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Effect of bio heating oil from biodiesel production on rheological behaviour of bitumen

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ABSTRACT: Bio-materials can replace traditional binders and additives used in the paving industry, which can help to build more resilient transport infrastructures and increase the consumption of secondary materials. Bio Heating Oil (BHO) is a residue of biodiesel production from waste cooking oil and animal fats. It has physical and chemical properties that allow the paving bitumen to improve binder-aggregate adhesion or rejuvenate aged bitumen. The objectives of this work are the study of the rheological and performance characteristics of bio-oil-modified bitumen. A 35/50 paving grade bitumen was modified with five contents of BHO (1, 2, 3, 5, and 10%). The bitumen was studied in unaged and aged conditions to evaluate the BHO ability to act as a rejuvenator of aged bitumen. Ageing treatment was induced by the Rolling Thin Film Oven and Pressure Ageing Vessel protocols. Physical and rheological tests were performed on unaged and aged bitumen. Physical tests include penetration, softening point and viscosity. Rheological behaviour was characterised through strain and frequency sweep tests at different temperatures. Viscoelastic Continuum Damage model was used to analyse the fatigue performance.

Keywords: Bio Heating Oil, Short-term ageing, Long-term ageing, Rejuvenator, Rheological test, Viscoelastic continuum damage

1 INTRODUCTION

The chemical composition of bitumen includes Saturates, Aromatics, Resins, and Asphaltenes (SARA) chemical groups. It can be considered a two-phase material with a liquid phase, called maltenes (saturates, aromatics, and resins), and a solid phase, called asphaltenes (Brovelli et al. 2013). When bitumen starts to be aged (liquid phase oxidised), the bitumen becomes dry and brittle (García et al., 2010; Alakhrass, 2018). Rejuvenators are used to restore the ratio of asphaltenes to maltenes of aged bitumen (García, Schlangen, and van de Ven 2011). The bio heating oil (BHO) is the residue obtained from the distillation of waste cooking oil and animal fats in biodiesel production (Santos et al. 2020). It is composed of less volatile organics and contains some residual Fatty Acid Methyl Ester (FAME).

Researchers have evaluated the effects of some bio-oils as a rejuvenator of aged bitumen. Zargar et al. (2012) and Sun et al. (2016) investigate the use of waste cooking oil as an aged bitumen rejuvenator. Zhang et al. (2018) used wasted wood bio-oil in their

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research. Zeng et al. (2018) carried out studies using residue in castor oil, and Zhang et al. (2019) used sawdust bio-oil as a bio-based rejuvenator. It was proven that bio-oil softens aged binders.

Overall, it was found that bio-oil derived from several biomass sources can successfully be used as a bitumen modifier, and it is expected bio-oil to soften the bitumen (Lei et al. 2017; Lei et al. 2018; Zhang, Wang, et al., 2018; Zhang et al., 2019).

Asadi, Tabatabaee and Hajj (2021) concluded that the stiffness of the bitumen could be reduced with low rejuvenator content that may not produce a significant effect on the failure. Some researchers have determined the optimum rejuvenator content based on the penetration or dynamic viscosity of the binder. In contrast, other researchers have investigated rejuvenated binder performance according to the Superpave criteria, Linear Amplitude Sweep (LAS), or Multiple Stress Creep Recovery (MSCR) tests.

The primary purpose of modifiers is to improve the rheological properties of bitumen binders and increase their resistance to fatigue cracking (Hassanpour-Kasanagh et al. 2020). So, this study aimed to (1) investigate the effects of BHO on the rejuvenation of aged bitumen, (2) analyse the fatigue performance of BHO-modified bitumen, and (3) determine the better BHO content to be used in aged bitumen.

2 MATERIALS AND METHODS

2.1 *Materials*

The study was carried out using a typical paving bitumen, namely a 35/50 pen bitumen which properties are presented in Table 1 in terms of needle penetration, softening point and viscosity. It is a semi-hard bitumen used for hot climatic or heavy traffic conditions.

Table 1. Properties of 35/50 penetration grade bitumen (EN 1426; EN 1427; EN 13302).

Property	Unit	Specification	Measured
Penetration at 25ºC	0.1 mm	35-50	36.6
Softening point	ºC	50-58	53.2
Complex Viscosity at 150ºC	Pa.s	–	0.68

The BHO used in this study resulted from biodiesel production and presented high viscosity due to the reduced amount of FAME and high glycerol. The presence and amount of these two components in the BHO depend on the quality and proportion of used cooking oil and animal fats used in biodiesel production.

To be used at room temperature, 50% of FAME was added to the BHO to produce the rejuvenator/modifier used in this study, which allowed the viscosity presented in Figure 1.

The rejuvenator was applied to three types of samples: unaged, short-term aged, and long-term aged bitumens. Short-term ageing was simulated through the Rolling Thin Film Oven (RTFO) (EN 12607-1), while the long-term ageing was simulated by the Pressure Ageing Vessel (PAV) (EN 14769). The RTFO simulates the binder aged after the mixing, transporting, and compacting processes, and the PAV simulates the long-term ageing equivalent to 5–10 years of in-service pavements.

Unaged bitumen is referred to as BU, RTFO aged bitumen is referred to as BR and PAV aged bitumen is referred to as BP. For each of these bitumens, 0, 1, 2, 3, 5 and 10% of rejuvenator was added, producing 15 binders. Thus, the previous terminology is followed by the rejuvenator content in the sample. For example, an unaged bitumen with 3% of rejuvenator is referred to as BU3.

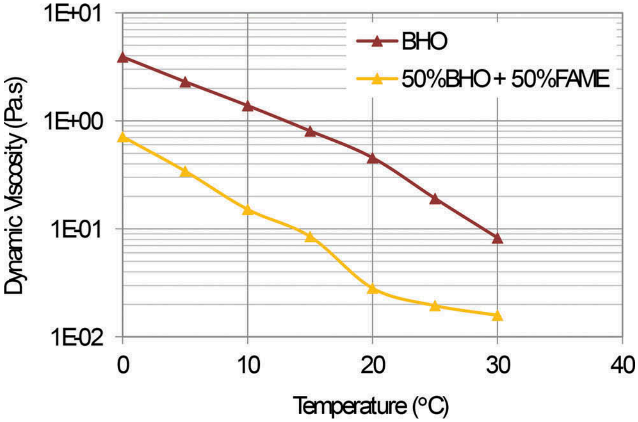


Figure 1. Rotational viscosity of the rejuvenator.

Samples were prepared in a low-shear mixer by blending the bitumen and rejuvenator under 150°C for 5 minutes at 350 rpm.

2.2 *Test methods*

2.2.1 *Physical property tests*

The physical properties of bitumen samples were investigated using needle penetration, ring and ball and dynamic viscosity tests. The needle penetration and ring and ball tests were performed following the European standard EN 1426 and EN 1427, respectively. The dynamic viscosity test was measured every 10°C from 100°C to 160°C, following EN 13302.

2.2.2 *Linear amplitude sweep test*

The mechanical behaviour was evaluated using the Linear Amplitude Sweep (LAS) test consisting of two phases. The first one is a frequency sweep test to determine rheological properties to determine the damage parameter. Then, a second phase consists of a series of oscillatory load cycles at linearly increasing amplitudes at a constant frequency to cause accelerated fatigue damage. The continuum damage approach is used to calculate the fatigue resistance from rheological properties and amplitude sweep results. Results from the LAS test can be used in Viscoelastic Continuum Damage (VECD) model. AASHTO-TP101 presents the test method and VECD analysis. The LAS test was performed at 20 ºC.

3 RESULTS AND DISCUSSION

3.1 *Physical properties*

Penetration test results of unaged and aged bitumens are presented in Figure 2. Penetration increases with the rejuvenator content, meaning that the BHO acts as a bitumen rejuvenator. The evolution of penetration with the rejuvenator content follows an exponential law, with excellent correlation coefficients. It is possible to see the influence of the rejuvenator on the penetration. Based on the obtained results, the increase of penetration (Δpen) can be expressed as:

$$\Delta pen = e^{a \times \Delta RC} \tag{1}$$

where, Δpen is the increase of penetration (mm/10); e is the Euler number; ΔRC is the increase of rejuvenator content (%); a is a statistically determined coefficient. For 35/50 unaged and short-term aged bitumen, *a* = 0.30; for long-term aged bitumen, *a* = 0.27. On average, the penetration increases about 30% for each 1% rejuvenator added.

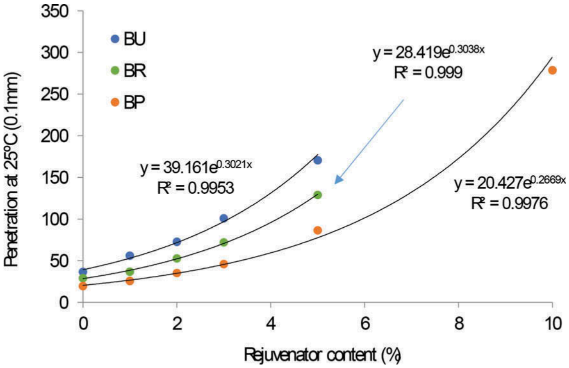


Figure 2. Penetration of the 35/50 bitumen with different rejuvenator content.

The softening point results of unaged and aged bitumens are shown in Figure 3, following a linear law with an excellent correlation coefficient. It is observed a decrease of the softening point when the rejuvenator content increases. On average, the softening point decreases 2.5 ºC for each 1% rejuvenator added to the long-term aged bitumen. The softening point decreasing ratio of the other bitumens is 2.3 ºC and 2.1 ºC for short-term ageing and unaged bitumen, respectively.

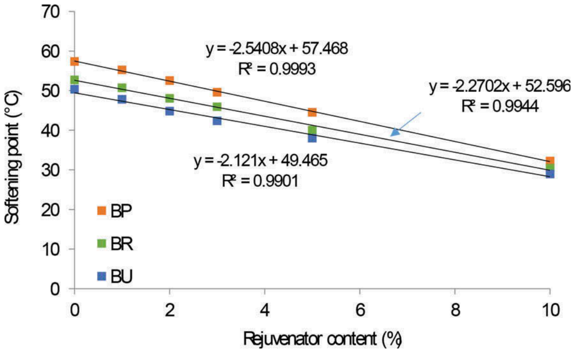


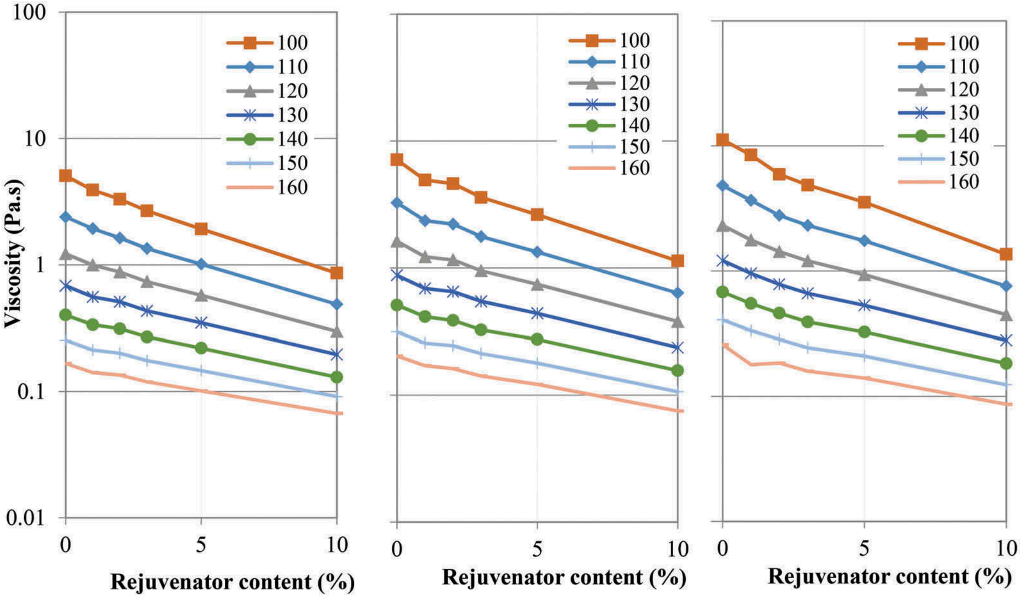
Figure 3. Softening point of the 35/50 bitumen with different rejuvenator content.

Therefore, the higher the dosage of rejuvenator is, the more significant the increase in penetration and the decrease in softening point. It occurs because the volatile component of the bitumen increased and, consequently, a reduction in its stiffness.

The viscosity test results are shown in Figure 4 against the rejuvenator content for temperatures ranging from 100 to 160 °C. These temperatures cover the production, laying and compaction of bituminous mixtures. Results show that the viscosity decreases with the addition of the rejuvenator and follows a perfect exponential law (a straight line in semi-log scale). Also, in a semi-log scale, the viscosity has a linear reduction with the temperature increase. These two trends allowed developing a model to predict the viscosity for 35/50 pen bitumen modified by BHO rejuvenator, as follows:

$$\text{Visco} = a \times e^{b \times RC} \times \left(\frac{t}{100}\right)^{c} \tag{2}$$

where, Visco is the dynamic viscosity in Pa.s; e is the Euler number; RC is the rejuvenator content in %; t is the temperature em °C; a, b, c are statistically determined coefficients. Based on this model (fitting coefficients listed in Table 2), one can conclude that the viscosity decreases about 15% per each 1% of added rejuvenator.



(a)

(b)

(c)

Figure 4. Viscosity of the (a) unaged; (b) short-term aged; and (c) long-term aged bitumen.

Table 2. Coefficients for Equation 2.

Bitumen	a	b	c	R^2
Unaged	3.7022	-0.1367	-6.4446	0.964
RTFO aged	4.8671	-0.1441	-6.5574	0.937
PAV aged	6.7283	-0.1495	-6.8431	0.884

3.2 *Rheological and fatigue resistance characterisation*

Figure 5 shows the black diagram (complex modulus G* versus phase angle δ) of the unaged bitumen (BU), short-term aged (BR), and long-term aged (BP) with different rejuvenator content obtained from the frequency sweep at low applied strain that precedes the damage induced strain sweep in LAS test. As expected, the complex modulus increased, and the phase decreased with the ageing treatment. Differently, the rejuvenator caused a reduction of the complex modulus and the increase of the phase angle. This means that the curves move to left and upwards with ageing and to right and downwards with the rejuvenator content added. However, it is noted that the rejuvenator affects more the stiffness modulus than the phase angle. Also, the bitumens with 10% rejuvenator show non-smooth curves that is an indication of non-linear effects and that these binders are more rheologically complex.

Table 3 shows the data of complex modulus G* and phase angle δ at 10Hz of the unaged bitumen (BU), short-term aged (BR), and long-term aged (BP) with different rejuvenator content. At 10 Hz, the rejuvenator content required to obtain the same level of complex modulus and phase angle of unaged bitumen is 1-2% and 2-3% with short- and long-term aged bitumens, respectively.

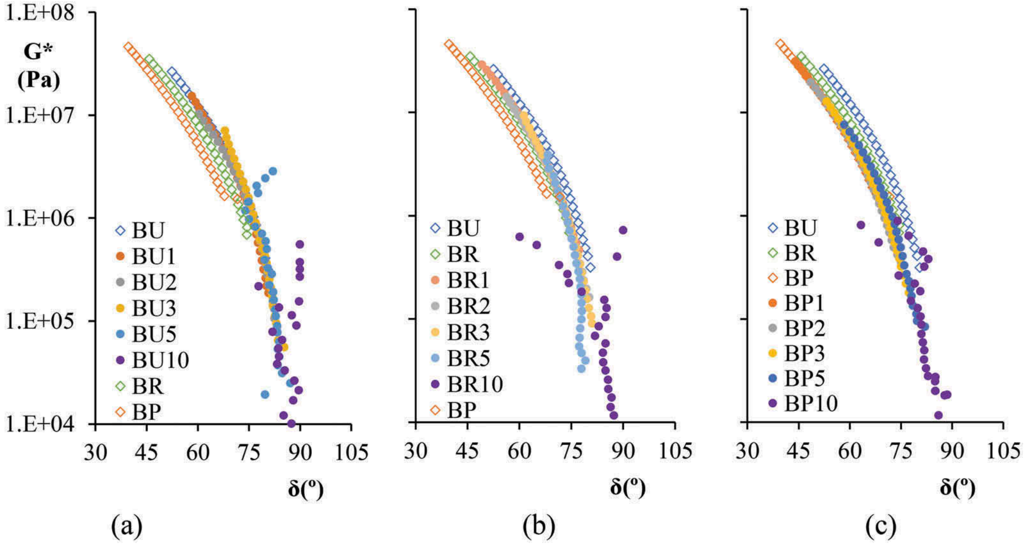


Figure 5. Black diagram of the unaged (BU); short-term aged (BR); and long-term aged (BP) bitumen.

Table 3. Complex Modulus (G*) and Phase Angle (δ) at 10Hz.

Sample	G* (Pa)	δ (°)	Sample	G* (Pa)	δ (°)	Sample	G* (Pa)	δ (°)
BU0	1.37E+07	58.95	BR0	1.95E+07	51.98	BP0	2.75E+07	45.22
BU1	7.50E+06	63.95	BR1	1.54E+07	55.63	BP1	1.76E+07	49.96
BU2	4.66E+06	67.27	BR2	7.04E+06	62.03	BP2	1.03E+07	54.77
BU3	3.16E+06	71.59	BR3	4.51E+06	65.76	BP3	6.57E+06	58.88
BU5	1.18E+06	74.00	BR5	1.74E+06	71.20	BP5	3.50E+06	65.10
BU10	1.55E+05	89.75	BR10	2.71E+05	73.94	BP10	3.80E+05	83.08

The shear stress versus shear strain of the samples (BU, BR and BP) is shown in Figure 6. It can be seen that stresses sustained by specimens during cyclic loading increase with ageing treatment and decrease with the rejuvenator, which is related with the specimens's stiffness. To determine the specimens's failure in the test is recommended in the literature (Bahia et al. 2013; Micaelo et al. 2015; Zhang et al. 2020) the adoption of the point at which the maximum stress level is attained. In Figure 6(a), the failure strain of BU0 is 8.4% and increases to 9.3%, 9.4%, 10.2%, 10.2% and 13% with the addition of 1%, 2%, 3%, 5% and 10% of rejuvenator, respectively. In Figure 6(b), the failure strain of BR0 is 7.4% and increases to 7.5%, 8.3%, 9.3%, 10% and 10% with increasing rejuvenator content. In Figure 6(c), the failure strain of BP0 is 7.3% and increases to 9.5%, 9.4%, 10.5%, 8.3% and 10.1% with increasing rejuvenator content. From this, it is concluded that ageing treatment made, as expected, the bitumen harder and less ductile, and this is reverted with the addition of bio-oil. The effect of the rejuvenator on the ductility of modified bitumen is especially noted for small rejuvenator contents (1-3%).

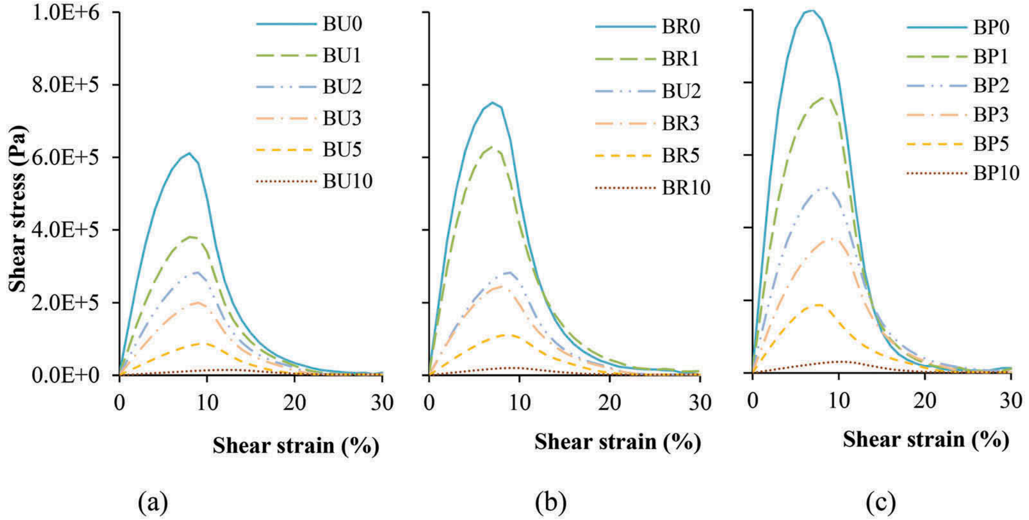


Figure 6. Stress-Strain curves (a) unaged (BU); (b) short-term aged (BR); and (c) long-term aged (BP) bitumen.

To assess the fatigue resistance of bitumens from LAS test the VECD methodology is adopted. In this analysis protocol are firstly determined the material integrity (C) versus damage (S) curves, also referred to as “damage characteristic curves”. Figure 7 shows the effect of the rejuvenator on the damage characteristic curves of bitumens aged to different levels. In the LAS test, the C value (initially 1.0) decreases with increasing loading cycles, and this is related with the growth of the S variable. It is observed that the S value required to attain the same level of C reduction is lower in aged bitumens, and increases with the rejuvenator content. A similar trend is seen with the three bitumens. To quantify the variation in C-S curves with both effects, it was determined the areas under the curves, which are shown in Table 4. The ageing treatment caused a reduction of about 22% in the C-S area, and 2% bio-oil is sufficient to attain the unaged bitumen level.

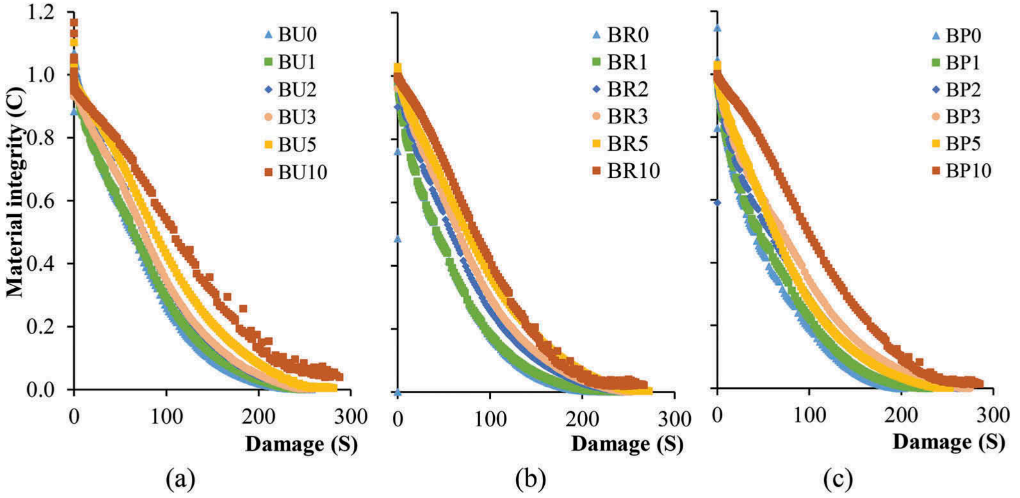


Figure 7. LAS: Material integrity (C) versus damage (S) for (a) unaged (BU); (b) short-term aged (BR); and (c) long-term aged (BP) bitumen.

Table 4. Area bellow 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
BU5	98	BR5	88	BP5	74
BU10	151	BR10	102	BP10	112

Then, the number of loading cycles (N_f) that the material can sustain at different loading amplitudes are calculated in the VECD methodology. Figure 8 compares the number of loading cycles for two different strain amplitudes (2.50% and 5.00%) of different bitumens. In general, and surprisingly, the fatigue resistance was greater for the long-term aged bitumen, with and without rejuvenator. On opposition, the short-term aged bitumen performed poorly than the unaged bitumen. The results are explained by the sensitivity of the method to the strain level at failure and the C(S) model. BP binders were stiffer than less aged binders (BU and BR), but the strain at failure did not change (reduction) as much as the complex modulus varied. Thus, the determined S value at the failure point was larger than in other bitumens. It is also noteworthy mentioning that the power-law model used to fit C-S curves did not fit well the full range of the curve for some bitumens. Nevertheless, it is observed for the three bitumens (BU, BR and BP) that a small addition of bio-oil increases the fatigue resistance. For the larger rejuvenation contents (5% and 10%), the bitumens performed differently.

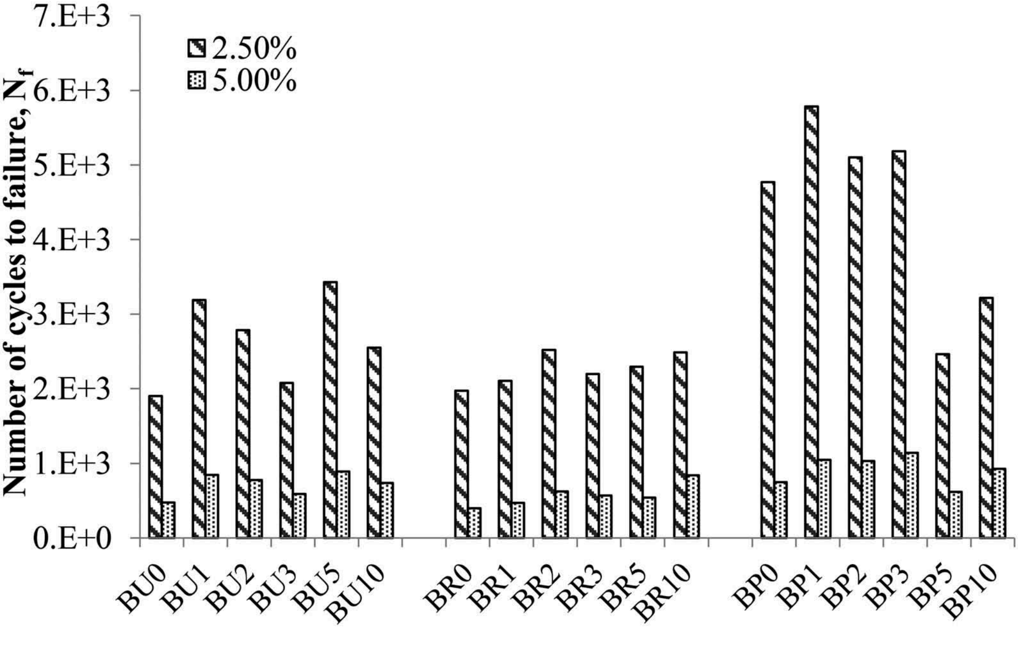


Figure 8. Fatigue resistance of bitumens at two strain levels (2.00% and 5.00%).

4 CONCLUSIONS

This paper aimed to investigate the effects of BHO as a rejuvenator on the physical and rheological properties of aged bitumen. The following conclusions can be drawn:

- The laboratory short-term and long-term ageing processes significantly changed the physical and rheological properties of the unaged bitumen, namely, BU0 that is a 35/50 bitumen grade, were changed after short-term ageing (BR0) to a 20/30 bitumen grade. The increase of the rejuvenator made the higher changed in the physical and rheological properties.
- Adding 1-3% of rejuvenator on the bitumen can restore the physical properties of the aged bitumen, namely, improve the penetration and reduce the softening point and viscosity. Their efficiency to restore the physical properties of the bitumen increase the complex modulus. So, it is also verified that the amount of rejuvenator added must be at least 1 to 3% after ageing to guarantee an adequate penetration value and allow good paving.
- Adding 1-2% rejuvenator in the bitumen, both complex modulus and phase angle of the long-term ageing can be restored to the level of short-term ageing without rejuvenator, which means the optimum dosage of the rejuvenator should be between 1 to 2%.
- Through VECD analyses, the addition of the rejuvenator into the bitumen can increase the fatigue life of the sample. The right amount of 2% until 3% of rejuvenator in the bitumen can increase material integrity.

Based on these findings, the selected amount of rejuvenator that can soften the aged bitumen to a required level, restore the physical properties, and increase the fatigue life, are 2% (by volume of bitumen).

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