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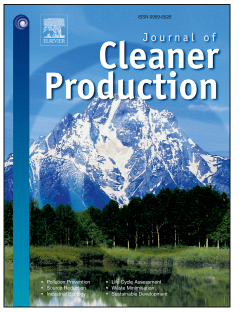

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Yang Yang: Investigation, Methodology, Resources.

Yangming Gao: Investigation, Methodology, Resources.

Yuqing Zhang: Funding acquisition, Methodology, Supervision, Writing - review & editing.

All authors interpreted and reviewed the results and approved the manuscript.

Sincerely yours,

Signed by Corresponding Author on behalf of all authors.

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Healing Characterisations of Waste-derived Bitumen Based on Crack Length: Laboratory and Modelling

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Healing Characterisations of Waste-derived Bitumen Based on Crack Length: Laboratory and Modelling

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Abstract: The accumulations of waste plastics and municipal solid wastes from the resident groups and industrial companies have been causing serious environmental issues in the UK, due to difficulties in logistics, sorting, and reuse. In this sense, this paper is stimulated to develop novel approaches to convert some wastes into eco-friendly infrastructural materials, which provides a new way of waste reduction and reuse and extending the service life of asphalt pavement. This study aims to characterise the healing performance of waste-derived bitumen before and after pressure ageing vessel (PAV) ageing based on the crack length. One waste-derived bitumen was fabricated by blending the bio-oil pyrolysed from the organic fraction of municipal solid waste (5wt.%) with a control bitumen (X70) using a high shear mixer at a speed of 150 revolutions per minute (RPM) for 30 minutes at 150°C under nitrogen atmosphere. A second waste-derived bitumen was fabricated using the low-density polyethene (LDPE) and mixed with the control bitumen at a concentration of 6wt.% at a speed of 900RPM for 90 minutes at 180°C under nitrogen atmosphere. Crack length-based healing index and Ramberg-Osgood model were employed to characterise the healing rate and healing capability of the bitumen, respectively. Material properties (e.g., relaxation modulus and surface energy) of the bitumen used in the healing models were calibrated by linear amplitude sweep test (10Hz and 20°C), frequency sweep test (10Hz, 10~70°C), and time sweep fatigue-healing test (10Hz and 20°C) at a controlled strain level of 5% with different rest durations. Advancing contact angles used in the healing models of the bitumen were measured by a sessile drop tensiometer based on the tilting cradle method. Results show that the bio-oil productively promotes the healing potential and capability of the unaged bitumen, and the LDPE slightly strengthens them. The PAV ageing process evaporates most of the modifier bio-oil; hence, the PAV-aged bio-oil modified bitumen does not show better healing performance than that of the PAV-aged control bitumen. The PAV-aged LDPE modified bitumen has much better healing performance than those of the PAV-aged control bitumen and the PAV-aged bio-oil modified bitumen. The short-term healing rate and healing potential dominate the healing behaviours of the bitumen. The unaged bio-oil modified bitumen heals the fastest (having the highest short-term healing rate) and most (having the highest healing potential), followed by the unaged LDPE modified bitumen, and the unaged control bitumen heals the least and slowest. The PAV-aged LDPE modified bitumen heals the fastest and most, followed by the PAV-aged bio-oil modified bitumen and the PAV-aged control bitumen. The fundamental reason for LDPE's enhancement to bitumen's healing is that the LDPE increased the deformation recovery ability of the bitumen, leading to a higher wetting rate of the cracked surfaces and thus a higher short-term healing rate. However, the LDPE in bitumen cannot accelerate the molecular diffusion to increase the intrinsic healing or the long-term healing rate.

Keywords: Waste-derived Bitumen, Ageing, Healing, Plastics

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1. Introduction

Petroleum-derived bitumen, the residue from crude oil refining, is used as a binder in asphalt concrete for constructing and maintaining over 95% of the UK's transport infrastructures, such as roads (Lvel et al., 2020), highways (Li et al., 2015), airport runways (Li, Q. et al., 2018) and car parks (Azam et al., 2018). However, there has been increased concerns over the negative environmental effects (e.g., global greenhouse gas emissions) of the petroleum industry (Ali et al., 2019), which increases the demand for eco-friendly binders for pavement construction. Furthermore, petroleum-derived bitumen makes up about 5% of the road construction but accounts for approximately half of the cost of materials used to produce an asphalt mixture. Hence, there would be substantial benefits to the environment and economic cost if an eco-friendly and renewable bitumen is developed and implemented.

There are a couple of waste materials that have been effectively utilised in asphalt pavement, which substantially enhances engineering performances of asphalt mixture and reduces the environmental harm. Marble wastes are commonly used as fine aggregate and filler in asphalt mixture. Kofteci et al. (2018) evaluated the usability of marble waste in asphalt mixture. They reported that marble waste can effectively increase the Marshall stability and reduce the flow value of the asphalt mixture. In addition, they found that the involvement of marble waste promoted the indirect tensile strength and resistance to moisture damage and abrasion wear. Recycled paper mill sludge is also documented to partially substitute the mineral filler to improve the asphalt performance. Chew et al. (2020) investigated the mechanical properties of asphalt mixture modified by recycled paper (dry process) mill sludge from the microscopic perspective. They found that recycled paper mill sludge can effectively promote the mechanical properties (e.g., resilient modulus, Leutner shear and dynamic creep) of asphalt mixture. The fundamental reason of the above observations is that recycled paper mill sludge forms a type of lapped antenna, which promotes mechanical performance and binder-aggregate adhesion bonding of asphalt mixture. In addition to marble waste and paper sludge waste, solid waste of restaurant (de Azevedo et al., 2020) can also be utilised in asphalt mixture to enhance its performance. Jalkh et al. (2018) evaluated the impacts of oxidation on physicochemical and rheological performances of waste cooking oil and coffee grounds oil for potential use as rejuvenators of reclaimed bitumen from aged and damaged asphalt pavements. They concluded that the waste cooking oil and coffee grounds oil can be used as sustainable rejuvenators for reclaimed bitumen. In addition, the viscosity of the above oils can be customised by oxidation process (e.g., temperature and duration) to work well with the reclaimed bitumen.

Meantime, in the UK there are approximately 8 million tonnes of waste plastics and municipal solid waste (MSW) needed to be processed each year. Due to the difficulties in their logistics, sorting and reuse, almost all of them are landfilled, sea-dumping or incinerated. With the development of waste management techniques and implementation of a circular economy, people gradually find that one of the promising options to recycle most of these wastes is to convert them physically or chemically into durable construction materials for transport infrastructures (Abo El-Naga and Ragab, 2019; Romeo et al., 2018). Fethiza Ali et al. (2020) studied the effect of waste plastic on thermal-oxidative ageing of the bitumen. They found that waste plastic modified bitumen presented higher resistance to thermal-oxidative ageing (i.e., short-term ageing and long-term ageing) compared to the control bitumen. Tauste-Martínez et al. (2021) conducted an assessment of the effect of recycled low-density polyethene (LDPE) on the long-term performance of the bitumen. The Atomic Force Microscopy (AFM) and Multiple Stress Creep and Recovery (MSCR) results proved that recycled LDPE can effectively improve the durability of the bitumen. Karmakar and

Kumar Roy (2021) investigated the influence of plastic waste on moisture damage of bituminous materials with the test methods of FTIR, AFM, modified Marshall immersion, and indirect tensile strength. The results indicated that plastic waste can be utilised as an effective moisture resistive modifier to fabricate a durable asphalt mix in the wearing course of asphalt pavement. Ramli et al. (2021) designed a modified asphalt mixture with waste plastic polypropylene to enhance its performance of horizontal deformation. The horizonal deformation measured from the test showed that waste plastic polypropylene strengthened the deformation properties of the mixture without any negative effect.

Regarding the MSW, more commonly known as trash or garbage, the main components are the product packaging, grass clippings, furniture, clothing, bottles, food scraps, newspapers, appliances, and paint, all of which comes from the residential homes, schools, hospitals, and businesses. In recent years, the possibility of recycling the above MSW in pavement engineering attracts increased attention and quickly leads to hot issues. Spreadbury et al. (2021) evaluated the field performances (i.e., resilient modulus and permanent deformation) of MSW incineration bottom ash as a base material of a road. They found that the resilient modulus and permanent deformation of this base layer were affected by the thickness, compaction effort, and moisture content of this layer. Based on these results, the authors proposed an optimal performance guideline regarding compact energy, thickness, and moisture control of the MSW incineration bottom ash. Yan et al. (2019) investigated properties (e.g., penetration, soft point, complex modulus, and creep stiffness) of asphalt mortar modified by MSW incineration fly ash. They concluded that: 1) MSW incineration fly ash slightly decreased the low-temperature performance of the mortar, and this negative effect could be ignored; 2) MSW incineration fly ash significantly enhanced the high-temperature properties of the mortar. Hence, compared with the traditional filler (e.g., limestone mineral), MSW incineration fly ash could be a better option for asphalt fabrication and implementation due to its advantages of waste management, energy conservation, and performance enhancement.

Pyrolysis, which is a thermochemical decomposition of organic material that occurs at designed temperatures in the absence of oxygen, is employed as a method for waste disposal and energy recovery. There have been increasing research activities and industrial developments of pyrolysis of unrecycled waste plastics by using different types of reactors to produce pyrolysis bio-oil. Hariadi et al. (2021) quantified the effects of bio-oils pyrolysed from waste LDPE in three different reactor outlets. They found that the quality and quantity of the bio-oil were essentially affected by pyrolysis duration and temperature. The optimal pyrolysis temperature for the selected waste LDPE was 250 °C. Baena-González et al. (2020) reported the recovery of bitumen, olefinic solvents, aromatic compounds, and recycled polystyrene from pyrolysis oil from waste plastics. Their results showed that waste-derived bitumen (i.e., bitumen obtained from the pyrolysis oil) had a high potential to be a modifier for traditional petroleum bitumen by reducing its viscosity and soft point because the waste-derived bitumen contained 55.05 wt% of aromatics and 33.41 wt% of saturates. Moreover, due to its great application potential showed by the altered physical, chemical, mechanical and economical properties, pyrolysis oil derived from the MSW also has been regarded as a promising candidate to enhance the bitumen’s engineering performance. Yang et al. (2018) presented an investigation on ageing and rheological properties of bio-oil from intermediate pyrolysis of the organic part of the MSW. They observed an obvious decrease in dynamic viscosity of the bio-oil after accelerated ageing, which was due to the decomposition of the semisolid organic agglomerates in the MSW during the intermediate pyrolysis. The reduced dynamic viscosity of the bio-oil (after ageing) indicated that it can be selected as a substitute for the light component in the petroleum bitumen for road construction and maintenance.

It is observed that the bitumen in the asphalt mixture, when exposed to cracking damage caused by thermal, vehicle and other loadings, can heal the cracks and restore partially or fully the original set of their physical, chemical, and mechanical properties. The healing can defer the initiation and evolution of the material deteriorations (e.g., fatigue crack) and eventually result in an extension of the service life of the asphalt pavement. A road performance prediction without accurately modelling the healing process in the bitumen will lead to a significant systematic error which could cause misleading conclusions or completely wrong decisions in material selections, road structural design or techno-economic analyses. Xu et al. (2021) employed three types of rejuvenators to quantify the healing effect on performance recoveries of the damaged bitumen. They found that the selected rejuvenators encapsulated in calcium alginate can effectively restore the physical, chemical, and rheological performances of the damaged bitumen. The fundamental reason for this phenomenon is that the released rejuvenator wets the cracks, diffuses into the damaged bitumen, and heals the bitumen eventually. Grossegger (2021) investigated the occurrence of an optimal healing time in the asphalt. He concluded that 1) healing potential was related to healing method, healing duration, and crack type; 2) optimal healing time only can be determined in a range due to the measurement uncertainty introduced by the heterogeneity of the asphalt. Chen et al. (2002) proposed a method for surface energy measurement of the asphalt, based on which they predicted the fatigue and healing performances of the asphalt. They concluded that healing performances of asphalt pavement were strongly correlated with the fundamental material properties such as relaxation modulus and advancing surface energy of the bitumen. The well-designed asphalt mixtures with the bitumen of better healing properties (e.g., modulus and surface energy) have been proved to provide better healing performance during the service life of the asphalt pavement. Thus, an increasing demand is substantially raised for a comprehensive understanding and accurate prediction of the healing performance of the bitumen, particularly for that novel bitumen modified by the waste plastics and MSW pyrolysis liquid, where their healing potential is completely unknown.

The healing of the bitumen is commonly quantified by the healing index, which is normally defined by a per cent ratio of the recovery of a material parameter after a rest interval to the one before the rest interval. The healing index was a highly empirical-based parameter because there were no agreed conclusions on which material parameter should be used in defining the healing index. It was regarded as an empirical indicator of the rate and capability at which healing proceeded (Little et al., 1999). Miglietta et al. (2021) assessed two types of healing index with the magnitude of stiffness and fatigue endurance gain, respectively. They emphasised the importance of considering the coupled effect between rest time and healing temperature to get a reliable evaluation of healing performance. Gallego et al. (2021) employed a thermomechanical method to evaluate the healing performance of the asphalt mixture. They defined the healing index by a ratio of initial indirect tensile strength of the undamaged asphalt to finial indirect tensile strength of the healed asphalt, based on which the authors optimised the heat and re-compaction energy for the assisted healing of the asphalt. Yamaç et al. (2021) characterised the healing of asphalt mastic by the capsule containing waste oil, during which the healing index was defined by a ratio of maximum breaking load after the healing process to the one prior to the healing process. They concluded that the amount of capsule added into the asphalt and healing temperature were two critical factors affecting the healing performance of the asphalt. Li et al. (2020) proposed that the healing can be directly defined by crack length as the healing is a process of crack reduction, resulting in the recovery of the other material properties. They concluded that the crack length-based healing index was more fundamental and reliable to characterise healing properties of the bitumen, because it can eliminate the effects of nonlinear viscoelasticity, frictional heat loss, and thixotropy.

This study aims to characterise the healing performance of two kinds of waste-derived bitumen, including bio-oil modified bitumen using MSW pyrolysis liquid and plastic modified bitumen by LDPE. The theoretical models of the healing of the bitumen based on the DSR tests were firstly presented, followed by the DSR fatigue-healing tests and surface energy experiments. This consisted of the fabrications of bio-oil modified bitumen and LDPE modified bitumen, preparation of testing specimens, and cracking/healing and contact angle tests of the virgin and PAV-aged waste-derived bitumen. Then, characterisations of the healing performances of the virgin and PAV-aged control and waste-derived bitumen were analysed in detail, based on which the effects of the bio-oil and waste plastics on the healing rate and healing potential were quantified. The last section summarised the main contributions of this paper.

2. Theoretical Models for Healing of Bitumen

2.1 Healing Characterisation Based on Crack Length

Dynamic shear rheometer (DSR) is commonly utilised to characterise viscoelastic properties of the bitumen; additionally, it can be effectively used to evaluate and predict fatigue crack performance of the bitumen by applying a rotational fatigue load. Zhang and Gao (2019) proposed and successfully verified a damage mechanics-based crack growth model, which was employed to calculate the crack length of the bitumen under a rotational shear fatigue load based on the DSR test. The crack length in a strain-controlled DSR time sweep fatigue test can be shown by **Equation (1)**:

$$CL = \left\{ 1 - \left[\frac{\left|G_N^*\right| / \sin\left(\delta_N\right)}{\left|G_0^*\right| / \sin\left(\delta_0\right)} \right]^{1/4} \right\} r_0 \tag{1}$$

Where, CL is crack length of the bitumen at the N^{th} load cycle; $|G^*_0|$ and δ_0 are dynamic shear modulus and phase angle of the bitumen in the undamaged state, respectively; $|G^*_N|$ and δ_N are dynamic shear modulus and phase angle of the bitumen at the N^{th} load cycle in the damaged state, respectively; r_0 is original radius of the bitumen sample (i.e., 4mm in this study).

Healing is a process of crack reduction; hence, it is reasonable to define the healing index using the crack length, a justification of which can be found in the authors' previous paper (Li et al., 2021). Based on **Equation (1)**, a new healing index defined in **Equation (2)** has been successfully utilised to characterise the healing property of the bitumen (Li et al., 2020). This newly defined parameter excludes the effects of viscoelasticity and thixotropy during the rest period of rotational fatigue loads.

$$\%HI = \frac{CL_D - CL_H}{CL_D} \times 100\% \tag{2}$$

Where, $\%HI$ is healing index; CL_D is crack length at the last load cycle prior to the rest duration; CL_H is crack length after the healing rest time. **Figure 1** shows an example of a typical crack length before and after the healing time in a strain-controlled time sweep fatigue-healing test.

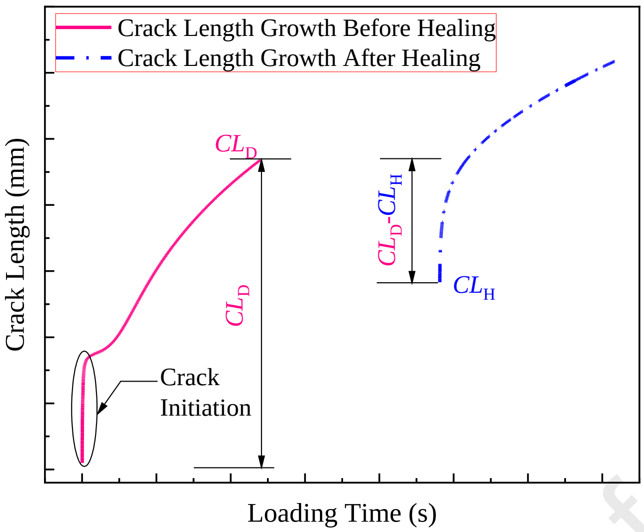

Figure 1. Crack length growth in a strain-controlled time sweep fatigue-healing test

Lytton (2000) proposed that healing of the bitumen depended on its surface energy (Γ_h) calculated from advancing contact angles, in which the non-polar component (Γ_h^{LW}) sourced from Lifshitz-Van der Waals force and the polar component (Γ_h^{AB}) resulted from Lewis acid-base force. There existed two healing mechanisms including short-term healing and long-term healing in its whole process. He noted that the short-term healing rate ($\dot{h}_1$) depended primarily on $1/\Gamma_h^{LW}$, and the long-term healing rate ($\dot{h}_2$) depended mainly on Γ_h^{AB}. Based on the above conclusions, Lytton proposed two useful models shown in **Equation (3)** to evaluate $\dot{h}_1$ and $\dot{h}_2$.

$$\begin{cases} \dot{h}_1 = a_1 \left(\dfrac{1}{\Gamma_h^{LW} G_1} \right)^{b_1 m'} \\ -\log\left(\dot{h}_2\right) = a_2 \left[-\log\left(\dfrac{\Gamma_h^{AB}}{G_1} \right) \right]^{b_2 m'} \end{cases} \quad (3)$$

Where, a_1 and b_1 are fitting parameters for the short-term healing rate $\dot{h}_1$; a_2 and b_2 are fitting parameters for the long-term healing rate $\dot{h}_2$.

Many researchers (Cheng, D. et al., 2002; Luo and Lytton, 2016; Si et al., 2002) also noted that the two healing rates occur simultaneously, and the real healing mechanism is the result of their coactions. They furtherly recommended that actual healing rate $d(HI)/dt$ could be expressed by the Ramberg-Osgood model (Ramberg and Osgood, 1943) shown in **Equation (4)**:

$$\frac{d(HI)}{dt} = \dot{h}_2 + \frac{\dot{h}_1 - \dot{h}_2}{1 + \dfrac{\dot{h}_1 - \dot{h}_2}{h_\beta} (\Delta t)_h} \quad (4)$$

Where, $(\Delta t)_h$ is the rest period between load applications; and h_β is the factor that varies between 0 and 1 and represents the healing potential, which is the maximum percentage of bitumen healing that can be achieved. The value of h_β is also empirically found to be related with $\Gamma_h^{AB}/\Gamma_h^{LW}$, and can be determined by **Equation (5)** (Luo, 2012):

$$h_\beta = \alpha_\beta \left(\frac{\Gamma_h^{AB}}{\Gamma_h^{LW} G_1^2} \right)^{b_\beta m'} \tag{5}$$

Where, α_β and b_β are fitting parameters for the healing potential, h_β.

It should be noted that although the Ramberg-Osgood model was originally used to describe the nonlinear relationship between stress and strain in materials near their yield points, there is no constitutive interpretation herein, and it is believed that the Rambert-Osgood model was simply used to characterise healing rate and healing potential of the selected bitumen.

2.2 Calculation of Surface Energy of Bitumen

Surface energy (more correctly, surface free energy) is one of the critical parameters affecting the healing performance of the bitumen as shown in **Equation (3)**. However, it is not feasible to directly measure the bitumen surface energy, and one proven method to estimate it is to measure contact angles between the bitumen and selected probe liquids with the sessile drop method and then calculate the surface energy. In this approach, the drops of a small number of probe liquids are deposited on the surface of the bitumen sample and the contact angles between the liquids and the bitumen surface are captured and measured, based on which the surface energy can be calculated using the Young-Dupre equation (van Oss, 2002):

$$(1+\cos\theta)\Gamma_{liquid} = 2\left[\sqrt{\Gamma^{LW}\Gamma_{liquid}^{LW}} + \sqrt{\Gamma^{+}\Gamma_{liquid}^{-}} + \sqrt{\Gamma^{-}\Gamma_{liquid}^{+}}\right] \tag{6}$$

Where, θ is contact angle between the bitumen and the probe liquid drop; Γ_{liquid}, Γ^{LW}, Γ_{liquid}^{LW}, Γ^{+}, Γ^{-}, Γ_{liquid}^{+}, and Γ_{liquid}^{-} are the surface energy of the probe liquid, the Lifshitz-van der Waals component of the bitumen, the Lifshitz-van der Waals component of the probe liquid, the Lewis acid component of the bitumen, the Lewis base component of the bitumen, the Lewis acid component of the probe liquid, and the Lewis base component of the probe liquid, respectively.

After obtaining the values of Γ^{LW}, Γ^{+}, and Γ^{-}, the total surface energy Γ of a material can be calculated by:

$$\Gamma = \Gamma^{LW} + 2\sqrt{\Gamma^{+}\Gamma^{-}} \tag{7}$$

3. Materials and Experimental Characterisation

3.1 Fabrication of Waste-derived Bitumen

Bitumen X70 was selected as a control and base bitumen to develop the waste-derived bitumen by mixing with waste materials (i.e., bio-oil or LDPE). Details of the production of the bio-oil can be found in the authors' previous publication (Yang et al., 2018). The concentrations of bio-oil and LDPE in the modified bitumen were 5wt. % and 6wt. %, respectively. The detailed characterisation of the control bitumen, bio-oil, and LDPE can be found in **Table 1**.

Table 1. Characterisations of X70, bio-oil, and LDPE

		Measure	Value
X70 [a]	Penetration @25°C	dmm	45-80
	Softening Point	°C	≥45

	Force Ductility @5°C	J/cm^2	>3
	Flash Point (Cleveland)	°C	>250
	Fraas Breaking Point	°C	≤-12
bio-oil [b]	Water Content	wt. %	25.4
	Solid Content	wt. %	19.3
	Higher Heating Value	MJ/kg	28.0
	Density	g/cm^3	0.972
LDPE [c]	Melting Point	°C	126
	Melting Heat	MJ/kg	0.141
	Density	g/cm^3	0.934
	Thermal Degradation Point	°C	220

[a] Data provided by the supplier. [b] Data referenced from the author's previous work (Yang et al., 2018). [c] Data obtained from DSC and density tests.

To fabricate 5wt. % bio-oil modified bitumen, bio-oil was firstly put into a clean beaker followed by adding a well-calculated mass of hot control bitumen. Then, a high shear mixer is used to mix them homogenously at a speed of 150RPM for 30min at 150°C under a nitrogen atmosphere. Regarding the 6wt. % LDPE modified bitumen, hot control bitumen was firstly put into another clean beaker followed by adding accurately calculated mass of the LDPE. Then, the high shear mixer is utilised to blend the LDPE at a speed of 900RPM for 90min at 180°C under a nitrogen atmosphere. The blending speed and time were selected to ensure the LDPE was completely melted and distributed within the hot bitumen.

Then, part of three types of unaged bitumen (i.e., unaged control bitumen, newly fabricated bio-oil modified bitumen and LDPE modified bitumen) were distributed into bitumen sample bottle retainers to conduct the rolling thin-film oven (RTFO) ageing test at 163°C for 85min, which is in consistent with the standard of AASHTO Designation T240-09 and ASTM Designation D2872-04. RTFO ageing test provides simulated short-term aged bitumen for engineering performance evaluations. The bitumen residue, from the RTFO ageing test, was then placed in stainless steel pans and aged at 100°C for 20 hours in a vessel pressurised with air to 2.10MPa (i.e., PAV ageing), which is in accordance with the standard of AASHTO Designation R28-09. PAV ageing test provides simulated long-term (7 to 10 years) aged bitumen for engineering performance evaluations, such as fatigue cracking and healing.

3.2 Preparations of DSR and Surface Energy Test Specimens

As mentioned in Section 2, the designed tests mainly include the DSR tests and surface energy tests. The major goal of the DSR tests is to accurately measure the dynamic moduli and phase angles of the undamaged, damaged, and healed bitumen. In terms of the surface energy tests, the main objective is to obtain the advancing contact angles between the bitumen and the probe liquids.

Before starting the tests, the bitumen samples stored in the containers were heated in the laboratory oven at 165°C for 30min to reduce the material viscosity. Then hot bitumen was carefully distributed into the silicon mould with a cavity of 4mm in diameter and 2 mm in depth. After 15 minutes, the DSR sample was carefully demoulded from the mould and installed and trimmed on the surfaces of the bottom and top plates to conduct the DSR tests shown in **Figure 2**. The LAS, frequency sweep and healing tests were conducted on the DSR using an 8 mm diameter parallel plate geometry and 2 mm gap setting, as shown in **Figure 2**. To make the bitumen samples contact well with the DSR plates and reduce the heterogeneity due to the fabrications of these samples, all bitumen samples were preheated to 80°C before the tests started. After the DSR tests, both plates were checked carefully to make sure the adhesions between them were still excellent. To examine the repeatability of the experiments, three replicates were tested at each condition and additional replicates were added when the repeatability COV of the target dynamic moduli and phase angles were greater than 5%, 10%, and 5%, respectively, which is in consistent with the standard of EN 14770: 2012.

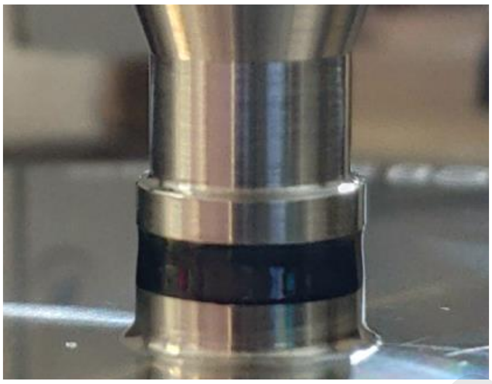

Figure 2. Trimmed configuration of an undamaged cylindrical bitumen sample

In this paper, surface energy for cracking and healing was measured by a versatile optical tensiometer, where the Attension Theta Flex tensiometer was used. Microscope slides with 76 × 26 ×1mm dimensions were cleaned with acetone and distilled water, and then dried by the laboratory oven at 60°C for 30min. After that, the slides were dipped into the melted bitumen for 10 seconds and then held out of the container for another 10 seconds to make the extra hot bitumen drop off the slide. To get a flat and smooth surface of the bitumen sample, the above process needs repeating at least 3 times, if necessary. Then the bitumen sample was cooled to ambient temperature in a desiccator with anhydrous calcium sulphate crystals for 24 hours. **Figure 3** presents the bitumen in the container and on the microscope slides, and the installation of microscope slide coated with the bitumen on the Attension Theta Flex tensiometer.

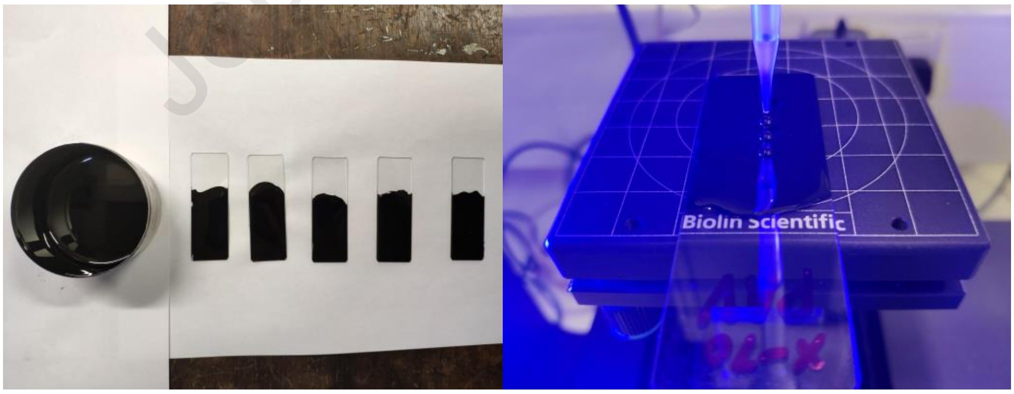

Figure 3. Bitumen in a container and on microscope slides, and contact angle test of the bitumen by Attension Theta Flex tensiometer

3.3 Experimental Characterisation of Bitumen

a) Linear amplitude test to determine dynamic shear modulus and phase angle

To calculate the crack length, healing index and other material parameters described in Section 2, the dynamic shear modulus and phase angle of the undamaged bitumen need to be calibrated firstly. **Figure 4** presents the dynamic shear moduli and phase angles from the linear amplitude sweep (LAS) test of the unaged control bitumen conducted at 10Hz and 20°C. In this LAS test, the start and end complex shear strain levels were selected as 0.01% and 100%, respectively.

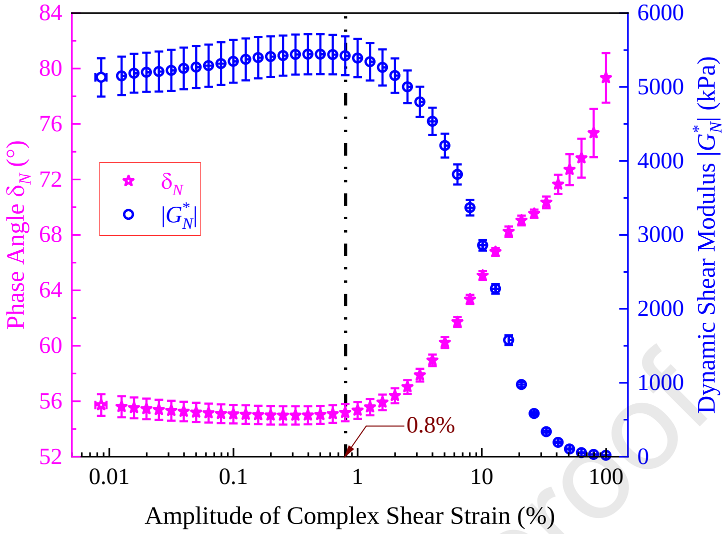

Figure 4. Curves of shear modulus and phase angle versus shear strain from the LAS test (10Hz and 20°C)

Figure 4 shows that, in the LAS test, both dynamic shear modulus and phase angle vary little or remain the same when the amplitude of the shear strain is low, e.g., less than 0.8%. This implies that 0.8% is a critical threshold strain level, below which the unaged control bitumen is undamaged at 10Hz and 20°C. When the amplitude of shear strain is over 0.8%, dynamic shear modulus decreases and phase angle increases dramatically, which indicates that cracks appear in the control bitumen. Theoretically, the $|G^*_0|$ and δ_0 can be obtained by averaging the $|G^*_N|$ and δ_N, respectively, in a strain level between 0.01% and 0.8%, within which the bitumen is in an undamaged condition. But practically, the sample-to-sample variation of the bitumen needs to be considered, because the $|G^*_0|$ and δ_0 measured herein will be used to characterise the healing performance of the bitumen by integrating the following results of frequency sweep tests and fatigue-healing tests. The DSR bitumen sample after the LAS tests which is already damaged cannot be reused to investigate the healing property of the unaged control bitumen. Therefore, a new DSR bitumen sample was employed to measure the $|G^*_0|$ and δ_0 with the LAS start and end strain levels ranging from 0.1% and 0.5% (both less than 0.8%). $|G^*_0|$ and δ_0 of the PAV-aged control bitumen, unaged and PAV-aged bio-oil modified bitumen, and unaged and PAV-aged LDPE modified bitumen were obtained using the same method.

b) Frequency sweep tests and fatigue-healing tests to determine viscoelasticity and healing properties

Frequency sweep tests using the DSR were conducted at temperatures of 10°C, 20°C, 30°C, 40°C, 50°C, 60°C, and 70°C, and in a frequency range from 0.1Hz to 25Hz. The dynamic shear moduli and phase angles were obtained at the above temperatures and frequencies, and the master curves of the dynamic shear modulus and phase angle can be accurately constructed (Li, L. et al., 2018a). Then, by using the interconversion equations for linear viscoelastic material (Park and Schapery, 1999), shear relaxation modulus can be accurately calculated, where the model parameter G_1 and m' can be determined.

A fatigue-healing test was employed to characterise the fatigue-healing performance of the bitumen. It consisted of a strain-controlled time sweep fatigue test (5%,10Hz, 20°C) plus a rest duration and followed by another strain-controlled time sweep fatigue test (5%, 10Hz, 20°C). The first part of the fatigue test was utilised to generate cracks in the bitumen and its duration was 20 min. Different rest durations were used including 10s, 0.5min, 1min, 5min, 10min, and 20min (For the unaged control bitumen, 5s, 2min, and 40min were also used for the rest durations). The second part of the fatigue test was used to obtain the cracking performance after the healing rest was applied. The testing temperature was selected as 20°C. The schematic plot of the loading sequences employed in the frequency sweep and the fatigue-healing tests can be found in **Figure 5**.

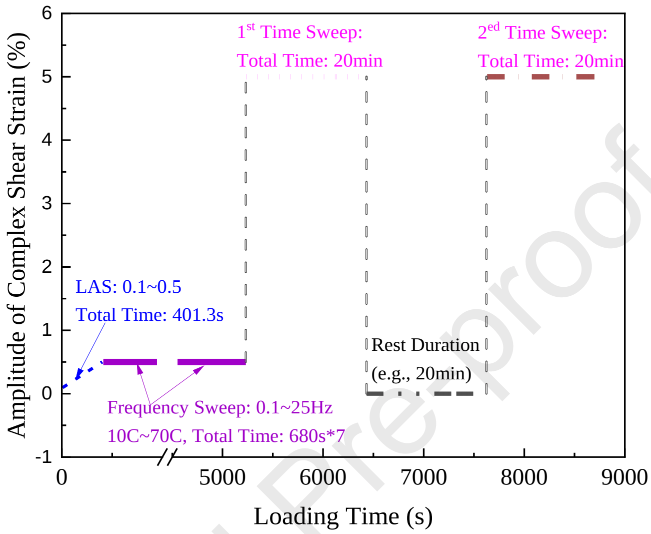

Figure 5. Loading sequences of frequency sweep testing and fatigue-healing tests

c) Contact angle tests to determine surface energy of the bitumen

The sessile drop method (using Attension Theta Flex optical tensiometer) was adopted to measure contact angles of bitumen surfaces with different probe liquids. The contact angle results were then used to calculate surface energy components using **Equation (6)**. There are two types of dynamic contact angles (i.e., advancing, and receding contact angles) that can be measured by the mode of automatic dynamic contact angle built-in the tensiometer. Lytton et al. (2005) stated that the surface energy calculated from the advancing contact angle contributed to crack surface wetting and was related to the healing process; the receding contact angle was associated with the de-wetting thus linked to the crack opening process. For the healing purpose, this study used the advancing contact angles to determine the surface energies of the materials.

Automatic dynamic contact angle experiments are conducted to obtain the advancing contact angles between the bitumen samples and five preselected probe liquids (i.e., Ethylene glycol, Water, Formamide, Glycerol, and Diiodomethane) with known surface energy components. For each liquid, three clean and dry microscope slides coated with the bitumen were utilised to reduce the variation of contact angle measurements. An automatic dispenser with a gauge needle and an adapter containing different probe liquids was utilised herein. The adapter was mounted to the dispenser and the needle was connected to the adapter. When the sample for the experiment was in place of the tensiometer, key configurations of built-in software (i.e., OneAttension software) were set up immediately, including the creation of the user level, selection of the experimental mode (i.e., automatic dynamic contact angle), completion of the recipe (critical parameters of the experiment can be transmitted to the computer). Once all the above controls have been set up, the experiment can be started and continued until its completion.

There are two options (i.e., tilting cradle and thin needle method) available to obtain the dynamic contact angles with the tensiometer. The tilting cradle method was used herein to measure the advancing contact angles, which mainly includes the following steps: 1) A droplet of the probe liquid is placed on the bitumen sample surface and tilting starts; 2) Once the droplet starts moving on the bitumen sample surface, the advancing contact angle has been reached, and the roll-off angle of the surface is also detected; 3) After setting the baseline to the bitumen surface, contact angles can be imaged directly by the image recording system of the tensiometer. The built-in software of the test equipment automatically recognises and calculates the contact angles between the selected probe liquid and the bitumen.

Figure 6 summarises the experimental methodology employed in this section.

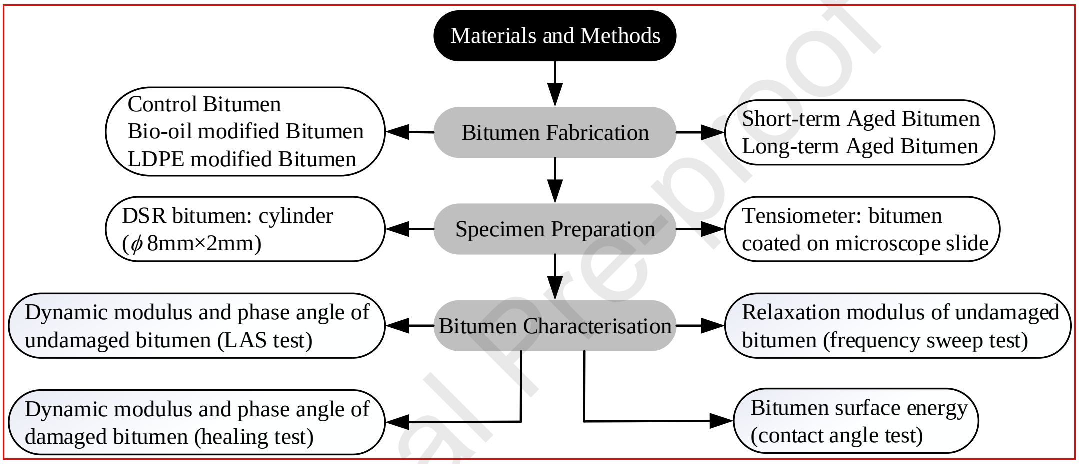

Figure 6. Flow chart of experimental methodology on healing characterisation

4. Results and Discussion

4.1 Enhancements of Healing Performances of Unaged Bitumen Modified by Bio-oil or LDPE

Figure 7 shows that healing indices of all three unaged bitumen increase with healing time, which is consistent with the Ramberg-Osgood model shown in **Equation (4)**. Both the bio-oil and LDPE can strengthen the healing performance of the unaged bitumen. Compared with the LDPE, the enhancements due to the inclusion of bio-oil to the healing rate and healing potential are more pronounced. The fundamental mechanism for this observation can be explained that, compared to the bitumen molecules, the bio-oil is a kind of less viscous fluid pyrolysed from municipal solid waste which can effectively soften the bitumen. Hence, compared with the unaged control bitumen, the molecules of the bio-oil modified bitumen are more diffusible, thus the cracks in the bio-oil modified bitumen are more healable. The above explanation is well consistent with the results obtained by Sun and Zhou (2018).

The LDPE slightly enhances, not weakens, the healing performance of the unaged control bitumen, which has been verified by the authors' previous paper (Li et al., 2021) using the fundamental properties (e.g., surface energy) of the material to predict its healing performance. This may be due to the diffusion of those short polymer chains in the LDPE to the bitumen molecules to stimulate the healing of the material. Previous studies (Ahmedzade et al., 2013; Farahani et al., 2017) show that the LDPE modified bitumen undergoes no observable changes in functional groups relative to the control bitumen, which confirms that this kind of modification of the control bitumen with LDPE is overwhelmed by physical process. The short chains of the LDPE melted in the unaged bitumen tends to aggregate and form a kind of network structure within a continuous polymer phase, which will potentially increase the healing performance of the unaged bitumen. Therefore, the new formation of the micro-network structure physically contributed by the melted LDPE is expected to be the critical reason that slightly increase the healing performance of the unaged bitumen. More details of fundamental understanding of LDPE's enhancements on bitumen healing will be presented in the last part of this section.

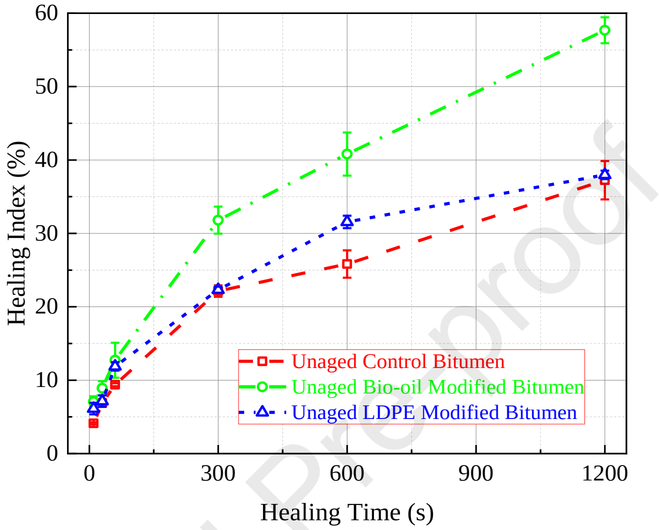

Figure 7. Healing curves (healing index versus healing time) of the control bitumen, bio-oil modified bitumen, and LDPE modified bitumen in unaged condition

To further quantify the enhancements of the bio-oil and the LDPE to the healing performance of the unaged bitumen, healing speed and healing potential models shown in **Equations (3)~(5)** are determined and interpreted in detail.

According to the Ramberg-Osgood model shown in **Equation (4)**, the healing index can alternatively be expressed by **Equation (8)**:

$$HI = \dot{h}_2 \left(\Delta t\right)_h + h_\beta \ln\left[1 + \frac{\dot{h}_1 - \dot{h}_2}{h_\beta}\left(\Delta t\right)_h\right] \quad (8)$$

Substituting the measured healing index shown in **Figure 7**, $\dot{h}_1$, $\dot{h}_2$, and h_β can be back-calculated and presented in **Figure 8**.

Figure 8 shows the results of the short-term healing rate, long-term healing rate and healing potential of the three kinds of unaged bitumen. It is found that the long-term healing rate is substantially smaller than the short-term healing rate, which means healing occurs mainly in a short term. This observation is consistent with the one found in the existing research (Cheng, D. et al., 2002). The short-term healing rate and healing potential dominate most of the healing behaviours of the unaged bitumen. **Figure 8** also indicates that the unaged bio-oil modified bitumen heals the most (having the highest h_β of the three), followed by the unaged LDPE modified bitumen, and the unaged control bitumen heals the least. Both the bio-oil and LDPE can increase the short-term healing rate of the unaged bitumen, and the unaged bio-oil modified bitumen heals faster than the unaged LDPE modified bitumen in a short term.

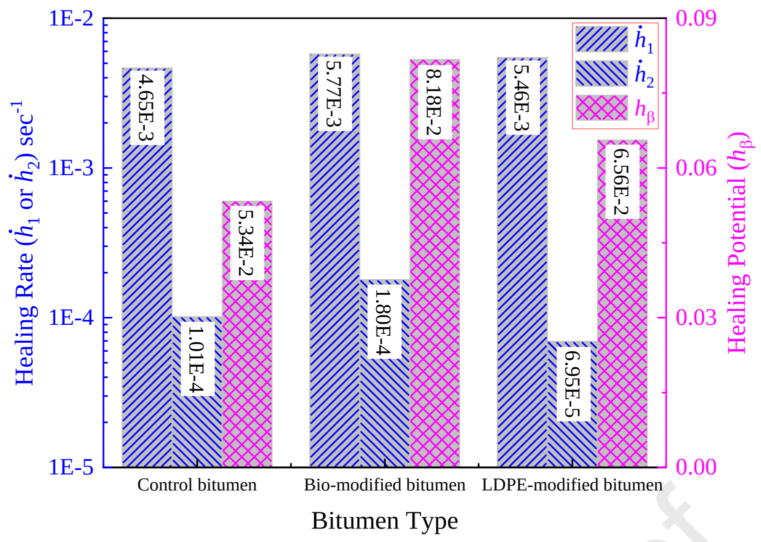

Figure 8. Short-term healing rate ($\dot{h}_1$), long-term healing rate ($\dot{h}_2$) and healing potential (h_β) of different kinds of unaged bitumen

The short-term healing rate, long-term healing rate and healing potential can also be predicted using **Equation (3)** and **Equation (5)**. The parameters of m', G_1 shown in these models are presented in **Table 2**, which are calculated from the testing results of the frequency sweep test (10Hz, 10~70°C) and LAS test (0.1%~0.5%, 20°C) of the unaged bitumen. The calculation steps of determining m' and G_1 are as follows: 1) Construction of the master curve of shear dynamic modulus and phase angle of the unaged bitumen at the reference temperature of 20°C by the time-temperature superposition principle (e.g., WLF equation); 2) Calculation of Prony model parameters of shear relaxation modulus by the collocation method (Park and Schapery, 1999) and the least squared regression minimization (Li, L. et al., 2018b); 3) Interconversion between Prony model parameters and power model parameters of shear relaxation modulus (i.e., $G(t) = G_1 t^{-m'}$).

Table 2. Slope of double logarithmic relaxation modulus curve m', initial shear relaxation modulus G_1, Non-polar component Γ_h^{LW}, polar component Γ_h^{AB}, and total Γ_h advancing surface energies of the unaged bitumen

	m'	G_1 (kPa)	Γ_h (mJ/m^2)	Γ_h^{LW} (mJ/m^2)	Γ_h^{AB} (mJ/m^2)
Control bitumen	0.7978	2398.8	14.36	13.95	0.41
Bio-modified bitumen	0.8290	480.2	12.50	12.39	0.11
LDPE-modified bitumen	0.7154	3680.2	27.16	27.09	0.07

The advancing contact angles between the unaged bitumen and the selected probe liquids are measured by the tensiometer to calculate the advancing surface energy contributing to the wetting and interdiffusion processes of the bitumen healing. **Table 2** also gives the experimental results of the non-polar component, polar component, and total advancing surface energies of the unaged bitumen.

Substituting the fundamental material constants (m', G_1 and Γ_h) to **Equations (3)** and **(5)**, constants of a_i and b_i (i=1, 2, and β) can be calculated with the aid of the Solver function in Microsoft Excel. More importantly, with all the material constants known, **Equations (3)~(5)** can be utilised to predict the healing index. **Figure 9** shows the measured and predicted healing indices of the three bitumen samples in the unaged condition. It can be found that

Equations (3)~(5) can be effectively used to predict the healing index of the unaged bitumen. This means, the healing curve of bitumen can be efficiently obtained once the material properties (including relaxation modulus and surface energy) are known, which can substantially reduce the experimental effort of the healing tests for the bitumen.

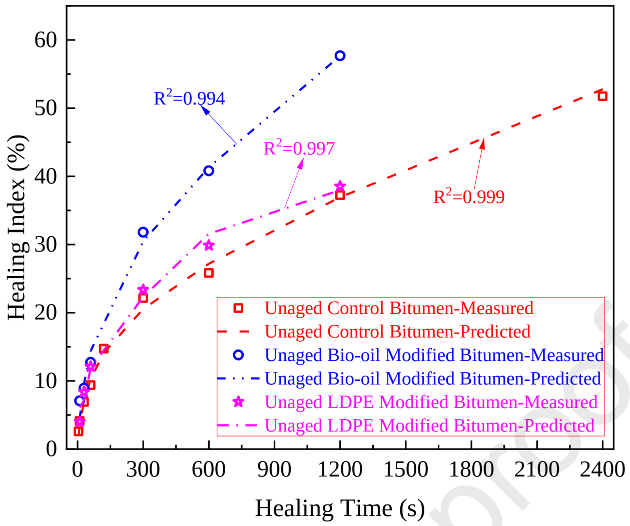

Figure 9. Comparisons between the predicted healing index using material properties (relaxation modulus and surface energy) and the measured results from healing tests for different bitumen materials in unaged condition.

4.2 Characterisations of Healing Performances of PAV-aged Control Bitumen, Bio-oil Modified Bitumen, and LDPE Modified Bitumen

Figure 10 shows that healing indices of the PAV-aged bitumen increase with the healing time, which agrees well with the Ramberg-Osgood model shown in **Equation (4)**. The PAV-aged bio-oil modified bitumen does not show better healing performance than that of the PAV-aged control bitumen, the reason of which is that most of the bio-oil has evaporated during the ageing process. Additionally, **Figures 7** and **10** show that the ageing process substantially reduces the healing performances of the bio-oil modified bitumen and control bitumen. The LDPE productively enhances the healing performance of the PAV-aged bitumen, one of the key reasons of which is that the LDPE substantially reduces the ageing rate of the bitumen (Nouali et al., 2020). Compared with the unaged LDPE modified bitumen, it is worthy to notice that the ageing process does not effectively reduce the healing ability of the LDPE modified bitumen. This observation can be interpreted as follows: 1) the molecular chain of the LDPE in the control bitumen is very stable, and the ageing process does not effectively decrease its activity and polarity (Nouali et al., 2020); 2) the PAV-aged control bitumen itself has a relatively low healing ability, which implies that any changes to its microstructure can potentially enhance its healing capability; and 3) the dispersed LDPE molecular chain (observed by SEM in a previous study (García-Morales et al., 2004)) can be swollen by the bitumen light components (i.e., saturate and aromatic), and the deformed LDPE molecular chains due to rotational shear loads will recover their original shape during the healing time. This significantly increases the motions of the light components of the PAV-aged bitumen and then enhances the healing performance of the PAV-aged bitumen. More details of the fundamental understanding of LDPE's enhancements on bitumen healing will be presented in the last part of this section.

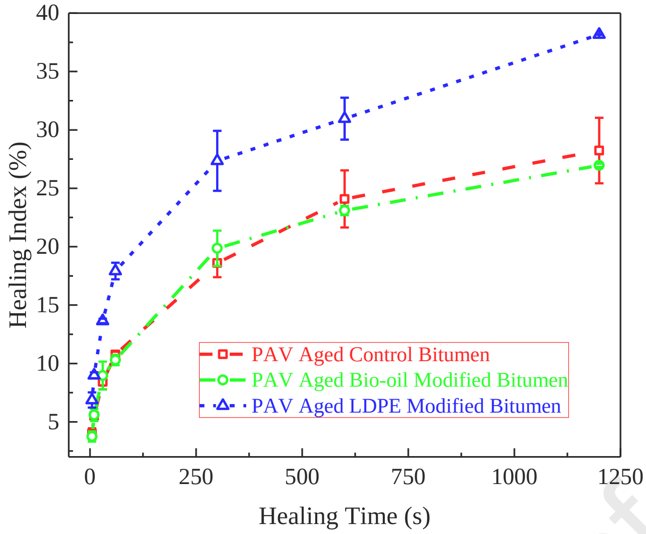

Figure 10. Healing curves of PAV-aged control bitumen, bio-oil modified bitumen, and LDPE modified bitumen

Substituting the measured healing indices shown in **Figure 10** into **Equation (8)**, the short-term healing rate $\dot{h}_1$, long-term healing rate $\dot{h}_2$, and healing potential h_β can be back-calculated and the results are presented in **Figure 11**.

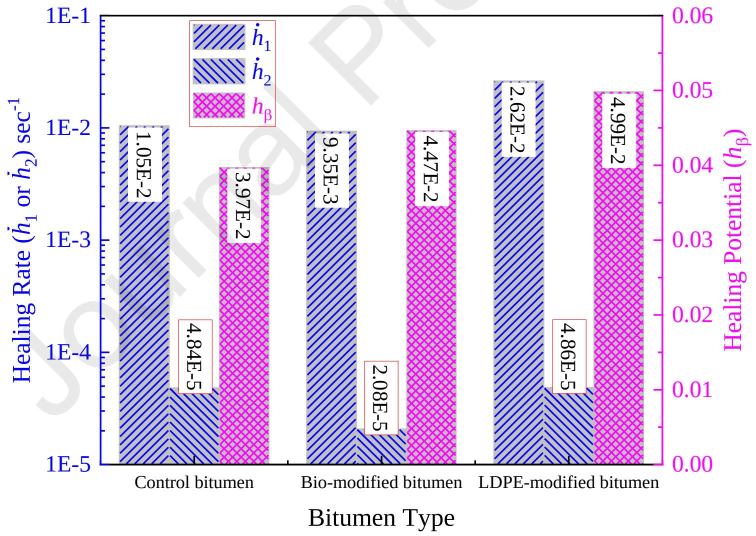

Figure 11. Short-term healing rate ($\dot{h}_1$), long-term healing rate ($\dot{h}_2$) and healing potential (h_β) of the PAV-aged bitumen

Figure 11 shows that the long-term healing rate is significantly smaller than the short-term healing rate, which indicates healing occurs mainly in a short term. The short-term healing rate and healing potential dominate the healing behaviours of the PAV-aged bitumen. Since most of the bio-oil has evaporated during the ageing process, differences in the short-term healing rate and healing potential between the aged control bitumen and the aged bio-oil modified bitumen can be neglected. **Figure 11** also shows that the PAV-aged LDPE modified bitumen heals the most (having the highest h_β), followed by the PAV-aged bio-oil bitumen and control bitumen. The PAV-aged LDPE modified bitumen heals faster (having the highest $\dot{h}_1$) than the PAV-aged control bitumen and the PAV-aged bio-oil modified bitumen.

As done in Section 4.1, healing speed and healing potential models shown in **Equations (3)~(5)** are reutilised to verify the healing performances shown in **Figure 10**. **Table 3** presents the values of m', G_1, Γ_h^{LW}, Γ_h^{AB}, and Γ_h of the control bitumen, bio-oil modified bitumen, and LDPE modified bitumen after the PAV ageing.

Table 3. Slope of double logarithmic relaxation modulus curve m', initial shear relaxation modulus G_1, Non-polar component Γ_h^{LW}, polar component Γ_h^{AB}, and total Γ_h advancing surface energies of the PAV-aged bitumen

	m'	G_1 (kPa)	Γ_h (mJ/m^2)	Γ_h^{LW} (mJ/m^2)	Γ_h^{AB} (mJ/m^2)
Control bitumen	0.7064	12251.3	17.62	17.25	0.36
Bio-modified bitumen	0.6598	12198.3	17.58	17.50	0.08
LDPE-modified bitumen	0.6399	21656.6	37.16	37.10	0.06

Substituting the fundamental material constants mentioned above to **Equations (3)** and **(5)**, fitting constants of a_i and b_i (i=1, 2, and β) can be back-calculated with the aid of the Solver function in Microsoft Excel. After that, the healing index of the PAV-aged bitumen can be predicted by **Equations (3)~(5)**. **Figure 12** presents the measured and predicted healing indices of the PAV-aged bitumen. It can be concluded that **Equations (3)~(5)** can be effectively used to predict the healing index of the PAV-aged bitumen once the material properties (e.g., relaxation modulus and surface energy) are measured, which are expected to substantially reduce the experimental effort of the healing tests of the PAV-aged bitumen.

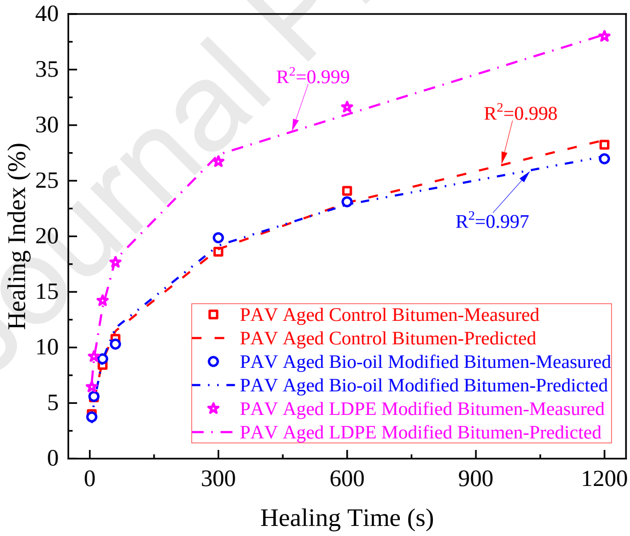

Figure 12. Comparisons between the predicted healing index using material properties (relaxation modulus and surface energy by **Equations (3)~(5)**) and the measured ones using healing tests for different PAV-aged bitumen materials

4.3 Fundamental Understanding of LDPE's Enhancements on Healing Performance of Bitumen

This section aims to provide a theoretical explanation of the LDPE's enhancements on bitumen's healing performance. **Figure 13** shows the healing process of cracked surfaces in a bitumen sample, which includes four steps, namely, surface rearrangement and approach, wetting, diffusion, and randomisation. Note the four healing steps exist simultaneously for the molecular chains of the bitumen (Bommavaram et al., 2009; Little et al., 2015). A well-known theory of polymer healing was first ingeniously delivered by Wool and O'Connor (Wool and O'connor, 1981). They employed two functions (i.e., wetting distribution function $\varphi(t)$ and intrinsic healing function $R_h(t)$) to obtain the healing index %*HI* (originally called recovery ratio) of mechanical properties of the polymer.

$$HI\%(t)=\int_{-\infty}^{t}R_h(t-\tau)\frac{\partial\varphi(\tau)}{\partial\tau}d\tau \tag{9}$$

Where, t is present time; τ is time history at which wetting distribution function and intrinsic healing function are obtained. The polymer healing theories shown in **Equation (9)** are demonstrated to be applicable for modelling bitumen healing as bituminous components and behaviours are comparable to a complex polymer (Bhasin et al., 2011).

Crack wetting of the bitumen depicts the contact and cohesion of two approached crack surfaces driven by the surface energy and bonding strength, leading to a definition of healing potential. Hence, the crack wetting happens at the interfaces, briefly depicted as wetting nucleation pools (Wool and O'connor, 1981), of the cracked surfaces without inclusions of bitumen chains' interdiffusion. Intrinsic healing of the bitumen describes the rate at which a wetted crack interface recovers the mechanical performance of the intact material, leading to a healing rate driven by Brownian motions of the bitumen molecules. The intrinsic healing develops on account of the motions of bitumen chains, which are significantly affected by the interdiffusions of neighbouring chains of the bitumen.

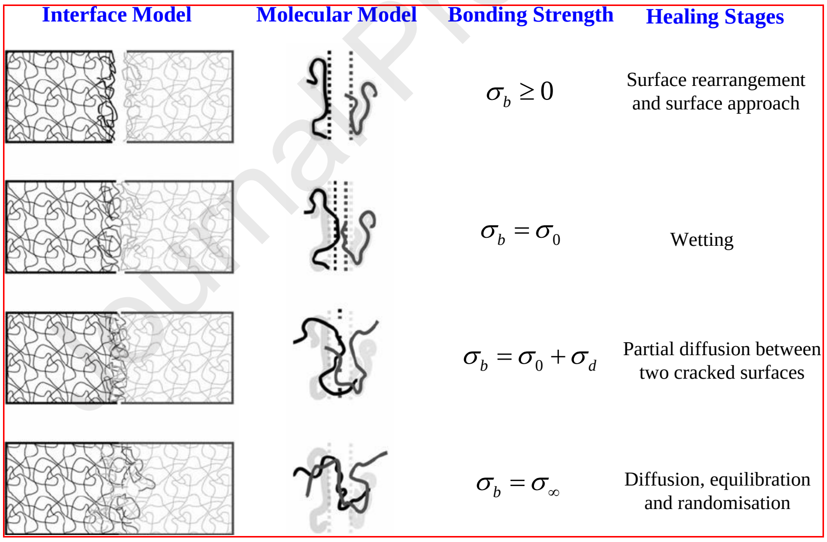

Figure 13. Schematic diagram of bitumen healing processes (σ_b is the bonding stress which derives the healing of the cracked surfaces; σ_0 is the recovered bonding strength due to wetting; σ_d is the recovered bonding strength due to interdiffusions of material chains between two cracked surfaces; and σ_∞ is the original bonding strength of the intact bitumen)

The LDPE modified bitumen shows better deformation recovery property than the control bitumen because 1) the inclusion of LDPE makes the bitumen much stiffer than the control one, as indicated by the increased modulus (G_1) in **Table 2** for the unaged bitumen and **Table 4** for the aged bitumen; 2) the elongated LDPE molecular chains in the bitumen recovers to their original shape when the external load is removed, which enhances the ability of the bitumen to restore to its original state (García-Morales et al., 2004); and 3) the surface energies (**Tables 2** and **3**) of the LDPE modified bitumen under unaged and aged conditions are higher than those of the control bitumen. Because of the increased deformation recovery and elevated surface energy, the LDPE modified bitumen has a reduced time to complete the surface rearrangement, approach and wetting in the healing processes. This means the LDPE in bitumen can accelerate the crack wetting process, thus leads to a higher wetting rate (i.e., the time derivative of the wetting distribution function $\varphi(t)$ is higher) and eventually results in higher healing. This is demonstrated by the higher short-term healing rate of the LDPE modified bitumen than that of the control bitumen. For instance, **Figure 8** shows that the short-term healing rate increased from $4.65\times10^{-3}sec^{-1}$ to $5.46\times10^{-3}sec^{-1}$ for the unaged bitumen, and **Figure 11** shows that it increased from $1.05\times10^{-3}sec^{-1}$ to $2.62\times10^{-3}sec^{-1}$ for the aged bitumen.

However, the LDPE in bitumen cannot accelerate the diffusion process to lead to faster healing, as demonstrated by the much less changed long-term healing rate in **Figure 11** between LDPE modified bitumen ($4.86\times10^{-3}sec^{-1}$) and the control bitumen ($4.84\times10^{-3}sec^{-1}$). The fundamental reason is that the main diffusible components in bitumen to lead healing are the light parts (i.e., saturate and aromatics). Some existing works of literature can support this explanation, such as the one completed by Yang et al. (2020). Based on the dynamic shear rheometer, they measured the data of complex viscosity and flow activation energy and found that saturates and aromatics diffused much better than resins and asphaltenes in the bitumen. The LDPE's molecular chains are much longer and heavier in molecular weight than those light bitumen components. This makes the LDPE almost non-diffusible and contributes little to long-term healing. In other words, the intrinsic healing function $R_h(t)$ remains unchanged between the LDPE modified bitumen and the control bitumen. Thus, the healing cannot be enhanced by the LDPE by the improved molecular diffusion to increase the intrinsic healing.

In sum, the increased deformation recovery ability induced by LDPE in bitumen improves the wetting rate of the cracked surfaces and leads to a higher short-term healing rate and bigger healing. However, the LDPE in bitumen cannot accelerate the molecular diffusion to increase the intrinsic healing or the long-term healing rate.

5. Summary and Conclusions

This paper characterised the healing performances of the unaged and PAV-aged waste-derived bitumen (i.e., bio-oil modified bitumen, and LDPE modified bitumen) based on crack length. The designed fatigue-healing test consisted of a strain-controlled time sweep fatigue test plus a rest duration and followed by another strain-controlled time sweep fatigue test. Crack length-based healing index and Ramberg-Osgood model were effectively utilised to characterise the healing rate and healing potential of the unaged and PAV-aged bitumen. The main findings and conclusions of this paper are summarized as follows:

(1) Both the bio-oil and LDPE can strengthen the healing performance of the unaged bitumen. The unaged bio-oil modified bitumen heals the most (having the highest healing potential) and fastest (having the highest short-term healing rate), followed by the unaged LDPE modified bitumen, and the unaged control bitumen heals the least and slowest.

(2) After the PAV ageing, the LDPE modified bitumen has much better healing performance than the control bitumen and the bio-oil modified bitumen. The PAV-aged bio-oil modified bitumen does not show any enhanced healing performance than the PAV-aged control bitumen. The PAV-aged LDPE modified bitumen heals the most and fastest among the above three types of PAV aged bitumen.

(3) The increased deformation recovery ability induced by LDPE in bitumen improves the wetting rate of the cracked surfaces and leads to a higher short-term healing rate and bigger healing. However, the LDPE in bitumen cannot accelerate the molecular diffusion to increase

the intrinsic healing or the long-term healing rate.

Conflict of interest

The authors declared that there is no conflict of interest.

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- Two waste materials were firstly used to enhance healing ability of bitumen
- Healing properties of bitumen was characterised based on crack length
- Healing rate and potential was modelled using Ramberg-Osgood Equation
- Bio-oil productively promotes healing performance of unaged bitumen
- LDPE strengthens healing performances of unaged and PAV-aged bitumen

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Statement of Conflict of Interest

Re: submission of paper (revised version) entitled "Healing Characterisations of Waste-derived Bitumen Based on Crack Length" by Linglin Li, Yang Yang, Yangming Gao, and Yuqing Zhang to Journal of Cleaner Production.

We (all authors) wish to confirm that there are no known conflicts of interest associated with this publication, and there has been no significant financial support for this work that could have influenced its outcome.

We confirm that the manuscript has been read and approved by all named authors and that there are no other persons who satisfied the criteria for authorship but are not listed. We further confirm that the order of authors listed in the manuscript has been approved by all of us.

We confirm that we have given due consideration to the protection of intellectual property associated with this work and that there are no impediments to publication, including the timing of publication, with respect to intellectual property. In doing so, we confirm that we have followed the regulations of our institutions concerning intellectual property.

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