Arabian Journal for Science and Engineering
https://doi.org/10.1007/s13369-021-05413-0

RESEARCH ARTICLE-CIVIL ENGINEERING

Impacts of Maltene on the Wettability and Adhesion Properties of Rejuvenated Asphalt Binder

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Received: 14 August 2020 / Accepted: 28 January 2021

Abstract

In recent years, the use of reclaimed asphalt pavement (RAP) has gained much attention and is widely accepted. However, the rejuvenating agents which are usually used to reduce the rigidity of the aged asphalt are subjected to diverse climate circumstances. The present work used maltene as a rejuvenator to investigate several measurements regarding stripping failure. The evaluation of wettability and work of adhesion (W_A) was assessed using the sessile drop method. Meanwhile, asphalt and asphalt-water aggregate systems were tested for acid and water resistance using chemical and water immersion tests. Next, atomic force microscopy (AFM) was used to evaluate the changes in the microstructures of the asphalt binders. The experimental results revealed that the ideal percentages of maltene which should be added to 30% and 50% aged asphalt were 8% and 16%, respectively. Meanwhile, the wettability, W_A and resistance to stripping differed depending on the percentage of aged asphalt in the blend. However, the inclusion of maltene has improved samples containing high percentages of aged asphalt. On the other hand, the resistance to boiling water containing acid decreased slightly with the addition of maltene. Nevertheless, all the rejuvenated samples exhibited better results than virgin asphalt. Moreover, the AFM results were in line with the observations, suggesting the suitability of maltene for the functional application of pavement.

Keywords Aged asphalt · RAP · Maltene · Moisture damage · Stripping · Contact angle

1 Introduction

The transport agencies and pavement construction companies, together with the asphalt suppliers, have proposed several initiatives to produce green paving technology [1]. The use of reclaimed asphalt pavement (RAP) can be considered as one of the main elements of such initiatives [1] since it is economically and environmentally beneficial [2, 3]. However, RAP binder (aged asphalt) which is subjected to the different conditions can affect the properties of virgin asphalt when mixed at high percentages [4]. It is because from the construction and throughout the service life, RAP is exposed to a combination of traffic loading and environmental influences [5]. Environmental conditions such as fluctuating temperatures, humidity, precipitation, oxygen, freeze–thaw cycles and ultraviolet (UV) radiation can deteriorate the material properties and cause asphalt ageing [6]. Usually, the rheology of the mixture is not affected by the addition of a low percentage of RAP [2]. However, when the level of RAP reaches 20%, the viscosity and asphalt rheology can be affected. In this case, many factors should be considered, such as susceptibility to ravelling, cracking and stripping which can destruct asphalt pavements [7]. The stripping phenomenon is represented by the loss of adhesion as a result of the asphalt film separation from the aggregate interface, or due to the loss of cohesion which occurs in the binder phase [6]. Consequently, the adhesion of asphalt mixture constituents (asphalt and aggregate) and their resistance to the effects of moisture become important in the long-lasting road design [8]. The level of adhesion failure within the asphalt mixture depends on the degree of adhesion between asphalt and aggregate [9]. Furthermore, the quality of the aggregate and asphalt plays a major role when it comes to adhesion strength [10]. Prior studies have reported that the interaction between asphalt and aggregate is mainly

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Published online: 01 March 2021

ascribed to the micro-nano physical and chemical actions on the contact interface [11, 12]. The micro-nano morphology primarily affects the physical actions which include the exertion of mechanical interlocking force between the materials through interlock and anchoring action on the micro-nano interface [11, 13]. Meanwhile, the chemical activities rely on the material micro-surface energy which correlates with the chemical composition of the materials [11]. The interfacial adhesion of the asphalt binder with aggregate determines how much the pavement can withstand fatigue cracking and moisture-induced damage [14].

As regard to ageing, the opinions associated with the impact of ageing on adhesion behaviour and asphalt's moisture sensitivity still vary. Having said that, Ji et al. [15] demonstrated that measurement of surface free energy (SFE) and associated asphalt binder parameters influence the wettability of asphalt to aggregate and its sensitivity towards moisture. Therefore, these elements were used to evaluate the water damage potential in the asphalt mixture [15]. For instance, Zhang et al. [16] evaluated asphalt's water resistance using SFE analysis and revealed that the ratio of SFE component rose as the ageing duration increased. This brought greater stripping work between the asphalt and aggregate. However, since asphalt's polar components increase its vulnerability to water, the asphalt stripping off from the aggregate was increased along with poor water resistance.

Despite the introduction of several methods to characterise the ability of asphalt to maintain contact with aggregate, the contact angle test is the commonest due to its measurement simplicity [17, 18]. The contact angle describes the tendency of a fluid phase to disperse onto a solid surface in the presence of another immiscible fluid phase [18]. It can be determined by the shape of the liquid droplet with defined surface energy [14]. The formation of a spherical drop shape can be affected by several factors such as temperature, surface exchangeable cations, particle size, surface roughness and surface heterogeneity [17, 19]. The higher the contact angle, the lower the wettability and adhesion property. On the contrary, lower contact angle will lead to higher wettability and higher adhesion property. Han et al. [20] investigated the adhesion mechanism and revealed that the contact angle of the asphalt subjected to short-term ageing (STA) was lower than that of unaged asphalt, which reflects the link between high adhesion and ageing. However, the asphalt binder subjected to the long-term ageing (LTA) exhibited a higher contact angle than that of unaged asphalt. The reason can be related to the volatilisation of the small molecular substances in the asphalt after STA. Besides that, the increment in asphaltene content can enhance the polarity and improve the adhesion with the aggregate. Following the LTA, the increased viscosity of the asphalt along with deteriorated fluidity could prevent of the granite surface from moistening by increasing the contact angle. Consequently, the adhesion work deteriorates. Moreover, Yao et al. [21] demonstrated that the ageing of asphalt can decrease the susceptibility to moisture damage in wet conditions. However, the bonding strength of asphalt with aggregates, together with fatigue life and resistance to moisture damage can deteriorate through ageing over time. Ghabchi et al. [1] evaluated the moisture susceptibility of asphalt mixes containing aged asphalt. They concluded that the addition of 10% aged asphalt did not affect the W_A, whereas the addition of 25% aged asphalt and more was beneficial in improving the adhesion between the aggregate and asphalt binder.

On the contrary, Kamaruddin et al. [22] investigated the effects of chemical additive on the wettability of modified aged asphalt with waste engine oil. They reported that the incorporation of aged asphalt into virgin asphalt may result in an increased contact angle. Nevertheless, the addition of chemical additive can decrease the contact angle besides improving the adhesion and cohesion function. Hossain et al. [6] also noticed that as asphalt ages, the work of adhesion (W_A) values drop and the energy required to initiate a cohesive failure is reduced. However, the addition of a rejuvenating agent to aged asphalt can improve the adhesive bonding and the cohesive energy of asphalt binder. Thus, the resistance to moisture damage is improved.

Cao et al. [23] evaluated the effects of cashew shell oil and active isocyanate rejuvenators on the resistance to water damage. Their results revealed that the SFE of polymer modified asphalt decreased after ageing, while the rejuvenators enhanced the SFE. Meanwhile, the work of cohesion (W_C) in aged polymer modified asphalt, together with the W_A between aged polymer modified asphalt and granite increased with the addition of rejuvenators. On the other hand, the water boiling test indicated that the adhesion grade of the modified asphalt on granite reduced from grade 5 to grade 3 with ageing. Nonetheless, the adhesion grade of aged polymer modified asphalt on granite was restored to the original level with the addition of rejuvenator. In a more recent study, Zhang et al. [24] proved that the ageing could increase the W_C and W_A of the virgin binder, while the incorporation of rejuvenators with an appropriate viscosity can lead to a further enhancement in these parameters.

Based on the aforementioned literature, a few studies emphasised the asphalt-aggregate stripping of aged asphalt containing rejuvenating agents. Therefore, the current study was undertaken to evaluate the adhesion behaviour and moisture sensitivity of the aged and rejuvenated asphalt binders, with maltene as a rejuvenator. The tests included contact angle, static and total water immersion tests as well as the chemical immersion test. The surface morphology and micromechanical characteristics of the binders were also investigated via Atomic Force Microscopy (AFM).

2 Experimental Procedure

2.1 Materials

Pen. 60–70 asphalt provided by Kemaman Bitumen Company (KBC) Malaysia was chosen as the virgin asphalt, whereas the virgin aggregate (granite) was obtained from stockpiles at Hanson Quarry in Johor Bahru, Malaysia. The RAP was procured from a stockpile of pavement materials reclaimed through milling of the Yong Peng highway heading to Pagoh, Malaysia. The aged asphalt was separated from the RAP using trichloroethylene and a centrifuge extraction method as per the ASTM D2172 [25]. Then, the asphalt binder was recovered using a rotary evaporator as per the ASTM D5404 [26]. The physical properties of the virgin and aged asphalt binders are displayed in Table 1

The rejuvenator employed in this study was maltene. It consists of 56.26% aromatics, 35.1% resins and 8.3% saturates. These values were calculated using ASTM D4124 [27]. On the other hand, the viscosity of maltene was estimated at 42.97 mPa.s at 95 °C with a density of 0.955 gm/cm^3 at 20 °C. The energy-dispersive X-ray (EDX) analysis of maltene (Fig. 1) recorded high content of carbon element (93.6%) along with other chemical elements such as sulphur (5.5%) and oxygen (0.8%). However, hydrogen and nitrogen were not detected due to EDX limitations, as it only detects elements present on the surface of maltene.

2.2 Preparation of Samples

The proportions of RAP binder (aged asphalt) used in this study were 30% and 50% by weight of the total mass. Based on the physical and rheological properties tests performed in this study, the optimal doses of maltene required to rejuvenate the blend containing 30% and 50% of aged asphalt were 8% and 16% by weight of the total asphalt binder, respectively. At these dosages, the penetration, softening point, ductility and viscosity values of the rejuvenated asphalt binders were similar to that of the penetration grade 60–70 virgin asphalt (see Table 2). Previous studies also proved that the percentage of rejuvenating agents that improves the rheological and physical properties of aged asphalt is considered as the ideal rejuvenator content [28–30]. Table 3 provides the types of samples prepared in this study.

Table 1 The characteristics displayed by virgin and aged asphalts

Properties	Virgin asphalt	Aged asphalt	Standard method
Density (gm/cm^3)	1.02	1.03	ASTM D-70
Penetration (dmm) at °C	64	18.5	ASTM D-5
Softening Point (°C)	51.5	73	ASTM D-36
Ductility (cm)	116	9	ASTM D-113
Viscosity @ 135 °C (mPa s)	650	3500	ASTM D-4402
Viscosity @ 165 °C (mPa s)	200	700	ASTM D-4402

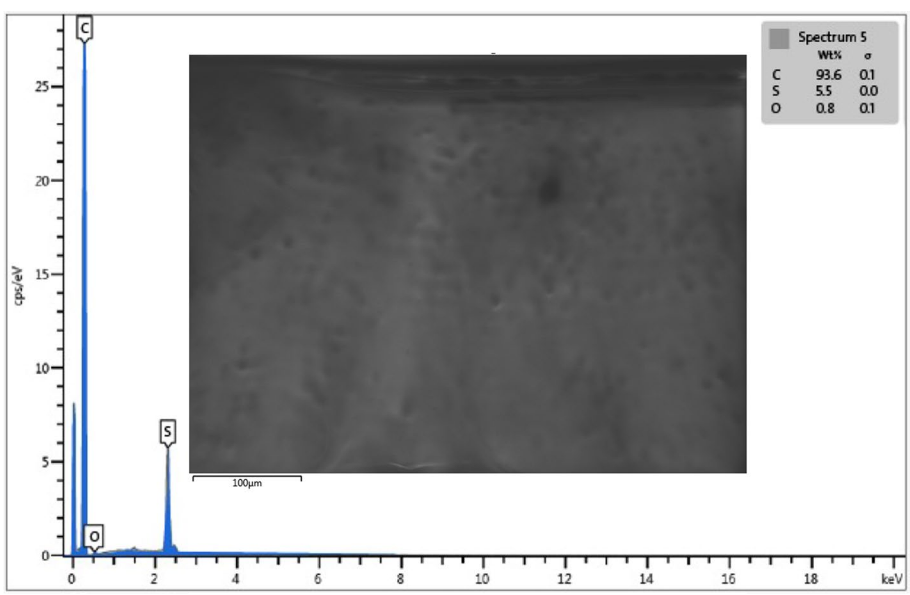

Fig. 1 EDX analysis of maltene

Table 2 Penetration, softening point, ductility and viscosity results of asphalt binders

Types of asphalt	Penetration (dmm)	Softening point (℃)	Ductility (cm)	Viscosity (mPa s)	
				At 135 ℃	At 165 ℃
VA	64	51.5	116	650	200
R30	44.6	59	78	1400	350
R30-4M	52.7	55.7	97	950	300
R30-8M	63.1	51.7	119	625	200
R30-12M	74.7	48	137	350	125
R30-16M	82	42.5	149	250	100
R50	31.6	63.5	64	1725	450
R50-4M	37.8	60.3	80	1450	400
R50-8M	44.2	57	97	1100	325
R50-12M	53.8	55.5	110	925	275
R50-6M	62.9	51.3	125	575	183

Table 3 The types of asphalt binders used in this study

Types of asphalt	Asphalt binder details
VA	Virgin asphalt binder, penetration grade (60–70)
R100	100% aged asphalt binder
R30	30% aged asphalt + 70% virgin asphalt
R30-8M	(30% aged asphalt + 70% virgin asphalt) + 8% maltene
R50	50% aged asphalt + 50% virgin asphalt
R50-16M	(50% aged asphalt + 50% virgin asphalt) + 16% maltene

2.3 Tests

2.3.1 Contact Angle and Work of Adhesion (W_A) Measurements

Wetting refers to a liquid's proclivity to maintain contact with a solid surface stemming from intermolecular reactions when the solid and liquid are mixed [7]. Figure 2 shows that the measurement of contact angle (θ) is undertaken by calculating of the tangent's slope to the droplet (at the interface of solid and liquid) for the purpose of mathematically define droplet shape. It is possible to balance the extent of wetting by utilising both cohesive and adhesive forces [31]. If the surface energy of the substrate is relatively high but lower than the surface tension of the liquid, the liquid will wet the solid surface. The contact angle (θ), hence, would be less

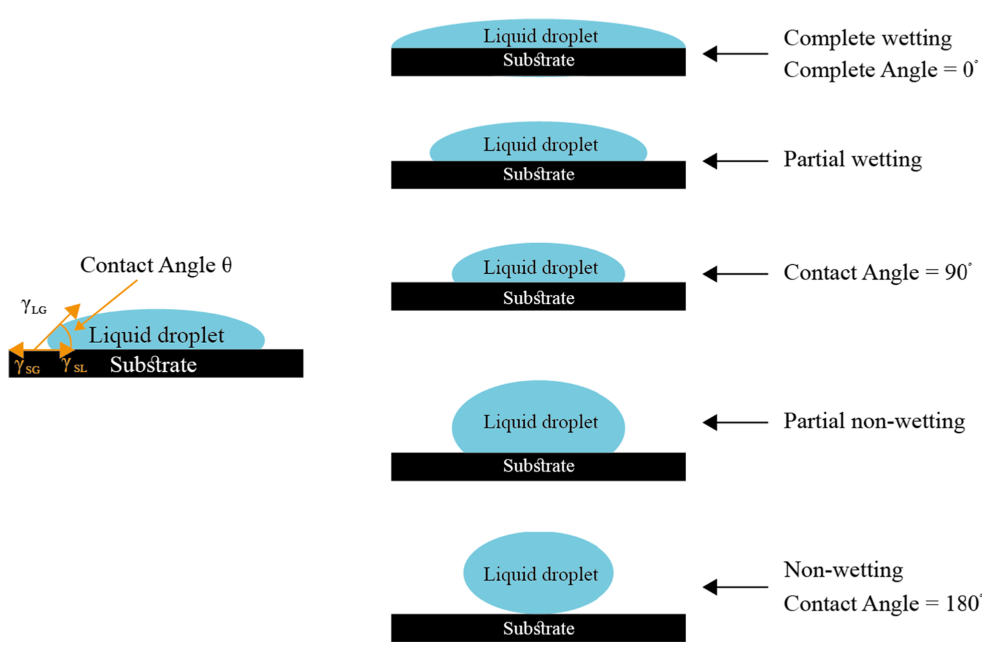

Fig. 2 The cases of liquid droplets on a solid substrate [31]

than 90° and higher than 0°. Contrastingly, in the opposite scenario, the liquid would exhibit inadequate adhesiveness and would be unable to wet the solid surface in an effective manner. This explains why the contact angle would exceed 90°. In this paper, the measurement of contact angle was undertaken by utilising the method of sessile drop, which has been extensively mentioned in extant literature through its application in myriad fields, including geology, painting, mining, coating, and chemistry, among others [9, 16, 20].

Prior to this test, the asphalt samples were heated in an environment free of dust at a temperature of 150 °C before being placed onto a glass slide. Thereafter, the holder was covered for preventing exposure to dust by keeping it horizontal for 24 h at 25 °C. In the second stage, the liquid was sucked into the injector, subsequent to which the bubbles were eradicated in entirety. Then, the syringe was loaded in the contact angle tester. The glass slide was placed on top of an adjustable platform, subsequent to which adjustments were made in the focal length and the sample's position to ensure that the quality of the image remains high. The contact angle between the surface of asphalt and the water droplets was determined at 25 °C through the VCA Optima device shown in Fig. 3. In order to assess the changes in wettability, the data were saved after being processed with a view to assess the polarity properties of various kinds of asphalt binders. Three different samples were sourced from each group, with each sample being tested using three points to ascertain the accuracy of the findings. The adhesion work was measured by formula (1) [32] to measure the bond strength between asphalt binder and aggregate.

$$W_{\mathrm{A}} = \gamma_l(1 + \mathrm{COS}\theta) \quad (1)$$

γ_l Surface energy of liquid (72.8 mj/m^2) [33, 34], θ contact angle, W_{A} work of adhesion.

Fig. 3 Contact angle measurements of asphalt surface

2.3.2 Water Immersion Tests

2.3.2.1 Static Immersion Test This test was undertaken to determine the stripping and binding forces between asphalt and aggregate in compliance with AASHTO T182 [35]. Approximately 100 g of loose asphalt mixture sample was immersed into a 500 ml glass bottle containing distilled water for 16 to 18 h at 25 °C. The loose mixture was then placed onto a wet paper towel and left to cool at room temperature. Next, the mixture was visually analysed to determine the amount of asphalt binder coating maintained on aggregate surfaces. The percentage of the total visible coated aggregate should be higher than 95%, and the mixture is deemed as a failure if it does not meet the criteria of these measures. Thus, it should be avoided because it can be susceptible to stripping [36].

2.3.2.2 Total Water Immersion Test It is a modified form of the static immersion test [37]. It uses distilled water at 40 °C instead of 25 °C to achieve improved performance. The test aimed to assess the percentage of the asphalt-coated aggregate after the sample was submerged in water at 40 °C for 3 h.

2.4 Chemical Immersion Test

The chemical immersion test was conducted to assess the bonding strength between the asphalt and aggregate under the influence of sodium carbonate (Na_2CO_3) and boiling water as per Road Research Laboratory (RRL)-England [38]. The Riedel and Weber (R&W) number refers to the number at which the asphalt starts to separate from the aggregate interface (stripping phenomenon). This test required immersing 10 g of loose asphalt mixture in 50 ml of distilled water with the inclusion of Na_2CO_3, and boiling the mixture for 60 s. The procedure started with 0.41 g of Na_2CO_3/L (signified by No. 1), whereby the concentrations regularly increased until stripping took place. The maximum amount of Na_2CO_3 being 106 g/L of distilled water was denoted by No. 9.

2.5 Atomic Force Microscopy (AFM)

The AFM imaging technique was used to identify morphology and micromechanical changes caused by ageing and rejuvenation processes. Three scanning modes which can be used in the AFM experiments include contact, intermittent contact and non-contact modes. This study employed the non-contact mode using a Smart SPM 1000 scanning probe microscope AIST-NT. Using this mode, the sample surface will not come into contact with the probe tip. This mode can restrict the normal and lateral forces of asphalt, making it suitable to examine materials characterised by viscosity and softness. Meanwhile, the preparation for AFM test involved heating different asphalt samples at 125 °C to melt them to facilitate pouring. A 1 cm droplet was obtained by dropping a heated asphalt bead onto a glass slide. The holder was subsequently covered to prevent exposure to dust and was maintained horizontally at 25 °C for at least one day prior to AFM analysis. The morphology of the asphalt binders was determined at 300 kHz drive frequency and a 0.5 Hz scan rate at room temperature and atmospheric pressure. As a result, 50×50 µm AFM images were acquired to observe the microstructure of asphalt binders.

3 Results and Discussion

3.1 Contact Angle (θ)

The contact angle between the distilled water and the asphalt differed depending on the adhesive strength between the asphalt sample and the test liquid. Figures 4 and 5 depict the

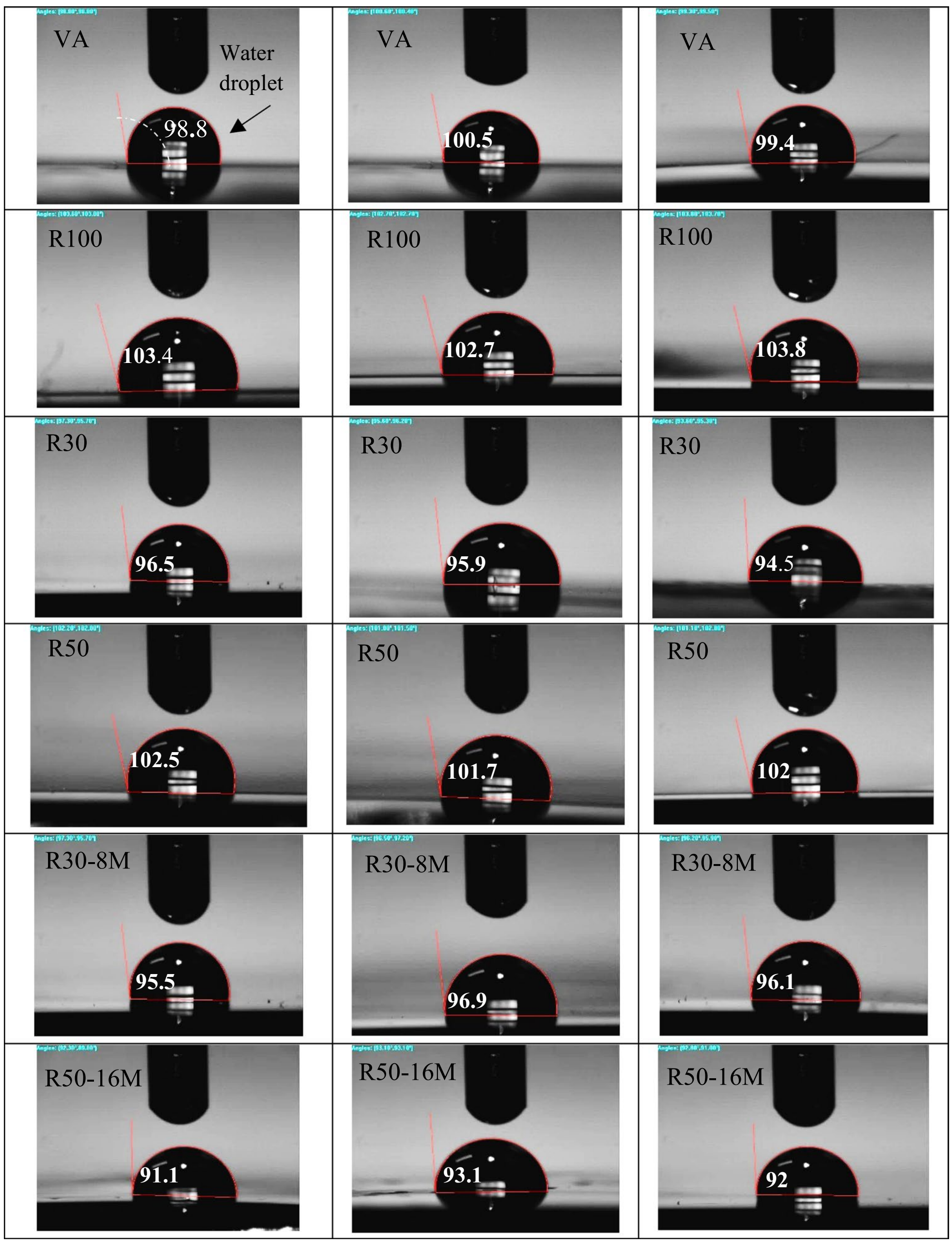

Fig. 4 Contact angle results of the specimens

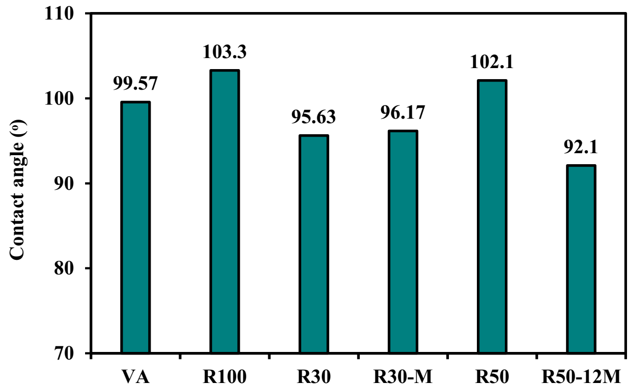

Fig. 5 The average contact angle values

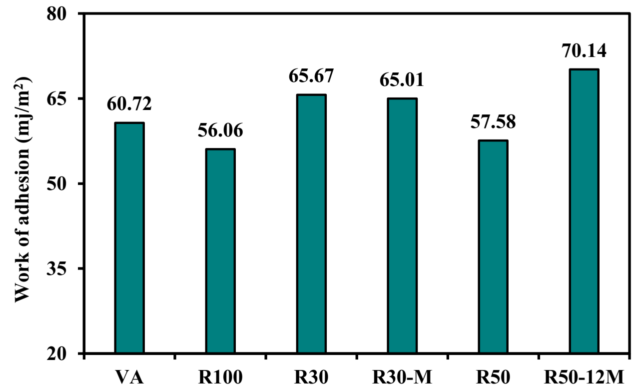

Fig. 6 Work of adhesion results

contact angle (θ) values obtained from the software. Based on the observation, the addition of 30% the aged asphalt to the blend caused the contact angle of virgin asphalt (VA) binder to decrease from 99.57° to 95.63°. The decline in contact angle corresponded to an increase in the wettability of asphalt. This phenomenon is related to the volatilisation of small molecular substances in the aged asphalt during the service life. Furthermore, when asphalt ages, the light chemical components inside it will transform into heavy chemicals, leading to the increased ring-like structures that are packed and condensed, such as asphaltene [39]. This condition can lead to an increment in the ratio of C=O and S=O content to the C–H, and thus improve the adhesion of asphalt with the aggregate.

On the contrary, R50 and R100 exhibited the highest contact angle values compared to the other samples, 102.1° and 103.3°, respectively. These results can be attributed to the loss of cohesion related to the presence of high percentages of aged asphalt. The findings were consistent with the results discussed by Kamaruddin et al. [22], who demonstrated that the blend with high percentages of aged asphalt was vulnerable to stripping.

Meanwhile, the addition of maltene to R30 did not show a significant trend, where the contact angle value of R30-8M was 96.17°. This value was higher than that of R30 by 0.54°. Conversely, the addition of maltene to R50 reduced the contact angle, where the contact angle value of R50-16 M dropped to 92.1° along with increased W_A. The recorded contact angle was less than R50 by 10°, and less than VA by 7.74°. These results indicated that R50 and R100 have a lower grade of binder for highway applications due to the possibility of stripping. The reduction in the contact angle of the rejuvenated binders is attributed to the presence of resin in maltene. A recent study reported that the ratio of resins/asphaltenes, colloid index and resins can exert positive effects on the SFE of the asphalt binders [40]. Typically, SFE increases with increasing wettability. Meanwhile, the elemental composition such as carbon (C), nitrogen (N), hydrogen (H) and sulphur (S) of asphalt binders were sensitive to the SFE parameters [41]. Therefore, it can be deduced that the increase or decrease in contact angle is dictated by multiple factors, such as the asphalt binder composition, its fraction contents (saturates, aromatics, resin, and asphaltene), the degree of oxidation, and the type of rejuvenator incorporated.

3.2 Work of Adhesion (W_A)

Figure 6 presents the results of the W_A, which is the energy required to separate an asphalt binder from aggregate interface [1]. Based on the data, the incorporation of 30% aged asphalt within the mix can increase the W_A. The W_A of R30 was 65.67 mj/m^2 along with the VA value of 60.72 mj/m^2. A higher W_A value is desirable to strengthen the bond between the asphalt binder and aggregate [1]. In other words, the larger the W_A of asphalt, the stronger ability to resist moisture damage [42]. On the other hand, electrostatic energy is the main component that contributes to an increase in adhesion energy, thereby strengthening the bond between asphalt and aggregate in the presence of oxygenated groups (ketone and sulphoxides) [43]. The ageing groups are highly active in the formation of hydrogen bonds with surface hydroxyl groups [43]. However, this enhancement of adhesion was decreased with the inclusion of a high percentage of aged asphalt in the mix (50% and more) which in turn negatively affected the W_A to be lower than that of VA. Therefore, R50 and R100 binders are vulnerable to stripping since they exhibited lower W_A (57.58 mj/m^2 and 56.06 mj/m^2, respectively). This means that the asphalt binder 's characteristics were deteriorated and caused more severe damage with the addition of very high percentage of aged asphalt. Nevertheless, the use of maltene as a rejuvenator compensated the negative effects of aged asphalt and improved the adhesive bond strength of the aged asphalt samples. Particularly, the

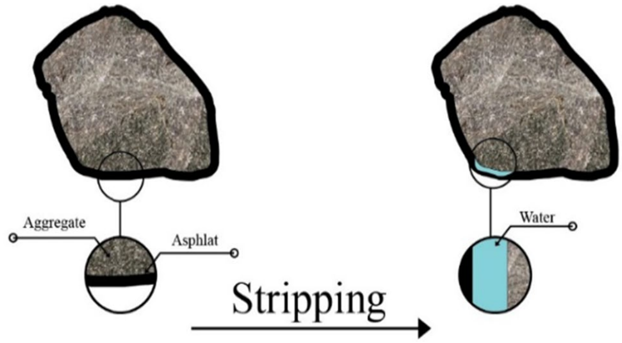

Fig. 7 Stripping phenomenon [48]

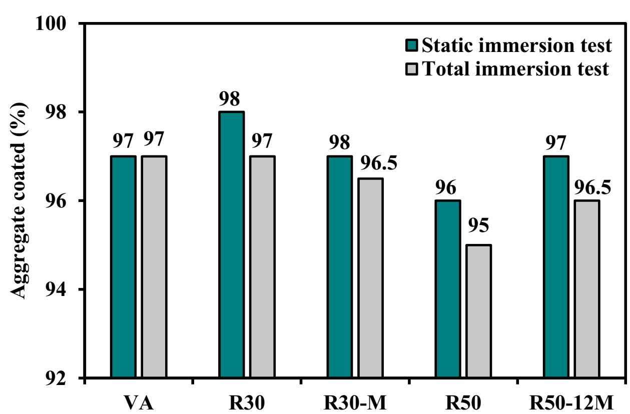

Fig. 8 Water immersions tests results

W_A of R50-16M yielded 70.14 mj/m^2 compared to 57.58 mj/m^2 before maltene addition. However, the addition of maltene to R30 did not result in a significant effect.

3.3 Water Immersion Tests

A loss of adhesion between the asphalt film and aggregate surface can be hastened when water penetrates the asphalt films and accesses the partially coated aggregate (Fig. 7). Figure 8 demonstrates that the majority of the asphalt-coated aggregate samples exhibited high stripping resistance (> 95%). The exception was R50, which had 95% aggregate-binder adhesion following the total water immersion test. In the case of VA, the aggregates remained coated at a rate of 97% (at both static and total immersion tests), while the coating was maintained at a rate of 98% after the static immersion test and 97% after the total immersion test for R30. The adhesive properties between asphalt and aggregate were closely related to polar chemical groups content [42]. More specifically, the resistance of R30 to water damage was caused by an increase in the C=O and S=O (which are considered as polar groups) and the decrease in the C–H functionalisation to a certain extent during asphalt oxidation [21]. In contrast, R50 exhibited stripping resistance of 96% after static immersion test and 95% after total immersion test. The decline in the moisture damage value of asphalt containing a very high percentage of the aged asphalt (R50) was caused by the chemical and microstructural changes that occurred during ageing, hence leading to an increase in the polar compounds to extremely high levels. The other reason is the addition of 50% aged asphalt, which stiffened and reduced the bonding between aged and virgin materials, adversely affected the moisture resistance. Moreover, the improper blending between the aged and virgin materials play significant role as well [44, 45]. Having said that, LTA is also known to harden asphalt content and facilitate the occurrences of micro-cracks which in turn increase the invasion of moisture and water damage [43]. Thus, it can be concluded that the appropriate percentages of aged asphalt in the mix should be considered to ensure the resistance of the mix to water damage. Moreover, the chemical composition of aggregate might play a fundamental role in moisture damage in HMA. This outcome indicated that the adhesion can be controlled by electrostatic forces when appropriate doses of aged asphalt are used in the mixture. The adhesion also depends on the mechanical behaviour of the material, especially when a very high percentage of aged asphalt is used. Similarly, Aguiar-Moya's et al. [46] demonstrated that adhesion is controlled by electrostatic forces during STA, but depended on the mechanical behaviour of the materials in LTA. Meanwhile, the findings obtained from the inclusion of the aged asphalt into the mix were in agreement with the recent study by Al-Saffar et al. [47], who revealed that the production of HMA mixture using 100% aged asphalt can produce a poor bonding strength between asphalt and aggregates, thus increasing the susceptibility to moisture.

On the other hand, the effects of adding 8% maltene to R30 was very low. Whilst the addition of 16% maltene to R50 regained the portion lost from ageing, further indicating significant improvement in the resistance to moisture damage based on the static and total immersion tests. In short, the rejuvenated asphalt binders (R30-8M and R50-16M) recorded aggregate coated values close to that of VA at the end of the water immersion tests. The improvement in moisture damage resistance following the addition of maltene to R50 was related to the presence of resin, a component of the maltene which behaves as a bridge between asphalt and aggregates to resist the displacing effects of water and improve the adhesion of asphalt on the aggregates. Moreover, the addition of maltene activated the aged asphalt, which improved the blending between the aged and virgin materials as a result of softness improvement. An appropriate viscosity of asphalt binder can also increase the stickiness between the aged asphalt and aggregates. In other words, the asphalt mixtures resistance to water damage is influenced by the adhesive strength between the asphalt binder and the aggregate under dry and wet conditions [9]. However, it is essential to mention that the softening and rejuvenating agents may interact differently with aged asphalt depending on the several factors such as binder source, rejuvenator type, and aggregate types [6].

3.4 Chemical Immersion Test

Acid rain, which is associated with urbanisation and industrialisation, can affect the asphalt-aggregate interface. Therefore, this test was performed to understand how asphalt adhered to aggregates with the presence of Na_2CO_3. Table 4 illustrates that increasing percentage of aged asphalt can also increase the resistance towards the effects of acid and boiling water. Based on the three replicate tests for every sample, the average failure was 7 for R50 and 6 for R30. The failure values varied as the asphalt binder formulation varied. Moreover, the failure points of aged samples declined with the addition of maltene, where the R&W numbers for R30-8M and R50-16M were 5 and 6, respectively. However, all the values were still higher than VA which failed at 4. The separation of asphalt film from the surface of the aggregate after test could be viewed in Fig. 9.

These unpredictable results can also be clarified in a number of ways. The existence of inorganic groups in the virgin asphalt reinforces the hydration interactions, where these groups also react with NA_2CO_3 [49]. Meanwhile, the hydration and sodium compounds had a low impact on aged asphalt due to the interactions of double bonds (C=C) and inorganic groups which occurred throughout the lifespan of pavement [49]. Typically, the oxygen, heat, humidity, ultraviolet light, tire pressure and lifespan can cause the majority of the functional groups to react, leading the asphalt binder to breakdown.

3.5 Atomic Force Microscopy (AFM)

Figure 10 illustrates the microstructure features of virgin, aged and rejuvenated asphalt binders. Three phases (catana, peri and para) were observed in the asphalt binder. The catana phase represented the archetypal bee structure (rigid phase), while the peri phase was peripheral to the catana phase. Lastly, the para phase was adjacent to the peri phase (soft phase) [50, 51]. Nonetheless, the software employed

Table 4 Chemical immersion test results

NA_2CO_3 concentration	R&W number	VA	R30	R30-8M	R50	R50-16M
1/1	9	–	–	–	–	–
½	8	–	–	–	–	–
¼	7	–	–	–	Fail	–
1/8	6	–	Fail	–	√	Fail
1/16	5	–	√	Fail	√	√
1/32	4	FAIL	√	√	√	√
1/64	3	√	√	√	√	√
1/128	2	√	√	√	√	√
1/256	1	√	√	√	√	√
Distilled water	0	√	√	√	√	√

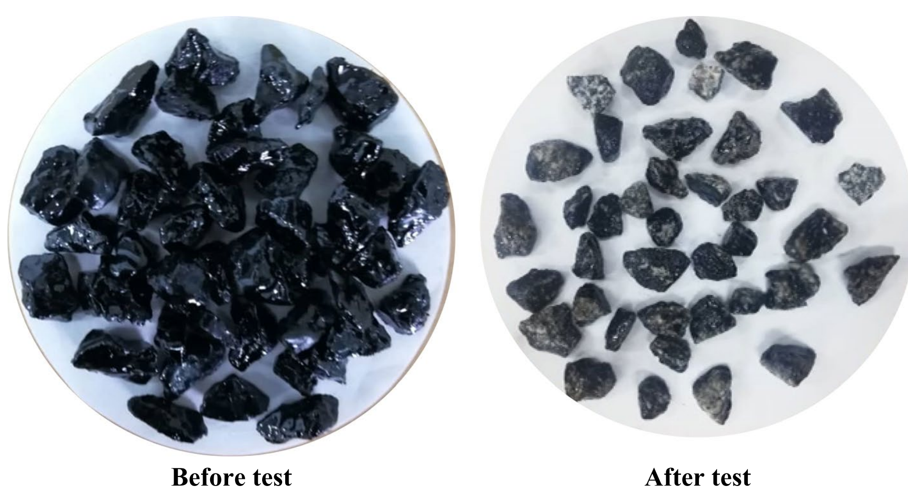

Fig. 9 Asphalt specimen before and after stripping

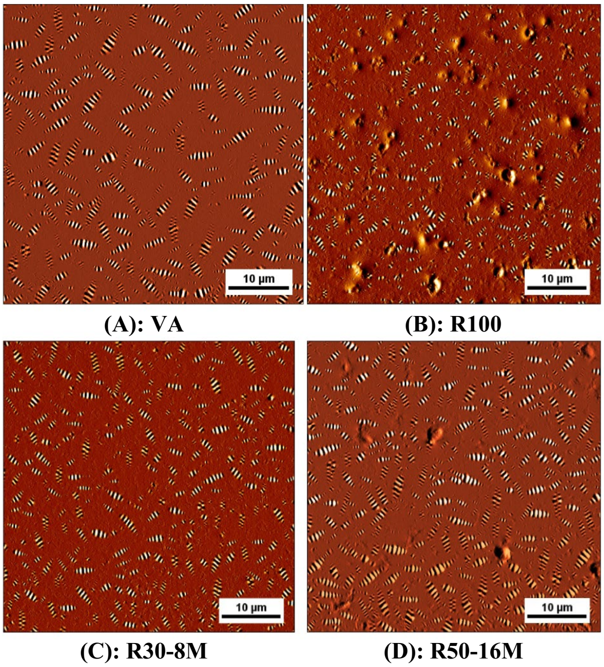

Fig. 10 AFM images of **a** VA, **b** R100, **c** R30-8M and **d** R50-16M samples (Scanned area: 50×50 µm^2)

in the present study was unable to distinguish the peri phase from the para phase in the images.

In the present study, the aged asphalt binder displayed large numbers of small fine-sized bee structure compared to virgin asphalt. Besides that, the protrusions and cavities were also randomly scattered in the matrix referring to the stiffness of the binder. According to the previous study, microcrystalline asphaltene and wax in asphalt are responsible for the formation of the "bee-like" structure [52]. Nevertheless, Aguiar-Moya et al. [53] reported that "bee-like" structure can be formed due to the presence of resins. The other fractions (like saturates, aromatic and asphaltene) in asphalt can play a significant role in the structural shapes, sizes, and topographies of asphalt [54].

The incorporation of maltene in aged asphalt samples led to significant changes in their morphology, where R30-8M and R50-16M images looked similar to those of virgin asphalt. In particular, R30-8M exhibited a big-sized bee structure with fewer numbers compared to the aged asphalt, whereas R50-16M demonstrated biggest bee structures in terms of size besides the presence of fewer protrusions. The change in the outer layer of the dispersed phase (bee-like structure) and continuous phase can be attributed to the different content of resin, saturates and aromatics in the asphalt binders. When the asphalt ages, some of its light components are polymerised or volatilised [55], followed by the transformation of the aromatics and saturates into resins, then the transformation of the resin into asphaltene [56]. In other words, the chemistry of asphalt and rejuvenator determine the microstructure of asphalt. Moreover, the finding obtained from AFM analysis was in agreement with a previous study by Gong et al. [57]. They revealed that following the ageing of asphalt, the bee structures increase greatly with significant size reduction and the formation of numerous black dots in the centre of the bee structures. The authors attributed this phenomenon to the increment in the viscosity of aged asphalt. After inserting the rejuvenating agent, the black dots in the rejuvenated asphalt decreased, while the bee structure size increased.

Based on results, it can be concluded that the inclusion of maltene as a rejuvenator yielded positive effects on the rheological and morphological properties of the aged asphalt by mitigating the aggregation of highly-oxidised components. Besides that, the diffusion ability of asphalt molecules was improved, whereby the properties of the rejuvenated asphalt were completely restored to that of the virgin asphalt. Nevertheless, the efficiency of the rejuvenating agent should be measured on the real performance of the rejuvenated asphalt, apart from its microstructure indices. This assumption is also in agreement with a previous study by Chen et al. [55].

4 Conclusion

The present study evaluated the wettability and stripping resistance of aged and rejuvenated asphalt binders. Based on the testing and analysis presented herein, the following conclusions are drawn:

1. The results demonstrated that the contact angle and W_A of asphalt binder were affected by the percentage of aged asphalt in the mixture. R30 exhibited better wettability and work of adhesion compared to R50 and R100.
2. The inclusion of maltene indicated a positive effect on the wettability and adhesion work of asphalt blend containing a very high percentage of the aged asphalt (R50).
3. The water and chemical immersion tests further revealed that compared to the virgin asphalt, the rejuvenated asphalt binders were less vulnerable to the impact of water and acids.
4. The AFM image analyses of the rejuvenated samples displayed improvement in the microstructure features of aged asphalts confirming with results of contact angle and water damage tests.
5. The results obtained in this study revealed that maltene could be used in industrial applications to increase the wettability and water damage resistance of binders containing high percentages of aged asphalt.

Funding This work was funded by the Universiti Teknologi Malaysia through the Fundamental Research Grant Scheme (Grant No. R.J130000.7851.5F019).

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