.2.3. Individual crushing strength of pellets (IPS)

a) FA-BT aggregates

As per the data presented in Fig. 2(c), the individual crushing strength of pellets measured for aggregate mix FABT 9 (marked in red) was found to be 7.2 MPa at 100 days of curing age. It is to be noted that this mix consists of the highest dosage contents of Na_2O and water, i.e., 5% and 21%, respectively, with a BT content of 10%. Whereas, aggregate mix FABT 1 (marked in blue) was found to be have the lowest crushing strength of pellets as 0.9 MPa at the curing age of 100 days. This is attributed to the lowest dosage content of Na_2O, water and BT content in the aggregate mix, i.e., 3%, 19% and 5%, respectively.

Previous research has reported that fly ash based pelletized aggregates produced by admixing clay minerals as an admixture has an individual crushing strength of 1.0-8.2 MPa (Gomathi and Sivakumar 2014).

The plotted relationship between the calculated response indices and governing factors in this type of aggregate (Fig. 8(a)) suggests that the individual crushing strength of FA-BT aggregates is directly proportional to Na_2O and water content dosages. In addition to this, dosage of BT content up to a certain percentage (i.e., 10%) showed an increment in individual crushing strength only. Further, with the increase in curing age, a relative superiority in crushing strength for FA-BT aggregates can be witnessed.

b) FA-BL aggregates

According to the measured values of individual crushing strength of FA-BL aggregates (shown in Fig. 3(c)), the aggregate mix FABL 5 (marked in red) had the highest crushing strength, i.e., 11.6 MPa after 100 days of curing. Whereas the lowest crushing strength value was obtained for FABL 1 (marked in blue) aggregate mix, i.e., 1.8 MPa at the curing age of 100 days. The variations in the dosage contents of Na_2O, water, and BL cause the resulting difference in individual crushing strength between these two mixes. Increased crushing strength for aggregate mix FABL 5 corresponds to dosage contents of Na_2O (5%), water (20%) with highest BL (15%). While, for FABL 1, the aggregate mix, which possesses lower crushing strength, consists of a combination of lower dosage contents of Na_2O (4%), water (19%), and BL (5%). However, it is understood from past research that fly ash-based aggregates produced using lime as an additive admixture have reported an individual pellet strength of 2.6 MPa (Gomathi and Sivakumar 2012).

Fig. 8(b) shows that the individual crushing strength values increase up to a certain dosage level of Na_2O (5%) and water content (20%), after which the individual crushing strength decreases. But, in the case of the governing factor BL content, an increase in individual crushing strength values was found to be proportional to BL content.

c) FA-GGBS aggregates

According to the results obtained on individual crushing strength of pellets of FA-GGBS aggregates (in Fig. 4(c)), the mix FAGGBS 7 (marked in red) demonstrated the highest individual crushing strength of 6.2 MPa at 100 days of curing age.

This increase in crushing strength of the aforementioned mix corresponds to high dosage contents of Na_2O (5%), with water (19%), and GGBS (15%). Whereas, the aggregate mix FAGGBS 1 (marked in blue) exhibited the lowest individual crushing strength of 0.8 MPa at 100 days of curing age, which corresponds to low dosage contents of Na_2O (3%), water (19%), and GGBS (5%).

The relationship between calculated response indices and governing factors in FA-GGBS aggregates is depicted in Fig. 8(c). From the figure, following interpretations can be drawn, a) with the increase in dosage contents of Na_2O, the individual crushing strength increased, b) marginal changes in individual crushing strength values were observed for different dosage contents of water; and d) as the GGBS dosage content increased (up to 10%), individual crushing strength decreased, which again increased with an increment in GGBS dosage contents.

4.3. Water absorption of produced aggregates

According to the results obtained on water absorption for produced FA-BT, FA-BL, and FA-GGBS aggregates (Figs. 2(d), 3(d), and 4(d), respectively), FABT 9, FABL 5, and FAGGBS 7 (all marked in red) had the lowest water absorption values, i.e., 15.3%, 14.1%, and 11.3%, respectively, at the curing age of 100 days. According to the plotted relationship between calculated response indices and governing factors for produced FA-BT, FA-BL, and FA-GGBS aggregates (Fig. 9 a-c), the governing factor Na_2O content had the greatest influence on water absorption of FA-BT, FA-BL, and FA-GGBS aggregates. It is reported that the artificially produced aggregates of structural grade were found to absorb 5-25 % by weight of dry aggregates (Holm and Ries 2006).

4.4. Size distributions of FA-BT, FA-BL and FA-GGBS aggregates

Particle size distribution results for FA-BT, FA-BL, and FA-GGBS aggregates as per the trial mixes specified in Table 3 are presented in Fig. 10 (a), (b), and (c), respectively. In order to check the suitability of the produced aggregates, the lower limit and higher limit for coarse aggregates as stated in IS 383-2016 specifications is plotted in these figures. However, the other mixes in FA-BT, FA-BL and FA-GGBS aggregates are not satisfying fully the required limits specified by IS 383-2016. This can be attributed to the inadequate dosage levels of the Na_2O, water, and mineral admixtures in their production.

Hence, it can be understood that selected governing factors in the production of FA-BT, FA-BL, and FA-GGBS aggregates have influenced the gradation of aggregates produced.

4.5. Pelletization efficiency of produced aggregates

Pelletization efficiency results for FA-BT, FA-BL, and FA-GGBS aggregates are shown in Figs. 2(e), 3(e), and 4(e), respectively (as specified in Table 3). It can be observed from Figs. that aggregate mixes FABT 9, FABL 5, and FAGGBS 7 are found to have maximum pelletization efficiency compared to other aggregate mixes of FA-BT, FA-BL, and FA-GGBS, respectively. It has been reported that the aggregate production stability differs during the process of pelletization with the incorporation of mineral admixtures (Gomathi and Sivakumar 2015). Therefore, this could be one of the possible reasons for obtaining a varied pelletization efficiency for FA-BT, FA-BL and FA-GGBS aggregates comprising different mineral admixtures.

However, it is important to understand the influence of governing factors on the production efficiency of produced FA-BT, FA-BL, and FA-GGBS aggregates. Keeping this in view, three dimensional (3-D) plots are developed for FA-BT, FA-BL, and FA-GGBS and the same is presented in Figures 11, 12 and 13, respectively. It should be noted that developed 3-D graphs can be used to understand how specific parameters affect production efficiency.

4.6.Morphology of produced aggregates

SEM studies on different sets of produced fly ash aggregates gave an insight into the effect of raw materials used such as BT, BL, and GGBS on the microstructure at different levels of replacement and alkali binder ratio. The micrographs taken in secondary electron mode for the set of FA-BT, FA-BL, and FA-GGBS mixes at the curing age of 100 days are presented in Figs. 14-16 respectively.

It can be observed from Fig. 14 that all the nine mixes of the FA-BT set showed different morphologies. FABT 1 to FABT 9 mixes showed large traces of unreacted spherical fly ash particles. It can also be noticed that mixes comprise plate-like structures representing the presence of unreacted bentonite particles in the mixes. However, it can be seen from Fig. 14 that traces of spherical and plate-like structures are found to be reduced for FABT 9 aggregate mix, which indicates the involvement of fly ash and bentonite particles in the process of polymerization. Further, the image displays a more compact and denser grey microstructure owing to the larger formation of hydration products (N-A-S-H/C-A-S-H). SEM images of FABL 1-4 and FABL 6-FABL 9 mixes, as well as FAGGBS 1-FAGGBS 6 and FAGGBS 8-FAGGBS 9 mixes, show large amounts of unreacted fly ash particles (spherical particles) and mushy lime particles adhering over the surface of fly ash particles (FABL mixes) and granular particles of GGBS (FA-GGBS mixes). Among FA-BL and FA-GGBS aggregate mixes, unreacted particles are found to be minimised in the micrograph of FABL 5 and FAGGBS 7 mixes that represent the improved microstructure with a denser and more homogeneous matrix of hydration products.

4.7. Thermogravimetric analysis (TGA)

TG-DTG plots for FABT, FABL and FAGGBS aggregates are presented in the Figs. 17 (a), (b) and (c) respectively.

In Fig. 17, TG curve represents the occurrence of thermogravimetric mass loss for geopolymerized samples during the process of heating from the temperature range of 25-850 °C, whereas the derivative of thermogravimetry (DTG) curve signifies the temperature boundaries for the decomposition of specific compounds. According to Fig 17, there is a series of endothermic peaks in the temperature range of 25-850 °C. First, a significant endothermic peak at 25-120 °C indicates the loss of physically absorbed free water molecules on the pores and surfaces of the samples (Adriano et al. 2013; Longhi et al. 2019; Wuddivira et al. 2012). The next significant peak was noticed at a temperature range of 120-225 °C that was associated with the thermal degradation of chemically bound water from sodium aluminosilicate gel (N-A-S-H) or calcium-aluminosilicate gel (C-A-S-H) (Adesanya et al. 2018; Ismail et al. 2014; Palomo et al. 2015). The endothermic peak at the temperature boundaries of 600-700 °C indicates the associated decomposition of carbonates (C) (Abdullah et al. 2018; Cornejo et al. 2018; Everaert et al. 2017). It is worth noting that in the presence of BL and GGBS, the DTG curve exhibits an additional endothermic peak at 400-550 °C, representing the dehydroxylation of calcium hydroxide (Ca $(OH)_2$) (Palomo et al. 2015).

By adopting the mass loss from the TG-DTG, the following equation at certain boundaries of temperature is proposed below.

$$\Delta M_{N-A-S-H/C-A-S-H}\,\% = M_{120°C} - M_{225°C} \quad (7)$$

where, $\Delta M_{N-A-S-H/C-A-S-H}$ is change in mass loss percentage of N-A-S-H/C-A-S-H and $M_{120°C}$, $M_{225°C}$ is the mass loss at the temperatures of 120 and 225 °C.

The decomposition of N-A-S-H/C-A-S-H for the obtained TG-DTG curves for all the three aggregates, i.e., FABT, FABL, and FAGGBS, was quantified following the mathematical equation proposed, Eq. 7, and the same is presented in Fig. 18 (a-c).

It can be observed from Fig. 18 that the quantified amount of major reaction product that is sodium aluminium silicate hydrate gel (N-A-S-H) is found to be intensified in proportion to Na_2O dosages, irrespective of the additives used in this study. It is reported that the amount of N-A-S-H formed in mixes indicates the extent of geopolymerization reaction (Garg et al. 2019). In the case of FABT aggregates, Fig. 18(a), the amount of N-A-S-H produced was found to be in the range of 0.18-0.2% for 3% Na_2O mixes (FABT 1-FABT 3) and increased by 30-40% and 40-60% for 4% (FABT 4-FABT 6) and 5% Na_2O (FABT 7-FABT 9) aggregate mixes, respectively.

Figures 18(b) (FABL aggregate mixes) and 18(c) (FAGGBS aggregate mixes) show that calcium rich mixes have a higher percentage of mass loss at temperature boundaries of 120-225 °C, indicating the formation of reaction products related to both C-A-S-H and N-A-S-H (Rafeet et al. 2019). Mass loss associated with C-A-S-H/N-A-S-H was found to be increased with the increase in burnt lime (15%) and GGBS (15%) content. FABL 5 (5% Na_2O, 20% water, and 15% burnt lime content) and FAGGBS 7 (5% Na_2O, 19% water, and 15% GGBS content) have the highest amount of hydration products (i.e., C-A-S-H and N-A-S-H) of all FA-BL and FA-GGBS aggregate mixes.

In order to understand the influence of the formation of N-A-S-H/C-A-S-H on the individual pellet strength of all the mixes, the relationship between the percentage of mass loss associated with N-

A-S-H/C-A-S-H ($\Delta M_{N\text{-}A\text{-}S\text{-}H/C\text{-}A\text{-}S\text{-}H}$) and individual crushing strength of pellets (IPS) is plotted and presented in Fig. 19 (a-c).

It can be observed from Fig.19 (a-c), that there exists a linear relationship between the individual crushing strength of pellets and the quantified amount of N-A-S-H/C-A-S-H for FABT, FABL, and FAGGBS aggregate mixes. The coefficient of correlation (R^2) values for FABT, FABL, and FAGGBS aggregate mixes were found to be 0.82, 0.91, and 0.92, respectively. This clearly indicates that N-A-S-H/C-A-S-H content in geopolymerized mixes greatly influences the individual pellet strength of the produced aggregates.

4.8. Fourier transform infrared spectroscopy

The FTIR spectra for FA, BT, BL and GGBS are presented in Fig. 20(a-d).

From Fig. 20 (a), it can be observed that the bands at 794.37 cm^{-1} and 599.37 cm^{-1} are ascribed to α-quartz and mullite, respectively (Coates 1977; Hlavay et al. 1978; Lee and Van Deventer 2002). A prominent band related to the geopolymerization of fly ash exists around wavenumber 1075.58 cm^{-1}, which is an asymmetric vibrational band of Si-O-Si and Si-O-Al compounds (Khale and Chaudhary 2007). The next bands at wavenumbers 2933.22 cm^{-1} and 3444.32 cm^{-1} correspond to organic carbon (C-H stretching) (Saikia et al. 2008) and O-H groups of silanols and hydrogen bonds between water-bound molecules and silanols, respectively (Summer 1995).

In Fig. 20 (b), the FTIR spectra for BT is presented. The bands at 3620.85 cm^{-1} and 917.52 cm^{-1} are mainly of dioctahedral smectites (Caillère et al. 1982; Dixon et al. 1977). The bands at 849.79 cm^{-1} and 3672.16 cm^{-1} are attributed to Al-OH-Mg bonds. The bands at 628.10 cm^{-1} and 671.21 cm^{-1} correspond to Si-O-Al and Si-O-Mg respectively. The 3454.59 cm^{-1} band is attributed to O-H frequencies of water.

Fig. 20 (c) represents an FTIR spectra of BL. From the figure, it can be observed that the strongest band located at 3643.43 cm^{-1} represents O-H bonds from the leftover hydroxide (Park et al. 2002). The occurrence of the spectrum at the wavelengths of 1418.27 cm^{-1} and 870.21 cm^{-1} (centered band) is attributed to the C-O bond. The strong bond at around 599.37 cm^{-1} shows Ca-O bonds.

Fig. 20 (d) represents the FTIR spectra of GGBS. From the figure, it can be observed that the important vibration band exists at around 901.10 cm^{-1}, which is allied with the asymmetric stretching vibration of the terminal Si-O bond (Zhang et al. 2012). The next important band is located at 985.26 cm^{-1}, which is associated with the asymmetric stretching vibration of the Si-O-

T bond. The existence of peaks at 3733.74 cm^{-1} is allotted to O-H stretching vibration, which indicates moisture in the raw material.

Fig. 21 (a), (b) and (c) represent the spectra of FABT, FABL, and FAGGBS aggregates, respectively.

While comparing Fig. 21 (a) with Figs. 20 (a) and (b), Fig. 21 (b) with Figs. 20 (a) and (c), and Fig. 21(c) with Figs. 20 (a) and (d), it can be observed that with the completion of the geopolymerization process, the strong band located at 1073.52 cm^{-1} for unreacted fly ash gets broadened and it shifts to lesser frequencies by more than 40-50 cm^{-1} for FA-BT, FA-BL, and FA-GGBS aggregates, consisting of fly ash as a precursor. Based on this observation, it can be attributed that the geopolymerization process increases the aluminosilicates (amorphous by nature) substantially (Bernal et al. 2011). However, it also indicates that the highly crosslinked networks with Si-O bonds (as connectors) are not formed because of the existence of high amounts of calcium at initial stages, which uses the Si and Al units and mostly forms C-A-S-H gels, so that the building of Si and Al units is hampered (Lee and Van Deventer 2002).

An important band representing the products formed in the geopolymerization reaction is in the produced FA-BT, FA-BL and FA-GGBS aggregates, which is associated with the asymmetric stretching vibration of Si-O terminal bonds, representing that the geopolymerization reaction produces N-A-S-H/C-A-S-H gel (chain-structured) (Gao et al. 2015).

It is also worth noting that the bands in Figs. 21(a), (b) are nearly identical at 1009.89 cm^{-1} and 990 cm^{-1} when compared to the unreacted fly ash band (Fig. 20(a)), where the bands shifted to lower frequencies for FA-BT and FA-BL aggregates, respectively (Gao et al. 2015). However, it can be understood from Figs. 21 (c) and 20 (d) that the band Si-O located at 901.10 cm^{-1} in raw GGBS is shifted to higher wavenumbers, which signifies the formation of an extra highly polymerized network (Gao et al. 2015). The bands attributed to the vibrations of O-C-O in carbonates (Gao et al. 2015) in the produced FA-BT, FA-BL, and FA-GGBS aggregates are presented in Tables 4, 5, and 6, respectively. It can be observed that, as the dosage content of lime increases, the transmittance band intensity, which represents carbonates, also increases (Gao et al. 2015).

Based on the above observations of IR spectra, it can be inferred that the spectra of produced FA-BT, FA-BL, and FA-GGBS are completely different as compared to the raw materials. That indicates a substantial amount of amorphous natured aluminosilicates are being produced in the geopolymerization process (Gao et al. 2015).

4. Grey relational analysis

As explained in the previous sections, the characteristic properties of FA-BT, FA-BL, and FA-GGBS aggregates are aggregate impact and crushing value, individual crushing strength of pellets, and water absorption. These were determined by carrying out the experiments and were termed as responses. In order to employ grey relational analysis for this experimental study, the responses which were yielded through the nine trial sets of Taguchi's L_9 orthogonal array presented in Table 3 were used.

4.1.1 Normalizing the data and generating the grey relational generations

Experimentally obtained responses for Taguchi's L_9 orthogonal array design (depicted in Table 3) are presented in Figs. 2, 3 and 4 for FA-BT, FA-BL, and FA-GGBS aggregates, respectively, were used for obtaining an unbiased analysis for determining the order of influence of governing factors in the produced aggregates (Sahoo et al. 2017). Firstly, the obtained results for FA-BT, FA-BL and FA-GGBS aggregates were normalized and grey relational generations were calculated. For FA-BT, FA-BL, and FA-GGBS aggregates, the grey relational generations for properties like aggregate impact value, aggregate crushing value, and water absorption at all the three curing were calculated using Eqn. 1, respectively. However, for individual pellet strength properties, the grey relational generations at 14, 28, and 100 days were calculated using Eqn. 2 for FA-BT, FA-BL, and FA-GGBS aggregates, respectively.

The grey relational generations for the obtained characteristic properties of produced FA-BT, FA-BL and FA-GGBS aggregates for 14, 28 and 100 days of curing are presented in Tables 4-6, respectively. However, for FA-BL and FA-GGBS aggregates, the obtained responses for all the characteristic properties and their respective grey relational generations at all the three curing ages is presented in Appendix A (Tables A1-A3: for FA-BT aggregates and A4-A6: for FA-BL aggregates).

4.1.2 Assessment of grey relational coefficients

The deviation sequences Δ_{0i} and grey relational coefficients ξ_i (k) for FA-BT, FA-BL, and FA-GGBS aggregates at all the three curing ages were computed by using Eqn. 3. For FA-BT aggregates, the values for deviation sequence and grey relational coefficients for 14, 28, and 100 days of curing are presented in Tables 7-19, respectively. For FA-BL and FA-GGBS aggregates, the deviation sequence and grey relational coefficient values (for 14, 28, and 100 days) are presented in Appendix B (B1-B3: for FA-BL aggregates and B4-B6: for FA-GGBS aggregates).

4.1.3 Grey relational grade

The grey relational grade γ_i for FA-BT, FA-BL, and FA-GGBS aggregates at all the three curing ages was computed using Eqn. 4.

The grey relational grade at all curing ages for all the respective governing factors in FA-BT aggregates is presented in Table 10. For FA-BL and FA-GGBS aggregates, the grey relational grades for all the three curing ages are presented in Appendix C (C1: for FA-BL and C2: for FA-GGBS aggregates).

Tables 11-13 represent the average values of the response characteristics, levels, and the governing factors in FA-BT, FA-BL, and FA-GGBS aggregates, respectively. By observing the delta values or ranks obtained for FA-BT, FA-BL, and FA-GGBS aggregates, it can be inferred that the factor Na_2O content is having a significant impact among all the other factors considered in the production of three kinds of aggregates. The next most significant ones were found to be BT and GGBS content, followed by water content in the production of FA-BT and FA-GGBS aggregates, respectively. However, for FA-BL aggregates, water content was found to be more significant than BL content. As a result of summarising this analysis, it is possible to generalise that the production as well as the characteristics of FA-BT, FA-BL, and FA-GGBS aggregates are highly susceptible to the factors considered in their manufacturing process.

5. Conclusions

This study contributed a systematic methodology through which production of pelletized fly ash based aggregates with the utilization of fly ash and additives such as bentonite (BT), burnt lime and ground granulated blast furnace slag (GGBS) can be achieved. The produced fly ash based aggregates are well characterized and obtained through ambient curing that is a step forward and this will also help in mitigating the huge scarcity of natural available aggregates in the construction industry.

Further, based on the obtained results the following conclusions can be drawn.

- The incorporation of admixtures such as BT, BL, and GGBS improved the engineering properties of produced fly ash-based aggregates. In the production of fly ash based aggregates admixed with BT and GGBS (FA-BT and FA-GGBS), Na_2O and mineral admixture (BT/GGBS) content are found to be more influential factors (represented as water < BT/GGBS < Na_2O). Whereas, for fly ash based aggregates admixed with burnt lime (FA-BL), Na_2O and water content were found to be the principal factors (represented as BL < water < Na_2O).
- Among FA-BT, FA-BL, and FA-GGBS aggregates, the results obtained for aggregate impact value, aggregate crushing value, individual crushing strength of pellets, and pelletization efficiency were found to be superior for aggregates admixed with burnt lime (BL), which indicates that calcium rich additives can be admixed with fly ash to obtain the desired results. This behavior of burnt lime addition is completely relatable to the inferences drawn from TGA analysis. However, the water absorption of aggregates with GGBS addition fared better in comparison with values obtained for FA-BT and FA-GGBS aggregates.
- Morphological studies revealed that aggregates produced with higher dosages of Na_2O and mineral admixture possess a more homogenous and denser microstructure.
- The quantified amount of geopolymerization products (N-A-S-H/C-A-S-H) was found to be the maximum for fly ash aggregates produced with 5% Na_2O content and 15% mineral admixture. However, mass loss associated with N-A-S-H/C-A-S-H was found to be intensified for fly ash aggregates admixed with calcium-rich minerals, i.e., BT and GGBS.
- Individual pellet strength of the produced aggregates directly relates to the extent of N-A-S-H/C-A-S-H formation.
- It can be concluded from FTIR analysis that strong bands of Si-O-T and Si-O observed in raw FA, BT, BL, and GGBS particles were found to be broadened and shifted towards lower wavelength, which signified the formation of amorphous, chain-structured geopolymerization reaction products (N-A-S-H/C-A-S-H gel). Grey relational analysis states that the influence of Na_2O is one of the dominant factors of geopolymerization.

Future scope for research

- Mass scale production of these aggregates and the uniformity in achieving the quality of the aggregate properties is the need of the hour and Authors are looking forward to explore the possibilities.
- There is a huge scope available to find out the possible utilization of these produced pelletized fly ash based coarse aggregates in concrete production with partial or full replacement with the natural aggregates.
- Further, it is also essential to understand the behavior of structural members under various stress states that is made up of concrete with artificial aggregates.

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Data Availability Statement

All data, models, and code generated or used during the study are presented in the manuscript suitably.

References

Abdul Rahim, R. H., K. A. Azizli, Z. Man, T. Rahmiati, and L. Ismail. 2014. "Effect of solid to liquid ratio on the mechanical and physical properties of fly ash geopolymer without sodium silicate." *Appl. Mech. Mater.*, 46–49. Trans Tech Publ.

Abdullah, M., L. Y. Ming, H. C. Yong, and M. F. M. Tahir. 2018. "Cement Based Materials." InTechOpen London, UK:

Adesanya, E., K. Ohenoja, T. Luukkonen, P. Kinnunen, and M. Illikainen. 2018. "One-part geopolymer cement from slag and pretreated paper sludge." *J. Clean. Prod.*, 185: 168–175. Elsevier.

Adriano, A., G. Soriano, and J. Duque. 2013. "Characterization of water absorption and desorption properties of natural zeolites in Ecuador." *Fifth Int. Symp. Energy*, 1–9.

Aggregates, A. C. C.-9 on C. and C. 2017. *Standard specification for lightweight aggregates for structural concrete*. ASTM International.

Aslam, M., P. Shafigh, M. Z. Jumaat, and M. Lachemi. 2016. “Benefits of using blended waste coarse lightweight aggregates in structural lightweight aggregate concrete.” *J. Clean. Prod.*, 119: 108–117. Elsevier.

ASTM, A. I. 2018. “ASTM C618-19 Standard Specification for Coal Fly Ash and Raw or Calcined Natural Pozzolan for Use in Concrete.” *ASTM Int. West Conshohocken, PA*.

Ayati, B., V. Ferrándiz-Mas, D. Newport, and C. Cheeseman. 2018. “Use of clay in the manufacture of lightweight aggregate.” *Constr. Build. Mater.*, 162: 124–131. Elsevier.

Baykal, G., and A. G. Döven. 2000. “Utilization of fly ash by pelletization process; theory, application areas and research results.” *Resour. Conserv. Recycl.*, 30 (1): 59–77. Elsevier.

Bernal, S. A., J. L. Provis, V. Rose, and R. M. De Gutierrez. 2011. “Evolution of binder structure in sodium silicate-activated slag-metakaolin blends.” *Cem. Concr. Compos.*, 33 (1): 46–54. Elsevier.

Bijen, J. M. J. M. 1986a. “Manufacturing processes of artificial lightweight aggregates from fly ash.” *Int. J. Cem. Compos. Light. Concr.*, 8 (3): 191–199. https://doi.org/10.1016/0262-5075(86)90040-0.

Bijen, J. M. J. M. 1986b. “Manufacturing processes of artificial lightweight aggregates from fly ash.” *Int. J. Cem. Compos. Light. Concr.*, 8 (3): 191–199. Elsevier. https://doi.org/10.1016/0262-5075(86)90040-0.

Bui, L. A., C. Hwang, C. Chen, K. Lin, and M. Hsieh. 2012a. “Manufacture and performance of cold bonded lightweight aggregate using alkaline activators for high performance concrete.” *Constr. Build. Mater.*, 35: 1056–1062. Elsevier.

Bui, L. A. T., C. L. Hwang, C. T. Chen, and M. Y. Hsieh. 2012b. “Characteristics of cold-bonded lightweight aggregate produced with different mineral admixtures.” *Appl. Mech. Mater.*, 978–983. Trans Tech Publ.

Bureau of Indian Standards (BIS). 2002. “IS : 2366 (Part IV)-1963-Methods of test for Aggregates for Concrete, part 4 : Mechanical properties.” *Indian Stand.*, 1–37.

Caillère, S., S. Hénin, and M. Rautureau. 1982. *Minéralogie des argiles: structure et propriétés physico-chimiques*. Masson Paris.

Cavazzuti, M. 2013. “Design of experiments.” *Optim. methods*, 13–42. Springer.

Chi, J. M., R. Huang, C.-C. Yang, and J. J. Chang. 2003. “Effect of aggregate properties on the strength and stiffness of lightweight concrete.” *Cem. Concr. Compos.*, 25 (2): 197–205. Elsevier.

Chindaprasirt, P., P. De Silva, K. Sagoe-Crentsil, and S. Hanjitsuwan. 2012. “Effect of SiO2 and Al2O3 on the setting and hardening of high calcium fly ash-based geopolymer systems.” *J. Mater. Sci.*, 47 (12): 4876–4883. Springer.

Coates, J. P. 1977. “The IR analysis of Quartz and Asbestos.” *Nelioth Offset Ltd., Chesham, Engl.*

Colangelo, F., R. Cioffi, F. Montagnaro, and L. Santoro. 2012. “Soluble salt removal from MSWI fly ash and its stabilization for safer disposal and recovery as road basement material.” *Waste Manag.*, 32 (6): 1179–1185. Elsevier.

Cornejo, M. H., J. Elsen, B. Togra, H. Baykara, G. Soriano, and C. Paredes. 2018. “Effect of calcium hydroxide and water to solid ratio on compressive strength of mordenite-based geopolymer and the evaluation of its thermal transmission property.” *ASME Int. Mech. Eng. Congr. Expo.*, V012T11A022. American Society of Mechanical Engineers.

Davidovits, J. 1989. “Geopolymers and geopolymeric materials.” *J. Therm. Anal.*, 35 (2): 429–441. Springer.

DIN, E. N. 2015. “13055: 2015-11: Leichte Gesteinskörnungen.” *Dtsch. und Englische Fassung FprEN*, 13055.

Dixon, J. B., S. B. Weed, and R. C. Dinauer. 1977. “Minerals in soil environments.” Soil Science Society of America,.

Everaert, C., M. Luypaert, J. L. V Maag, Q. X. Cheng, M. E. Dinger, J. Hellemans, and P. Mestdagh. 2017. “Benchmarking of RNA-sequencing analysis workflows using whole-transcriptome RT-qPCR expression data.” *Sci. Rep.*, 7 (1): 1–11. Nature Publishing Group.

Fernández-Jiménez, A., and A. Palomo. 2005. “Composition and microstructure of alkali activated fly ash binder: Effect of the activator.” *Cem. Concr. Res.*, 35 (10): 1984–1992. Elsevier.

Franus, M., D. Barnat-Hunek, and M. Wdowin. 2016. “Utilization of sewage sludge in the manufacture of lightweight aggregate.” *Environ. Monit. Assess.*, 188 (1): 1–13. Springer.

Freedonia-Group. 2016. “The Freedonia Group: World Construction Aggregates to Reach 51.7 Billion Metric Tons.” The Freedonia Group Cleveland, OH, USA.

Gao, X., Q. L. Yu, and H. J. H. Brouwers. 2015. “Properties of alkali activated slag–fly ash blends with limestone addition.” *Cem. Concr. Compos.*, 59: 119–128. Elsevier.

Garg, N., V. O. Özçelik, J. Skibsted, and C. E. White. 2019. “Nanoscale ordering and depolymerization of calcium silicate hydrates in the presence of alkalis.” *J. Phys. Chem. C*, 123 (40): 24873–24883. ACS Publications.

Geetha, S., and K. Ramamurthy. 2010. “Reuse potential of low-calcium bottom ash as aggregate through pelletization.” *Waste Manag.*, 30 (8–9): 1528–1535. Elsevier.

Geetha, S., and K. Ramamurthy. 2011. “Properties of sintered low calcium bottom ash aggregate with clay binders.” *Constr. Build. Mater.*, 25 (4): 2002–2013. Elsevier.

Geetha, S., and K. Ramamurthy. 2013. “Properties of geopolymerised low-calcium bottom ash aggregate cured at ambient temperature.” *Cem. Concr. Compos.*, 43: 20–30. Elsevier.

Gesoğlu, M., E. Güneyisi, and H. Ö. Öz. 2012. “Properties of lightweight aggregates produced with cold-bonding pelletization of fly ash and ground granulated blast furnace slag.” *Mater. Struct.*, 45 (10): 1535–1546. Springer.

Gomathi, P., and A. Sivakumar. 2012. “Characterization on the strength properties of pelletized fly ash aggregate.” *ARPN J. Eng. Appl. Sci.*, 7 (11): 1523–1532.

Gomathi, P., and A. Sivakumar. 2014. “Fly ash based lightweight aggregates incorporating clay binders.” *Indian J. Eng. Mater. Sci.*, 21 (2): 227–232.

Gomathi, P., and A. Sivakumar. 2015. “Accelerated curing effects on the mechanical performance of cold bonded and sintered fly ash aggregate concrete.” *Constr. Build. Mater.*, 77: 276–287. Elsevier.

Görhan, G., and G. Kürklü. 2014. “The influence of the NaOH solution on the properties of the fly ash-based geopolymer mortar cured at different temperatures.” *Compos. part b Eng.*, 58: 371–377. Elsevier.

Hardjito, D., C. C. Cheak, and C. H. L. Ing. 2008. “Strength and setting times of low calcium fly ash-based geopolymer mortar.” *Mod. Appl. Sci.*, 2 (4): 3–11. Citeseer.

Hardjito, D., S. E. Wallah, D. M. J. Sumajouw, and B. V. Rangan. 2004. “On the development of fly ash-based geopolymer concrete.” *Mater. J.*, 101 (6): 467–472.

Harikrishnan, K. I., and K. Ramamurthy. 2006. “Influence of pelletization process on the properties of fly ash aggregates.” *Waste Manag.*, 26 (8): 846–852. Elsevier.

Hlavay, J., K. Jonas, S. Elek, and J. Inczedy. 1978. “Characterization of the particle size and the

crystallinity of certain minerals by ir spectrophotometry and other instrumental methods—II. Investigations on quartz and feldspar." *Clays Clay Miner.*, 26 (2): 139–143. Springer.
Hoff, G. C. 1993. "High Strength Lightweight Aggregate Concrete for Arctic Applications--Part 1: Unhardened Concrete Properties." *Spec. Publ.*, 136: 1–66.
Holm, T. A., and J. P. Ries. 2006. "Lightweight concrete and aggregates." *Significance tests Prop. Concr. Concr. Mater.* ASTM International.
Hwang, C.-L., and V.-A. Tran. 2015. "A study of the properties of foamed lightweight aggregate for self-consolidating concrete." *Constr. Build. Mater.*, 87: 78–85. Elsevier.
IS: 3812 (Part-1). 2003. "Pulverized fuel ash — specification. Part 1: For use as Pozzolana in cement, Cement Mortar and Concrete (Second Revision)." *Bur. Indian Stand.*, (October): 1–14.
IS:383-2016. 2016. "Indian Standard Coarse and Fine aggregate for Concrete- Specification." *Bur. Indian Stand. New Delhi, India*, (January): 1–21.
Ismail, I., S. A. Bernal, J. L. Provis, R. San Nicolas, S. Hamdan, and J. S. J. van Deventer. 2014. "Modification of phase evolution in alkali-activated blast furnace slag by the incorporation of fly ash." *Cem. Concr. Compos.*, 45: 125–135. Elsevier.
Kayali, O. 2008. "Fly ash lightweight aggregates in high performance concrete." *Constr. Build. Mater.*, 22 (12): 2393–2399. Elsevier.
Khale, D., and R. Chaudhary. 2007. "Mechanism of geopolymerization and factors influencing its development: a review." *J. Mater. Sci.*, 42 (3): 729–746. Springer.
Kockal, N. U., and T. Ozturan. 2011. "Characteristics of lightweight fly ash aggregates produced with different binders and heat treatments." *Cem. Concr. Compos.*, 33 (1): 61–67. Elsevier.
Komljenović, M., Z. Baščarević, and V. Bradić. 2010. "Mechanical and microstructural properties of alkali-activated fly ash geopolymers." *J. Hazard. Mater.*, 181 (1–3): 35–42. Elsevier.
Krishnan, T., and R. Purushothaman. 2017. "Optimization and influence of parameter affecting the compressive strength of geopolymer concrete containing recycled concrete aggregate: using full factorial design approach." *IOP Conf. Ser. Earth Environ. Sci.*, 12013. IOP Publishing.
Lee, W. K. W., and J. S. J. Van Deventer. 2002. "Structural reorganisation of class F fly ash in alkaline silicate solutions." *Colloids Surfaces A Physicochem. Eng. Asp.*, 211 (1): 49–66.

Elsevier.

Lo, T. Y., H. Cui, S. A. Memon, and T. Noguchi. 2016. “Manufacturing of sintered lightweight aggregate using high-carbon fly ash and its effect on the mechanical properties and microstructure of concrete.” *J. Clean. Prod.*, 112: 753–762. Elsevier.

Longhi, M. A., Z. Zhang, E. D. Rodríguez, A. P. Kirchheim, and H. Wang. 2019. “Efflorescence of alkali-activated cements (geopolymers) and the impacts on material structures: A critical analysis.” *Front. Mater.*, 6: 89. Frontiers.

Manikandan, R., and K. Ramamurthy. 2007. “Influence of fineness of fly ash on the aggregate pelletization process.” *Cem. Concr. Compos.* https://doi.org/10.1016/j.cemconcomp.2007.01.002.

Manikandan, R., and K. Ramamurthy. 2008. “Effect of curing method on characteristics of cold bonded fly ash aggregates.” *Cem. Concr. Compos.*, 30 (9): 848–853. Elsevier.

Manikandan, R., and K. Ramamurthy. 2009. “Swelling characteristic of bentonite on pelletization and properties of fly ash aggregates.” *J. Mater. Civ. Eng.*, 21 (10): 578–586. American Society of Civil Engineers.

Mannan, M. A., and K. Neglo. 2010. “Mix design for oil-palm-boiler clinker (OPBC) concrete.” *J. Sci. Technol.*, 30 (1).

Montgomery, D. C. 2017. *Design and analysis of experiments*. John wiley & sons.

Mustafa Al Bakri, A. M., H. Kamarudin, M. Bnhussain, A. R. Rafiza, and Y. Zarina. 2012. “Effect of Na 2 SiO 3/NaOH Ratios and NaOH Molarities on Compressive Strength of Fly-Ash-Based Geopolymer.” *ACI Mater. J.*, 109 (5).

Narattha, C., and A. Chaipanich. 2018. “Phase characterizations, physical properties and strength of environment-friendly cold-bonded fly ash lightweight aggregates.” *J. Clean. Prod.*, 171: 1094–1100. Elsevier.

Nath, P., P. K. Sarker, and V. B. Rangan. 2015. “Early age properties of low-calcium fly ash geopolymer concrete suitable for ambient curing.” *Procedia Eng.*, 125: 601–607. Elsevier.

Nor, A. M., Z. Yahya, M. M. A. B. Abdullah, R. A. Razak, J. J. Ekaputri, M. A. Faris, and H. N. Hamzah. 2016. “A Review on the Manufacturing of Lightweight Aggregates Using Industrial By-Product.” *MATEC Web Conf.*, 1067. EDP Sciences.

Oktay, H., R. Yumrutaş, and A. Akpolat. 2015. “Mechanical and thermophysical properties of lightweight aggregate concretes.” *Constr. Build. Mater.*, 96: 217–225. Elsevier.

Palomo, Á., E. Kavalerova, A. Fernández-Jiménez, P. Krivenko, I. García-Lodeiro, and O. Maltseva. 2015. “A review on alkaline activation: new analytical perspectives.” CSIC-Instituto de Ciencias de la Construcción Eduardo Torroja (IETCC).

Park, J. H., D. J. Min, and H. S. Song. 2002. “Structural investigation of CaO–Al2O3 and CaO–Al2O3–CaF2 slags via Fourier transform infrared spectra.” *ISIJ Int.*, 42 (1): 38–43. The Iron and Steel Institute of Japan.

Patankar, S. V, Y. M. Ghugal, and S. S. Jamkar. 2014. “Effect of concentration of sodium hydroxide and degree of heat curing on fly ash-based geopolymer mortar.” *Indian J. Mater. Sci.*, 2014. Hindawi.

Priyadharshini, P., M. G. Ganesh, and A. S. Santhi. 2011. “Experimental study on cold bonded fly ash aggregates.” *Int. J. Civ. Struct. Eng.*, 2 (2): 493. Integrated Publishing Association.

Rafeet, A., R. Vinai, M. Soutsos, and W. Sha. 2019. “Effects of slag substitution on physical and mechanical properties of fly ash-based alkali activated binders (AABs).” *Cem. Concr. Res.*, 122: 118–135. Elsevier.

Rahmiati, T., K. A. Azizli, Z. Man, L. Ismail, and M. F. Nuruddin. 2015. “Effect of solid/liquid ratio during curing time fly ash based geopolymer on mechanical property.” *Mater. Sci. Forum*, 120–124. Trans Tech Publ.

Ramamurthy, K., and K. I. Harikrishnan. 2006. “Influence of binders on properties of sintered fly ash aggregate.” *Cem. Concr. Compos.*, 28 (1): 33–38. Elsevier.

Rangan, B. V., D. Hardjito, S. E. Wallah, and D. M. J. Sumajouw. 2005. “Studies on fly ash-based geopolymer concrete.” *Proc. World Congr. Geopolymer, Saint Quentin, Fr.*, 133–137.

Rattanasak, U., and P. Chindaprasirt. 2009. “Influence of NaOH solution on the synthesis of fly ash geopolymer.” *Miner. Eng.*, 22 (12): 1073–1078. Elsevier.

Reddy, M. V. S., M. Nataraja, K. Sindhu, V. Harani, and K. Madhuralalasa. 2016. “Performance of light weight concrete using fly ash pellets as coarse aggregate replacement.” *Int. J. Eng. Res. Technol.*, 9: 95–104.

Sahoo, S., B. B. Das, and S. Mustakim. 2017. “Acid, alkali, and chloride resistance of concrete composed of low-carbonated fly ash.” *J. Mater. Civ. Eng.*, 29 (3): 4016242. American Society of Civil Engineers.

Saikia, B. J., G. Parthasarathy, N. C. Sarmah, and G. D. Baruah. 2008. “Fourier-transform infrared spectroscopic characterization of naturally occurring glassy fulgurites." *Bull. Mater. Sci.*, 31 (2): 155–158. Springer.

Sharath, B. P., and B. B. Das. 2021. "Production of Artificial Aggregates Using Industrial By-Products Admixed with Mine Tailings—A Sustainable Solution." *Recent Trends Civ. Eng.*, 383–397. Springer.

Shivaprasad, K. N., and B. B. Das. 2018. "Determination of optimized geopolymerization factors on the properties of pelletized fly ash aggregates." *Constr. Build. Mater.*, 163: 428–437. Elsevier.

Shivaprasad, K. N., and B. B. Das. 2021. "Study on the Production Factors in the Process of Production and Properties of Fly Ash-Based Coarse Aggregates." *Adv. Civ. Eng.*, 2021. Hindawi.

Somayaji, S. 1985. *Civil engineering materials*. Pearson Education India.

Soudki, K. A., E. F. El-Salakawy, and N. B. Elkum. 2001. "Full factorial optimization of concrete mix design for hot climates." *J. Mater. Civ. Eng.*, 13 (6): 427–433. American Society of Civil Engineers.

Sumer, M. 2012. "Compressive strength and sulfate resistance properties of concretes containing Class F and Class C fly ashes." *Constr. Build. Mater.*, 34: 531–536. Elsevier.

Summer, M. E. 1995. "Hand Book of Soil Science. University of Georgia." Boca Raton Hondor press, New York.

Tang, P., M. V. A. Florea, and H. J. H. Brouwers. 2017. "Employing cold bonded pelletization to produce lightweight aggregates from incineration fine bottom ash." *J. Clean. Prod.*, 165: 1371–1384. Elsevier.

Terzić, A., L. Pezo, V. Mitić, and Z. Radojević. 2015. "Artificial fly ash based aggregates properties influence on lightweight concrete performances." *Ceram. Int.*, 41 (2): 2714–2726. Elsevier.

Videla, C., and P. M. Martinez. 2002. "Fly ash lightweight aggregates produced by cold bonding for sustainable concrete construction." *Challenges Concr. Constr. Vol. 5, Sustain. Concr. Constr. Proc. Int. Conf. held Univ. Dundee, Scotland, UK 9–11 Sept. 2002*, 363–372. Thomas Telford Publishing.

Wasserman, R., and A. Bentur. 1997. "Effect of lightweight fly ash aggregate microstructure on the strength of concretes." *Cem. Concr. Res.*, 27 (4): 525–537. Elsevier.

Wuddivira, M. N., D. A. Robinson, I. Lebron, L. Bréchet, M. Atwell, S. De Caires, M. Oatham, S. B. Jones, H. Abdu, and A. K. Verma. 2012. “Estimation of soil clay content from hygroscopic water content measurements.” *Soil Sci. Soc. Am. J.*, 76 (5): 1529–1535.

Yang, K.-H., J.-H. Mun, J.-I. Sim, and J.-K. Song. 2011. “Effect of water content on the properties of lightweight alkali-activated slag concrete.” *J. Mater. Civ. Eng.*, 23 (6): 886–894. American Society of Civil Engineers.

Zhang, Z., H. Wang, J. L. Provis, F. Bullen, A. Reid, and Y. Zhu. 2012. “Quantitative kinetic and structural analysis of geopolymers. Part 1. The activation of metakaolin with sodium hydroxide.” *Thermochim. Acta*, 539: 23–33. Elsevier.

Zhao, X., C. Liu, L. Zuo, L. Wang, Q. Zhu, and M. Wang. 2019. “Investigation into the effect of calcium on the existence form of geopolymerized gel product of fly ash based geopolymers.” *Cem. Concr. Compos.*, 103: 279–292. Elsevier.

Zion (2019) Global Construction Aggregates Market Will Reach USD 490 Billion by 2025, Feb 2019, Zion Market Research Report. Zion Market Research, New York, NY, USA, research reports, Suite N202.

Table 1: Physical and chemical characteristics of FA, BT, BL and GGBS

Characteristics	FA	BT	BL	GGBS
Fineness (m^2/kg)	368	-	-	350
Retained on 45 µm sieve (%)	14.62	-	-	
$SiO_2 + Al_2O_3 + Fe_2O_3$ (%)	84.16	81.75	9.3	45.98
MgO (%)	1.20	2.95	4.45	4.43
CaO (%)	6.31	1.80	75.80	44.8
SO_3 (%)	0.57	-	-	2.26
Loss on ignition (%)	1.68	8.10	2.00	1.32

Table 2: Levels of factors considered for the production of FA-BT, FA-BL and FA-GGBS aggregates

Factors	Level 1			Level 2			Level 3		
	FA-BT	FA-BL	FA-GGBS	FA-BT	FA-BL	FA-GGBS	FA-BT	FA-BL	FA-GGBS
I: Na_2O content (%)	3	4	3	4	5	4	5	6	5
II: Water content (%)	19	19	19	20	20	20	21	21	21
III: Admixture content (%)	5	5	5	10	10	10	15	15	15

Table 3: Experimental sets of trial mixes as per L_9 orthogonal array in the production of FA-BT, FA-BL and FA-GGBS aggregates

Na_2O content (%)			Water content (%)			Admixture content (%)		
FA-BT	**FA-BL**	**FA-GGBS**	**FA-BT**	**FA-BL**	**FA-GGBS**	**FA-BT**	**FA-BL**	**FA-GGBS**
3.0	4.0	3.0	19.0	19.0	19.0	5.0	5.0	5.0
3.0	4.0	3.0	20.0	20.0	20.0	10.0	10.0	10.0
3.0	4.0	3.0	21.0	21.0	21.0	15.0	15.0	15.0
4.0	5.0	4.0	19.0	19.0	19.0	10.0	10.0	10.0
4.0	5.0	4.0	20.0	20.0	20.0	15.0	15.0	15.0
4.0	5.0	4.0	21.0	21.0	21.0	5.0	5.0	5.0
5.0	6.0	5.0	19.0	19.0	19.0	15.0	15.0	15.0
5.0	6.0	5.0	20.0	20.0	20.0	5.0	5.0	5.0
5.0	6.0	5.0	21.0	21.0	21.0	10.0	10.0	10.0

Table 4: Grey relational generations for aggregate impact and crushing value, individual crushing strength and water absorption of FA-BT aggregates for 14 days of curing

Trial No.	Grey relational generations			
FABT 1	0.378	0.366	0.032	0.437
FABT 2	0.037	0.025	0.129	0.239
FABT 3	0.000	0.000	0.000	0.000
FABT 4	0.627	0.626	0.226	0.704
FABT 5	0.568	0.584	0.323	0.831
FABT 6	0.402	0.420	0.065	0.592
FABT 7	0.610	0.752	0.742	0.761
FABT 8	0.822	0.744	0.484	0.563
FABT 9	1.000	1.000	1.000	1.000

Table 5: Grey relational generations for aggregate impact and crushing value, individual crushing strength and water absorption of FA-BT aggregates for 28 days of curing

Trial No.	Grey relational generations			
FABT 1	0.376	0.365	0.000	0.357
FABT 2	0.046	0.037	0.043	0.125
FABT 3	0.000	0.000	0.021	0.000
FABT 4	0.629	0.614	0.255	0.696
FABT 5	0.586	0.589	0.213	0.857
FABT 6	0.380	0.378	0.043	0.518
FABT 7	0.679	0.705	0.596	0.304
FABT 8	0.705	0.697	0.426	0.857
FABT 9	1.000	1.000	1.000	1.000

Table 6: Grey relational generations for aggregate impact and crushing value, individual crushing strength and water absorption of FA-BT aggregates for 100 days of curing

Trial No.	Grey relational generations			
FABT 1	0.399	0.508	0.000	0.286
FABT 2	0.082	0.176	0.032	0.129
FABT 3	0.000	0.000	0.063	0.000
FABT 4	0.635	0.619	0.175	0.743
FABT 5	0.609	0.566	0.222	0.757
FABT 6	0.339	0.340	0.111	0.557
FABT 7	0.734	0.721	0.540	0.386
FABT 8	0.815	0.775	0.413	0.814
FABT 9	1.000	1.000	1.000	1.000

Table 7: Δ_{0i} and grey relation coefficients for 14 days of curing with respect to governing factors in FA-BT aggregates

Trial No	Δ_{0i}				Grey relational coefficients			
	Aggregate impact value (%)	Aggregate crushing value (%)	Individual Crushing Strength (MPa)	Water absorption (%)	Aggregate impact value (%)	Aggregate crushing value (%)	Individual Crushing Strength (MPa)	Water absorption (%)
FABT 1	0.622	0.634	0.968	0.563	0.445	0.441	0.341	0.470
FABT 2	0.963	0.975	0.871	0.761	0.342	0.339	0.365	0.397
FABT 3	1.000	1.000	1.000	1.000	0.333	0.333	0.333	0.333
FABT 4	0.373	0.374	0.774	0.296	0.572	0.572	0.392	0.628
FABT 5	0.432	0.416	0.677	0.169	0.537	0.546	0.425	0.747
FABT 6	0.598	0.580	0.935	0.408	0.456	0.463	0.348	0.550
FABT 7	0.390	0.248	0.258	0.239	0.562	0.669	0.660	0.676
FABT 8	0.178	0.256	0.516	0.437	0.737	0.661	0.492	0.534
FABT 9	0.000	0.000	0.000	0.000	1.000	1.000	1.000	1.000

Table 8: Δ_{0i} and grey relation coefficients for 28 days of curing with respect to governing factors in FA-BT aggregates

Trial No	Δ_{0i}				Grey relational coefficients			
	Aggregate impact value (%)	Aggregate crushing value (%)	Individual Crushing Strength (MPa)	Water absorption (%)	Aggregate impact value (%)	Aggregate crushing value (%)	Individual Crushing Strength (MPa)	Water absorption (%)
FABT 1	0.624	0.635	1.000	0.643	0.445	0.441	0.333	0.438
FABT 2	0.954	0.963	0.957	0.875	0.344	0.342	0.343	0.364
FABT 3	1.000	1.000	0.979	1.000	0.333	0.333	0.338	0.333
FABT 4	0.371	0.386	0.745	0.304	0.574	0.564	0.402	0.622
FABT 5	0.414	0.411	0.787	0.143	0.547	0.549	0.388	0.778
FABT 6	0.620	0.622	0.957	0.482	0.446	0.445	0.343	0.509
FABT 7	0.321	0.295	0.404	0.696	0.609	0.629	0.553	0.418
FABT 8	0.295	0.303	0.574	0.143	0.629	0.623	0.465	0.778
FABT 9	0.000	0.000	0.000	0.000	1.000	1.000	1.000	1.000

Table 9: Δ_{0i} and grey relation coefficients for 100 days of curing with respect to governing factors in FA-BT aggregates

Trial No	Δ_{0i}				Grey relational coefficients			
	Aggregate impact value (%)	Aggregate crushing value (%)	Individual Crushing Strength (MPa)	Water absorption (%)	Aggregate impact value (%)	Aggregate crushing value (%)	Individual Crushing Strength (MPa)	Water absorption (%)
FABT 1	0.601	0.492	1.000	0.714	0.454	0.504	0.333	0.412
FABT 2	0.918	0.824	0.968	0.871	0.352	0.378	0.341	0.365
FABT 3	1.000	1.000	0.937	1.000	0.333	0.333	0.348	0.333
FABT 4	0.365	0.381	0.825	0.257	0.578	0.567	0.377	0.660
FABT 5	0.391	0.434	0.778	0.243	0.561	0.535	0.391	0.673
FABT 6	0.661	0.660	0.889	0.443	0.431	0.431	0.360	0.530
FABT 7	0.266	0.279	0.460	0.614	0.653	0.642	0.521	0.449
FABT 8	0.185	0.225	0.587	0.186	0.730	0.689	0.460	0.729
FABT 9	0.000	0.000	0.000	0.000	1.000	1.000	1.000	1.000

Table 10: Grey relational grades for three curing ages with respect to governing factors in FA-BT aggregates

Trial No	Grey relational grade		
	14 Days	28 Days	100 Days
FABT 1	0.424	0.406	0.426
FABT 2	0.361	0.343	0.359
FABT 3	0.333	0.335	0.337
FABT 4	0.541	0.513	0.546
FABT 5	0.564	0.495	0.540
FABT 6	0.454	0.412	0.438
FABT 7	0.642	0.597	0.566
FABT 8	0.606	0.572	0.652
FABT 9	1.000	1.000	1.000

Table 11: Response table for grey relational grade for three curing ages with respect to governing factors in FA-BT aggregates

Factors	Curing Ages	Mean grey Relational Grade			Maximum value – minimum value	Rank
		Level 1	Level 2	Level 3		
Na_2O content (%)	14 days	0.373	0.520	0.749	0.377	1
Water content (%)		0.536	0.510	0.596	0.086	3
BT (%)		0.495	0.634	0.513	0.139	2
Na_2O content (%)	28 days	0.361	0.473	0.723	0.362	1
Water content (%)		0.506	0.470	0.582	0.112	3
BT (%)		0.463	0.619	0.476	0.155	2
Na_2O content (%)	100 days	0.374	0.508	0.739	0.366	1
Water content (%)		0.513	0.517	0.592	0.079	3
BT (%)		0.505	0.635	0.481	0.154	2

Table 12: Response table for grey relational grade for three curing ages with respect to governing factors in FA-BL aggregates

Factors	Curing Ages	Mean grey Relational Grade			Maximum value – minimum value	Rank
		Level 1	Level 2	Level 3		
Na_2O content (%)	14 days	0.394	0.829	0.524	0.435	1
Water content (%)		0.493	0.627	0.628	0.135	2
BL (%)		0.631	0.543	0.573	0.088	3
Na_2O content (%)	28 days	0.387	0.839	0.522	0.452	1
Water content (%)		0.502	0.635	0.610	0.133	2
BL (%)		0.606	0.546	0.596	0.060	3
Na_2O content (%)	100 days	0.390	0.838	0.507	0.448	1
Water content (%)		0.507	0.642	0.586	0.136	2
BL (%)		0.582	0.541	0.612	0.071	3

Table 13: Response table for grey relational grade for three curing ages with respect to governing factors in FA-GGBS aggregates

Factors	Curing Ages	Mean grey Relational Grade			Maximum value – minimum value	Rank
		Level 1	Level 2	Level 3		
Na_2O content (%)	14 days	0.406	0.579	0.820	0.414	1
Water content (%)		0.639	0.581	0.585	0.059	3
GGBS (%)		0.600	0.570	0.634	0.064	2
Na_2O content (%)	28 days	0.404	0.586	0.807	0.404	1
Water content (%)		0.650	0.568	0.579	0.082	3
GGBS (%)		0.595	0.554	0.647	0.093	2
Na_2O content (%)	100 days	0.359	0.548	0.765	0.407	1
Water content (%)		0.623	0.518	0.531	0.106	3
GGBS (%)		0.534	0.513	0.624	0.111	2

Appendix A: Grey relational generations for FA-BL and FA-GGBB aggregates

Table A1: Grey relational generations for aggregate impact and crushing value, individual crushing strength and water absorption of FA-BL aggregates for 14 days of curing

Trial No.	Grey relational generations			
FABL 1	0.000	0.000	0.000	0.370
FABL 2	0.355	0.177	0.603	0.222
FABL 3	0.336	0.192	0.175	0.000
FABL 4	0.767	0.690	0.635	0.815
FABL 5	1.000	0.882	0.762	1.000
FABL 6	0.973	1.000	1.000	0.951
FABL 7	0.389	0.236	0.143	0.728
FABL 8	0.653	0.483	0.302	0.852
FABL 9	0.496	0.330	0.317	0.877

Table A2: Grey relational generations for aggregate impact and crushing value, individual crushing strength and water absorption of FA-BL aggregates for 28 days of curing

Trial No.	Grey relational generations			
FABL 1	0.000	0.000	0.000	0.307
FABL 2	0.330	0.146	0.505	0.216
FABL 3	0.374	0.196	0.172	0.000
FABL 4	0.793	0.744	0.699	0.807
FABL 5	1.000	0.955	1.000	0.875
FABL 6	0.993	1.000	0.731	1.000
FABL 7	0.378	0.347	0.204	0.705
FABL 8	0.626	0.492	0.366	0.795
FABL 9	0.522	0.352	0.409	0.830

Table A3: Grey relational generations for aggregate impact and crushing value, individual crushing strength and water absorption of FA-BL aggregates for 100 days of curing

Trial No.	Grey relational generations			
FABL 1	0.000	0.000	0.000	0.291
FABL 2	0.350	0.390	0.459	0.038
FABL 3	0.315	0.341	0.153	0.000
FABL 4	0.797	0.756	0.663	0.848
FABL 5	0.969	0.961	1.000	1.000
FABL 6	1.000	1.000	0.653	0.911
FABL 7	0.385	0.439	0.133	0.709
FABL 8	0.664	0.488	0.378	0.658
FABL 9	0.524	0.390	0.378	0.696

Table A4: Grey relational generations for aggregate impact and crushing value, individual crushing strength and water absorption of FA-GGBS aggregates for 14 days of curing

Trial No.	Grey relational generations			
FAGGBS 1	0.000	0.000	0.000	0.284
FAGGBS 2	0.301	0.237	0.030	0.162
FAGGBS 3	0.213	0.128	0.000	0.000
FAGGBS 4	0.728	0.699	0.152	0.432
FAGGBS 5	0.682	0.689	0.182	0.297
FAGGBS 6	0.812	0.836	0.273	0.554
FAGGBS 7	1.000	1.000	0.909	1.000
FAGGBS 8	0.728	0.790	1.000	0.676
FAGGBS 9	0.728	0.767	0.758	0.838

Table A5: Grey relational generations for aggregate impact and crushing value, individual crushing strength and water absorption of FA-GGBS aggregates for 28 days of curing

Trial No.	Grey relational generations			
FAGGBS 1	0.000	0.000	0.025	0.261
FAGGBS 2	0.252	0.219	0.075	0.159
FAGGBS 3	0.165	0.143	0.000	0.000
FAGGBS 4	0.704	0.683	0.200	0.391
FAGGBS 5	0.696	0.665	0.250	0.319
FAGGBS 6	0.857	0.853	0.400	0.551
FAGGBS 7	1.000	1.000	1.000	1.000
FAGGBS 8	0.709	0.759	0.900	0.739
FAGGBS 9	0.704	0.679	0.675	0.841

Table A6: Grey relational generations for aggregate impact and crushing value, individual crushing strength and water absorption of FA-GGBS aggregates for 100 days of curing

Trial No.	Grey relational generations			
FAGGBS 1	0.000	0.000	0.000	0.188
FAGGBS 2	0.265	0.250	0.037	0.116
FAGGBS 3	0.139	0.167	0.019	0.000
FAGGBS 4	0.700	0.697	0.204	0.420
FAGGBS 5	0.704	0.684	0.333	0.275
FAGGBS 6	0.839	0.816	0.370	0.449
FAGGBS 7	1.000	1.000	1.000	1.000
FAGGBS 8	0.748	0.741	0.833	0.594
FAGGBS 9	0.713	0.697	0.667	0.783

Appendix B: Grey relational coefficients for FA-BL and FA-GGBS aggregates

Table B1: Δ_{0i} and grey relation coefficients for 14 days of curing with respect to governing factors in FA-BL aggregates

Trial No	Δ_{0i}				Grey relational coefficients			
	Aggregate impact value (%)	Aggregate crushing value (%)	Individual Crushing Strength (MPa)	Water absorption (%)	Aggregate impact value (%)	Aggregate crushing value (%)	Individual Crushing Strength (MPa)	Water absorption (%)
FABL 1	1.000	1.000	1.000	0.630	0.333	0.333	0.333	0.443
FABL 2	0.645	0.823	0.397	0.778	0.437	0.378	0.558	0.391
FABL 3	0.664	0.808	0.825	1.000	0.430	0.382	0.377	0.333
FABL 4	0.233	0.310	0.365	0.185	0.682	0.617	0.578	0.730
FABL 5	0.000	0.118	0.238	0.000	1.000	0.809	0.677	1.000
FABL 6	0.027	0.000	0.000	0.049	0.949	1.000	1.000	0.910
FABL 7	0.611	0.764	0.857	0.272	0.450	0.396	0.368	0.648
FABL 8	0.347	0.517	0.698	0.148	0.590	0.492	0.417	0.771
FABL 9	0.504	0.670	0.683	0.123	0.498	0.427	0.423	0.802

Table B2: Δ_{0i} and grey relation coefficients for 28 days of curing with respect to governing factors in FA-BL aggregates

Trial No	Δ_{0i}				Grey relational coefficients			
	Aggregate impact value (%)	Aggregate crushing value (%)	Individual Crushing Strength (MPa)	Water absorption (%)	Aggregate impact value (%)	Aggregate crushing value (%)	Individual Crushing Strength (MPa)	Water absorption (%)
FABL 1	1.000	1.000	1.000	0.693	0.333	0.333	0.333	0.419
FABL 2	0.670	0.854	0.495	0.784	0.427	0.369	0.503	0.389
FABL 3	0.626	0.804	0.828	1.000	0.444	0.383	0.377	0.333
FABL 4	0.207	0.256	0.301	0.193	0.707	0.661	0.624	0.721
FABL 5	0.000	0.045	0.000	0.125	1.000	0.917	1.000	0.800
FABL 6	0.007	0.000	0.269	0.000	0.985	1.000	0.650	1.000
FABL 7	0.622	0.653	0.796	0.295	0.446	0.434	0.386	0.629
FABL 8	0.374	0.508	0.634	0.205	0.572	0.496	0.441	0.710
FABL 9	0.478	0.648	0.591	0.170	0.511	0.435	0.458	0.746

Table B3: Δ_{0i} and grey relation coefficients for 100 days of curing with respect to governing factors in FA-BL aggregates

Trial No	Δ_{0i}				Grey relational coefficients			
	Aggregate impact value (%)	Aggregate crushing value (%)	Individual Crushing Strength (MPa)	Water absorption (%)	Aggregate impact value (%)	Aggregate crushing value (%)	Individual Crushing Strength (MPa)	Water absorption (%)
FABL 1	1.000	1.000	1.000	0.709	0.333	0.333	0.333	0.414
FABL 2	0.650	0.610	0.541	0.962	0.435	0.451	0.480	0.342
FABL 3	0.685	0.659	0.847	1.000	0.422	0.432	0.371	0.333
FABL 4	0.203	0.244	0.337	0.152	0.711	0.672	0.598	0.767
FABL 5	0.031	0.039	0.000	0.000	0.941	0.928	1.000	1.000
FABL 6	0.000`	0.000	0.347	0.089	1.000	1.000	0.590	0.849
FABL 7	0.615	0.561	0.867	0.291	0.448	0.471	0.366	0.632
FABL 8	0.336	0.512	0.622	0.342	0.598	0.494	0.445	0.594
FABL 9	0.476	0.610	0.622	0.304	0.513	0.451	0.445	0.622

Table B4: Δ_{0i} and grey relation coefficients for 14 days of curing with respect to governing factors in FA-GGBS aggregates

Trial No	Δ_{0i}				Grey relational coefficients			
	Aggregate impact value (%)	Aggregate crushing value (%)	Individual Crushing Strength (MPa)	Water absorption (%)	Aggregate impact value (%)	Aggregate crushing value (%)	Individual Crushing Strength (MPa)	Water absorption (%)
FAGGBS 1	1.000	1.000	1.000	0.716	0.500	0.333	0.333	0.411
FAGGBS 2	0.699	0.763	0.970	0.838	0.589	0.396	0.340	0.374
FAGGBS 3	0.787	0.872	1.000	1.000	0.560	0.364	0.333	0.333
FAGGBS 4	0.272	0.301	0.848	0.568	0.786	0.624	0.371	0.468
FAGGBS 5	0.318	0.311	0.818	0.703	0.759	0.617	0.379	0.416
FAGGBS 6	0.188	0.164	0.727	0.446	0.842	0.753	0.407	0.529
FAGGBS 7	0.000	0.000	0.091	0.000	1.000	1.000	0.846	1.000
FAGGBS 8	0.272	0.210	0.000	0.324	0.786	0.704	1.000	0.607
FAGGBS 9	0.272	0.233	0.242	0.162	0.786	0.682	0.673	0.755

Table B5: Δ_{0i} and grey relation coefficients for 28 days of curing with respect to governing factors in FA-GGBS aggregates

Trial No	Δ_{0i}				Grey relational coefficients			
	Aggregate impact value (%)	Aggregate crushing value (%)	Individual Crushing Strength (MPa)	Water absorption (%)	Aggregate impact value (%)	Aggregate crushing value (%)	Individual Crushing Strength (MPa)	Water absorption (%)
FAGGBS 1	1.000	1.000	0.975	0.739	0.500	0.333	0.339	0.404
FAGGBS 2	0.748	0.781	0.925	0.841	0.572	0.390	0.351	0.373
FAGGBS 3	0.835	0.857	1.000	1.000	0.545	0.368	0.333	0.333
FAGGBS 4	0.296	0.317	0.800	0.609	0.772	0.612	0.385	0.451
FAGGBS 5	0.304	0.335	0.750	0.681	0.767	0.599	0.400	0.423
FAGGBS 6	0.143	0.147	0.600	0.449	0.875	0.772	0.455	0.527
FAGGBS 7	0.000	0.000	0.000	0.000	1.000	1.000	1.000	1.000
FAGGBS 8	0.291	0.241	0.100	0.261	0.774	0.675	0.833	0.657
FAGGBS 9	0.296	0.321	0.325	0.159	0.772	0.609	0.606	0.758

Table B6: Δ_{0i} and grey relation coefficients for 100 days of curing with respect to governing factors in FA-GGBS aggregates

Trial No	Δ_{0i}				Grey relational coefficients			
	Aggregate impact value (%)	Aggregate crushing value (%)	Individual Crushing Strength (MPa)	Water absorption (%)	Aggregate impact value (%)	Aggregate crushing value (%)	Individual Crushing Strength (MPa)	Water absorption (%)
FAGGBS 1	1.000	1.000	1.000	0.812	0.333	0.333	0.333	0.381
FAGGBS 2	0.735	0.750	0.963	0.884	0.405	0.400	0.342	0.361
FAGGBS 3	0.861	0.833	0.981	1.000	0.367	0.375	0.338	0.333
FAGGBS 4	0.300	0.303	0.796	0.580	0.625	0.623	0.386	0.463
FAGGBS 5	0.296	0.316	0.667	0.725	0.628	0.613	0.429	0.408
FAGGBS 6	0.161	0.184	0.630	0.551	0.757	0.731	0.443	0.476
FAGGBS 7	0.000	0.000	0.000	0.000	1.000	1.000	1.000	1.000
FAGGBS 8	0.252	0.259	0.167	0.406	0.665	0.659	0.750	0.552
FAGGBS 9	0.287	0.303	0.333	0.217	0.635	0.623	0.600	0.697

Appendix C: Grey relational grade for FA-BL and FA-GGBS aggregates

Table C1: Grey relational grades for three curing ages with respect to governing factors in FA-BL aggregates

Trial No	Grey relational grade		
	14 Days	**28 Days**	**100 Days**
FABL 1	0.361	0.355	0.353
FABL 2	0.441	0.422	0.427
FABL 3	0.381	0.384	0.389
FABL 4	0.652	0.678	0.687
FABL 5	0.872	0.929	0.967
FABL 6	0.965	0.909	0.860
FABL 7	0.466	0.473	0.479
FABL 8	0.568	0.555	0.533
FABL 9	0.538	0.538	0.508

Table C2: Grey relational grades for three curing ages with respect to governing factors in FA-GGBS aggregates

Trial No	Grey relational grade		
	14 Days	28 Days	100 Days
FAGGBS 1	0.394	0.394	0.345
FAGGBS 2	0.425	0.422	0.377
FAGGBS 3	0.398	0.395	0.353
FAGGBS 4	0.562	0.555	0.524
FAGGBS 5	0.543	0.547	0.520
FAGGBS 6	0.633	0.657	0.601
FAGGBS 7	0.962	1.000	1.000
FAGGBS 8	0.774	0.735	0.656
FAGGBS 9	0.724	0.686	0.639