Moisture damage evaluation of aggregate–bitumen bonds with the respect of moisture absorption, tensile strength and failure surface

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

Keywords: pull-off system; aggregate–bitumen bonds; moisture damage; adhesion; cohesion

1. Introduction

Asphalt mixtures are widely used as pavement construction materials. During their service life, asphalt pavements have to sustain harsh traffic loads and environmental conditions and deteriorate with the passage of time. The effect of moisture on asphalt mixtures is recognised as a major cause of pavement failure. The penetration of moisture through asphalt mixtures can increase the pavements’ vulnerability to traffic loading and thermal stress (Kim, Little, & Lytton, 2004; Mehrara & Khodaii, 2013). Moisture damage in asphalt pavement is defined as the loss of strength, stiffness and durability because of the presence of moisture, resulting in adhesive failure at the aggregate–bitumen interface and/or cohesive failure within the bitumen or mastic (Airey, Collop, Zoorob, & Elliott, 2008). With the presence of moisture, water may enter the aggregate–bitumen interface by diffusion through bitumen films, seepage into the film through micro voids or cracks, and through direct access in partially coated aggregates (Stuart, 1990). It is noticeable

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that the existence of moisture may only weaken the asphalt mixture by emulsifying or softening the bitumen film but without removing it from aggregate surfaces. Also, when the moisture is removed from the asphalt mixture, the stiffness loss is reversible. However, when the pavement is loaded during the weakened condition, the moisture damage is accelerated and may become irreversible (Santucci, 2002). Although not all damage is caused directly by moisture, its presence increases the extent and severity of already-existing distresses such as cracking, potholes and rutting (Grenfell et al., 2014).

According to previous researchers (Caro, Masad, Bhasin, & Little, 2008; Kakar, Hamzah, & Valentin, 2015), the moisture in either a liquid or vapour state infiltrates the asphalt mixture as well as the bitumen film or mastic and reaches the aggregate–bitumen interface so as to change the internal structure and finally results in the degradation of mechanical properties of the material. In addition, the moisture may also invade the asphalt mixture system by seeping through the already-existing cracks in the mixture or by diffusing outward from the aggregate pores. Once moisture has come into contact and interacted with the asphalt mixture, the moisture damage could be developed in the following mechanisms: detachment, displacement, spontaneous emulsification, pore pressure and hydraulic scour (Grenfell et al., 2014). It should be mentioned that the moisture damage is not limited to only one mechanism but is the result of a combination of several mechanisms.

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

By measuring the adhesive bond strength of coatings between bitumen and aggregate, several testing techniques have been developed but the most commonly used methods include the pull-off test and peel test. Normally, the pull-off test is conducted by measuring the tensile stress necessary to detach the adhesive materials in a direction perpendicular to the substrates (Apeagyei, Grenfell, & Airey, 2014; Harvey & Cebon, 2003). In terms of the peel test, a thin flexible peel arm and a rigid substrate are bonded using the adhesive material. During testing, the peel arm is pulled from the substrate at a specified angle and speed while the peel force is recorded. The recorded peel force in steady-state conditions is then used to calculate the fracture energy of the adhesive (Blackman, Cui, Kinloch, & Taylor, 2013; Horgnies, Darque-Ceretti, Fezai, & Felder, 2011). These two methods have been successfully used to evaluate the moisture sensitivity of the aggregate–bitumen bond by immersing specimens in water for a range of times before testing. However, the limitations of these established tests are very obvious. First of all, the mechanical evaluation only reflects the influence of conditioning time but not the amount of moisture absorbed. Secondly, manual control of bitumen film thickness makes it hard to obtain the required thickness and results in big deviations in measured strength. Thirdly, these studies were limited in their ability to control bitumen film thickness to submicron level and hence cannot simulate the real bitumen thickness in mixtures. For a better understanding of the performance of the aggregate–bitumen interface when exposed to moisture, this paper presents the development of a suitable procedure consisting of innovative sample preparation, controlled moisture conditioning, pull-off test set-up and failure surface evaluation.
2. Materials

Two base bitumens named B1 and B2 with penetration grades of 40/60 pen and 70/100 pen, respectively, were selected. These two binders were from the same crude source and therefore had similar chemical compositions (Zhang, Airey, & Grenfell, 2016). The fundamental physical properties of the bitumen were measured using softening point (ASTM D36) and penetration (ASTM D5) tests with the results shown in Table 1.

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

3. Methodology

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

3.1. MLA test

It has been accepted that the mineralogical compositions of aggregates have a profound influence on moisture sensitivity of asphalt mixtures. By measuring the mineralogical properties of aggregates, an MLA device was used in this research. The MLA is an automated mineral analysis system that can identify minerals in polished sections of drill cores, particulates or lump materials, and quantify a wide range of mineral characteristics such as mineral abundance, grain size and liberation. Before testing, the aggregate need to be polished and carbon coated to get an electron conductive surface. During testing, the Back-scattered Electron image is combined with Electron Dispersive X-ray analysis for the specimen surface for a series of frames step by step. Finally, the MLA’s data-view software allows presenting the digital results in a graphical format. A detailed introduction about the testing procedure can be seen in previous publications (Grenfell et al., 2012; Zhang, Apeagyei, Airey, & Grenfell, 2015).

| Bitumen | Softening point (°C) | Penetration (0.1 mm) |
|---------|----------------------|---------------------|
| B1      | 51.2                 | 46                  |
| B2      | 45.2                 | 81                  |
3.2. **Moisture absorption of aggregates**

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

\[
\text{Mass uptake(\%)} = M_t = \frac{w_t - w_0}{w_0},
\]

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

3.3. **DSR test**

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

3.4. **Pull-off test**

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

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

| Testing condition               | Bitumen |
|-------------------------------|---------|
| 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       | 8 mm 25 mm |
| Sample thickness              | 2 mm 1 mm |
| Frequency                     | 0.1–10 Hz |
3.4.1. Aggregate–bitumen specimen preparation

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 min and dried in an oven at a temperature of 40°C for 24 h. 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 min 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.

Figure 1. Aggregate–bitumen specimen preparation procedures.
3.4.2. **Moisture conditioning of adhesion specimen**

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 and 168 h) with the temperature maintained at 20°C. The schematic diagram of the moisture conditioning is shown in Figure 2. During the moisture conditioning, moisture could reach the aggregate–bitumen interface in three different ways: through the top and bottom aggregate, through the edge of aggregate–bitumen interface and through the bitumen film.

3.4.3. **Bond strength evaluation using pull-off test**

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

![Figure 2. Specimen of aggregate–bitumen adhesion submerged into water.](image)

![Figure 3. Procedures for pull-off test.](image)
load divided by the cross-sectional area of the bitumen film as follows:

\[ TS = \frac{F}{1000 \times \pi \times r^2}, \]  

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

4. Results

4.1. Mineralogy of aggregates

Figure 4 shows the mineralogical distribution on aggregate surfaces obtained from the MLA with the mineralogical composition shown in Table 3. As shown in Figure 4, the mineral distribution of G1 and G2 is much more complex than that of L1. The two granite aggregates G1 and G2 were made up of a large number of different mineral phases, while there are very few mineral phases in limestone L1. As shown in Table 3, chlorite and albite are the foremost minerals in G1 with quantities of 31.53% and 27.13% by weight, followed by quartz, epidote and K-feldspar, which account for 19.11%, 11.11% and 4.82%, respectively. Albite and anorthite are the predominant

| Mineral type       | 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  |

Figure 4. Mineral distribution of the three aggregates.
minerals in G2, which account for 32.73% and 18.54% by weight, but quartz and chlorite also have significant quantities. In terms of limestone L1, calcite is the dominant phase with 96.98% by weight.

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

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

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.

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

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

4.4.2. Effect of moisture conditioning on aggregate–bitumen bond strength
The tensile strength of each aggregate–bitumen bond in the dry condition and after moisture damage was calculated based on Equation (3) with the average values and standard deviation for all specimens depicted in Table 2. From this table it can be seen that specimens prepared with the same bitumen tend to yield similar tensile strength in the dry condition, no matter which aggregate substrate was used. This could be attributed to the cohesive failure surface observed in the dry condition. In terms of the same aggregate substrate in the dry condition, samples prepared with B1 bitumen exhibited higher tensile strength than those with B2 bitumen. This phenomenon correlates well with the bitumen fundamental properties where the softening point and complex
Figure 7. Development of stress–strain behaviour of aggregate–bitumen combined samples with respect to moisture conditioning time. Samples were conditioned in water at 20°C; loading rate was 10 mm/min.

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

| Sample ID | Dry   | 24 h  | 168 h |
|-----------|-------|-------|-------|
| 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  |

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

modulus are higher for B1 than B2. So, it can be concluded that, in dry conditions, the tensile strength of the aggregate–bitumen bond is dominated by the bitumen rather than by the aggregate (Table 4).

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

Retained strength, the ratio of bond strength after a given level of moisture conditioning to the dry bond strength, is considered to be a common parameter to measure the moisture sensitivity of asphalt mixtures. High retained tensile strength demonstrating better moisture resistance of the specimen. Figure 8 shows the effect of conditioning time on retained tensile strength of different aggregate–bitumen bonds. By using the pull-off test results, it is possible to identify “good” and “bad” mixtures. It can be seen that samples prepared with L1 and G1 show good moisture resistance with over 70% tensile strength retained after 168 h conditioning. However, samples prepared with G2 are more sensitive to moisture attack as the tensile strength decreased by over 80% and 40% for B1 and B2 bitumen, respectively. The aggregates L1 and G2 have similar moisture absorption, meaning similar time will be taken to transport moisture to aggregate–bitumen interface, but they show a significant difference in retained strength. This is because the bonds formed between bitumen and G2 are quickly degraded once in contact with moisture due to the large amount of albite and quartz. However, calcite, being the dominant mineral in L1, can form water-insoluble bonds with bitumen that retain better moisture resistance. In terms of G1, due to its lower water absorption, it will take a much longer time for water to reach the aggregate–bitumen interface. On this basis, there is limited chance for water to attack the bonds even though G1 contains several moisture-sensitive minerals. The difference in retained strengths between G1 and G2 could be attributed to higher moisture absorption of the latter. This later result combined with the L1 results previously discussed leads one to conclude that for susceptible aggregates, the amount of moisture absorption is a significant factor. In summary, the moisture-induced damage of the aggregate–bitumen bond is not only controlled by the mineralogical composition, but the moisture absorption of aggregate should also be considered.

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

![Figure 8. Retained tensile strength of different aggregate–bitumen combinations with the passage of conditioning time.](image-url)
4.4.3. **Effect of moisture on failure surface**

Figure 9 shows the fracture surface photographs of the aggregate–bitumen specimens, taken immediately after the pull-off tensile test. The failure surfaces could be visually defined into three types, which are cohesive, adhesive and adhesive–cohesive mix. From this figure, it can be seen that all specimens show cohesive failure in the dry condition. After moisture conditioning, the failure tends to transform from cohesive to adhesive–cohesive mix and even adhesive failure. It can be seen that specimens prepared with L1 aggregate retained the most cohesive failure, followed by G1, while specimens with G2 showed the least cohesive failure.

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

![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.](image-url)
5. Discussion

5.1. Relationship between moisture absorption and retained tensile strength

The results presented in Section 4.4 showed the deterioration of tensile strength and failure surface development of the aggregate–bitumen bonds with the respect to moisture conditioning time. However, due to the disparity in physico-chemical properties, different aggregates may absorb different amounts of water to attack the aggregate–bitumen interface. Just using conditioning time may not be adequate to evaluate the sensitivity of aggregate–bitumen bonds to moisture. Based on previous research (Apeagyei et al., 2015; Kringos, Scarpas, & De Bondt, 2008), the relationship between retained strength and moisture uptake has been considered more realistic to characterise the moisture damage. Figure 11 shows the retained tensile strength of the specimens prepared with B1 bitumen versus the square root of moisture absorption. From this figure it can be seen that the retained tensile strength and square root of moisture content show a negative relationship. This demonstrated that by using the same type of aggregate, moisture uptake dominated the degradation of the aggregate–bitumen bonds. The bigger slope of granite
than limestone suggesting it is more sensitive to moisture attack. In short, the deterioration of aggregate–bitumen bonds correlate well with the moisture uptake.

5.2. Relationship between retained tensile strength and failure surface

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

6. Conclusions

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

- For the three aggregates used in this research, the water absorption and mineralogical compositions showed different results. The differences of these fundamental properties are considered important to evaluate the moisture sensitivity of asphalt mixtures.
- The pull-off testing system used in this research was found to be effective in characterising the tensile strength of aggregate–bitumen bonds. The system is capable of controlling the bitumen film thickness with a resolution of 1 μm.
- Before moisture conditioning, the bitumen grade dominated the tensile strength with 40/60 pen bitumen giving higher values than the 70/100 pen bitumen. The results suggested the bitumen stiffness controls the aggregate–bitumen bond strength in the dry state to a higher extent than aggregate type.
- Based on the pull-off results, the moisture resistance of different aggregate–bitumen bonds could be explained by the moisture uptake and the mineralogical compositions of aggregates. With the same moisture absorption, limestone tends to have better resistance to moisture damage than granite. Furthermore, in terms of similar mineralogical compositions, lower moisture absorption may result in better moisture resistance.
The two bitumens used in this research showed similar ranking in terms of the moisture resistance demonstrating the effect of bitumen on moisture damage was lower than the effect of aggregate.

For both the limestone and granite used in this research, the square root of moisture content and retained tensile strength correlated well. The significant correlation between the moisture uptake and retained tensile strength suggests that the water absorption process of the aggregate affects the degradation of the aggregate–bitumen 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.

Disclosure statement
No potential conflict of interest was reported by the authors.

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