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VARIABILITY OF RAP BINDER MOBILISATION IN HOT MIX ASPHALT MIXTURES

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Abstract

Assuming the full availability of aged binder in Reclaimed Asphalt Pavement (RAP) is one of the considerations in the design of bituminous mixtures with RAP. However, many studies have shown that in reality only a part of this binder should be reflected in the mixing process. The extent of the RAP binder mobilised during mixing could have a significant effect on the performance properties of mixtures. Very little is currently known regarding the variability of this mobilisation subject to the type of RAP used. In this study, the extents of RAP binder mobilisation for 3 different RAP sources were compared using attenuated total reflectance Fourier transform infrared (ATR-FTIR) spectroscopy as a major assessment tool. The results of the laboratory tests showed that there is substantial variability in mobilisation capability, up to 50% depending on RAP source. Viscosity tests on the binders indicated that the workability of the RAP binders might play a significant role in the mobilisation process. Further, chemical composition analysis through saturates, aromatics, resins and asphaltene (SARA) fractionation and microstructural characterisation using optical microscopy implied that the mobile fractions of the respective RAP binder will likely influence its ability to be active in a mixing process. Overall, it is recommended that this variability be taken into consideration and provisions, such as improved specifications, or accommodation of special mixing conditions, such as increased temperature or mixing time depending on the rheology of RAP used, be provided.

Keywords: RAP, ATR-FTIR, Binder Mobilisation, RAP Mixing

1. Introduction

The use of reclaimed asphalt pavement (RAP) in bituminous mixtures has turned into a customary practice around the world to increase the sustainability of pavement. However, many concerns still persist regarding the unpredictable performance of RAP mixtures and uncertain elements in RAP mixture design (1, 2). One of the main apprehensions hindering the applicability of high RAP mixtures is the uncertainty regarding the extent of RAP binder that is active or mobilised during the mixing process, and then accessible to form a binder layer around aggregates. Many current specifications regarding RAP mixture design assume that all the binder covering RAP aggregates are available to be mixed with the virgin binder during mixing. Further, it is presumed that the RAP binder completely homogenises with the virgin binder to generate a homogenous mixture blend with target properties (1). According to this theory, the RAP binder achieves adequate workability during the mixing process to contribute and fully blend with the added virgin binder (1). Nevertheless, many research studies have indicated this may not be the exact case in practice. Some studies have reported a high level of mobilisation and blending may be possible (3). On the other hand, studies have also indicated limited or partial mobilisation is the likely scenario under normal mixing conditions (4). Researchers have also reported a “black rock” effect when RAP is used, which insinuates the possibility of a double coating of RAP binder and virgin binder (5). Overall, RAP binder mobilisation is an important design issue, especially for high RAP mixtures, as underasphalted mixtures could increase the possibility of fatigue cracking and moisture damage (1). The significant variability of different results available in literature also indicates that seemingly looking for universally applicable quantitative answers may not be the correct approach as there is substantial inconsistency in materials used. Nearly all studies conducted so far have characterised the level of blending and mobilisation with the respective materials of the study, while none have analysed their materials characteristics in depth, which in turn could have prompted the observed results. One of the main material characteristics of importance is the RAP material used. It is likely that the binders in different types of RAP will be mobilised and blended differently with virgin binders, depending on their rheology and chemical properties. Hence, the aim of this study is to assess the variability in mobilisation using RAP from different sources and identify the probable mechanisms involved in the process. It is anticipated that the outcome of such studies will allow practitioners and engineers to produce RAP mixtures with more reliable performance and better understand RAP material at a microscopic level, and thereby help facilitate the improvement of current specifications in relation to RAP mixture design.

2. Methods to Characterise RAP Binder Mobilisation and Blending

In literature, numerous rheological and chemical methods have been applied to study the mobilisation and blending of RAP mixtures. One of the earliest relevant studies presumed a preliminary percentage of mobilised RAP binder active in the mix and recommended an iterative procedure to estimate the level of blending centred on rutting factor values from the recovered mixtures (6). Rheological models, such as the Hirsch model, have also been used to forecast the dynamic modulus of blended binders using the master curves obtained from mixture tests (7, 8). Another study defined the blending level as the mobilisation percentage of RAP binder and estimated the mobilisation extent based on air void content. Using 25% RAP, they reported a mobilisation level of around 90% (9). The chemical methods that have been reported to characterize mobilisation and blending mainly include gel permeation chromatography (GPC) and Fourier transform infra-red (FTIR) spectroscopy. One study examined the amount of recycled binder that could be mobilised during mixing, using GPC as a tool (3). The results disclosed that the RAP binder mobilisation rate declined with the rise in RAP percentage. However, it could be close to 100% at RAP contents of up to 20%. The authors of this paper used FTIR in ATR mode to determine the extent of RAP binder mobilisation in mixtures with and without warm mix additives (4). It was found that the mobilisation is dependent on temperature and increases with the use of warm mix additives. In this study, a similar method based on ATR-FTIR was chosen as a means of estimating the RAP binder mobilisation for mixtures prepared with different RAP sources.

3. Experimental Mix Design and Materials

The mixtures in this study were prepared using the Marshall mix design method. The virgin binder and aggregates used were of penetration grade of 60/70 (Pen 60/70) and local granite based rocks, respectively. The three different sources of RAP were obtained locally from wearing course milling and named RAP-1, RAP-2 and RAP-3, respectively. The specific gradations of the three RAP mixtures are presented in Table 1. However, only RAP above 0.6mm was used in this study to prevent excessive clustering of RAP fines. The binders from each RAP were extracted according to the procedure specified in AASHTO T164 and the conventional properties of the extracted binders and virgin binder are presented in Table 2. In terms of mixture design, the mixtures in this study were prepared with consideration of the whole contribution of the recycled binder. AASHTO T308 standard was followed to determine the binder contents of the RAP, which were found to be 5.4%, 5.7 %, and 5% for RAP-1, RAP-2 and RAP-3, respectively. For all the mixtures with 15%, 30% and 50% RAP materials, a total binder content based on the binder content of the respective RAP was chosen. Lastly, a mechanical mixer was used to mix all the samples for a period of 2 min to reduce variability between samples with regards to preparation.

Table 1. Gradation of the Various Mixtures

	Mixture Gradation	RAP Gradation		
Sieve Size (mm)	Passing ratio (%)	RAP-1	RAP-2	RAP-3
14.00	100.00	100.00	100.00	100.00
10.00	85.00	90.70	95.60	94.23
5.00	58.00	68.15	73.46	58.87
2.36	38.00	49.02	50.22	35.46
1.18	26.00	33.81	32.73	25.54
0.60	17.90	23.31	21.80	19.32
0.30	11.00	14.84	16.13	14.60
0.15	3.40	8.31	11.96	9.85
0.08	3.00	4.27	6.92	5.42

Table 2. Conventional Rheological Properties

	RAP-1	RAP-2	RAP-3	Virgin Binder
Penetration (0.1 mm, 25°C)	34.0	39.0	46.0	69.0
Softening Point (°C)	61.5	57.5	53.5	48.0

In a normal mixing process, it would be practically impossible to distinguish between RAP and virgin aggregate after mixing. However, in order to estimate RAP binder mobilisation, it is necessary to analyse at least some of the virgin aggregates whose original identity is known.

To differentiate between RAP aggregates and virgin aggregates, this study used borosilicate glass beads as artificial aggregates to replace a small part of the virgin aggregates in the mix (10). Figure 1 shows a picture of the glass beads used in this study. The beads of sizes 7.5mm, 10mm and 12mm constituted around a 2% fraction of the complete gradation of the mixes, similar to the method reported and evaluated in a previous study (4). The chemical composition and the weight of glass beads used in each mixture are shown in Table 3 and Table 4, respectively. Prior to use in tests, the glass beads were dry blended with aggregates to roughen its surface and simulate its rough texture.

Figure 1. Picture of glass beads.

Table 3. Properties of Glass Beads

Chemical Composition	SiO_2-82% B_2O_3-12.4% Na_2O-3% Sb_2O_3/As_2O_3 - <0.01%
Density	2,230 kg/m^3
Melting Point	1,500°C

Table 4. Weight of Glass Beads Used in Each Mixture

Glass Bead Types (*Number)	Weight (g)
12mm (*2)	7.17
10mm (*4)	8.88

7.5mm (*6)	7.08
Sum	23.13

4. Experimental Procedure and Methods

4.1 Preparation of Samples and Mixing Process

Mixtures with 15%, 30% and 50% RAP were prepared using each type of RAP and virgin materials. All samples were prepared at the standard hot mixing temperature of 165^{o}C. Regarding the mixing process, the glass beads were heated with the virgin aggregates at approximately 10^{o}C higher than the mixing temperature before adding the pre-heated RAP (11). After mixing with the virgin binder, all the glass beads were collected from the loose mixtures. The binders were removed from the glass beads of each respective mixture as a single entity using trichloroethylene (TCE) solvent for further analysis (12).

4.2 Method for Evaluating RAP Binder Mobilisation

In this study, RAP mobilisation was estimated using a method that employed the use of an FTIR spectrometer in ATR mode, and has been applied in a previous work by the authors (4). The main concept revolves around the fact that the FTIR spectra of RAP binders exhibits significantly higher levels of carbonyl bands (C=O) at a wavelength of around 1700 cm^{-1}, due to the natural oxidation of asphalt binder over time. In comparison, virgin binders exhibit little or no carbonyl bands at the respective wavelength. After a mixing process, the binder coating the virgin aggregates is theoretically a blend of mobilised RAP binder and virgin binder. This blend should naturally exhibit a different intensity in carbonyl band as compared to the RAP binder and virgin binder. The difference in oxidation between the RAP binder, virgin binder and the recovered binder is then utilised to estimate the amount of RAP mobilisation. Based on this, a mathematical parameter for estimating the RAP mobilisation was developed. As per the method, the analysis of a single FTIR spectra involves the integration of areas, normalised spectra and absolute baseline (13). Figure 2 shows the FTIR spectrum of a sample virgin binder and RAP binder, demonstrating the oxidation peak at the wavelength of around 1,700 cm^{-1} for the RAP binder (4). The Figure also shows the analysis area of interest in the FTIR spectra.

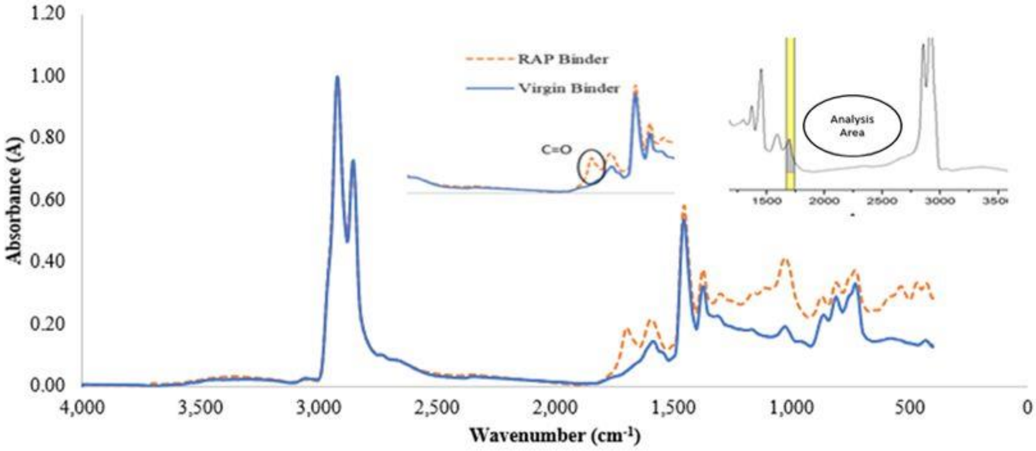


Figure 2. Sample FTIR spectra of virgin binder and RAP binder (4)

Mathematically, the area of analysis of a single spectrum can be calculated using the following equation:

$$\text{IA}=\int_{w_{l,oa}}^{w_{u,oa}} VA_{norm}(w)\,dw \qquad (1)$$

where IA is the normalised integrated area; $w_{u,oa}$ is the upper wavenumber limit for the structural group; $w_{l,oa}$ is the lower wavenumber limit for the structural group and $VA_{norm}(w)$ is the normalised absorbance at wavenumber w. The carbonyl band was charted from the wavelengths of 1666 to 1746 cm^{-1} (13).

Using Equation (1), the parameters for estimating the extent of RAP binder mobilisation were developed. The recovered binder for each mixture is anticipated to have a sizable proportion of RAP binder mixed in it as a result of the mixing process. The IA of the RAP binder is considered to have 100% RAP character whereas the IA of the virgin binder has 0% of this

RAP character. A parameter was then developed to estimate the percentage of RAP binder in the retrieved binders and used to represent the RAP mobilisation, which can be described mathematically as follows:

$$\text{Percentage of RAP Binder Recovered (\%)} = \frac{IA_{BS}-IA_{VB}}{IA_{RAP}-IA_{VB}}*100 \qquad (2)$$

where,

IA_{BS} is the IA of the recovered binder from the respective sample;

IA_{VB} is the IA of the virgin binder;

IA_{RAP} is the IA of the RAP binder.

In this study, the binders were examined using a Bruker Vertex 70 Hyperion 1000 spectrometer mounted with a diamond ATR module to obtain this parameter. The spectra of each binder were measured in reflective mode from 4000 to 4 cm^{-1} with a resolution of 4 cm^{-1}.

4.3 Viscosity

To measure the viscosities of various RAP binders, Brookfield viscosity tests were conducted in accordance with ASTM D4402 using a DV-II Brookfield rotational viscometer. The samples were conditioned in the thermo-container for about 30 min at the chosen test temperature before testing.

4.4 Saturates, Aromatics, Resins and Asphaltene (SARA) Fractionation

The SARA fractionation into saturates, aromatics, resins and asphaltene components was conducted as per ASTM D2007 to determine the chemical compositions of the different binders. In this test, the asphaltenes and maltenes were firstly separated using isooctane. The separation of the maltenes fraction into saturates, aromatics and resins was conducted using a glass liquid chromatography column with chromatographic grade activated alumina. The three fractions were subsequently eluted using different solvents, such as n-heptane, methanol, toluene and trichloroethylene.

4.5 Optical Microscopy

To study the microstructures of the various binders, an optical microscope from Leitz Ergolux equipped with a 100X objective was used, following the procedure described in a previous study (14). The samples were illuminated using a 20W halogen lamp emitting a spectrum peaked at 630 nm. To prepare the testing samples, a tiny amount of binder was placed onto a microscopic slide and then heated to around 140°C using a heater. The heating was stopped once the bitumen melted and flowed. The samples were then left to cool to room temperatures before testing.

5. Results

5.1 RAP Binder Mobilisation

The binders recovered from the prepared mixtures were evaluated using Eq (2) and the results are presented in Figure 3. It can be seen that RAP-1 and RAP-2 showed similar mobilisation trends, whereas in comparison, RAP-3 showed higher mobilisation tendencies. In general, the results suggest that the mobilisation of RAP binder is lesser when the RAP content in the mixture is higher. At 50% RAP content, mixtures prepared with RAP-1 and RAP-2 , showed around 20% and 22% of RAP character in the recovered binders, whereas the mixtures prepared with RAP-3 showed a higher percentage of 28%. At all percentages of RAP, the binder in RAP-3 was able to be mobilised at a higher rate as compared with RAP-1 and RAP-2. RAP-3 showed on average around 35% -50% higher mobilisation rate than RAP-1 whereas the difference between RAP-1 and RAP-2 was only around 10%. However, from these results, it can be inferred that a sizeable percentage of RAP binder seems to be unavailable to be mixed with the virgin binder under these mixing conditions. Thus, there is a high possibility of producing under-asphalted mixtures at high RAP contents if full blending is assumed, which may lead to several pavement distresses (15).

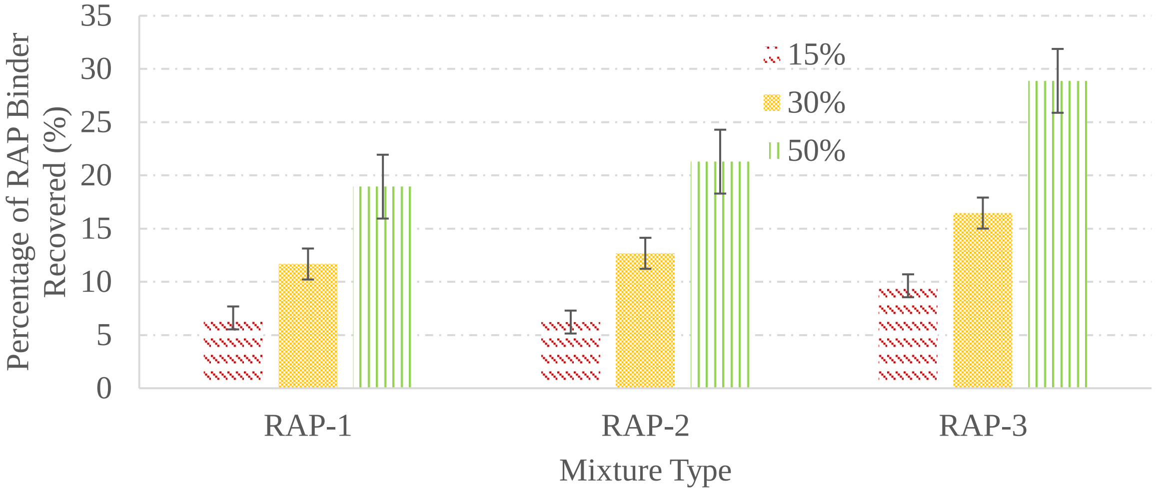


Figure 3. Percentage of RAP binder recovered from binder samples.

5.2 Viscosity

The FTIR test results of the recovered binders indicated that different RAP materials exhibit different binder mobilisation capabilities. Therefore, it is important to understand the various influences that govern the RAP binder activation process so that specific considerations can be taken into account to design mixes. One of the most apparent factors that could be involved in the mobilisation process is the workability of the RAP binder. If the RAP binder is unworkable and highly viscous at the mixing temperature, it is unlikely that it would contribute much in a mixing process, especially in order to create a homogenous film around virgin aggregates.

In this study, viscosity tests were conducted to evaluate if an obvious relationship existed between binder workability and mobilisation capability, and the results are presented in Figure 4. RAP-1 was the stiffest binder exhibiting the highest viscosities at all testing temperatures, followed by RAP-2 and RAP-3. At the mixing temperature of 165°C, RAP-1 had a viscosity of around 540 cP, RAP-2 had a viscosity of 470 cP and RAP-3 had the lowest viscosity of 210 cP. As expected, the RAP binders that are more workable may be more susceptible to be mobilised during a mixing process with virgin binder and aggregate. The viscosity results obtained may also explain the effect observed by other researchers wherein the mobilisation capability increased with temperature, which in essence is a reduction in viscosity and increase in workability of RAP binders (4,12).

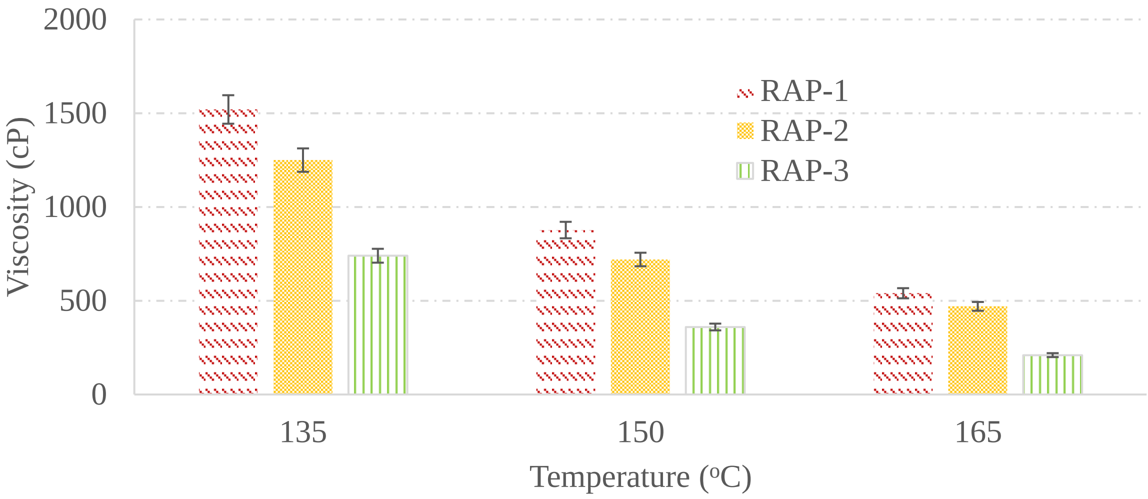


Figure 4. Viscosities of RAP binders

5.3 SARA Fractionation

Although viscosity may be a good indicator for RAP mobilisation capability, it can only explain the mobilisation phenomenon partially. In order to fully comprehend the mechanism involved, it is necessary to analyse the materials from microscopic and chemical perspectives. Ultimately, the chemical compositions of binders dictate their field performance when used in an asphalt mixture. The precise chemical composition of bitumen is complicated and impractical to interpret experimentally. Thus, it is common to separate bitumen into its polar fractions i.e., saturates, aromatics, resins and asphaltenes, which have shown to have a convincing relationship with rheological properties (16, 17). These chemical fractions represent a continuous spectrum of polar molecules and are not isolated units in the bitumen matrix. Although the original sources of bitumen are different in the case of the RAP binders, the SARA fractions can still give some useful information regarding its chemical nature and polarity based mobility tendencies. The SARA fractionation for the various binders were conducted as per the method described in section 4.4 and the results are presented in Figure 5. The ageing of binders generally first leads to a decline in aromatic content and subsequent increase in resin and asphaltene content. It is hypothesised that the aromatics generate resins which are then converted into asphaltenes (18). When comparing the different binders, one obvious difference is the asphaltene content. The asphaltene contents of RAP-1, RAP-2, and RAP-3, were 15.89%, 13.24%, and 12.5%, respectively. But as the original fractions of the binders are unknown, it is difficult to suggest any additional correlation other than this deviation, which is known to influence binder stiffness. Moreover, the asphaltenes produced during ageing can differ than those present initially as reported by some studies (19).

As per the colloidal model of bitumen, the micelles of asphaltene are dispersed in an oily medium of maltenes, i.e., saturates, resin and aromatics. These micelles are thought to be asphaltenes along with a stablising layer of aromatic resins of high molecular weight. The viscosities of the maltene components are dependent on the molecular weight distribution. This viscosity accords a fundamental viscosity to bitumen which is then increased by the asphaltene components. It is the interesting to note that the saturate content in RAP-3 was around 24%, which is considerably higher than the other RAP binders. Higher fractions of saturates have been known to soften bitumen as it can increase the inherent mobility of the chemical fractions (18). Looking factually at the SARA fractions of the different RAP binders, it is reasonable to conclude that RAP-3 will have higher mobile tendencies than the other RAP binders studied here and this phenomenon would be accentuated with increase in temperature and mixing time. Although the SARA compositions and viscosity tests can shed some light into the RAP binder activation phenomenon, it cannot however fully explain the specific interaction of the RAP binder with the virgin binder during mixing. This is discussed further in Section 6.

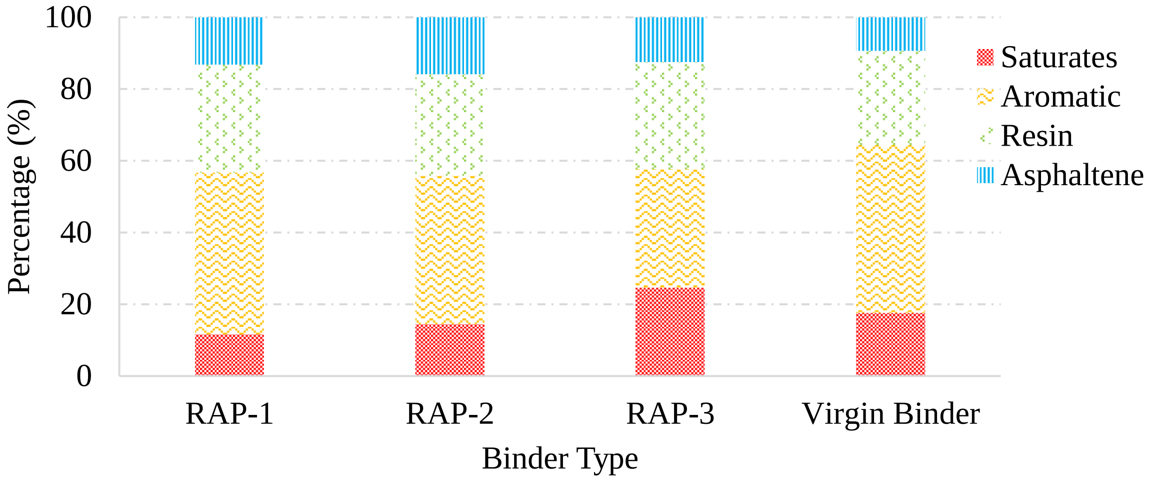
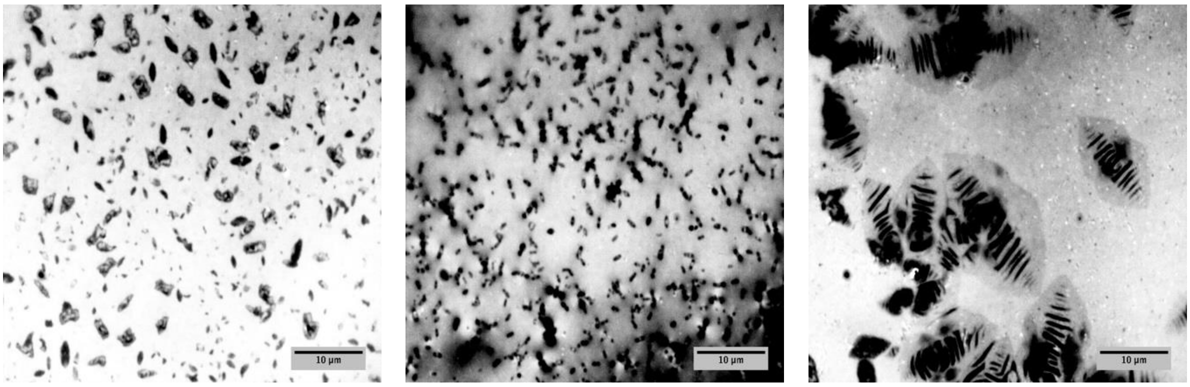


Figure 5. SARA fraction of binders.

5.4 Optical Microscopy

Microscopic studies were conducted using an optical microscope to study the various microstructural features of the RAP binders as illustrated in Figure 6. The characteristic distinct domains or “bee” structures were observed in all 3 binders. However, their size, appearance and dimensions differed significantly. Smaller and less defined bee structures were observed on the surface of RAP-1 and RAP-2, whereas RAP-3 showed more distinct and sharper structures. The bee structures are commonly attributed to the “catana” phase which is known to be surrounded by the dispersed “peri” phase, both phases representing apparent interfacial differences in the bitumen matrix (20). The exact chemistry and rheology of the phases are still being debated. However, it is agreed that the properties of the microstructures influence the bulk properties of the binder, such as stiffness, and viscoelasticity (21). Long-term ageing has been known to significantly affect the microstructural features of asphalt binders, and the smaller bee structures observed here at 100X magnification are characteristic of aged binders reported in literature (22). The appearances of these structure and their differences most certainly depend on different properties of the binder, such as composition and intermolecular mobility. For RAP-3, the formation of relatively larger structural entities suggests that the mobility of its molecules is relatively higher. Conversely, for the stiffer binders, such as RAP-

2 and RAP-1, the structure formation is much smaller and more dispersed. This suggests that either the composition of these binders is not amenable to form structures or the intermolecular mobility is low or both, but their mobility is lower and they cannot reach their low-energy position, creating indented and detached borders, and hence more domain structures (23). Examining these differences in light of other information such as the differences in viscosity and relative SARA fractions suggests that RAP-3 has higher molecular mobility compared to the other two RAP binders. It is also important to note that although these structural features are primarily surface entities, it is the macroscopic properties of the bulk that induces these attributes.

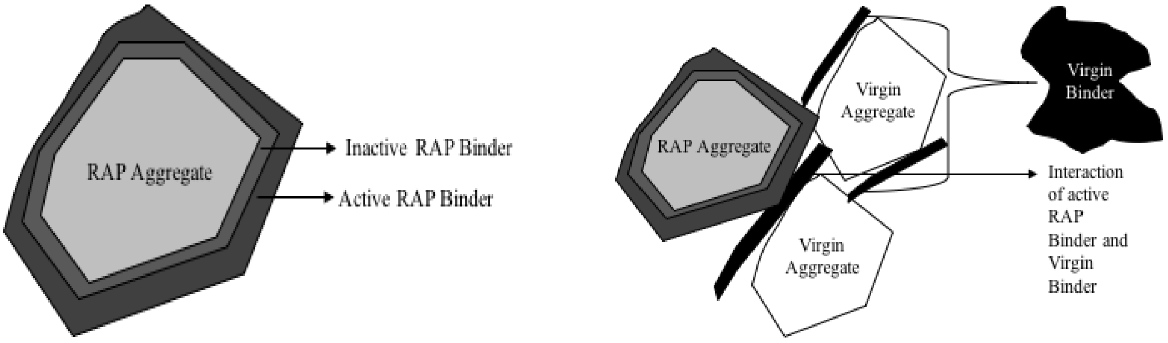


a) RAP-1 b)RAP- 2 c) RAP-3

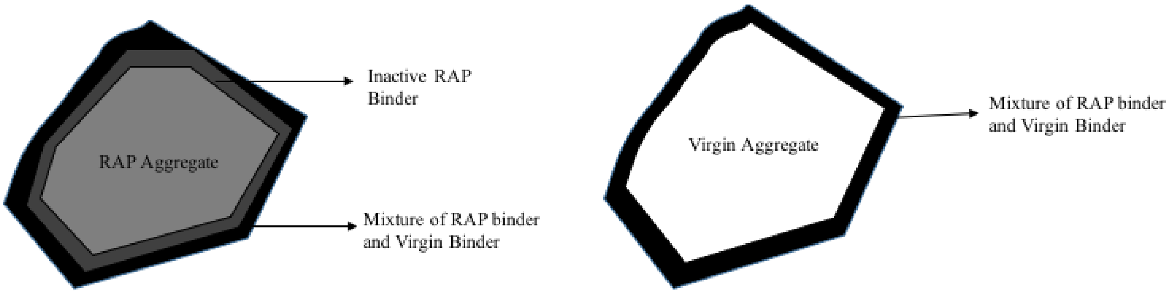
Figure 6. Microstructure images of RAP binders.

6. Discussion

From the tests conducted in this study, it can be ascertained that in conventional hot mixing process, the rate of mobilisation of RAP binder is subject to the specific chemical and rheological properties of the RAP used. Using these results, it is possible to predict the mechanism of RAP binder mobilisation. As reported by many other researchers, there is likely a layer of active binder that exists around RAP binder which can be mobilised during the mixing process with virgin binder, as shown in Figure 7 (24). During the mixing process, the mobilised RAP binder interacts with the virgin binder and virgin aggregates and form a binder blend that will subsequently coat all aggregates in the mixture. However, the amount of mobilised RAP binder will differ, contingent on the type of RAP used and other conditional variables, such as temperature and mixing time, which in principle can also alter RAP properties to induce mobilisation (4, 25).



a) RAP aggregate b) Mixing Process



c) After Mixing

Figure 7. Mixing process of RAP with virgin binder and aggregate.

Although viscosity may be good practical indicator to ascertain the ability of RAP binder to mobilise, it can only partially explain the complete RAP mobilisation phenomenon. It is important to also comprehend the effect of the virgin binder on RAP. In a mixing process, the virgin binder acts a solvent of sorts and “rejuvenates” the RAP binder. Depending on this interaction or compatibility between the binders, some RAP binder can also be activated in this manner. Further, compatibility between two binders can be defined in terms of solubility. If a virgin binder is completely or partially soluble in RAP binder or vice versa, it is rational to conclude that some RAP binder is mobilised in this process. The solubility parameters for bitumen have been previously described for representing the solubility of solvents in bitumen. However, it can also be applied to appreciate the solubility of various bitumen fractions with each other, which is essentially what occurs during a mixing process (26). The basis of solubility parameters can be traced back to the single parameter solubility factor (δ_T), which is based on liquid cohesion energy and molar volume (27). Single parameter solubility factors can be further divided into Hansen solubility parameters (HSP): dispersion interaction (δ_D $MPa^{1/2}$), dipole interaction (δ_P $MPa^{1/2}$) and hydrogen-bonding interaction (δ_H $MPa^{1/2}$). Using this, the relative solubility between two substances can be estimated using the 'distance' between corresponding HSPs in three-dimensional space and referred to as Hansen space (28) . A previous study investigated 15 different binders and interestingly reported different solubility parameters for each Hansen space (29). Although this idea is not further explored in this study, it is recommended for future work and could be a good tool for researchers to estimate the solubility and compatibility between RAP and virgin binders, which will likely affect the mobilisation and blending process.

7. Findings And Conclusion

In this study, the difference in RAP binder mobilisation with the use of variable RAP materials was investigated. Artificial glass beads were used in the prepared mixtures as tracers to separate some virgin aggregates after mixing. The following conclusions could be drawn after the laboratory tests:

- Considerable amount of RAP binder remains immobilised regardless of the RAP source.
- RAP binder mobilisation considerably varies, depending on the nature of RAP used in the mix.

- The RAP binders exhibiting a lower viscosity at mixing temperature showed higher tendencies to be activated and mobilised.
- From the SARA fractionation tests, no clear trend with mobilisation could be gathered. However, the chemical composition of a binder can indicate the mobilisation potential of its fractions.
- From the optical studies, it was seen that the binders exhibiting lower viscosity showed a less degraded surface microstructure and higher molecular mobility.
- It was suggested to extend this study to solubility parameters, which may be applicable to investigate the compatibility between RAP binders and virgin binders and its effect in the mobilisation process.

A limitation of this study was that the glass beads used may not represent the precise nature of real aggregates in terms of texture and absorption. Moreover, only one type of virgin binder and RAP mixtures with non-polymer modified binder were used in this study. It is recommended to conduct further tests using a range of different RAP materials, additives and virgin binders to obtain a wider spectrum of data.

8. Implication For Practitioners

As the quantity of RAP used in mixtures continues to increase, it is critically important for practitioners to consider the actual amount of active and reusable RAP binder available in these mixtures. As seen from the results obtained, there is significant variability in mobilisation between RAP samples. Therefore, a one fits all approach may not be appropriate. At this stage, it would be most suitable to use viscosity as a parameter to assess RAP variability. Binders exhibiting high viscosity may be regarded as incompatible to be used as active binder replacement in RAP mixtures or may require special mixing conditions such as increased mixing time and temperature. Moving forward, there is also a need to develop specifications exclusive to RAP wherein the amount of residual binder added could be catered to the detailed chemical and rheological properties of the RAP used.

9. Acknowledgement

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