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Blending Efficiency of Reclaimed Asphalt Rubber Pavement Mixture and Its Correlation with Cracking Resistance

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ABSTRACT

Despite significant number of studies on recycling of conventional reclaimed asphalt pavement (RAP) mixtures into new asphalt mixture construction, research on recycling reclaimed asphalt rubber pavement (RARP) is still quite limited. The special multiphase system of asphalt rubber (AR) binder may cause a more complicated blending condition during the recycling of RARP due to the existence of rubber particles, and the blending efficiency of RARP and virgin AR mixtures is still unclear. The main objective of this study is to investigate the mobilization tendency of aged AR binder during the recycling of RARP mixture considering both the potential mobilizations of bitumen phase and rubber particles, as well as their correlation with the cracking resistance of the RARP mixture. AR mixtures containing 40% RARP were prepared at three different mixing temperatures and one warm mix case with the inclusion of a foam-based additive. A special gradation design was used to track the mobilization of aged AR binder from RARP to virgin coarse aggregates. Mobilization indexes of bitumen and rubber were established based on Fourier transform infrared spectroscopy and thermal gravimetric analysis tests, while the cracking resistance of the RARP mixture was evaluated by the semi-circular bending test. The mobilization indexes indicated that rubber and bitumen were simultaneously mobilized, and a higher mixing temperature led to stronger mobilization tendencies. In addition, a positive correlation was found between both mobilization indexes and the cracking resistance of the RARP mixture. The foam-based WMA additive markedly improved the blending efficiency but compromised the cracking resistance of the RARP.

Key Words: Reclaimed Asphalt Rubber Pavement, Blending Efficiency, Binder Mobilization, Tire Rubber, Chemical Analysis

1 Introduction

With the aggravating environmental concerns such as the greenhouse effect and resource shortage, the sustainability of infrastructure has become a critical issue worldwide. As the most commonly used materials in pavement industry, asphalt mixture suffers from various distresses such as rutting and cracking throughout the service period (Behnood, 2019). The removal of old asphalt pavement generates a substantial amount of reclaimed asphalt pavement (RAP) materials. It was estimated that in China the amount of RAP produced per year had reached about 790 million tons and may continuously increase within the next five years (Gao et al., 2021). To comply with the reclaim, recycle, and reduce (3R) policies, the use of RAP in new asphalt mixture has become a prevailing practice with numerous merits, including energy and emission reduction, non-renewable materials saving, landfill space conservation, and lower production costs (Jamshidi et al., 2016; Aurangzeb., 2014).

When mixing RAP with new asphalt mixture, one critical consideration is the extent of the active aged asphalt binder that can be mobilized from RAP to mingle with the virgin asphalt binder, because the blending efficiency between aged and virgin binders strongly influences the performances of RAP mixture (Roja et al., 2020). According to the findings of *NCHRP 9-12 Recommended Use of RAP in Superpave Mix Design Method*, it is believed that a partial blending can occur to a significant extent during the mixing (Sreeram and Leng, 2019). To track the binder mobilization from RAP aggregates to virgin aggregates, special mix designs were developed by different researchers, such as using different size ranges of RAP and virgin aggregates (Ma et al., 2021) or incorporating artificial aggregates (Sreeram et al., 2018), so that the targeted aggregates can be identified for binder extraction. Subsequently, the composition of the extracted asphalt binder can be characterized by chemical tests, such as gel permeation chromatography (GPC), fluorescence microscopy, and Fourier transform infrared spectroscopy (FTIR), to evaluate the mobilization extent of aged asphalt binder from RAP to virgin aggregates (Bowers et al., 2014; Ding et al., 2018; Hettiarachchi et al., 2020). Taking the FTIR test as an example, Carbonyl peak is a functional group of asphalt whose concentration increases with aging. This merit of Carbonyl peak makes it an indicator to distinguish the aging degree of asphalt binder, which can be further utilized to establish a mobilization index for RAP mixture. The concept is that RAP binder is considered to have a carbonyl index of 100%, the virgin binder has a carbonyl index of 0%, and the blended binder of RAP and virgin binder would have a carbonyl index between 0% and 100%, so in this scenario the carbonyl index can be used to symbolize the ratio of RAP binder in the blended binder. Therefore, the mobilization rate of RAP binder at different mixing conditions can be quantified and compared through establishing mathematical equations between the three carbonyl indexes. It has been found that the mixing temperature is a dominant factor determining the mobilization extent of RAP binder, which can also be promoted by the addition of warm mix asphalt (WMA) additive (Ma et al., 2020; Sreeram et al., 2019).

Recycling waste tires into crumb rubber modifier (CRM) to produce the rubberized asphalt is another 3R-compliant asphalt paving technology. Among the different technologies of rubberized asphalt, asphalt rubber (AR) is a commonly used technology in which more than 15wt% of CRM is incorporated into raw asphalt binder through wet process (ASTM Standard D6114, 2009). Due to its enhanced durability and environmental benefits, AR has gained increasing interest from the industry and government in recent years (Yu et al., 2020a; Xu et al., 2021). As more and more constructed AR pavements will come to the end of service period, their recyclability becomes an inevitable problem. Asphalt binder becomes stiffer after aging with increased complex modulus and decreased phase angle due to oxidation and the loss of light fractions in bitumen, which causes durability concerns when a high RAP content is used (Apostolidis et al., 2017). By comparison, AR binder has been proved to own superior aging resistance compared to conventional asphalt binder (Wang et al., 2020a). Previous studies found that the incorporation of CRM can mitigate the oxidation of asphalt aging and the absorption behavior of rubber can maintain a softer mechanical property of the swelling rubber during aging to offset the stiffening effect of asphalt aging (Li et al., 2021; Li et al., 2022). Therefore, AR is regarded to be a paving material with superior aging resistance because its rheological properties are more stable and less susceptible to aging effects (Li et al., 2021; Wang et al., 2020a). Such properties of AR may reduce the concern of cracking potential when reclaimed asphalt rubber pavement (RARP) is incorporated in new asphalt pavement, but the recyclability of RARP is unclear yet.

During the recycling process of RARP, the existence of rubber particles may complicate the blending condition. As the absorption behavior of rubber transforms the raw asphalt binder into a binary phase system of swelling rubber and liquid phase, the mobilizations of bitumen and CRM during the blending between RARP and the fresh prepared virgin asphalt rubber (VAR) mixture should be both considered. For clarity, the term AR or RARP binders refer to the whole binder including bitumen phase and rubber particles, while the term bitumen only refers to the bitumen phase in AR or RARP binders excluding the rubber particles. However, very limited attention has been paid to the mobilization of aged AR binder in the research concerning the recycling of RARP. Little is known about whether the aged CRM can be mobilized simultaneously with aged bitumen, and an appropriate index for evaluating the mobilization of aged CRM is missing. The influences of temperature on binder mobilization and the resultant performances of RARP mixture are still uncharted territory. To fill these gaps, this study aims to 1) develop an index for the evaluation of rubber mobilization; 2) investigate the mobilization of aged bitumen and rubber from RARP binder under different mixing conditions; and 3) explore the relationship between the binder mobilization and cracking resistance of RARP mixture. Fig.1 shows the flowchart of this study. To better distinguish the mobilization indexes of RARP binder at different mixing temperatures, a relatively high RARP content (40%) was adopted, because a previous study reported that the mobilization level can be higher than 90% when RAP is up to 20%, which may eclipse the potential impact of mixing temperature on the mobilization of aged asphalt binder (Zhao et al., 2015). AR mixtures containing 40% RARP were mixed at different mixing temperatures (160/175/190 ºC), with or without the participation of WMA additive. A special gradation design was adopted to track the mobilization of aged bitumen and rubber. A series of extraction methods were developed to separate the bitumen and CRM from the aggregates. Chemical tests including FTIR and thermal gravimetric analysis (TGA) were used to establish the mobilization indexes of bitumen and CRM, respectively. Finally, the cracking resistance of different RARP-VAR mixtures was evaluated by the semi-circular bending (SCB) test to reveal the correlation between the mechanical performance and binder mobilization extent. The findings are expected to provide reference for the recycling design of RARP and promote its application.

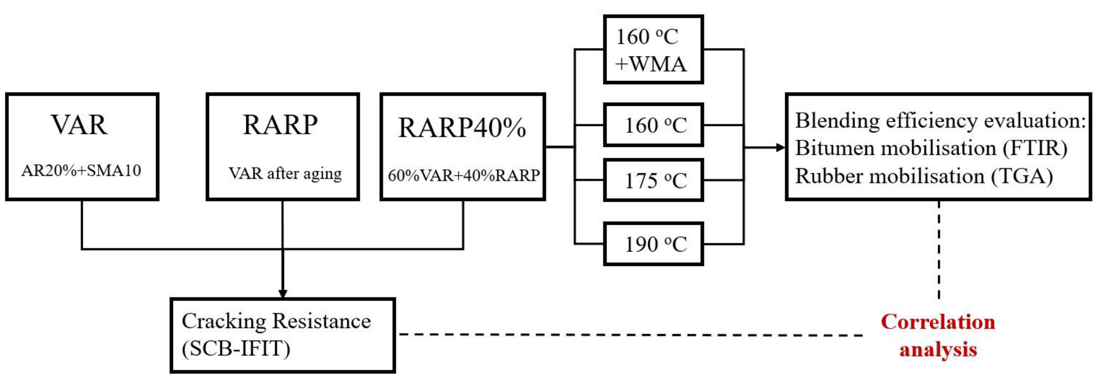


Figure 1. Flowchart of research plan

2 Experimental Program

2.1 Materials and sample preparations

A raw asphalt binder with a penetration grade of 60/70 (Pen60/70), crumb rubber modifier (CRM) with a size of 0.3-0.5 mm ambiently ground from end-of-life truck tires, and granite aggregates were used as the raw materials, all provided by local companies. The AR binder was produced by using a high shear mixer to blend 20% CRM (by mass of raw asphalt binder) into raw asphalt binder at 180 °C and 4000 rpm for 60 min. The basic physical and rheological properties of the AR binder are shown in Table 1. Stone mastic asphalt with a nominal maximum aggregate size of 10 mm (SMA10) was selected for preparing the AR mixture, which is a commonly-used gradation design for the wearing course in Hong Kong. Figure 2 illustrates the gradation of the mixture. According to the standard Marshall method, the optimum asphalt content was 6.6% by the weight of mixture and the target air void was around 4.5%. Since temperature is an important factor of this study, a rigorous heating pattern was conducted throughout the experiments. For the VAR mixture, the aggregates and AR binder were pre-heated and mixed at 175 °C. To prepare the RARP, a laboratory loose mix aging procedure was used. The VAR was uniformly spread on a plate in loose state, which can achieve an accelerated and uniform aging of the VAR mixture. The loose VAR was first aged at 135 °C for 4 hours as short-term aging and then another 8 hours as long-term aging, which was reported to simulate the asphalt binder aged by 20 hours of pressure vessel aging (PAV) (Chen et al., 2021). To alleviate the possible variability caused by the different gradations of RARP, every set of RARP to be mixed into the RARP-VAR mixture was individually prepared (weighing and mixing) and aged. Afterwards, three mixing temperatures (160/175/190 °C) were adopted for the preparation of AR mixtures containing 40% RARP (RARP40%). For each case of mixing, the following heating and mixing procedure was adopted: 1) pre-heat the virgin aggregates and AR binders at the corresponding mixing temperature and pre-heat the RARP at 135 °C to avoid excessive aging; 2) mix the virgin aggregates larger than 0.075 mm and RARP (if any) in the mixing pot for 30 seconds; 3) add the AR binder and mix for another 60 seconds; and 4) add the aggregates smaller than 0.075 to the mixture and mix for 90 seconds. Besides, a warm mix case was also investigated, as a comparative group to examine the effect of binder's viscosity on the blending efficiency. A foam-based synthetic zeolite WMA additive was directly mixed with AR and aggregates at a lower mixing temperature of 160 ºC. The dosage of the WMA additive is 0.3wt% as recommended by the manufacturer. The foam-based WMA additive was selected because other types of WMA additives may contain light components which could affect the composition of bitumen and FTIR results, thus interfering with the evaluation of bitumen mobilization (Yu et al., 2020b). The WMA additive was added after the blending of AR binder and before the addition of fillers.

Table 1 Basic properties of the AR binder

Measuring Index	Method	Unit	Value
Penetration at 25 ºC	ASTM D5	0.1 mm	40.5
Softening point	ASTM D36	ºC	64.2
Rutting factors (G*/sinδ) at 64 ºC	AASHTO M320	kPa	23.246
Rutting factors (G*sinδ) at 25 ºC	AASHTO M320	MPa	1.327
Viscosity at 135 ºC	AASHTO T316	cP	10377
Viscosity at 160 ºC	AASHTO T316	cP	3308
Non-recoverable creep compliance (J_{nr}) at 64 ºC	AASHTO MP19	/3.2 kPa	0.326

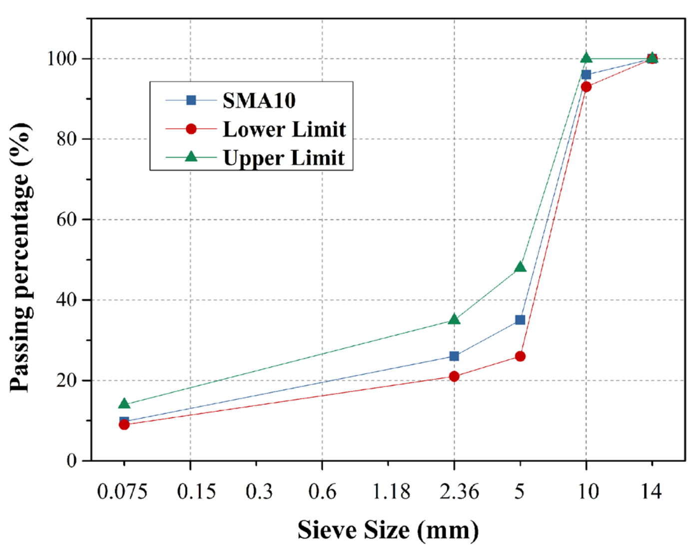


Figure 2. The aggregate gradation of AR and RARP mixture

2.2 Extraction of bitumen and CRM for chemical characterization

To characterize the mobilization of aged AR binder from aged to virgin aggregates, a special mix design was used where the RARP of size between 10-15 mm were first excluded before mixing with virgin aggregates. The missing part of coarse aggregates was supplemented by the virgin aggregates. By doing this, the large virgin aggregates can be easily differentiated and picked out after mixing with RARP. It should be noted that the research targets were not the coarse aggregates but the AR mastic containing AR binder and finer aggregates adhering to the surface of the coarse aggregates, which can reflect how much aged AR binders have been transferred from RARP to virgin aggregates. A recent study has used the similar method to quantify the effective mobilized RAP binder content in hot-in-place recycling (Ma et al., 2021). It should also be noted that this special mix design was only used in the analysis of binder mobilization. In the performance evaluation of mixtures containing different RARP contents, all sizes of RARP were included.

The separation process of CRM and bitumen is shown in Figure 3. Firstly, the coated coarse aggregates were picked out from the RARP-VAR mixture, which were then wrapped by a 200-mesh sieve net and put into the Soxhlet extraction device. The Soxhlet extraction is a method of separating soluble substances from insoluble solids using specific solvents based on the solvent reflux and siphonage which allows an unmonitored and unmanaged operation while efficiently recycling a small amount of solvent (Jensen, 2007). The device is composed of a Soxhlet extractor connected to a condenser on the top and a flask in the bottom soaked into an oil-bath heater. The solvent used in this study is dichloromethane (DCM) because of its very low boiling point (39.5 ^{o}C) and negligible influence on the properties of bitumen and rubber (Li et al., 2022). The extraction process ended when the DCM solvent condensed in the Soxhlet extractor became colorless. Subsequently, the sieve net containing CRM and aggregates can be removed from the extractor. The mixture of CRM and aggregates was then soaked into DCM solvent for separation utilizing their density difference, as the densities of CRM, DCM, and aggregates are around 1.15 g/cm^3, 1.33

g/cm^3, and 2.8 g/cm^3, respectively. Meanwhile, the bitumen was obtained from the DCM solvent in the flask after centrifugation and evaporation.

In this study, chemistry-based evaluation methods were used to characterize the mobilization of bitumen and CRM. The functional group distribution of extracted bitumen was estimated by a PerkinElmer FTIR Spectrometer in the attenuated total reflectance (ATR) mode. The wavenumber range is from 4000 to 400 cm^{-1} with a resolution of 4 cm^{-1}. Five replicates were scanned for each sample. On the other hand, the chemical composition of extracted CRM was determined by TGA. Sufficient blending of the extracted CRM was conducted for a more uniform sampling. A Rigaku instrument (Thermos Plus Evo 8121) with nitrogen gas at a flow rate of 250 ml/min and a heating rate of 10 ^{o}C/min was used. Three replicates were prepared for each test. About 10 mg of sample was heated from ambient temperature to 700 ^{o}C and its weight loss was recorded in this process.

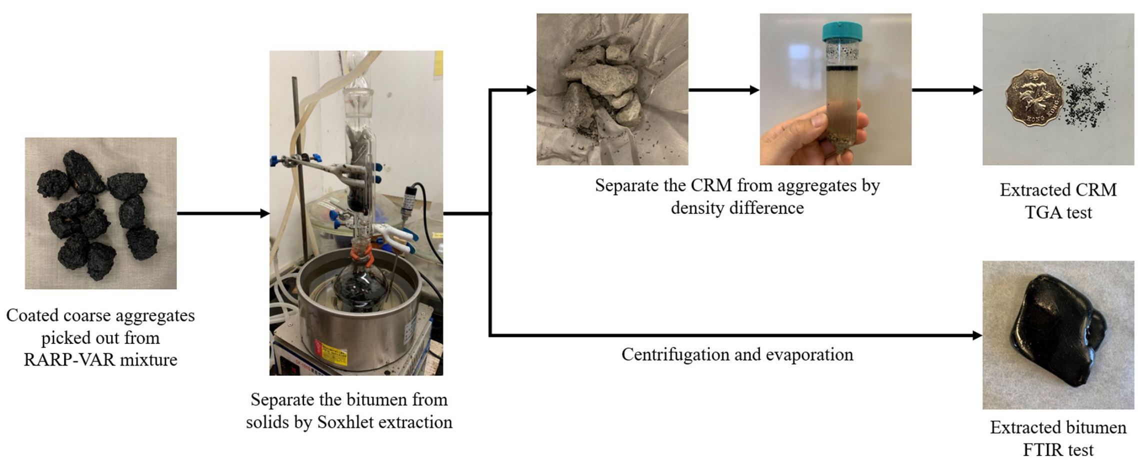


Figure 3. The separation process of CRM and bitumen from mixture

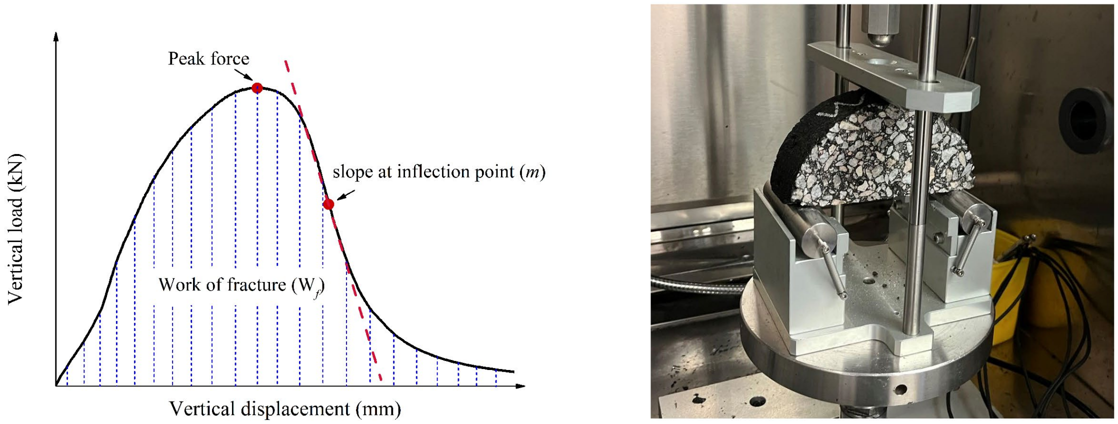
2.3 Cracking resistance test

When RAP is included in new asphalt mixture, the cracking resistance of the new asphalt mixture is the major concern due to the presence of aged/stiffened asphalt binder (Silva et al., 2012). To characterize the cracking resistance of asphalt concrete, SCB tests have become a popular method for its merits of rapid and simple tests and clear observation of crack propagation (Jiang et al., 2020). In Illinois, the cracking resistance is evaluated by the Illinois flexibility index, which is calculated from the fracture energy and post-peak slope using a notched SCB specimen (Ozer et al., 2016a). The proposed Illinois flexibility index has been reported to show considerable consistency and repeatability with various mix design (Ozer et al., 2016b). More recently, discussions have been made on the requirement of notch in the SCB sample by researchers because it is time-consuming and labor intensive. Yan et al. (2020) pointed out that the SCB test using unnotched samples can present results with higher repeatability and less variance than the notched samples. Therefore, the SCB samples without notch were prepared in this study. For each type of mixture, a Superpave gyratory compacted (SGC) specimen with a height of 150 mm and a diameter of 150 mm was prepared and then cut into 4 SCB specimens with a thickness of 50 mm. The SCB tests were conducted at 25 ºC with a constant load-line displacement rate of 50 mm/min using a closed-loop, feedback-controlled servo-hydraulic loading device. Figure 4 shows the schematic of the SCB test. Based on the Illinois flexibility index, a non-notched based flexibility index is calculated using Equations (1) and (2).

$$G_f = \frac{W_f}{Area_{lig}} \times 10^6 \tag{1}$$

$$FI = \frac{G_f}{|m|} \times A \tag{2}$$

where FI is the flexibility index; G_f is the fracture energy (J/m^2) calculated by dividing the work of fracture (W_f) (integral area of the load-displacement curve) by the ligament area (the product of the specimen thickness and the ligament length); m is the slope at the inflection point on the load-displacement curve after the peak load point; and A is for unit conversion, which is 0.01 for lab-compacted specimen.



(a) Typical load-displacement curve of SCB test (b) Testing set-up of SCB test

Figure 4. Schematics of the SCB test: (a) load-displacement curve; (b) testing set-up

2.4 Method for evaluating binder mobilization

2.4.1 Bitumen mobilization index based on FTIR test

As an effective and efficient tool to detect the change of functional group distribution of asphalt binder, FTIR is usually used in combination with asphalt binder extraction methods in research concerning the blending efficiency between RAP and virgin asphalt binders. As mentioned in the literature review, the carbonyl group has been commonly used as indicator of the aging and oxidation of asphalt binder in asphalt chemistry (Hou et al., 2018). For example, Figure 5 compares the carbonyl areas of bitumen extracted from VAR and RARP. A significant augment of the carbonyl area can be found after aging. Therefore, the sensitivity of the carbonyl peak to aging has been utilized to develop the index for quantifying the extent of asphalt binder mobilization (Sreeram et al., 2018). Mathematically, the carbonyl index can be calculated from Equation 3:

$$CI = \frac{A_{C=O}}{A_{ref}} \tag{3}$$

$$A_{ref} = A_{1750/1660} + A_{1030} + A_{(1375,1455)} + A_{1600} + A_{966} + A_{(2953,2862)} + A_{(863,810,750,720)} \tag{4}$$

where $A_{C=O}$ is the normalized integrated area of the carbonyl peak within the wave number range of 1660-1750 cm^{-1}; and A_{ref} is the sum of total considered peak areas for reference (Tarsi et al., 2018). Furthermore, the mobilization index of bitumen (MI_B) can be calculated by the following equation:

$$MI_B = \frac{CI_{Mix} - CI_{VAR}}{CI_{RARP} - CI_{VAR}} \quad (5)$$

where CI is the carbonyl index of bitumen extracted from the coarse aggregates in different mixtures; the subscripts Mix, $RARP$, and VAR refer to the RARP-VAR mixture, RARP, and VAR, respectively. A higher MI_B represents a higher content of aged bitumen mobilized from RAP to virgin aggregates.

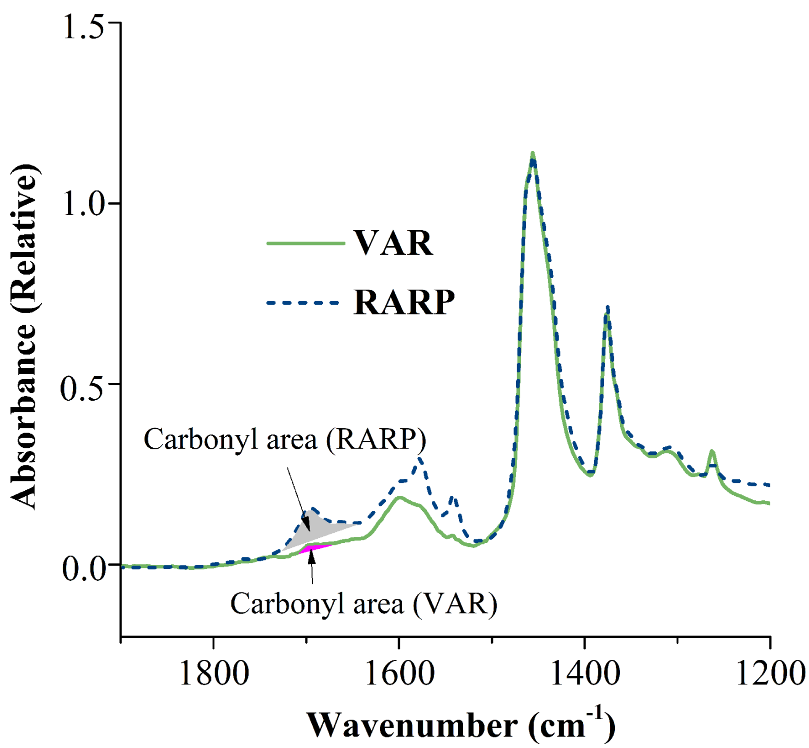


Figure 5. Comparison of the carbonyl peak between VAR and RARP bitumen

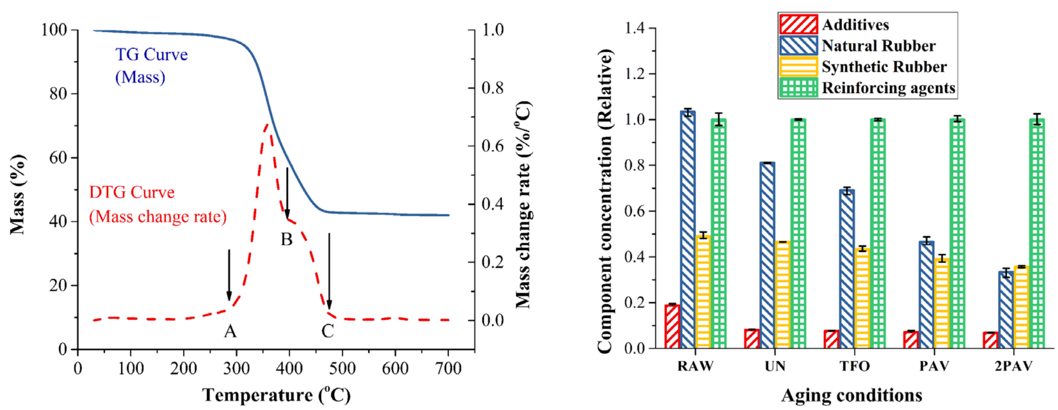
2.4.2 Rubber mobilization index based on TGA test

As aforementioned, the mobilizations of both bitumen and rubber deserve attention during the recycling of RARP because the swelling rubber carries a certain amount of bitumen fractions, and it plays an important role in the mechanical property of AR binder. To evaluate the mobilization of rubber, an aging index of rubber is needed first so that the mobilization index of rubber can be subsequently established, similar to the mobilization index of bitumen built based on the carbonyl index for evaluating asphalt aging. In a previous study, TGA tests were conducted on CRMs extracted from AR binders with different aging conditions to investigate their compositional change during aging (Li et al., 2022). Figure 6(a) shows the TG curve of CRM, which represents the mass loss pattern of CRM during the thermal decomposition process when temperature increased from ambient temperature to 700 ºC. After taking the derivative of the TG curve, the DTG curve shows the mass change rate of CRM, and the three inflection points, A, B, and C can represent the demarcation points of the different components in CRM, namely additives (0-300 ºC), natural rubber (300-400 ºC), synthetic rubber (400-500 ºC), and reinforcing agents including carbon black and inorganic fillers (>500 ºC) (Yao et al., 2016). Therefore, the concentration of each component can be obtained by the mass loss in the corresponding temperature range. In a previous study, the undissolved rubber particles were extracted from AR binders at 5 different aging conditions, including raw state before mixed with bitumen (RAW), unaged (UN), thin film oven short-term aged (TFO), 20 hours of PAV aged (PAV), and 40 hours of PAV aged (2PAV) (Li et al., 2022). Figure 6(b) shows the concentration change of each component from UN to 2PAV conditions when normalized to the content of reinforcing agents. It was found that the content of natural rubber (NR) decreased consistently and rapidly with aging while that of synthetic rubber (SR) only slightly decreased. The different decreasing rates of NR and SR can be attributed to their different molecular structures. The main component of NR is the cis-1,4-polyisoprene, which has a linear, regular, and simple molecular structure. By comparison, SR is synthesized from petroleum byproducts through a polymerization process with more complex molecular networks. The simple structure of NR makes it easier to swell and more vulnerable to chemical degradation, so the proportion of NR decreased faster as shown in the TGA test results (Wang et al., 2020b). More importantly, the discrepant decreasing rates of NR and SR during aging may enable the establishment of an index for evaluating the aging degree of CRM. Thus, an index for evaluating the aging degree of CRM in AR is proposed as follows:

$$RAI = \frac{C_{SR}}{C_{NR}} \tag{6}$$

where RAI is the rubber aging index; C_{NR} is the content ratio of natural rubber; and C_{SR} is the content ratio of synthetic rubber. It is worth noting that the content of reinforcing agents was not included in the calculation of CRM aging, because some extremely small mineral fillers may still be embedded in the CRM after the extraction process, which would interfere with the residual mass of CRM in the temperature range larger than 500 °C, while only using the mass change during the temperature range of rubber degradation can exclude such influences. Therefore, based on the developed rubber aging index, a mobilization index of the CRM (MI_R) can be calculated to assess the extent of aged CRM moving from RARP to virgin aggregates, as shown in Equation 5. A higher MI_R refers to a higher content of mobilized CRM.

$$MI_R = \frac{RAI_{MIX} - RAI_{VAR}}{RAI_{RARP} - RAI_{VAR}} \tag{7}$$



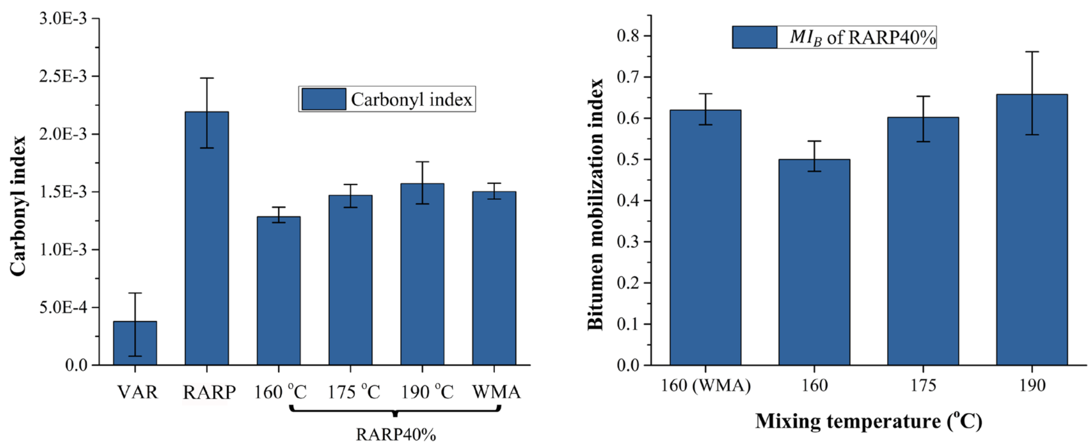
(a) TGA Curve of CRM (b) Compositional change of CRM during aging

Figure 6. Establishment of CRM aging index

3 Results and Discussion

3.1 FTIR test results

The mobilization of bitumen during the mixing of RARP and VAR were analyzed based on the FTIR test results. Figure 7(a) presents the carbonyl indexes of the bitumen extracted from the coarse aggregates of different mixtures. The much larger carbonyl index of RARP compared with VAR indicates the aging level of bitumen during the laboratory loose mix aging procedure. By comparison, the carbonyl indexes of different RARP40% fall between the values of VAR and RARP, indicating partial blending during the mixing. The blending degree was further analyzed by the mobilization indexes of each RARP40% shown in Figure 7(b). When the mixing temperature increased from 160 °C to 190 °C, the bitumen mobilization index gradually increased, which indicates that more aged bitumen can be mobilized from RARP to virgin aggregates. It is also interesting to notice that when the WMA was incorporated in the mixing at 160 °C, the bitumen mobilization index of RARP40% is in the same statistical range with that at 175 °C without the WMA. This is because the foam-based WMA can create a rapid volume expansion of bitumen at high temperatures, which can temporarily decrease the viscosity of bitumen and promote the asphalt coating (Woszuk and Franus, 2017; Zou et al., 2022). The decreased viscosity of bitumen promotes the blending between aged and virgin bitumen. The influences of mixing temperature and WMA additives demonstrate that the aged bitumen has a higher tendency to mobilize at lower viscosity.



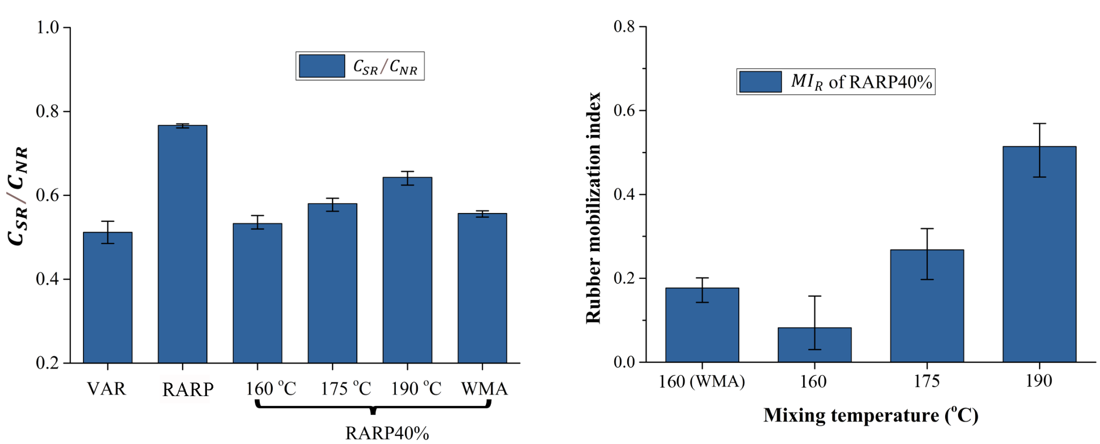
(a) Carbonyl index

(b) Bitumen mobilization index of RARP40%

Figure 7. FTIR test results

3.2 TGA test results

The TGA test results of the CRMs extracted from different mixtures are shown in Fig. 8. The ratio of synthetic rubber to natural rubber (C_{SR}/C_{NR}) can reflect the aging degree of CRM based on their degradation rates. As shown in Fig 8(a), RARP shows much larger C_{SR}/C_{NR} than VAR, which can be attributed to the faster degradation of natural rubber compared to synthetic rubber during the aging process. A series of chemical degradation occurred to the rubber polymer network during aging due to the attacks of oxygen and high temperature, including disentanglement, devulcanization, and depolymerizations (Wang et al., 2020b). By comparison, natural rubber has much simpler molecular structures than synthetic rubber, so the degradation of natural rubber was more severe. These degradation behaviors broke the constraints between rubber polymer chains to let them diffuse out and disperse into the bitumen, which significantly reduced the content of natural rubber in CRM. For RARP40%, their C_{SR}/C_{NR} fall in between the values of VAR and RARP. These results interpret the mobilization of aged rubber during the mixing between RARP and VAR. Subsequently, the rubber mobilization index was calculated as presented in Fig 8(b). It can be found that the rubber mobilization index increased with temperature as well, which indicates that the aged CRMs were more prone to mobilize to virgin aggregates at higher temperatures. When the WMA additive was used, the rubber mobilization index was also improved for the sample at 160 °C, although the increment is not so obvious as the case in the bitumen mobilization index. Therefore, it is reasonable to infer that the viscosity of bitumen can also affect the mobilization of rubber particles in RARP.



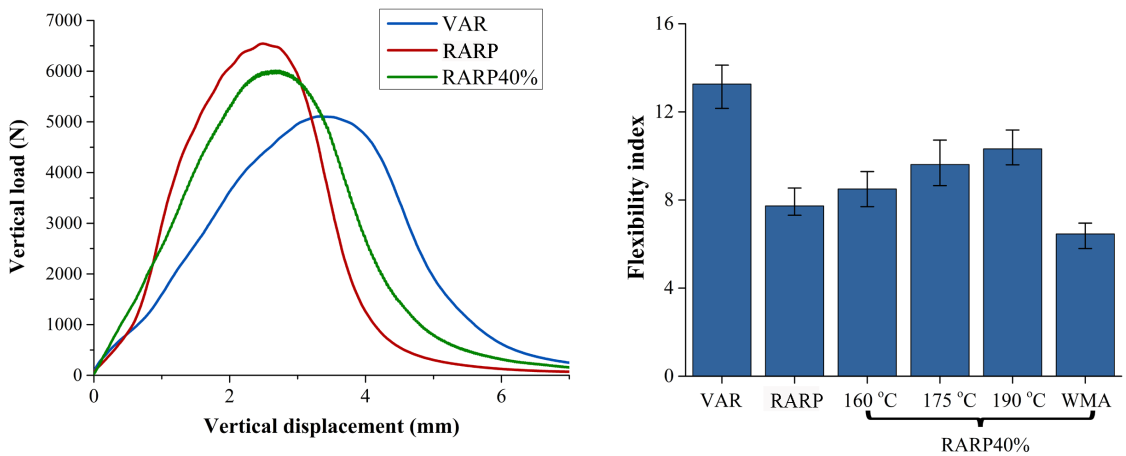
(a) Rubber aging index

(b) Rubber mobilization index of RARP40%

Figure 8. TGA test results

3.3 Cracking resistance

The chemical analysis revealed that the increasing mixing temperature facilitated the mobilization of aged bitumen and rubber. Subsequently, whether the different mixing temperatures affect the cracking resistance was characterized by the flexibility index from the SCB test. Figure 9(a) compares the load-displacement curves of VAR, RARP, and RARP40% mixed at 175 ºC. It is obvious that the peak vertical load of RARP is much larger than that of VAR due to the stiffer AR binder in RARP. The stiffened AR binder lost certain relaxation capacity and became brittler after aging, which lead to a faster deteriorating rate of the post-peak load referring to the faster crack propagation (Jiang et al., 2018). The flexibility indexes of different samples are shown in Fig 9(b), where a higher flexibility index indicates a better resistance of the asphalt mixture to fracture at an intermediate temperature. VAR owned the largest flexibility index of 14.24, while that of RARP was only 7.72. The decreasing of flexibility index indicates that the stiffening of AR binder during the loose mix aging resulted in the decay of cracking resistance of AR mixture. When it comes to RARP40%, the cracking resistance is between those of RARP and VAR. It is interesting to notice the upward tendency of the flexibility index when the mixing temperature increased from 160 ºC to 190 ºC. However, the warm mix RARP40% shows the worst cracking resistance among all samples. It was unexpected because the chemical analysis revealed that the incorporation of WMA significantly improved the binder mobilization. Thus, it can be deduced that the foam-based WMA impaired the cracking resistance of RARP40%. The foam-based WMA used in this study is a very fine powder of porous and hydrated aluminosilicates containing about 21% crystalline water by weight. When added to the asphalt mix at high temperatures, the crystalline water is released into the mixture, causing foaming effect and reducing the viscosity of asphalt binder. However, it may also damage the adhesive capacity between aggregate and asphalt binder, thus resulting in the poorer cracking resistance of asphalt mixture (Cui et al., 2020). Excluding the influence of WMA, the increased flexibility index of RARP40% with temperature preliminarily proved the role of binder mobilization in affecting the cracking resistance of RARP mixture, but further correlation analysis is required.



(a) Load-displacement curves

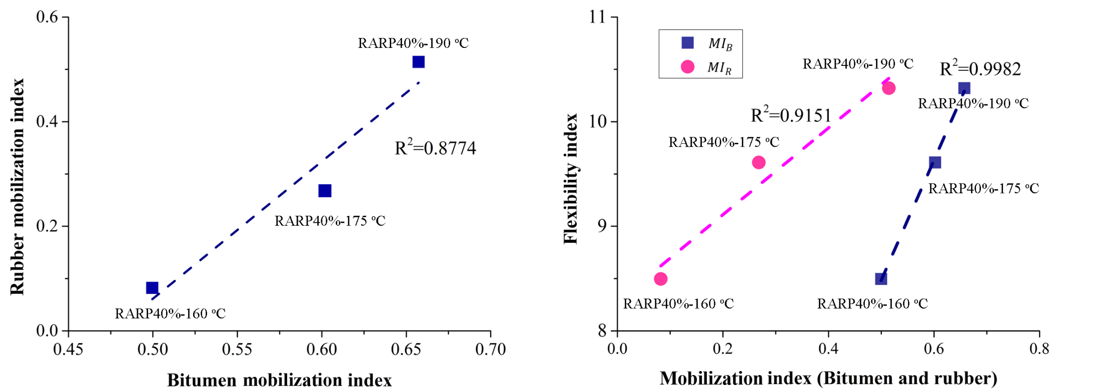
(b) Flexibility index

Figure 9. SCB test results

4 Correlation Analysis between Binder Mobilization and Cracking Resistance

The discussion above has exhibited the different mobilizations of binder and CRM during the blending of RARP and VAR and the resultant different cracking resistances of the RARP40% mixtures produced under different conditions. A follow-up correlation analysis was further conducted to understand the dependence of the performance of RARP40% on the blending efficiency.

Fig 10(a) illustrates the correlation between the mobilization indexes of bitumen and rubber. The common growth of these two indexes with temperature proves that the CRM can mobilize synchronously with bitumen from RARP to the virgin aggregates. The correlations between the flexibility index and these two mobilization indexes are shown in Fig 10(b), respectively. Both indexes show good correlations with the flexibility index while the bitumen mobilization index is even better. Such good correlations reveal the potential effect of blending efficiency on the cracking resistance of AR mixture containing RARP. A higher mixing temperature can activate more aged bitumen and CRM to mobilize from aged aggregates to coat the virgin aggregates, so as to promote the diffusion between the aged and virgin bitumen, and in the case of AR mixture, the blending of aged and virgin CRM (Ding et al., 2016). The improved blending efficiency between aged and virgin AR binders partially rejuvenated the aged AR binder, thus positively affecting the cracking resistance of RARP40%. Comparing the two mobilization indexes, the bitumen mobilization index based on FTIR test is still recommended for evaluating the blending efficiency of RARP mixture because of its easier sample collection, rapid testing, and better correlation with the cracking resistance.



(a) Correlation between MI_B and MI_R (b) Correlations between cracking resistance and blending efficiency

Figure 10. Correlation analysis

5 Findings and Recommendations

This study investigated the mobilization of aged bitumen and CRM during the recycling of RARP at different mixing temperatures and its influence on the cracking resistance of RARP mixture. The mobilizations of bitumen and CRM were characterized by FTIR and TGA tests while the cracking resistance was evaluated by SCB test, followed by a correlation analysis. The following points summarize the main findings of this study:

- The mobilization indexes of bitumen and rubber were established based on FTIR and TGA tests. The former increased from 0.50 to 0.66 while the latter increased from 0.08 to 0.51 when the mixing temperature increased from 160 ºC to 190 ºC. The R^2 between the two indexes is 0.8774, which proved that the aged rubber can be mobilized simultaneously with the aged bitumen from RARP to virgin aggregates.

- When the same proportion of RARP is incorporated into AR mixture, a higher mixing temperature could improve the cracking resistance of the RARP mixture, as indicated by the SCB test. The flexibility index of RARP40% increased from 8.50 to 10.32 when the mixing temperature increased from 160 ºC to 190 ºC.
- Both mobilization indexes correlate well with the flexibility index, which indicates the positive dependence of cracking resistance on the blending efficiency of RARP. By comparison, the bitumen mobilization index is recommended for its higher R^2 (0.9982) and easier sample preparation.
- The foam-based WMA additive markedly improved the blending efficiency but compromised the cracking resistance of RARP40% due to the negative effect of released water on the adhesive capacity of AR binder. The WMA-RARP40% shows the lowest flexibility index of 6.46.

To sum up, this study serves as a critical step before the investigation of the rejuvenation design of RARP. The synchronous mobilization of bitumen and CRM has important implications that the rejuvenation of both aged bitumen and rubber polymers should be considered when recycling RARP into new AR pavement. However, the findings of this study are based on laboratory-prepared artificial RARP due to the practical constraints. In-situ RARP should be used when it is available. Although the improved blending efficiency at higher temperature can contribute to a better cracking resistance, the stiffened aged AR binder still inevitably deteriorated the cracking resistance of RARP40% compared to the virgin AR mixture. In future studies, the rejuvenation of the aged AR binder can be attempted by combing rejuvenators and extra rubber to properly supplement the bitumen light fractions and effective rubber content in the RARP mixture, which can repair the rheological property of RARP binder and further promote the recycling of waste tires.

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References

ASTM Standard D 6114, 2009. Standard specification for asphalt-rubber binder. American Society for Testing and Materials.

Apostolidis, P., Liu, X., Kasbergen, C., Scarpas, A.T., 2017. Synthesis of asphalt binder aging and the state of the art of antiaging technologies. Transp. Res. Rec.: J. Transp. Res. Board. 2633, 147–153. https://doi.org/10.3141/2633-17.

Aurangzeb, Q., Al-Qadi, I.L., Ozer, H., Yang, R., 2014. Hybrid life cycle assessment for asphalt mixtures with high RAP content. Resources, Conservation and Recycling 83, 77–86. https://doi.org/10.1016/j.resconrec.2013.12.004

Bowers, B.F., Moore, J., Huang, B., Shu, X., 2014. Blending efficiency of reclaimed asphalt pavement: an approach utilizing rheological properties and molecular weight distributions. Fuel 135, 63-68.

Behnood, A., 2019. Application of rejuvenators to improve the rheological and mechanical properties of asphalt binders and mixtures: A review. Journal of Cleaner Production 231, 171–182. https://doi.org/10.1016/j.jclepro.2019.05.209

Chen, H., Zhang, Y., Bahia, H.U., 2021. The role of binders in mixture cracking resistance measured by ideal-CT test. International Journal of Fatigue 142, 105947. https://doi.org/10.1016/j.ijfatigue.2020.105947

Cui, W., Huang, W., Hu, B., Xie, J., Xiao, Z., Cai, X., Wu, K., 2020. Investigation of the Effects of Adsorbed Water on Adhesion Energy and Nanostructure of Asphalt and Aggregate Surfaces Based on Molecular Dynamics Simulation. Polymers 12, 2339.

https://doi.org/10.3390/polym12102339
Ding, Y., Huang, B., Shu, X., Zhang, Y., Woods, M.E., 2016. Use of molecular dynamics to investigate diffusion between virgin and aged asphalt binders. Fuel 174, 267–273. https://doi.org/10.1016/j.fuel.2016.02.022
Ding, Y., Huang, B., Shu, X., 2018. Blending efficiency evaluation of plant asphalt mixtures using fluorescence microscopy. Construction and Building Materials 161, 461–467. https://doi.org/10.1016/j.conbuildmat.2017.11.138
Gao, J., Yang, J., Yu, D., Jiang, Y., Ruan, K., Tao, W., Sun, C., Luo, L., 2021. Reducing the variability of multi-source reclaimed asphalt pavement materials: A practice in China. Construction and Building Materials 278, 122389. https://doi.org/10.1016/j.conbuildmat.2021.122389
Hou, X., Lv, S., Chen, Z., Xiao, F., 2018. Applications of Fourier transform infrared spectroscopy technologies on asphalt materials. Measurement 121, 304–316. https://doi.org/10.1016/j.measurement.2018.03.001
Hettiarachchi, C., Hou, X., Xiang, Q., Yong, D., Xiao, F., 2020. A blending efficiency model for virgin and aged binders in recycled asphalt mixtures based on blending temperature and duration. Resources, Conservation and Recycling 161, 104957. https://doi.org/10.1016/j.resconrec.2020.104957
Jamshidi, A., Kurumisawa, K., Nawa, T., Igarashi, T., 2016. Performance of pavements incorporating waste glass: The current state of the art. Renewable and Sustainable Energy Reviews 64, 211–236. https://doi.org/10.1016/j.rser.2016.06.012
Jensen, W.B., 2007. The Origin of the Soxhlet Extractor. J. Chem. Educ. 84, 1913. https://doi.org/10.1021/ed084p1913
Jiang, J., Dong, Q., Ni, F., Zhao. Y., 2018. Effects of Loading Rate and Temperature on Cracking Resistance Characteristics of Asphalt Mixtures Using Nonnotched Semicircular Bending Tests. Journal of Testing and Evaluation 47, 4. https://doi: 10.1520/JTE20170711

Jiang, J., Li, Y., Zhang, Y., Bahia, H.U., 2020. Distribution of mortar film thickness and its relationship to mixture cracking resistance. International Journal of Pavement Engineering 0, 1–10. https://doi.org/10.1080/10298436.2020.1774767

Kim, M., Mohammad, L.N., Elseifi, M.A., 2012. Characterization of Fracture Properties of Asphalt Mixtures as Measured by Semicircular Bend Test and Indirect Tension Test. Transportation Research Record 2296, 115–124. https://doi.org/10.3141/2296-12

Li, D., Leng, Z., Zou, F., Yu, H., 2021. Effects of rubber absorption on the aging resistance of hot and warm asphalt rubber binders prepared with waste tire rubber. Journal of Cleaner Production 303, 127082. https://doi.org/10.1016/j.jclepro.2021.127082

Li, D., Leng, Z., Wang, H., Chen, R., Wellner, F., 2022. Structural and Mechanical Evolution of the Multiphase Asphalt Rubber during Aging based on Micromechanical Back-Calculation and Experimental Methods. Materials & Design 110421. https://doi.org/10.1016/j.matdes.2022.110421

Ma, Y., Polaczyk, P., Hu, W., Zhang, M., Huang, B., 2021. Quantifying the effective mobilized RAP content during hot in-place recycling techniques. Journal of Cleaner Production 314, 127953. https://doi.org/10.1016/j.jclepro.2021.127953

Ma, Y., Polaczyk, P., Park, H., Jiang, X., Hu, W., Huang, B., 2020. Performance evaluation of temperature effect on hot in-place recycling asphalt mixtures. Journal of Cleaner Production 277, 124093. https://doi.org/10.1016/j.jclepro.2020.124093

Ozer, H., Al-Qadi, I.L., Lambros, J., El-Khatib, A., Singhvi, P., Doll, B., 2016a. Development of the fracture-based flexibility index for asphalt concrete cracking potential using modified semi-circle bending test parameters. Construction and Building Materials 115, 390–401. https://doi.org/10.1016/j.conbuildmat.2016.03.144

Ozer, H., Al-Qadi, I.L., Singhvi, P., Khan, T., Rivera-Perez, J., El-Khatib, A., 2016b. Fracture Characterization of Asphalt Mixtures with High Recycled Content Using Illinois Semicircular Bending Test Method and Flexibility Index. Transportation Research Record 2575, 130–137.

https://doi.org/10.3141/2575-14

Roja, K.L., Masad, E., Mogawer, W., 2021. Performance and blending evaluation of asphalt mixtures containing reclaimed asphalt pavement. Road Materials and Pavement Design 22, 2441–2457. https://doi.org/10.1080/14680629.2020.1764858

Silva, H.M.R.D., Oliveira, J.R.M., Jesus, C.M.G., 2012. Are totally recycled hot mix asphalts a sustainable alternative for road paving? Resources, Conservation and Recycling 60, 38–48. https://doi.org/10.1016/j.resconrec.2011.11.013

Sreeram, A., Leng, Z., Yuan, Z., Padhan, R.K., 2018. Evaluation of RAP binder mobilisation and blending efficiency in bituminous mixtures: An approach using ATR-FTIR and artificial aggregate. Constr. Build. Mater. 179, 245-253. https://doi.org/10.1016/j.conbuildmat.2018.05.154

Sreeram, A., Leng, Z., 2019. Variability of rap binder mobilisation in hot mix asphalt mixtures. Constr. Build. Mater. 201, 502–509. https://doi.org/10.1016/j.conbuildmat.2018.12.212

Tarsi, G., Varveri, A., Lantieri, C., Scarpas, A., Sangiorgi, C., 2018. Effects of Different Aging Methods on Chemical and Rheological Properties of Bitumen. Journal of Materials in Civil Engineering 30, 04018009. https://doi.org/10.1061/(ASCE)MT.1943-5533.0002206

Wang, H., Liu, X., Apostolidis, P., van de Ven, M., Erkens, S., Skarpas, A., 2020a. Effect of laboratory aging on chemistry and rheology of crumb rubber modified bitumen. Mater Struct 53, 26. https://doi.org/10.1617/s11527-020-1451-9

Wang, H., Apostolidis, P., Zhu, J., Liu, X., Skarpas, A., Erkens, S., 2020b. The role of thermodynamics and kinetics in rubber–bitumen systems: A theoretical overview. Int. J. Pavement Eng. https://doi.org/10.1080/10298436.2020.1724289.

Woszuk, A., Franus, W., 2017. A Review of the Application of Zeolite Materials in Warm Mix Asphalt Technologies. Appl. Sci. 7, 293. https://doi:10.3390/app7030293.

Xu, X., Leng, Z., Lan, J., Wang, W., Yu, J., Bai, Y., Sreeram, A., Hu, J., 2021. Sustainable Practice in Pavement Engineering through Value-Added Collective Recycling of Waste Plastic and Waste Tyre Rubber. Engineering 7, 857–867. https://doi.org/10.1016/j.eng.2020.08.020

Yu, H., Chen, Y., Wu, Q., Zhang, L., Zhang, Z., Zhang, J., Miljković, M., Oeser, M., 2020a. Decision support for selecting optimal method of recycling waste tire rubber into wax-based warm mix asphalt based on fuzzy comprehensive evaluation. Journal of Cleaner Production 121781. https://doi.org/10.1016/j.jclepro.2020.121781

Yu, H., Leng, Z., Zhang, Z., Li, D., Zhang, J., 2020b. Selective absorption of swelling rubber in hot and warm asphalt binder fractions. Constr. Build. Mater. 238, 117727. https://doi.org/10.1016/j.conbuildmat.2019.117727.

Yan, C., Zhang, Y., Bahia, H.U., 2020. Comparison between SCB-IFIT, un-notched SCB-IFIT and IDEAL-CT for measuring cracking resistance of asphalt mixtures. Construction and Building Materials 252, 119060. https://doi.org/10.1016/j.conbuildmat.2020.119060

Yao, H., Zhou, S., Wang, S., 2016. Structural evolution of recycled tire rubber in asphalt. Journal of Applied Polymer Science 133. https://doi.org/10.1002/app.42954

Zou, F., Leng, Z., Cao, R., Li, G., Zhang, Y., Sreeram, A., 2022. Performance of zeolite synthesized from sewage sludge ash as a warm mix asphalt additive. Resources, Conservation and Recycling 181, 106254. https://doi.org/10.1016/j.resconrec.2022.106254