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The use of bio-oil from biodiesel production for enhancing the bitumen healing

Marina Cabette [a], Rui Micaelo [b,*], Jorge Pais [c]

[a] Dept. of Civil Engineering, University of Minho, Guimarães, Portugal
[b] CERIS, Dept. of Civil Engineering, NOVA School of Science and Technology, Caparica, Portugal
[c] ISISE, Dept. of Civil Engineering, University of Minho, Guimarães, Portugal

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ABSTRACT

Road pavements are exposed to traffic and temperature effects that produce distress, mainly cracking that leads to pavement failure, after which rehabilitation actions are necessary to extend pavement life. Bio-rejuvenators have been used to recover the bitumen properties lost due to fatigue of the asphalt mixtures and ageing due to temperature variations and oxidation. Thus, this paper investigates the healing of bitumen modified with bio-oil derived from biodiesel production. One base bitumen in three different ageing stages (unaged, short and long-term ageing) was used for adding up to 3% of bio-oil to the bitumen. In addition to consistency and rheological tests to characterise the modified bitumen, fatigue resistance tests with and without rest periods were performed to assess the healing through three indexes, relating the complex shear modulus, the number of cycles to failure and the amount of damage, after a rest period. It was verified that the healing follows an exponential law with the rest period, but the number of cycles in each loading period decreases compared to the previous period. For a 15-minute rest period, the healing was limited to 26.9%, but the bio-oil contributed significantly to that restoration.

1. Introduction

When exposed to traffic and temperature variations, pavements are subject to distress, mainly fatigue and thermal cracking. Cracking constitutes one of the main concerns for the asphalt pavement's integrity and leads to disrepairs associated with discomfort and safety for users. In addition, cracking allows water entrance to the pavement layers and subgrade, consequently reducing the support capacity and promoting other types of distress. Road administrations are focused on delaying or, when possible, avoiding cracking in asphalt pavements to prevent the reduction of functional and structural performance [1–3].

Bitumen has a significant influence on asphalt mixtures and pavement performance. Bitumen properties evolve during production, storage, transport, and placement of the asphalt mixture and in-service, resulting in adverse physical and chemical changes (ageing process) [4–6]. The gradual variation of viscoelastic properties due to ageing makes bituminous binders harder and more brittle [6], leading to the fatigue and thermal cracking of asphalt pavements. Excessive hardening must be reduced to avoid pavement life reduction [4]. To address bitumen hardening, researchers have investigated the natural ageing resistance of different bitumens and additives to increase its resistance or to revert ageing. To the latter, one promising solution is adding the bitumen of softening or rejuvenating agents, which, due to its lower viscosity, can revert ageing-related changes in bitumen properties [7,8]. Rejuvenators and softening agents can be used in different ways, namely in the recycling of reclaimed asphalt pavements [9], applied directly onto the asphalt surface [10] or inside capsules embedded in asphalt mixtures [11]. Among different agents, bio-oils derived from organic sources or industrial by-products are preferred [12].

The use of petroleum-based products is associated with several environmental issues, such as large greenhouse gas emissions, depletion of natural non-renewable resources, and soil and water pollution. As an alternative to these products, biomass and waste from different industries are processed with different thermochemical processes to obtain dense oils that can be used in asphalt materials, and the research reported so far is generally favourable to its adoption [13]. Bio-oils and their derivatives can be used as modifiers or rejuvenators in asphalt materials, offering promising performance, such as durability, flexibility, and resistance to ageing and cracking [8,14,15]. However, the bio-oil properties vary with the source and processing technique used

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* Corresponding author.
E-mail addresses: ruilbm@fct.unl.pt (R. Micaelo), jpais@civil.uminho.pt (J. Pais).

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[16]. Thus, further investigation is required to characterize these materials and understand the interaction mechanism of bitumen and bio-oil.

Zargar *et al.* [17] and Sun *et al.* [18,19] explored the effects of waste cooking oil (WCO) and the residue of WCO processing for biodiesel production on aged bitumen using various rheological tests. Zargar *et al.* found that adding 3–4 % of WCO to aged bitumen (40/50) could soften it to a level similar to the original bitumen (80/100). At the same time, Sun et al. discovered that the WCO biodiesel-derived bio-oil could decrease the complex modulus and creep stiffness and increase the bitumen's phase angle and *m*-value, leading to improved stress relaxation and thermal cracking resistance. However, the Fourier transform infrared spectroscopy (FT-IR) analysis [19] revealed no significant chemical reactions between the control bitumen and the biodiesel by-product. While the thermal stability of the by-product was somewhat lower than that of the control bitumen, it was still suitable for mixing and testing procedures. Also, the storage stability tests showed that the by-product-modified bitumens remained stable during hot storage and transportation, with differences in complex shear modulus values being less than 20 %.

Gong *et al.* [20] extended these findings by examining the use of a similar WCO biodiesel-derived bio-oil as a softener for bitumen. They found that the bio-oil improves the rheological properties of aged bitumen, which can reduce energy consumption and carbon dioxide emissions during production. They found that short-term aged 50-pen bitumen and styrene–butadiene–styrene (SBS) modified bitumen could be recovered to their original level with a bio-oil content of 1.75 % and 2.00 %, respectively. However, adding bio-oil also increased the susceptibility of aged binder to moisture damage. The oil compensates for the loss of light components with ageing, and the aged bitumen with oil had less aggregation of polar components based on the Atomic Force Microscopy images.

In the same way, Zadshir et al. [4] investigated the impact of three modifiers (petroleum-based oil, modified vegetable oil and swine manure-derived oil) on the chemical and thermo-mechanical properties of aged bitumens and found that the rejuvenators effectively reduce the increased molecular size ratio caused by oxidation. Then, molecular dynamics simulations revealed that amide groups in bio-rejuvenators interact with asphaltene molecules, altering their conformational packing, which they related to the observed improvements in the bitumen's rheological properties. However, Ren et al. [21] found that the diffusion capacity of the rejuvenator in the bitumen decreases with the ageing level, which was justified with the variation of the free volume fraction distribution and the intermolecular force between rejuvenator and bitumen molecules. Thus, the intermolecular force also varies with the rejuvenator.

In addition, Gao *et al.* [22] evaluated the penetration, ductility, softening point, and temperature and carried out frequency sweep tests on bitumen modified with a different type of bio-oil (wood-chip-derived bio-oil). Their results indicated that increasing the bio-oil content increases the penetration, ductility, and anti-cracking performance. The softening point and the anti-rutting ability of unaged bio-bitumen decreased when compared with the non-modified bitumen. Zhang *et al.* [23] analysed the effect of a sawdust-derived bio-oil on two long-term aged bitumens (PG 58–28 and PG 64–22 binders). They found that the rotational viscosity of aged bitumen could be decreased to the same level, or close to that, of virgin bitumen with 15–20 % bio-oil incorporation. So, using bio-oil, the rutting index of aged PG 58–28 and PG 64–22 binders decreased by 75.5 % and 77.2 % on average, respectively, for temperatures from 52 °C to 76 °C. The bio-oil also restored the low-temperature crack resistance of aged binders to their original properties. Asadi *et al.* [24] described that adding bio-oil to bitumen can improve its fatigue-fracture behaviour and performance due to its increased elasticity and damage tolerance.

Related to cyclic loading response and damage, in [25,26] is reported that bio-oils can also improve the self-healing properties of bitumen. Generally, healing materials are defined as being able to restore their original properties after damage through thermal, mechanical, or other means [27]. Specifically for bituminous materials, based on the work of the Rilem Technical Committee TC 278-CHA (Crack-Healing of Asphalt Pavement Materials), Leegwater *et al.* [28] distinguished two types of healing: intrinsic when the restoration is provided from the material's inherent properties; and extrinsic when the restoration, crack-healing, or prevention of crack propagation is improved by adding a phase or external action. The healing phenomenon of bituminous materials is currently a hot research topic [27].

The healing assessment is usually carried out by comparing the behaviour of the damaged material after applying internal or external factors to restore the initial condition. Then, the healing index is calculated as the ratio of the quantities describing the material's behaviour in the two states, which can be determined by testing it with monotonic or cyclic tests. The process is rather straightforward because only one test configuration is used. Nevertheless, due to the viscoelastic nature of bituminous materials, the loading response, especially in cyclic tests, is affected by non-damage-related mechanisms, which stop or decrease its effect with time when the test is interrupted. To this end, Leegwater *et al.* [28] proposed to adopt the term "restoration" to the total change in properties after the rest period, and "recovery" and "self-healing" to the components of restoration due to loading-related reversible mechanisms and (micro)-cracks repair, respectively.

Several tests have been used to investigate bitumen healing [29–31]. However, the most used tests consist of constant-amplitude cyclic loading protocols with the introduction of rest periods that can be frequent short periods or a few long periods. Often, with the latter type of resting in the cyclic test, the amount of healing is quantified based on the fatigue life or restoration of the stiffness modulus before and after the rest period [27]. Differently, Xie *et al.* [32] proposed a healing test protocol for a linear-increasing amplitude cyclic test, with rest periods applied before and after cohesive failure, and quantified the healing index from the restoration of the accumulated damage growth that was calculated based on the simplified-viscoelastic continuum damage (S-VECD) model. Also, Baglieri *et al.* [33] adopted a visco-elastic continuum damage mechanics approach to characterise bitumen healing using oscillatory shear tests with rest periods.

Relative to the effect of bio-oils on healing, Gaudenzi *et al.* [34] evaluated the healing capacity of two bitumens and one bio-oil-modified bitumen obtained from tall crude oil. The authors conducted time sweep tests (TST) in strain-controlled mode, in both *iso*-stiffness and *iso*-thermal conditions, and the healing contribution was defined as the variation of the number of loading cycles required to reach the same damage level after each rest period. The healing index results were considerably low (2–8 %) for the tested materials, however, the authors of the referred study concluded that healing depends mainly on the binder consistency, which decreases with bio-oil and increases with ageing. Sun *et al.* [26] also investigated the effects of ageing and bio-oil (WCO biodiesel-derived and a petroleum-based rejuvenator) regeneration on the healing behaviour of two types of bitumen with TST but calculated the healing index with the stiffness modulus. The healing results at 25 °C were significantly higher for the bio-oil-modified binders, reaching 25 % with one bitumen and 61 % with the other.

Previous studies found that bio-oil derived from several sources have the potential to be a bitumen modifier and rejuvenator [35–37]. In particular, bio-oil derived from the biodiesel production process increases the proportion of aromatics, resins and saturates in bitumen which prevents the aggregation of highly oxidised components within the aged bitumen, however, the interaction of oil and bitumen function groups is not fully understood [18,20,38]. The oil-modified bitumen has lower viscosity and greater low-temperature performance. Recently, Pais *et al.* [38] investigated a bio-oil from biodiesel production that uses WCO and animal fat and concluded that the properties (moisture sensitivity, dynamic modulus and phase angle, fatigue resistance and rutting resistance) of asphalt mixtures with bio-oil modified bitumen were compatible with those of asphalt mixtures with traditional bitumens. However, despite the mixture with the largest bio-oil content having the longest fatigue life, the other mixtures with less bio-oil performed worse than the reference mixture without oil. In summary, there is limited research regarding the effect of bio-oils, namely the ones derived from the biodiesel production process, on the fatigue-fracture behaviour and self-healing properties, and their evolution with oxidative ageing, which are especially important for the in-service performance.

2. Objectives

This research study aimed to investigate the fatigue and healing behaviour of bitumen modified with bio-oil derived from biodiesel production and to evaluate the ability of this bio-oil to revert the ageing effects on the bitumen. A paving grade bitumen (35/50) in three ageing states (unaged, short- and long-term aged) was blended with 1–3 % bio-oil. The fatigue behaviour is evaluated with constant- and increasing loading amplitude cyclic tests, and for the healing behaviour, the loading phases are alternated with rest periods at a specified damage level. Also, the healing index calculation is assessed by considering different test quantities, namely the complex shear modulus, the fatigue life and the VECD-derived damage intensity.

3. Materials and methods

3.1. Materials

A standard paving grade bitumen, 35/50, was used. Table 1 presents the bitumen's main properties, namely the needle penetration, softening point, and viscosity. The 35/50 grade is a semi-hard bitumen suitable for use in hot climates or under heavy traffic conditions.

The bio-oil used in this work is a by-product of biodiesel production from waste cooking oil and animal fats, containing a range of organic compounds that are less volatile than those found in biodiesel and some residual Fatty Acid Methyl Ester (FAME). The specific composition of bio-oil varies depending on the feedstocks used to produce the biodiesel and the distillation process [38]. The bio-oil sample used in the study had high viscosity; therefore, it was blended with FAME in equal proportions (50/50) by weight before its incorporation in bitumen.

The bio-oil was incorporated in three bitumen-aged conditions: unaged, short-term, and long-term. Short-term ageing was simulated through the Rolling Thin Film Oven (RTFOT) based on the EN 12607–1 test protocol [39], and the long-term ageing was simulated by the Pressure Ageing Vessel (PAV) following the EN 14769 test protocol [40]. The RTFOT treatment aims to induce in the bitumen a similar ageing to that occurs during asphalt mixing, transporting, and compaction, and the PAV treatment aims to simulate in the material the ageing level after 5 to 10 years in service. Unaged bitumen is referred to as BU, RTFOT aged bitumen is referred to as BR and PAV aged bitumen is referred to as BP. For each bitumen, 1, 2 and 3 % of bio-oil was added by weight of the total mix. Binders were prepared in a low-shear mixer by blending the bitumen and the bio-oil at 150 °C for 5 min at 350 rpm. Thus, the previous terminology is followed by the bio-oil content. For example, the unaged bitumen with 3 % bio-oil is referred to as BU3.

Table 1
Properties of 35/50 penetration grade bitumen.

Property	Test method	Value
Penetration at 25 °C (0.1 mm)	EN 1426	37
Softening point (°C)	EN 1427	53
Dynamic viscosity at 150 °C (Pa.s)	EN 13,302	0.68

3.2. Consistency and rheological characterisation

The consistency of the bio-oil-modified bitumen was measured through the needle penetration and ring and ball tests, following the European standards EN 1426 [41] and EN 1427 [42], respectively. A frequency sweep test at low strain amplitude was conducted using a dynamic shear rheometer to perform a linear viscoelastic rheological characterisation of the material at an intermediate temperature (20 °C).

3.3. Linear amplitude sweep test

The fatigue resistance, without healing contribution, of the bio-oil-modified bitumens was assessed using the Linear Amplitude Sweep (LAS) test (AASHTO-TP 101–14 [43]), which involves the application of a series of oscillatory load cycles at linearly increasing amplitudes to a disc-shaped specimen (diameter of 8 mm and height of 2 mm). The strain amplitude starts at 0.1 %, then 1 %-30 %, with a constant increase of 1 % every 10 s loading. The loading frequency was constant throughout the test (10 Hz). Hence, this test can induce fatigue damage in the specimen in less time than the constant amplitude time sweep test. The LAS test was performed at the intermediate temperature of 20 °C.

To determine the fatigue resistance, the LAS test results were analysed using the Viscoelastic Continuum Damage (VECD) approach defined in AASHTO-TP 101–14 [43]. Briefly, the material integrity C at cycle i is given by

$$C_i = \frac{|G^*|_i}{|G^*|_{ini}} \tag{1}$$

and the damage intensity at time t (N cycles) is

$$S_t = \sum_{i=1}^{N} \left[\pi \times \gamma^2 \times (C_{i-1} - C_i)\right]^{\frac{\alpha}{1+\alpha}} \times (t_i - t_{i-1})^{\frac{1}{1+\alpha}} \tag{2}$$

where, $|G^*|_i$ is the shear complex modulus at cycle i, $|G^*|_{ini}$ is the initial shear complex modulus (undamaged state), γ is the loading amplitude, and α is the inverse of the maximum slope (m) of the relaxation modulus (E(t) vs t) on the log–log scale. The m value was determined from the frequency sweep test described in section 3.2. The specimen's failure is defined at the point where the shear stress is maximum, and the fatigue life is determined by

$$N_f = \frac{f\left(\frac{1-C_{\tau\,max}}{C1}\right)^{\frac{1+(1-C2)\alpha}{C2}}}{[1+(1-C2)\alpha](\pi \times C1 \times C2)^{\alpha}}\gamma^{-2\alpha} \tag{3}$$

where N_f is the number of cycles to failure, f is the loading frequency, $C1$ and $C2$ are the power law constants of C vs S, and $C_{\tau\,\mathrm{max}}$ is the integrity at the shear stress peak (specimen's failure point).

3.4. Time sweep test with rest periods

The healing capacity of the bitumen was evaluated from time sweep tests in strain-controlled mode, using a procedure similar to the research described in [44–46], which considers the total change in mechanical properties after the rest period as healing.

The base procedure consists of sequential loading and resting periods (see Fig. 1), where the loading period ($i = 1, 2, \ldots$) is interrupted when the complex shear modulus ($|G^*|$) decreases to 50 % of the initial value (undamaged condition) and the duration of the resting period ($j = 1, 2, \ldots$) is constant. The test was conducted at 20 °C, and the specimen size was the same as used with LAS. Three healing indexes were determined from test results, which differ in the test quantity used, i.e. the complex shear modulus, the number of cycles to failure and the amount of damage.

The index *HIm* after the rest period j is obtained with the complex

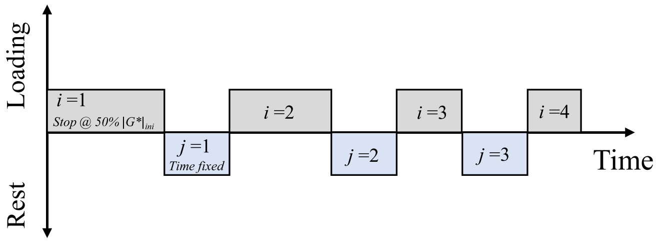


Fig. 1. The sequence of the time sweep test with rest periods.

shear modulus as:

$$HIm_j = \frac{|G^*|_{i+1_{initial}} - |G^*|_{i_{final}}}{|G^*|_{i_{final}}} \times 100 = \frac{\Delta|G^*|}{|G^*|_{i_{final}}} \times 100 \tag{4}$$

where $|G^*|_{i+1_{initial}}$ is the initial complex shear modulus value of loading period $i + 1$ (post rest j), and $|G^*|_{i_{final}}$ is the final complex shear modulus of loading period i (before rest j). The index *HIc* after the rest period j is determined with the number of loading cycles as:

$$HIc_j = \frac{\Delta N_{i+1}}{N_1} \times 100 \tag{5}$$

where N_1 is the number of cycles of the first loading period, and ΔN_{i+1} is the number of cycles of loading period $i + 1$ (post rest j). The index *HId* after the rest period j is determined with the damage as:

$$HId_j = \frac{S^F_{i+1} - S^F_i}{S^F_i} \times 100 \tag{6}$$

where S^F_i and S^F_{i+1} are the final S values of loading periods i and i-1, respectively. Fig. 2 illustrates the quantification of *HIm*, *HIc* and *HId* for the first rest period ($j = 1$).

The bitumen's behaviour under cyclic loading is affected by numerous factors, such as the material's stiffness, temperature, loading frequency, etc. Therefore, the effect of the temperature, the strain amplitude, the rest time and the number of loading-resting phases were analysed to define the test conditions for healing tests.

Fig. 3 shows the effect of the strain amplitude ($\gamma = 1.0$ %, 2.5 % and 5.0 %) on the decrease of the complex shear modulus, normalised to the initial value ($|G^*|/|G^*|_{ini}$), with the loading cycles for bitumen BU0, and as expected, the highest the strain amplitude is, the shortest the fatigue life of the material.

Fig. 4 shows the evolution of the normalised complex shear modulus for three specimens throughout 12 loading phases with resting periods of 15 min in between. The complex shear modulus restoration after each rest period decreased rapidly with the accumulation of the loading cycles, meaning that the healing contribution decreased with it also.

Fig. 5a and 5b present the test results of two binders (BU0 and BU2) for rest times varying between 5 min and 120 min. During the rest period, or when a sample is allowed to relax after applying a constant strain, the bitumen molecules can rearrange and recover their original structure, eventually increasing the complex shear modulus [47].

To aid the interpretation of the effect of the rest time, Fig. 6 shows the healing index values of the first rest period (HIm_1, HIc_1 and HId_1) versus the rest time. Despite the differences between the two binders, the three indexes show increased healing with rest time. The relationship between the healing index and the rest time followed an exponential law with excellent fitting to the data, as demonstrated in Fig. 6. As the rest time increases, the healing index also tends to increase, meaning that the material can partially recover the lost stiffness and increase fatigue after being subjected to deformation when given sufficient rest time.

Given these preliminary test results to the effect of the strain amplitude, the rest time and the number of rest periods, and for practical reasons of the laboratory work, the time sweep test with rest periods was implemented with a strain amplitude of 2.5 %, a rest time of 30 min and a sequence of 4 loading and 3 rest periods.

4. Results

4.1. Consistency and rheological characterisation

Fig. 7a and 7b show the effect of the bio-oil on the penetration and the softening point of the bitumen aged to different levels. The higher the ageing level is, the lowest the penetration and the highest the softening point. The bio-oil significantly affected the consistency of bitumen, and on average, 1 % bio-oil resulted in an increase of 30 % in penetration and a decrease of 2 °C in the softening point. Also, the penetration increased exponentially with the bio-oil content, whereas the softening point linearly decreased.

To understand the effect of the bio-oil on the viscoelastic rheological behaviour of the bitumen at intermediate conditions, Fig. 8 shows the results of the frequency sweep test performed at 20 °C.

As anticipated, the ageing treatment increased complex shear modulus and decreased phase angle, which can be reverted with the bio-oil. Also, except for the binders BP0 and BP1 at the highest loading frequencies, the bitumens showed a viscoelastic behaviour dominated by its viscous part, which occurs when the δ is higher than 45°, and the importance of the viscous behaviour increased with the bio-oil content

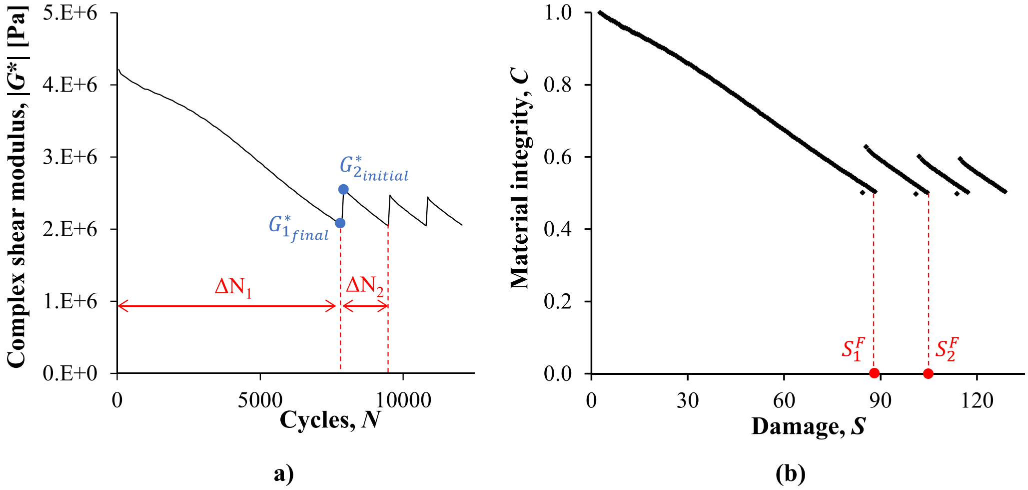


Fig. 2. Calculation of healing indexes: (a) HIm_1 and HIc_1; (b) HId_1.

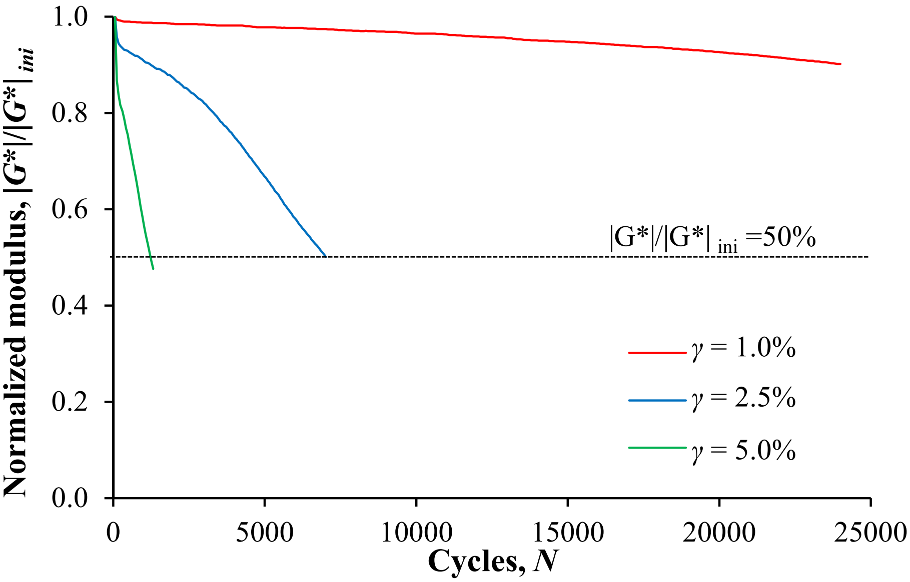


Fig. 3. Effect of the strain level on the normalised complex shear modulus (sample BU0).

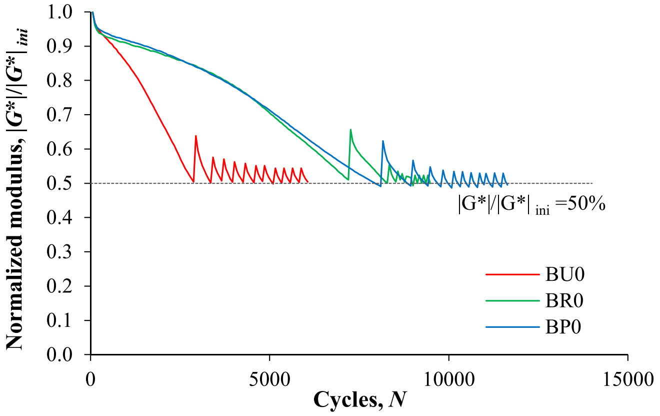


Fig. 4. Effect of the number of rest periods on the normalised complex shear modulus.

and decreased with the ageing level. In the literature, the temperature at which the δ overpasses 45° is referred to as the viscoelastic transition temperature [26,48]. Therefore, for most of the tested binders, this temperature is below 20 °C. Notably, the bio-oil had a more significant effect on the stiffness modulus than on the phase angle, e.g. for 3 % bio-oil, at 10 Hz, $|G^*|$ reduced 77 % and δ increased 18–30 % depending on the ageing level of the bitumen.

4.2. Fatigue resistance without rest periods

The LAS test was performed primarily to evaluate the fatigue resistance of the binders without the beneficial effect of the rest period. Fig. 9 illustrates the shear stress versus shear strain on the LAS test. The curve is similar in shape for all tested binders, but the stress level held by the specimens during cyclic loading increased with the ageing treatment (from unaged up to PAV treatment) and decreased with the addition of the bio-oil, which is related to the stiffness of the material. In the literature, different studies [49–51] suggested using the point at which the maximum stress level is reached to determine the failure point of the specimen in the test. From this, the results show that the ageing

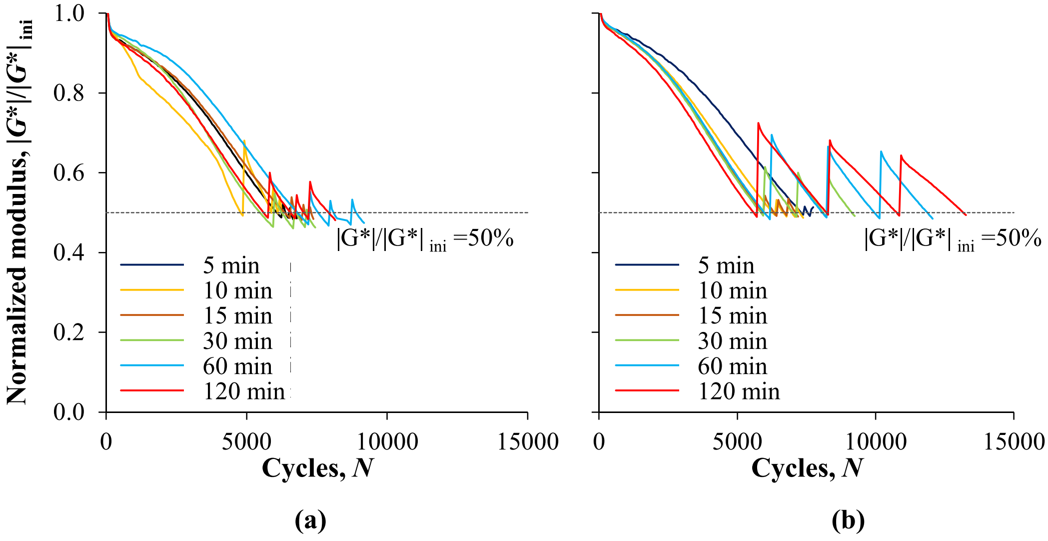


Fig. 5. Effect of rest time on the normalised complex shear modulus: (a) BU0; (b) BU2.

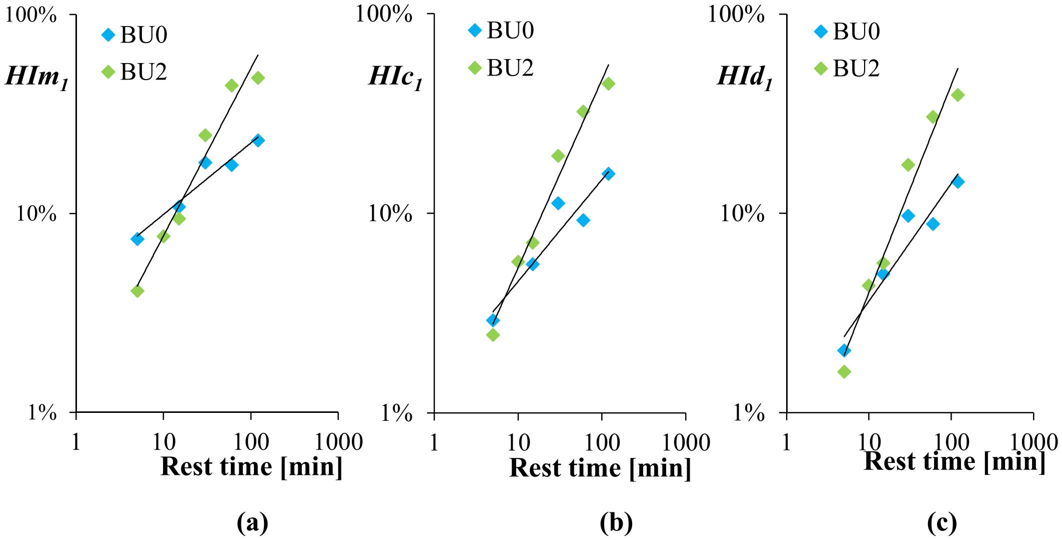


Fig. 6. Healing index of first rest period with different rest periods for BU0 and BU2: (a) HIm_1; (b) HIc_1; (c) HId_1.

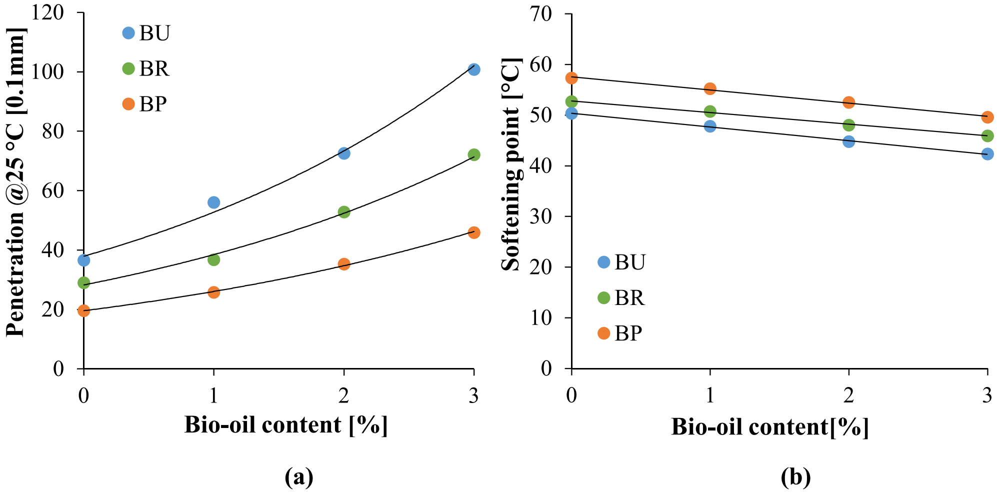


Fig. 7. Test results: (a) penetration; (b) softening point.

treatment made the bitumen harder and less ductile, and this effect was reversed with the addition of the bio-oil. On average, the tangent modulus in the initial part of the test ($\gamma < 4$ %) increased by 47 % with each ageing treatment and decreased by 75 % with the incorporation of 3 % bio-oil.

The fatigue resistance of bitumens from the LAS test was then evaluated using the VECD methodology, which involves determining the material integrity (C) versus damage (S) curves, also known as “damage characteristic curves”. Fig. 10 illustrates the impact of the bio-oil on the damaged characteristic curves of bitumens subjected to different ageing levels. In the LAS test, the C value, which starts at 1.0, decreased as the number of loading cycles increased, which is related to the growth of the S variable. It is observed that the S value required to achieve the same level of C reduction was lower in aged bitumens and increased with the bio-oil content. This trend is seen in all three bitumens. C-S curves were compared by calculating the areas under the curves, presented in Table 2. The short and long-term ageing without bio-oil resulted in around a 22 % decrease in the C-S area, and adding just 2 % of bio-oil on short and long-term ageing samples is enough to achieve the same level as the unaged bitumen (BU0).

The number of loading cycles that the bitumen can support was determined with Eq. (3), and Fig. 11 compares the estimated resistance for two strain amplitudes (2.5 % and 5.0 %). Surprisingly, the long-term aged bitumen, with and without rejuvenator, exhibited greater fatigue resistance, while the short-term aged bitumen performed worse than the unaged bitumen. These results are attributed to the sensitivity of the C-S model to the strain level. Although the power-law model used to fit C-S curves did not accurately capture the entire C-S curve for some bitumens, it is observed that a small addition of bio-oil increased the fatigue resistance for all bitumens (BU, BR, and BP).

In this study, the testing protocol of the time sweep test was adapted (described in section 3.4) to investigate the healing capacity of studied binders. However, the number of cycles applied in the first loading phase, which was interrupted when the complex shear modulus decreased to less than 50 % of the initial value, can be compared with the fatigue resistance estimated from the LAS test for the same strain amplitude (2.5 %), see Fig. 12. In all cases, the fatigue resistance estimated from LAS was significantly lower than that measured in the time sweep test (average values). Also, the results show a smaller variation in the fatigue resistance estimated from LAS for the different materials. Differently, Safaei *et al.* [52] reported a good correlation between these two tests; however, they used different analysis protocols for both tests.

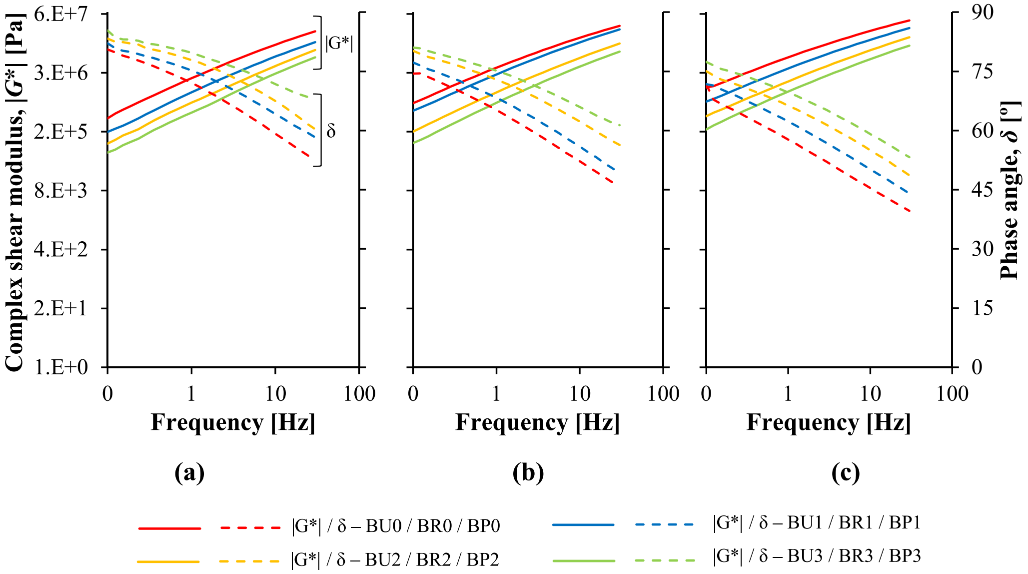


Fig. 8. Variation of $|G^*|$ and δ with the frequency, at 20 °C: (a) BU; (b) BR; (c) BP.

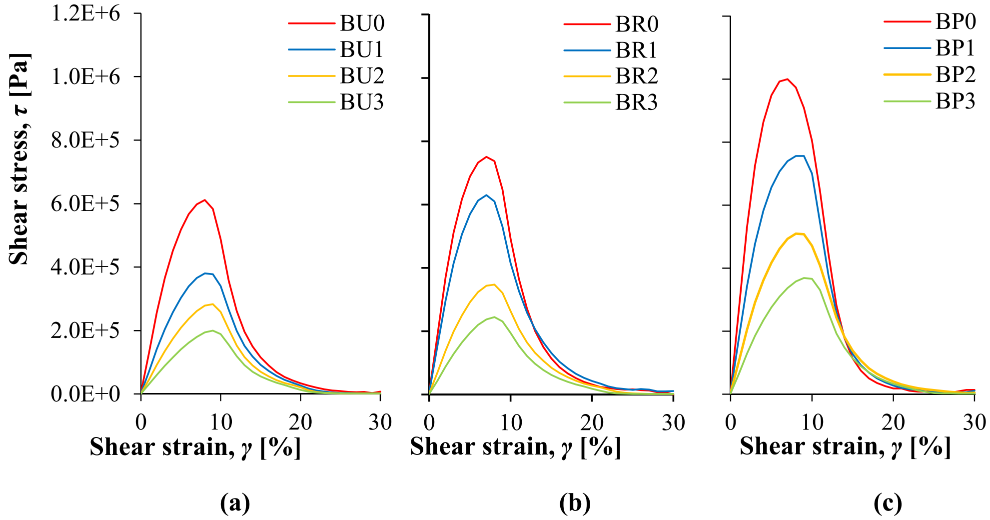


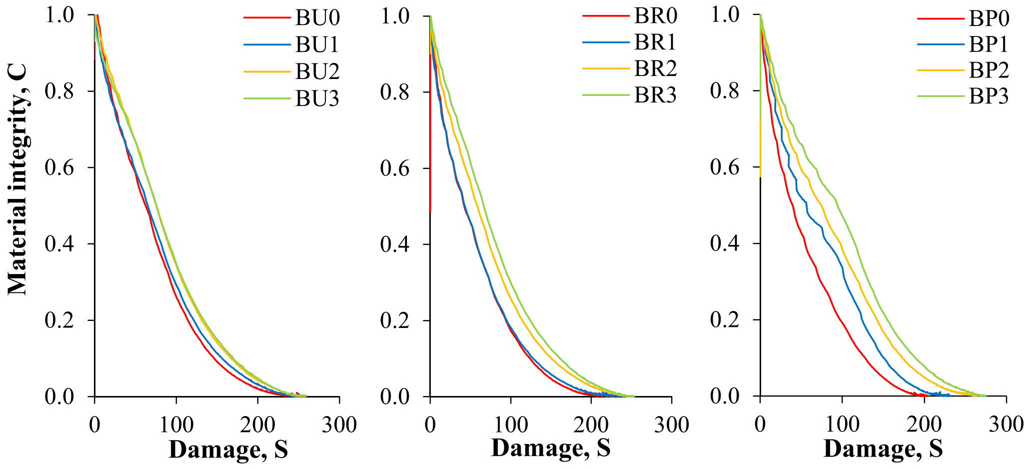
Fig. 9. Stress-Strain curves on LAS tests: (a) BU; (b) BR; (c) BP.

4.3. Healing potential from time sweep testing with rest periods

The fatigue life is an essential factor in the design of asphalt pavements, as it ensures that the pavements will have a long service life and will require few maintenance and repair operations. Continuous loading used in most cyclic tests is convenient for reducing the testing time, but it does not realistically represent the real traffic patterns with short and long rest periods between loads [53]. Thus, the inclusion of rest periods between loads is essential to evaluate the healing contribution to fatigue resistance. Fig. 13 illustrates the evolution of the complex shear modulus, normalised to the initial value, with the loading cycles in the four loading periods. As expected, it is observed that the ageing level and the bio-oil content affected the number of cycles held by the specimens in the test. For the same specimen, the number of cycles in each loading period decreased in comparison with the previous period, which is related to the accumulation of damage that is not fully recovered in the rest period. In general, for the same ageing level, the number of loading cycles increased with the bio-oil content, which means that adding bio-oil to the binder has a positive effect on the fatigue life of the material. Differently, the effect of ageing is less clear because, for the same oil content, the number of loading cycles can decrease or increase with increasing ageing.

It is noteworthy to mention that, as shown in Fig. 14, the limit of 50 % reduction in the complex shear modulus that was used as a stopping condition for the loading periods was insufficient to obtain a peak in the graph of the $N \times [|G^*|/|G^*|_{ini}]$ versus N. This peak has been associated before with the specimen's failure in cyclic tests and is considered a better failure indicator than the 50 % reduction in $|G^*|$ [54]. In the first loading period, the test is stopped just before reaching the peak. However, in the following loading periods, the curves show constant slope lines throughout the period. Also, it is observed in Fig. 14 a similar effect of the bio-oil and ageing to that found in Fig. 13.

This study evaluated the contribution of healing during rest periods to the loading response with three indicators (*HIm*, *HIc* and *HId*) because there is no unanimous method to quantify it. It should be noted that the bitumen's response to cyclic loading is affected by multiple phenomena, including stress relaxation/strain recovery, thixotropy, steric hardening and healing, which also influence restoration after resting [27,55]. However, it is not straightforward to separate their individual effects and evaluate their differences among bitumens. So, the healing indexes



(a) **(b)** **(c)**

Fig. 10. Damage characteristic curves (C-S): (a) BU; (b) BR; (c) BP.

Table 2
Area measured below C-S curve.

Sample	Area (-)	Sample	Area (-)	Sample	Area (-)
BU0	71	BR0	55	BP0	54
BU1	74	BR1	56	BP1	60
BU2	84	BR2	69	BP2	70
BU3	84	BR3	77	BP3	80

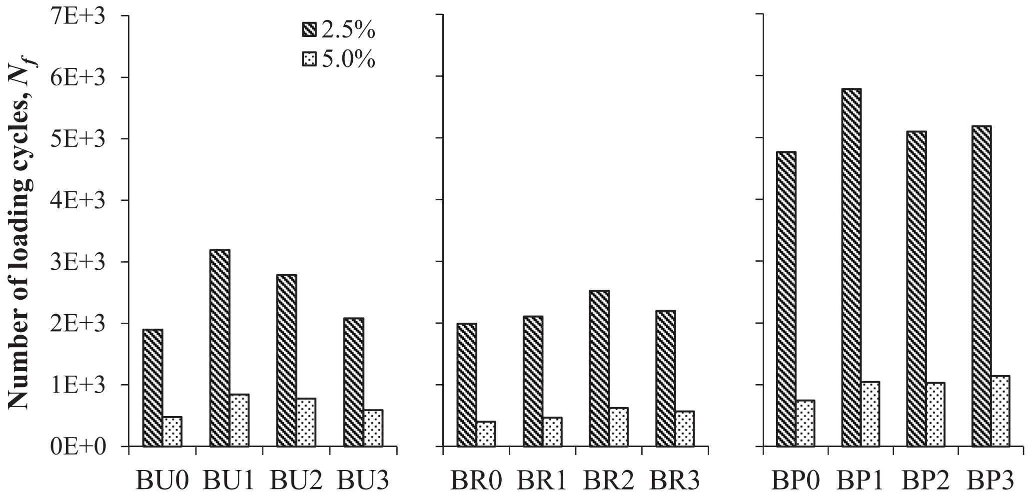
calculation considers the total observed change in mechanical properties, i.e. the loading response restoration.

Fig. 15 shows the variation of the healing indexes (average values) in the three rest periods (*j* = 1, 2 and 3), with the bio-oil content for the different ageing levels. Overall, the amplitude of variation in *HIm*, *HIc* and *HId* values was (min–max) 7.7–26.9 %, 1.1–20.0 % and 1.1–18.4 %, respectively. The indexes HIc and HId results were very similar (average difference of 1.3 %) regardless of the ageing level and bio-oil content. This means that, after the rest periods, the number of cycles sustained in the loading period is proportional to the damage intensity increase in that same loading period, which is used to calculate *HId*. Thus, it can be seen in Fig. 2a and 2b that, for both $|G^*|$-N and C-S curves, the curve slope is very similar in the different loading periods. Differently, the healing index based on the complex shear modulus restoration after the rest period, *HIm*, was higher than *HIc* and *HId* in all cases. The average difference was 7.2 % for *HIc* and 8.4 % for *HId*.

Nevertheless, it is observed a common effect of the bio-oil content, the ageing level and the number of load/rest repetitions from the analysis of the three healing indexes. First, the healing indexes values reduced by an average of 1.1 % with every increase in the ageing level, i.e. from BU to BR and BR to BP, but the largest differences were found for the highest oil content (-4.3 % BR3 and −3.2 % BP3).

Oxidative ageing changes the chemistry of the bitumen, especially the proportion of the chemical groups, which gives the viscoelastic behaviour, ductility, elastic recovery, flowability, etc., to the bitumen to perform well under very different conditions [47,56,57]. To address this problem, the rejuvenator should change the aged bitumen by rebalancing the chemical composition of bitumen and the rheological and performance-related properties eventually.

Hence, the healing index increased with the bio-oil content. For every 1 % increase in bio-oil, the average increase was 3.3 %, 2.4 % and 1.1 % for BU, BR and BP, respectively. Moreover, these results show the limitations of the bio-oil to rejuvenate aged bitumen. In the literature,



(a) **(b)** **(c)**

Fig. 11. Fatigue resistance estimated from LAS test: (a) BU; (b) BR; (c) BP.

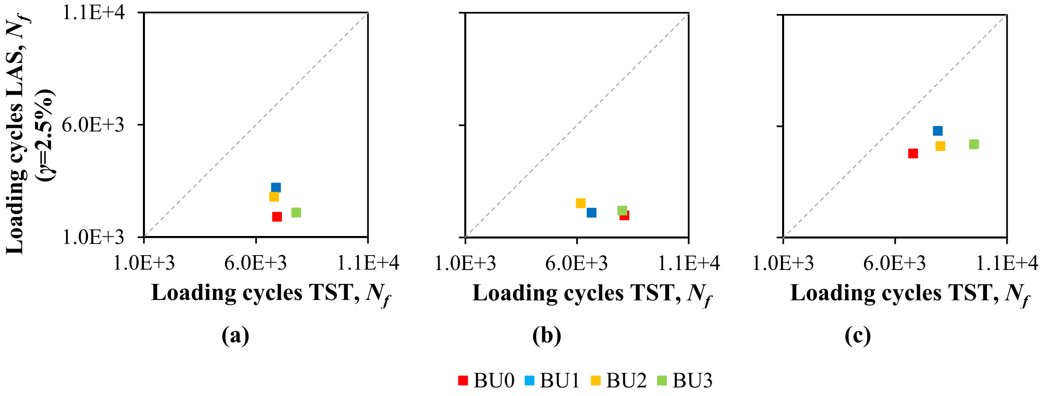


Fig. 12. Fatigue resistance (γ = 2.5 %), LAS vs. TST (1st loading period): (a) BU; (b) BR; (c) BP.

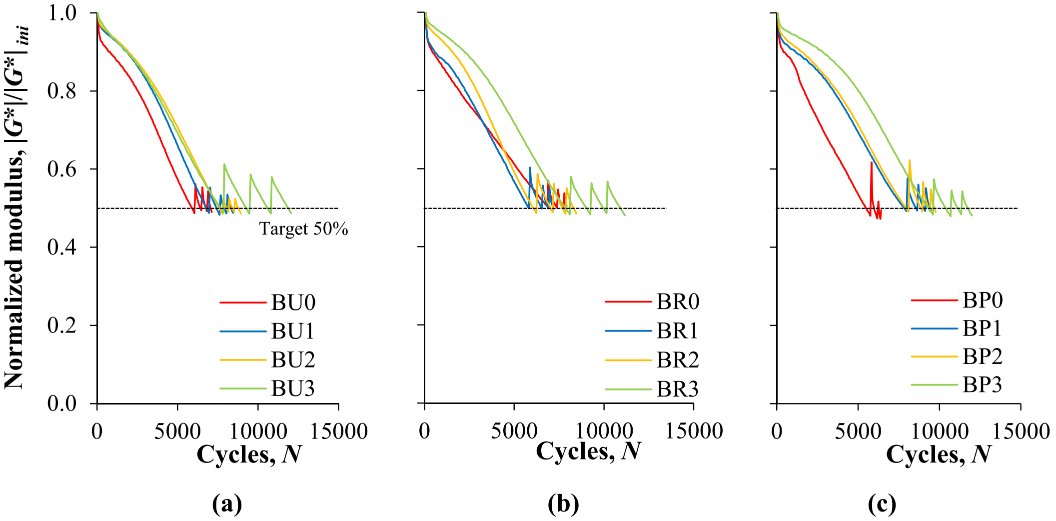


Fig. 13. Evolution of the normalised complex shear modulus with loading cycles: (a) BU; (b) BR; (c) BP.

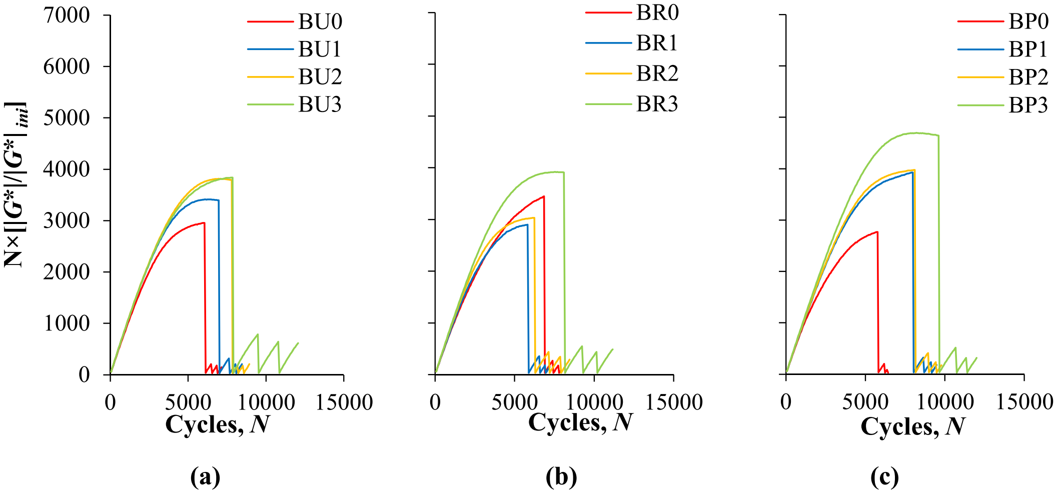


Fig. 14. Evolution of $N \times [|G^*|/|G^*|_{ini}]$ with loading cycles: (a) BU; (b) BR; (c) BP.

Sun *et al.* [26] and Gaudenzi *et al.* [34] reported a positive effect of both ageing and rejuvenator content on the healing index at a similar intermediate temperature (~20 °C). However, Sun *et al.* also showed that at higher temperatures (25–30 °C), the effect of ageing was negative, from which they formulated the hypothesis of a healing-temperature curve for each bitumen where the healing behaviour changes could be related to the elastic recovery ability at low to intermediate temperatures and the flow ability at higher temperatures. Moreover, according to these authors, the proposed healing-temperature curve would be dislocated into the lower temperatures with ageing and in the opposite direction

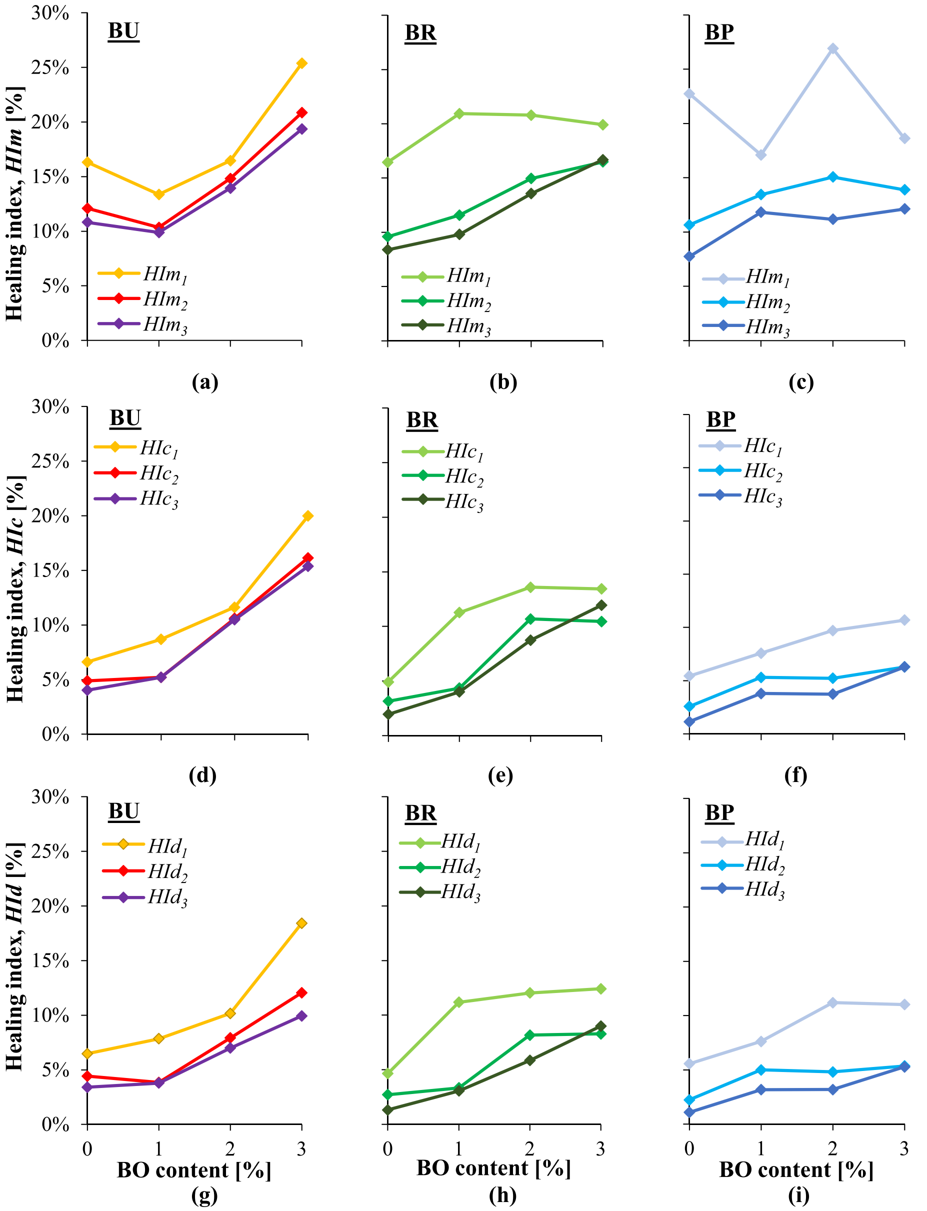


Fig. 15. Healing indexes: (a) *HIm* – BU; (b) *HIm* – BR; (c) *HIm* – BP; (d) *HIc* - BU; (e) *HIc* - BR; (f) *HIc* - BP; (g) *HId* - BU; (h) *HId* - BR; (i) *HId* - BP.

with the rejuvenator.

As expected, the loading-rest sequence, and the accumulated damage, affected the healing indexes values. For the first rest period, the average values of *HIm*, *HIc*, *HId* were 19.6 %, 10.3 % and 9.9 %, respectively, and decreased 3.2–5.9 % and 0.7–1.5 % in the second and third rest periods. During the rest period, different mechanisms, damage-related (e.g. internal structural rearrangements) and non-damage-related (e.g. relaxation and redistribution of stress, temperature decrease, etc.), contribute to the loading behaviour and are included in the calculated healing index; however, these results show that the healing ability decreases with the accumulated damage. Also, the differences in healing indexes values between the first and the second rest periods were larger for the aged bitumens (BR and BP) in comparison with the unaged bitumen, which further demonstrates the detrimental effect of oxidative ageing on the fatigue resistance of bituminous materials.

Oxidative ageing and bio-oil have opposite effects on the stiffness of the bitumen. Hence, to understand the relationship between the bitumen's stiffness modulus and the healing ability, Fig. 16 plots one variable against the other, and the results were fitted with an exponential model. Thus, with the exception of *HIm* in the first rest period (HIm_1), the healing index is mostly influenced by the complex shear modulus, and this relationship increases with the rest periods (or the accumulated damage). Obviously, these models are material-specific, and it should be investigated the variation with the bitumen and rejuvenator properties.

5. Conclusions

This paper presents a study that investigated the healing behaviour of bitumen modified with bio-oil derived from biodiesel production. One base bitumen in three different ageing stages (unaged, short and long-term ageing) was used for adding up to 3 % of bio-oil. In addition to consistency and rheological tests to characterise the modified bitumen, fatigue resistance tests with and without rest periods were performed to assess the healing capacity through three indexes, relating the complex shear modulus, the number of cycles to failure and the amount of damage, after a rest period.

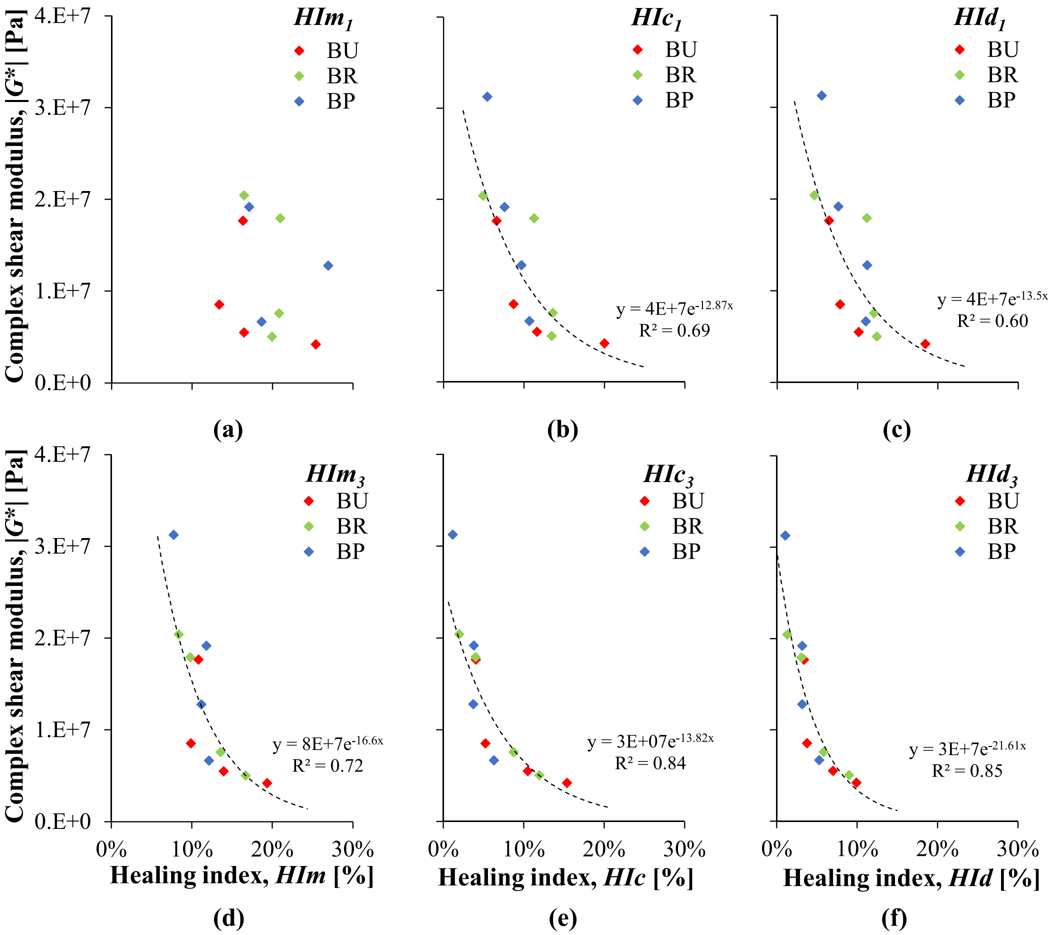


Fig. 16. Complex shear modulus versus healing indexes: (a) *HIm*$_1$; (b) *HIc*$_1$; (c) *HId*$_1$; (d) *HIm*$_3$; (e) *HIc*$_3$; (f) *HId*$_3$;

From the general characterisation of the bitumen modified with bio-oil, it was possible to conclude that: i) the addition of the bio-oil produced a binder softening, expressed by the increase of the penetration and a decrease in the softening point for all stages of ageing considered (1 % bio-oil increased 30 % the penetration and decreased 2 °C the softening point); ii) in terms of the rheological behaviour, the bio-oil produced a decrease of the complex shear modulus and an increase of the phase angle, function of the bio-oil content and ageing induced (for 3 % bio-oil, $|G^*|$ reduced 77 % and δ increased 18–30 % depending on the ageing level of the bitumen); iii) LAS test results proved the increase of the binder hardening with the ageing that was reversed with the addition of the bio-oil. Regarding fatigue resistance, the bio-oil allows its improvement, as proved by the area below the C-S curves.

Regarding the healing of the binder modified with bio-oil, it is possible to state that the healing indexes considered in this work follow an exponential law with the duration of the rest period. However, the number of cycles in each loading period decreased compared to the previous period, meaning that the damage was not fully recovered in the rest period. Depending on the healing index considered, the material restoration after a 15-minute rest period was limited to 26.9 %, meaning that the material integrity is not all recovered by adding the bio-oil. However, the bio-oil contributed significantly to that restoration by an increase of the restoration with the increase of the bio-oil added to the binder. For every 1 % increase in bio-oil, the average increase in healing index was 3.3 %, 2.4 % and 1.1 % for unaged, short- and long-term aged binders, respectively. The results of the bitumens in different ageing states also showed that the healing index is mostly influenced by the complex shear modulus, which the studied bio-oil is very effective in decreasing.

Bio-oils can serve both the purpose of recovering bitumen properties to allow its persistent use in pavement structures and of adopting more sustainable solutions in practice. However, further studies need to be conducted, such as examining how the studied bio-oil interacts with aggregate and filler in the mixture. Also, depending on the method used to incorporate bio-oils in pavement structures, adequate studies may be necessary to ensure that its constituents do not pose environmental risks in the long term.

CRediT authorship contribution statement

Marina Cabette: Writing – original draft, Investigation, Formal analysis. **Rui Micaelo:** Writing – review & editing, Validation, Supervision, Methodology, Investigation, Conceptualization. **Jorge Pais:** Writing – review & editing, Validation, Supervision, Resources, Methodology, Investigation, Conceptualization.

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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References

[1] D. Llopis-Castelló, T. García-Segura, L. Montalbán-Domingo, A. Sanz-Benlloch, E. Pellicer, Influence of pavement structure, traffic, and weather on urban flexible pavement deterioration, Sustainability 12 (2020) 9717, https://doi.org/10.3390/su12229717.

[2] C. Santos, M. Cabette, J. Pais, V. Carvalho, P. Pereira, Assessing self-healing asphalt by the heating of asphalt mixtures, Lect. Notes Civil Eng. 76 (2020) 253–261, https://doi.org/10.1007/978-3-030-48679-2_25.

[3] C.R. Santos, M.M. Cabette, J.C. Pais, P.A. Pereira, The Heating and Compaction of Asphalt Mixtures as a Self-healing Mechanism, in: H. Di Benedetto, H. Baaj, E. Chailleux, G. Tebaldi, C. Sauzéat, S. Mangiafico (Eds.), Proceedings of the RILEM International Symposium on Bituminous Materials, Springer International Publishing, Cham, 2022: pp. 1799–1805. https://doi.org/10.1007/978-3-030-46455-4_228.

[4] J. Ali, S.B. Dhobale, A Study on aging Behavior of paving Grade Bitumen using Filler Material, (2018) 1370–1375.

[5] Y. Hu, W. Si, X. Kang, Y. Xue, H. Wang, T. Parry, G.D. Airey, State of the art: Multiscale evaluation of bitumen ageing behaviour, Fuel 326 (2022), 125045, https://doi.org/10.1016/j.fuel.2022.125045.

[6] M. Iwański, M. Cholewińska, G. Mazurek, Impact of the Ageing on Viscoelastic Properties of Bitumen with the Liquid Surface Active Agent at Operating Temperatures, IOP Conf. Ser.: Mater. Sci. Eng. 245 (2017), 032049, https://doi.org/10.1088/1757-899X/245/3/032049.

[7] H. Wang, Z. Ma, X. Chen, M.R. Mohd Hasan, Preparation process of bio-oil and bio-asphalt, their performance, and the application of bio-asphalt: A comprehensive review, J. Traffic Transp. Eng. (English Ed.) 7 (2020) 137–151, https://doi.org/10.1016/j.jtte.2020.03.002.

[8] Z. Zhang, Y. Fang, J. Yang, X. Li, A comprehensive review of bio-oil, bio-binder and bio-asphalt materials: Their source, composition, preparation and performance, J. Traffic Transp. Eng. (English Ed.) 9 (2022) 151–166, https://doi.org/10.1016/j.jtte.2022.01.003.

[9] M. Zahoor, S. Nizamuddin, S. Madapusi, F. Giustozzi, Recycling asphalt using waste bio-oil: A review of the production processes, properties and future perspectives, Process Saf. Environ. Prot. 147 (2021) 1135–1159, https://doi.org/10.1016/j.psep.2021.01.032.

[10] J. Jiang, Z. Leng, B. Yang, G. Lu, Z. Tan, M. Han, Z. Dong, Penetration mechanism of the emulsion-based rejuvenator in damaged porous asphalt mixture: Microstructure characterization and 3D reconstruction, Mater. Des. 221 (2022), 111014, https://doi.org/10.1016/j.matdes.2022.111014.

[11] T. Ribeiro, A.C. Freire, M. Sá-da-Costa, J. Canejo, V. Cordeiro, R. Micaelo, Investigating asphalt self-healing with colorless binder and pigmented rejuvenator, Sustainability 15 (2023) 4556, https://doi.org/10.3390/su15054556.

[12] I.P. Pérez, A.M. Rodríguez Pasandín, J.C. Pais, P.A. Alves Pereira, Use of lignin biopolymer from industrial waste as bitumen extender for asphalt mixtures, J. Clean. Prod. 220 (2019) 87–98, https://doi.org/10.1016/j.jclepro.2019.02.082.

[13] A. Weir, A. Jiménez Del Barco Carrión, C. Queffélec, B. Bujoli, E. Chailleux, C.N. Uguna, C. Snape, G. Airey, Renewable binders from waste biomass for road construction: A review on thermochemical conversion technologies and current developments, Constr. Build. Mater. 330 (2022) 127076. https://doi.org/10.1016/j.conbuildmat.2022.127076.

[14] C. Chen, J. Lu, T. Ma, Y. Zhang, L. Gu, X. Chen, Applications of vegetable oils and their derivatives as Bio-Additives for use in asphalt binders: A review, Constr. Build. Mater. 383 (2023), 131312, https://doi.org/10.1016/j.conbuildmat.2023.131312.

[15] M. Zadshir, D.J. Oldham, S. Hosseinnezhad, E.H. Fini, Investigating bio-rejuvenation mechanisms in asphalt binder via laboratory experiments and molecular dynamics simulation, Constr. Build. Mater. 190 (2018) 392–402, https://doi.org/10.1016/j.conbuildmat.2018.09.137.

[16] X. Zhang, K. Zhang, C. Wu, K. Liu, K. Jiang, Preparation of bio-oil and its application in asphalt modification and rejuvenation: A review of the properties, practical application and life cycle assessment, Constr. Build. Mater. 262 (2020), 120528, https://doi.org/10.1016/j.conbuildmat.2020.120528.

[17] M. Zargar, E. Ahmadinia, H. Asli, M.R. Karim, Investigation of the possibility of using waste cooking oil as a rejuvenating agent for aged bitumen, J. Hazard. Mater. 233–234 (2012) 254–258, https://doi.org/10.1016/j.jhazmat.2012.06.021.

[18] Z. Sun, J. Yi, Y. Huang, D. Feng, C. Guo, Properties of asphalt binder modified by bio-oil derived from waste cooking oil, Constr. Build. Mater. 102 (2016) 496–504, https://doi.org/10.1016/j.conbuildmat.2015.10.173.

[19] Z. Sun, J. Yi, Y. Huang, D. Feng, C. Guo, Investigation of the potential application of biodiesel by-product as asphalt modifier, Road Mater. Pavement Design. 17 (2016) 737–752, https://doi.org/10.1080/14680629.2015.1096819.

[20] M. Gong, J. Yang, J. Zhang, H. Zhu, T. Tong, Physical–chemical properties of aged asphalt rejuvenated by bio-oil derived from biodiesel residue, Constr. Build. Mater. 105 (2016) 35–45, https://doi.org/10.1016/j.conbuildmat.2015.12.025.

[21] S. Ren, X. Liu, Y. Gao, R. Jing, P. Lin, S. Erkens, H. Wang, Molecular dynamics simulation and experimental validation on the interfacial diffusion behaviors of rejuvenators in aged bitumen, Mater. Des. 226 (2023), 111619, https://doi.org/10.1016/j.matdes.2023.111619.

[22] J. Gao, H. Wang, Z. You, M.R.M. Hasan, Y. Lei, M. Irfan, Rheological behavior and sensitivity of wood-derived bio-oil modified asphalt binders, Appl. Sci. (Switzerland) 8 (2018), https://doi.org/10.3390/app8060919.

[23] R. Zhang, Z. You, H. Wang, M. Ye, Y. Khin, C. Si, The impact of bio-oil as rejuvenator for aged asphalt binder, Constr. Build. Mater. 196 (2019) 134–143, https://doi.org/10.1016/j.conbuildmat.2018.10.168.

[24] B. Asadi, N. Tabatabaee, R. Hajj, Use of linear amplitude sweep test as a damage tolerance or fracture test to determine the optimum content of asphalt rejuvenator, Constr. Build. Mater. 300 (2021), 123983, https://doi.org/10.1016/j.conbuildmat.2021.123983.

[25] E. Gaudenzi, F. Cardone, X. Lu, F. Canestrari, Analysis of fatigue and healing properties of conventional bitumen and bio-binder for road pavements, Materials 13 (2020) 420, https://doi.org/10.3390/ma13020420.

[26] G. Sun, B. Li, D. Sun, J. Zhang, C. Wang, X. Zhu, Roles of aging and bio-oil regeneration on self-healing evolution behavior of asphalts within wide temperature range, J. Clean. Prod. 329 (2021), 129712, https://doi.org/10.1016/j.jclepro.2021.129712.

[27] R. Varma, R. Balieu, N. Kringos, A state-of-the-art review on self-healing in asphalt materials: Mechanical testing and analysis approaches, Constr. Build. Mater. 310 (2021), 125197, https://doi.org/10.1016/j.conbuildmat.2021.125197.

[28] G. Leegwater, A. Taboković, O. Baglieri, F. Hammoum, H. Baaj, Terms and Definitions on Crack-Healing and Restoration of Mechanical Properties in Bituminous Materials, RILEM Bookseries. 27 (2022) 47–53, https://doi.org/10.1007/978-3-030-46455-4_6.

[29] L. Shan, Y. Tan, Y. Richard Kim, Establishment of a universal healing evaluation index for asphalt binder, Constr. Build. Mater. 48 (2013) 74–79, https://doi.org/10.1016/j.conbuildmat.2013.06.039.

[30] E. Santagata, O. Baglieri, D. Dalmazzo, L. Tsantilis, Investigating cohesive healing of asphalt binders by means of a dissipated energy approach, Int. J. Pavement Res. Technol. 10 (2017) 403–409, https://doi.org/10.1016/j.ijprt.2017.06.004.

[31] L. Zhou, W. Huang, Y. Zhang, Q. Lv, C. Yan, Y. Jiao, Evaluation of the adhesion and healing properties of modified asphalt binders, Constr. Build. Mater. 251 (2020), 119026, https://doi.org/10.1016/j.conbuildmat.2020.119026.

[32] W. Xie, C. Castorena, C. Wang, Y. Richard Kim, A framework to characterize the healing potential of asphalt binder using the linear amplitude sweep test, Constr. Build. Mater. 154 (2017) 771–779, https://doi.org/10.1016/j.conbuildmat.2017.08.021.

[33] O. Baglieri, L. Tsantilis, E. Santagata, Evaluation of healing potential of bituminous binders using a viscoelastic continuum damage approach, Constr. Build. Mater. 184 (2018) 344–350, https://doi.org/10.1016/j.conbuildmat.2018.05.228.

[34] E. Gaudenzi, F. Cardone, X. Lu, F. Canestrari, Analysis of fatigue and healing properties of conventional bitumen and bio-binder for road pavements, Materials 13 (2020), https://doi.org/10.3390/ma13020420.

[35] Z. Lei, H. Bahia, T. Yi-qiu, C. Ling, Effects of refined waste and bio-based oil modifiers on rheological properties of asphalt binders, Constr. Build. Mater. 148 (2017) 504–511, https://doi.org/10.1016/j.conbuildmat.2017.05.101.

[36] R. Zhang, H. Wang, J. Gao, Z. You, X. Yang, High temperature performance of SBS modified bio-asphalt, Constr. Build. Mater. 144 (2017) 99–105, https://doi.org/10.1016/j.conbuildmat.2017.03.103.

[37] R. Zhang, H. Wang, X. Jiang, Z. You, X. Yang, M. Ye, Thermal Storage stability of bio-oil modified asphalt, J. Mater. Civ. Eng. 30 (2018), https://doi.org/10.1061/(ASCE)MT.1943-5533.0002237.

[38] J. Pais, C.R. Santos, M. Cabette, L. Hilliou, J. Ribeiro, H. Wang, M. Rosli, M. Hasan, Feasibility of using bio-oil from biodiesel production for bio-bitumen creation, (2023). https://doi.org/10.1080/14680629.2023.2180305.

[39] Cen, En 12607–1: Bitumen and bituminous binders - Determination of the resistance to hardening under the influence of heat and air - Part 1: RTFOT method, European Committee for Standardization, Brussels, 2014.

[40] Cen, En 14769: Bitumen and bituminous binders - Accelerated long-term ageing conditioning by a Pressure Ageing Vessel (PAV), European Committee for Standardization, Brussels, 2012.

[41] Cen, En 1426: 2015 . Bitumen and bituminous binders – Determination of needle penetration, European Committee for Standardization, Brussels, Belgium, 2015.

[42] Cen, En 1427: 2015 . Bitumen and bituminous binders – Determination of the softening point: Ring and Ball method, European Committee for Standardization, Brussels, Belgium, 2015.

[43] Aashto, TP 101–14: Estimating Damage Tolerance of Asphalt Binders Using the Linear Amplitude Sweep, American Association of State Highway and Transportation Officials, Washington, D.C., 2014.

[44] F. Canestrari, A. Virgili, A. Graziani, A. Stimilli, Modeling and assessment of self-healing and thixotropy properties for modified binders, Int. J. Fatigue 70 (2015) 351–360, https://doi.org/10.1016/j.ijfatigue.2014.08.004.

[45] G. Mazzoni, A. Stimilli, F. Cardone, F. Canestrari, Fatigue, self-healing and thixotropy of bituminous mastics including aged modified bitumens and different filler contents, Constr. Build. Mater. 131 (2017) 496–502, https://doi.org/10.1016/j.conbuildmat.2016.11.093.

[46] A. Stimilli, G. Ferrotti, C. Conti, G. Tosi, F. Canestrari, Chemical and rheological analysis of modified bitumens blended with “artificial reclaimed bitumen”, Constr.

Build. Mater. 63 (2014) 1–10, https://doi.org/10.1016/j.conbuildmat.2014.03.047.

[47] D. Lesueur, The colloidal structure of bitumen: Consequences on the rheology and on the mechanisms of bitumen modification, Adv. Colloid Interface Sci. 145 (2009) 42–82, https://doi.org/10.1016/j.cis.2008.08.011.

[48] A. Padmarekha, J.M. Krishnan, Viscoelastic Transition of Unaged and Aged Asphalt, J. Mater. Civ. Eng. 25 (2013) 1852–1863, https://doi.org/10.1061/(ASCE)MT.1943-5533.0000734.

[49] H. Bahia, H.A. Tabatabaee, T. Mandal, A. Faheem, Field Evaluation of Wisconsin Modified Binder Selection Guidelines - Phase II, Wisconsin Highway Research Program Project, University of Wisconsin Madison. (2013) 13–86.

[50] R. Micaelo, A. Pereira, L. Quaresma, M.T. Cidade, Fatigue resistance of asphalt binders: Assessment of the analysis methods in strain-controlled tests, Constr. Build. Mater. 98 (2015) 703–712, https://doi.org/10.1016/j.conbuildmat.2015.08.070.

[51] H. Zhang, K. Shen, G. Xu, J. Tong, R. Wang, D. Cai, X. Chen, Fatigue resistance of aged asphalt binders: An investigation of different analytical methods in linear amplitude sweep test, Constr. Build. Mater. 241 (2020), 118099, https://doi.org/10.1016/j.conbuildmat.2020.118099.

[52] F. Safaei, C. Castorena, Y.R. Kim, Linking asphalt binder fatigue to asphalt mixture fatigue performance using viscoelastic continuum damage modeling, Mech. Time-Depend. Mater. 20 (2016) 299–323, https://doi.org/10.1007/s11043-016-9304-1.

[53] A. Pereira, R. Micaelo, L. Quaresma, M.T. Cidade, Evaluation of Different Methods for the Estimation of the Bitumen Fatigue Life with DSR Testing, in: F. Canestrari, M.N. Partl (Eds.), 8th RILEM International Symposium on Testing and Characterization of Sustainable and Innovative Bituminous Materials, Springer, Netherlands, Dordrecht, 2016, pp. 1017–1028, https://doi.org/10.1007/978-94-017-7342-3_81.

[54] C. Wang, C. Castorena, J. Zhang, Y.R. Kim, Unified failure criterion for asphalt binder under cyclic fatigue loading, Road Mater. Pavement Design 16 (2015) 125–148, https://doi.org/10.1080/14680629.2015.1077010.

[55] R. Micaelo, F.P. Jiménez, R. Botella, R. Miró, A. Martínez, M.S. da Costa, On the analysis of uniaxial cyclic strain sweep test results to bituminous binders: stiffness-phase angle curve, Mater Struct. 55 (2022) 90, https://doi.org/10.1617/s11527-022-01936-6.

[56] C. Carl, P. Lopes, M. Sá da Costa, G. Canon Falla, S. Leischner, R. Micaelo, Comparative study of the effect of long-term ageing on the behaviour of bitumen and mastics with mineral fillers, Constr. Build. Mater. 225 (2019) 76–89, https://doi.org/10.1016/j.conbuildmat.2019.07.150.

[57] J. Petersen, A Review of the Fundamentals of Asphalt Oxidation. Chemical, Physicochemical, Physical Property, and Durability Relationships., Transportation Research Board, Washington, D.C, 2009.