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Optimising the dosage of bio-rejuvenators in asphalt recycling: A rejuvenation index based approach

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ABSTRACT

While significant research efforts have been devoted to the rejuvenation of aged bitumen using bio-rejuvenators, a universally recognised approach for dosage optimisation is still yet to be established. Aiming to develop standardised methods to determine the optimal dosages of six types of bio-rejuvenators, this study validated the use of a rheology-based rejuvenation index as a potential universal method. Rheological properties of rejuvenated binders, determined through frequency sweep tests, multiple stress creep and recovery tests, linear amplitude sweep tests, and bending beam rheometer (BBR) tests were carried out with the properties of virgin binder as reference targets. The results indicated that the applied rheology-based rejuvenation index is a feasible approach. This index can ensure improved low- and intermediate-temperature (fatigue) performance of aged bitumen while maintaining high-temperature performance within acceptable limits. Further characterisation of the binders by Fourier Transform Infrared Spectroscopy (FTIR) analysis suggested that the bio-rejuvenation processes will commonly only involve physical softening rather than a chemical reversal of ageing. Therefore, the use of chemistry-based indices is likely not feasible.

1. Introduction

Roads are one of the most important infrastructure assets for the development of a country's economy and society [1]. The design life of roads is limited by the deterioration in performance of bitumen or asphalt binders, especially related to ageing [2,3]. This is mainly the result of the exposure to the environment and the combined effects of temperature, oxygen, UV radiation etc [4], which results in significant physical and chemical changes in the properties of bitumen and inevitably leads to deterioration of the performance and usability of asphalt pavements, such as cracks and potholes [5].

Hardening due to ageing of bitumen can be broken down into two different categories, oxidisation, and steric hardening [6,7]. Oxidisation is the primary attribute of ageing, involving chemical reactions between the atmospheric oxygen and bitumen. This reaction results in two major changes: a large difference in the ratio between molecule sizes and ratio of polar and non-polar compounds which reduces solubility and ductility, consequently diminishing bitumen performance [8]. Steric hardening is the secondary contributing factor to bitumen ageing, which is a slow developing, heat-reversible hardening that impacts the molecular structure of the binder [9]. In terms of the chemical composition, ageing of bitumen results in a reduction in the content of aromatics and the increase of resins and asphaltenes, inducing instability of its colloidal system [10]. The decrease in content of aromatics contributes to a reduction in ductility and other elastic properties, eventually leading to cracking and other failures [11].

Fortunately, there are methods to renew the properties of aged bitumen and restore its ductility related properties using additives known as "rejuvenators". When considering different types of rejuvenators, bio-rejuvenators, which are derived from bio-sources, are becoming more and more popular nowadays as a result of its increased sustainability as a waste derived material. [12]. Bio-rejuvenators are products from nonpetroleum-based renewable resources, such as wood, grain, livestock manure etc., and its potential use shown to have economic and environmental benefits [13]. The rationale for using such rejuvenators in bitumen rejuvenation is that their lighter fractions such

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Table 1
Rejuvenators used in this study.

Code	Rej-1	Rej-2	Rej-3	TO	RBO	WCO
Description	Commercial bio-based rejuvenator	Commercial bio-based rejuvenator	Commercial bio-based rejuvenator	Distilled tall oil	Recycled biofuel oil	Recycled very viscos cooking oil residue
Density (kg/l)	0.915	0.890	0.874	0.908	0.865	0.878
Viscosity at 25 °C (cP)	60	60	46	42	39	876
Appearance						

as high saturates and aromatics content can replenish the loss of components caused by ageing, and thereby, recover physical properties [14]. However, existing research has mainly focused on the feasibility of using different types of rejuvenators and the dosages used have been a subject of ambiguity. In many studies, the dosages have been selected randomly or employed the use of just one modification dosage [15–18]. For the studies using varying dosages, the dosages were often based on manufacturers recommendations [19]. Minimal studies have investigated the use of universally applicable dosage optimisation approaches, especially in relation to fully capturing the target performance.

Different indices based on the changes in penetration, softening point, ductility, viscosity [20], glass transition temperature [21], performance grade (PG) [22], rheological properties [23,24], SARA fractions [25], carbonyl index, sulfoxide index [26], morphological properties [27] have been used to evaluate the ageing degrees of bitumen. Rejuvenation is the method used to recover the properties of binders after ageing, therefore, some forms of these ageing indices are often used as guidelines for rejuvenation metrics and effectiveness.

Dosage optimisation is of crucial significance for the rejuvenation of aged bitumen. If the dosage of rejuvenator used is not inappropriate, the performance of rejuvenated asphalt pavements might be negatively affected. For example, if the dosage were too low, the performance of bitumen cannot be adequately recovered while if the dosages were too high, some additional issues such as rutting might be induced. The commonly used metrics of rejuvenator selection and dosage optimisation are not very comprehensive or sophisticated [28]. Penetration and viscosity are the most used indices for dosage optimisation [15,29]. However, both indices are single-temperature and single-reading based, which fails to evaluate the complete properties of the rejuvenated bitumen. Moreover, some studies have reported that the penetration and viscosity of bitumen are not necessarily related to binders field performance [30]. In addition to these two conventional indices, there is no other well recognised index to accurately optimise the dosage of rejuvenators. When considering more sophisticated methods, some rheology-based approaches have been proposed, such as the metrics based on performance grade [19,22,24,31]. Out of these, one of the most comprehensive rheology-based indices calculates the changes in complex modulus of bitumen for a whole temperature (frequency) range and was theorised to be capable of capturing the performance changes of bitumen during the ageing process. Lin et al. used this index as one of the indicators to illustrate the rejuvenation efficiency [28]. Combining the results of LAS, MSCR, G-R parameter and relaxation, an optimum range of dosage for rejuvenators was determined. However, the method was complex and required a lot of laboratory efforts to determine the range, and the dosage ranges for each rejuvenator used were broad, leading to limited applicability for practice. Rathore et al. also used this index for evaluating the softening effect of rejuvenation. It was found that rejuvenators can effectively soften aged binders, but correlations between this index and rejuvenator dosage was not investigated [32]. Additionally, both the aforementioned studies did not characterise the low-temperature performance by the means of experimental approaches, which might result in uncertainty since the low-temperature performance is one of the most important properties affected by ageing [33]. Moreover, the application of this index for different types of bio-rejuvenators has yet to be well understood. Overall, though some studies have attempted to use this index as one of the indicators to evaluate rejuvenation efficiency, there has been no investigation on the direct correlation between this index and dosage. Dosage optimisation of bio-rejuvenators is of crucial importance in current times as bitumen recycling is becoming more common, and inappropriate dosages could result in either insufficient rejuvenation or reduced high-temperature performance due to overdosing [34]. Methods using comprehensive rheology-based approaches are promising to develop a standard for a universally applicable rejuvenation index. Aiming to address the existing gap, this study will employ a rheology-based rejuvenation index and validated its feasibility. Additionally, the chemical properties of both rejuvenators and bitumen were also characterised to further comprehend the intrinsic characteristics of rejuvenation.

2. Scope

This study employs a rheology-based index to optimise the dosages of bio-rejuvenators for rejuvenating aged bitumen. Six bio-rejuvenators were used to validate the feasibility of the employed rejuvenation index, which was calculated using frequency sweep tests on binders. Bending beam rheometer (BBR) tests and multiple stress creep and recovery tests (MSCR) were carried out to identify the lower and higher limits of dosages for each rejuvenator to validate the feasibility and accuracy of the rejuvenation index. Results from these measurements were further corroborated using linear amplitude sweep (LAS) tests. Lastly, Fourier Transform Infrared Spectroscopy (FTIR) tests were carried out to characterise the rejuvenated binders and the potential use of chemistry-based indices to comprehend the rejuvenation processes. Overall, this work presents comprehensive evidence to validate the feasibility of using a rheology-based method to optimise the dosages of bio-rejuvenators reliably, for the efficient rejuvenation of bitumen.

3. Materials and methods

3.1. Materials

The bitumen used in this study was a 70/100 penetration grade bitumen with a penetration of 81 (0.1 mm) and softening point of 45.4 °C. The performance grade of this bitumen was PG 70–22. Six bio-rejuvenators were used in this study, three of them were commercial products while others were waste oils. Some information of the rejuvenators is shown in Table 1.

3.2. Methods

The virgin bitumen was aged using a rolling thin film oven (RTFO) at 163 ℃ for 75 mins as per BS EN 12607–1 [35]. The residue was then subjected to pressure ageing vessel (PAV) ageing at 100 ℃ and 2.1 MPa for 20 h as per BS EN 14769 [36]. Every batch of aged bitumen was poured into a large container and blended using a mixer at 160 ^{o}C and 3000 r/min for 10 min to ensure homogeneity. Subsequently, 100 g of aged bitumen was extracted from the large container and poured into a small container, then the accurately weighted rejuvenators at designed dosages were added to the aged binders and blended for another 10 mins at the temperature of 160 ^{o}C. The dosages of each rejuvenator were chosen following the manufacturers' manual and literatures [37,38].

Frequency sweep tests were conducted using a Kinexus Pro DSR at a constant stain level of 0.2 % over the temperature range of 10–70 ℃ at intervals of 10 ℃ and frequencies ranging from 0.1 rad/s to 100 rad/s with 10 readings per decade. The geometry configuration consisted of 25 mm parallel plates with a gap of 1 mm. The linear amplitude sweep (LAS) tests were carried out at 25 ℃ as per AASHTO T391–20 to determine the fatigue properties of binders [39]. The multiple stress creep and recovery (MSCR) tests were carried out at 70 ^{o}C as per BS EN 16659 to determine the high-temperature performance of bitumen in terms of ageing and rejuvenation [40]. The bending beam rheometer (BBR) tests were carried out using a Cannon BBR to measure the low-temperature properties of binders. The testing temperature ranged from -12 ℃ to -30 ℃ with an interval of 6℃ to obtain the critical temperatures and delta *Tc* for binders. A Bruker Tensor 27 FTIR spectrometer equipped with an attenuated total reflection (ATR) unit was employed to obtain the functional groups of rejuvenators and binders. The scanning range was 4000–400 cm^{-1} with the resolution of 4 cm^{-1} [41].

4. Data analysis

4.1. Frequency sweep

The master curve is commonly used to represent the relationship between the complex modulus (and phase angle) and frequency of a bitumen [42]. The shift factor $\alpha(T)$for building master curves was obtained with Eq. (1):

$$\log \alpha(T) = -\frac{C_1(T-T_R)}{C_2+(T-T_R)} \tag{1}$$

Where,

C_1 and C_2 = Shifting parameters

T and T_R = Testing and reference temperature,"°C

The modified Christensen-Anderson-Marasteanu (CAM) model in Eq. (2) was employed to build master curves of the complex modulus (G^*) of all binders [43]. The master curves for phase angle were built as per Eq. (3). At the present study, 25 ℃ was selected as the reference temperature.

$$G* = \frac{G_g^*}{\left[1+(f_c/f_r)^k\right]^{m/k}} \tag{2}$$

$$\delta = 90I - \frac{90I-\delta_m}{\left\{1+\left[\frac{\log(f_d/f_r)}{R_d}\right]^2\right\}^{\frac{m_d}{2}}} \tag{3}$$

Where,

G^*_g = glass complex modulus

f_c = crossover frequency

f_r = reduced frequency

m,k = fitting parameters

δ_m= phase angle constant at f_d

f_d = location parameter with dimensions of frequency

R_d, m_d = shape parameters

$I = 1$ if $f \le f_d$, $I = 0$ if $f > f_d$

Table 2
Assigned vibration associated with chemical functional groups [26].

Appearing bands (cm^{-1})	Assigned vibrations	Functional groups	Related fractions
720	r(CH_{2n})	Alkyls	Saturates
750	γ CH_{aro}	Aromatics	Aromatics, Asphaltenes
810	γ CH_{aro}	Aromatics	Aromatics, Asphaltenes
860	γ CH_{aro}	Aromatics	Aromatics, Asphaltenes
1030	v S=O	Sulfoxides	Resins, Asphaltenes
1080	v_s SO_3	Sulfate Ester	Asphaltenes
1270	v_{as} SO_3	Sulfate Ester	Asphaltenes
1380	δ_{as} CH_2/CH_3	Alkyls	Saturates
1455	δ_{as} CH_2/CH_3	Alkyls	Saturates
1600	v C=C	Aromatic	Aromatics
1655	v C=O	2-Quinolone	Resins
1700	v C=O	Ketone	Aromatics, Resins, Asphaltenes
1730	v C=O	Carboxylic acid	Resins
2850	v_s CH_2/CH_3	Alkyls	Saturates
2920	v_{as} CH_2/CH_3	Alkyls	Saturates

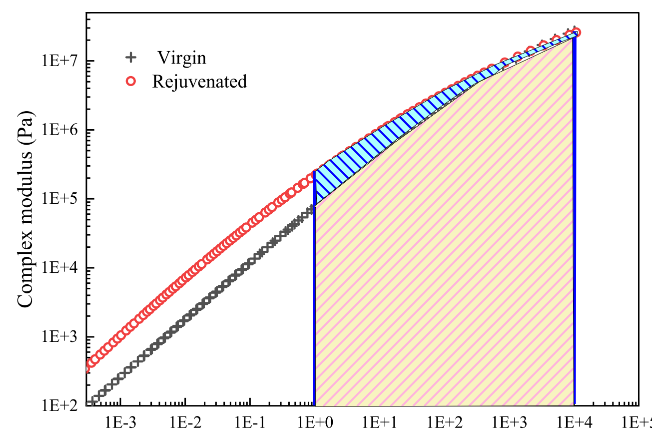


Fig. 1. Schematic of rejuvenation index.

4.2. Fourier-transform infrared spectroscopy (FTIR)

The spectra were normalised based on the aliphatic band, whose wavenumber is around 2920 cm^{-1} to increase the visibility of the peaks [44]. The carbonyl (C=O) with wavenumbers between 1800 and 1660 cm^{-1} was picked to evaluate the changes in chemical properties of bitumen during ageing and rejuvenation using the carbonyl index, which is the ratio between carbonyl and the peak at wavenumber of 2920 cm^{-1}, whose intensity does not change with ageing. The assigned vibration associated with different functional groups found in bitumen and its fractions are shown in Table 2.

5. Results and discussion

5.1. Rejuvenation effect evaluation and dosage optimisation

To quantify the holistic impact of bio-rejuvenation on the rheological properties of bitumen, the rejuvenation index (*RI*) was proposed, as defined in Eq. (4), which is based on previous works as detailed in

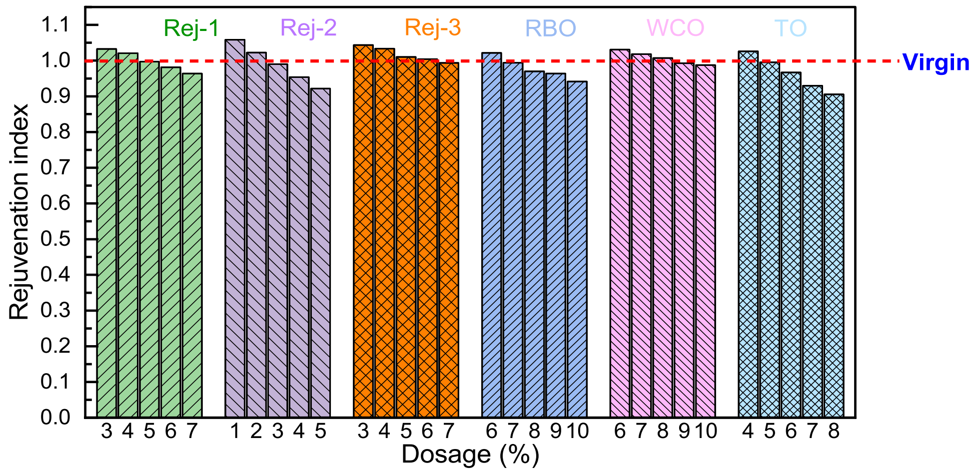


Fig. 2. Rejuvenation index of all binders using different rejuvenators at different dosages.

Table 3
Relationship between dosages and rejuvenation indices.

Rejuvenators	Fitting models	Fitting accuracy (R^2)	Predicted optimal dosage (%)
Rei-1	*RI=1.08768–0.0177x*	0.99	5.0
Rei-2	*RI=1.09217–0.0343x*	1.00	2.7
Rei-3	*RI=1.08189–0.0130x*	0.97	6.3
RBO	*RI=1.13034–0.0190x*	0.96	6.9
WCO	*RI=1.09729–0.0113x*	0.98	8.6
TO	*RI=1.14897–0.0307x*	0.99	4.9

literature [24,45]. Ageing mainly has a negative effect on the low-temperature performance of bitumen, therefore, the boundaries of the integration were set as 0 and 4 (1E0 to 1E4 of the frequencies) respectively to target the complex modulus values found at low temperatures (high frequencies). Fig. 1 provides a schematic representation of the *RI* using Rej-1 rejuvenated bitumen at a dosage of 3 % as an illustrative example.

$$RI = \frac{\int_0^4 \log \left|G*_{aged/rejuvenated}\right| \xi d\xi}{\int_0^4 \log \left|G*_{virgin}\right| \xi d\xi} \quad (4)$$

Mathematically, *RI* values larger than one indicates the bitumen is aged or insufficiently rejuvenated, while on the contrary, the *RI* values smaller than one indicates over-rejuvenation. Therefore, the closer *RI* is to one, the better the rejuvenation effectiveness.

The area encompassed by the rejuvenated binder's master curve was found to be larger than that of the virgin binder, indicating suboptimal rejuvenation. A comprehensive depiction of the rejuvenation indices for all rejuvenated binders, using varying rejuvenators at different dosages, are shown in Fig. 2.

The results first and foremost demonstrated the efficacy of these bio-based rejuvenators in restoring the rheological properties of aged bitumen. All rejuvenators showed efficacy in recycling aged bitumen, with the tall oil even demonstrating performance on par with commercially available products. As shown in Fig. 2, all rejuvenated binders exhibit rejuvenation indices greater than one when dosages were relatively low, suggesting incomplete rejuvenation. Conversely, increasing the dosages led to a consistent decrease in the rejuvenation index, which might induce over-rejuvenation and result in unnecessary material consumption and even high-temperature related issues such as rutting.

Observations from Fig. 2 suggested that there are clear linear correlations between dosages and rejuvenation indices, indicating the possibility of predicting the ideal dosage for each combination of aged bitumen and rejuvenators with just two sets of testing. The linear regression models were derived and displayed in Table 3, which illustrated the predicted ideal or optimal dosages for each rejuvenator.

High fitting accuracies, as shown in Table 3, suggested that the linear models are suitable for calculating optimal dosages of rejuvenators to restore the rheological properties of aged bitumen. Notably, the optimal dosage for Rej-2 was only 2.7 %, achieving a rejuvenation effect comparable to waste cooking oil, which required a significantly higher dosage of 8.6 %. Moreover, binders rejuvenated by Rej-2 had the highest fitting accuracy, showcasing the efficiency of this rejuvenator. Surprisingly, Tall oil performed as well or even better than some commercial bio-rejuvenators, indicating its substantial potential for broader use as a sustainable rejuvenator. However, the performance of waste cooking oil was subpar, as seen by the high dosages required to achieve sufficient rejuvenation.

In accordance with the fitting results in Table 3, the proposed rejuvenation index can potentially be used as a tool for optimising the dosage of rejuvenators. Since the rejuvenation index has a linear correlation with the dosage, a fitting function can be obtained. Based on this function, the optimal dosage for any rejuvenators can be back calculated with reference to the virgin binder. Though the proposed rejuvenation index has the potential to optimise the dosage accurately, it's feasibility still needs to be further validated.

The rejuvenation index is mainly based on the low-temperature performance of bitumen. However, the low-temperature performance was estimated from master curves rather than actual measured results at negative temperature. Though normally the estimated results from master curve are reliable, to solidify the evidence, experimental measurements are still required since the frequency sweep cannot capture some critical properties such as m-value, critical temperatures and delta T_c, which have direct impact on the performance of rejuvenated bitumen [46]. Therefore, the low-temperature performance of rejuvenated binders with varying dosages should be evaluated experimentally to validate this rheology-based index and assure that the low-temperature performance can be recovered sufficiently. Moreover, the high-temperature performance should also be examined to validate that the recovery of low temperature will not diminish the high-temperature performance, such as the resistance to permanent deformation, which can be indicated by the means of MSCR tests [47]. Lastly, the fatigue performance should also be validated to have a comprehensive evaluation of the rejuvenation efficiency.

5.2. Low-temperature performance evaluation

Thermal cracking of asphalt pavement is one of the most severe distress caused by bitumen ageing [48]. When the extent of cracking is high, pavements then need to be milled, resulting in materials that are as

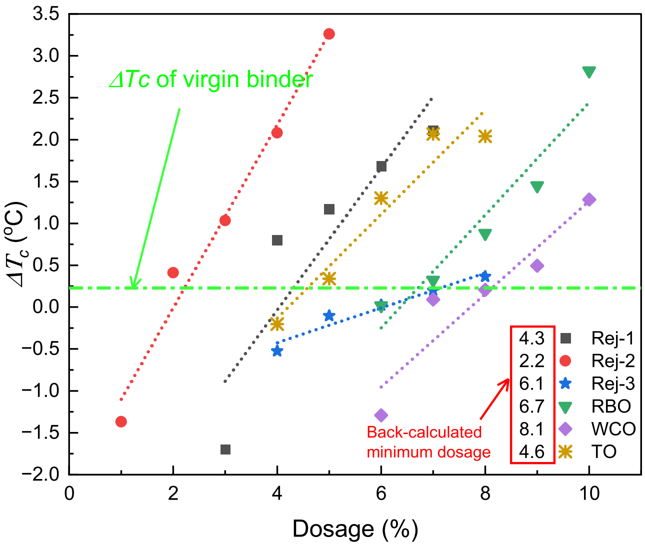


Fig. 3. Correlation between dosage and delta T_c.

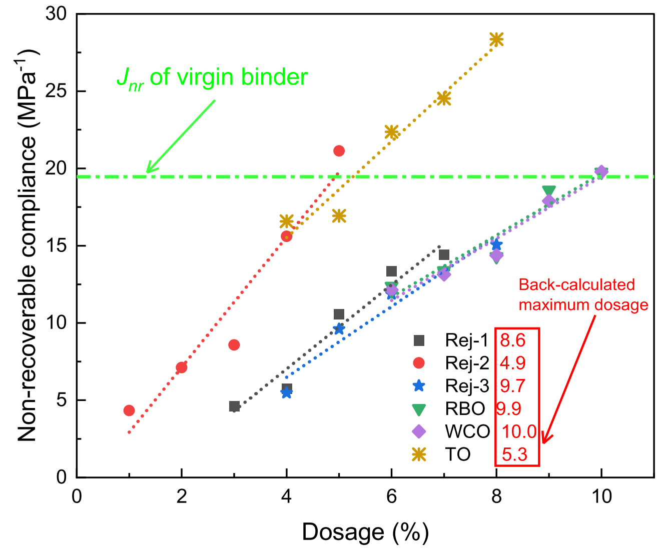


Fig. 4. Correlations between dosage and nonrecoverable compliance.

termed as reclaimed asphalt pavement (RAP). Using RAP in new pavement construction has now become a necessity to achieve high levels of circular use in pavement materials [49,50]. However, the bitumen present in RAP is aged, therefore some measures need to be taken to recover the properties of aged bitumen to ensure that new mixtures meet mix and performance target requirements, especially for the low-temperature performance which is closely related to thermal cracking susceptibility [51,52]. Rejuvenation is a possible solution for recovering the low-temperature performance of aged binder and thereby to assure the low-temperature performance of new pavements with high RAP content [49].

The BBR tests were carried out on virgin bitumen, aged bitumen, and rejuvenated bitumen with six types of rejuvenators, each rejuvenator with five dosages. The lower Performance Grade (PG) of bitumen, the critical temperatures as well as the ΔT_c were measured and calculated. Previous studies have reported that for the low-temperature performance of bitumen, the virgin binders are stiffness controlled while the aged binders are m-value controlled [53]. Therefore, ΔT_c normally is positive for virgin binders while negative for aged binders. Studies showed that ΔT_c has a direct influence on block cracking and indirect influence on fatigue, longitudinal and transverse cracking etc. [54]. Therefore, this study employed ΔT_c as the target for rejuvenation of the low-temperature performance of bitumen. The results are shown in Fig. 3.

As shown in Fig. 3, dosage has a linear correlation with ΔT_c, with the fitting accuracies (R-square) ranging from 0.81 to 0.98. As expected, the ΔT_c of rejuvenated binders changed from negative to positive with higher dosages ultimately resulting in more positive values of ΔT_c. The ΔT_c for the virgin binder was 0.23 ^{o}C while it was -2.31 ^{o}C for the laboratory aged binder. Based on the regression functions, the equivalent dosages for Rej-1, Rej-2, Rej-3, RBO, WCO and TO were 4.3 %, 2.2 %, 6.1 %, 6.7 %, 8.1 % and 4.6 % respectively. These dosages were the minimum dosages for each rejuvenator to recover the low-temperature performance of rejuvenated binders sufficiently. Comparing the dosages calculated from the rejuvenation indices and from the low-temperature performance, it was found that the dosages calculated from the rejuvenation indices were a slightly higher than the lower limit of dosages, indicating the rejuvenation index is reasonable and applicable.

5.3. High-temperature performance evaluation

Ageing leads to the stiffening of bitumen and improves its high-temperature performance such as rutting resistance [38]. Rejuvenation generally has a negative effect on the high-temperature performance of bitumen since rejuvenation leads to the softening of aged bitumen. Previous studies reported that if the dosages of rejuvenators were too high, the high-temperature performance might be negatively impacted, which is unfavourable [55]. Therefore, the upper limit of dosage of rejuvenators is controlled by the high-temperature performance of bitumen [28,52,56].

The Multiple Stress Creep and Recovery (MSCR) Test is currently being considered as a replacement for the Superpave high temperature binder criteria G*/ sin δ since it provides a better correlation to permanent deformation of asphalt mixtures [57]. The nonrecoverable compliance (J_{nr}) and recovery percent ($\%R$) are the parameters reported from this test. The J_{nr} is a measure of the amount of residual strain left in the specimen after repeated creep and recovery, relative to the amount of stress applied. The $\%R$ is a measure of how much the sample returns to its previous shape after being repeatedly stretched and relaxed [58]. It was reported that the J_{nr} at 3.2 kPa is more efficient in determining the dosages of rejuvenators [28,37], therefore, this study employed J_{nr} at 3.2 kPa as the index to determine the upper limit of the dosages for each rejuvenator. Similar to the lower limits of the dosages, upper limits showed linear correlations with J_{nr} at 3.2 kPa, as shown in Fig. 4.

Regression functions were obtained to back calculate the dosage relevant to the value of virgin binder. The fitting accuracies were reasonably acceptable, with the R-squares for Rej-1, Rej-2, Rej-3, RBO, WCO and TO rejuvenated bitumen being 0.95, 0.94, 0.95, 0.92, 0.95 and 0.95 respectively. As expected, the upper limits of dosages for every rejuvenator were higher than those determined based on the rheology-based rejuvenation index, which again showed that the rejuvenation index is reliable since it falls between the lower and upper limits determined by low-temperature and high-temperature performance respectively.

5.4. Fatigue properties of rejuvenated binders

To assess fatigue performance, the LAS test was employed to accurately simulate damage. The data from this test allows the estimation of the approximate number of cycles to failure, N_f, for any desired strain. The AASHTO T391–20 standard recommends strains of 2.5 % and 5 % without specifying values for fatigue life [39]. However, an increasing number of publications have pointed out that these relatively low strains fail to determine the fatigue life of binders, resulting in a poor correlation with the fatigue performance of asphalt mixtures [59,60].

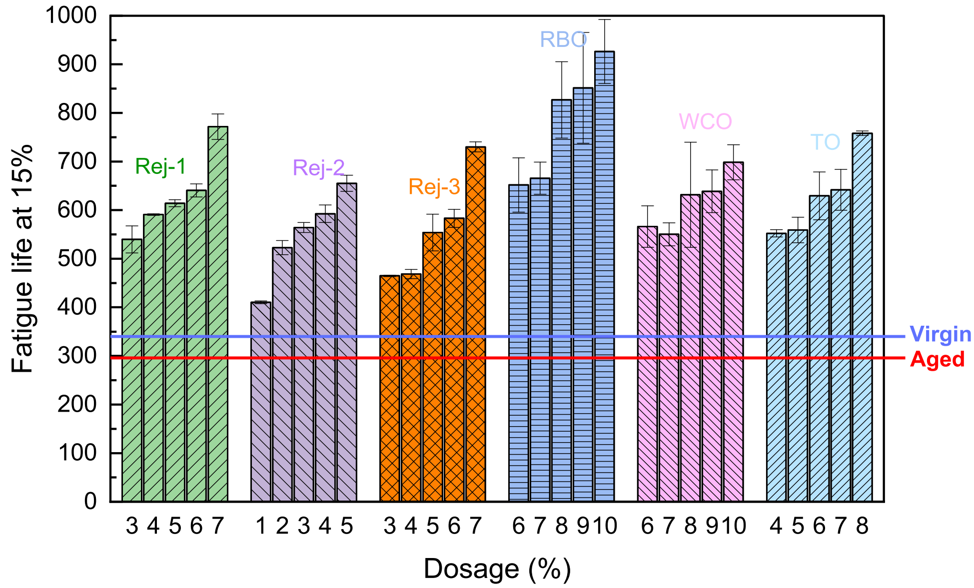


Fig. 5. Fatigue life of binders at strain level of 15 %.

Table 4
The rankings of the fatigue lives of rejuvenated binders in terms of dosages.

	Rej-1	Rej-2	Rej-3	RBO	WCO	TO
Dosage I	5	5	4	5	5	5
Dosage II	4	4	5	4	4	4
Dosage III	3	3	3	3	3	3
Dosage IV	2	2	2	2	2	2
Dosage V	1	1	1	1	1	1

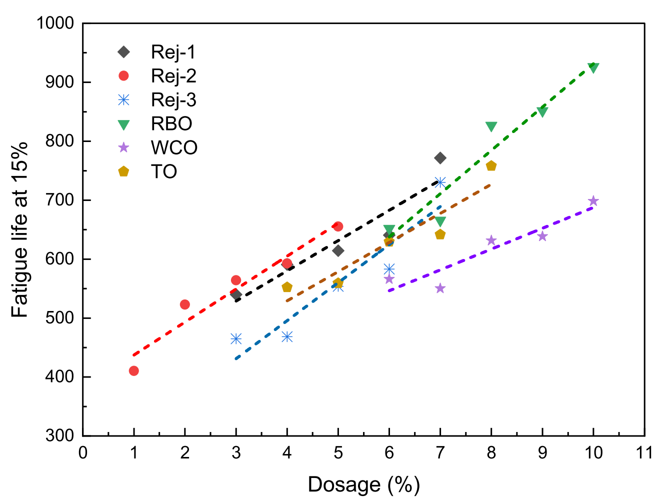


Fig. 6. Correlations between dosage of rejuvenators and fatigue life of rejuvenated binders.

Moreover, fatigue life at lower strains tends to be inconsistent in relation to the modification or ageing of bituminous materials [18]. In response to this, a broader range of strains was selected in this study, with the aim of producing more accurate results. The strain levels specifically employed in this study were 2.5 %, 5 %, 7.5 %, 10 %, 12.5 %, and 15 % respectively. The fatigue lives of all binders calculated at strain level of 15 % are depicted in Fig. 5.

Data at 15 % strain of LAS tests demonstrated a clear and strong proportional relationship. An increase in rejuvenator content resulted in an increase in fatigue life – with all dosages exceeding the original

Table 5
Fitting models for fatigue life against dosage.

Rejuvenators	Fitting models	Fitting accuracy (R^2)
Rei-1	N_f=51x+375	0.88
Rei-2	N_f=56x+381	0.94
Rei-3	N_f=65x+237	0.89
RBO	N_f=73x+196	0.93
WCO	N_f=35x+335	0.87
TO	N_f=49x+332	0.89

fatigue life of the virgin binder. To further summarise the relationship between the dosages and the fatigue life of bitumen, the fatigue life of each binder was ranked in Table 4. A ranking of 1 suggests that the best performance or highest fatigue life, and a ranking of 5 suggests the worst performance or lowest fatigue life. Considering that the dosages for each rejuvenator were different, codes representing the dosages from lowest to highest were allocated, which were Codes I, II, III, IV, and V respectively.

As illustrated in Table 4, when the strain level was set at 15 %, the ranking shows an organised trend. Binders with higher dosages exhibited a higher fatigue life, with only one exception which might be due to instrumental or experimental errors. Furthermore, the correlations between the dosages of rejuvenators and fatigue life of binders were established, as shown in Fig. 6 and Table 5.

Fig. 6 illustrates a general linear correlation between the dosages of the rejuvenators and the fatigue life of rejuvenated binders. The best fit was observed with Rej-2-rejuvenated binder, featuring a fitting accuracy (R^2) of 0.94. Conversely, the poorest fit was observed for the waste cooking oil rejuvenated binder, with an R^2 value of 0.87. Fig. 6 indicates that at 15 % strain, the rejuvenators Rej-1, Rej-2, and tall oil require lower dosages than other rejuvenators to get a higher value of fatigue life. This suggests that these types of rejuvenators are more effective at higher strains, thus requiring less rejuvenator to restore the fatigue resistance. This conclusion aligns with that drawn from the rejuvenation index, validating the reliability of the rejuvenation index as a suitable indicator. Moreover, these results indicated that fatigue life of rejuvenated bitumen can be improved even when the dosages of rejuvenators were relatively low.

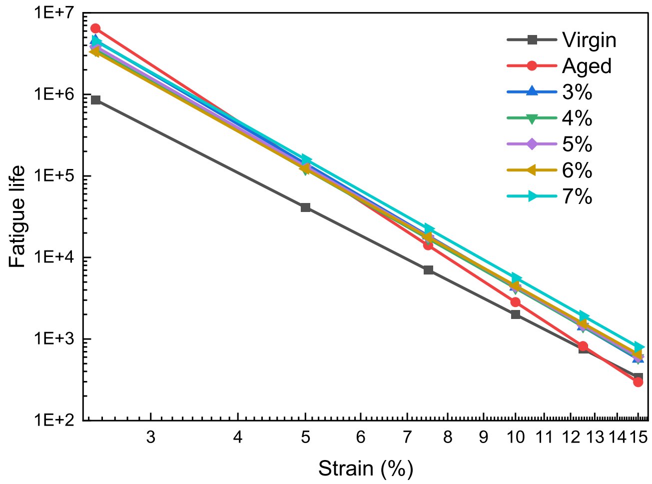


Fig. 7. Correlation between fatigue life and strain.

5.5. Strain sensitivity

The fatigue life of bituminous materials is strain-dependent, with higher strain typically resulting in lower fatigue life. As six different strain levels were applied in this study, the relationships between these strain levels and fatigue life were visualised and plotted in Fig. 7.

Taking Rej-1 rejuvenated binders as an example, it can be observed from Fig. 7 that when the strains were relatively low, the fatigue life of virgin binder was lower than that of aged binder. However, when the strains were relatively high, the fatigue life of virgin binder was higher than that of aged binder. Notably, the fatigue life of rejuvenated binders was consistently higher than that of virgin binder, irrespective of the strain levels. Further analysing the trend of fatigue life change with strain, it was found that the fatigue life of aged binders changed dramatically with strain, indicating that the aged binder was the most strain-dependent one. Moreover, the strain-fatigue life curves for rejuvenated were parallelly with increased dosage and showed less strain-dependency compared to aged binder. A potential reason is that rejuvenators enhance compatibility between new and recycled binders by reducing the intermolecular cross-linking between chemical fractions such as asphaltene and resins, which increases the mobility of the bitumen molecules and reduces its strain sensitivity [61].

5.6. Chemical characterisation of rejuvenated binders

The FTIR technique can provide insights into the chemical composition of a material and can be used to determine whether the rejuvenation process of aged bitumen is primarily physical or chemical [62]. In general, the presence of new peaks suggests chemical interaction between binders and bio-rejuvenator, which would support the hypothesis that the rejuvenation process is at least partially chemical [31]. If there is no significant change in functional groups, it might suggest that the process is mainly physical [62]. Prior to determining the chemical changes of bitumen, the chemical profiles of rejuvenators were measured, as shown in Fig. 8.

Rej-1 primarily consists of saturated hydrocarbons like alkyls, a finding substantiated by the five spectral bands attributed to the alkyl functional group (2920, 2855, 1460, 1380, and 720 cm^{-1}) [63] and a substantial concentration of carboxylic acids and sulfones, with a minimal presence of alkenyl, sulfate ester, 2-Quinolone, and aromatic compounds. Rej-2 shares significant chemical similarities with Rej-1, the primary difference being a considerably reduced sulfone content in Rej-2 as well as the presence of secondary alcohol. Rej-3, on the other hand, is chemically simpler, comprising mainly of alkyls and aliphatic ether, along with trace amounts of carboxylic acids, sulfate ester and sulfones. Tall oil is rich in alkyls and ketones, also contains a substantial quantity of alkenes and aromatic ester and small amount of sulfate ester, 2-Quinolone, and alkenyls. Recycled biofuel oil presents the simplest chemical structure among all rejuvenators, primarily includes alkyls with scant quantities of carboxylic acids and sulfones. Waste cooking oil comprises mainly alkyls, carboxylic acids, sulfate ester, and sulfones, with slight aromatics and alkenes. The FTIR results of rejuvenated binders with lowest and highest dosages of each rejuvenator, for the sake of brevity, are shown in Fig. 9.

Fig. 9 illustrated that the rejuvenated binders encompass the chemical functional groups originating from both the rejuvenators and the aged bitumen. Notably, while some peak intensities of the spectra were altered, no new functional groups emerged. However, some functional groups found in rejuvenators disappeared after blending with bitumen. The reason behind the peak disappearance is that the intensities of these disappeared functional groups were extremally weak, after being blended with aged bitumen, the intensities were even weaker, which the

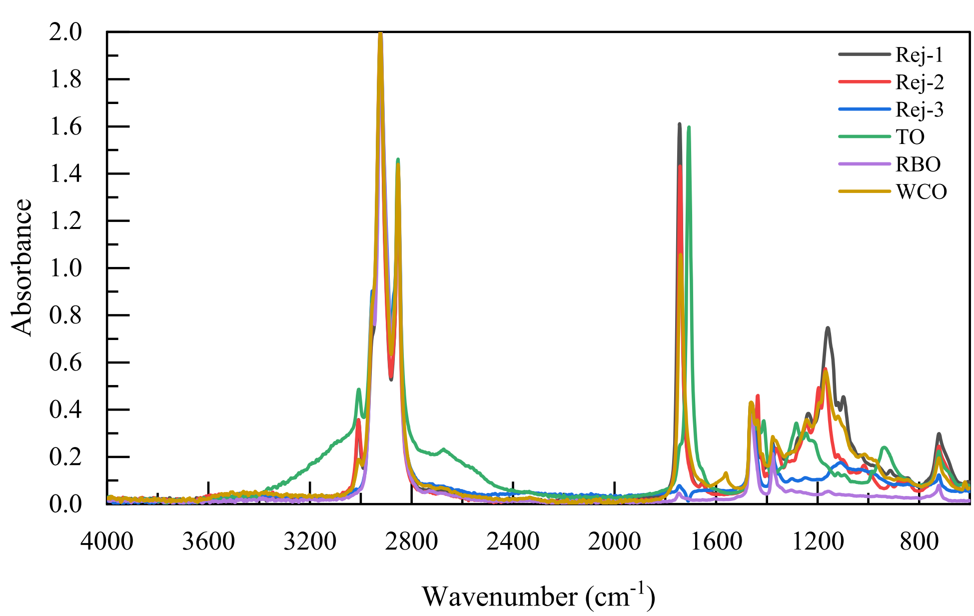
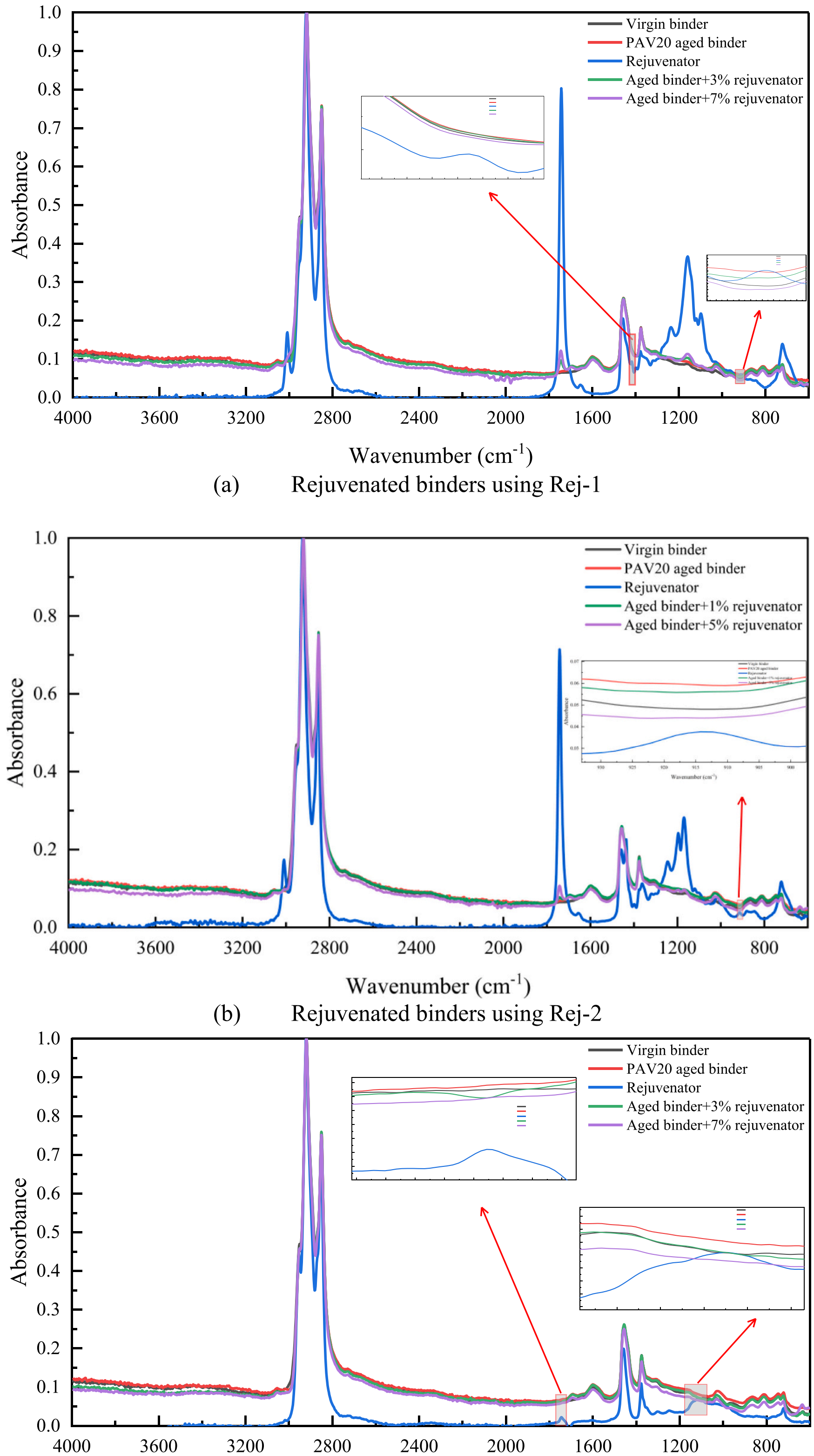


Fig. 8. FTIR spectra of rejuvenators.



(a) Rejuvenated binders using Rej-1

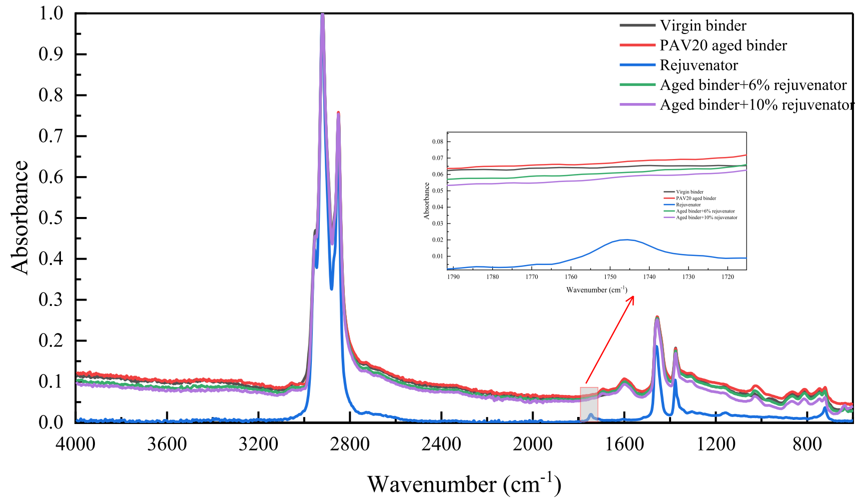
(b) Rejuvenated binders using Rej-2

(c) Rejuvenated binders using Rej-3

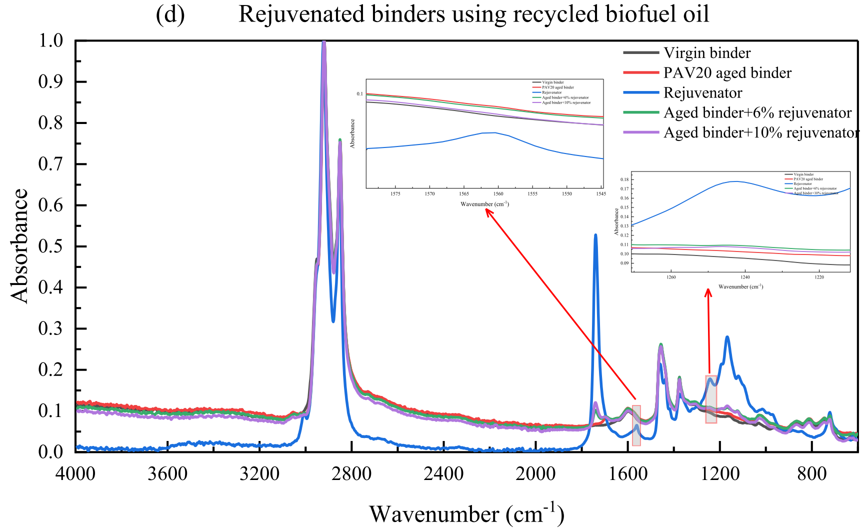
Fig. 9. Chemical properties of bitumen and rejuvenators during rejuvenation.

equipment cannot recognise.

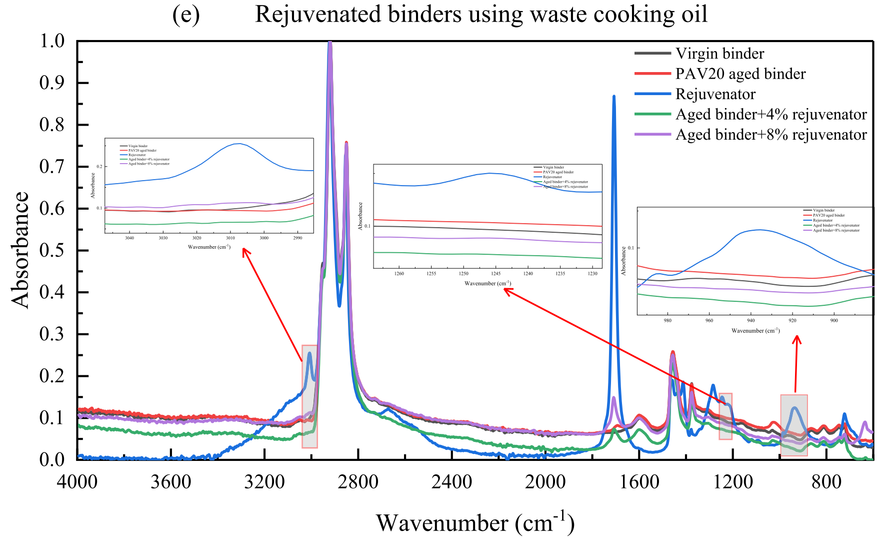
Carbonyls are considered as an oxidation signature and the carbonyl index is typically used to illustrate the chemical changes in bitumen in terms of ageing [63]. This study employed this index to represent the changes in the oxidation signature, attempting to uncover whether rejuvenation can reduce the extent of ageing. The results are depicted in



(d) Rejuvenated binders using recycled biofuel oil



(e) Rejuvenated binders using waste cooking oil



(f) Rejuvenated binders using tall oil

Fig. 9. (*continued*).

Fig. 10. Though sulfoxide is also regarded as an ageing marker, its high sensitivity and simultaneous formulation and degradation during ageing process make it an unreliable marker for characterising the ageing process of bitumen [64].

It was seen from Fig. 10 that carbonyl indices for two out of the six rejuvenators decreased with increased dosages, while the remaining four showed an increase. Looking closely at the chemical functional groups of these two rejuvenators, it is noticeable that they either do not contain or scarcely contain carbonyls. The decrease in the carbonyl index was likely extensively due to the physical dilution of carbonyls

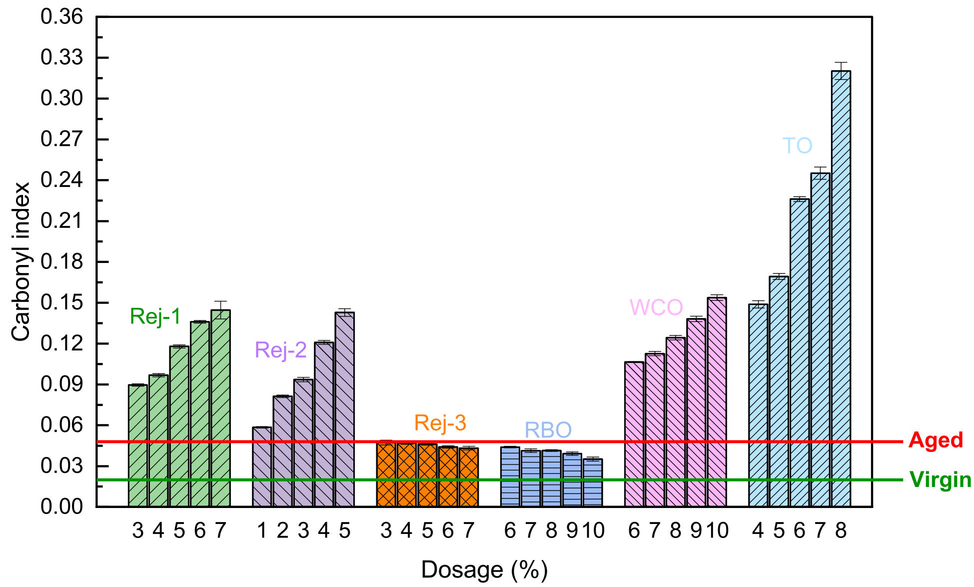


Fig. 10. Carbonyl index of binders before ageing, after ageing and after rejuvenation.

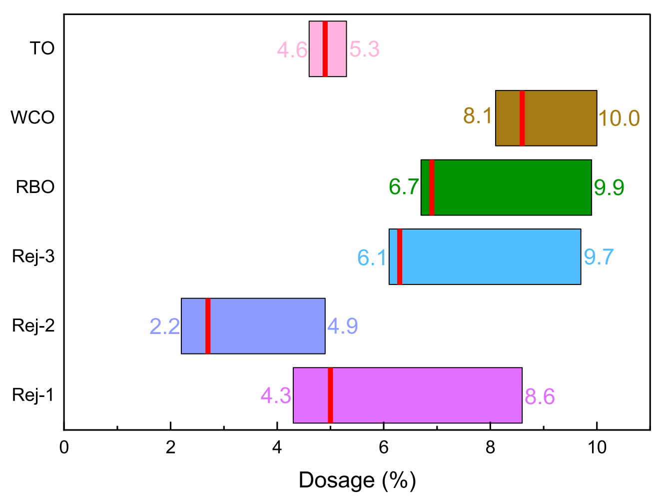


Fig. 11. Dosage optimisation approach considering low- and high-temperature performance.

[17]. For example, if the carbonyl index of aged bitumen is "a" and the carbonyl index of the rejuvenator is 0, with a mass ratio of aged bitumen to rejuvenator of 100:3, the carbonyl index of the rejuvenated binder should be $(100a+0)/103=0.971a$.

In that sense, the carbonyl index of binders rejuvenated with RBO was lower than those rejuvenated with Rej-3 as the dosages of recycled biofuel oil were higher than that of Rej-3. When Rej-3 and RBO were used at the same dosages, the carbonyl indices of the rejuvenated binders were nearly identical, reinforcing the validity of this assumption. For the binders rejuvenated by the other four rejuvenators, their carbonyl indices increased continuously with dosages as these rejuvenators are rich in carbonyls, which increases the carbonyl index of the mixture, and similar results were reported from literature [16,65]. Overall, rejuvenators softened the aged bitumen by adding light components with smaller molecules, as all rejuvenators are found to be rich in alkyls. Therefore, using simple FTIR test based parameters for developing rejuvenation target is not feasible since there is no direct correlation between the chemical properties (or the fingerprint functional groups) of rejuvenated bitumen and the rejuvenation efficiency.

5.7. Recommendation for the dosage optimisation of bio-rejuvenators

The previous subsections revealed that the rheology-based rejuvenation index is a reliable and practical. To have a clear understanding of this rejuvenation index and further validate its feasibility, the dosage limits determined by both low-temperature and high-temperature performance are plotted together in Fig. 11.

Fig. 11 visually showcases the relationship between the determined optimal dosages and the measured limits. Clearly, the optimal dosages (red vertical lines in Fig. 11) fall into the upper and lower limits, which demonstrates the reliability and feasibility of the rheology-based rejuvenation index. Moreover, it was observed that the optimal dosages were closer to the lower limits, which suggests that using this approach can effectivity save the quantity of rejuvenators and simultaneously leave enough safe leeway for uncertainties during the production and construction processes.

Overall, the rheology-based rejuvenation index has significant advantages: 1) it's a comprehensive index considering a range of frequencies and temperatures, which can overcome the deficiencies associated with traditional single point based indices; 2) Only very small quantity of materials are needed for testing, which is time and material efficient; 3) Using this approach, the low-temperature performance of aged bitumen can be efficiently recovered without compromising the high-temperature performance; 4) Take advantage of linear correlation with dosage, only two trials would be required to calculate the optimal dosage.

6. Conclusion

The objective of this study was to validate a rheology-based rejuvenation index by comprehensively evaluating its feasibility to predict the optimal dosages of bio-rejuvenators for asphalt recycling. Based on the results, the following conclusions can be drawn.

- ➢ The proposed rheology-based rejuvenation index, based on the master curve of complex modulus, proved to be an accurate tool for evaluating rejuvenation efficiency and predicting optimal rejuvenator dosages. The rejuvenator dosage is linearly proportional to its efficiency, and it was feasible to predict the optimal rejuvenator dosage with just two trials.
- ➢ Both commercial bio-rejuvenators and waste oils successfully recovered the properties and fatigue performance of binders

adversely affected by ageing and reduced the strain sensitivity of binders. A linear correlation was found between the increase in rejuvenator content and fatigue life of bitumen at a strain of 15 % in the LAS tests. This supports the feasibility of a sustainable approach to bitumen rejuvenation, creating a synergistic benefit for both waste bituminous materials and industrial or household oil wastes.
- ➢ Using waste cooking oils and recycled biofuel oils was seen to be less effective for rejuvenating the aged bitumen, while tall oil exhibited high potential. A significant disparity was observed in the effectiveness of different rejuvenators.
- ➢ FTIR results indicated that the chemical properties of rejuvenated bitumen were mostly unchanged. Therefore, the use of simple chemistry-based rejuvenation indices is not feasible.

This study evaluated and validated a rheology-based rejuvenation index for optimising the dosage of bio-rejuvenators. The results illustrated that this index works adequately for rejuvenated binders and can assure that the low-temperature performance is recovered while the high-temperature performance is not diminished. It should be noted that this index was only validated at the binder scale and further study is recommended to evaluate the feasibility of this index at the mixture level.

CRediT authorship contribution statement

Bhupendra Singh: Writing – review & editing. **Haopeng Wang:** Writing – review & editing, Investigation, Formal analysis. **Lu Zhou:** Writing – review & editing, Formal analysis. **Yongping Hu:** Writing – original draft, Methodology, Investigation, Funding acquisition, Formal analysis, Data curation, Conceptualization. **Gordon Airey:** Writing – review & editing, Supervision, Resources, Methodology, Investigation, Formal analysis. **Max Allanson:** Investigation, Formal analysis, Data curation. **Ana R. Pasandín:** Resources, Data curation. **Jack Ryan:** Investigation, Formal analysis, Data curation. **Anand Sreeram:** Writing – review & editing, Supervision, Methodology, Investigation, Formal analysis.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data Availability

Data will be made available on request.

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