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Utilization of Distilled Water to Observe Surface Contact Angle of Polymer-Modified Bitumen

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Abstract Stable sub-grade and reliable structural layers are required in road pavements to ensure long serviceability. One of the road distresses associated with the sub-grade (unbound soil) or top (bituminous) structural layer failure is rutting in flexible pavement. This is the reason why better rutting resistance is highly desirable in bitumen to prevent potential damage as well as to ensure higher durability for the pavement. Studies on surface material identified the concept of contact angle by testing liquids on solid surface. This concept can also be applied to bitumen to determine its ability to repel or attract a particular liquid such as water in line with the modification rate. Therefore, this study was conducted in an effort to improve rutting and fatigue through low-density polyethylene (LDPE) polymer-based modified bitumen. The focus was to determine the ability of measuring water contact angle (WCA) to differentiate several types of modified bitumen. Polymer-modified bitumen was produced through high shear mixing using virgin and recycled LDPE to improve its rutting resistance. Moreover, standard tests such as ring and ball softening points were conducted to determine the penetration number and softening point to confirm the modification results. WCA tests were applied to both the unmodified and the modified bitumen, and some differences were observed in the results.

Keywords Polymer-Modified Bitumen, Low-Density Polyethylene, Water Contact Angle, Moisture Damage

1. Introduction

Several cities in the world are experiencing an increasing demand for transportation and this is causing traffic congestion [1] and different infrastructural problems. Traffic problems are generally related to insufficient road infrastructures as opposed to demands. Meanwhile, infrastructural challenges are linked to the quality and durability of pavement structures for accommodating different types of vehicles. Indonesia, as one of the developing countries in the world, is constructing massive road infrastructures including local and arterial roads, and highway to compete and fulfill transportation demands. However, the two main issues generally identified in road pavements that require continuous study are (1) the subgrade or issues related to soil and (2) the pavement or road structure itself.

Soil subgrade aspects are issues related to the existence of soft soils such as soft clay, peat, and loose sandy types with low bearing capacity. There are several soils on road construction sites requiring improvement before the actual work is initiated. Several studies have proposed pre-loading, chemically-based soil improvements, or physical compaction to improve the bearing capacity of soils. Moreover, different types of substances have been proposed for chemical grouting and these include cement, lime, or other solutions, depending on the stabilization as well as the characteristics of the soil [2–4]. Bio-grout-based ground improvement practices such as calcite precipitation and bio-polymer have also been proposed as potential and effective techniques, specifically for soil strengthening under existing infrastructures [5].

Several studies have also been conducted on the structure of road pavement to accommodate the need for heavier vehicles and busier traffic. This is observed in the continuous focus of flexible pavement studies on normal and polymer-modified bitumen as one of the major materials for road construction. This includes an insightful observation of the characteristics of bitumen considered important to the performance of the pavement. One of the methods to analyse the surface of materials is contact angle and this focuses on providing a quick and easy initial description of the behaviour of such material as either hydrophilic (attract moisture) with contact angle of Θ<90°or hydrophobic (repel moisture) with Θ>90° [6,7] as presented in Figure 1. This method can also be applied in bitumen modification studies to understand the effect of polymer-modified. A further deep analysis such as surface free energy [6,8] can also be implemented based on a similar principle using different probe liquids such as Glycerol, Formamide, or Diiodomethane. The application of these liquid probes for the initial analysis can be too expensive. Therefore, distilled water is hypothesized to be sufficient for the early determination of the characteristics of bitumen. This has led to the wide application of contact angle method to water and other probe liquids but none has focused on low-density polyethylene (LDPE)-modified bitumen, specifically using distilled water for the initial characterisation. It is also important to reiterate that pure or distilled water is considered practically usable in contact angle tests and relatively cheaper. This study was conducted as part of the efforts to improve the rutting and fatigue resistance of LDPE-modified bitumen with a focus on the use of distilled water in contact angle analysis. The main aim was to understand the effectiveness of distilled water in differentiating the level of bitumen modification, specifically in the case of LDPE-modified bitumen. Contact angle tests were conducted on the Goniometer while a simple set of standard tests related to penetration number and softening points were used to indicate a successful modification process. The different contact angle test results of the tested bitumen indicate the successful modification of the LDPE-modified bitumen.

2. Method

2.1. Materials and Equipment

This study incorporated 50/70 penetration grade bitumen and LDPE plastic as bitumen modifiers. LDPE plastic also consisted of pure or virgin LDPE (VL) and recycled LDPE (RL). VL was typically white or opaque round pellets with a diameter of 3-4 mm produced by Sigma Aldrich (Merck) from Germany. RL was flat pellets with a diameter of 4-5 mm in a mixed colour of light to dark blue produced by BJ Parr recycling company, Mansfield, United Kingdom. The two types are presented in the following Figure 2. Distilled water was applied as water contact angle (WCA) in the Goniometer while Glycerol was used as a liquid bath in the ring and ball test for highly modified bitumen and the softening point was expected to exceed 80 ℃.

The equipment used was a high-shear mixer for bitumen modification, an oven for different heating purposes, a bitumen standard needle penetration test, a ring and ball softening point test, and a set of Goniometers for contact angle measurements.

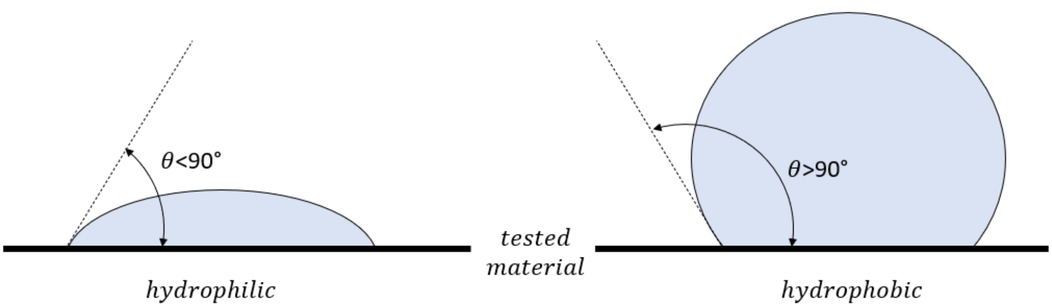
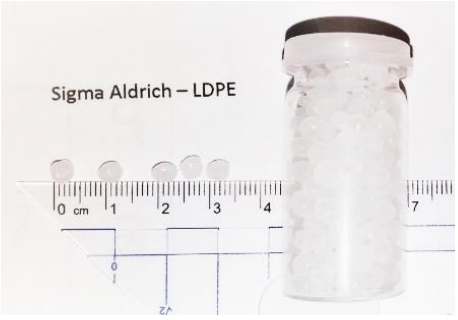


Figure 1. Hydrophilic and Hydrophobic Behaviour of Materials



Produced by Sigma-Aldrich (Merck)
Form, diameter: round pellets, 3-4 mm
Colour: white/opaque

Produced by BJ Parr recycling company
Form, diameter: round/flat pellets, 4-5 mm
Colour: mixed light to dark blue

Figure 2. The Two Types of LDPE Applied for Bitumen Modification

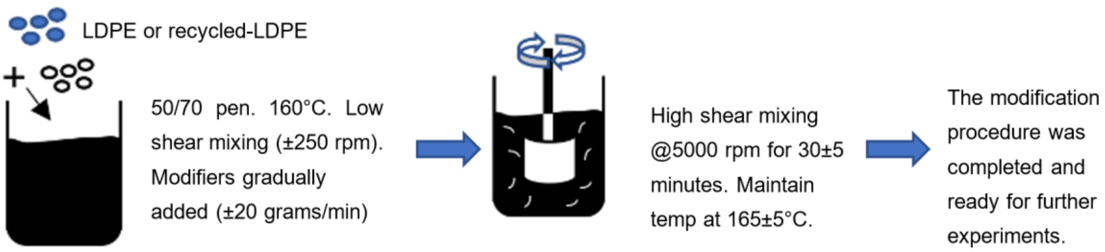


Figure 3. High Shear Mixing Method For LDPE-Modified Bitumen Production

2.2. Procedure

The study was initiated with an LDPE polymer-modified process using a high-shear mixer followed by standard tests on both the normal and modified bitumen, the preparation of Goniometer samples on microscopy glass, and the measurement of distilled WCA.

2.2.1. Preparation and Modification Process

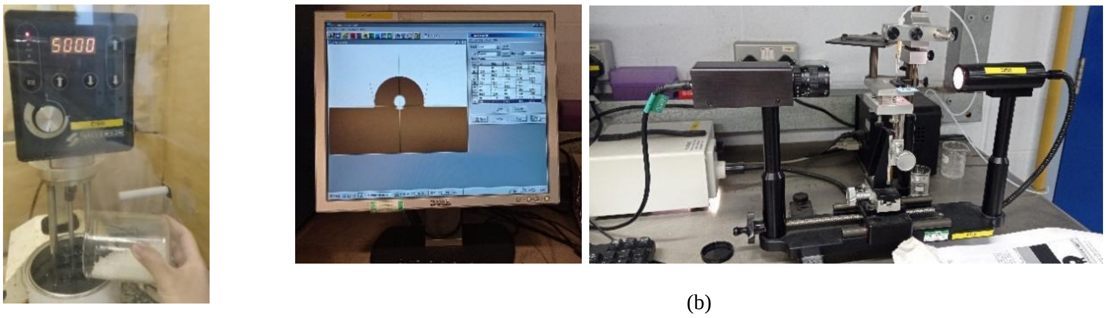
Bitumen modifications were conducted using high shear mixing at 5000 rpm, 165 ℃ for 30 minutes. The mixing duration and method were selected based on successful results in a previous study and the insignificant improvements recorded in terms of viscosity for a longer mixing duration [9]. The modifications of 2%, 4%, 6%, 8% and 10% of bitumen weight were applied to VL to produce VL1, VL2, VL3, VL4, and VL5, and also to RL to produce RL1, RL2, RL3, RL4 and RL5, respectively. Meanwhile, the normal or unmodified bitumen was used as the control and was labelled R0.

The high shear mixing modification process was initiated with the preparation of the sample by oven heating the neat 50/70 penetration grade bitumen at 165 ℃ until it turned to liquid form. Bitumen was poured approximately 1700 grams into a 2000mL tin and this quantity was required for each modification level to ensure the high mixing shear head was fully submersed in the blends. The weight of VL and RL modifiers was also calculated and prepared based on the actual bitumen poured.

Bitumen prepared at ±1700 gram was sheared at lower rate of ±250 rpm while maintaining the temperature at 165 ℃. During this low shearing rate, LDPE was poured gradually at ±20 grams/minute into the blends. This was followed by an increase in the speed to 5000 rpm and the blending process was continued for another 30 minutes while maintaining the 165 ±5 ℃ temperature as presented in Figure 3. The same process was conducted for all ten samples of modified bitumen and the high-shear mixer used is presented in Figure 4 (a).

2.2.2. Standard Bitumen Tests on Penetration and Softening Point

Standard tests were conducted on the penetration number based on BS EN 1426-2015 and the softening point on BS EN 1427-2015 for both unmodified (R0) and LDPE-modified bitumen (VL and RL). The results were expected to be used in differentiating between the modified and unmodified bitumen samples. Several previous studies have incorporated LDPE as a bitumen modifier and it was generally agreed to have lower penetration number and higher softening point [9–11]. Moreover, R&B softening point results were believed to have the capacity to provide an insightful explanation of the success rate of the modification procedure for modified bitumen [12]. This was confirmed by the results that 2% and 4% modifications had R&B t<80 ℃ while the higher modified bitumen had t>80 ℃ with the use of glycerol as the liquid bath.



(a) (b)

Figure 4. (a) The High Shear Mixing and (b) WCA Goniometer Testing Equipment

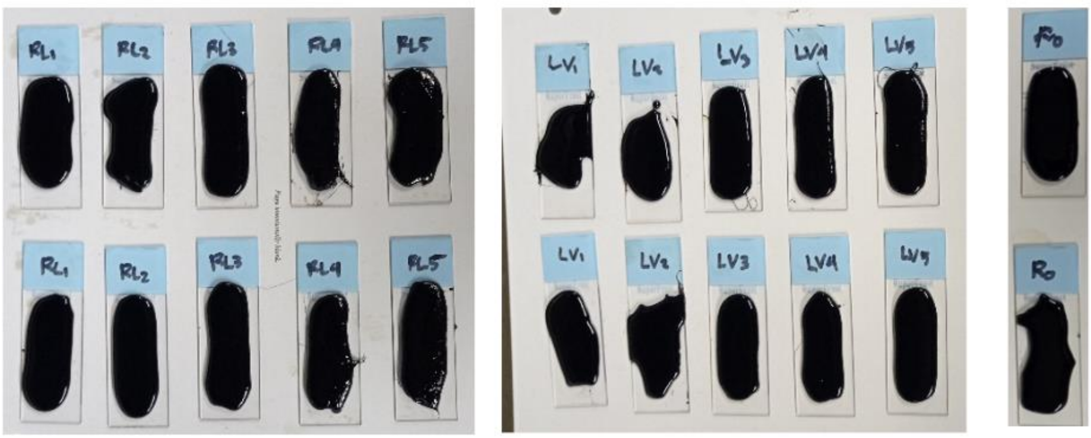


Figure 5. WCA Test Specimens

2.2.3. Contact Angle Sample Preparation and Measurement

WCA specimens were prepared by pouring hot bitumen on top of a microscopy observation glass, and the process was repeated twice for each type of bitumen. The specimens were made to have sufficient fluidity by conducting the pouring process at approximately 180 ℃ for modified and 160 ℃ for unmodified bitumen. Moreover, the specimen glasses were heated to 150-160 ℃ before the pouring process to prevent any cracks due to sudden temperature changes. Extra care was ensured to achieve the appropriate level of safety and successful results considering the high temperature of the specimens.

The Goniometer test equipment used is presented in Figure 4 (b) and WCA specimen is in Figure 5. The dimension of the specimens was estimated at 10mm x 40mm x 2mm. Moreover, RL was used to indicate RL-modified bitumen, VL for VL-modified bitumen, and the R0s for the unmodified bitumen.

The pouring process was used to produce the flat-surface specimen for the two types of bitumen analysed. For highly modified bitumen such as VL4, VL5, RL4 and RL5, the process led to a slightly wavy surface and this required special attention during the application of the Goniometer to ensure water dropped in the flat area of the specimen.

3. Results and Discussion

3.1. Penetration Number

The penetration number test showed noticeable differences between the unmodified and both VL and RL modified bitumen. Figure 6 shows that 2% of VL and RL produced relatively the same penetration number but the value obtained at 4% was slightly lower for RL compared to VL which had a higher modification effect. Further effect was also observed at higher rates where RL had lower penetration number than VL at the same %w of modification.

Penetration number was defined as a simple test to provide an early indication of the changes in the physical property of the modified bitumen. At room temperature, LDPE was widely known to be sturdy, impact-resistant and withstand tensile strength at approximately 9 MPa [13]. The modification of bitumen was observed to have led to swelling and dispersion of the particles of polymer based on the compatibility factor between the two materials [14,15]. Therefore, the lower penetration number of the modified bitumen at room temperature after the modification process indicated the proportions of the physical properties in LDPE that changed those of bitumen-polymer system. A higher content of LDPE led to higher changes or lower penetration numbers of the modified product. It was also observed that RL reduced the penetration number more than VL at the same %bitumen weight. This indicated better modification in RL due to its better dispersion and swelling in bitumen compared to VL.

3.2. Softening Point

The softening point test also showed some changes due to polymer modification just as observed with the penetration number. It was discovered in Figure 6 that the softening points were increasing in line with the decreasing penetration number. The ring and ball softening points were used to measure the compatibility of polymer blends. According to [12], the increasing number of softening points for the modified bitumen represented a better modification blend. Several studies on the materials modified using polyethylene reported higher softening points and this was also indicated in the changes recorded in the high-temperature complex modulus which was an important rheological property [3,9,10,16–19]. The R&B obtained for the modified bitumen in Figure 6 showed the typical results of highly modified bitumen, specifically VL4, VL5, RL3, RL4 and RL5 and were also expected to be observed in WCA measurements.

3.3. Distilled WCA

WCA was measured after bitumen specimens were cooled to room temperature and rested for 24 hours. Approximately 200 mL of distilled water was prepared and pumped to the Goniometer. Each specimen had three drops of distilled water on three separate spots to be measured as presented in Figure 7 followed by the calculation of the average angle. Figure 5 shows an example of the typical water drops and their WCA measurements. It was observed that two specimens were produced for each bitumen type, leading to a total of 6 measurements per type. Figure 7 shows the measurement of water drops and the average WCA from the six specimens for each bitumen type.

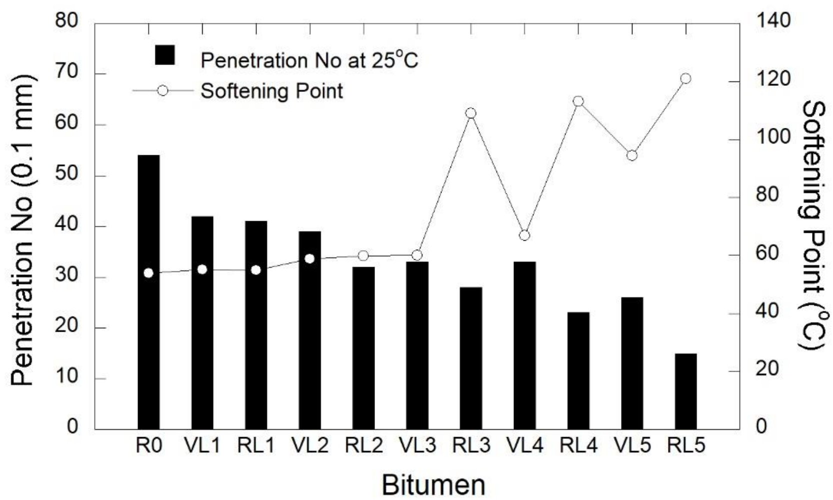


Figure 6. Penetration Number and Softening Point

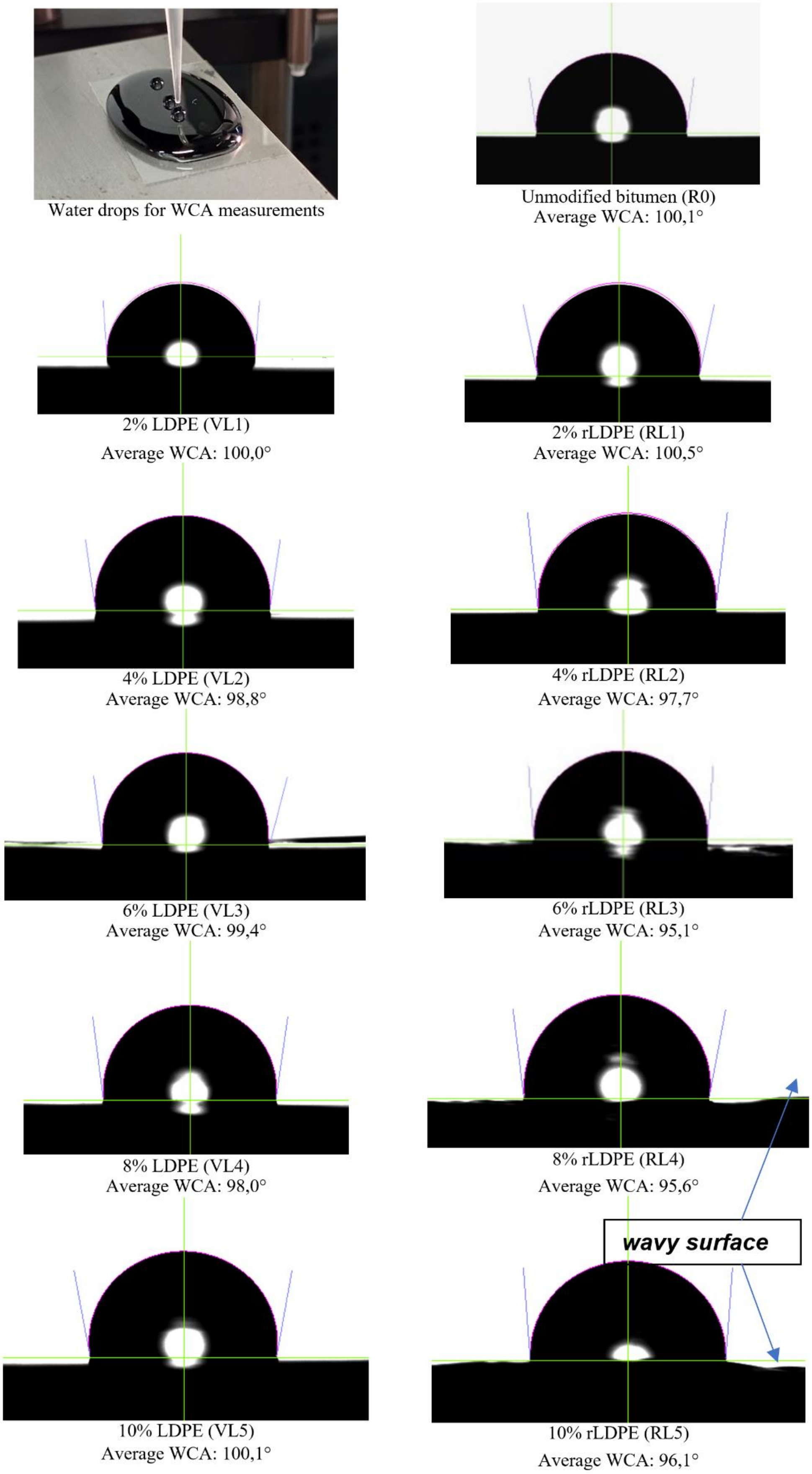


Figure 7. Three Drops of WCA Test and Observed Contact Angle

A consistent result was observed in WCA measured for the two specimens of each type of bitumen. However, very little difference was recorded in the case of 2% modified compared to the unmodified bitumen R0 (R0=100,1 ° compared to VL1=100,0 °and RL1=100,5 °). This indicated insignificant changes in the bitumen surface despite the inclusion of a low amount of LDPE polymer. A further higher modification with VL up until 10% also produced negligible changes. This further confirmed that LDPE modification had an insignificant effect on surface morphology of the material, thereby leading to unnoticeable changes in the hydrophobicity.

Lower number of average WCA was obtained in the case of RL. It was observed that higher modification led to further reduction of WCA. At the same modification rate, RL produced lower WCA compared to VL, and this was possibly due to the significant impact of including RL polymers in both the inner body structure and surface morphology of bitumen. The changes in surface morphology were observed in the spots of non-flat or wavy surface of the specimen. This was discovered to have affected waterdrop and subsequently influenced the measurement. The biased result was minimised by measuring the waterdrops on the flat surface of the specimen. Meanwhile, the flatness factor in the analysis of bitumen specimen in the Goniometer was not covered deeply in this study. Another important observation was that WCA changes were relatively low and the value for all RL was>90 °, leading to their consideration as hydrophobic materials [7]. This showed that lower WCA measured was possibly due to the biased measurement caused by the wavy surface and not because of the changes in hydrophobicity or wettability values of RL-modified bitumen.

3.4. The Effect of the Modification on WCA Results

Lower WCA indicated lesser hydrophobicity of bitumen and this was translated to the reduction in its ability to repel moisture. In other words, the materials tended to be more hydrophilic, which could lead to a higher susceptibility to moisture damage during asphalt application. A hydrophilic or higher-wettability bitumen could cause premature failure of the mixture due to the strong adhesion of bitumen to water after bonding to the aggregates [6]. The changes in WCA value of VL were relatively low compared to the original bitumen R0. Meanwhile, RL showed lower WCA compared to VL at the same modification rate.

Figure 8 shows WCA value as a function of the modification rate. This was indicated by the fact that VL1 and RL1 showed insignificant changes compared to R0 at low modification. Meanwhile, RL consistently had lower WCA compared to VL at the same percentage of modification at higher rates. Similar results of lower WCA or higher wettability were reported by [6] using natural rubber latex (NRL) as bitumen modifier.

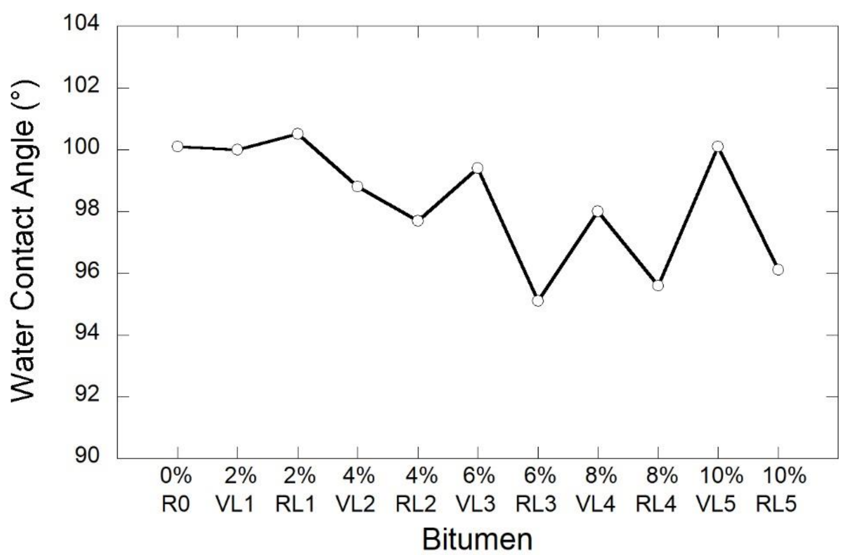


Figure 8. WCA Value Compared To %Modification

The adhesion between bitumen and aggregates is an important factor in determining the mixture's performance during the application of asphalt [20]. However, the potential of stripping occurring due to the presence of water [6] is dependent on the natural chemistry of the aggregates which can be either hydrophobic (non-polar) or hydrophilic (polar). This means the slightly lower WCA measured for RL is possibly better for improving bitumen-aggregate adhesive bonding systems [6,21] due to the changes in the hydrophobic nature of bitumen to slightly hydrophilic. However, WCA of all RL remains in the hydrophobic state and is slightly lower due to biased calculation caused by the wavy surface of the specimen.

Several studies also suggested that bitumen-aggregate bonding was affected by the possibility of changing the interfacial bonding associated with the chemical functional group [6,22,23] but this was not covered in this study. Therefore, there is a need for a study on the intrinsic bitumen-aggregate bonding to conclude the real effect of RL modification on the adhesion between bitumen and aggregates.

4. Conclusions

In conclusion, LDPE polymer-based modified bitumen was able to successfully produce a material with lower penetration number and significantly higher softening point. These standard test results were further confirmed by the changes in the rheological performance reported in previous studies. Moreover, VL showed insignificant changes in WCA for all five types of bitumen while RL had slightly lower values. Several detailed conclusions can be drawn as follows:

- The standard tests were observed to have indicated observable changes in penetration number and softening point of the modified bitumen compared to the original. This was a clear sign of successful modification but further rheological property tests of the modified bitumen could be beneficial to understand the exact changes in the properties.
- WCA of bitumen modified using VL was observed to be relatively unchanged, which indicated the lack of any significant changes in the surface morphology of the material. Meanwhile, those modified using RL showed a noticeable reduction in WCA compared to R0 and VL. This showed that bitumen samples modified using high content of RL had an interesting wavy surface indicating the significant influence of polymers on both the internal structure and surface of the material.
- Lower WCA value measured was probably due to (1) the wavy surface that led to a biased result, (2) the hydrophobicity changes in the material, or the combination of both. These results were unable to answer the cause, thereby indicating the need for further bitumen-aggregate affinity tests to understand the phenomenon.
- Several studies have already reported the possibility of lower WCA causing a higher susceptibility to moisture damage. However, this was not confirmed for RL inclusion due to the influence of other factors such as the different surface flatness of the specimens and the hydrophilicity of bitumen considered beneficial to some aggregates in the mixture. Both modified bitumen, including VL and RL, showed WCA of >90°, which showed that the materials remained hydrophobic and were able to repel moisture.

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