Characterization of hydrophobic-treated recycled paper mill sludge in bituminous materials

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Abstract. The experimental investigation of the performance of hydrophobic-treated recycled paper mill sludge (RPMS) incorporated into asphalt mixtures is presented in this paper. This research implements RPMS as a solid waste additive to partially replace the mineral filler in the asphalt mixture while practicing green asphalt technology. The raw RPMS required mechanical pre-treatments and its hydrophilic property was modified chemically. The hydrophobicity was assessed by Hydrophilic Coefficient, Water Contact Angle (WCA) and Scanning Electron Microscopy (SEM). The ethanol method, which involved the esterification of ethyl esters that utilized 7ml of waste cooking oil (WCO) and 50ml of ethanol, was adopted. In the Marshall mix design, RPMS was incorporated at 0.5% and 1.0% of the weight of aggregates. Conventional 60/70 PEN bitumen and granite aggregates were used. The optimum binder content (OBC) was evaluated and justified by its adsorption strength. The mechanical properties of asphalt mixtures were determined and compared with the Public Work Department (PWD) specifications. All the volumetric properties satisfied the standard specification by PWD for 0.5% modified RPMS asphalt mixture, and thus it is preferable as it also involved lower binder cost due to the lower OBC achieved.

1 Introduction

In the past decades, Malaysia has been experiencing remarkable growth in population, urbanization, and industrialization, which consequently reflected in the significant amount of solid waste generated [1], which then caused a scarcity of available landfill capacity as it is the primary disposal method [2]. Therefore, an increasing number of pavement engineers or researchers have begun to focus on asphalt modification with waste materials [3] such as plastic waste, recycled polymer, rubber waste, steel slag, glass waste, fly ash, cement kiln dust, asphalt shingle, bio-oil, construction and demolition waste, rice husk and straw, paper waste, waste cooking oil, food waste, cigarette butts, etc. [3,4,5,6]. Likewise, the significant amount of sludge waste generated by paper mill industries after several phases, such as
sorting, pulping, screening, cleaning, deinking, refining, colour stripping, and bleaching processes [7] also causes environmental consequences. As a result, this waste, known as recycled paper mill sludge or RPMS, which is composed of fibrous recycled paper, chemical additives, organic matter originating from cellulose fiber (from wood or recycled paper), and inorganic compounds (mainly consisting of calcium carbonate, kaolinite, and talc), has also been utilized as a filler portion for asphalt mixture production.

However, the hydrophilic properties of RPMS can cause premature damage to the asphalt mixtures due to moisture-induced damage mechanisms. The low indirect tensile strength of the asphalt mixture with incorporated RPMS induced poor water resistance due to the presence of water-soluble mineralogical compounds, namely anhydrite, halite, sylvine, quartz, and periclase [7]. Moisture percolated within the macropores and existing cracks and impaired the short- and long-term pavement performance, leading to high moisture susceptibility during cyclic loading moisture damage acceleration [8, 9]. Hence, it is crucial to develop mitigation actions to improve the pavement and prolong its service life.

A longer life expectancy asphalt mixture is notable for sustainable consumption and production (SCP), stated in the eleventh Malaysia plan by the Economic Planning Unit [10]. From this concern, efficient use of natural resources such as better curing materials and practices should be adopted for sustainable pavement. Therefore, the use of recycled waste materials to substitute the use of virgin materials should be promoted to minimize the exploitation of natural resources, lower energy demand and greenhouse gas emissions, as well as improve safety and health.

In this study, hydrophobic-treated RPMS was used as the asphalt modifier. Hydrophobic indicates the surfaces and structures of molecules having the property of repelling water and hence reducing water or dissolved substances absorption, which could affect any material's structural integrity. For this reason, hydrophobic treatment can enhance the resistance to moisture damage [11]. From the literature, there are a lack of studies that implement treatment on the fillers to improve their moisture susceptibility. Therefore, the asphalt modifier RPMS is required to undergo a prerequisite treatment method by introducing hydrophobic treatment and the mixtures will undergo laboratory assessments to determine their physical and mechanical properties.

### 2 Materials and methods

#### 2.1 Materials

**2.1.1 Asphalt binder**

A conventional asphalt binder with a 60/70 penetration grade was used and its physical performance was evaluated through several property tests and analysed in accordance with Malaysia's Public Work Department (PWD). The results are shown in Table 1.

<table>
<thead>
<tr>
<th>Properties</th>
<th>Test Results</th>
<th>Specification Range</th>
<th>Standard Specification</th>
</tr>
</thead>
<tbody>
<tr>
<td>Penetration (dmm)</td>
<td>62</td>
<td>60-70</td>
<td>ASTM D5</td>
</tr>
<tr>
<td>Penetration index</td>
<td>-1.78</td>
<td>-2 to +2</td>
<td>-</td>
</tr>
<tr>
<td>Softening point (s)</td>
<td>46</td>
<td>46-57</td>
<td>ASTM D36</td>
</tr>
<tr>
<td>Ductility (cm)</td>
<td>&gt;100</td>
<td>&gt;100</td>
<td>ASTM D113</td>
</tr>
<tr>
<td>Specific gravity</td>
<td>1.034</td>
<td>1.01 to 1.06</td>
<td>ASTM D70</td>
</tr>
</tbody>
</table>
2.1.2 Aggregates

Granite aggregates were used and subjected to physical tests to assess their physical strength and proper gradation. The gradation was referred to the Public Works Department of Malaysia’s Standard Specification Road Works with a 14 mm maximum aggregate size (AC14-JKR/SPJ/2008). The properties were required to conform with Malaysian Public Work Department specifications of version 2008 as listed in Table 2.

<table>
<thead>
<tr>
<th>Properties</th>
<th>Test Results (%)</th>
<th>Standard Specification (%)</th>
<th>Standard</th>
</tr>
</thead>
<tbody>
<tr>
<td>Los Angeles Abrasion Value (LAAV)</td>
<td>23.07</td>
<td>&lt; 25</td>
<td>ASTM C131</td>
</tr>
<tr>
<td>Aggregate Crushing Value (ACV)</td>
<td>23.26</td>
<td>&lt; 25</td>
<td>BS 812-110</td>
</tr>
<tr>
<td>Flakiness Index (FI)</td>
<td>22.81</td>
<td>&lt; 25</td>
<td>BS 812-105.1</td>
</tr>
<tr>
<td>Elongation Index (EI)</td>
<td>31.19</td>
<td>&lt; 25</td>
<td>BS 812-105.2</td>
</tr>
<tr>
<td>Water absorption of coarse aggregate</td>
<td>0.635</td>
<td>&lt; 2</td>
<td>AASHTO T85</td>
</tr>
<tr>
<td>Water absorption of fine aggregate</td>
<td>1.825</td>
<td>&lt; 2</td>
<td>AASHTO T84</td>
</tr>
</tbody>
</table>

2.1.3 RPMS

RPMS was obtained from a local company in the northern state of Peninsular Malaysia. The raw RPMS was oven-dried at 105°C for 24 hours to remove all the moisture content. The weight after the oven-dried process was weighed and the loss of weight was indicated as the moisture content. The oven-dried RPMS in Fig. 1 (b) was granulated, ground, and sieved prior to further implementation. The particles that were retained in the 75 micron and pan sieve sizes were used as mineral fillers in asphalt mixtures. Their physical properties were determined prior to the hydrophobic treatment.

![Raw RPMS](image1)

![Oven-dried RPMS](image2)

Fig. 1. The appearance of RPMS.

2.2 Hydrophobic treatment of unmodified RPMS

2.2.1 Utilization of ethanol method

Waste cooking oil was subjected to ultrasonic transesterification to attain a uniform dispersion of CNC and minimize the oil-to-alcohol ratio [12]. This treatment method performed a primary transesterification of WCO into fatty acid ethyl esters (WCO-FEEs), which was commenced by adding 7ml of WCO to 50ml of ethanol and sonicating at room temperature for 15 minutes. The mixture was then mixed with a ratio of 2:1 to distilled water at 70°C for 5 minutes and stirred at 300 rpm. After mixing, crude glycerol was
separated using a separatory funnel and 5g of grounded RPMS was added to the WCO fatty acid ethyl esters (WCO-FEEs) for second transesterification by attaching FEEs to hydroxyl groups chemically on the CNC surface. The mixture was stirred and sonicated at room temperature for 15 mins and then left for overnight drying at room temperature without closing the lid. The mixture was then oven-dried at 110°C for 2 hours to dry the ethanol. The dried samples were centrifuged 3 times with 50ml of ethanol to eliminate unreacted fatty acids and filtrated with ethanol and distilled water to ensure the fatty acids were fully removed. The filtrated residue was collected and identified as modified RPMS.

2.3 Evaluation of effectiveness of treatment for modified RPMS

2.3.1 Hydrophilic coefficient (HC)

HC test indicates the adhesive power of chemical and mineral content of fillers to bitumen after the sedimentation process for 30 minutes as referred to in JTG E42-2005 T0353. This test involved mixing 5g of fillers with 15-30ml of water/kerosene into the mortar and then grinding it for 5 mins to ensure thorough mixing, and then pouring it into a measuring cylinder which was then filled to the maximum scale with water or kerosene. It was kept at a constant temperature and the result is defined by the

\[
\eta = \frac{V_{\text{distilled water}}}{V_{\text{kerosene}}}
\]

Where, \( \eta \) is hydrophilic coefficient, \( V_{\text{distilled water}} \) and \( V_{\text{kerosene}} \) is the volume of RPMS in water and kerosene after 30 minutes of sedimentation, respectively.

2.3.2 Water contact angle (WCA)

The WCA test conducted based on ASTM D7334 covers a measurement of the angle of contact of an applied drop of liquid or water with a volume no greater than 20uL deposited on the coated surface. The water drop was observed by using a digital microscope and analysed with a software named ImageJ. During the test, the angle must be measured rapidly or within 30 seconds of depositing the drop to prevent any changes in angle due to evaporation.

2.3.3 Surface morphology – Scanning electron microscopy (SEM)

The SEM test in accordance with ASTM E 986 was for surface examination by analysing the grain size and morphology of the RPMS by using a SEM machine named QUANTA FEG 450. The electrons in RPMS that are placed on small SEM stubs interact with the sample via the gold coating layer and the signals generated are detected. The detected signals were displayed on the screen of a cathode ray tube and developed an image that illustrated the information about the fillers' surface topography and composition.

2.4 Preparation of asphalt mixture

2.4.1 Preparation of asphalt specimen

The asphalt mixture was prepared by referring to AASHTO TP4 and AASHTO PP2. Two types of mixtures which incorporate unmodified and modified RPMS were prepared. The
aggregate gradations as shown in Table 3 were prepared. For RPMS incorporated asphalt mixtures, RPMS was separated and mixed with the aggregates. Then, heated aggregates and fillers were placed into the preheated mechanical mixer and blended dry for 1 to 2 minutes before an appropriate amount of heated asphalt binder was added. The loose asphalt mix was transferred to a tray and conditioned at 150°C in the oven for 2 hours to ease the binder absorption. The batching mass involved 1150g and 100 gyrations.

Table 3. Aggregates gradation for asphalt mixtures.

<table>
<thead>
<tr>
<th>B.S Sieve Size</th>
<th>% Passing (By Weight)</th>
<th>AC 14 Mix Design (%)</th>
<th>0.5% RPMS</th>
<th>1.0% RPMS</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>Control</td>
<td>Aggregate</td>
<td>RPMS</td>
</tr>
<tr>
<td>20 mm</td>
<td>100</td>
<td>0</td>
<td>0</td>
<td>-</td>
</tr>
<tr>
<td>14 mm</td>
<td>90 – 100</td>
<td>5</td>
<td>5</td>
<td>-</td>
</tr>
<tr>
<td>10 mm</td>
<td>76 – 86</td>
<td>14</td>
<td>14</td>
<td>-</td>
</tr>
<tr>
<td>5 mm</td>
<td>50 – 62</td>
<td>25</td>
<td>25</td>
<td>-</td>
</tr>
<tr>
<td>3.35 mm</td>
<td>40 – 54</td>
<td>9</td>
<td>9</td>
<td>-</td>
</tr>
<tr>
<td>1.18 mm</td>
<td>18 – 34</td>
<td>21</td>
<td>21</td>
<td>-</td>
</tr>
<tr>
<td>425µm</td>
<td>12 – 24</td>
<td>8</td>
<td>8</td>
<td>-</td>
</tr>
<tr>
<td>150 µm</td>
<td>6 – 14</td>
<td>8</td>
<td>8</td>
<td>-</td>
</tr>
<tr>
<td>75 µm</td>
<td>4 – 8</td>
<td>4</td>
<td>3.8</td>
<td>0.2</td>
</tr>
<tr>
<td>Mineral Filler</td>
<td></td>
<td></td>
<td>6.0</td>
<td>0.3</td>
</tr>
<tr>
<td>Total</td>
<td>100</td>
<td>99.5</td>
<td>0.5</td>
<td>99</td>
</tr>
</tbody>
</table>

2.4.2 Marshall mix design

Optimum binder content was obtained through the parameters obtained from plotted graphs that were calculated from the Marshall stability and mixture volumetric tests. The parameters are required to conform with PWD specifications.

2.5 Adsorption strength of fillers

The porosity or void content of RPMS on its surface was identified by equations that were interpreted from the Rigden void test. This test was commenced by adding kerosene to 15g of RPMS in a mortar, which was accompanied by a continuous shaking simultaneously. The process proceeded until the mixture lost cohesion as all the added kerosene was attached between the voids of RPMS and achieved the maximum limit for filling the voids. The maximum volume of kerosene to fill up the voids used to calculate bulk concentration and Rigden voids by using Equations (2) and (3) [13].

\[
\text{Bulk concentration, } x (\%) = \{(V_{\text{before}} - V_{\text{after}})/(V_{\text{before}})\} \times 100 \tag{2}
\]

\[
\text{Rigden voids, } y (\%) = -1.05x + 101.75 \tag{3}
\]

The results from the Rigden voids test that indicate the absorption of bitumen into the fillers were utilized in determining the adsorption of fillers, which indicated the amount of bitumen that adheres to the filler particles. The adsorbed binder layer may change the viscosity drastically, hence Equations (4) to (6) were used for the analysis.

\[
V_f = \left(\frac{M_f}{G_{sf}}\right)/\left[\left(\frac{M_f}{G_{sf}}\right) + \left(\frac{M_b}{G_{sb}}\right)\right] \tag{4}
\]

Where \(M_f\) is mass of filler, \(G_{sf}\) is specific gravity of filler, \(M_b\) is mass of binder, \(G_{sb}\) is specific gravity of binder.
\[ V_{fa} = (100 \times V_f)/(Rigden Void) \] \hspace{1cm} (5) \\
\[ V_{bfree} = (100 - V_f) \] \hspace{1cm} (6)

In Equation (5), \( V_{bfree} \) is the binder that suspended within the mixtures after the adsorption process of binder has stopped. Meanwhile, the effective particle concentration \( (\phi_e) \) could be figured out by the ratio of particle concentration \( (\phi) \) to the maximum concentration without binder \( (\phi_m) \) in Equation (9). The \( \phi_m \) and \( \phi \) was determined based on Equations (7) and (8), meanwhile \( V_{fa} \) is effective filler volume, \( V_{bfree} \) is volume of free binder.

\[ \phi_m = V_f / V_{fa} \] \hspace{1cm} (7) \\
\[ \phi = V_f / (V_{fa} + V_{bfree}) \] \hspace{1cm} (8) \\
\[ \phi_e = \phi / \phi_m \] \hspace{1cm} (9)

Lastly, the amount of adsorbed binder layer, \( \sigma \) in which the binder layer was turned out to be a part of the filler was determined by using Equation (10) [14, 15]. Based on the equation, \( \phi_e \) is the effective particle concentration, \( A_s \) and \( G_s \) represents as specific surface area and specific gravity respectively. The specific surface area can be obtained from the particle size distribution analysis in accordance with ASTM D 422.

\[ \text{Adsorbed binder layer, } \sigma = (\phi_e - \phi)/(\phi \times G_s \times A_s) \] \hspace{1cm} (10)

### 3 Results and discussion

#### 3.1 Physical properties of RPMS

The raw RPMS that obtained from factory was moist in nature. An evaluation on physical properties of RPMS is summarized and tabulated in Table 4.

**Table 4.** Physical properties of RPMS.

<table>
<thead>
<tr>
<th>Physical Property</th>
<th>Unit</th>
<th>Test Results</th>
</tr>
</thead>
<tbody>
<tr>
<td>Water Content</td>
<td>%</td>
<td>67.00</td>
</tr>
<tr>
<td>Time Taken to Eliminate Water Content</td>
<td>H</td>
<td>24</td>
</tr>
<tr>
<td>Density</td>
<td>kg/m³</td>
<td>1.7776</td>
</tr>
<tr>
<td>Coefficient of continuity, ( C_u )</td>
<td>0.075 mm</td>
<td>12.25</td>
</tr>
<tr>
<td>Pan size</td>
<td>mm</td>
<td>10.00</td>
</tr>
</tbody>
</table>

#### 3.2 Hydrophobic treatment of RPMS

The RPMS filler has been decided to be treated by utilizing ethanol in producing WCO-SFEEs to improve compatibility with the RPMS. This method was selected from trial-and-errors based on its optimum hydrophilic coefficient of 1 and the highest water contact angle as recorded. The modification was finalised by involving 7ml of WCO, ethanol as well as 0.075mm or pan size RPMS fillers in the step of preparing fatty acid esters.

#### 3.3 Evaluation of effectiveness of pre-treatment

##### 3.3.1 Hydrophilic coefficient

Theoretically, hydrophilic materials have HC that is greater than one and the higher value indicates that there is a higher volume of filler in water than kerosene. The filler has a
hydrophobic property when its HC is below 1. The modified RPMS showed an improved HC with a value of 1.00 and this indicates an improvement in its hydrophobicity as compared to the unmodified RPMS with a HC of 1.33. Further treatment optimization will be carried out to improve the hydrophobicity of RPMS, whereby the HC should be below 1.

3.3.2 Water contact angle (WCA)

A WCA that is less than 90° indicates the tested specimen is hydrophilic. A surface is hydrophobic when the WCA is greater than 90° but smaller than 145°, whereby a super-hydrophobic surface exhibits a WCA that is greater or equal to 145° [16]. The modified RPMS achieved a higher WCA of 87.949° than the unmodified RPMS (83.823°). The higher WCA may cause by its more evenly particle size distribution or higher -OH substituted degree thus increasing its hydrophobicity [17]. Furthermore, fillers with higher roughness surface may experience a higher WCA as investigated by Dalhat [18]. Concisely, the ethanol method with 7ml WCO is accepted as it has significantly highest WCA among the fillers.

3.3.3 Surface morphology – scanning electron microscopy (SEM)

The test was conducted under magnification rates of 500x, 700x and 1000x. For 0.075mm RPMS, coarse surface textures with pores, attachment of fine particles, and loose antenna-like structures were observed in the unmodified sample. These provide large surface areas and enhance the adsorption of asphalt binder, which results in an increased binder amount required [19]. The modified RPMS (Fig. 2 (b)) possessed a more noticeable angular shape with smoother surfaces due to low attachment of fine particles onto the surfaces. However, the high porosity on the particles surface increases the absorption of optimum binder content as it requires more asphalt binder to fill the voids to achieve better bonding.

![0.075mm unmodified RPMS](image1)

![0.075mm modified RPMS](image2)

![Pan size unmodified RPMS](image3)

![Pan size modified RPMS](image4)

**Fig. 2:** Surface morphology of fillers.

For pan size RPMS, agglomeration occurred within the unmodified particles (Fig. 2 (c)) and made the binder coating more difficult. Irregular shapes and attachment of fine
particles provide higher surface area, which prompts increase in the adsorption of binder [20]. The modified pan size RPMS in Fig. 2 (d) are more regular and spherical in shape. In conjunction with this, a larger surface area provided by spherical particles also increases the binder adsorption but is able to provide an even coating of the particles. Individual grains can be spotted clearly and well dispersed. Less fine particles are found as compared to unmodified pan size RPMS.

3.4 Marshall mix design

3.4.1 Marshall properties

This research indicated both asphalt mixtures incorporated with 0.5% and 1.0% modified RPMS have an increased OBC. The increment is believed to be affected by skipping the heating process before mixing due to its combustible property, and the strong adsorption strength of RPMS.

Table 5. Summary of Marshall mix design parameters.

<table>
<thead>
<tr>
<th>Marshall Mix Design Parameter</th>
<th>Unit</th>
<th>PWD Specification for AC14 (%)</th>
<th>Results</th>
</tr>
</thead>
<tbody>
<tr>
<td>OBC</td>
<td>%</td>
<td>4.0 – 6.0</td>
<td>Controlled Mixture</td>
</tr>
<tr>
<td>Stability, S (N)</td>
<td>N</td>
<td>&gt; 8000</td>
<td>16.56</td>
</tr>
<tr>
<td>Flow, F</td>
<td>mm</td>
<td>2.0 – 4.0</td>
<td>3.46</td>
</tr>
<tr>
<td>Stiffness, S/F</td>
<td>N/mm</td>
<td>&gt; 2000</td>
<td>4.82</td>
</tr>
<tr>
<td>Air Voids</td>
<td>%</td>
<td>3.0 – 5.0</td>
<td>4.01</td>
</tr>
<tr>
<td>Voids in Aggregate Filled with Bitumen, VFB</td>
<td>%</td>
<td>70 - 80</td>
<td>78.94</td>
</tr>
<tr>
<td>Density</td>
<td>g/cm³</td>
<td>-</td>
<td>2.41</td>
</tr>
<tr>
<td>Voids in Mineral Aggregates, VMA</td>
<td>%</td>
<td>-</td>
<td>15.39</td>
</tr>
</tbody>
</table>

The asphalt mixture with modified RPMS as filler had a higher optimum binder content (OBC) as compared to the unmodified mixture. The increase in air voids for modified asphalt is probably due to the agglomeration of particles after treatment that leads to unequal particle sizes, which affects the compaction of the mixture and causes the mixture to have more air voids than the controlled mixture. The VFB has decreased with the addition of modified RPMS and reduced with the increased incorporated modified RPMS. The 1.0% modified RPMS with insufficient VFB indicated that there was not enough asphalt to fill the voids and thus affected the durability of the mixture [21]. The increasing VMA with the addition of modified RPMS provides more available spaces to accommodate the asphalt and promotes the entrance of air and water to enter via the high air-void content. This has increased the optimum binder content as there are more air-void spaces that exist in between the aggregate particles in a compacted paving mixture to be filled.

3.4.2 Mechanical properties of asphalt mixtures

The mechanical properties such as stability, flow, and stiffness of the asphalt mixture are analysed and tabulated in Table 6 and compared with the PWD standard specification.
Table 6. Mechanical properties of mixtures.

<table>
<thead>
<tr>
<th>Sample</th>
<th>OBC %</th>
<th>Stability (kN)</th>
<th>Flow (mm)</th>
<th>Stiffness (kN/mm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Controlled Mixture</td>
<td>5.18</td>
<td>16.56</td>
<td>3.46</td>
<td>4.80</td>
</tr>
<tr>
<td>0.5% Modified RPMS</td>
<td>5.44</td>
<td>12.74</td>
<td>3.23</td>
<td>3.97</td>
</tr>
<tr>
<td>1.0% Modified RPMS</td>
<td>5.49</td>
<td>13.64</td>
<td>3.00</td>
<td>4.52</td>
</tr>
</tbody>
</table>

It can be noticed that the incorporation of RPMS caused a decrease in stability as compared to the controlled mixture. It significantly increased the OBC, and the high bitumen content has developed thicker film around the aggregates which resulted in displacing the contacted aggregates. From Fig. 3, it illustrated that the stability value for each mixture has satisfied the minimum standard requirement for stability from PWD (8kN). Therefore, the stability value of the three mixtures is accepted.

From Fig. 4, a constant reduction in flow value has been noticed in accordance with the increase in RPMS. The trend implied that the higher the percentage of incorporated RPMS, the lower the vertical deformation. In general, the flow results are within the PWD standard requirement range for flow value, which is 2mm to 4mm, and thus the flow of the mixtures is accepted.

The high stiffness as well as the high stability that is achieved by controlled and 1.0% modified RPMS may contribute to better bonding and adhesion between aggregates and thus improve the mixture strength. In contrast, the 0.5% modified RPMS with the lowest stiffness value reduced the rutting resistance and led to increased rut depth [22]. Generally, stiffness in mixture is influenced by stability and flow, and thus, higher stability and lower flow are more preferred to achieve high stiffness performance. In addition, the minimum requirement for stiffness that is fixed by PWD is 2kN/mm, which has been satisfied by the three mixtures in this study, as shown in Fig. 5.
Fig. 5. Stiffness for each bituminous mixture.

3.5 Adsorption of binder layer

Kandhal [23] deduced that several parameters, such as particle shape, particle size distribution, and surface texture, influence the void properties of mineral fillers. This consequently impacts the thickness of the bitumen layer attached around the filler. Table 7 shows that the 0.075mm modified RPMS has the highest adsorption of binder layer due to its high porosity on the surface of particles, as shown in Fig. 2 (b). A higher amount of bitumen is required to fill up the voids on the surface of the fillers in order to achieve thorough coverage and adequate adhesion. On the contrary, hydrated lime and granite with lower adsorption of bitumen have fewer voids presented and a smoother surface as shown in Fig. 6 (c) and (d), thus less binder is required.

![Image](a) Limestone [24]

![Image](d) Granite [25]

Fig. 6. Surface morphology of fillers.

<table>
<thead>
<tr>
<th>Properties</th>
<th>0.075mm Modified RPMS</th>
<th>Pan size Modified RPMS</th>
<th>Hydrated Lime</th>
<th>Granite</th>
</tr>
</thead>
<tbody>
<tr>
<td>Porosity</td>
<td>High</td>
<td>High</td>
<td>Low</td>
<td>Low</td>
</tr>
<tr>
<td>Surface morphology (SEM)</td>
<td>Particle’s agglomerate and irregular sizes</td>
<td>Rough surface texture with voids</td>
<td>Fine particles attached</td>
<td>Smooth surface with less particles attached</td>
</tr>
<tr>
<td>Adsorption of binder layer (%)</td>
<td>4.43</td>
<td>2.43</td>
<td>1.62</td>
<td>0.33</td>
</tr>
</tbody>
</table>

Table 7. Adsorption value for fillers.

4 Conclusion

This study presented the utilization of WCO and ethanol as the hydrophobic treatment for RPMS mineral fillers due to its optimum HC as well as the highest WCA. To ensure the presence of RPMS will not affect the resistance to moisture damage of the RPMS modified
asphalt mixture, other potential treatment optimizations will be performed to increase the hydrophobicity of RPMS, with the HC value set to be less than 1. The modified RPMS-incorporated asphalt mixtures resulted in a higher OBC than the controlled mixture due to the high adsorption strength of RPMS particles. All the volumetric properties showed a value that satisfied the standard specification by PWD for 0.5% modified RPMS asphalt mixture, and thus it is preferable as it also involved lower binder cost due to the lower OBC achieved. The mechanical properties of the modified asphalt mixture that were determined from the Marshall stability test have achieved the standard specification of PWD and hence the results are accepted. On the other hand, there will be more research into its direct impact, including skid resistance and cost effectiveness, of the RPMS modified asphalt mixture as a road surfacing material to be done in the future.

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