Effects of aging on the damage tolerance of Polymer Modified Bitumens investigated through the LAS test and fluorescence microscopy.

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ABSTRACT

The effects of polymer modification on the durability of PMBs remain relatively unclear due to the partial understanding of the combined effects of aging and loading on the damage accumulation. This paper investigates the effects of aging on the cumulative resistance of PMBs by evaluating the damage tolerance of laboratory aged SBS and EVA modified bitumens. Results of Linear Amplitude Sweep (LAS) tests were corroborated with their morphology as determined using fluorescence microscopy. Aging increases the capacity of bitumens to maintain integrity with damage and reduces their time-temperature susceptibility. The damage tolerance derives from the combination of the two mechanisms as a function of the strain considered. Polymer modification influences the magnitude of this phenomenon. The damage tolerance of SBS modified bitumens is a function of polymer concentration, dispersion, and integrity of the polymer network. The EVA PMBs show low PAV-susceptibility, which contributes to increasing their damage tolerance at low strain levels.

Key words

Aging, Damage tolerance, PMB, LAS test, Fluorescence microscopy

1. INTRODUCTION

Long-term pavement performance (LTPP) needs to account for the combined effects of aging and loading (American Association of State Highway and Transportation Officials – AASHTO, 1993). Fatigue cracking is one of the major distresses that affects the LTPP of flexible pavements and depends on the accumulation of damage under repeated loads resulting in the formation and propagation of cracks in the compacted mixture (Molenaar, 2007). These cracks initiate and propagate either in the bitumen (bitumen-filler) phase or at the aggregate-bitumen interface and are influenced by the degree of aging of the bitumen. Oxidative aging results in the hardening of the bitumen and the subsequent loss of its strain tolerance. It is a long-term chemical process caused by the diffusion of oxygen molecules throughout the film of bitumen, which coats the aggregates in asphalt mixtures. Oxidation is responsible for changes in the microstructure of bitumen, leading to the formation of heavier and more polar molecules that have reduced mobility. This results in an increase in viscosity and stiffness, together with the loss of strain tolerance under repeated loading (Petersen, 1984, 2009; Page, Murphy, Ruth, & Roque, 1985). For this reason, oxidative effects should be considered in the evaluation of the fatigue resistance of bitumens.

Polymer additives have been used to improve the mechanical performance of bitumens, including their resistance to damage. Recently, polymers have also found applications as aging inhibitors of bitumen (Bahia et al., 2001; Zhu, Birgisson, & Kringos, 2014 Yan, Huang, Lv, & Lin, 2019; Yan, Huang, Zheng, Zhang, & Lin, 2020). Examples of typically used polymers include Styrene Butadiene Styrene (SBS), Ethylene Vinyl Acetate (EVA) and Styrene Butadiene Rubber (SBR) (Bahia et al., 2001). However, the effects of oxidative aging on the damage tolerance of polymer-modified bitumens (PMBs) still remain relatively unclear due to various reasons.

Firstly, PMBs are the result of complex thermodynamic interactions between the two phases (bitumen and polymer). These interactions occur at the macroscopic level and result in the polymer swelling in the bitumen phase prompted by the absorption of the light and low-polar bitumen fractions. Polymers capable of maintaining their structure during swelling positively influence the rheological response of PMBs, providing higher stiffness at high temperatures, lower stiffness at low temperatures, and reduced temperature susceptibility. The quality of modification depends on the compatibility between the polymer and the bitumen, which is a function of the type of polymer and the source and refining process of the bitumen (Polacco, Filippi, Merusi, & Stastna, 2015). Furthermore, the effects of cross-linking agents need to be considered (Mandal, Sylla, Bahia, & Barmand, 2015; Cuciniello et al., 2019). For these reasons, the definition of a unique aging mechanism is unclear since it is polymer-dependent.

Secondly, aging and damage produce opposite effects on bitumen stiffness. The first causes an increase in stiffness of the bitumen. The second can be defined as the "*Deterioration of internal microstructure of material (breaking of molecular bonding) with a consequent worsening of mechanical response due to the accumulation of micro-cracks and defects. Specifically, damage can be seen as a reduction in the stiffness of materials*" (Johnson, 2013). In terms of the rheological response of a bitumen, measurements using a dynamic shear rheometer (DSR) under linear viscoelastic (LVE) conditions demonstrate an increase in complex modulus (|G*|) and a decrease in phase angle indicating an increase in overall stiffness with aging. However, as the stiffness increases, aged pavements tend to be more susceptible to cracking (e.g., low temperature cracking, fatigue cracking) and, therefore, damage. There is evidence that some polymers (e.g., SBS) mitigate the effects of hardening of the bitumen phase on the rheological response of the PMB. The polymer backbone degrades under the thermo-oxidative processes occurring during production (thermal processes) and in the field (oxidation). Such degradation has been considered, in some cases, as beneficial since it provides a viscous effect that helps in compensating for the bitumen hardening, thus retaining the rheological properties with time (Lu, Soenen, Heyrman, & Redelius, 2013; Lu, Sandman, & Redelius, 2010; Lu & Isacsson, 2000; Cortizo, Larsen, Bianchetto, & Alessandrini, 2004; Airey & Brown, 1998). In recent studies, microscopy approaches have been used to evaluate the effects of aging on the polymer dispersion (Cuciniello et al., 2018; Mouillet, Lamontagne, Durrieu, Planche, & Lapalu, 2008; Handle et al., 2014). Findings from these studies provide two main interpretations. On one hand, polymers undergo thermo-oxidative degradation and reduce their dispersion in the bitumen-rich phase. This conclusion is supported by rheological findings and by measurements of molecular weight distribution (Cuciniello et al., 2018, 2019). The alternative hypothesis is represented by the de-swelling of polymer (such as SBS) during aging (Mouillet et al., 2019). That is, the polymer phase de-swells by desorbing part of the aromatic portion (having a marked fluorescence), which leaves the polymer phase due to the increase in polarity during oxidative aging. In this way, the morphology observed at the unaged condition disappears with the de-swollen polymer phase providing a lower effect on the rheological response.

Damage tolerance was developed as a design philosophy for aeronautic structures in the 1970s as an improvement on the fail-safe principle for structural deterioration; this approach is based on the principle that while cracks due to fatigue and corrosion will develop in the aircraft structure, the process can be understood and controlled. That is, damage-tolerant structures are designed to sustain cracks without catastrophic failure until the damage is detected in scheduled inspections, and the damaged part is repaired or replaced. The concept of damage tolerance has been extended in other fields of engineering; specifically, in the field of pavement engineering, damage tolerance can be considered as the property of a pavement related to its ability to sustain defect (cracks) until the repair can be accomplished.

Within this context, the LVE characterization of bitumens is unlikely to provide insight on the actual damage tolerance, especially in the case of PMBs. The viscous modulus ($|G^*|\sin\delta$) is currently adopted in the Superpave Performance Grading (AASHTO M320) to control the fatigue resistance of bitumens. The applicability of this indicator to PMBs has been challenged since the late '90s because it is measured in the linear region and at a single strain level. Due to their structure, polymers show strain dependency that is unlikely to be captured at low strain levels. Furthermore, using a single strain level (i.e., 1%) does not consider the pavement structure, which affects the strain domain in the bitumen. Non-linearity is truly relevant as strains in the bitumen can be appreciably higher and therefore potentially in the non-linear region even at low strain levels in the asphalt mixture and pavement (D'Angelo & Kluttz, 2007; Kose, Guler, Bahia, & Masad, 2000; Masad & Somadevan, 2002; Airey, Rahimzadeh, & Collop, 2004). Aside from the strain level, the LVE characterization is conducted at a low number of loading cycles, which does not allow the effects of thixotropic and energy dissipation in damage (i.e., micro-cracks) to be evaluated (Bahia, Zhai, Zeng, Hu, & Turner, 2002; Bahia, Zhai, Bonnetti, & Klose, 1999; Bahia, Perdomo, & Turner, 1999; Hintz, Velasquez, Johnson, and Bahia, 2011; Underwood, Baek, & Kim, 2012;).

Therefore, according to the approach of the damage tolerance, any investigation of the durability of bitumen should focus on how oxidative aging affects their damage tolerance. In order to achieve this, the rheological characterization should be conducted outside the linear range to assess damage tolerance. The Linear Amplitude Sweep (LAS) test was developed with this purpose and represents an accelerated fatigue test, which generates damage by increasing the strain amplitude over the loading cycles (Johnson, 2013; Hintz et al., 2011). Damage is observed as the reduction of the viscous modulus with loading cycles due to the formation and the propagation of cohesive cracks. Results are analyzed using Viscoelastic Continuum Damage (VECD) theory which represents a theoretical approach to the analysis of fatigue cracking in asphalt materials, including PMBs, and allows the number of cycles to failure at different strain levels with a single test to be predicted (Schapery, 1975, 1984, 1999; Kim, Y., Lee, Little, Kim, Y.R., 2006; Hintz, Velasquez, & Bahia, 2011; Safaei & Castorena, 2016: Riccardi, Cannone Falchetto, Witsuba, & Losa, 2017).

In order to investigate the durability of different PMBs, this paper investigates the effects of aging on the cumulative resistance of PMBs by analyzing their damage tolerance. Results of the LAS test were analyzed by using the VECD conceptual framework as detailed in AASHTO TP 101-12. Findings from the rheological test were corroborated by the qualitative analysis of the PMBs' microstructure observed by fluorescence microscopy.

2. MATERIALS and METHODS

2.1. Materials

Eight types of bitumen were prepared by combining one base bitumen (penetration grade 70/100) with a radial SBS copolymer (polystyrene content 29-31%) and a plastomer EVA (vinyl acetate content 28%). In the case of the SBS modified bitumens, sulfur (S) was also used as a cross-linking agent. The details of the eight bitumens are given in Table 1.

Table 1. Bitumens used in the experiment.

Bitumen	Polymer concentration (w/w) [%]	Sulphur (w/w/) [%]
Pen 70-100 (Pen)	0	-
Pen/S	0	0.1
SBS 2/S	2% SBS	0.1
SBS 4/S	4% SBS	0.1
SBS 6/S	6% SBS	0.1
EVA 2	2% EVA	-
EVA 4	4% EVA	-
EVA 6	6% EVA	-

The concentration of sulphur is 0.1% by weight of the blend (bitumen + polymer). An additional bitumen modified with sulphur (Pen/S) was prepared to highlight any potential effects of sulphur. Details of the bitumen preparation are given in (Cuciniello et al., 2018, 2019).

2.2. Bitumen aging

The bitumens were short-term aged by using the Rolling Thin Film Oven (RTFO) as per the UNI EN 12607-1 standard and long-term aged by using multiple cycles of the Pressure Aging Vessel (PAV) as per AASHTO R 28. Multiple cycles of PAV were applied to simulate different levels of oxidation (i.e., 20 hours – 1PAV; 40 hours – 2PAV; 80 hours – 4PAV).

2.3. Fluorescence microscopy

A Leica® DM LB microscope was used to capture images of the PMB morphology at 10X magnification. The microscope is connected to a camera to gather pictures of the various samples. Details about sample preparation are given in (Cuciniello et al., 2018, 2019).

2.4. LAS test

2.4.1. Test method

The LAS test was conducted as per AASHTO TP 101 – 12 using an Anton Paar MCR 301 Dynamic Shear Rheometer (DSR) with the 8 mm parallel plate geometry. The test includes a frequency sweep (logarithmic ramp) to determine the undamaged properties in the LVE region; and a strain sweep, which induces damage by increasing the strain level applied (linear ramp) at different loading cycles. Test conditions are given in Table 2.

Table 2. LAS test conditions

Frequency Sweep portion	
Strain [%]	1.0
Frequency [Hz]	0.1 to 30
Strain Sweep portion	
Strain [%]	0.1 to 30
Frequency [Hz]	10
Loading cycles	3,100

2.4.2. Analysis of data

The LAS test data were analyzed according to the VECD theory included in AASHTO TP 101-12. The application of the VECD theory is based on the work of potential theory (Schapery, 1975) in which the damage accumulation rate follows a Paris's Law of the variation of the strain energy versus the damage intensity (Equation (1)).

$$\frac{dD}{dt} = \left(-\frac{dW}{dD}\right)^{\alpha} \tag{1}$$

The term on the left side represents the damage (D) accumulation rate. The term on the right side is the variation of the strain energy (W) with damage intensity. The exponent of the power-law α determines the energy release rate, and it is measured in the linear region. This parameter is given by the inverse of the slope (m) of the isotherm of the logarithm of the storage modulus (G') versus the logarithm of the frequency obtained from the frequency sweep test. Parameter m is a function of the time-temperature susceptibility of the material.

In viscoelastic materials, damage results in changes in the dissipated strain energy, which, in the case of strain (γ_0) - controlled phenomenon, is proportional to the viscous modulus $|G^*|\sin\delta$ (Equation (2)).

$$W_d = \pi \cdot \gamma_0^2 \cdot G'' = \pi \cdot \gamma_0^2 \cdot |G^*|\sin\delta \tag{2}$$

Where $|G^*|$ is the complex shear modulus, and δ is the phase angle.

Therefore, the loss in material integrity (damage) can be represented by a reduction in the viscous modulus 0Kim et al., 2014). The damage intensity can be calculated at each loading cycle of the strain sweep by using Equation (3).

$$D(t_N) \cong \sum_{i=1}^{N} \left[\pi \cdot I_D \cdot \gamma_0^2 \left(|G^*|\sin\delta_{i-1} - |G^*|\sin\delta_i\right)\right]^{\frac{\alpha}{1+\alpha}} (t_i - t_{i-1})^{\frac{1}{1+\alpha}} \quad (3)$$

Where:

- N – number of data point (3,100);
- I_D – initial value of $|G^*|\sin\delta$ as proposed by Johnson (2013);
- t – is the testing time [sec];
- γ_0 – is the applied strain for a given data point [%];
- α - meaning clarified with Equation (1).

Experimental data can be used to fit a power-law model of $|G^*|\sin\delta$ versus D given by Equation (4) (Kim et al., 2014).

$$|G^*|sin\,\delta = C_0 - C_1(D(t))^{C_2} \quad (4)$$

Where C_0 is the average value of $|G^*|sin\,\delta$ during the 0.1 % strain interval, and (C_1, C_2) are the curve-fit coefficients derived from the linearization of the power-law form (Equation (4)) in the form suggested by (Hintz, Velasquez, Johnson, et al., 2011) and shown in Equation (5).

$$\log\left(C_0 - |G^*|sin\,\delta(t)\right) = \log C_1 + C_2 \log\left(D(t)\right) \quad (5)$$

After the calculation of the parameters (C_1, C_2) the derivative concerning the damage intensity (D) of Equation (4) can be computed and included in Equation (1). A closed-form of the solution provides the relation between the number of cycles to failure and the strain amplitude (γ_{max}) at a defined failure criterion (Equation (6)).

$$N_f = \frac{f(D_f)^k}{k(\pi I_D C_1 C_2)^\alpha} (\gamma_{max})^{-2\alpha} \quad (6)$$

Where:

- $k = 1 + (1 - C_2)\alpha$;
- f – is the loading frequency during the amplitude sweep portion (10 Hz);

- $D_f = \left(\frac{0.35 \cdot C_0}{C_1}\right)^{1/C_2}$ - is the damage intensity corresponding to a reduction of 35% of the initial viscous modulus (35% is selected as the criterion, according to Johnson, 2013).

Equation (6) can be simplified to achieve the typical form of the fatigue law of bituminous materials (Wohler curve – Equation (7)) (Monismith, Epps, Kasianchuk, & McLean, 1970).

$$N_f = A(\gamma_{max})^B \tag{7}$$

Where

$$A = \frac{f(D_f)^k}{k(\pi I_D C_1 C_2)^\alpha} \tag{8}$$

Which represents the value of N_f at a strain level of 1%. It depends on material integrity (i.e.,$|G^*|$ sin δ) versus the damage curve (D), on the initial undamaged modulus (I_D), and the criterion selected as a failure. And where

$$B = -2\alpha = -2 \cdot \frac{1}{m} \tag{9}$$

Which is the slope of the Wohler curve and is a function of the parameter m that depends on the time-temperature dependency of the material. A decrease in the time-temperature dependency of the material (decrease in m) corresponds to a reduction in B.

An example of the fatigue law is given in Figure 1.

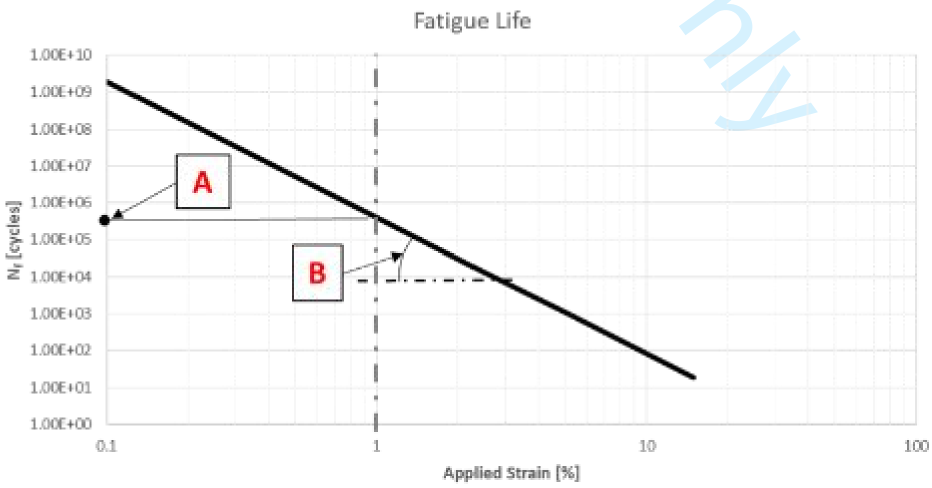


Figure 1. Example of the fatigue law of bituminous materials (Equation (7)).

Considering the failure criterion as a reduction of 35% in the viscous modulus, Equation (7) can be written as Equation (10).

$$N_f = A_{35}(\gamma_{max})^B \quad (10)$$

Results are discussed in terms of the fatigue law, A_{35}, B, and N_f calculated at γ_{max} equal to 2.5% (typical of thick pavements) and 5.0% (typical of thin pavements) (Hintz, Velasquez, Johnson, et al., 2011; Hintz, Velasquez, Li, et al., 2011; Teymourpour & Bahia, 2016).

2.4.3. Selection of LAS temperature

Fatigue cracking of asphalt mixtures occurs within the range of intermediate pavement temperatures. In the case of the LAS test, it is recommended to conduct the test at the intermediate PG grade temperature (Safaei & Castorena, 2016; Teymourpour & Bahia, 2016). However, the bitumens in this study were all tested at the same temperature, although they are of different PG grades. Besides the evaluation of the intermediate grade temperatures, an alternative method to define the LAS test temperature was investigated.

Four different provinces in Italy were selected to be representative of intermediate climatic regions (Figure 2).

Figure 2. Italian climatic region considered to define LAS test temperature.

For each province, the data given in (http://www.centrometeo.com) was taken. The climatic and geographic data (Table 3) were used in the Bell2 model (Federal Highway Administration – FHWA,

2000) to estimate the annual average temperature in the center of a base course layer located to a depth of 14.5 cm.

Table 3. Climatic and geographic data used to calculate LAS test temperatures.

(Max. T)	Annual maximum air temperature (Average from the past ten years)
(Min. T)	Annual minimum air temperature (Average from the past ten years)
(Avg. T)	Annual average air temperature (Average from the past ten years)
Latitude	Latitude of the location

The average value from the four provinces was considered. The average temperature from this method results equal to 31°C compared to the average PG intermediate temperature of 28°C with the LAS test temperature selected as 31°C.

3. RESULTS AND DISCUSSION

3.1. Fluorescence microscopy

Images of the fluorescence microscopy of the SBS and EVA PMBs are given in Figures 3 to 8.

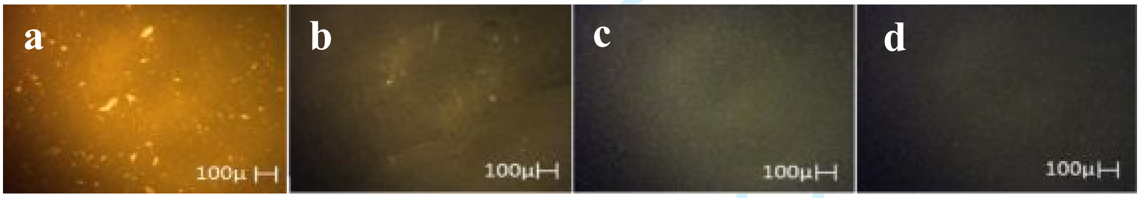


Figure 3. Morphology of SBS 2/S bitumen (10X magnification) (a – Unaged; b – RTFO; c – 2 PAV; d – 4 PAV).

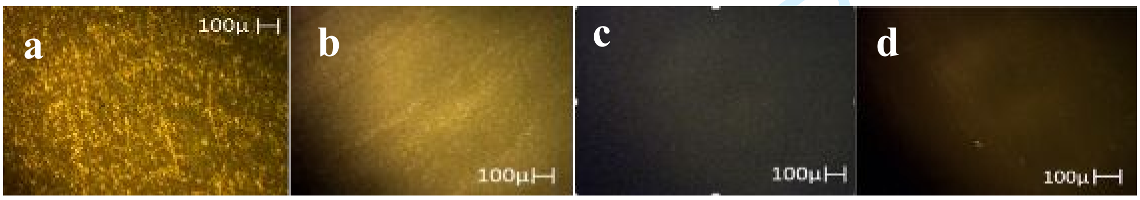


Figure 4. Morphology of SBS 4/S bitumen (10X magnification) (a – Unaged; b – RTFO; c – 2 PAV; d – 4 PAV).

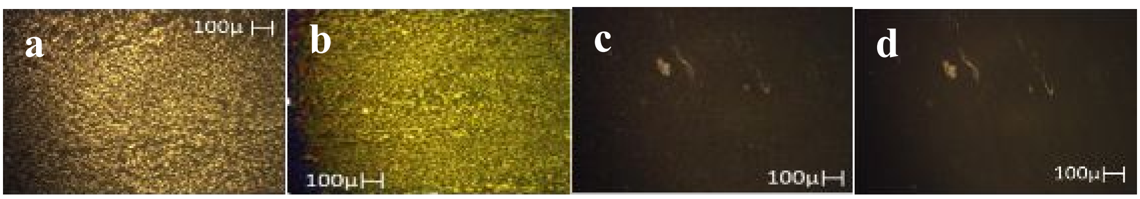


Figure 5. Morphology of SBS 6/S bitumen (10X magnification) (a – Unaged; b – RTFO; c – 2 PAV; d – 4 PAV).

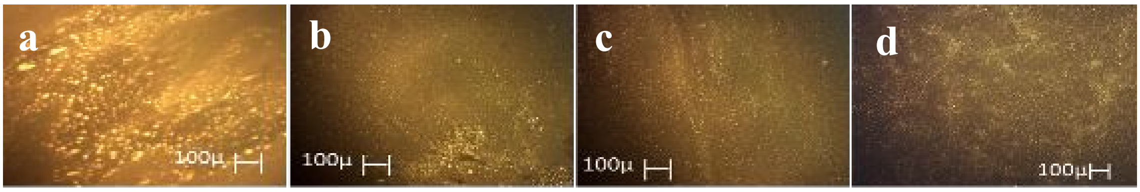


Figure 6. Morphology of EVA 2 bitumen (10X magnification) (a – Unaged; b – RTFO; c – 2 PAV; d – 4 PAV).

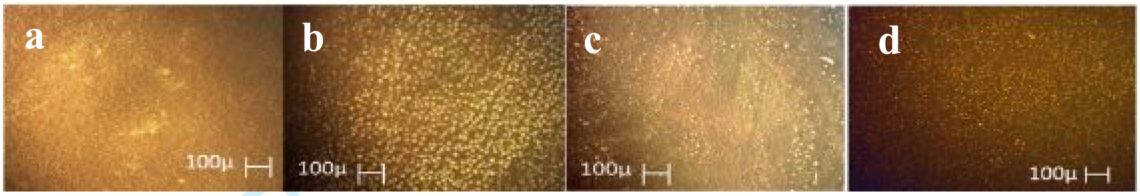


Figure 7. Morphology of EVA 4 bitumen (10X magnification) (a – Unaged; b – RTFO; c – 2 PAV; d – 4 PAV).

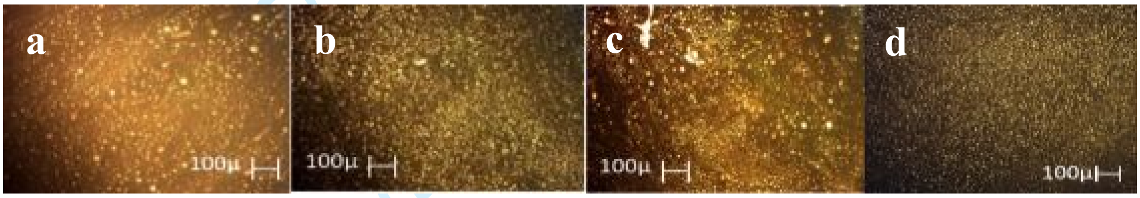


Figure 8. Morphology of EVA 6 bitumen (10X magnification) (a – Unaged; b – RTFO; c – 2 PAV; d – 4 PAV).

Details on the effects of bitumen-polymer compatibility and aging on the morphology have been discussed in Cuciniello et al. 2018 and 2019. The SBS PMBs show higher compatibility (prompted by the use of sulphur as a cross-linking agent) than the EVA PMBs, which tend to remain dispersed as disconnected spherical droplets. In terms of aging susceptibility, the changes in the morphology of the SBS PMBs are a function of the polymer concentration (and use of sulphur). At the same time, the EVA PMBs show a low aging susceptibility irrespective of the polymer concertation. This aspect is supported by the fluorescent microscopy images in Figure 6 to 8 that indicate the presence of EVA polymer as dispersed droplets even after 80 hours of PAV.

Unlike previous studies that only considered reduced levels of aging (i.e., 1 PAV in Cuciniello et al., 2018) and the link between morphology and rheological results obtained at high temperatures through the Multiple Stress Creep and Recovery (MSCR) test, this study showed extended aging profiles of the PMB morphology. This paper also focuses primarily on the rheological testing of the fatigue performance of SBS and EVA PMBs and the influence of aging while only using the morphology to corroborate the relations between aging and damage tolerance.

3.2. LAS test

The test data were analyzed following the theoretical framework streamlined from Equation (3) to Equation (7). Repeatability of the test was checked by considering the A_{35} and B values of Equation (7 that show a maximum variability (between two samples) of 15% (Hintz, Velasquez, Li, et al., 2011).

The fatigue laws of the Pen bitumen at different levels of aging are given in Figure 9.

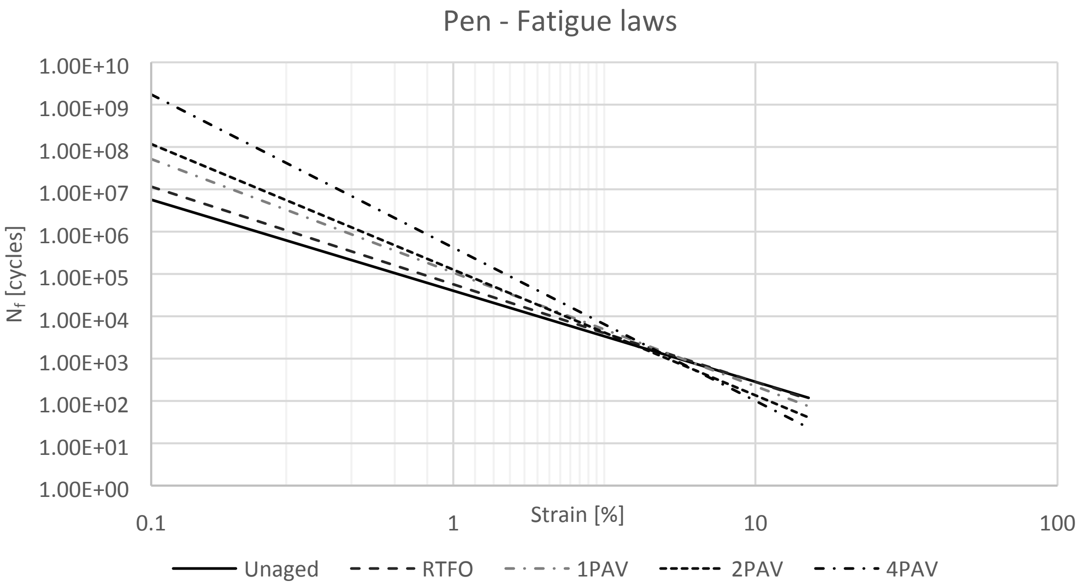


Figure 9. Fatigue laws of Pen bitumen at different levels of aging.

In the strain interval below 1%, the number of cycles to failure increases with aging (larger increases in N_f corresponding to lower strain values). However, this strain level is rarely experienced by the bitumen phase in pavements, which is exposed to higher strain levels than the bulk pavement. Such a difference depends on localized rotations of aggregates, on the different stiffness between the bitumen and the aggregates and the lack of uniformity of the bitumen film coating the aggregates (Airey et al., 2004; Masad & Somadevan, 2002; Kose et al., 2000). From 1% to 10% (and higher), the curves converge and overlap so that the fatigue functions shown in the figure at different aging levels intersect at a defined strain value, which has been qualitatively defined as the “pivot strain.” Its value decreases with aging (Figure 10). This strain interval (from 1% to 10% and higher) could be seen as the transition of the strain levels in the bitumen phase from thick to thin pavements. Fatigue cracking is more likely to occur in thin pavements (higher strain levels); for this reason, aging is seen to worsen the durability.

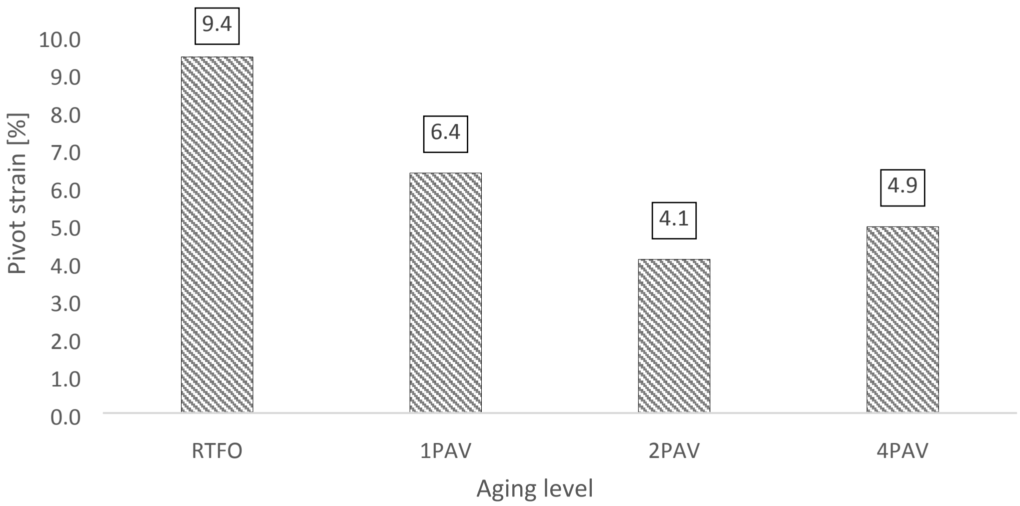


Figure 10. Pivot strains of Pen bitumen at different levels of artificial aging.

Polymer modification does not affect the behavior described for the Pen bitumen, with aging providing either a beneficial or a detrimental effect as a function of the strain level considered. An example is given in Figure 11.

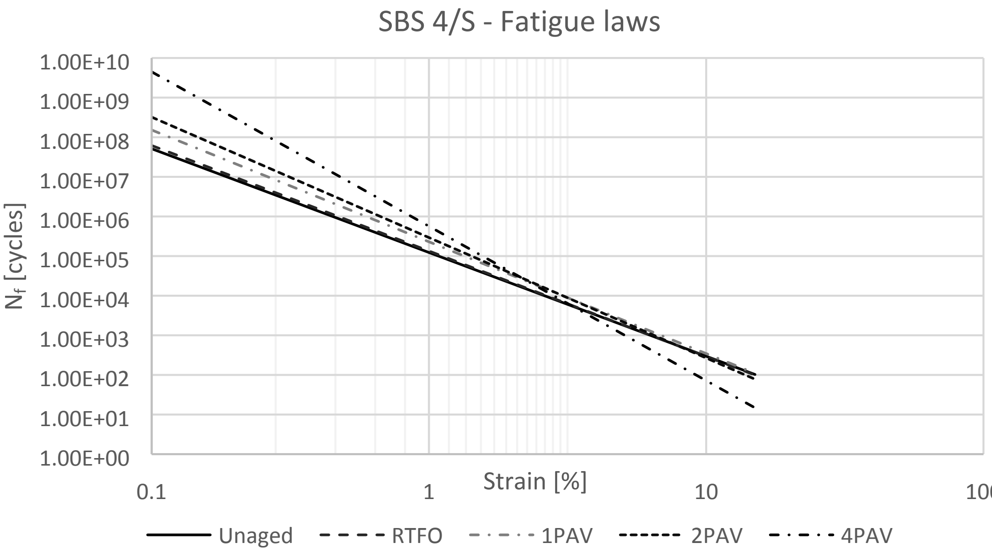
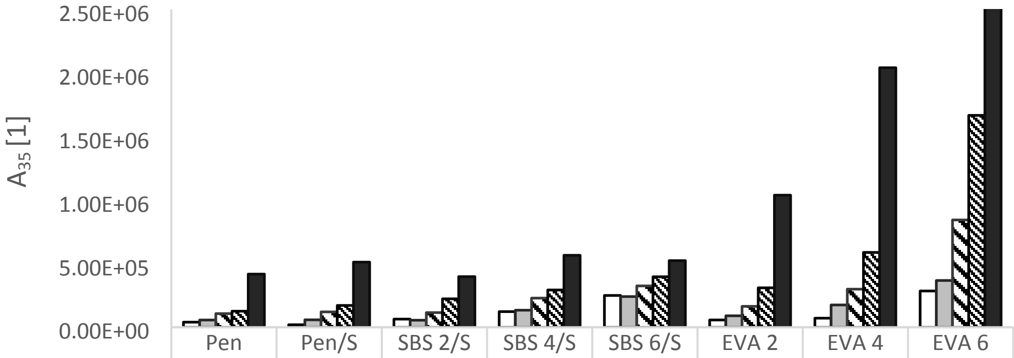


Figure 11. Fatigue laws of SBS 4/S bitumen at different levels of aging.

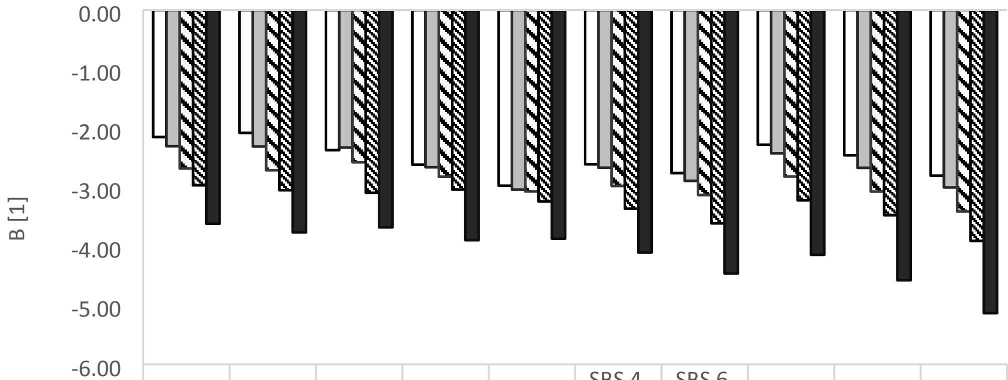
Polymer modification affects the magnitude of the phenomenon, and the pivot strain at the various levels of aging is a function of polymer type and polymer concentration. In the case of the SBS and EVA modified bitumens as well as the sulphur (Pen/S) bitumen, there are only minor differences compared to the Pen bitumen with similar results being seen for all eight bitumens used in the study. Although there are slight differences in the aging susceptibility of the bitumens as a function of the degree of aging, the strain dependency effect of aging on the fatigue cracking resistance is common to all the binders and in agreement with the work of Hintz, Velasquez, Li, and co-workers (2011).

The evaluation of the effects of aging on the resistance to damage accumulation of bitumen needs to account for the strain level considered, and the use of a single strain level (i.e., 1%) may not be representative of the actual performance of bitumens. The values of the parameters of Equation (10) (i.e., A_{35} and B) measured at the different levels of aging are given in Figures 12 and 13.



	Pen	Pen/S	SBS 2/S	SBS 4/S	SBS 6/S	EVA 2	EVA 4	EVA 6
Unaged	4.01E+04	1.99E+04	6.41E+04	1.23E+05	2.50E+05	5.73E+04	7.15E+04	2.85E+05
RTFO	5.70E+04	5.84E+04	5.54E+04	1.34E+05	2.41E+05	9.02E+04	1.75E+05	3.68E+05
1PAV	1.08E+05	1.21E+05	1.15E+05	2.29E+05	3.25E+05	1.65E+05	3.00E+05	8.44E+05
2PAV	1.26E+05	1.72E+05	2.24E+05	2.93E+05	3.97E+05	3.10E+05	5.89E+05	1.67E+06
4PAV	4.18E+05	5.12E+05	3.98E+05	5.66E+05	5.24E+05	1.04E+06	2.04E+06	8.57E+06

Figure 12. Values of A_{35} (Equation (10)) at different levels of aging.



	Pen	Pen/S	SBS 2/S	SBS 4/S	SBS 6/S	SBS 4 No S	SBS 6 No S	EVA 2	EVA 4	EVA 6
Unaged	-2.15	-2.08	-2.37	-2.62	-2.98	-2.61	-2.76	-2.28	-2.46	-2.80
RTFO	-2.31	-2.31	-2.33	-2.66	-3.04	-2.67	-2.89	-2.43	-2.67	-3.01
1PAV	-2.68	-2.71	-2.58	-2.82	-3.07	-2.98	-3.13	-2.82	-3.07	-3.41
2PAV	-2.97	-3.05	-3.10	-3.04	-3.24	-3.36	-3.61	-3.22	-3.48	-3.91
4PAV	-3.62	-3.77	-3.68	-3.90	-3.87	-4.11	-4.46	-4.15	-4.58	-5.13

Unaged RTFO 1PAV 2PAV 4PAV

Figure 13. Values of B (Equation (10)) at different levels of aging.

The fatigue parameters (A_{35}) and (B) show opposite trends with aging. A_{35} increases with aging, while B decreases. The first depends on the initial undamaged modulus (I_D - Equation (8)) and on the variation of the material integrity with damage ($|G^*|\sin\delta$ versus D – Equation (4)). Since aging produces an increase in I_D to which should correspond to a decrease in A_{35} (Equation (8)), the growth of the parameter A_{35} depends on how much the material is capable of maintaining integrity with damage. Therefore, it depends on the integrity versus damage curve. An example is given in Figure 14.

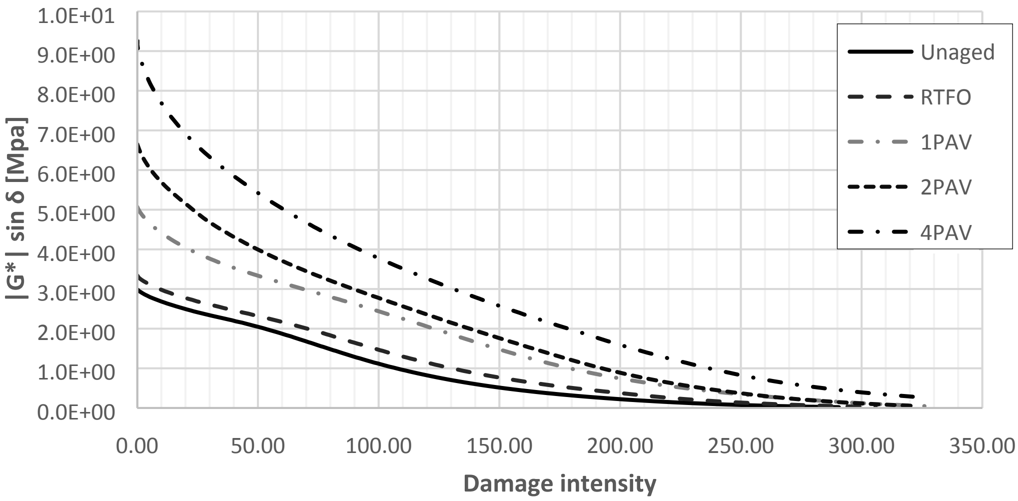


Figure 14. Material integrity versus damage curve Pen bitumen.

Based on the curves, it seems that the more the material is capable of maintaining its integrity (G'') against damage, the higher is the value of A_{35}. Similar conclusions are given in Hintz, Velasquez, Li, and co-workers (2011).

On the other hand, the parameter B decreases with aging as a function of the time-temperature susceptibility of the bitumens (Equation (9)).

The combination of the A_{35} and B trends contributes to defining the effects of aging on the damage tolerance of the bitumens. In the case of low strain levels, the capability of maintaining integrity (A_{35}) has a more considerable impact, and the number of cycles to failure increase with aging. At higher strain levels, the opposite occurs, and aging contributes to decreasing N_f. Polymer modification alters the magnitude of this mechanism (Figure 9 & Figure 11).

3.2.1 SBS modified bitumens

Results have been compared in terms of the number of cycles to failure at 2.5% strain (thick pavements) and 5.0% strain (thin pavements). Before discussing the results, it is worth recalling that the comparison among different bitumens has been conducted under the same test temperature. This condition does not account for the effects of different stiffness. However, the binders are tested at the same temperature to highlight the effects of aging, polymer type, and polymer concentration. Results at the strain level of 2.5% are given in Figure 15.

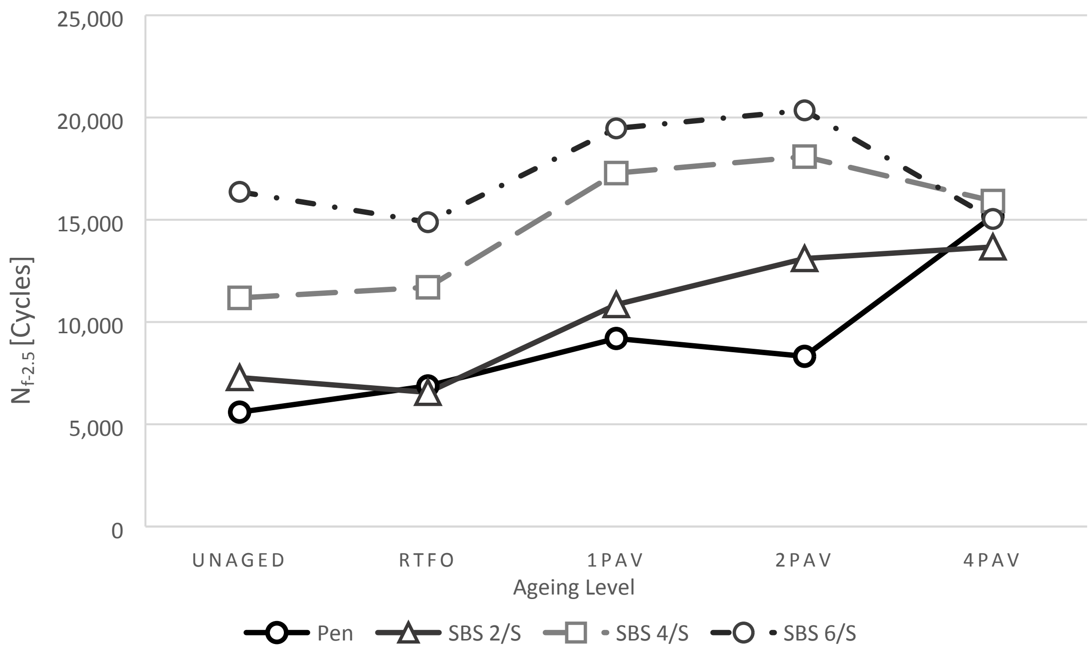


Figure 15. Number of cycles to failure at strain 2.5% of Pen and SBS modified bitumens at different levels of aging.

For the unaged binders, the effect of SBS modification is visible, with the number of cycles to failure increasing with the polymer content. Aside from the polymer content, Figures 3 to 5 show that at higher concentrations, the polymer-phase achieves a higher level of dispersion (and structure), which improves the fatigue resistance. The effects of SBS modification are visible between RTFO and 2PAV, where N_f increases with aging while the Pen bitumen shows a limited variation between 1PAV and 2PAV. At 4PAV, the effects of the modification are likely to have vanished with the bitumens showing similar N_f for both the control and SBS PMBs. The morphologies in Figures 3 to 5 display the reduction in the polymer dispersion that qualitatively indicates the degradation of the polymer network with this degradation being in terms of molecular weight distribution (Cuciniello et al., 2019).

The fatigue results at the strain level equal to 5% are given in Figure 16.

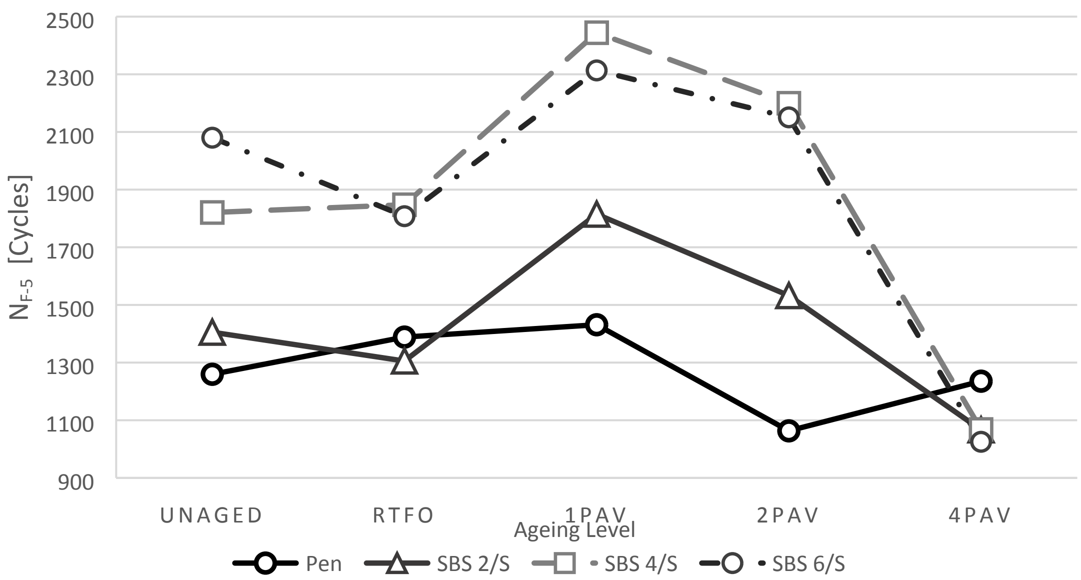


Figure 16. Number of cycles to failure at strain 5.0% of Pen and SBS modified bitumens at different levels of aging.

The effects of polymer modification are also visible at this strain level. However, at this higher strain level, the effects of aging on N_f start to be pejorative at a lower PAV exposure (degree of aging). A comparison between the curves at 2.5% (Figure 15) and those at 5.0% (Figure 16) show that at the higher strain level (i.e., 5.0%) the number of cycles to failure decreases between 1PAV and 2PAV; while at the lower strain level (i.e., 2.5%), the reduction occurs between 2PAV and 4PAV. Also in this case, the bitumens have a similar number of cycles to failure at 4PAV.

It appears that once the polymer network is damaged, as shown in Figures 3-5 (d – 4PAV), the polymer modification does not affect the rheological response with the behavior being controlled by the PAV-oxidised bitumen-rich phase. The fact that after four cycles of PAV, the unmodified bitumen and the SBS PMBs have the same fatigue life indicates that the bitumen (i.e., Pen) and the bitumen-rich phase of the SBS modified bitumens have undergone similar PAV-oxidative hardening irrespective of the presence (and the concentration) of the polymer. That is, once the polymer network is damaged, the bitumen-rich phase controls the rheological response, and the level of aging is similar to that of the base bitumen under the same aging conditions. Similar conclusions were drawn by Cotte and Such (1996) and by Airey and Brown (1998). However, both these studies focused on the linear region with no damage tolerance investigated.

3.2.2 EVA modified bitumens

The number of cycles to failure at 2.5% strain level of the Pen and the EVA modified bitumens at different levels of aging are given in Figure 17.

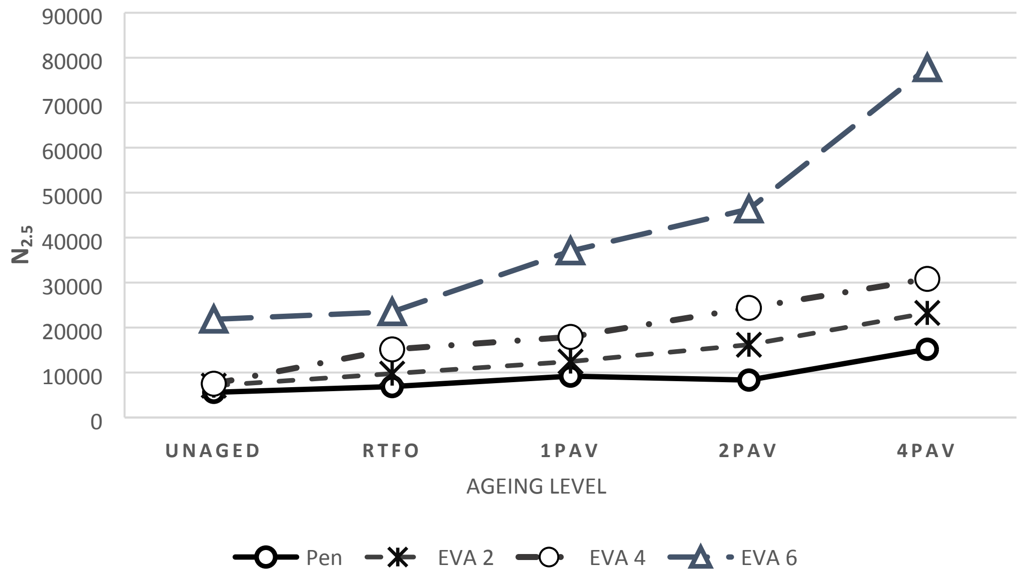


Figure 17. Number of cycles to failure at strain 2.5% of Pen and EVA modified bitumens at different levels of aging.

At the unaged condition, the EVA modification increases the fatigue life but only significantly at the very high polymer concentration. The number of cycles to failure of the EVA 2 and EVA 4 bitumens are close to those of the Pen (below 10,000 cycles). Considering the results of the SBS modified bitumens (Figure 15), the EVA 6 bitumen is the one providing the highest number of cycles to failure at all levels of aging, with aging that increases the number of cycles to failure. Furthermore, under the same level of aging, the number of cycles increases with polymer concentration. At this strain level, N_f of the modified bitumens is larger than the Pen bitumen, with the EVA 6 achieving more than 70,000 cycles to failure.

Results of the fluorescence microscopy (Figures 6 to 8) show that the EVA PMBs have a reduced aging susceptibility. Specifically, the images have highlighted that the polymer dispersion remains unvaried with PAV aging. It can be hypothesized that the microstructure of the EVA modified bitumens evolves towards a highly PAV-hardened bitumen-rich phase and an oxidative resistant polymer-rich phase, which provides an additional contribution to the bitumen integrity. The bitumen-rich phase hardens because the polymer does not mitigate the oxidative effects (a consequence of low-compatibility). The polymer-rich phase has a very low PAV susceptibility and resists oxidation. Therefore, the system might evolve towards a "biphasic structure" very resistant to damage where the highly hardened bitumen phase is reinforced by the presence of the polymer that maintains its dispersed structure. The formation of this system increases the integrity of the bitumen and its cumulative damage resistance. The analysis of the fatigue life at 2.5% strain shows the consequence on fatigue life of the variation in the microstructure of EVA modified bitumens.

Results at the higher strain level (5%) are given in Figure 18.

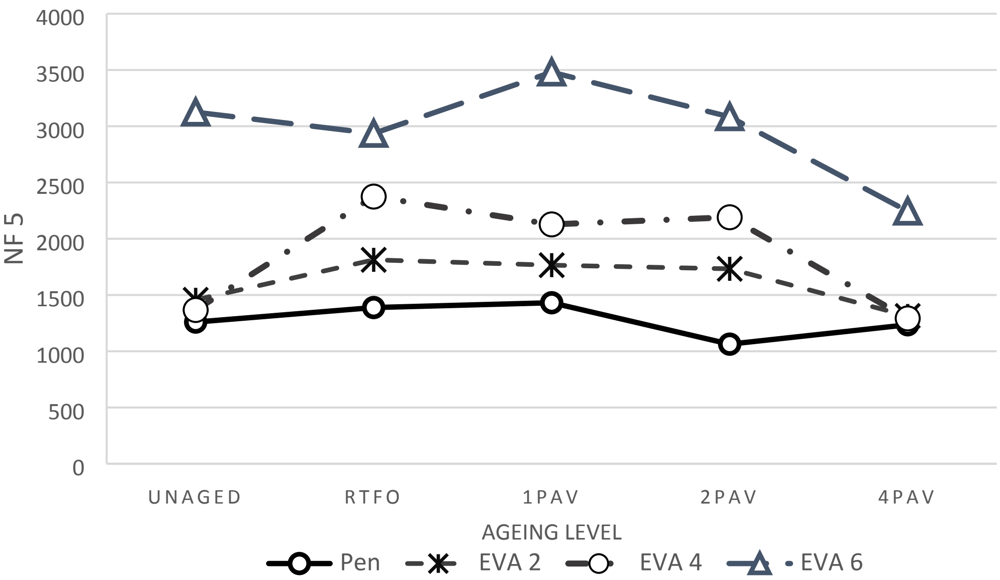


Figure 18. Number of cycles to failure at strain 5.0% of Pen and EVA modified bitumens at different levels of aging.

The increase in the strain level reduces the number of cycles to failure of the bitumens. Similar to the lower strain level, at the unaged condition, the polymer modification increases the fatigue life only at the very high concentration (i.e., 6% EVA). The higher strain level reduces the effect of aging on N_f that shows minimal variation for all the bitumens. To some extent, it can be concluded that in the case of thicker pavements, the use of EVA polymer might contribute to increased durability with aging.

4. CONCLUSIONS

Results from this study into the effect of aging and polymer modification on the damage tolerance of different bitumens have produced the following key conclusive remarks.

(a) Aging increases the stiffness of bitumens and contributes to maintaining their integrity against damage (A_{35} grows). On the other hand, aging decreases their time-temperature susceptibility (B decreases). The first mechanism is visible in the intercept with the y-axis of the fatigue law at a 1% strain level that increases with aging (Wohler curve). The second increases the magnitude of the slope of the Wohler curve. To estimate the contribution of aging to the damage tolerance, the two effects need to be combined by considering the maximum strain level experienced by the bitumen phase in asphalt mixtures.

(b) The results of this combination show that for each bitumen, it is possible to define a strain value that represents a threshold for ranking the effects of aging as positive or negative. Below this strain level, the cumulative damage resistance increases with aging, while above this range, it decreases. Other researchers have observed these results. Below a strain of 1%, all the bitumens show an increase in the cumulative damage resistance with aging. However, this strain value might not be representative of the strain experienced by the bitumen phase in pavements. Polymer modification does not alter the mechanism observed but affects the magnitude of the changes with similar conclusions being drawn for each polymer.

(c) For the cross-linked SBS modified bitumens, the higher the polymer concentration (and dispersion), the higher is the number of cycles to failure. The gain in the damage tolerance is a function of the level of integrity of the polymer network that is qualitatively visible when looking at the binder morphology. Between the unaged and 2PAV-aged conditions, the bitumens show higher fatigue cracking resistance than the unmodified bitumen. After 4PAV, when the polymer has undergone extensive damage, this difference becomes negligible. There is a reason to believe that at 31°C, the rheological response is more controlled by the bitumen-rich phase. Therefore, at 4PAV, the latter has undergone the same PAV-oxidative hardening of the control bitumen irrespective of the presence of the polymer.

(d) For the EVA modified bitumens, polymer modification increases the damage tolerance as a function of the polymer concentration. The higher the polymer concentration, the higher is the resistance. At 2PAV-aging, EVA 6 shows the highest material capacity to maintain integrity against damage and the highest number of cycles to failure at the strain level of 2.5%. The damage tolerance is observed to increase with aging as a function of the polymer concentration at the strain of 2.5%, while it has shown a low aging susceptibility at the strain of 5%. However, at both strain levels, the EVA PMBs have shown a higher number of cycles to failure compared to the unmodified bitumen. These results are indicative of a low PAV-susceptibility of the polymer network that has also been observed by fluorescence microscopy.

Disclosure statement

No potential conflict of interest was reported by the authors.

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