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DOI

[10.1201/9781003156222-02](https://doi.org/10.1201/9781003156222-02)

Publication date

2021

Document Version

Accepted author manuscript

Published in

Functional pavement

Citation (APA)

Ren, S., Liu, X., Lin, P., & Erkens, S. (2021). Use of waste oil/styrene-butadiene-rubber blends as rejuvenators for aged bitumen. In X. Chen, J. Yang, M. Oeser, & H. Wang (Eds.), *Functional pavement: Proceedings of the 6th Chinese-European Workshop on Functional Pavement Design, CEW 2020* (pp. 7-12). CRC Press / Balkema - Taylor & Francis Group. <https://doi.org/10.1201/9781003156222-02>

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Use of waste oil/styrene-butadiene-rubber blends as rejuvenators for aged bitumen

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ABSTRACT: This study elaborates the influence of blends formulated by waste oil (WO) and styrene-butadiene rubber (SBR) on thermo-mechanical and chemical compositional characteristics of rejuvenated bitumen. The results show that both WEO (waste engine oil) and WCO (waste cooking oil) can compensate viscous component for reclaimed bitumen, and the effect of WCO is more significant. However, WO-rejuvenated bitumen has considerable drawbacks in terms of temperature susceptibility and flow resistance, but dramatic improvement after being added with SBR has been noticed. Furthermore, the rejuvenation mechanism of WO/SBR blends is revealed by using FTIR tests, which shows that the physical blending mainly determines the rejuvenation mechanism when the WO/SBR blends are used in bitumen. The combination of WO and SBR is beneficial for improving the temperature susceptibility and rutting resistance of rejuvenated bitumen.

1. INTRODUCTION

At the end-of-life of asphalt pavements, the aged bitumen in old asphalt, which is named reclaimed asphalt pavement (RAP), is a degraded material of high modulus [4] but with the potential to be re-used with the proper treatment [1-3]. Recently, although many highway agencies allow the incorporation of RAP in new pavements [4], the pavement materials consisting of RAP could show high thermal and fatigue cracking vulnerability [5].

Nowadays, the most commonly used rejuvenation method for aged RAP binders is the addition of softening additives of light fractions which can balance the chemical composition and thus the physio-mechanical properties of aged binders [3]. Among those additives, waste oil (WO) is commonly used for the aged bitumen, including WCO and WEO [6]. Although many studies have reported that adding WO could compromise the physio-mechanical properties for aged bitumen, there are inevitable issues of using WO into the rejuvenation of aged bitumen. The main challenge of WO-rejuvenated bitumen is on the determination of rejuvenator dosage, which can not only restore the physio-mechanical properties but strengthen the anti-rutting ability for aged bitumen as well [7, 8]. Hence, the motivation and objective of this study is to explore the potential of using waste oil and SBR to restore and improve the rheological and chemical properties of aged bitumen.

2. MATERIALS AND METHODS

2.1. Materials and sample preparation

Base bitumen ZH70[#] is used to prepare the aged bitumen with air-blowing process at 260 °C for 48 hours. The conventional performance for neat bitumen and waste oil are displayed in Table 1 and 2, respectively. Meanwhile, the 60 °C viscosity and 25 °C penetration value of aged bitumen is 32359 Pa·s and 10-dmm, while the softening point and 5 °C ductility is 84.1 °C and 0.1-cm, respectively.

Table 1 Conventional properties and chemical components of aged asphalt

Items	Aged bitumen	Test methods
Penetration (25 °C, 0.1mm)	10	ASTM D5
Softening point (°C)	84.1	ASTM D36
Ductility (10 °C, cm)	0.2	ASTM D113
Ductility (15 °C, cm)	1.3	ASTM D113
Viscosity (60 °C, Pa·s)	32359	AASHTO T316
Saturate (S, wt%)	10.31	ASTM D4124
Aromatic (A, wt%)	8.19	
Resin (R, wt%)	42.83	
Asphaltene (At, wt%)	38.67	
Colloidal index CI *	0.96	

* Colloidal index CI = (Asphaltene + Saturate) / (Aromatic + Resin)

The aged bitumen is mixed with WO for about 15min with the rotation speed of 1000 r/min and mixing temperature of 150 °C. The dosage of waste oil is 5, 10 and 15wt%, which is referred as WCO5 (WEO5), WCO10 (WEO10) and WCO15 (WEO15), respectively.

Further, both WCO10 and WEO10 rejuvenated bitumen are further modified by SBR with various content of 1, 2 and 3wt% to obtain WO/SBR rejuvenated bitumen, which are called as WCOSBR1 (WEOSBR1), WCOSBR2 (WEOSBR2) and WCOSBR3 (WEOSBR3).

Table 2 Basic properties of WCO and WEO

Items	WCO	WEO
Specific gravity ($\text{g}\cdot\text{cm}^{-3}$)	0.926	0.884
Viscosity (60°C, $\text{mPa}\cdot\text{s}$)	28.75	55.00
Flash point (°C)	171	173

2.2. Rheological and chemical properties characterization

Dynamic shear rheometer (DSR, TA-HR1) is performed to determine these viscoelastic behaviors for bitumen with 25-mm diameter and 1-mm gap. Both complex modulus (G^*) and phase angle (δ) of aged and rejuvenated bitumen are obtained through frequency sweep tests with the frequency increasing from 10^{-1} rad/s to 10^2 rad/s at the various temperature of 45, 50, 55, 60 and 65 °C, respectively.

FTIR is used to explore the rejuvenation and modification mechanism of rejuvenated bitumen with the WO/SBR blends. The molecular interaction of bitumen sample in IR ranges of $400\text{--}4000\text{ cm}^{-1}$ is observed.

3. RESULTS AND DISCUSSION

3.1. Temperature susceptibility

In this study, the complex number index (CNI) parameter is selected to evaluate the temperature susceptibility of aged and rejuvenated bitumen as follows:

$$\text{CNI} = G'_{\text{TS}} + G''_{\text{TS}} \cdot i \quad (1)$$

The relationship between temperature (T) with storage modulus (G') and loss modulus (G'') of aged, WCO- and WCO/SBR-rejuvenated bitumen is displayed in Fig.1. Obviously, the increasing of temperature remarkably reduces both storage and loss modulus of bitumen. There exists fairly good linear-relation between temperature with $\lg G'$ and $\lg G''$ values. The G'_{TS} and G''_{TS} values of bitumen can be obtained from the absolute slope of regression equations of $\lg G'-T$ and $\lg G''-T$, severally.

Fig.1. Storage modulus (G') and loss modulus (G'') of aged bitumen, WCO and WCO/SBR rejuvenated bitumen (a), and WEO and WEO/SBR rejuvenated bitumen (b)

From Table 3, WCO and WEO increases the G'_{TS} as well as G''_{TS} values of aged and rejuvenated bitumen, indicating that adding WCO and WEO can rise the temperature susceptibility for aged and rejuvenated binders. This phenomenon is attributed to the temperature-dependency of light components in WO. Meanwhile, the temperature susceptibility of WO-rejuvenated bitumen improves after being added with SBR copolymer. Moreover, WCO/SBR-rejuvenated bitumen has superior thermal susceptibility to that of WEO/SBR-rejuvenated bitumen.

Table 3 CNI values of aged and rejuvenated bitumen

Bitumen samples	$\text{CNI} = G'_{\text{TS}} + G''_{\text{TS}} \cdot i$	Bitumen samples	$\text{CNI} = G'_{\text{TS}} + G''_{\text{TS}} \cdot i$
RA	$0.06427 + 0.04414i$	RA	$0.06427 + 0.04414i$
WCO5	$0.06630 + 0.04642i$	WEO5	$0.06775 + 0.05316i$
WCO10	$0.07564 + 0.05316i$	WEO10	$0.07259 + 0.05339i$
WCO15	$0.07752 + 0.05197i$	WEO15	$0.07520 + 0.05201i$
WCOSBR1	$0.06970 + 0.04688i$	WEOSBR1	$0.06628 + 0.05084i$
WCOSBR2	$0.06140 + 0.04488i$	WEOSBR2	$0.06417 + 0.04951i$
WCOSBR3	$0.06040 + 0.04359i$	WEOSBR3	$0.06298 + 0.04891i$

3.2. Viscoelastic properties

Fig.2 presents the effects of WO type and concentration, SBR copolymer dosage as well as frequency on the complex modulus of aged and rejuvenated bitumen. This result indicates that the complex modulus value of bitumen increases linearly as the testing frequency increases. Over the whole frequency range, the aged bitumen has the highest modulus values because of the loss of light compounds. Meanwhile, adding WCO and WEO could remarkably reduce complex modulus of aged bitumen, which continues to decrease as the WO dosage increases. Obviously, adding WCO and WEO has adverse influence on anti-deformation ability for aged and rejuvenated bitumen. The modulus value of rejuvenated bitumen is improved by incorporating the SBR. With the increase of SBR dosage, the complex modulus of rejuvenated bitumen increases gradually, which is lower than that of WCO5 and WEO5 rejuvenated bitumen.

Fig.2. Complex modulus (a) and phase angle (b) of aged and rejuvenated bitumen

Compared to aged bitumen, WCO and WEO rejuvenated bitumen has higher phase angle value, showing that WO can restore the viscous component and modulus of aged bitumen dramatically. However, the increase of WO dosage weakens the elastic properties of rejuvenated bitumen, which is importance of improving both high- and low-temperature performance for bitumen. Moreover, adding SBR can remarkably decline phase angle value and strengthen elastic property of WO-rejuvenated bitumen. Furthermore, the phase angle for WCO-rejuvenated binder shows larger than WEO-rejuvenated bitumen. This result indicates that WCO can recover the viscous characteristic for aged bitumen more easily than WEO. It also can be seen that the impact of SBR on improving elastic properties for WEO-rejuvenated binder is more obvious.

3.3. Activation energy

The relationship between temperature and shift factor α of aged and rejuvenated bitumen is analyzed by the Arrhenius formula to assess the influence of WO and SBR on the viscoelastic properties of aged and rejuvenated bitumen. The activation energy of bitumen can be obtained as

$$\ln \alpha = \frac{Ea}{R} \left(\frac{1}{T} - \frac{1}{T_0} \right) \quad (2)$$

where T represents the testing temperature; T_0 shows the reference temperature; α is the shift factor; Ea is the activation energy and $R=8.314\text{J/mol}\cdot\text{K}$.

Table 4 presents the Ea values of aged and rejuvenated bitumen, which are obtained by calculating from Arrhenius formula. The aged bitumen has the highest Ea value and the most excellent performance stability. Meanwhile, the addition of WCO and WEO decreases the Ea value of aged bitumen dramatically, which means WCO and WEO both make negative influence on temperature sensibility for rejuvenated bitumen. Moreover, Ea value of rejuvenated bitumen can be further enhanced by adding SBR, which can strengthen the temperature sensitivity for WO-rejuvenated binder obviously. Further, WEO rejuvenated bitumen has better temperature susceptibility performance than WCO rejuvenated bitumen.

Table 4 The Ea values of aged and rejuvenated bitumen

Codes	RA	WCO5	WCO10	WCO15	WCO5BR1	WCO5BR2	WCO5BR3
Ea/KJ·mol ⁻¹	67.09	62.22	60.27	55.92	57.28	58.96	58.58
Codes	RA	WEO5	WEO10	WEO15	WEO5BR1	WEO5BR2	WEO5BR3
Ea/KJ·mol ⁻¹	67.09	66.50	65.96	63.37	60.56	61.56	61.54

3.4. Rejuvenation mechanism of WO/SBR blends in bitumen

Aim to further characterize the rejuvenation mechanisms of WO and SBR on the

microstructure of recycled bitumen, the main IR vibrational peaks of aged and rejuvenated bitumen are shown in Fig.3. It is well known that FTIR results can determine whether or not chemical reaction happens during the mixing of aged asphalt, WO and SBR. Compared with reclaimed asphalt, waste cooking oil rejuvenated asphalt has additional characteristic peaks at 1160, 1257 and 1745 cm^{-1} , which is attributed to the addition of waste oil. Besides, WO/SBR rejuvenated bitumen has the additional characteristic peak at 966 cm^{-1} , which are assigned as $-\text{CH}=\text{CH}-$ out-of-plane deformation. Furthermore, new absorption peaks of WEO/SBR rejuvenated bitumen appear at 966 cm^{-1} and 1747 cm^{-1} , which could characterize the existence of WEO and SBR. However, compared with the absorption peaks of raw materials, there is no distinct formation or disappearance of characteristic peaks for WO and SBR rejuvenated bitumen. In a word, the rejuvenation mechanisms of WO and WO/SBR rejuvenated bitumen mainly focus on the physical blending.

Fig.3. FTIR plots of aged and rejuvenated bitumen

4. CONCLUSIONS

In this study, waste oil (WCO and WEO) and SBR were used as rejuvenators to restore and enhance the viscoelastic of rejuvenated bitumen. Blending WCO and WEO significantly decreases the complex modulus of aged bitumen, which can be enhanced by the addition of SBR. Meanwhile, WCO and WEO can restore the viscous components and modulus of aged bitumen dramatically but weakens the elastic behavior of rejuvenated bitumen, which can be remarkably enhanced by SBR. Based on master-curves, WCO and WEO both have adverse influence on the rutting resistance, activation energy as well as the temperature susceptibility of rejuvenated bitumen, which can be improved by incorporating the SBR copolymer in WO. Further, FTIR tests have shown that the rejuvenation mechanism occurs mostly due to the physical blending of WO/SBR blends in aged binders.

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