

1 **Moisture damage evaluation of aggregate-bitumen bonds with the respect**  
2 **of moisture absorption, tensile strength and failure surface**

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1 **Abstract:** The moisture-induced deterioration of asphalt mixture is because of the loss of  
2 adhesion at the aggregate-bitumen interface and/or the loss of cohesion within the bitumen  
3 film. An experimental study was undertaken in this paper to characterise the effects of  
4 moisture on the direct tensile strength of aggregate-bitumen bonds. The aim of this paper was  
5 to evaluate the moisture sensitivity of aggregate-bitumen bonds in several different aspects,  
6 which included moisture absorption, tensile strength and failure surface examination.  
7 Moisture absorption and mineralogical compositions of aggregate were measured using  
8 gravimetric techniques and a Mineral Liberation Analyser (MLA), respectively, with the  
9 results being used to explain the moisture sensitivity of aggregate-bitumen bonds. Aggregate-  
10 bitumen bond strength was determined using a self-designed pull-off system with the  
11 capability of accurately controlling the bitumen film thickness. The photographs of the failure  
12 surfaces were quantitatively analysed using Image-J software. The results show that the  
13 magnitude of the aggregate-bitumen bonding strength in the dry condition is mainly  
14 controlled by bitumen. However, the retained tensile strength after moisture conditioning was  
15 found to be influenced by the mineralogical composition as well as the moisture diffusion  
16 properties of the aggregates. The linear relationship between retained tensile strength and the  
17 square root of moisture uptake suggests that the water absorption process controls the  
18 degradation of the aggregate-bitumen bond. The results also suggested that the deterioration  
19 of aggregate-bitumen bonds is linked to the decrease of cohesive failure percentage.

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21 **Keywords:** pull-off system, aggregate-bitumen bonds, moisture damage, adhesion, cohesion

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## 1 **1. Introduction**

2

3 Asphalt mixtures are widely used as pavement construction materials. During their service  
4 life, asphalt pavements have to sustain harsh traffic loads and environmental conditions and  
5 deteriorate with the passage of time. The effect of moisture on asphalt mixtures is recognised  
6 as a major cause of pavement failure. The penetration of moisture through asphalt mixtures  
7 can increase the pavements vulnerability to traffic loading and thermal stress (Kim, Little &  
8 Lytton, 2004, Mehrara & Khodaii 2013). Moisture damage in asphalt pavement is defined as  
9 the loss of strength, stiffness and durability because of the presence of moisture resulting in  
10 adhesive failure at the aggregate-bitumen interface and/or cohesive failure within the bitumen  
11 or mastic (Airey, Collop, Zoorob & Elliott, 2008). With the presence of moisture, water may  
12 enter the aggregate-bitumen interface by diffusion through bitumen films, seepage into the  
13 film through micro voids or cracks, and through direct access in partially coated aggregates  
14 (Stuart, 1990). It is noticeable that the existence of moisture may only weaken the asphalt  
15 mixture by emulsifying or softening the bitumen film but without removing it from aggregate  
16 surfaces. Also, when the moisture is removed from the asphalt mixture, the stiffness loss is  
17 reversible. However, when the pavement is loaded during the weakened condition, the  
18 moisture damage is accelerated and may become irreversible (Santucci, 2002). Although not  
19 all damage is caused directly by moisture, its presence increases the extent and severity of  
20 already existing distresses like cracking, potholes and rutting (Grenfell et al., 2014).

21

22 According to previous researchers (Kakar, Hamzah & Valentin, 2015, Caro, Masad, Bhasin  
23 & Little, 2008), the moisture in either a liquid or vapour state infiltrates the asphalt mixture as  
24 well as the bitumen film or mastic and reaches the aggregate-bitumen interface so as to  
25 change the internal structure and finally results in the degradation of mechanical properties of

1 the material. In addition, the moisture may also invade the asphalt mixture system by seeping  
2 through the already existing cracks in the mixture or by diffusing outward from the aggregate  
3 pores. Once moisture has come into contact and interacted with the asphalt mixture, the  
4 moisture damage could be developed in the following mechanisms: detachment,  
5 displacement, spontaneous emulsification, pore pressure, and hydraulic scour (Grenfell et al.,  
6 2014). It should be mentioned that the moisture damage is not limited to only one mechanism  
7 but is the result of a combination of several mechanisms.

8

9 The resistance of asphalt mixtures to moisture attack has been related to aggregate  
10 mineralogy, surface texture of aggregate, bitumen chemistry and the compatibility between  
11 bitumen and aggregate (Terrel & Al-Swailmi, 1994, Abo-Qudais & Al-Shweily, 2007).  
12 However, it has been suggested that in a susceptible mixture, the effect of aggregate is more  
13 influential than the effect of mastic (Apeagyei, Grenfell & Airey, 2015). In addition, factors  
14 such as permeability of the asphalt mixtures, volumetric properties of binder and the ambient  
15 conditions are all important when considering the susceptibility of asphalt mixture (Grenfell,  
16 Ahmad, Airey, Collop & Elliott, 2012). For susceptible asphalt mixtures, the failure surfaces  
17 tend to transform from cohesive to adhesive after moisture damage. So, the adhesive strength  
18 of the aggregate-bitumen interface and its sensitivity to moisture attack are considered to be  
19 vital parameters in moisture damage evaluation.

20

21 By measuring the adhesive bond strength of coatings between bitumen and aggregate, several  
22 testing techniques have been developed but the most commonly used methods include the  
23 pull-off test and peel test. Normally, the pull-off test is conducted by measuring the tensile  
24 stress necessary to detach the adhesive materials in a direction perpendicular to the substrates  
25 (Harvey & Cebon, 2003, Apeagyei, Grenfell & Airey, 2014). In terms of the peel test, a thin

1 flexible peel arm and a rigid substrate are bonded using the adhesive material. During testing,  
2 the peel arm is pulled from the substrate at a specified angle and speed while the peel force is  
3 recorded. The recorded peel force in steady state conditions is then used to calculate the  
4 fracture energy of the adhesive (Horgnies, Darque-Ceretti, Ferzai & Felder, 2011, Blackman,  
5 Cui, Kinloch & Taylor, 2013). These two methods have been successfully used to evaluate  
6 the moisture sensitivity of the aggregate-bitumen bond by immersing specimens in water for  
7 a range of times before testing. However, the limitations of these established tests are very  
8 obvious. First of all, the mechanical evaluation only reflects the influence of conditioning  
9 time but not the amount of moisture absorbed. Secondly, manual control of bitumen film  
10 thickness makes it hard to obtain the required thickness and results in big deviations in  
11 measured strength. Thirdly, these studies were limited in their ability to control bitumen film  
12 thickness to submicron level and hence cannot simulate the real bitumen thickness in  
13 mixtures. For a better understanding of the performance of the aggregate-bitumen interface  
14 when exposed to moisture, this paper presents the development of a suitable procedure  
15 consisting of innovative sample preparation, controlled moisture conditioning, pull-off test  
16 set-up and failure surface evaluation.

17

## 18 **2. Materials**

19

20 Two base bitumens named B1 and B2 with penetration grades of 40/60 pen and 70/100 pen,  
21 respectively, were selected. These two binders were from the same crude source and  
22 therefore had similar chemical compositions (Zhang, Airey & Grenfell, 2015a). The  
23 fundamental physical properties of the bitumen were measured using softening point (ASTM  
24 D36) and penetration (ASTM D5) tests with the results shown in Table 1.

25

1 Three types of aggregate from different quarries were selected as substrates. They included  
2 one limestone aggregate (L1) and two granite aggregates (G1 and G2). These aggregates are  
3 known to have different moisture sensitivity due to their moisture absorption and  
4 mineralogical composition. Based on their mineral compositions, the two granite aggregates  
5 G1 and G2 can be classified as acidic and the L1 is defined as basic.

6

### 7 **3. Methodology**

8

9 The aim of this research is to characterise the moisture deterioration of aggregate-bitumen  
10 bond through different aspects, such as moisture absorption, bonding strength and failure  
11 surface, using a new pull-off test. As the performance of the aggregate-bitumen combined  
12 specimen is dominated by the physical and chemical properties of the original materials, the  
13 fundamental properties of the aggregates and bitumen were first analysed. Then, the  
14 deteriorations of the aggregate-bitumen bonds under moisture attack have been evaluated  
15 using a new pull-off test. The pull-off test set-up consists of three main parts: accurate control  
16 of the bitumen film thickness using self-designed DSR fixtures, a moisture conditioning step  
17 which can allow the moisture to diffuse into aggregate-bitumen interface and a direct tension  
18 test with accurate control of loading rate and testing temperature.

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#### 20 3.1 Mineral Liberation Analyser (MLA) test

21

22 It has been accepted that the mineralogical compositions of aggregates have a profound  
23 influence on moisture sensitivity of asphalt mixtures. By measuring the mineralogical  
24 properties of aggregates, an MLA device was used in this research. The MLA is an  
25 automated mineral analysis system that can identify minerals in polished sections of drill

1 cores, particulates or lump materials, and quantify a wide range of mineral characteristics,  
2 such as mineral abundance, grain size and liberation. Before testing, the aggregate needs to  
3 be polished and carbon coated to get an electron conductive surface. During testing, the  
4 Back-scattered Electron (BSE) image is combined with Electron Dispersive X-ray (EDX)  
5 analysis for the specimen surface for a series of frames step by step. Finally, the MLA's data-  
6 view software allows presenting the digital results in a graphical format. A detailed  
7 introduction about the testing procedure can be seen in previous publications (Grenfell et al.,  
8 2012, Zhang, Apeageyi, Airey & Grenfell, 2015b)

9

### 10 3.2 Moisture absorption of aggregates

11

12 An important factor which affects the moisture-induced deterioration of asphalt mixtures is  
13 the speed and amount of moisture absorbed by the aggregates. Therefore, a robust procedure  
14 was developed to characterise the moisture absorption and moisture diffusion properties of  
15 the aggregates during laboratory conditioning. This approach is different from most previous  
16 studies that only consider conditioning time when evaluating the moisture damage. To  
17 measure moisture absorption, big aggregate boulders were first trimmed into rectangular  
18 beams with the dimensions of 100 mm × 20 mm × 10 mm. It should be noted that any  
19 regularly shaped aggregate specimens can be used. After cleaning and drying the beams, the  
20 weight of each in the dry condition was measured using a sensitive balance with a resolution  
21 of 0.1 µg. Then, the aggregates were moisture conditioned by totally immersing them in  
22 deionised water at 20°C and measuring moisture uptake periodically. The results were used  
23 to calculate the mass of water absorbed by aggregate as a percentage of the dry aggregate  
24 weight (Eq.1).

25

1  $Mass\ uptake\ (\%) = M_t = \frac{w_t - w_0}{w_0}$  (1)

2

3 where  $M_t$  is the moisture uptake at time  $t$ ,  $w_0$  is the initial mass of the aggregate in the dry  
4 condition,  $w_t$  is the mass of aggregate after time  $t$ .

5

### 6 3.3 Dynamic Shear Rheometer (DSR) test

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8 The DSR was adopted to characterise the visco-elastic behaviour of bitumen in the  
9 temperature range from 10°C to 80°C. Before the frequency sweep test, a strain sweep test  
10 needs to be done so as to define the linear visco-elastic region (LVE) at each temperature.  
11 Based on the strain sweep tests, the strain levels were defined before the frequency sweep  
12 tests. Table 2 shows the testing conditions at different temperatures for the frequency sweep  
13 tests.

14

### 15 3.4 Pull-off test

16

17 The motivation for developing the pull-off test is the lack robust yet simple and reliable test  
18 with the capability to precisely control loading rate. Currently, the most common pull-off test  
19 (PATTI) is limited in the sense that the stress rate cannot be controlled. The innovation of  
20 this test is the ability to accurately determine bitumen film thickness using a modified  
21 dynamic shear rheometer, the small aggregate substrate size that permits realistic moisture  
22 conditioning and the simplified custom-made direct tension fixtures that can be easily  
23 mounted on a Universal Testing Machine (UTM).

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#### 25 3.4.1 Aggregate-bitumen specimen preparation

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To prepare the aggregate-bitumen adhesion specimen, the cylindrical aggregate substrates need to be prepared first. Samples of boulder sized aggregates were drilled using an electrically operated water-cooled core drill to get aggregate cylinders with 25 mm diameter. A trimming saw was then used to cut the aggregate cylinders into discs with 5 mm thickness. The top and bottom surfaces of the discs were polished using a rotary polishing machine, to remove all blemishes left by the sawing process and get parallel surfaces to ensure complete adhesion between aggregate and bitumen. All discs were cleaned in an ultrasonic cleaning machine for 15 minutes and dried in an oven at a temperature of 40°C for 24 hours. Two aluminium holding plates were specially designed (diameter and thickness) and fabricated to fit in a standard Bohlin Gemini DSR. With a view to precisely control the bitumen film thickness, the two holding plates were designed to clamp the discs and then fixed into the DSR machine. After establishing the zero gap and ensuring that the discs are parallel, a small amount of hot bitumen was placed on the lower aggregate surface and then pressed with the upper aggregate disc to achieve the required bitumen film thickness of 20 µm, with a gap resolution of 1 µm. The sample was removed from the DSR after about 15 minutes of cooling and then the excess bitumen removed by means of a heated pallet knife. Figure 1 shows the whole procedure of the sample preparation.

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### 3.4.2 Moisture conditioning of adhesion specimen

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To evaluate the deterioration of the aggregate-bitumen interface after moisture damage, the prepared aggregate-bitumen adhesion specimens were immersed in distilled water to simulate the moisture damage process. Moisture conditioning was performed by storing specimens in water (24 hours and 168 hours) with the temperature maintained at 20°C. The schematic

1 diagram of the moisture conditioning is shown in Figure 2. During the moisture conditioning,  
2 moisture could reach the aggregate-bitumen interface in three different ways: through the top  
3 and bottom aggregate, through the edge of aggregate-bitumen interface and through the  
4 bitumen film.

5

### 6 3.4.3 Bond strength evaluation using pull-off test

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8 The aggregate-bitumen interfacial bond strength in the dry condition and after periods of  
9 moisture conditioning (24 hours and 168 hours) were determined by using a pull-off tensile  
10 test with detailed procedures shown in Figure 3. Before the pull-off test, the specimen was  
11 first fixed by two direct tension fixtures with three screws on each. These two fixtures  
12 combined with the aggregate-bitumen specimen were then mounted on the UTM. An  
13 extension speed of 10 mm/min and a temperature of 20°C were applied to break the interface.  
14 However, depending on the equipment limitations, any available loading rate can be used.  
15 During the test, the pull force as a function of elongation was recorded and the failure  
16 surfaces of each broken sample were photographed with a digital camera to characterise the  
17 loci of failure as either adhesive or cohesive. At least four repeat tests were made for each  
18 aggregate-bitumen combination. The results were used to calculate the tensile strength.  
19 Tensile strength TS (kPa) was computed as the ratio of the peak load divided by the cross-  
20 sectional area of the bitumen film as follows:

21

$$22 \quad TS = \frac{F}{1000 \times \pi \times r^2} \quad (3)$$

23

24 where  $F$  is the peak tensile force (N) and  $r$  is the radius of the aggregate substrate (0.0125m).

25

## 1 4. Results

2

### 3 4.1 Mineralogy of aggregates

4

5 Figure 4 shows the mineralogical distribution on aggregate surfaces obtained from the MLA  
6 with the mineralogical composition shown in Table 3. As shown in Figure 4, the mineral  
7 distribution of G1 and G2 is much more complex than that of L1. The two granite aggregates  
8 G1 and G2 were made up of a large number of different mineral phases, while there are very  
9 few mineral phases in limestone L1. As shown in Table 3, chlorite and albite are the foremost  
10 minerals in G1 with quantities of 31.53% and 27.13% by weight, followed by quartz, epidote  
11 and K-feldspar, which account for 19.11%, 11.11% and 4.82%, respectively. Albite and  
12 anorthite are the predominant minerals in G2, which account for 32.73% and 18.54% by  
13 weight, but quartz and chlorite also have significant quantities. In terms of limestone L1,  
14 calcite is the dominant phase with 96.98% by weight.

15

### 16 4.2 Moisture absorption of aggregates

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18 Water absorption data were obtained from these three aggregates used for substrates in this  
19 research. In order to know how much water was diffused into the aggregate during the  
20 immersion time, a water absorption test was performed and the results are shown in Figure 5.  
21 As shown in this figure, more than 80% of the moisture was absorbed during the first 24  
22 hours of conditioning. After that, the water uptake of L1 and G2 experienced a slow growth  
23 and finally reached slightly over 0.5% although the water absorption of L1 and G2 still seems  
24 to be increasing and has probably not reached equilibrium. G1 showed the lowest water  
25 uptake with the result being only 0.13% after 600 hours conditioning. The obvious difference

1 in terms of the moisture absorption could be attributed to the mineralogical composition and  
2 the structural arrangement of the aggregates.

3

#### 4 4.3 Rheological properties of bitumen

5

6 It has been suggested that bitumen is the only agent that binds aggregates together in asphalt  
7 mixtures and its properties directly affect the performance of asphalt pavements. So,  
8 rheological measurements were conducted at eight temperatures from 10°C to 80°C in the  
9 frequency range of 0.1-10 Hz. Figure 6 shows the shear complex modulus of the two  
10 bitumens used in this research, and the values increase with increased frequency. It was found  
11 that bitumen B1 and B2 exhibit similar complex modulus values at 10°C. As the temperature  
12 increases above 20°C, the B1 bitumen shows higher complex modulus results in comparison  
13 with the B2 bitumen. Bitumen showing higher complex modulus is likely to form a stiffer  
14 bond to resist the direct tensile forces. Based on the penetration classification of the binders  
15 B1 (40/60) and B2 (70/100), the results are as expected and reliable.

16

#### 17 4.4 Aggregate-bitumen bond strength

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19 To measure the effect of moisture on the mechanical performance of different aggregate-  
20 bitumen combinations, the direct tensile strength test was conducted at 20°C with an  
21 extension speed of 10 mm/min. For each aggregate-bitumen combination, four replicate tests  
22 were performed in the dry condition and after moisture conditioning for 24 hours and 168  
23 hours. The results were evaluated by considering factors such as bitumen type, aggregate  
24 type, moisture conditioning, loading behaviour, retained strength and failure surface.

25

1 4.4.1 Effect of moisture conditioning on stress-strain behaviour

2

3 To simulate the effect of moisture on the stress-strain properties of the aggregate-bitumen  
4 bonds, the pull-off tests were performed by continuously recording the tensile load and the  
5 displacement of cross-head. Figure 7 shows the development of the stress-strain behaviour  
6 with the respect to the moisture conditioning time. The tensile loads of all specimens  
7 experienced a decrease after moisture conditioning with the B1-G2 combination obtaining the  
8 biggest decline. The stress-strain curve of B1-L1 and B1-G2 after 24 hours moisture  
9 conditioning experienced a sharp decline once the peak load was reached, showing totally  
10 different behaviour from other specimens. This could be because the short-term moisture  
11 conditioning makes the bitumen harder so that it has no chance to release during the loading  
12 process. Due to the lower moisture absorption of G1 aggregate as shown in Figure 5, it is  
13 difficult for moisture to reach the aggregate-bitumen interface in such a short period of time  
14 so that the sharp drop of the tensile load does not appear in the B1-G1 combination. After 168  
15 hours of conditioning, the peak load for B1-G2 decreased from about 900 N to less than 100  
16 N demonstrating its poor resistance to moisture attack. In contrast, B1-L1 and B1-G1  
17 experienced much less decrease of peak loads meaning better moisture resistance.

18

19 4.4.2 Effect of moisture conditioning on aggregate-bitumen bond strength

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21 The tensile strength of each aggregate-bitumen bond in the dry condition and after moisture  
22 damage was calculated based on Equation 3 with the average values and standard deviation  
23 for all specimens depicted in Table 2. From this table it can be seen that specimens prepared  
24 with the same bitumen tend to yield similar tensile strength in the dry condition, no matter  
25 which aggregate substrate was used. This could be attributed to the cohesive failure surface

1 observed in the dry condition. In terms of the same aggregate substrate in the dry condition,  
2 samples prepared with B1 bitumen exhibited higher tensile strength than those with B2  
3 bitumen. This phenomenon correlates well with the bitumen fundamental properties where  
4 the softening point and complex modulus are higher for B1 than B2. So, it can be concluded  
5 that, in dry conditions, the tensile strength of the aggregate-bitumen bond is dominated by the  
6 bitumen rather than aggregate.

7

8 After moisture conditioning, the tensile strengths experienced a steady decrease with G2  
9 aggregate showing the most significant reduction. The difference in moisture sensitivity of  
10 the different aggregates could be attributed to several factors. It is clear that these three  
11 aggregates have different water absorption and mineralogical composition. With higher water  
12 absorption meaning more air voids in the aggregate and therefore probable shorter time it  
13 takes to allow the moisture to be transported to the aggregate-bitumen interface. In addition,  
14 the dominant mineral in L1 (calcite) is considered moisture resistant, but there are some  
15 moisture sensitive minerals in G1 and G2, such as albite, quartz and K-feldspar. So, when  
16 explaining the moisture damage of different samples, parameters including moisture  
17 absorption and mineralogical composition should be considered together.

18

19 Retained strength, the ratio of bond strength after a given level of moisture conditioning to  
20 the dry bond strength, is considered to be a common parameter to measure the moisture  
21 sensitivity of asphalt mixtures. High retained tensile strength demonstrating better moisture  
22 resistance of the specimen. Figure 8 shows the effect of conditioning time on retained tensile  
23 strength of different aggregate-bitumen bonds. By using the pull-off test results, it is possible  
24 to identify 'good' and 'bad' mixtures. It can be seen that samples prepared with L1 and G1  
25 show good moisture resistance with over 70% tensile strength retained after 168 hours

1 conditioning. However, samples prepared with G2 are more sensitive to moisture attack as  
2 the tensile strength decreased by over 80% and 40% for B1 and B2 bitumen, respectively.  
3 The aggregates L1 and G2 have similar moisture absorption, meaning similar time will be  
4 taken to transport moisture to aggregate-bitumen interface, but they show a significant  
5 difference in retained strength. This is because the bonds formed between bitumen and G2  
6 are quickly degraded once in contact with moisture due to the large amount of albite and  
7 quartz. However, calcite, being the dominant mineral in L1, can form water insoluble bonds  
8 with bitumen that retain better moisture resistance. In terms of G1, due to its lower water  
9 absorption, it will take a much longer time for water to reach the aggregate-bitumen interface.  
10 On this basis, there is limited chance for water to attack the bonds even though G1 contains  
11 several moisture sensitive minerals. The difference in retained strengths between G1 and G2  
12 could be attributed to higher moisture absorption of the latter. This later result combined with  
13 the L1 results previously discussed leads one to conclude that for susceptible aggregates, the  
14 amount of moisture absorption is a significant factor. In summary, the moisture-induced  
15 damage of the aggregate-bitumen bond is not only controlled by the mineralogical  
16 composition, but also the moisture absorption of aggregate should be considered.

17

18 In terms of the same aggregate, specimens prepared with B2 bitumen show higher retained  
19 strengths in comparison with B1. This is in contrast to previous studies indicating stiffer  
20 binders have better resistance. Therefore, more tests need to be done so as to confirm this.

21

#### 22 4.4.3 Effect of moisture on failure surface

23

24 Figure 9 shows the fracture surface photographs of the aggregate-bitumen specimens, taken  
25 immediately after the pull-off tensile test. The failure surfaces could be visually defined into

1 three types which are cohesive, adhesive and adhesive-cohesive mix. From this figure it can  
2 be seen that all specimens show cohesive failure in the dry condition. After moisture  
3 conditioning, the failure tends to transform from cohesive to adhesive-cohesive mix and even  
4 adhesive failure. It can be seen that specimens prepared with L1 aggregate retained the most  
5 cohesive failure, followed by G1, while specimens with G2 showed the least cohesive failure.

6  
7 Based on the photographs obtained, the damage proportions of all specimens were analysed  
8 using Image-J software. The percentage of the cohesive section of each specimen was  
9 calculated by identifying the grayscale levels, with results shown in Figure 10. The results  
10 shown in Figure 10 are important for two main reasons. Firstly, it allows a quantitative  
11 comparison of different specimens to identify the best aggregate-bitumen combination.  
12 Secondly, the results could be used to correlate with other parameters such as water  
13 absorption and retained tensile strength so as to confirm the identification. From this figure it  
14 can be seen that the retained cohesive failure was aggregate type dependent. Specimens  
15 prepared with L1 aggregate show the highest retained cohesive failure while G2 show the  
16 lowest value. In terms of the same aggregate, the influence of bitumen on the failure surface  
17 was not significant.

18

19 **5. Discussion**

20

21 5.1 Relationship between moisture absorption and retained tensile strength

22

23 The results presented in Section 4.4 showed the deterioration of tensile strength and failure  
24 surface development of the aggregate-bitumen bonds with the respect to moisture  
25 conditioning time. However, due to the disparity in physico-chemical properties, different

1 aggregates may absorb different amounts of water to attack the aggregate-bitumen interface.  
2 Just using conditioning time may not be adequate to evaluate the sensitivity of aggregate-  
3 bitumen bonds to moisture. Based on previous research (Apeagyei et al., 2015, Kringos,  
4 Scarpas & De Bondt, 2008), the relationship between retained strength and moisture uptake  
5 has been considered more realistic to characterise the moisture damage. Figure 11 shows the  
6 retained tensile strength of the specimens prepared with B1 bitumen versus the square root of  
7 moisture absorption. From this figure it can be seen that the retained tensile strength and  
8 square root of moisture content show a negative relationship. This demonstrated that by using  
9 the same type of aggregate, moisture uptake dominated the degradation of the aggregate-  
10 bitumen bonds. The bigger slope of granite than limestone suggesting it is more sensitive to  
11 moisture attack. In short, the deterioration of aggregate-bitumen bonds correlate well with the  
12 moisture uptake.

13

## 14 5.2 Relationship between retained tensile strength and failure surface

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16 The relationship between retained tensile strength and cohesive failure percentage of all  
17 specimens is shown in Figure 12. It can be seen that all results are located near the equality  
18 line with a higher percentage of cohesive surface achieving higher retained tensile strength.  
19 The results suggested that the deterioration of tensile strength is due to the transformation  
20 from cohesive failure to adhesive failure. So, the cohesive failure percentage could reflect the  
21 deterioration of the aggregate-bitumen bond the same way as retained tensile strength.

22

## 23 6. Conclusions

24

25 The following conclusions were reached based on the results presented in this study:

- 1       • For the three aggregates used in this research, the water absorption and mineralogical  
2       compositions showed different results. The differences of these fundamental  
3       properties are considered important to evaluate the moisture sensitivity of asphalt  
4       mixtures.
- 5       • The pull-off testing system used in this research was found to be effective in  
6       characterising the tensile strength of aggregate-bitumen bonds. The system is capable  
7       of controlling the bitumen film thickness with a resolution of 1  $\mu\text{m}$ .
- 8       • Before moisture conditioning, the bitumen grade dominated the tensile strength with  
9       40/60 pen bitumen giving higher values than the 70/100 pen bitumen. The results  
10      suggested the bitumen stiffness controls the aggregate-bitumen bond strength in the  
11      dry state to a higher extent than aggregate type.
- 12      • Based on the pull-off results, the moisture resistance of different aggregate-bitumen  
13      bonds could be explained by the moisture uptake and the mineralogical compositions  
14      of aggregates. With the same moisture absorption, limestone tends to have better  
15      resistance to moisture damage than granite. Furthermore, in terms of similar  
16      mineralogical compositions, lower moisture absorption results in better moisture  
17      resistance.
- 18      • The two bitumens used in this research showed similar ranking in terms of the  
19      moisture resistance demonstrating the effect of bitumen on moisture damage was  
20      lower than the effect of aggregate.
- 21      • For both the limestone and granite used in this research, the square root of moisture  
22      content and retained tensile strength correlated well. The significant correlation  
23      between the moisture uptake and retained tensile strength suggests that the water  
24      absorption process of the aggregate affects the degradation of the aggregate-bitumen  
25      bond.

- The failure surface was shown to transform from cohesive to a cohesive-adhesive mix and even adhesive failure with extended conditioning time. The quantified cohesive failure surface percentage was found to be correlated with the retained tensile strength. This result suggested that the deterioration of the aggregate-bitumen bond is directly linked to the decrease of the cohesive failure percentage.

## References

Abo-Qudais, S., & Al-Shweily, H. (2007). Effect of aggregate properties on asphalt mixtures stripping and creep behaviour. *Construction and Building Materials*, 21(9), 1886-1898.

Airey G. D., Collop A. C., Zoorob S. E., & Elliott R. C. (2008). The influence of aggregate, filler and bitumen on asphalt mixture moisture damage. *Construction and Building Materials*, 22(9), 2015-2024.

Apeageyi A. K., Grenfell J. R. A., & Airey G. D. (2015). Influence of aggregate absorption and diffusion properties on moisture damage in asphalt mixtures. *Road Materials and Pavement Design*, 16(1), 404-422.

Apeageyi A. K., Grenfell J. R. A., & Airey G. D. (2014). Moisture-induced strength degradation of aggregate–asphalt mastic bonds. *Road Materials and Pavement Design*, 15(1), 239-262.

Blackman B. R. K., Cui S., Kinloch A. J., & Taylor A. C. (2013). The development of a novel test method to assess the durability of asphalt road-pavement materials. *International Journal of Adhesion and Adhesives*, 42, 1-10.

Caro S., Masad E., Bhasin A., & Little D. N. (2008). Moisture susceptibility of asphalt mixtures, Part 1: mechanisms. *International Journal of Pavement Engineering*, 9(2), 81-98.

Grenfell J. R. A., Ahmad N., Airey G. D., Collop A. C., & Elliott R. C. (2012). Optimising the moisture durability SATS conditioning parameters for universal

1 asphalt mixture application. *International Journal of Pavement Engineering*, 13(5),  
2 433-450.

3 Grenfell J. R. A., Ahmad N., Liu Y., Apeageyi A. K., Large D. & Airey G. D. (2014).  
4 Assessing asphalt mixture moisture susceptibility through intrinsic adhesion, bitumen  
5 stripping and mechanical damage. *Road Materials and Pavement Design*, 15(1), 131-  
6 152.

7 Harvey J. A. F., & Cebon D. (2003). Failure mechanisms in viscoelastic films.  
8 *Journal of Materials Science*, 38, 1021–1032.

9 Horgnies M., Darque-Ceretti E., Fezai H., & Felder E. (2011). Influence of the  
10 interfacial composition on the adhesion between aggregates and bitumen:  
11 Investigations by EDX, XPS and peel tests. *International Journal of Adhesion and*  
12 *Adhesives*, 31(5), 238–247.

13 Kakar M. R., Hamzah M. O., & Valentin J. (2015). A review on moisture damages of  
14 hot and warm mix asphalt and related investigations. *Journal of Cleaner Production*,  
15 99, 39-58.

16 Kim Y. R., Little D. N., & Lytton R. L. (2004). Effect of moisture damage on material  
17 properties and fatigue resistance of asphalt mixtures. *Transportation Research Record*,  
18 1891, 48–54.

19 Kringos N., Scarpas A., & De Bondt A. (2008). Determination of moisture  
20 susceptibility of mastic-stone bond strength and comparison to thermodynamical  
21 properties. *Journal of the Association of Asphalt Paving Technologists*. 77, 435-478.

22 Mehrara A. & Khodaii A. (2013). A review of state of the art on stripping  
23 phenomenon in asphalt concrete. *Construction and Building Materials*, 38, 423-442.

24 Santucci L., (2002). Moisture Sensitivity of Asphalt Pavements. Technical Transfer  
25 Program, Institute of Transportation Studies, University of California, Berkeley,  
26 California.

27 Stuart K. D., (1990). Moisture Damage in Asphalt Mixture-State of the Art, Report  
28 No. FHWA-RD-90-019, FHWA, 6300, VA 22101-2296.

1 Terrel R. L., & Al-Swailmi S. (1994). Water sensitivity of asphalt-aggregate mixes:  
 2 test section. SHRPA-403, Strategic highway research program. Washington, DC:  
 3 National Research Council.

4 Zhang J., Airey G. D. & Grenfell J. R. A. (2015a). Experimental evaluation of  
 5 cohesive and adhesive bond strength and fracture energy of bitumen-aggregate  
 6 systems. *Materials and Structures*, 1-15.

7 Zhang J., Apeageyi A. K., Airey G. D., & Grenfell J. R. A. (2015b). Influence of  
 8 aggregate mineralogical composition on water resistance of aggregate-bitumen  
 9 adhesion. *International Journal of Adhesion and Adhesives*, 62, 45-54.

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18 **Tables**

19

20 Table 1. Softening point and penetration results of bitumen

Property	Bitumen	
	B1	B2
Softening point (°C)	51.2	45.2
Penetration (0.1 mm)	46	81

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23 Table 2. Frequency sweep testing condition to establish LVE limit of bitumen

Testing condition	Bitumen							
	10	20	30	40	50	60	70	80
Temperature (°C)	10	20	30	40	50	60	70	80
Target Strain (%)	0.5	0.5	0.5	1	1	1	1	1
Parallel Plate Diameter	8mm			25mm				
Sample Thickness	2mm			1mm				
Frequency	0.1-10 Hz							

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1 Table 3 Mineral composition of aggregates identified by MLA analysis

Mineral type	Composition (Wt%)		
	G1	G2	L1
Chlorite	31.53	13.52	-
Albite	27.13	32.73	-
Quartz	19.11	15.86	0.49
Epidote	11.11	1.37	-
K-feldspar	4.82	9.64	-
Muscovite	2.39	3.43	-
Hornblende	1.88	2.57	-
Biotite	0.99	0.34	-
Other	0.74	1.91	0.30
Calcite	0.20	0.08	96.98
Anorthite	0.10	18.54	-
Dolomite	-	-	1.30
Clay	-	-	0.93
Total	100	100	100

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3 Table 4. Tensile strength (kPa) of aggregate-bitumen in both dry and wet conditions at 20°C  
4 with loading rate 10 mm/min

Sample ID	Mean ± Std (kPa)		
	Dry	24 hours	168 hours
B1-L1	1920 ± 103	1390 ± 206	1384 ± 196
B1-G1	1947 ± 199	1351 ± 113	1293 ± 149
B1-G2	1938 ± 312	498 ± 143	371 ± 224
B2-L1	1425 ± 147	1203 ± 71	1078 ± 72
B2-G1	1386 ± 72	1248 ± 175	1062 ± 199
B2-G2	1413 ± 128	1042 ± 200	799 ± 185

5 Note: B1 = 40/60 pen bitumen; B2 = 70/100 pen bitumen; L1 = limestone; G1= granite 1; G2  
6 = granite 2; Mean = average value; Std = standard deviation

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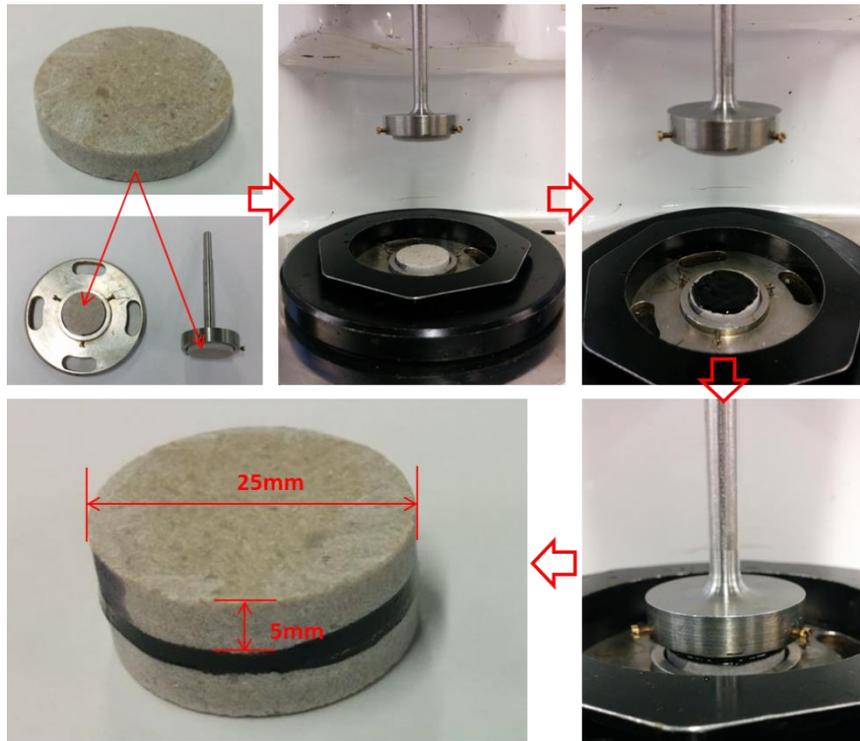
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1 **Figures**

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Figure 1. Aggregate-bitumen specimen preparation procedures

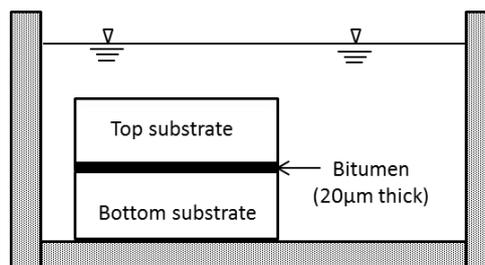
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Figure 2. Specimen of aggregate-bitumen adhesion submerged into water

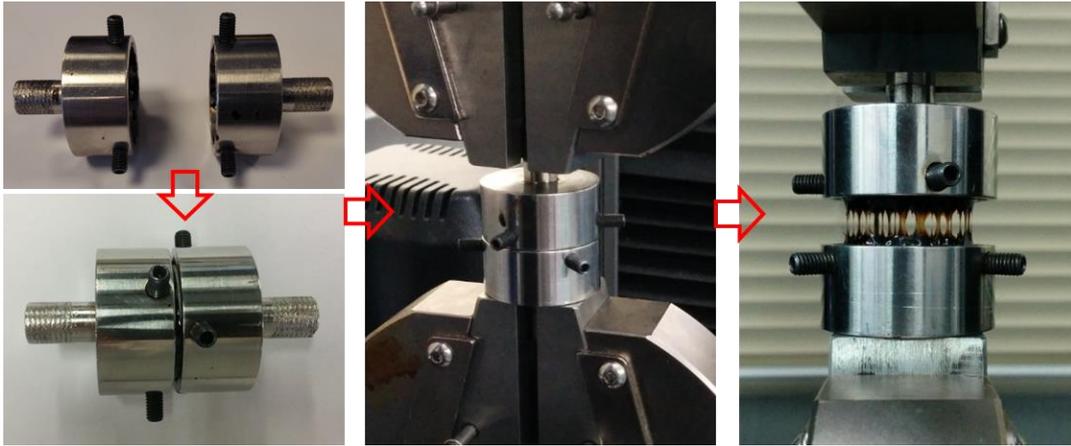


Figure 3. Procedures for pull-off test

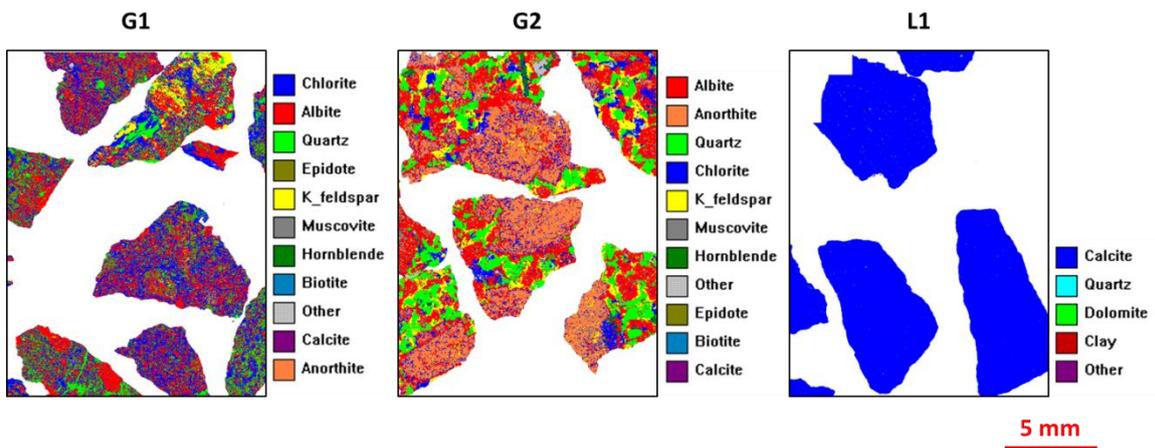


Figure 4. Mineral distribution of the three aggregates

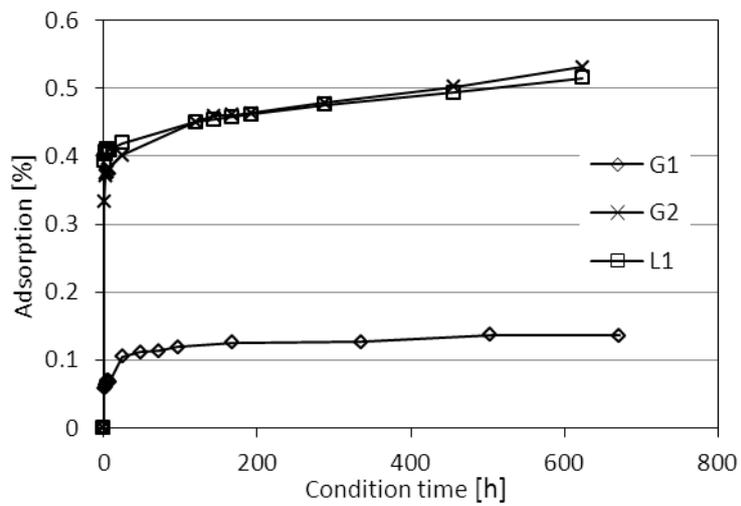


Figure 5. Moisture uptake of the three aggregates used in this research

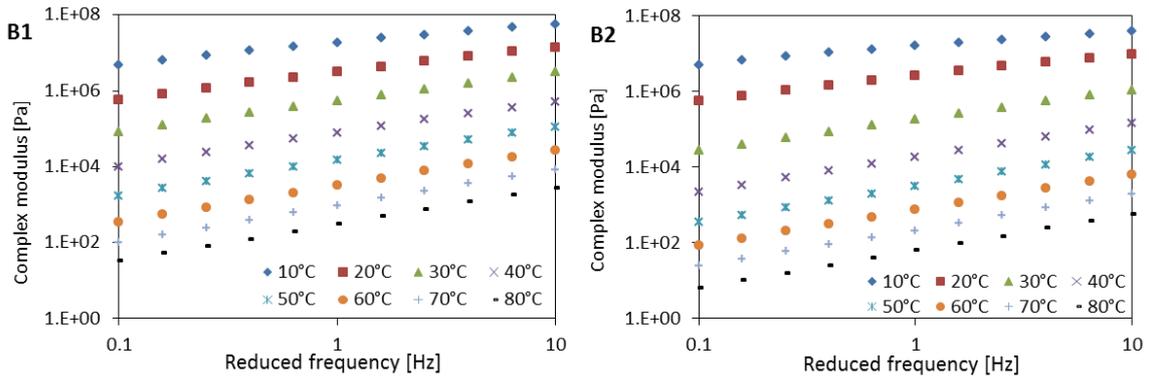


Figure 6. Complex modulus results for the two binders used in this research

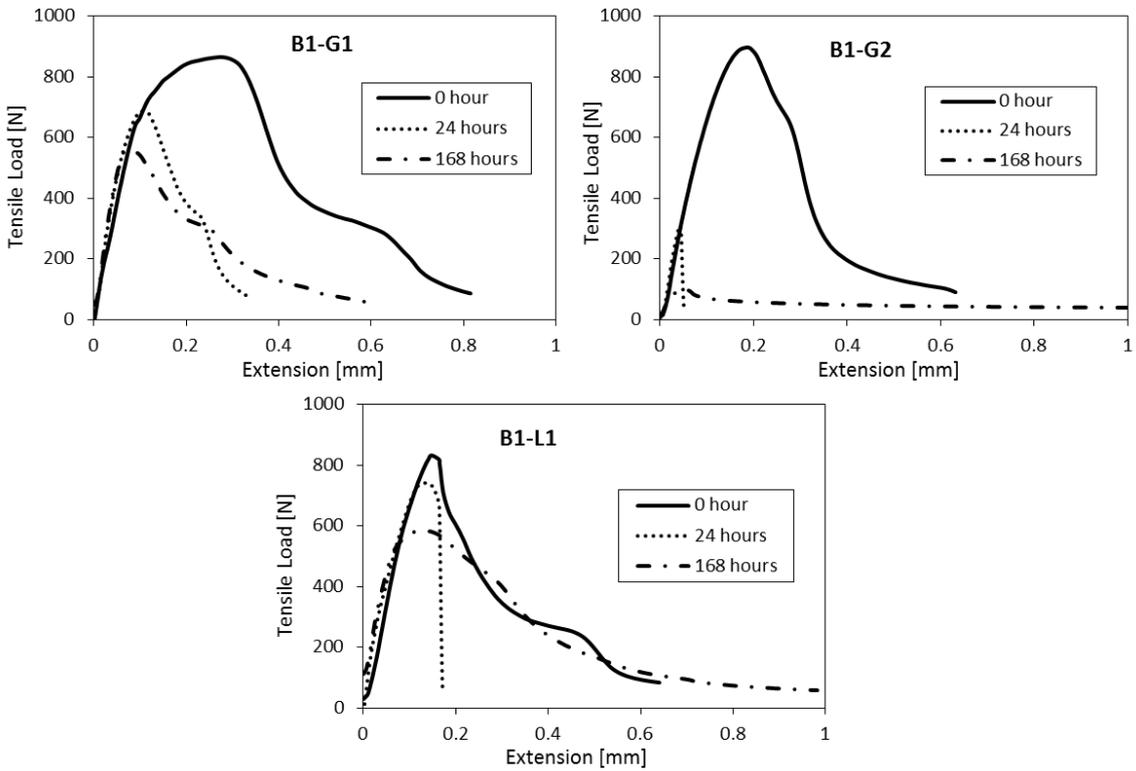
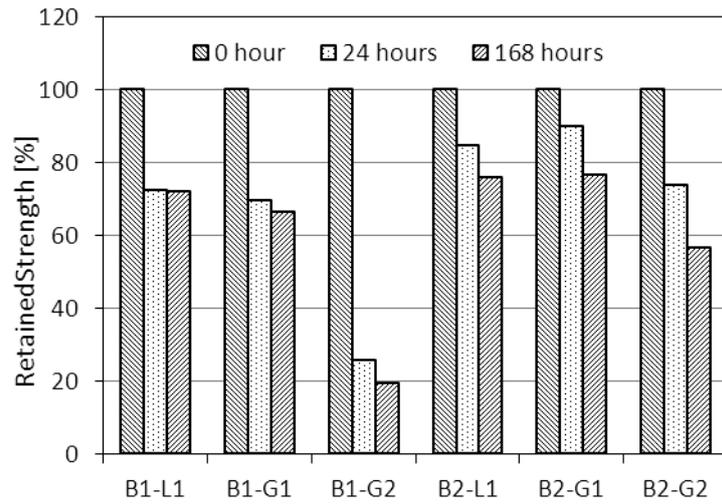
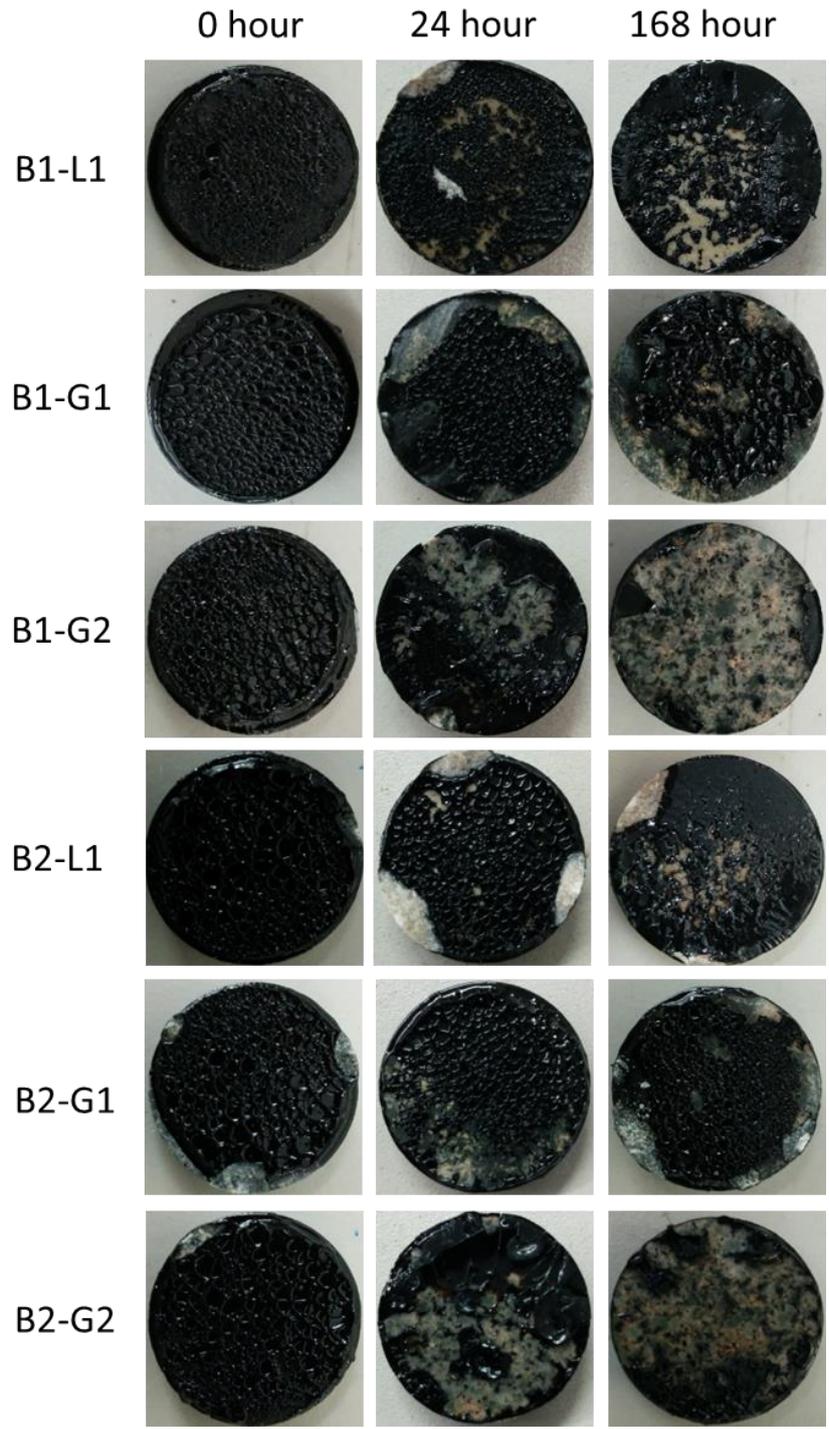


Figure 7. Development of stress-strain behaviour of aggregate-bitumen combined samples with respect to moisture conditioning time. Samples were conditioned in water at 20C; loading rate was 10mm/min.



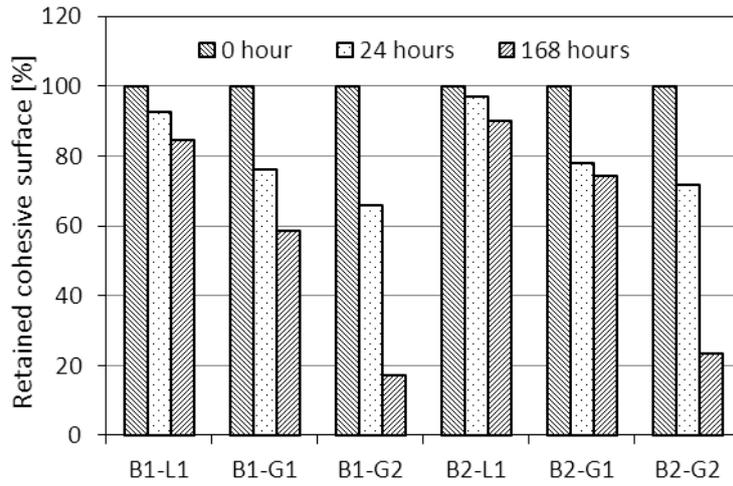
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Figure 8. Retained tensile strength of different aggregate-bitumen combinations with the passage of conditioning time



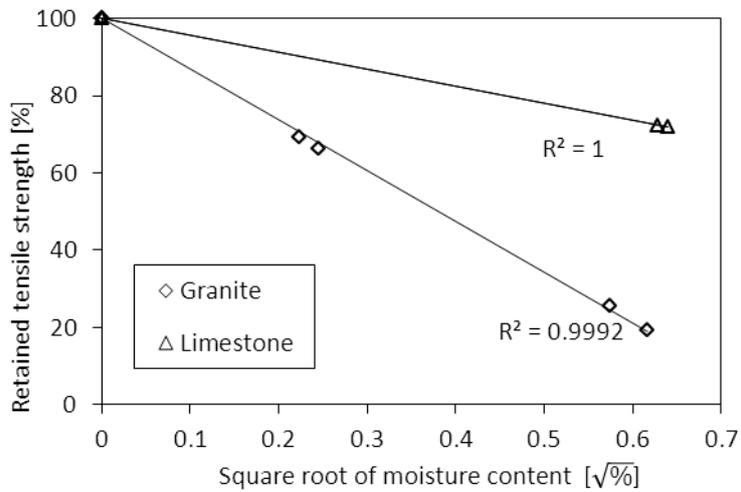
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Figure 9. Failure surface photographs of aggregate-bitumen bonds before and after moisture conditioning. The effect of bitumen type is minimal compared with the effect of aggregate type



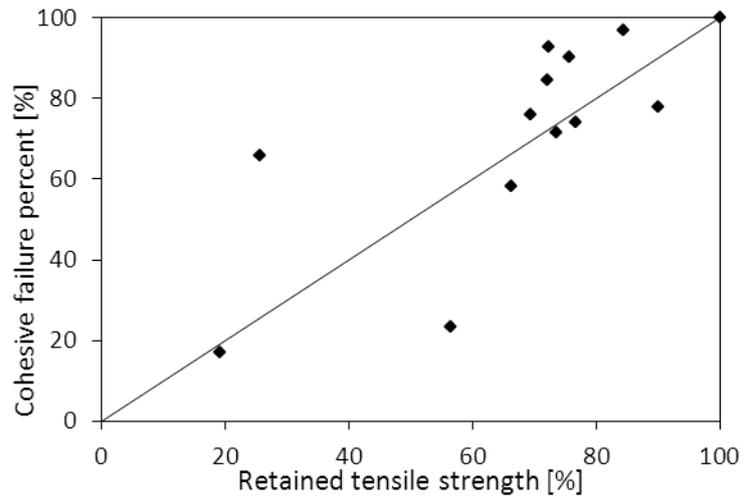
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Figure 10. Retained cohesive surface percentage of different aggregate-bitumen combinations with the passage of conditioning time



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Figure 11. Correlation between retained tensile strength and square root of moisture uptake: specimens prepared with bitumen B1



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Figure 12. Correlation between retained tensile strength and cohesive failure percentage: all specimens (B1-L1, B1-G1, B1-G2, B2-L1, B2-G1 and B2-G2) and conditioning time (0 hour, 24 hours and 168 hours) included.