Investigation of Rejuvenation Mechanisms of Reclaimed Asphalt Rubber Pavement through Mortar Tests

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Conventional rejuvenation methods recover the rheological properties of aged bitumen by adding rejuvenators to supplement the content of light fractions of bitumen. However, these methods cannot fully restore the microstructure and advantages of aged asphalt rubber (AR) binder due to the dissolution of rubber particles. Aiming at discovering a more suitable rejuvenation method for the reclaimed asphalt rubber pavement (RARP), this study investigates the rejuvenation mechanisms of RARP through a series of chemical and rheological tests at the mortar level. A chemistry-based method was proposed to estimate the rubber dissolution in RARP mortar. AR mortars containing 40% of RARP mortar (RARP40%) were prepared. Three rejuvenation schemes were adopted for RARP40%, namely recovering the light fractions, supplementing the swelling rubber content, and both recovering and supplementing, respectively. The results indicate that adding rejuvenator only can effectively soften the aged RARP binder but it fails to slow down the rheological evolution of RARP40% mortar in the secondary aging. The incorporation of AR binder with extra rubber content notably improved the aging resistance but significantly compromised the workability of RARP40% mortar. The compound rejuvenation scheme was the optimum solution that balanced the rejuvenation effectiveness, aging resistance, and workability. The findings of this study led to a new strategy for the rejuvenation design of RARP.

Keywords: Recycling, Tire Rubber, Reclaimed Asphalt Rubber Pavement, Rejuvenation, Mortar, Rheological properties.

1 Introduction

The global production of waste tires has constantly grown due to the increasing demand for transportation in parallel with the developing economy. It is estimated that more than 1.5 billion waste tires are generated annually worldwide (Mohajerani et al. 2020). Known as the “black pollution,” the inherent characteristics of waste tires make them very difficult to degrade, and the stockpiles of waste tires can result in worrying problems, such as fire accident and mosquito breeding. To properly dispose of waste tires, one prevailing technology in the pavement industry is to recycle them into asphalt paving materials. Asphalt rubber (AR) is a composite of raw bitumen and crumb rubber modifier (CRM) recycled from waste tires, in which the CRM content is at least 15% by weight of the total bitumen blend (ASTM 2009). As a sustainable paving material, AR provides excellent rutting and cracking resistances, in addition to helping recycle waste tires (Xu et al. 2021). Thus, it has gained fast-growing interest during the past decades in many regions. However, the widespread application of AR faces one inevitable problem: the unclear recyclability of AR pavement at the end of its service life.

Recycling reclaimed asphalt pavement (RAP) into new asphalt mixture has become routine in pavement industry for its significant economic and environmental benefits (Aurangzeb et al. 2014). From the perspective of mechanical property, the incorporation of RAP has its pros and cons because the rheological properties of asphalt binder change significantly after aging. The chemical composition of bitumen changes dramatically during aging due to oxidation, loss of volatile components, and increase of asphaltene content (Subhy et al. 2018). The increased polar components cause an increase of the colloidal agglomerates, which significantly stiffens bitumen. The increased stiffness of aged asphalt binder can improve the resistance of asphalt mixture to permanent deformation, but it also compromises its resistance to cracking damage (Aghapour et al. 2020). Therefore, when a high percentage of RAP is incorporated or the RAP binder is severely aged, a rejuvenation design is needed to recover the rheological properties of aged asphalt binder. The aim of rejuvenation is to restore the microstructure of aged asphalt binder by decreasing the asphaltenes to maltenes ratio, so rejuvenators are usually softening additives composed of light oily components (Behnood 2019). Through recovering the maltenes content, the relaxation capacity and flexibility of RAP binder can be repaired to improve the cracking resistance of RAP-included mixture.

When it comes to the recycling of reclaimed asphalt rubber pavement (RARP), the complicated aging mechanisms of AR binder need to be understood first. As illustrated in Figure 1, AR binder has a special binary system due to the swelling of rubber polymer network absorbing bitumen light fractions during the high-temperature blending (Yu et al. 2020). The absorbed bitumen fractions were reported to maintain a relatively softer environment around the swelling rubber to offset the stiffening effect of bitumen aging (Li et al. 2021). Previous studies have also found that the aging of AR binder is more than just the evolution of mechanical property, but also the continuous change of inner structure with decreasing swelling rubber content due to the chemical degradation of rubber polymer network (Li et al. 2022a). The chemical degradation refers to the breakage of crosslink bonds and backbone chain bonds as well as the chain disentanglement of rubber polymer network during aging under the thermal and oxygen attacks, which causes the dissolution of rubber particles (Wang et al. 2020a). The molecular weight of bitumen increases during aging, while the degradation of swelling rubber gradually releases the absorbed bitumen light fractions into the liquid phase to function as softening agent (Noorvand et al. 2021). Meantime, the fragment of devulcanized natural rubber released in bitumen may also soften the aged bitumen due to its linear and flexible molecular structure (Wang et al. 2020a). Together with the other components in CRM like carbon black and antioxidants that can prevent the oxidation of bitumen, AR binder has been found to own superior aging resistance with more stable rheological properties throughout aging than the conventional bitumen and polymer modified bitumen (Wang et al. 2020b). However, it is inevitable that the enhanced aging resistance of AR will decay after the excessive dissolution of rubber polymer. In short, the aging of AR binder is a process where the bitumen hardening and loss of effective rubber content occur simultaneously. Such complicated aging evolutions bring more uncertainties to the rejuvenation design of RARP compared with the conventional RAP. Given the circumstances, the conventional rejuvenation method of simply supplementing light components may not effectively recover the microstructure and main advantages of RARP binder (Faisal Kabir and Fini 2021). Simply adding rejuvenator cannot reverse the reduced rubber content in RARP, so the aging resistance of the new AR mixture containing RARP may be compromised. Limited attempts have been made to explore a customized rejuvenation method for the RARP to fill this gap. Whether adding extra CRM can restore the structural and rheological properties of RARP is a noteworthy question for improving the aging resistance of RARP and further promoting the recycling of waste tires.

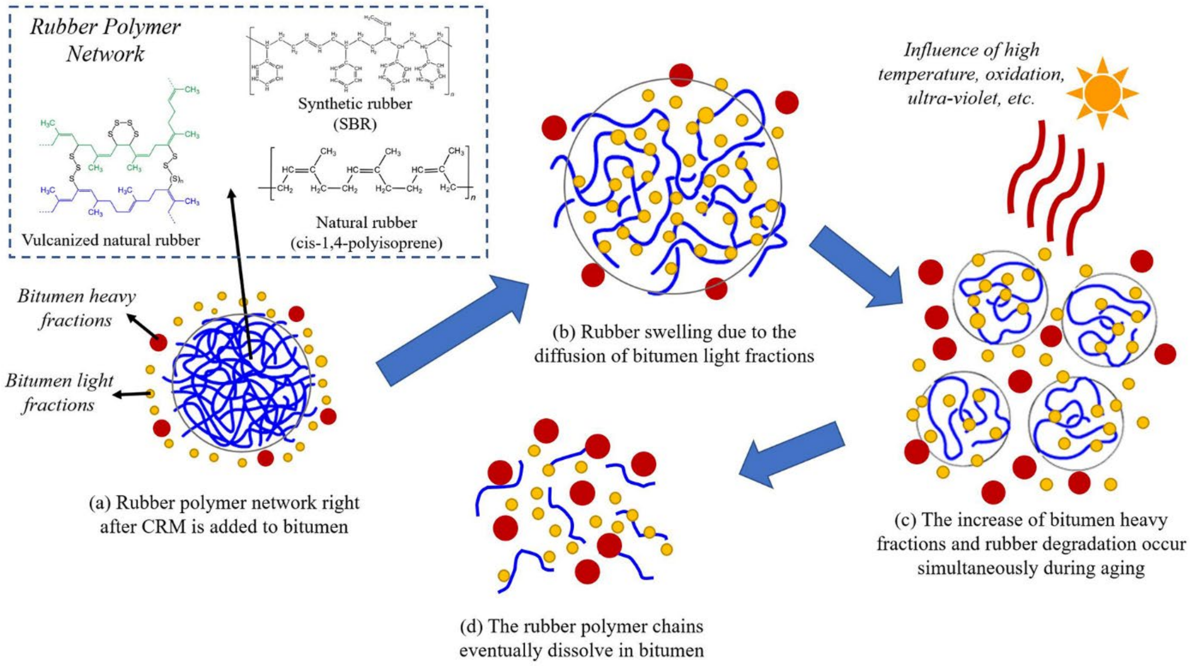


Figure 1. Schematic of the aging behaviors of AR binder.

On the other hand, the binary system of AR binder also leads to difficulties in the extraction and evaluation of RARP binder. The common extraction method using chemical solvent has the problem of destroying the gel structure in swelling rubber, and only the bitumen phase can be dissolved and extracted from the aggregates after centrifugation. Consequently, the RARP binder cannot be intactly extracted for the evaluation of rheological properties through this method, which also limits the common use of blending chart to determine the dosage of rejuvenator. To overcome this limitation, tests conducted on asphalt mortar, also known as fine aggregate matrix (FAM), can be considered an alternative approach. Asphalt mortar consists of asphalt binder, filler, and fine aggregates smaller than the minimum aggregate size in the aggregate skeleton, which is regarded as an intermediate state between the asphalt mixture and asphalt binder (Underwood and Kim 2013). Asphalt mortar tests are more labor-saving than asphalt mixture tests and offer a solvent-free method to measure the rheological property of asphalt binder in RAP-included mixture. He et al. used the mortar test to characterize the rheological property of blended binders in mixes containing high quantities of RAP (He et al. 2016). Zhang and Leng evaluated the effects of aging on the mortars prepared with aggregates smaller than 0.5 mm and different kinds of asphalt binders (Zhang and Leng, 2017). It was found that the mortar testing showed high repeatability in testing results, and the effect of aging can be clearly identified.

Therefore, asphalt mortar test can also be regarded as a promising solution for the assessment of RARP binder, so as to facilitate the rejuvenation design of RARP mixture.

The objective of this study is to investigate the rejuvenation mechanisms of RARP with different rejuvenation schemes through mortar level tests. Figure 2 shows the flowchart of this study. AR mortar was prepared, and artificial RARP mortar was produced by pressure aging vessel (PAV) of different durations. The CRM dissolution extent in RARP mortar was indirectly estimated by the Soxhlet extraction and thermal gravimetric analysis (TGA) tests. AR mortars containing 40% of RARP (RARP40%) were prepared for comparing the effects of different rejuvenation schemes, which include two individual rejuvenation schemes and one compound rejuvenation scheme. The idea was to rejuvenate the RARP by either or both the following methods: adding rejuvenator to recover the bitumen light fractions and incorporating AR binder with extra rubber content to supplement the degraded rubber content. Frequency sweep tests were conducted to assess the effectiveness of different rejuvenation schemes. All mortar samples were subjected to a secondary aging to evaluate the effects of different rejuvenation schemes on aging resistance. The workability was evaluated by the mortar viscosity tests. The findings of this study are expected to provide the theoretical foundation for a more sustainable and durable rejuvenation design of RARP.

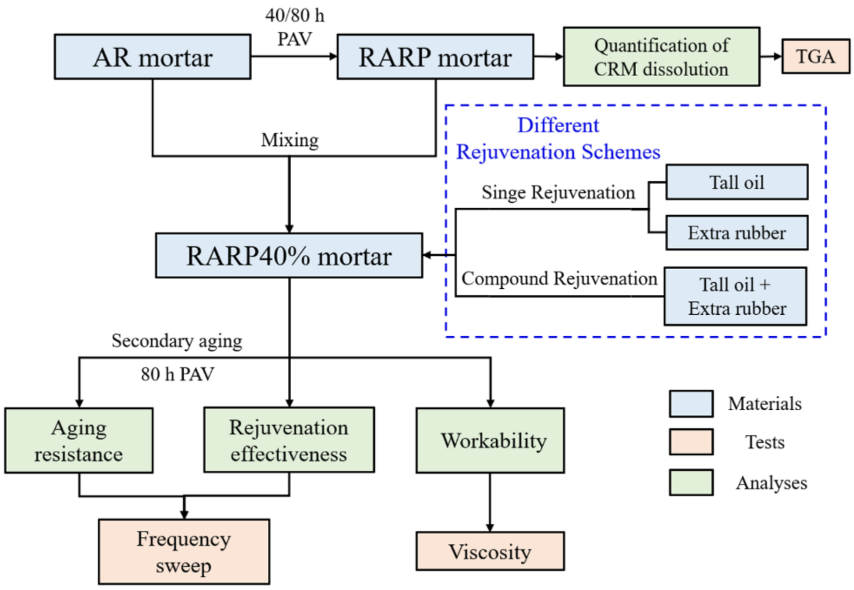


Figure 2. Flowchart of research plan.

2 Experimental Program

2.1 Materials

In this study, AR mortar was prepared with a high-viscosity AR binder and granite aggregates passing the No. 16 sieve (<1.18 mm). The raw materials included CRM ambiently ground from end-of-life truck tires with a size of 0.3-0.6 mm and raw bitumen with a penetration grade of 60/70 (Pen60/70). The AR binder was then produced by using a high shear mixer to blend 20% CRM (by mass of raw bitumen) into raw bitumen at 180 ºC and 4,000 rpm for 60 min, whose basic physical and rheological properties are shown in Table 1. In Hong Kong, AR is usually used in wearing course with gap-graded mix designs, so the proportion of aggregates smaller than 1.18 mm in a stone mastic asphalt mix with a nominal maximum aggregate size of 10 mm (SMA10) was adopted as the gradation of AR mortar. When performing the mortar level evaluation, the composition of the fabricated mortar sample should be as close as possible to its actual state in the asphalt mixture. Thus, the determination of filler and binder contents in mortar is of great significance because part of them are adhering to the coarse aggregates rather than existing in mortar. Underwood and Kim developed a microstructural hypothesis based on meso-gravimetric and sieved ignition tests to calculate the effective binder contents in asphalt mortar, which assumed that the asphalt binder coats all aggregate particles with a uniform film thickness (Underwood and Kim 2013). For the SMA10 mix design with an AR binder content of 6.7% used in Hong Kong, the AR mortar with aggregates smaller than 1.18 mm was calculated to contain 24.26% of binder and 30.41% of filler. The composition of AR mortar is summarized in Table 2. To prepare the AR mortar, the AR binder, fine aggregates, and fillers were first pre-heated at 175 ºC for 1 hour. Then, the fine aggregates and AR binder were blended for 30 s, followed by adding the fillers for another 60 s of blending.

After the preparation of AR mortar, RARP mortar was artificially prepared by laboratory aging methods. 50 g of AR mortar was poured on a plate with a diameter of 140 mm. The short-term aging was conducted by storing the loose AR mortar in an oven of 163 ºC for 2 hours. As for the long-term aging, Ding et al. suggested that when the aging duration is longer than 40 h, asphalt mortar can be reliably aged by PAV where the binder in mortar can be sufficiently aged (Ding et al. 2022). Thus, the short-term aged AR mortar was aged by PAV at 100 °C with an air pressure of 2.1 MPa for 40 h and 80 h, respectively.

Table 1 Basic properties of the original AR binder.

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

Table 2 Material composition of AR mortar.

Components	Fine aggregates (mm)				Filler	Binder content
	1.18-0.6	0.6-0.3	0.3-0.15	0.15-0.075		
Proportion (%)	18.50	12.95	8.33	5.55	30.41	24.26

2.2 Estimation of the undissolved CRM content in AR mortar

As aforementioned, the swelling rubber content decreases during aging due to the dissolution of CRM particles. The dissolution of CRM particles refers to their size reduction attributed to the degradation of rubber polymers, physical split, and component loss during mixing and aging. In many studies considering AR binder as a binary system, the undissolved CRM particles are defined as the CRM particles that cannot pass the 200-mesh sieve net (CRM particle size > 0.075 mm). Li et al. found that the Soxhlet extraction method using dichloromethane (DCM) solvent can extract and measure the undissolved CRM contents in AR binders at different aging conditions (Li et al. 2022a). Soxhlet extraction is a conventional laboratory method used for separating soluble components from insoluble solids by a small amount of specific solvent based on solvent reflux and siphonage (Jensen 2007). However, the direct measurement of undissolved CRM content in AR mixture is difficult because the Soxhlet extraction can only separate the bitumen phase that is dissolved in the solvent while the insoluble CRM remains with aggregates. Although CRM can be further separated from aggregates based on their density difference, a complete separation is challenging because some extremely fine aggregates and fillers may be embedded in the CRM particles. Therefore, this study proposes a chemistry-based method to indirectly estimate the dissolution extent of CRM in AR and RARP mortar.

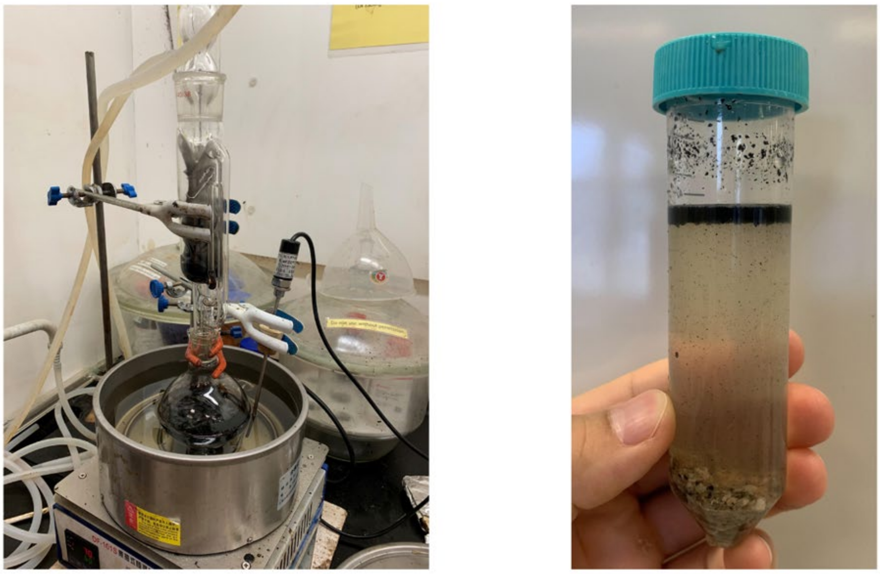
To do so, the relationship between the undissolved CRM content and rubber degradation index of AR binder in a wide range of aging conditions should be established first. A series of laboratory aging was conducted on the same AR binder used for preparing the AR mortar. The undissolved CRM in AR binders of different aging conditions were then extracted by the Soxhlet extraction device shown in Figure 3(a). A Soxhlet extractor was connected to a flask in the bottom and a condenser on the top. The flask was heated by an oil-bath of 60 °C of the condenser was circulating with cooling water. AR binder was wrapped by a 200-mesh sieve net inside the Soxhlet chamber. The undissolved CRM content can be calculated using the following equation.

$$C_{UR} = \frac{m_{sox} - m_{net}}{m_{AR}} \times 100\% \quad (1)$$

where C_{UR} is the undissolved CRM content; m_{sox} is the mass of sieve net containing undissolved CRM after the Soxhlet extraction; m_{net} is the mass of the sieve net; and m_{AR} is the mass of the whole AR binder before Soxhlet extraction. In total six aging conditions of CRM were obtained, including the raw CRM (before mixing with bitumen), unaged, short-term aged (thin film oven aged, TFO), and three different durations of PAV aged (20/40/80 hours of PAV). Furthermore, TGA tests were conducted on the undissolved CRM particles using nitrogen gas at a flow rate of 250 ml/min and a heating rate of 10 °C. About 10 mg of sample was used in each test and three replicates were prepared for each kind of CRM particles. Figure 4(a) shows a typical mass change curve of CRM and its mass change rate curve in TGA test. It can be seen that the mass of CRM rapidly decreased in the temperature range of 300-500 °C due to the thermal decomposition of rubber polymers. Three inflection points A, B, and C on the mass change rate curve can be used to demarcate the different degradation stages of natural rubber (300-400 °C) and synthetic rubber (400-500 °C) (Yao et al. 2016). Figure 4(b) shows the concentration changes of natural rubber and synthetic rubber in CRM during aging. From raw state to 80 hours of PAV aged condition, the concentration of natural rubber rapidly decreased while that of synthetic rubber barely changed due to the more complex molecular structure of synthetic rubber (Wang et al. 2020a). Such a clear trend has been used in a previous study to establish the degradation index of CRM for quantifying the mobilization extent of aged CRM particles during the recycling of RARP mixture (Li et al. 2022b). The rubber degradation index evaluates the degradation degree of rubber polymers in CRM particles, which can be calculated by Equation 2.

$$RDI = \frac{C_{SR}}{C_{NR}} \tag{2}$$

where RDI is the rubber degradation index; C_{NR} is the concentration of natural rubber in CRM; and C_{SR} is the concentration of synthetic rubber. A higher RDI refers to a more severe degradation degree of the CRM particles. In this study, this index was used to indirectly quantify the dissolution of CRM in AR mortar by following steps. First of all, the relationship between undissolved CRM content and rubber degradation index in AR binders at different aging conditions was obtained by nonlinear fitting. The same Soxhlet device was used for AR and RARP mortars to first remove the bitumen from undissolved CRM and aggregates, which were then removed from the Soxhlet extractor and soaked into DCM solvent for separation based on their different densities, because CRM (1.15 g/cm^3) has smaller density than DCM solvent (1.33 g/cm^3) and granites (2.8 g/cm^3), as shown in Figure 3(b). TGA tests were conducted on undissolved CRM extracted from mortar to obtain its rubber degradation index. With the established relationship between rubber degradation index and undissolved CRM content, the undissolved CRM content in mortar can be indirectly estimated.



(a) Soxhlet extraction device

(b) Separate the CRM from aggregates by density difference

Figure 3. Extraction of undissolved CRM from AR binder and AR/RARP mortar

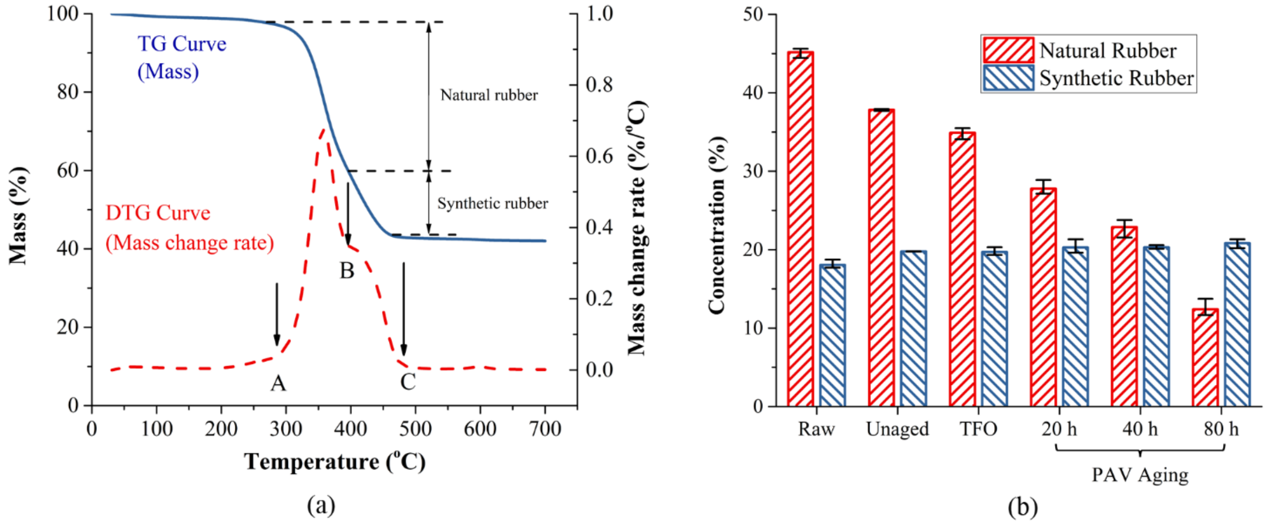


Figure 4. Establishment of rubber degradation index: (a) TGA and DTG curve of CRM; (b) concentration change of natural and synthetic rubber during aging.

2.3 Fabrication and testing of cylindrical mortar specimens

In this study, the rheological properties of mortar were measured by dynamic shear rheometer (DSR) with a special measuring system designed for solid specimens. To accommodate the test conditions, the mortar was fabricated into cylindrical specimens. A specially designed Teflon mould shown in Figure 5(a) was assembled with its accessories including two screws for fixing the mould and two caps of the cylindrical mortar. As shown in Figure 5(b), The assembled moulds were put into a self-developed rack where the two bars could hold the moulds still. The whole racks were put into an oven of 185 °C for pre-heating. During this time, the mortar was manually kneaded into cylindrical strips so they could be inserted into the mould through the upper cap. The mass of each sample was strictly controlled at 2.40 ± 0.01 g (excluding the mass of two caps) to improve the repeatability of results. The cylindrical specimen can be naturally formed due to the flowability of mortar at high temperatures. Figure 5(c) shows the end product of the cylindrical mortar sample and its dimensions with a unit of mm. Figure 5(d)&(e) show the loading and testing of cylindrical mortar sample, where the two caps were clamped by the DSR measuring system.

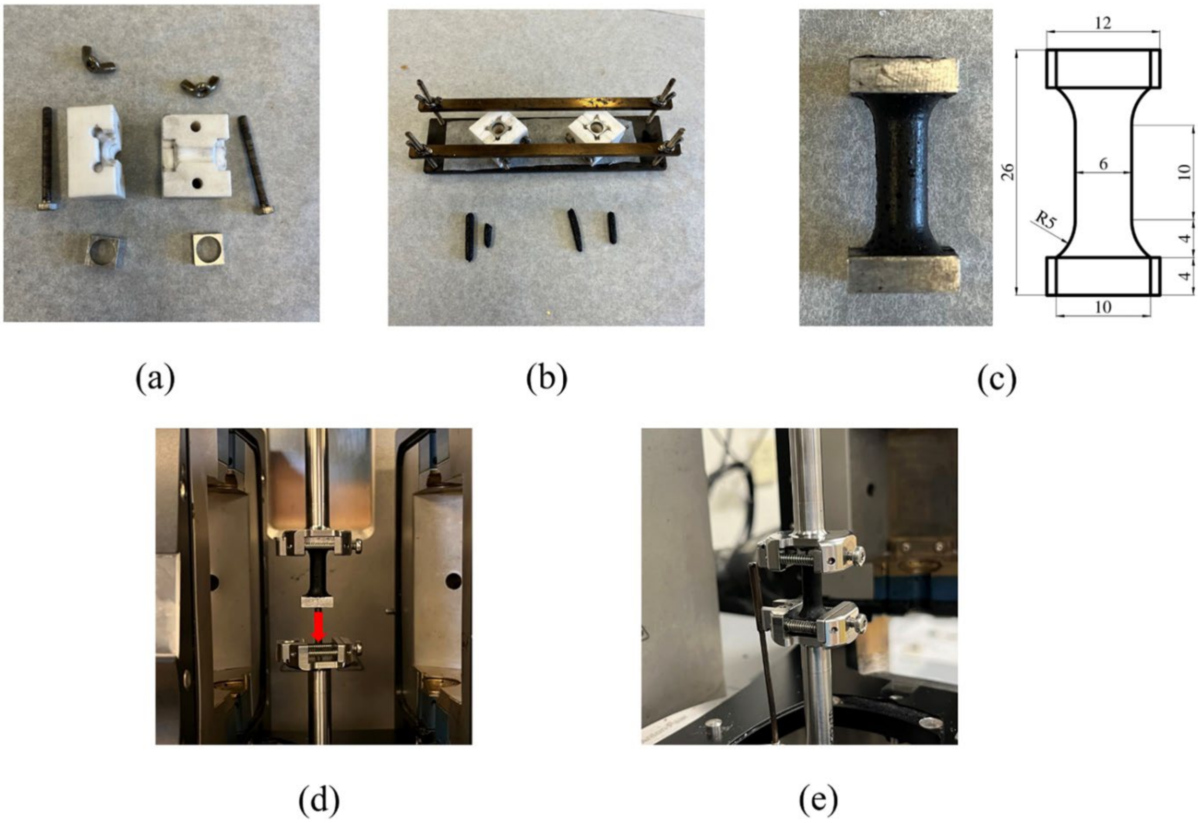


Figure 5. Fabrication and testing of cylindrical mortar specimen: (a) Teflon mould and accessories; (b) filling mortar strips into the mould at high temperatures; (c) schematic of the mortar cylindrical sample; (d) & (e) loading and testing of mortar sample.

2.4 Frequency sweep

The rheological properties of the AR and RARP mortar were evaluated by frequency sweep test in the strain-controlled mode using a DSR. The complex shear moduli and phase angles of different mortar samples in the frequency range of 100-0.01 Hz were measured in shear at 10, 20, 30, 40, and 50 ^{o}C, respectively. Three replicates were prepared for each type of sample. A strain amplitude sweep test was carried out at each testing temperature to determine the strain level in the frequency sweep test where the mortar maintains linear viscoelasticity. Master curves of the complex shear modulus and phase angle versus reduced frequency were constructed at a reference temperature of 25 ^{o}C. Based on the time-temperature superposition principle, the shift factors were determined by the Williams-Landel-Ferry (WLF) equation as shown in Equation 3.

$$\log \alpha_T(T) = \frac{-C_1(T-T_{ref})}{C_2+(T-T_{ref})} \tag{3}$$

where $\alpha_T(T)$ is the shift factors; C_1 and C_2 are coefficients of regression; T is the experimental temperature; and T_{ref} is the reference temperature. The viscoelastic responses of mortar were fitted by the Modified Huet-Sayegh (MHS) model developed by Woldekidan et al, whose physical representation is shown in Figure 6 (Woldekidan et al. 2012). A linear dashpot is connected in series with the original Huet-Sayegh (HS) model, which improves its ability to simulate the viscous behavior of mortar and the accuracy of data fitting in low frequency region (Zhang and Leng 2017). The mathematical expression of the MHS model is shown in Equation 4.

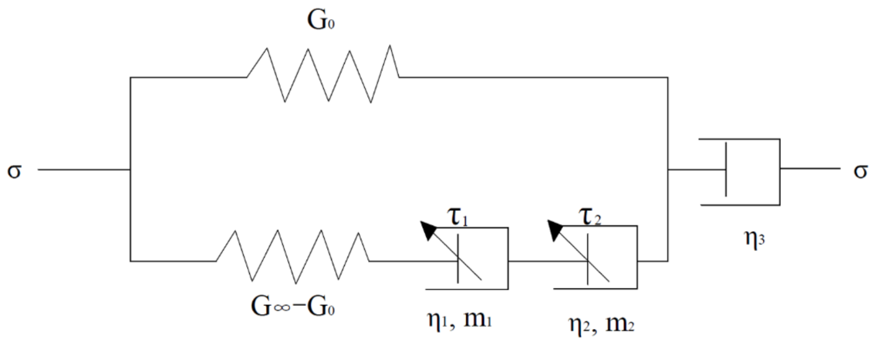


Figure 6. Physical representation of MHS model

$$J^*(\omega) = \frac{G'}{|G^*(\omega)|^2} - i\left[\frac{G''}{|G^*(\omega)|^2} + \frac{1}{\eta_3\omega}\right] = J'(\omega) - iJ''(\omega) \tag{4}$$

where $J^*(\omega)$ is the complex creep compliance of the MHS model; ω is the reduced frequency; $G^*(\omega)$ is the complex shear modulus of original HS model; G' and G'' are the storage shear modulus and loss shear modulus of original HS model, respectively; η_3 is the linear dashpot parameter; and $J'(\omega)$ and $J''(\omega)$ are the storage creep compliance and loss creep compliance of the MHS model, respectively. Furthermore, the expressions of $G^*(\omega)$, G', and G'' are listed as follows.

$$G^*(\omega) = G_0 + \frac{G_\infty - G_0}{1+\delta_1(i\omega\tau_1)^{-m_1}+\delta_2(i\omega\tau_2)^{-m_2}} \tag{5}$$

$$\delta_i = \frac{\tau_i(G_\infty - G_0)}{\eta_i} \tag{6}$$

$$G' = G_0 + A\frac{G_\infty - G_0}{A^2+B^2} \tag{7}$$

$$G'' = B\frac{G_\infty - G_0}{A^2+B^2} \quad (8)$$

$$A = 1 + \delta_1\frac{\cos(m_1\frac{\pi}{2})}{(\omega\tau)^{m_1}} + \delta_2\frac{\cos(m_2\frac{\pi}{2})}{(\omega\tau)^{m_2}} \quad (9)$$

$$B = \delta_1\frac{\sin(m_1\frac{\pi}{2})}{(\omega\tau)^{m_1}} + \delta_2\frac{\sin(m_2\frac{\pi}{2})}{(\omega\tau)^{m_2}} \quad (10)$$

where G_0 is the rubbery shear modulus; G_∞ is the instantaneous shear modulus; δ_1 and δ_2 are model parameters; τ, τ_1 and τ_2 are time constants, whose values are identical for response modelling of asphalt materials; and η_i is the parameter of parabolic dashpot. Except for evaluating the overall viscoelastic property of AR and RARP mortar by the master curves, two mechanical indexes, namely the rutting parameter and Glover-Rowe (G-R) parameter were adopted to evaluate the effectiveness of different rejuvenation schemes. The rutting factors ($G^*/sin\theta$) is the representative of the resistance to high-temperature deformation, where G^* and θ are the complex shear modulus and phase angle of each mortar derived from the frequency sweep test at the frequency of 10 Hz and temperature of 50 ºC. The G-R parameters ($G^* \cdot (cos\theta)^2/sin\theta$) were adopted to evaluate the ductility and brittleness at low temperature, which were calculated using the rheological response of mortar at the frequency of 0.005 rad/s and temperature of 15 ºC (Mensching et al. 2015).

2.5 Workability

Workability is one indispensable consideration during the recycling of RARP. AR itself has worse workability than the conventional asphalt, so it is undesirable that the potential rejuvenation will further deteriorate the workability of RARP. Thus, the workability was characterized by the Brookfield rotational viscometer in this study. The rotational viscosity tests were directly conducted on different AR-RARP mortars at 176 ºC, which is the target mixing temperature of AR mixture in Hong Kong.

3 Determination of Rejuvenation Methods for RARP

3.1 Evolution of AR mortar during aging

As shown in Figure 7(a), a negative dependence of the undissolved CRM content on the rubber degradation index can be found in AR binders at different aging conditions, which can be fitted by an exponential function, as shown in Equation 11.

$$C_{UR} = 31.241 \times e^{\frac{-RDI}{0.482}} + 4.891 \quad (11)$$

where C_{UR} is the undissolved CRM content and RDI is the rubber degradation index, which can be calculated by Equation 2. It can be seen that the dissolution rate of rubber polymers decreased with increasing aging time, because at the later period, the re-crosslinking of natural rubber might occur simultaneously with its degradation (Azura and Thomas 2006). Based on this fitting expression, the undissolved rubber contents in AR and RARP mortar were indirectly estimated by their rubber degradation index. Figure 7(b) shows that the undissolved CRM content in AR mortar decreased from 13.91% to 9.48 % and to 6.75%, which reveals that more than half of the CRM was dissolved after 80 hours of PAV aging. The rheological evolutions of AR mortar during aging are shown in in Figure 8, and the master curves of Pen60/70 mortar at unaged condition and 40 hours of PAV aged are also plotted for reference. Based on the time-temperature-superposition principle, the increase of reduced frequency can be understood as the decrease of temperature, so the moduli of different mortar samples increased with the reduced frequency. A plateau area can be found on phase angle curves of AR and Pen60/70, respectively. Such plateau areas refer to the where the aggregates and binder taking control.

As expected, aging increased the complex shear modulus and decreased the phase angle of different kinds of mortars due to the stiffening of binder. It can be seen that the rheological properties of Pen60/70 mortar changed more severely after 40 hours of PAV aging than AR after 80 hours of PAV aging, which highlights the superior aging resistance of AR binder.

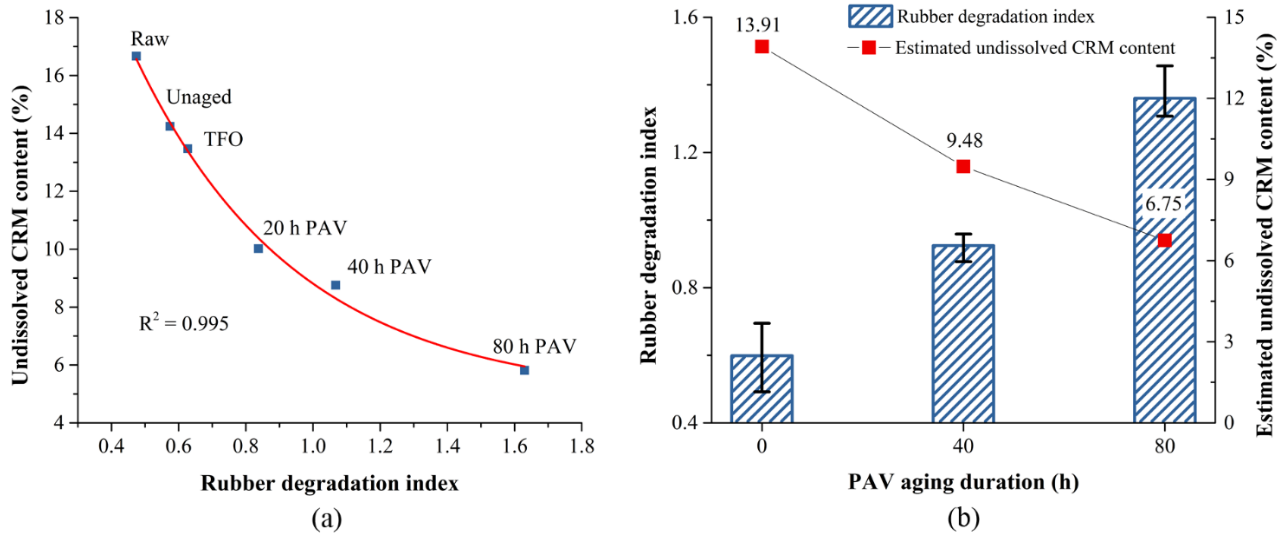


Figure 7. Indirect estimation of undissolved CRM content in AR mortar: (a) relationship between undissolved rubber content and rubber degradation index; (b) estimated undissolved CRM content in AR mortar after aging

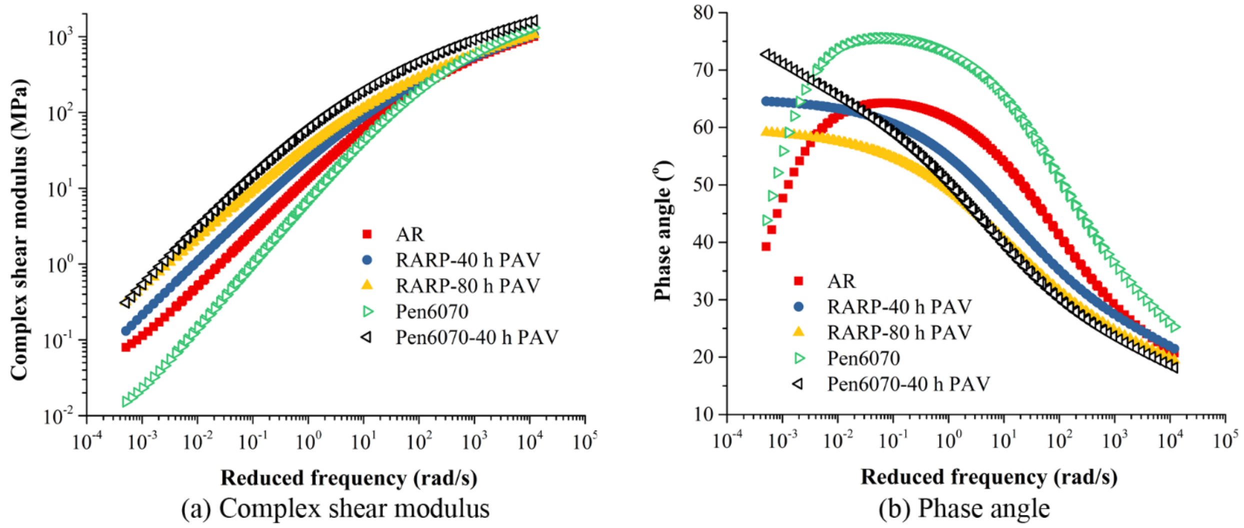


Figure 8. Rheological evolution of AR and Pen60/70 mortar during aging

3.2 Alternative Rejuvenation Methods

With known dissolution extents of CRM and rheological properties of the RARP mortar, the 80-hour aged RARP was selected for rejuvenation design because its rheological evolution was more severe than the 40-hour aged RARP, which can better highlight the influence of different rejuvenation schemes. It is worth noting that this study did not aim to determine the optimum dosage of rejuvenation but rather to investigate the rejuvenation mechanisms of RARP under different rejuvenation schemes. New AR mortar containing 40% of RARP mortar (RARP40%) with different rejuvenation schemes were prepared in this study to investigate the rejuvenation mechanisms of RARP. The proportion of RARP used in this study is relatively higher than common hot mix asphalt mixtures containing RAP considering that the RARP has less changed rheological property after aging. Aiming at the aging characteristics of AR binder, the purposes of the rejuvenation methods can be categorized into two groups: 1) softening the stiffened RARP binder by adding rejuvenator to recover the light fractions; and 2) supplementing the swelling rubber content by incorporating AR binder with extra CRM content. Three rejuvenation schemes can be developed by singly performing either rejuvenation method or performing them together as a compound rejuvenation. Table 3 summarizes the processing details of different rejuvenation schemes and their sample IDs. In the individual rejuvenation scheme, only rejuvenator or AR binder with extra CRM content was solely incorporated into the new AR mortar. In the compound rejuvenation scheme, both were incorporated. The tall oil used in this study is a commonly used bio-rejuvenator for aged asphalt binder containing fatty acids and surfactants, which is a byproduct derived during the kraft process (Prosperi and Bocci 2021). Three different dosages of tall oil (2/4/6) were engaged with the RARP by mass of the RARP binder. In addition, the AR binders with extra CRM were prepared using the same procedure as the original AR binder except for the different CRM dosages. Two CRM dosages were selected to restore the undissolved CRM content to different levels, namely 23% (AR23) and 26% (AR26) by mass of the raw bitumen. All RARP40% mortars were prepared by the same procedure and binder content. The RARP mortar was pre-heated and reacted with rejuvenator (if any) at 135 ºC, while the new aggregates and new AR binder were pre-heated at the mixing temperature of 175 ºC. Subsequently, the RARP mortar was first mixed with new aggregates for 30 s, then the new AR binder was added for another 30 s of mixing, followed by the addition of fillers for the final 60 s of mixing. The workability of each RARP40% mortar was measured before they were fabricated into cylindrical specimens for frequency sweep tests.

Table 3 Rejuvenation schemes of RARP mortar

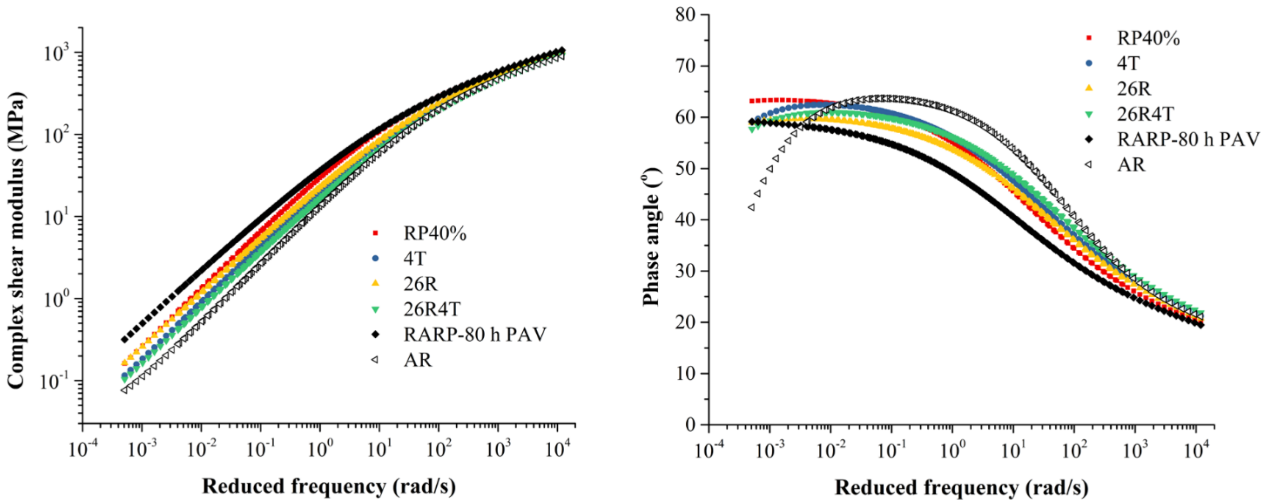
Rejuvenation scheme	**Purpose**	**Materials**	**Processing details**	**Sample ID**
Reference	Reference	Original AR binder	40% RARP mortar was mixed into	RP40%

group		(AR20)	new AR mortar using the original AR binder without special processing.	
Individual rejuvenation	Softening	Original AR binder (AR20), Rejuvenator (tall oil)	2/4/6wt% of tall oil was first reacted with 40% RARP mortar before mixing into new AR mortar using the original AR binder.	2/4/6T
	Recovering rubber	AR binder with extra CRM (AR23 & AR26)	40% RARP mortars were mixed into new AR mortar using AR binders with CRM contents of 23wt% and 26wt%	23/26R,
Compound Rejuvenation	Softening & recovering rubber	Rejuvenator (tall oil), AR binder with extra CRM	2/4/6wt% of tall oil was first reacted with 40% RARP mortar before mixing into new AR mortar using the AR23 & AR26.	23/26R2/4/6T

4 Results and Discussion

4.1 Comparison of rejuvenation effectiveness

The rejuvenation effectiveness is defined as the extent to which the rheological properties of RARP mortar were restored or improved. Figure 9 compares the master curves of AR, RARP, and RARP40%, where one case was selected from each rejuvenation scheme. As expected, the master curve of complex shear modulus of RP40% falls in between that of RARP and AR. In the individual rejuvenation scheme, the single addition of tall oil further softened the RARP40%, as reflected by the decreasing complex shear modulus and increasing phase angle. Meanwhile, it is surprising to observe that the modulus master curve of 26R is slightly lower than that of RP40%, which implies that the incorporation of extra CRM also softened the mortar. It can be attributed to the fact that the bitumen became stiffer than CRM after long-term aging. By comparison, the compound rejuvenation case 26R4T mortar shows the rheological properties closest to the initial AR mortar.



(a) Complex shear modulus (b) Phase angle

Figure 9. Master curves of RARP40% using different rejuvenation schemes

To more quantitatively compare the rejuvenation effectiveness, the rutting factors and G-R parameters were calculated to quantify the change of rheological properties when different rejuvenation methods were used for RARP40% mortar, as summarized in Table 4. As aforementioned, the rutting factor is the indicator of resistance to high-temperature deformation, while the G-R parameter is the representative of the ductility and cracking resistance at low temperatures. Both indexes increased with aging, indicating the stiffening of AR binder. In the individual rejuvenation group, the single addition of rejuvenator could significantly soften the RARP40%. The softening effect of rejuvenator was enhanced with the increasing dosage. Especially when the dosage of tall oil was 6%, both indexes dramatically decreased and rutting factor was even lower than that of the original AR. On the other hand, singly incorporating AR binder with extra CRM into RARP40% mortar only slightly softened the mortar, while 26R showed a stronger softening effect than 23R. In the compound rejuvenation group, the softening effect of rejuvenator was maintained when simultaneously incorporated into the RARP40% mortar. Similarly, the softening effect improved with the increasing dosage of tall oil and CRM. Overall, the results revealed that the rejuvenator was more effective in terms of the ability to soften the aged AR binder, but the contributions of rejuvenator and extra CRM to the aging resistance of RARP40% require further discussion.

Table 4 Mechanical indexes of different mortar samples

Reference group	**AR**	**RARP-40 h PAV**	**RARP-80 h PAV**	**RP40%**		
Rutting factor (MPa)	4.57	8.56	12.94	10.96		
G-R parameter (MPa)	0.53	1.43	3.43	1.60		
Individual Rejuvenation group	**2T**	**4T**	**6T**	**23R**	**26R**	
Rutting factor (MPa)	9.46	7.04	4.51	9.95	8.28	
G-R parameter (MPa)	1.7	1.04	0.61	1.53	1.57	
Compound Rejuvenation group	**23R2T**	**23R4T**	**23R6T**	**26R2T**	**26R4T**	**26R6T**
Rutting factor (MPa)	8.11	6.76	5.16	7.63	5.94	4.93
G-R parameter (MPa)	1.53	0.94	0.84	1.10	0.94	0.71

4.2 Comparison of aging resistance based on a secondary aging

When RARP is recycled into new AR mixture, it is preferable to avoid compromising the superior aging resistance of AR binder due to the existence of degraded RARP binder. To assess the aging resistance of RARP40% mortar rejuvenated by different methods, all RARP40% mortars were processed by a secondary PAV aging of 80 hours. The rutting factors and G-R parameters of different RARP40% mortars after the secondary PAV aging were calculated, and the aging extent was calculated by Equations 11 & 12.

$$Aging\ extent_{RT} = Rutting\ factor_{asa} - Rutting\ factor_{bsa} \quad (12)$$

$$Aging\ extent_{GR} = G - R\ parameter_{asa} - G - R\ parameter_{bsa} \quad (13)$$

where the aging extent is the discrepancy between the mechanical indexes after and before the secondary aging; the subscripts asa refers to after the secondary aging, and bsa refers to before the secondary aging. Figure 10 compares the aging resistances of different RARP40% mortars rejuvenated by different methods. The complete results of all mortars were not shown for the clarity of the figure, but the presented data are sufficient to reflect the overall pattern. Both indexes significantly increased after the secondary aging, symbolizing the stiffening of bitumen and degradation of CRM. The previous section has reported that the individual use of rejuvenator effectively softened the aged binder while the individual use of AR binder with extra rubber only provided very limited softening effect. However, it is interesting to find that the rutting factors and G-R parameters of 4T and 6T surpassed those of the 23R and 26R after the secondary aging. As shown in Figure 10(b), the G-R parameters of

4T and 6T had no statistical difference from those of the RP40% which did not adopt any particular rejuvenation approach. This phenomenon reveals the negligible contribution of rejuvenator to the aging resistance of RARP40% mortar, as reflected in another study (Ren et al. 2020). It might be attributed to the higher amount of oxygen in the bio-rejuvenators, which makes the bitumen more susceptible to aging (Cai et al., 2019). By comparison, all RARP40% mortars incorporated with extra CRM underwent fewer changes in their mechanical indexes after the secondary aging. 23R, 26R, and all of the compound rejuvenation mortars show smaller aging extents than the mortar samples without the incorporation of AR binder with extra CRM content. Therefore, the results demonstrate that the recovery of rubber content can help restore the aging resistance of AR mixture containing RARP.

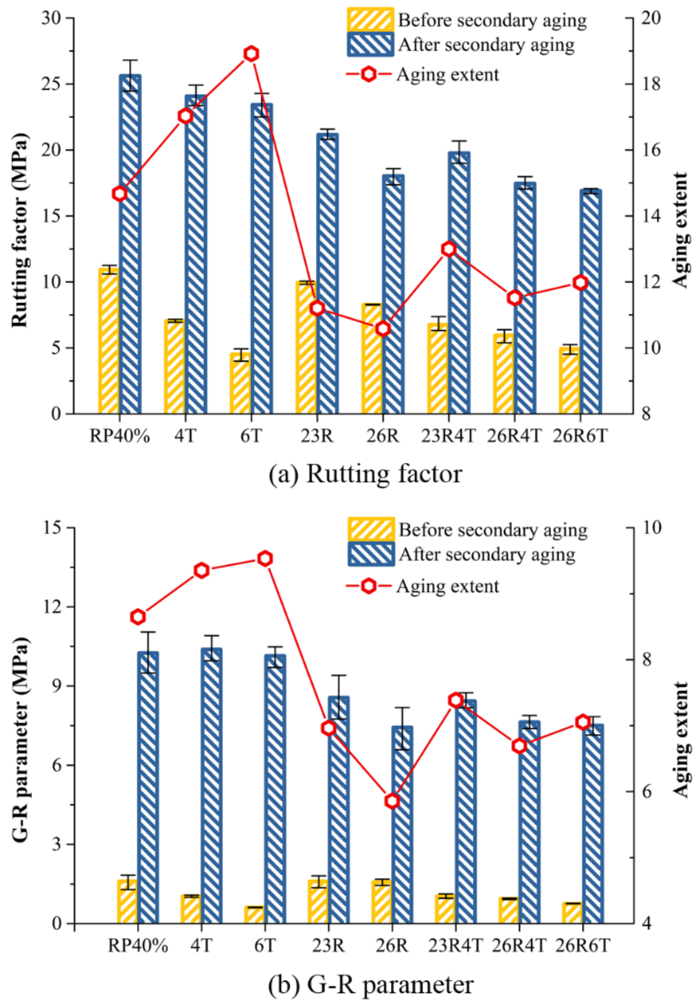


Figure 10. Aging resistance of RARP40% using different rejuvenation schemes

4.3 Workability

Viscosity tests were conducted to observe the influence of different rejuvenation schemes on the workability of RARP-included mixtures. Figure 11(a) shows that the viscosity of AR mortar first decreased after 40 hours of PAV aging. This phenomenon might be contrary to the previous perception because most asphalt binders gain increased viscosity after aging. However, it can be explained by the structural change of AR binder caused by the rubber degradation during aging. AR binder overgoes the decreasing volume of swelling rubber and increasing proportion of liquid phase (Li et al. 2022a). Such structural changes have certain similarities to the transition from AR binder to the terminal blend (TB) binder. TB is another type of rubberized asphalt, which emphasizes the excessive degradation of CRM during the preparation process to decrease the viscosity (Huang et al. 2017). The natural rubber and absorbed bitumen light fractions were released to reduce the viscosity of the AR binder. More importantly, the increasing liquid phase content also indirectly increased the binder content in the mortar, which might be a critical reason for the decreased viscosity of AR mortar after 40 hours of PAV aging. The viscosity increased again after 80 hours because the stiffening effect of bitumen aging may have overwhelmed the degradation effect of CRM at a specific time. Figure 11(b) compares the viscosity of RARP40% mortar using different rejuvenation methods. The workability of RP40% is similar to that of the original AR mortar. The two different individual rejuvenation schemes show completely distinct results. The addition of rejuvenator improved the workability of RARP40% mortar because the tall oil could work as surfactant to reduce the viscosity of the binder. By contrast, the viscosity of RARP40% mortar rapidly increased with the CRM content in the new AR binder, where 26R almost doubled the viscosity of the RARP mortar after 80 hours of PAV aging. Fortunately, it is promising to find that the combined use of rejuvenator can alleviate the workability concern when AR binder with extra CRM was included.

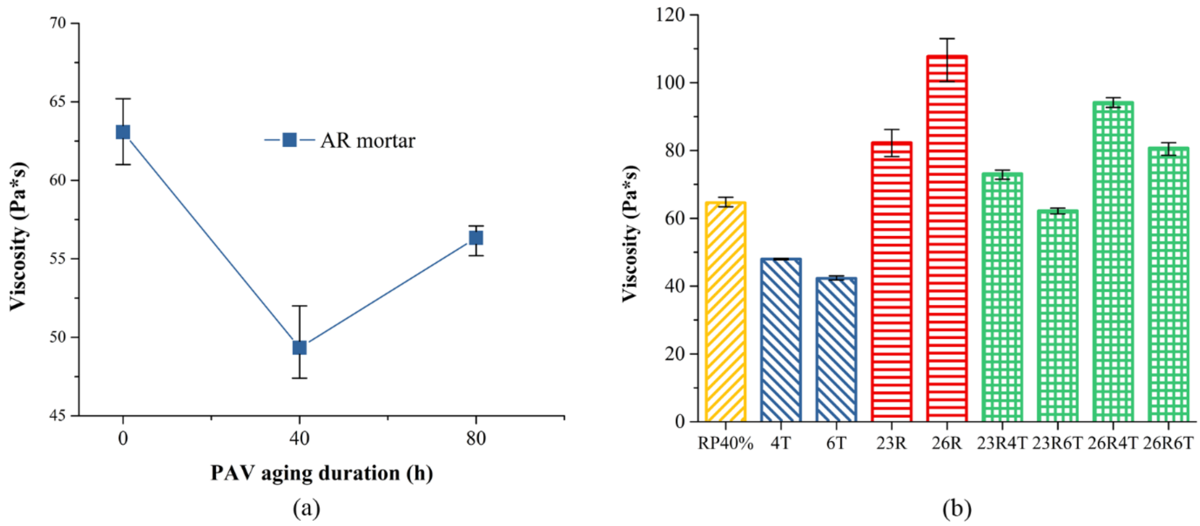


Figure 11. Viscosity test results: (a) viscosity change of AR mortar during aging; (b) viscosity of RARP40% using different rejuvenation schemes

4.4 Discussion and Generalization

After a series of testing and analyses on the RARP40% mortar in terms of the rejuvenation effectiveness, aging resistance, and workability, an overall picture can be obtained regarding the rejuvenation mechanisms of RARP. The findings can probably be generalized for more different material compositions that may happen in the recycling of RARP. If the RARP was treated as common RAP, the conventional rejuvenation method of adding rejuvenator can effectively soften the aged AR binder, but it was merely conducive to the aging resistance. Although the addition of rejuvenator could recover the light fractions in the aged AR binder, it was unable to slow down the aging evolution of AR binder and might be vulnerable to evaporation during aging (Ma et al. 2020). Meanwhile, the aging resistance is expected to deteriorate with the increasing proportion of RARP due to the reduced content of undissolved rubber. By comparison, the recovery of undissolved CRM content has been found to improve the aging resistance of the RARP-included mixture. The supplemented swelling rubber could replace the old degraded rubber to exert its role in maintaining the rheological properties of the mixed AR binder. The extra dosage of CRM can also further promote the recycling of waste tires. However, the single incorporation of AR binder with extra CRM content was considered impractical when a relatively high RARP proportion was used, because it could not effectively soften the binder and caused serious workability problem. Technically, the workability concern might be alleviated if a smaller RARP proportion was adopted where the extra CRM content in the AR binder to be mixed into the new AR mixture could be lower. The compound rejuvenation scheme appeared to be the optimum solution that balanced the rejuvenation effectiveness, aging resistance, and workability. The addition of rejuvenator could reduce the viscosity of RARP40% mortar containing AR binder with extra CRM to an acceptable level. For material selections, it is supposed that many other types of rejuvenators with a liquid form that can reduce the viscosity of asphalt binder can serve as alternatives for the tall oil used in this study, such as various vegetable oil, soft bitumen, and aromatic oil. Even some warm mix additives like Evotherm-DAT or Evotherm-3G containing surfactants might be used as a rejuvenator in the recycling of RARP (Li et al. 2021). To sum up, the rejuvenation design of RARP can be flexible in terms of the RARP proportion and material composition. The determination should be made based on a comprehensive consideration of target mixing and paving temperature as well as the rheological evolution and rubber dissolution extent of RARP binder.

5 Findings and Recommendations

This study investigated the rejuvenation mechanisms of RARP through a series of chemical and rheological analyses conducted at the mortar level. The following points summarize the main findings of this study.

- The relationship between undissolved CRM content and rubber degradation index was established through the Soxhlet extraction and TGA tests. This relationship can be applied to indirectly estimate the undissolved CRM content in RARP mortar, which helped to determine the required amount for supplementing the CRM content in RARP40% mortar.
- The individual addition of rejuvenator effectively softened the RARP binder and brought positive effect to the workability, but it did not slow down the rheological evolution of RARP40% mortar during the secondary aging.
- The incorporation of AR binder with extra CRM can recover the swelling rubber content in the RARP40% mortar, notably improving the aging resistance. However, the softening effect was limited, and the workability was significantly deteriorated.

- The compound rejuvenation scheme combined the advantages of both rejuvenation methods, balancing the rejuvenation effectiveness, aging resistance, and workability.

Overall, the findings of this study are expected to lead to a more customized rejuvenation strategy for RARP toward a more sustainable application of the AR pavement. Future work will be extended to the mixture level to verify the actual performance of the RARP-included mixtures using different rejuvenation methods and RARP proportions.

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Disclosure statement

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