

The effect of Selenice Natural Bitumen on un-modified and polymer-modified bitumen rheological properties

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ABSTRACT

Selenice Natural Bitumen (SNB) is a natural bitumen with specific properties, which make it a valuable additive in asphalt mixtures in order to reduce the dosage of virgin bitumen, thus reducing the environmental footprint. The current state of the art validated its use in unmodified bituminous blends, highlighting the positive influence as an ageing inhibitor only at low dosages ($\leq 10\%$). In order to consider higher dosages of SNB in asphalt pavement materials, the objective of this study is to investigate the rheological properties of bituminous blends composed of SBS polymer-modified bitumen and SNB.

The experimental program evaluated the addition of SNB in different dosages in an un-modified bitumen and an SBS polymer-modified bitumen. The analysis involved empirical and rheological properties at different ageing conditions. The main rheological characterization consisted in complex modulus tests, Multiple Stress-Creep Recovery (MSCR) tests and Linear Amplitude Sweep (LAS) tests.

Overall, the results related to un-modified bituminous blends are consistent with findings reported in previous studies, confirming that a maximum SNB content of 10 % by weight is recommended to limit performance loss. Conversely, the experimental data did not support the feasibility of increasing the SNB dosage beyond 10 % when using SBS polymer-modified bitumen. The presence of SBS polymers in the modified blends did not mitigate the fatigue performance loss associated with SNB use. Therefore, the use of SNB at dosages exceeding 10 % by weight of the total bituminous blend is not recommended, regardless of whether unmodified or SBS-modified bitumen is employed.

1. Introduction

The increasing demand of alternative solutions to reduce the global environmental footprint of industrial processes involves, among others, the asphalt industry. In Hot Mix Asphalts (HMA), this is possible by partially or totally replace the virgin aggregates and/or the virgin bitumen. In the last decades several studies focused on different possibilities, i.e. the optimized use of the reclaimed asphalt pavement (RAP) [1–3], the Warm Mix Asphalt (WMA) technology [4,5], cold recycled mixtures (CRM) [6–8], crumb-rubber asphalt mixtures (CRAM) [9,10], use of artificial aggregates [11,12] and the use of bio-binders [13–15]. However, another type of material can be considered as a possible additive in HMA, i.e. natural bitumens (or natural asphalts). A natural bitumen (NB) is available in nature and it is composed of highly oxidized bitumen and mineral parts. Although NB is not a recycled material, nowadays no specific employment of such resource is clearly recognized [16]. NB materials are valid additives for bituminous binders compared

to polymers, since the latter have the disadvantage of stability problems [17].

Several types of NBs are available worldwide, and their characteristics and possible applications may vary based on their characteristics.

A specific type of NB is mined in Albania, more specifically in the region of Selenice. The Selenice Natural Bitumen (SNB) is composed of 15–20 % of mineral materials, which is generally reduced during mining operations by means of a heating treatment process. Successively, the organic phase is dissolved in tetrachloroethylene and centrifuged to obtain the material intended for petroleum bitumens modification [18]. From an environmental point of view, the production of 1 ton of SNB causes around 130 kg of CO₂ emissions, while 1 ton of standard paving bitumen causes around 230 kg of CO₂ emissions. This significant difference is given by a lower impact of the SNB mining operations compared to the crude oil extraction process and the SNB storage conditions, since no additional heating is required [19].

A study from Zeng et al. (2016) showed that the use of SNB led to a

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decrease in penetration and an increase in softening point [20]. More specifically, Tartari (2019) reported that a decrease in one penetration grade can be expected if SNB is used at 8–10 %, depending on the original bitumen selected for modification [21]. The use of SNB was also discovered to be advantageous in terms of aging mechanisms. The effect of simulated aging in laboratory on the materials investigated were less consistent for the SNB modified binders than for the original, implying that the natural bitumen may operate as an aging inhibitor.

Another study deeply investigated the physical, rheological and mechanical performance of both SNB modified bitumen and the corresponding mixture, highlighting an optimal SNB dosage of 15 % by bituminous blend mass and a recommended dosage of 10 – 20 % by bituminous blend mass. Such findings suggested the possibility to advantageously use SNB to develop bio-binders combining SNB and rejuvenating oils. In particular, the rheological properties of bituminous blends composed of about 70 % of SNB, 10 % of a hard penetration grade bitumen and 20 % of two different waste vegetable oils were investigated. The mechanical properties of the bio-binder obtained were compared to those of a reference un-modified bitumen P35/50. The results on both binders and mixtures scale were promising in terms of rutting resistance, even though low temperature cracking and fatigue properties were not evaluated [22].

The current state-of-the-art is poor of information regarding field experience of pavements containing SNB. However, a study conducted by Santagata et al. in 2012 included a performance-based evaluation of two pavement layers (base and binder courses) containing 8 % of SNB by mass of the base bitumen. The results obtained were employed as input data for the calculation of the cumulative fatigue damage by means of a mechanistic procedure. It was observed that the considered bituminous mixtures containing natural asphalt were accepted for the construction of the desired highway pavement [23].

Overall, the state-of-the-art regarding SNB use in asphalt binders regards only un-modified binders, and confirms its beneficial use in terms of rutting behaviour, due to the stiffening effect conferred to the blends. On the other hand, this induces potential issues in terms of cracking behaviour at both intermediate (fatigue) and low temperatures, due to the poor relaxation properties conferred by SNB dosages higher than 10 % [24]. A recent study evaluated the addition of Styrene-Isoprene-Styrene (SIS) polymers to a PG 64–22 binder containing Trinidad Lake Asphalt (TLA) natural bitumen. It was obtained that 10 % TLA combined with 5 % SIS polymers offer an optimal balance between rigidity and flexibility, thanks to the contribution given by the polymers. However, the optimal dosage strongly depends on the environmental conditions and the performance needs [25].

Consequently, this motivates the need to investigate the rheological properties of bituminous blends composed of SNB in different dosages and an SBS polymer-modified bitumen, in order to reduce the negative influence of SNB thanks to the presence of SBS polymers. In particular, the study involved both un-modified and polymer-modified bituminous blends containing SNB, covering a wide range of properties assessed by empirical and rheological test methods performed at un-aged, short-term aged and long-term aged conditions.

2. Materials and methods

2.1. Selenice Natural Bitumen (SNB)

The SNB used in this research is a 0/2 fraction granular material (Fig. 1). The properties measured of the melted SNB are listed in Table 1. It is highlighted that the needle penetration, the softening point and the ash content were determined after a previous heating of the sample at 190 °C. In this manner, the volatile components trapped in the material did not affect the test results.

The results shown are reported as an average of six samples in order to represent the natural deposit from where the material was sampled.



Fig. 1. A sample of SNB used for the production of bituminous blends.

Table 1

Properties of the Selenice Natural Bitumen.

Property	Standard	Values
Particle size (mm)	UNI EN 933–1	0/2
Needle penetration @ 25 °C (dmm)	UNI EN 1426	0
Softening point (°C)	UNI EN 1427	114.6
Ash content (%)	UNI EN 12697–47	28.5

2.2. Un-modified and polymer-modified binders

A local producer of modified bitumen supplied the three binders used in this study. A preliminary characterization was conducted, and the key properties are summarized in Table 2. In Italy, Styrene-Butadiene-Styrene (SBS) polymer-modified bitumen, generally classified as PG 58E-22, is commonly employed for highways and high-traffic roadways. On contrary, two unmodified binders, classified as PG 58E-16 and PG 58V-22, are typically used for roads experiencing low to moderate traffic volumes.

In Table 2 it is possible to observe that the polymer-modified bitumen exhibits superior performance at high, intermediate, and low service temperatures compared to the unmodified binders.

Fig. 2 shows the results in terms of the complex modulus $|G^*|$ and phase angle δ master curves. Across the entire frequency range analyzed, the PG 58V-22 binder is characterized by lower stiffness compared to the unmodified PG 58E-16 binder. Simultaneously, the phase angle master curves highlight higher values for the PG 58V-22 binder. Furthermore, the PG 58E-22 polymer-modified bitumen exhibits higher complex modulus values at low frequencies (corresponding to high temperatures). In the same conditions, the phase angle behavior is significantly influenced by the presence of SBS polymers.

2.3. Bituminous blends investigated and production protocol

The experimental program involved six bituminous blends produced with different dosages of SNB material. The SNB material was combined with the un-modified bitumen PG 58V-22 resulting in the bituminous blends N1, N2 and N3. Due to the SNB properties (Table 1), it is reasonable to hypothesize that its inclusion may increase the stiffness of the resulting bituminous blends, as supported by existing literature [22]. Consequently, the three blends above-mentioned were compared with the stiffer un-modified bitumen, PG 58E-16.

Similarly, the SNB material was combined with the polymer-modified bitumen PG 58E-22 obtaining the bituminous blends identified as PM1, PM2 and PM3.

The composition of each bituminous blends produced is reported in

Table 2
Properties of the base binders used.

Property	Standard	Unit	Value		
			PmB	Un-modified	
Performance Grade (PG)	ASTM D8239	-	58E-22	58E-16	58V-22
Needle penetration @ 25 °C	UNI EN 1426	dmm	47	53	83
Softening point	UNI EN 1427	°C	71.0	52.3	45.8
Brookfield viscosity @ 160 °C	UNI EN 13302	Pa·s	0.490	0.096	0.090
Multiple Stress Creep Recovery (MSCR) @ 58 °C	UNI EN 16659	J_{nr} 0.1 kPa	0.12	0.33	0.84
		3.2 kPa	0.15	0.34	0.91
		%R 0.1 kPa	61.2	18.4	12.5
		3.2 kPa	53.6	13.8	5.7
Linear Amplitude Sweep (LAS) @ 25 °C	AASHTO T 391-20	N_f (2.5 %)	7.1E+ 06	3.6E+ 06	3.9E+ 06
		N_f (5.0 %)	9.7E+ 04	1.5E+ 04	4.0E+ 04
		N_f (15.0 %)	1.1E+ 02	2.6E+ 00	2.9E+ 01

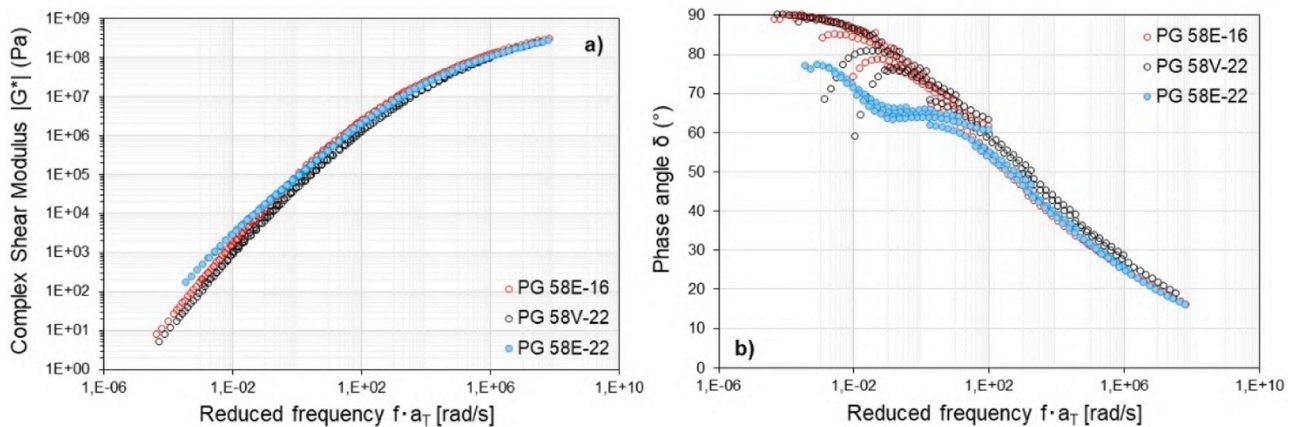


Fig. 2. Binders master curves ($T_{REF} = 30\text{ °C}$): a) Complex Shear Modulus $|G^*|$; b) Phase angle δ .

Table 3
Identification of the bituminous blends analyzed.

Bituminous blend	Base bitumen	SNB
N1	90 %	10 %
N2	85 %	15 %
N3	80 %	20 %
PM1	90 %	10 %
PM2	85 %	15 %
PM3	80 %	20 %

Table 3. The dosages of the base bitumen (PG 58V-22 and PG 58E-22) and the SNB material are expressed as a percentage of the total mass of the bituminous blends.

The laboratory production of the bituminous blends was carried out by means of a high-shear mixer for bitumen modification and an oil bath used to keep operating temperatures constant at 190 °C. The mixing process was performed for 1 h, which was determined to be sufficient to

achieve uniform dispersion of the SNB material, at a shear speed of 1800 RPM.

2.4. Experimental program

The research program was carried out considering empirical and rheological properties of the bituminous blends investigated at different ageing conditions. A summary of the experimental activities is reported in Table 4.

Rheological tests such as complex shear modulus, Multiple Stress-Creep Recovery (MSCR) tests, Linear Amplitude Sweep (LAS) tests and the determination of the Glover-Rowe (G-R) parameter were carried out by means of a Dynamic Shear Rheometer (DSR). Short-term ageing (STA) was simulated in accordance to the Rolling Thin Film Oven Test (RTFOT) procedure at a testing temperature of 163 °C (UNI EN 12607-1), followed by the Pressure Ageing Vessel (PAV) procedure at a testing temperature of 100 °C and pressure of 2.1 MPa for long-term ageing (LTA) simulation (UNI EN 14769).

Table 4
Experimental program.

Property	Ageing conditions ⁽¹⁾	Standard
Empirical tests:		
Needle Penetration @ 25 °C	UB + STA	UNI EN 1426
Softening Point	UB + STA	UNI EN 1427
Rheological characterization:		
Brookfield Viscosity @ 160 °C	UB	UNI EN 13302
Complex modulus $ G^* $ - δ -method	UB + STA	AASHTO T 315-12
Multiple Stress-Creep Recovery Test	STA	UNI EN 16659
Linear Amplitude Sweep (LAS) @ 25 °C	LTA	AASHTO T 391-20
G-R parameter	LTA	-

⁽¹⁾ UB = Un-aged binder, STA = Short-term aged binder, LTA = Long-term aged binder

In general, the effect of the SNB modification in the bituminous blends on the short-term ageing is investigated by performing needle penetration tests, softening point tests and complex modulus tests at both un-aged and short-term aged conditions.

Complex modulus tests were carried out with a 25 mm plate configuration for high testing temperatures ($> 30\text{ }^{\circ}\text{C}$) at a strain level of 0.1 % and with a 8 mm plate configuration for low testing temperatures ($\leq 30\text{ }^{\circ}\text{C}$) at a strain level of 0.05 %. The investigated temperatures ranged between $-10\text{ }^{\circ}\text{C}$ and $70\text{ }^{\circ}\text{C}$, with $10\text{ }^{\circ}\text{C}$ increase steps, whereas the investigated angular frequencies varied from 0.1 to 100 rad/s.

The bituminous blends investigated were initially validated as thermo-rheologically simple, allowing the application of the Time-Temperature Superposition Principle (TTSP). In this manner, the isothermal curves obtained for the complex shear modulus $|G^*|$ and the phase angle δ can be shifted in the time domain in order to form a single curve (master curve) characteristic of the material analysed at a specific reference temperature (T_{REF}). This is possible by the definition of the shift factors (a_T), specific for each isothermal curve. In case of viscous materials, shift factors values and the related temperatures are correlated, and the Williams-Landel-Ferry (WLF) model can accurately approximate the experimental values [26]. For bituminous materials, the WLF equation is:

$$\log(a_T) = -\frac{C_1(T - T_{\text{REF}})}{C_2 + T - T_{\text{REF}}} \quad (1)$$

Where a_T is the shift factor at the temperature T , T_{REF} is the reference temperature, C_1 and C_2 are constants.

The complex modulus results obtained were modelled by the rheological model 2S2P1D, which is composed by 2 springs, 2 parabolic elements and 1 linear dashpot to describe the complex behaviour of bituminous materials [27]. The 2S2P1D model consists in the following equation:

$$G^*(i\omega\tau) = G_0 + \frac{G_{\infty} - G_0}{1 + \delta(i\omega\tau)^{-k} + (i\omega\tau)^{-h} + (i\omega\tau\beta)^{-1}} \quad (2)$$

Where G_0 is the static shear modulus when $\omega \rightarrow 0$ ($G_0 = 0$ for binders); G_{∞} is the glassy shear modulus when $\omega \rightarrow \infty$; $\omega = 2\pi f$ is the angular frequency; i is the imaginary unit defined as $i^2 = -1$; τ is the characteristic time; δ , k and h are dimensionless constants ($0 < k < h < 1$); β is a function of the dashpot viscosity ($\eta = G_{\infty} \cdot \beta \cdot \tau$). Based on the TTSP, τ can be determined as:

$$\tau(T) = a_T \cdot \tau_0 \quad (3)$$

Results obtained from complex modulus tests allow to investigate the molecular weight distribution (MWD) of bitumens and polymers. In general, the MWD parameter is determined by chromatographic methods, as gel permeation chromatography. However, such methods highlight low accuracy mainly because of the presence of solvents. In alternative, several studies present in literature show the high sensitivity of phase angle master curves to bitumens MWD [18,28,29]. These two properties can be linked by the δ -method, which relies on the assumption that the phase angle δ at a given angular frequency ω_i is proportional to the fraction of relaxed components at the i^{th} frequency [30].

During dynamic measurements in the small-strain domain and with oscillatory loading configuration, viscous materials are characterized by a series of relaxation processes. In particular, the material investigated can be considered as a composite of different elements with different relaxation frequencies, which are associated to a characteristic single molecular weight [31]. In bituminous binders, it was found that as the angular frequency increases smaller and smaller components would relax, whereas the components already relaxed will behave as a diluent for the unrelaxed ones [32]. On contrary, as the angular frequency increases, bigger components will relax resulting in no contribution to the complex modulus.

MWD can be expressed in terms of cumulative molecular weight

distribution (CMWD), i.e. the semi-logarithmic plot of molecular weight (MW) in the abscise axis and the cumulative fraction, cumf(MW), in the ordinate. The CMWD and the phase angle master curve are linked as follows:

$$\text{CMWD} = A + B \cdot \delta(\text{MW}) \quad (4)$$

By differentiating Eq. (4), the differential molecular weight distribution (DMWD, or AMWD) is obtained:

$$\text{AMWD} = \frac{d(\text{CMWD})}{d(\log \text{MW})} \quad (5)$$

Although it was not possible to measure the fractional composition of the SNB sample used in this study, results from literature for the same material source report on average an asphaltenes content of 39.6 %, a resins content of 36.2 %, an aromatic components content of 22.8 % and saturates components content of 1.6 % [21]. On the other hand, results available in literature for un-modified and polymer-modified bitumen report an asphaltenes content between 12 % and 24 %, a resins content between 26 % and 41 %, an aromatic components content between 35 % and 50 % and a saturated components content between 5 % and 12 % [33]. According to these values, it is possible to observe that the two types of binders differ mostly in terms of asphaltenes (dispersed components) and aromatic (continuous phase) contents.

Hence, this approach was applied to better evaluate the introduction of SNB in un-modified and polymer-modified bitumens, considering at the same time the effect of short-term ageing.

To assess the implications of these compositional differences on the mechanical behavior of the resulting blends, rheological tests were carried out to investigate the bituminous blends performance in terms of permanent deformation and cracking resistance. In particular, the tests focused on the evaluation of key performance indicators such as elasticity and recovery capacity, as well as fatigue resistance properties.

Elastic recovery (ER) test is nowadays a common characteristic required by roadways and highways agencies for polymer-modified bitumens classification and verification. In Italy, an ER value greater than 80 % is generally required on un-aged bitumen samples collected at the production plant [34,35]. The test performed according to the standard UNI EN 13398 is run after the sample conditioning at intermediate temperatures (usually $25\text{ }^{\circ}\text{C}$) in a ductility water bath. The elastic recovery obtained with this procedure is named ER-DB. Although widely employed, ER tests are purely empirical and the results can be influenced by the operator experience (precision of the half-threads cut, recovery period, elastic recovery measurement, etc.). In addition, the long time required to perform the test and the poor relationship between ER results and the binder performance properties need to be highlighted.

Clopotel & Bahia (2012) proposed a new testing protocol, which aims to characterize elastic recovery behavior of asphalt binders by means of the DSR [36]. The test is performed on STA binders, at a temperature of $25\text{ }^{\circ}\text{C}$ with the 8 mm plate-plate configuration. A constant strain rate of 0.023 1/s is applied for 2 min, which is followed by a zero shear stress (relaxation) period of 1 h. The elastic recovery ER-DSR at the end of the test is calculated as:

$$\text{ER} - \text{DSR} = \frac{\text{recovered strain at the end of the relaxation step } (\epsilon_2)}{\text{strain at the end of loading step } (\epsilon_1)} \cdot 100 \quad (6)$$

Other studies found this procedure to be an excellent alternative to the more empirical ER-DB tests [37–40], whereas the correlation between the measured values of ER-DSR and ER-DB for the investigated STA bituminous blends are presented in Fig. 3. In the graph it is highlighted the good relationship between the two methodologies.

Another methodology to investigate the elastic properties of asphalt binders is the Multiple Stress-Creep Recovery (MSCR) Test. MSCR tests are performed to characterize the bituminous binders in terms of rutting resistance after short-term ageing simulation. In this study, MSCR tests

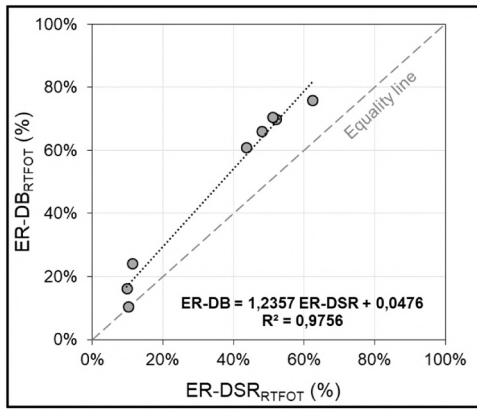


Fig. 3. Correlation between ER-DSR and ER-DB (STA condition).

were carried out following two procedures.

In the first case, the traditional method in accordance to the UNI EN 16659 was followed at a testing temperature of 60 °C, considered the maximum service temperature experienced by the pavement. With this procedure, the tested material is subjected to standardized loading-unloading conditions: 10 cycles of 10 s each divided in 1 s of loading followed by 9 s of recovery time, performed with two stress levels: 0.1 and 3.2 kPa. In the MSCR test, two parameters can be determined: the average non-recoverable creep compliance J_{nr} and the average percent recovery %R of the 10 loading cycles.

In the second case, a different experimental procedure was proposed. The MSCR test performed according to the standardized procedure described above consists in comparing different binders at the same testing temperature and by applying the same stress levels (0.1 kPa and 3.2 kPa). In this manner, a bitumen that is stiffer than another at that testing temperature would be characterized by lower values of the J_{nr} parameter and higher values of the %R parameter. For instance, results obtained from bituminous blends composed of polymer-modified bitumen and different dosages of RAP binder highlighted an increase of %R value (for the same testing temperature) [41]. These findings would suggest that the addition of RAP binder would enhance the elastic properties of the bituminous blends. On contrary, the %R observed when using RAP is higher because the increased stiffness of the blend results in a lower deformation during the loading steps. This lower peak is more likely to remain within the linear domain, making the deformation entirely (or mostly) recoverable.

Since the test is supposed to run in the non-linear domain, in case of very stiff bituminous blends at the testing temperatures, the results obtained only in terms of elastic recovery %R could be misleading. For these reasons, MSCR tests were also carried out in equi-stiffness conditions, i.e. varying the testing temperatures by keeping the norm of the complex modulus $|G^*|$ equal to 500 Pa and 2000 Pa for STA un-modified binders and polymer-modified binders, respectively. The $|G^*|$ threshold were chosen to perform the MSCR tests at testing temperatures equal or above 60 °C.

The testing temperatures were extrapolated from iso-chronic $|G^*|$ master curves defined at a reference frequency of 0.628 rad/s (or 0.1 Hz), assumed as a representative frequency of the traditional MSCR test (1 cycle every 10 s).

In terms of fatigue properties, these are commonly determined according to the Performance Grade classification method for bituminous binders by the $|G^*| \cdot \sin \delta$ parameter (ASTM D6373 and ASTM D8239), measured by means of the DSR with a 8 mm plate configuration on LTA binders. This procedure is performed in the linear visco-elastic domain, keeping the strain levels low enough to not damage the test specimens and varying the temperature by verifying at the same time the condition $|G^*| \cdot \sin \delta \leq 5000 \text{ kPa}$. The last testing temperature that satisfy this condition is considered to be the average medium temperature at

which the bituminous binder guarantees good fatigue properties.

In recent years, Linear Amplitude Sweep test (AASHTO T 391–20) has been widely used in terms of fatigue characterization since considered more accurate in capture the real behaviour of the bituminous binder tests [42,43]. In addition, recent studies validated the strong correlation between LAS tests results on binders and Four-Point Bending Tests results related to the bituminous mixtures [43–45]. The LAS test consists in characterizing the LTA material by an initial frequency sweep at one temperature and 12 frequencies (from 0.2 to 30 Hz). Successively, the test is run using oscillatory shear in strain-control mode at a frequency of 10 Hz. The loading scheme consists of 10 s interval of increased strain amplitude from 0.1 % to 30 %. The specimen failure is determined at a 35 % reduction of the parameter $|G^*| \cdot \sin \delta$.

The fatigue behavior is analyzed by means of the Visco-Elastic Continuum Damage (VECD) model [46]. In particular, fatigue curves were represented according to the following Eq. (7):

$$N_f = A \cdot \gamma_{\max}^{-2\alpha} \quad (7)$$

Where γ is the maximum expected binder strain for a given asphalt pavement; α is a parameter depending on the slope of the storage modulus G_1 vs angular frequency ω in the logarithmic plane determined in the preliminary frequency sweep; A is a coefficient depending on the test frequency, the value of damage at failure D_f , the value of $|G^*|$ when $\gamma = 1$ %, and the parameters of the power law fitting the $|G^*| \cdot \sin \delta$ vs damage D data. LAS tests were carried out at a fixed temperature of 25 °C.

Complex modulus and phase angle results allow to capture non-load associated cracking properties of the bituminous binders by defining the Glover-Rowe (G-R) parameter [47]. Initially, the approach evaluated the possibility to assess cracking properties by means of a DSR function in terms of binder ductility with cracking data related to cores obtained from field test sections. A few years later, the G-R parameter was simplified, and currently a single DSR measurement of the binder complex shear modulus $|G^*|$ and phase angle δ at 15 °C and 0.005 rad/s allows to discriminate different asphalt binders in terms of cracking resistance [48].

The G-R was calculated on LTA bituminous blends using the following Eq. (8):

$$G - R = \frac{|G^*| \cdot (\cos \delta)^2}{(\sin \delta)^2} \quad (8)$$

where $|G^*|$ and δ are the experimental complex shear modulus and phase angle measured at the temperature of 15 °C and angular frequency of 0.005 rad/s. G-R parameter discriminates binders performance in terms of non-load-associated cracking limiting the results with a “damage onset” area ($180 \text{ kPa} < G-R < 600 \text{ kPa}$) and a “significant cracking” area ($G-R > 600 \text{ kPa}$).

3. Results and discussion

The results related to the addition of SNB to the un-modified bitumen PG 58V-22 are compared to the bitumen PG 58E-16, whereas the influence on the polymer-modified bitumen addition is evaluated comparing the bituminous blends results with the base binder PG 58E-22.

3.1. Needle penetration, softening point and viscosity

Fig. 4 shows results in terms of needle penetration of the investigated binders. The values reported are related to both the un-aged and STA conditions, and are group for un-modified and polymer-modified binders. The effect conferred by the addition of SNB is reported in terms of percentage variation related to the binders PG 58E-16 and PG 58E-22 for un-modified and polymer-modified bituminous blends,

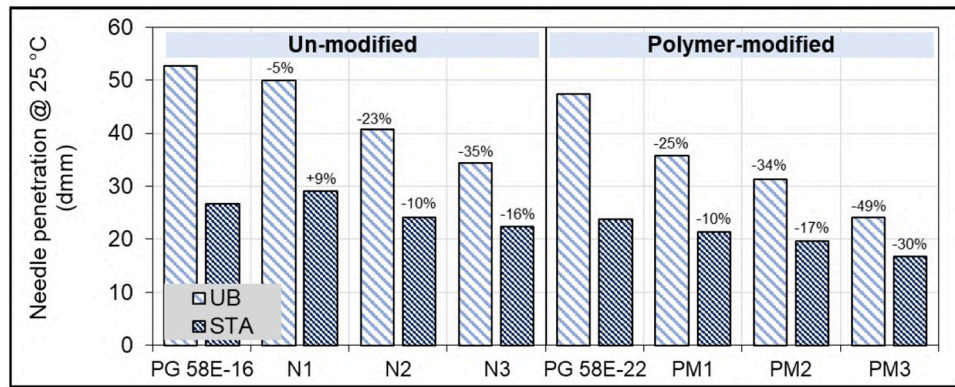


Fig. 4. Needle penetration results.

respectively.

For what concerns the un-aged conditions, SNB leads to a decrease of the penetration values between 5 % and 35 % for the un-modified bituminous blends and between 25 % and 49 % for the modified bituminous blends, related to the respective reference binder.

In terms of STA conditions, the addition of SNB causes a variation in the penetration value between + 9 % and –16 % in case of un-modified bituminous blends and between –10 % and –30 % in case of polymer-modified binders.

It is remarked that the effect of STA is less effective as the SNB dosage increases, for both un-modified and polymer-modified bituminous blends. In fact, results highlight that the higher the SNB dosage, the lower is the gap between the un-aged and the STA conditions of the bituminous blends.

In Fig. 5 the softening point results of the studied bituminous blends are reported, as well as the percentage variation between the SNB bituminous blends and the respective reference binders (PG 58E-16 and PG 58E-22).

In case of un-aged conditions, the addition of SNB results in a softening point variation between –2 % and + 7 % for un-modified bituminous binders and between + 14 % and + 23 % for polymer-modified bituminous blends.

In case of STA conditions, the variation in the softening point conferred by the SNB is between + 1 % and + 9 % for un-modified binders and between –1 % and + 5 % for modified binders.

Considering the effect of STA, softening point temperatures increase of + 15 %, + 14 % and + 15 % for bituminous blends N1, N2 and N3, whereas the values vary of + 3 %, –3 % and + 1 % for bituminous blends PM1, PM2 and PM3. Differently from penetration values, the short-term ageing values of softening point on binders containing SNB is not influenced by the dosage.

The viscosity values recorded at 135 °C and 165 °C on the un-aged

bituminous materials are collected in Fig. 6. The gradual addition of SNB leads to an increase in viscosity at both 135 °C and 165 °C for un-modified and polymer-modified bituminous blends. The increase in viscosity due to the SNB addition is similar at both 135 °C and 165 °C for un-modified binders, i.e. 40 %, 65 % and 100 % for N1, N2 and N3 blends respectively. On contrary, the testing temperature influenced the results of bituminous blends produced with the polymer-modified bitumen. At 135 °C, the increasing dosage of SNB leads to an increase in viscosity of 56 %, 85 % and 130 %, whereas at 165 °C the viscosity increases by 21 %, 38 % and 59 % for PM1, PM2 and PM3 bituminous blends, respectively.

3.2. Complex modulus tests and modelling

Complex modulus results in terms of master curves of the complex shear modulus and phase angles are reported for un-aged and STA binders in Fig. 7 and 8. The reference temperature is 30 °C and the 2S2P1D model is plotted for each bituminous blend. The model parameters adopted for all blends investigated are listed in Table 5.

The results related to the bituminous blends produced with the un-modified binder PG 58V-22 are reported in Fig. 7. The complex modulus master curves (Fig. 7a) of un-aged binders highlight the effect of SNB increasing dosage particularly at low frequencies (or high temperatures), leading to a gradual decrease in the model parameter h and in an increase in the parameter β , linked to the binder viscosity. In terms of phase angle results (Fig. 7b), the gradual addition of SNB confers lower phase angle values and the shifting of the master curves towards lower frequencies (or higher temperatures). This outcome can be linked to the increasing asphaltene content, as reported in literature [49]. In fact, by increasing the SNB content, the model parameter τ_E linked to the oxidative state increases. Furthermore, the glassy modulus G_∞ increases as the SNB dosage increases.

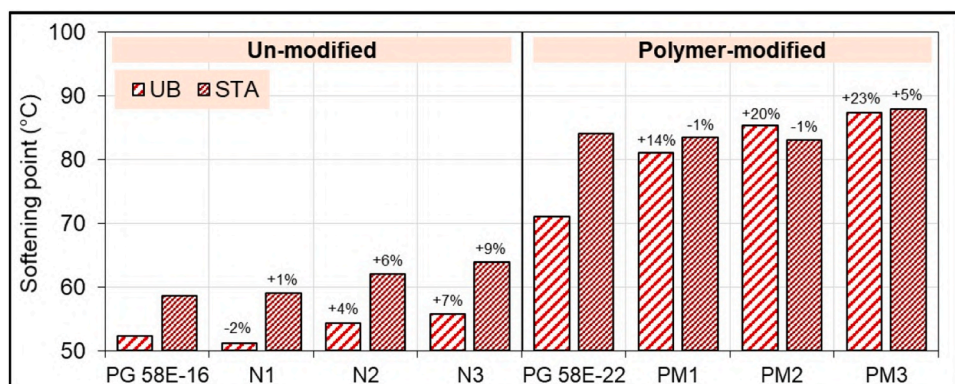


Fig. 5. Softening point results.

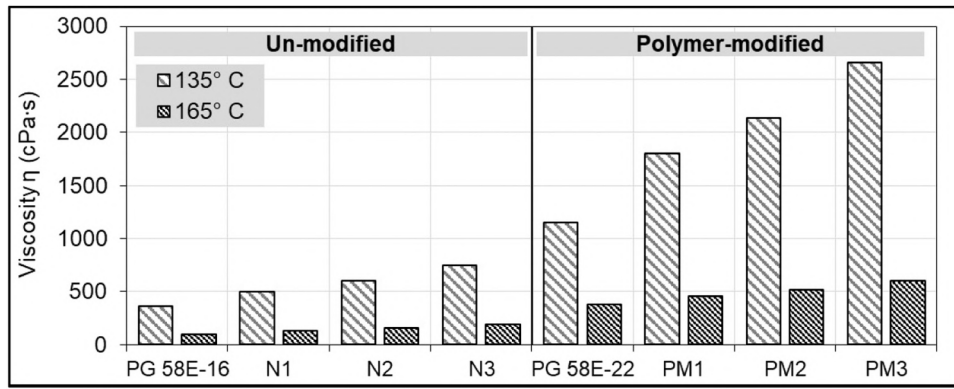


Fig. 6. Viscosity results for the bituminous blends investigated.

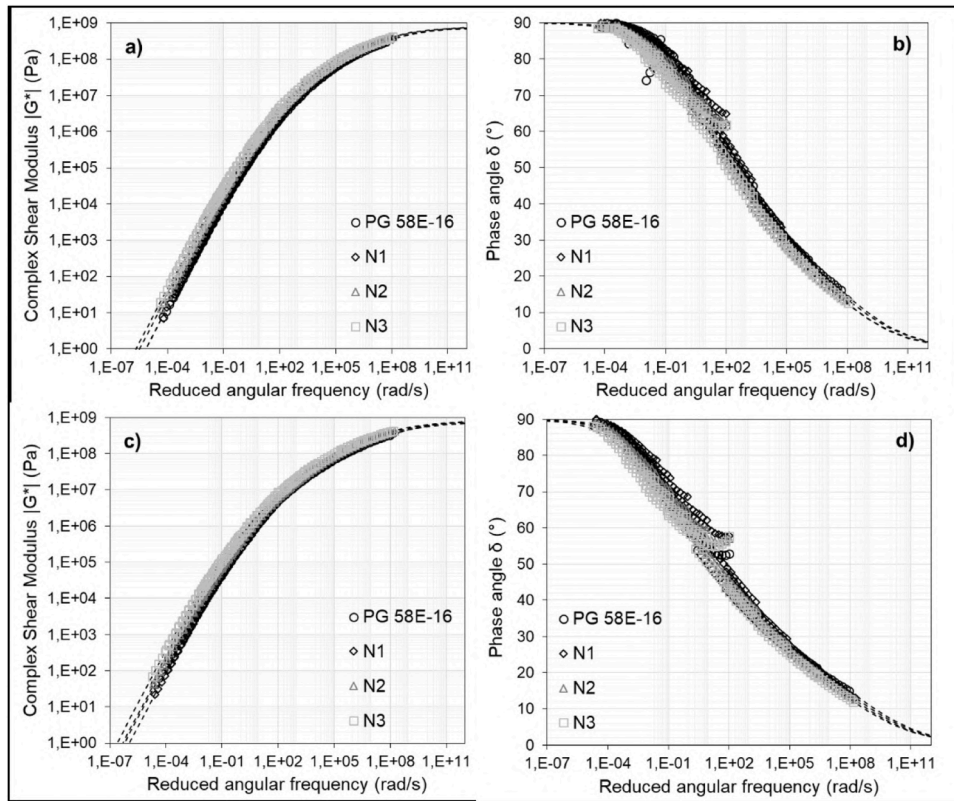


Fig. 7. Master curves and 2S2P1D model ($T_{REF} = 30^\circ\text{C}$) of un-modified binders: a) complex shear modulus $|G^*|$ of bituminous blends in un-aged conditions; b) phase angle δ of bituminous blends in un-aged conditions; c) complex shear modulus $|G^*|$ of bituminous blends in short-term conditions; d) phase angle δ of bituminous blends in short-term aged conditions.

Complex modulus and phase angle master curves after STA highlight different outcomes compared to the un-aged binders (Figs. 7c and d). The bituminous blend N1 shows a lower G_∞ , a higher h parameter at low frequencies/high temperatures and a lower β parameter compared to the reference binder PG 58E-16, meaning that the STA influenced more the reference binder than the bituminous blend N1. On contrary, the bituminous blend N2 and the binder PG 58E-16 were characterized by a similar behaviour and comparable parameters, whereas the bituminous blend N3 shows a higher oxidized state (higher G_∞ , τ_E and β parameters) than the reference binder.

The results related to the bituminous blends produced with the polymer-modified binder PG 58E-22 are reported in Fig. 8.

Complex modulus master curves in un-aged conditions show a marked increase in stiffness due to the SNB addition in the whole

frequency range analysed (Fig. 8a). At the same time, lower phase angle values are observed for SNB bituminous blends especially at low frequencies/high temperatures (Fig. 8b). As previously observed for un-modified blends, this result is linked to the higher asphaltenes content in the SNB bituminous blends [49]. However, in terms of model parameters, the SNB increasing content leads to higher values of G_∞ , even though the reference binder PG 58E-22 is characterized by the highest value. At the same time, the parameter h related to low frequencies/high temperatures is affected by the introduction and the increasing dosage of SNB, which penalized the viscous behaviour of the bituminous blends.

After short-term ageing, the differences among the bituminous blends produced with the polymer-modified binder are less significant (Figs. 8c and d). The bituminous blends containing SNB (PM1, PM2 and PM3) are characterized by similar values of stiffness and phase angle,

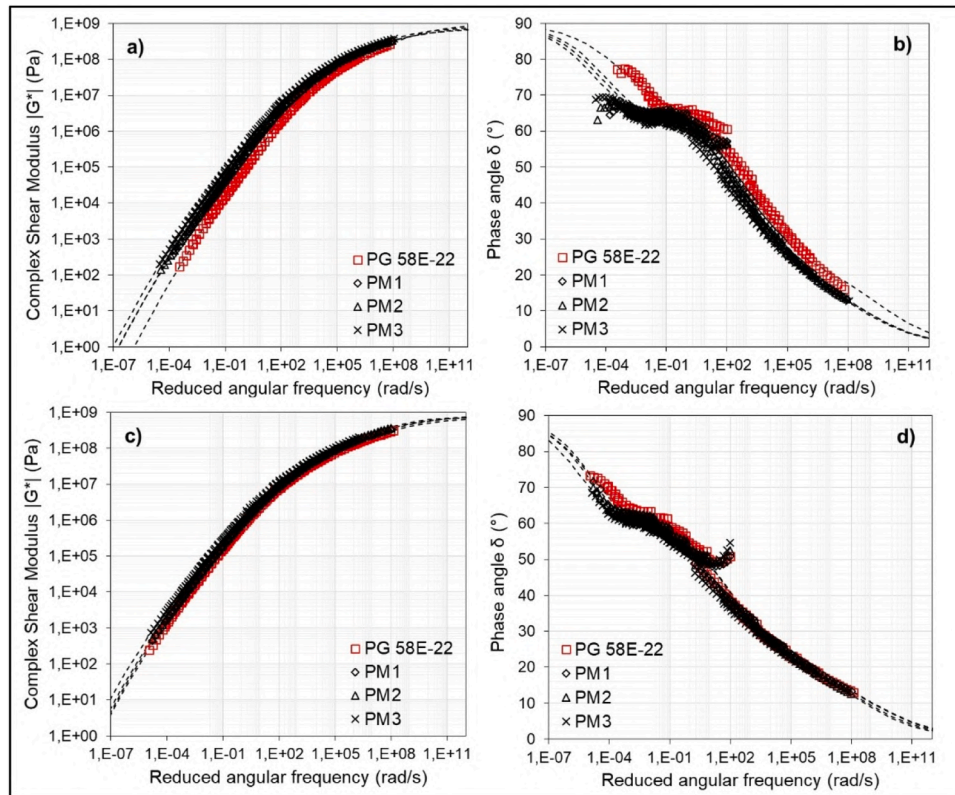


Fig. 8. Master curves and 2S2P1D model ($T_{REF} = 30\text{ }^{\circ}\text{C}$) of polymer-modified binders: a) complex shear modulus $|G^*|$ of bituminous blends in un-aged conditions; b) phase angle δ of bituminous blends in un-aged conditions; c) complex shear modulus $|G^*|$ of bituminous blends in short-term conditions; d) phase angle δ of bituminous blends in short-term aged conditions.

Table 5
2S2P1D model parameters.

	G_{00}	G_{∞}	k	h	δ	τ_E	β
	(Pa)	(Pa)	(-)	(-)	(-)	(s)	(-)
Un-aged binders							
PG 58E-16	0	7.6E+ 08	0.29	0.65	6.0	4.5E-07	65
N1	0	7.6E+ 08	0.28	0.65	5.3	4.5E-07	65
N2	0	7.7E+ 08	0.28	0.65	5.0	7.0E-07	90
N3	0	8.0E+ 08	0.27	0.63	5.0	7.2E-07	130
PG 58E-22	0	1.0E+ 09	0.27	0.70	10.0	3.0E-07	450
PM1	0	7.2E+ 08	0.25	0.66	5.2	6.5E-07	2135
PM2	0	7.5E+ 08	0.26	0.67	6.5	1.0E-06	1200
PM3	0	8.2E+ 08	0.26	0.67	7.5	1.8E-06	1200
Short-term aged binders							
PG 58E-16	0	8.2E+ 08	0.27	0.60	7.4	6.5E-07	487
N1	0	7.7E+ 08	0.27	0.64	7.4	9.1E-07	211
N2	0	8.1E+ 08	0.26	0.62	6.6	1.0E-06	401
N3	0	8.5E+ 08	0.26	0.62	6.6	1.5E-06	501
PG 58E-22	0	7.2E+ 08	0.25	0.65	10.3	4.2E-06	2500
PM1	0	8.3E+ 08	0.25	0.63	9.3	2.6E-06	4000
PM2	0	8.1E+ 08	0.25	0.63	8.5	3.4E-06	4000
PM3	0	8.2E+ 08	0.26	0.64	9.5	7.0E-06	3800

resulting in close values of the 2S2P1D model regardless the SNB dosage. The PG 58E-22 was more susceptible to ageing, confirmed by a significant decrease of the h and β parameters at high temperatures/low frequencies.

3.3. δ -method

Fig. 9 and 10 show results in terms of AMWD of un-modified and polymer-modified bituminous blends, respectively. The results shown are related to both the un-aged and STA conditions. The AMWD of the

investigated blends were obtained by the analysis of the phase angle master curves modelled through the 2S2P1D model.

In terms of un-modified bituminous blends (Fig. 9a), the un-aged binders analysis highlights a small peak related to fractions with low AMW ($\leq 300\text{ g/mol}$) and a major concentration of molecules fraction with AMW of $\approx 500\text{ g/mol}$, which could both be related to the maltene fractions. All the binders investigated, including the SNB bituminous blends, do not show peaks in the part of the graph related to higher AMW ($\approx 1000\text{ g/mol}$), generally associated to the presence of asphaltene agglomeration [31]. However, with the incremental addition of SNB in un-modified bitumen, it is possible to observe a shifting of the AMW distribution curves towards higher values, meaning that SNB leads to an increase of the global MW without asphaltene agglomeration.

After short-term ageing, more molecular fractions are highlighted in the bituminous blends studied, since three peaks are clearly visible (Fig. 9b). In addition to the fractions with AMW $\approx 500\text{ g/mol}$ and $\approx 300\text{ g/mol}$ previously detected, a significant portion of elements is characterized by a value of AMW $\approx 1000\text{ g/mol}$. The effect of ageing has indeed caused the asphaltene agglomeration in all the binders; it is observed that the higher concentration of molecules with AMW $\approx 1000\text{ g/mol}$ is related to the PG 58E-16 binder, whereas the bituminous blends with SNB are characterized by lower concentration values, confirming the lower ageing susceptibility of SNB.

In terms of polymer-modified bituminous blends, the influence of SNB addition is limited at un-aged conditions, except for the bituminous blend PM3 with the highest SNB dosage (Fig. 10a). The reference binder PG 58E-22 and the bituminous blends PM1 and PM2 show two main fractions concentrations, at AMW $\approx 300\text{ g/mol}$ and $\approx 500\text{ g/mol}$, and no presence of particles with higher AMW. The bituminous blend PM3 shows two major concentrated groups, characterized by an AMW of $\approx 200\text{ g/mol}$ and $\approx 500\text{ g/mol}$, with a lower concentration of fractions with AMW of $\approx 300\text{ g/mol}$ compared to the rest of the materials

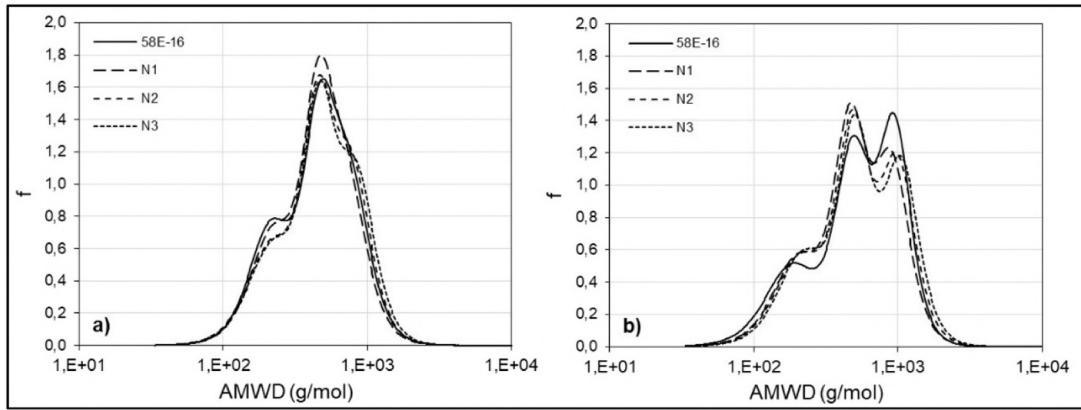


Fig. 9. AMWD of un-modified bituminous blends: a) un-aged conditions; b) STA conditions.

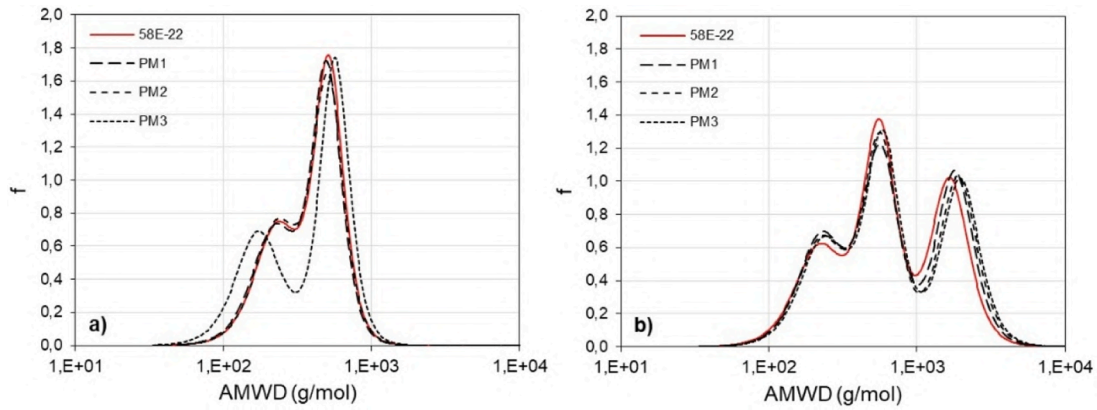


Fig. 10. AMWD of polymer-modified bituminous blends: a) un-aged conditions; b) STA conditions.

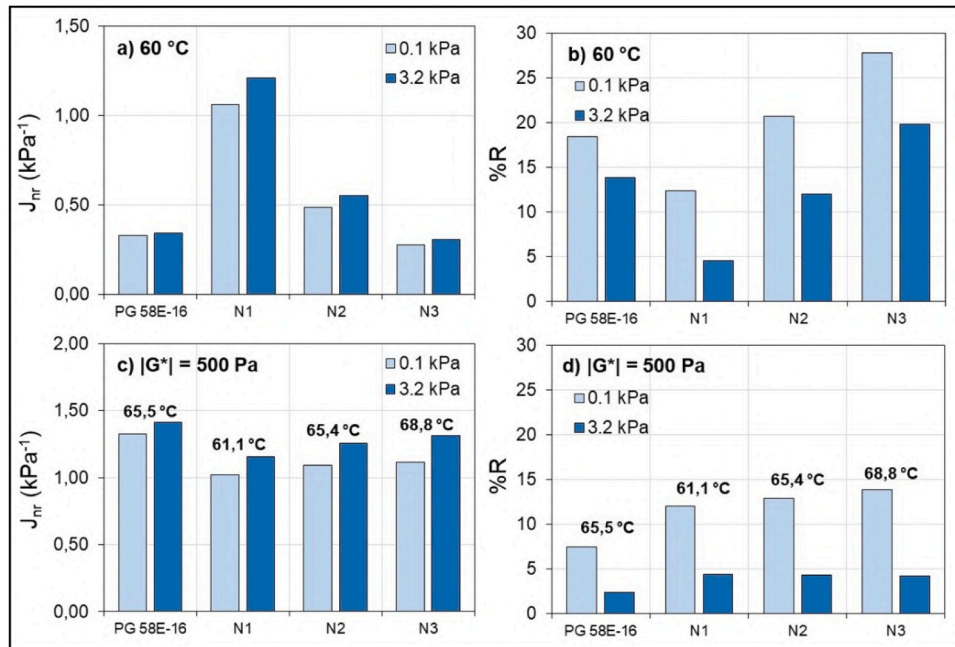


Fig. 11. MSCR tests results of un-modified binders: a) J_{nr} at 60 °C; b) %R at 60 °C; c) J_{nr} at $|G^*| = 500$ Pa; d) %R at $|G^*| = 500$ Pa.

investigated. This results could be caused by the contemporary presence of polymers and the maximum amount of SNB introduced (20 % by the bituminous blend mass), which led to a lower concentration of light fractions (maltenes).

Results in terms of STA conditions highlighted the presence of three main concentrated groups, with AMW values of ≈ 200 g/mol and ≈ 500 g/mol (related to lighter fractions), and ≈ 2000 g/mol (related to asphaltene agglomeration conferred by ageing) (Fig. 10b). However, the results obtained do not highlight significant difference among the blends investigated.

3.4. Elastic recovery characteristics

Results from MSCR tests performed on un-modified and polymer-modified bituminous blends are reported in Fig. 11 and 12, respectively. Values are shown in terms of non-recoverable creep compliance J_{nr} and percent recovery %R obtained from the tests performed at 60 °C and in equi-stiffness conditions ($|G^*|$ equal to 500 Pa and 2000 Pa obtained from complex modulus iso-chronic master curves). In the second case, the testing temperature adopted is also reported.

The results related to the un-modified bituminous blend obtained at 60 °C highlight that the addition of SNB leads to a decrease in the J_{nr} value (Fig. 11a) and in an increase of the %R values (Fig. 11b), because of the stiffening effect conferred by the increasing asphaltene content [50]. The bituminous blends N3 shows similar properties at 60 °C compared to the PG 58E-16 bitumen.

The results related to the equi-stiffness conditions highlight that the increase in stiffness of the bituminous blends containing SNB led to an increase of the testing temperature. In this case, the results obtained in terms of J_{nr} do not remark a significant difference among the mixtures containing SNB, which showed comparable performance at both stress levels (Fig. 11c). However, the PG 58E-16 bitumen is characterized by higher values of J_{nr} , highlighting lower performance in terms of permanent deformation compared to the SNB bituminous blends. On the other hand, the percent recovery %R related to the bituminous blends with SNB is significantly higher than the PG 58E-16 bitumen, and it is observed an increase in the value recorded with a higher SNB dosage (only in case of 0.1 kPa stress level) (Fig. 11d).

Figs. 12a and 12b highlight that, at the testing temperature of 60 °C,

Table 6

Percent recovery %R obtained from MSCR tests and elastic recovery ER-DSR.

Binders	MSCR @ 60 °C		MSCR @ equi - $ G^* $		ER-DSR @ 25 °C
	0.1 kPa	3.2 kPa	0.1 kPa	3.2 kPa	
PG 58E-16	18.4 %	13.8 %	7.5 %	2.3 %	9.8 %
N1	12.4 %	4.5 %	12.0 %	4.4 %	10.2 %
N2	20.7 %	12.0 %	12.9 %	4.3 %	12.1 %
N3	27.8 %	19.8 %	13.9 %	4.2 %	11.4 %
PG 58E-22	74.3 %	66.5 %	67.7 %	60.2 %	64.4 %
PM1	79.5 %	73.7 %	75.4 %	68.5 %	51.0 %
PM2	80.5 %	74.6 %	75.0 %	67.6 %	48.1 %
PM3	82.3 %	76.7 %	73.7 %	66.2 %	43.6 %

the addition of SNB to the PG 58E-22 modified binder causes a decrease in the J_{nr} value and an increase in the percent recovery %R. However, results are influenced by the increasing SNB dosage in PM1, PM2 and PM3 bituminous blends, due to the increase in asphaltene concentration [50].

On the other hand, results related to the MSCR test performed at equi-stiffness conditions on polymer-modified bituminous blends confirmed the outcomes from the previous section on un-modified binders (Figs. 12c and d). In this case, the addition of SNB to the PG 58E-22 binder leads to a decrease in the J_{nr} values and an increase in the %R, with no influence conferred by the varying SNB dosage.

In Table 6, the values of percent recovery %R obtained from the MSCR tests and the elastic recovery ER-DSR obtained with the procedure previously described are compared. For what concerns the MSCR tests performed at 60 °C, the %R values tend to increase with the increase in stiffness of the tested material. This is observed for both the un-modified and the polymer-modified bituminous blends. However, it is possible to observe that values obtained with a stress level of 0.1 kPa in equi-stiffness conditions are comparable to the ER-DSR values detected at 25 °C for un-modified bituminous blends. Since it was previously shown that the ER-DSR has a good correlation with the “traditional” elastic recovery values (ER-DB – Fig. 3), it is possible to assess that the bituminous blends containing SNB, regardless the dosage, have higher elastic recovery values than the un-modified bitumen PG 58E-16.

For what concerns polymer-modified bituminous blends, the results obtained from MSCR tests performed in equi-stiffness conditions at both

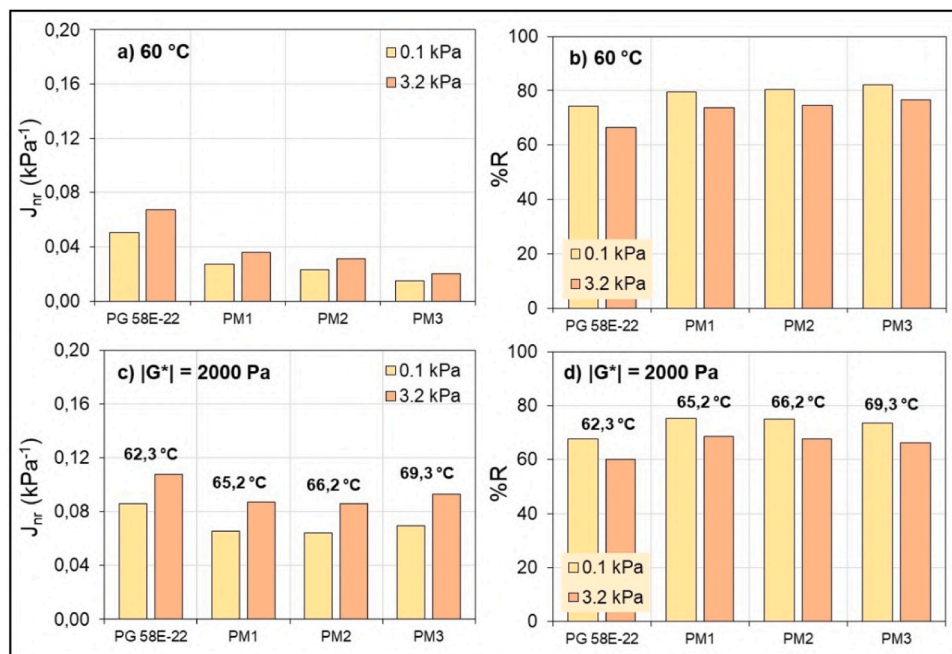


Fig. 12. MSCR tests results of polymer-modified binders: a) J_{nr} at 60 °C; b) %R at 60 °C; c) J_{nr} at $|G^*| = 2000$ Pa; d) %R at $|G^*| = 2000$ Pa.

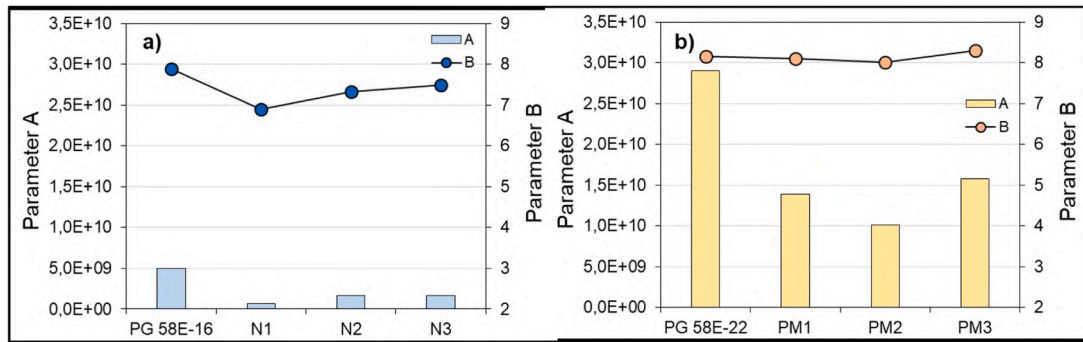


Fig. 13. LAS Test results: a) un-modified bituminous blends, b) polymer-modified bituminous blends.

stress levels show a decreasing value as the SNB dosage increases, as well as the results in terms of ER-DSR. Consequently, as for the un-modified bituminous binders, the SNB used with polymer-modified bitumen causes a lower elastic recovery performance, although in this case the results obtained between the two procedures are not directly correlated.

3.5. Linear Amplitude Sweep (LAS) Test

Fatigue characteristics of the investigated materials were assessed by Linear Amplitude Sweep (LAS) tests at 25 °C, which results are shown in Fig. 13. The fatigue resistance is reported in terms of parameters A and B, both related to the fatigue curves of the bituminous blends analysed. The parameter A is linked to the material ability to accumulate damage during the loading cycles keeping its integrity, while the parameter B represents the sensitivity of the material to the strain level change. In general, good fatigue performance of asphalt binders are given by higher values of parameter A and lower values of parameter B [42].

Fig. 13a shows results related to the un-modified bituminous blends. The values obtained for the A parameter highlight the higher values for the PG 58E-16 binder compared to the blends containing SNB, indicating that adding SNB penalizes the binders resistance to resist damage. At the same time it can be observed that the B parameter decreases by adding the SNB to the base binder, and it increases if the SNB dosages increases. Such results suggest that by adding SNB to the un-modified binder the blend obtained is less sensitive to the strain level change, but the increasing dosage of SNB (above 10 %) penalizes this characteristic.

Fig. 13b shows results related to the polymer-modified bituminous blends. Parameter A values show the highest result related to the PG 58E-22 binder and by adding SNB to the bituminous blends the ability to accumulate damage is penalized. On contrary, in this case parameter B results do not significantly discriminate the bituminous blends analysed.

In both cases analyzed (un-modified and polymer-modified blends), it was observed that the A parameter is more influenced by the addition of SNB than the B parameter, and the A parameter of the bituminous blends containing SNB resulted lower than the binders without SNB. As shown in other studies, the loss in fatigue properties could be linked to the higher asphaltenes concentration of the SNB bituminous blends [50].

3.6. Glover-Rowe (G-R) parameter

Results in terms of G-R parameter are shown in Fig. 14, where the two damage thresholds (onset and cracking) are also reported. In general, a higher G-R parameter represents worse non-load related cracking properties. All the bituminous binders analysed showed G-R values above the significant cracking limit, highlighting a global poor resistance to cracking.

The un-modified binder PG 58E-16 is characterized by the highest G-R value, meaning that among the un-modified bituminous blends it shows the worst cracking properties. At the same time, the bituminous blends produced with the PG 58V-22 bitumen and SNB showed an increase in the G-R value related to the incremental dosage of SNB, highlighting that the SNB addition has a negative influence on the cracking properties.

The polymer-modified bitumen PG 58E-22 and the bituminous blends produced (PM1, PM2 and PM3) are all characterized by close G-R values, significantly above the damage onset threshold. However, the use of SNB and its incremental dosage penalize the cracking properties of the polymer-modified binders.

4. Conclusions

The research focused on the effect conferred by the use of Selenice

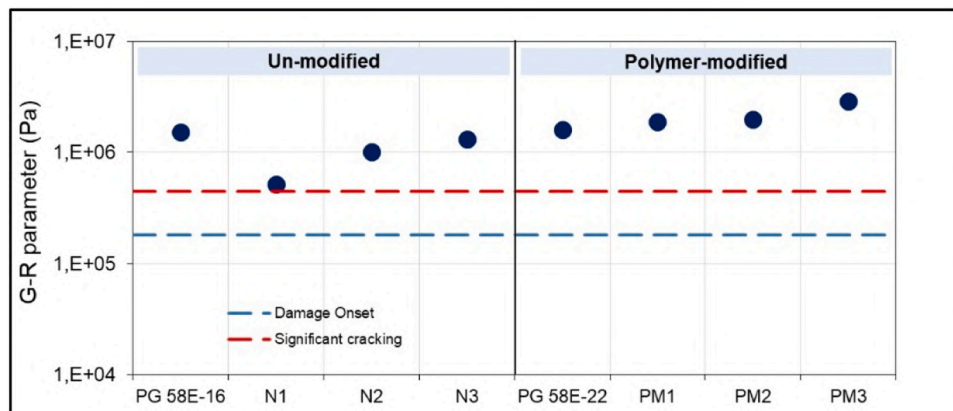


Fig. 14. G-R parameter values.

Natural Bitumen (SNB) to the rheological properties of bituminous blends produced with an un-modified and a polymer-modified binders (PG 58V-22 and PG 58E-22, respectively). In both cases, the SNB was dosed at 10 %, 15 % and 20 % by the total bituminous blend mass. The experimental program involved the investigation of different aspects, some of which analysed in un-aged and short-term ageing conditions of the binders. The study led to the following outcomes:

- Results related to un-modified bituminous binders highlighted that the incremental dosage of SNB is more influent in un-aged conditions rather than in STA conditions. At high temperatures, the use of SNB results in better rutting properties, by decreasing the permanent deformation and by not significantly varying the elastic recovery. In terms of cracking resistance, SNB penalizes the fatigue life at intermediate temperatures (especially at low and intermediate strain levels) but it confers better non-load related cracking properties (lower G-R parameter).
- Results related to SBS polymer-modified bituminous binders remarked a lower influence conferred by SNB in short-term aged conditions compared to un-aged conditions. In terms of rutting characterization, the increasing dosage of SNB leads to both lower permanent deformation and percent recovery values. At the same time, cracking resistance properties confirmed the detrimental effect of the use of SNB with the polymer-modified bitumen. It has to be highlighted that the results obtained only refer to one type of polymer, and different polymer types combined with SNB could lead to different outcomes.

Concluding, the analysis conducted after short-term ageing highlighted that the negative effect brought by the SNB introduction in bituminous blends is less remarkable. According to the results obtained, the SBS polymers included in the polymer-modified bitumen did not hinder the negative effect conferred by the SNB to the bituminous blend, even at low dosages (10 %).

Considering the petrographic nature of natural bitumens, further researches should focus on the use of different polymers and rejuvenating additives, in order to limit the negative effects related to the use of such materials. Of course, both alternatives shall be evaluated in terms of ecological, economical and performance outcomes.

CRediT authorship contribution statement

Simone Raschia: Methodology, Formal analysis, Data curation, Conceptualization, Writing – review & editing, Writing – original draft, Supervision. **Stefano Tattolo:** Supervision, Project administration, Funding acquisition, Conceptualization. **Giovanni Marchegiani:** Methodology, Investigation, Data curation.

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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