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Influence of mineral fillers on the rheological response of polymer-modified bitumens and mastics



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ABSTRACT

The rheological properties of the bituminous components (bitumen and bituminous mastic) within asphalt mixtures contribute significantly to the major distresses of flexible pavements (i.e. rutting, fatigue and low temperature cracking). Asphalt mixtures are usually composed of mastic-coated aggregates rather than pure bitumen-coated aggregates. The purpose of this study is to investigate the effects of mineral fillers on the rheological behaviour of several polymer-modified bitumens (PMBs) through laboratory mixing. A neat bitumen and two types of polymers (elastomeric and plastomeric) were used to produce PMBs, and two fillers with different minerals (limestone and basalt) were selected to obtain mastics. The dynamic shear rheometer (DSR) and bending beam rheometer (BBR) were used to characterize the rheological properties of PMBs and mastics. In particular, multiple stress creep recovery (MSCR) tests were performed to evaluate the rutting potential at high temperatures, whereas BBR tests were carried out to investigate the low temperature behaviour of these materials. BBR results for unmodified mastics show that the increase of stiffness is similar regardless of the filler type, whereas results for polymer-modified mastics indicate that the degree of stiffening depends on the combination of filler/polymer types. MSCR results show that adding filler leads to a reduced susceptibility of permanent deformation and an enhanced elastic response, depending on the combination of filler/polymer types. Overall results suggest that a physical–chemical interaction between the filler and bitumen occurs, and that the interaction level is highly dependent on the type of polymer modification.

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1. Introduction

The rheological properties of the bituminous components of asphalt mixture contribute significantly to the major distresses of flexible pavements. In particular, bitumen and mineral fillers create a blend known as bituminous mastic that, depending on its nature and composition, significantly influences the durability of pavement asphalt layers.

Playing a dual role in asphalt mixture, the filler is a mineral aggregate that fills the interstices between larger aggregates. It provides a higher number of contact points and, when mixed with bitumen, improves bitumen consistency by enhancing the bonds between larger aggregate particles. Presently, asphalt mixture should be considered a mixture comprising mastic-coated aggregates rather than pure bitumen-coated aggregates (Wang et al., 2011).

The main effect of mineral filler is to stiffen the bitumen, consequently to improve the asphalt mixture's mechanical properties. The stiffening effect of the mastic controls the mixture performance from the construction stage through the entire service life of the pavement. During transportation and paving processes, poor stiffness within the mastic can cause drain-down problems, whereas excessive stiffness may lead to compaction difficulties and workability reduction (Brown et al., 2009). The mastic stiffness affects the ability to resist permanent deformation at high temperatures, fatigue life at intermediate temperatures and cracking resistance at low temperatures (Airey et al., 2006; Johnsson and Isacson, 1998; Wang et al., 2011).

The stiffening effect of mineral fillers on bitumen has been extensively investigated by many researchers (Faheem et al., 2008; Faheem and Bahia, 2010; Grabowski and Wilanowicz, 2008; Harris and Stuart, 1995; Kim et al., 2003; Liao et al., 2012; Wang et al., 2011). These studies show that the filler volume concentration and physical properties, such as size distribution, shape, texture, void space and surface area, of mineral fillers, along with the physical–chemical interaction between the filler and bitumen, affect the stiffness of the mastics. According to Buttler et al. (1999), the behaviour of mastics can be divided into three reinforcement mechanisms: volume-filling reinforcement (i.e. the presence of a rigid inclusion in a less rigid matrix causes the stiffening effect), physical–chemical reinforcement (the stiffening is caused by interfacial effects between the bitumen and filler particles), and particle–interaction reinforcement. The last mechanism has a limited role in the stiffening effect of the filler and is usually considered negligible (Shashidhar and Romero, 1998). Furthermore, laboratory investigations based on rheological measurements show that mastics can be considered thermorheologically simple linear viscoelastic materials (Anderson and Goetz, 1973), and the mechanical influence of

fillers can be assessed through a viscoelastic analysis of the mastics (Kim and Little, 2004; Underwood and Kim, 2015).

Given the fundamental role of mastics in asphalt mixtures, their mechanical characterization is essential in improving the understanding of asphalt mixture performance with respect to common asphalt pavement distresses. This paper presents a laboratory investigation on different bituminous mastics using a rheology-based approach. In particular, the study investigates the effects of fillers with different mineralogy on the high and low temperature behaviour of both neat and polymer-modified bitumens (PMBs). Specifically, the use of both elastomeric and plastomeric polymers allowed the effects of the filler/polymer type combination on the rheological properties of the mastics to be evaluated. This line of research was chosen because current studies mostly focus on filler properties and place less importance on the polymer modification of bitumens.

2. Laboratory investigation

2.1. Materials

A neat bitumen, selected as the reference bitumen, was used to produce two PMBs by adding 2 types of polymers to the same content. Subsequently, the neat bitumen and PMBs were used to produce 6 mastics by adding 2 types of fillers, which were dosed at a constant filler/bitumen ratio according to the identification codes summarized in Table 1.

The neat bitumen was a 70/100 penetration-grade bitumen from an Italian oil refinery. Regarding the production of PMBs, a radial styrene-butadiene-styrene (SBS) polymer containing 30% styrene with a density of 0.94 g/cm³ and polyolefin (PO) polymer containing an ethylene and propylene blend with a density of 0.94 g/cm³ were selected as modifying agents. The two different types of polymers (elastomer and plastomer) were selected because of their different reinforcement mechanisms within bitumen. The polymer content adopted for the PMBs was 4% of the base bitumen's weight.

All modified bitumens were produced in the laboratory using a ROSS high-shear mixer operating at a rotation speed of 3000 rpm at 180 °C for 3 h. Initially, 700 g of bitumens in a 1000 mL cylindrical can were heated to a fluid condition. Upon reaching 180 °C, the polymer was added slowly to the bitumen to prevent any polymer separation during the mixing process. Table 2 shows the physical properties of the neat and polymer-modified bitumens.

The two natural fillers with different minerals (i.e. basalt and limestone) were selected and consisted of particles passing the 0.063 mm sieve. Table 3 lists some basic properties of the fillers.

Table 1 – Codes for bitumens and mastics.

Polymer type	Polymer content (%)	Bitumen code	Mastic code	
			Basalt filler	Limestone filler
No polymer	–	B_Neat	M1_Unmodified	M2_Unmodified
SBS	4	B_SBS4	M1_SBS4	M2_SBS4
PO	4	B_PO4	M1_PO4	M2_PO4

Table 2 – Physical properties of the neat and the polymer-modified bitumens.

Material	Penetration at 25 °C (0.1 mm) (EN 1426)	Softening point (°C) (EN 1427)	Dynamic viscosity at 135 °C (Pa·s) (EN 12595)
B_Neat	72	47.7	0.31
B_SBS4	54	69.5	0.91
B_PO4	53	55.8	0.78

The mastics were produced following an optimized protocol to obtain homogeneous bitumen-filler mixes. The mineral filler and bitumen were heated in the oven at 170 °C for 2 h. Then the filler was slowly added to the bitumen and blended with a mechanical mixer at 170 °C. The rotation speed was gradually increased in proportion to the increase of the material density. Filler addition was completed in about 10 min, and the mixing process was continued for 30 min to avoid filler segregation.

A constant filler-bitumen (by mass) ratio of 1.2 was selected in accordance with superpave specifications (Kennedy et al., 1994), which recommend a ratio within the range of 0.6–1.2. In terms of filler concentration by total mastic volume, the filler-bitumen ratio results in volume fractions of 31.3% for limestone and 31.5% for basalt. These values are so similar because of the close particle density of the fillers. According to the conceptual model proposed by Faheem and Bahia (2009), these filler concentrations fall within the diluted region, where the available bitumen is not completely influenced by the filler.

In order to reproduce real field conditions as accurately as possible, the materials produced were both short- and long-term aged. Bitumens were short-term aged using the rolling thin-film oven (RTFO) procedure, whereas the mastics were short-term aged using the thin-film oven (TFO) procedure. Finally, long-term ageing was simulated through the pressure ageing vessel (PAV) procedure for both the bitumens and mastics.

2.2. Testing program and protocols

A dynamic shear rheometer (DSR) was used to evaluate the rutting potential of laboratory produced bituminous mastics

at high temperatures, whereas a bending beam rheometer (BBR) was used to investigate their low temperature properties.

Rutting behaviour was evaluated through multiple stress creep recovery (MSCR) tests according to ASTM (2010). These tests provide the means to investigate reversible and non-reversible strain behaviours, as well as the stress dependence of bituminous materials. The MSCR tests were performed using parallel plate geometry (25 mm plate diameter) and a gap equal to 1 mm. Tests consisted of 10 creep-recovery cycles with a creep loading time of 1 s and recovery time of 9 s. The MSCR tests were conducted on short-term aged materials in accordance with specifications. In order to evaluate the temperature as well as the stress dependency of both bitumens and mastics, four testing temperatures (58 °C, 64 °C, 70 °C and 76 °C) and two stress levels (0.1 and 3.2 kPa) were selected. An additional stress level of 10 kPa was considered exclusively for the mastics.

BBR was used to investigate the propensity of bitumens and mastics to thermal cracking at low service temperatures in accordance with European Committee for Standardization (2012), including the measurement of a creep test in which the deflection of a beam specimen under a constant load of 0.98 N was applied for 240 s at a controlled temperature. By loading the beam and measuring the central deflection continuously, the creep stiffness ($S(t)$) was calculated as a function of the loading time and temperature. Moreover, the rate at which creep stiffness changes with loading time (m -value) is taken into account as a measure of the material's stress relaxation capability. After 1 h of conditioning, tests were carried out on long-term aged materials for bitumens at -6 °C, -12 °C, -18 °C and -24 °C, as well as for mastics at 0 °C, -6 °C, -12 °C, and -18 °C.

The two replicate performances were completed for each testing condition, and the average value was used for the results analysis.

Table 3 – Basic properties of mineral filler.

Property	Limestone	Basalt
Particle density (g/cm ³)	2.712	2.690
Carbonate content (% by mass)	95.4	6.8
Plasticity index	NP	NP
Rigden voids (%)	36.8	33.5
Particle size distribution (µm)	(% passing by mass)	
63.0	100.0	100.0
58.2	92.7	96.0
42.7	88.9	85.0
31.2	76.2	77.1
22.9	66.7	67.5
12.3	56.3	56.4
7.4	42.5	43.6
4.7	32.8	30.4
3.4	25.7	22.0
1.5	16.6	7.1
Fineness modulus	3.6	3.5

3. Results and analysis

3.1. Low temperature resistance

Table 4 reports the values for creep stiffness ($S(60)$) measured at 60 s and for the rate at which the creep stiffness changes with loading time ($m(60)$) for all materials at each testing temperature.

As expected, the creep stiffness decreases as the temperature increases for both bitumens and mastics, whereas the m -value tends to increase, indicating that the materials become less stiff and more prone to relax stress over time.

Table 4 – BBR test results in terms of $S(60)$ and $m(60)$ of bitumens and mastics.

Material	$S(60)$ (MPa)					$m(60)$				
	–24 °C	–18 °C	–12 °C	–6 °C	0 °C	–24 °C	–18 °C	–12 °C	–6 °C	0 °C
B_Neat	915	471	202	81	–	0.20	0.27	0.37	0.46	–
B_SBS4	679	436	186	80	–	0.18	0.25	0.34	0.42	–
B_PO4	961	543	291	105	–	0.16	0.24	0.31	0.40	–
M1_Unmodified	–	1572	854	371	132	–	0.21	0.30	0.40	0.50
M1_SBS4	–	1354	809	319	143	–	0.20	0.28	0.37	0.47
M1_PO4	–	1701	965	480	216	–	0.20	0.26	0.36	0.46
M2_Unmodified	–	1579	847	387	148	–	0.21	0.30	0.39	0.50
M2_SBS4	–	1284	738	359	143	–	0.20	0.28	0.39	0.48
M2_PO4	–	1663	1024	466	207	–	0.18	0.26	0.34	0.43

Regarding bitumens, the stiffness measured at each temperature is affected by the presence of polymers. At low temperatures, the SBS modified bitumen shows lower creep stiffness than the neat bitumen. However, the difference tends to decrease substantially as the temperature increases, implying that the presence of SBS helps prevent excessive thermal stress at low in-service temperatures without any significant influence on stiffness at higher temperatures. On the contrary, the PO polymer stiffens the bitumen at all testing temperatures, as observed from the highest $S(60)$ values, thus making the material more prone to brittle failure. The presence of both polymers tends to decrease the m -value, indicating that the stress relaxation capability of PMBs is lower than the corresponding bitumen. This effect is more evident with PO modified bitumens.

Regardless of bitumen and filler type, the effects of the mineral filler on low temperature properties of bitumen are evident at all testing temperatures for the mastics. In particular, the filler caused stiffening, as shown by higher $S(60)$ values, as well as a decrease in the relaxation capability (lower $m(60)$) of the materials. Moreover, the effect of polymers on mastics was found to be similar to that on bitumens, and the SBS polymer guarantees a softening of both mastics unlike the presence of polymer PO, which results in an increase in the stiffness of materials. A slight decrease in relaxation capability due to the polymers was also identified.

The $S(60)$ results in Table 4 show that the unmodified mastics demonstrate similar results, whereas differences in stiffness, especially at lower temperatures, occur between the two mastics produced with the same PMB type but

different fillers (i.e. M1_SBS4 vs. M2_SBS4, M1_PO4 vs. M2_PO4). No appreciable difference between the stress relaxation performances of mastics was noted. Overall, a clear and univocal filler type effect within the temperature range investigated cannot be highlighted.

In order to better understand the stiffening effect due to the mineral filler, a global rheological response of the bitumens over a wide range of loading times and temperatures is required. To that end, the master curves of the creep stiffness modulus were built using the time-temperature superposition principle for both bitumens and mastics. The isothermal curves were shifted to a reference temperature of –12 °C, and the creep stiffness modulus, as a function of the reduced loading time, was fitted using a second degree polynomial function in accordance with [European Committee for Standardization \(2012\)](#).

$$\lg(S(t)) = A + B \lg(t) + C(\lg(t))^2$$

where $S(t)$ is the measured creep stiffness at time t (MPa), t is the reduced loading time (s) obtained by the product of the test time and temperature shift factor a_T , A , B , C are experimental coefficients. The shift factors and coefficient values obtained by a regression analysis are reported in Table 5.

Regarding bitumens, the PMBs shift factors decreased at temperatures lower than –12 °C (reference temperature), whereas higher shift factors were observed at higher temperatures with respect to the neat bitumen. In particular, bitumen modified with polymer PO shows the lowest and highest values. Thus, the temperature susceptibility of bitumens is influenced by the polymer modification. A similar

Table 5 – BBR master curves: shift factors and regression coefficients of bitumens and mastics.

Material	$\lg(a_T)$				A	B	C
	–24 °C	–18 °C	–6 °C	0 °C			
B_Neat	–2.29	–1.12	0.98	–	5.84	–0.23	–0.04
B_SBS4	–2.26	–1.01	1.06	–	5.80	–0.21	–0.03
B_PO4	–2.22	–0.98	1.26	–	5.91	–0.19	–0.03
M1_Unmodified	–	–1.21	0.97	2.01	3.35	–0.16	–0.04
M1_SBS4	–	–0.95	1.08	2.02	3.27	–0.13	–0.04
M1_PO4	–	–1.00	0.92	1.71	3.35	–0.12	–0.05
M2_Unmodified	–	–1.04	0.98	1.86	3.34	–0.16	–0.04
M2_SBS4	–	–1.01	0.93	1.86	3.25	–0.13	–0.05
M2_PO4	–	–0.97	1.12	2.04	3.34	–0.12	–0.04

trend can also be identified for mastics such as limestone filler (M2), whereas a change in temperature susceptibility was noticed for mastics containing basalt filler (M1). In fact, the SBS polymer-modified mastics showed lower and higher shift factor values at temperatures lower and higher than $-12\text{ }^{\circ}\text{C}$ with respect to PO polymer-modified mastic. An analysis of regression coefficients (i.e. the shape of the curve) also indicates that the filler effect does not result in a mere vertical shifting of the corresponding base bitumen master curve, and reveals that the viscoelastic behaviour of mastics can be both bitumen and filler dependent.

In order to evaluate the stiffening effect due to the mineral filler type on both neat and polymer-modified bitumens, the master curves of all mastics at the reference temperature of $-12\text{ }^{\circ}\text{C}$ were compared. Fig. 1 shows the stiffness comparison in terms of the stiffness ratio defined as the ratio between mastic stiffness and corresponding bitumen stiffness versus reduced loading time.

Interestingly, the stiffness ratio of the unmodified mastics is higher than the stiffness ratios of both polymer-modified mastics overall loading times, indicating that the stiffening effect of fillers is more significant on the neat bitumen than on the PMBs. Moreover, in the case of unmodified mastics, the stiffness ratio tends to increase as the loading time increases, implying that the stiffening effect of the filler is more relevant at long loading times or high temperatures. In fact, at high temperatures, the difference in stiffness between the bitumen and filler becomes significant as the bitumen experiences viscous flow behaviour, whereas the filler, being the rigid component, is responsible for a stiffer response of the mastic (Chen et al., 2008).

On the other hand, both polymer-modified mastics show a different behaviour as a function of loading time with respect

to the unmodified mastics. In particular, the stiffness ratio tends to increase with the loading time until the curves approach the maximum values, after which the stiffness ratio starts to decrease as the loading time increases. This means that at a high loading time (i.e. higher temperature), the increase in relative stiffness caused by the filler diminishes in relation to the polymer contribution within the bitumen. Therefore, the behaviour of PMBs at high temperatures is governed by the polymer phase that enhances the stiffness properties since the bitumen phase behaves as a pure viscous flow material. Moreover, in the polymer-modified mastics, the addition of filler seems to restrain the stiffness contribution due to the polymeric network.

The comparison between the results of plots (b) and (c) in Fig. 1 allows the effect of filler type on the low temperature behaviour of mastics to be evaluated. For unmodified mastics, the increase in stiffness due to both fillers is similar, and the different fillers result in different stiffness properties depending on the type of polymer modification. For SBS polymer-modified mastics, the stiffness ratio trend shows higher values and approaches a maximum value at a higher loading time when basalt filler is used (unlike PO polymer-modified mastics that show two filler types leading to an opposite effect and indicating that limestone filler ensures better performance). Considering that both fillers are characterized by a similar fineness modulus (see Table 3), no stiffening effect from the particle size distribution can be inferred. Hence, the evidence suggests that a different physical–chemical interaction between bitumen and surface filler occurs, and it significantly depends on both filler type and base bitumen, as well as on the type of polymer modification (Frigo et al., 2015; Shivokhin et al., 2012). This experimental finding confirms previous results obtained by

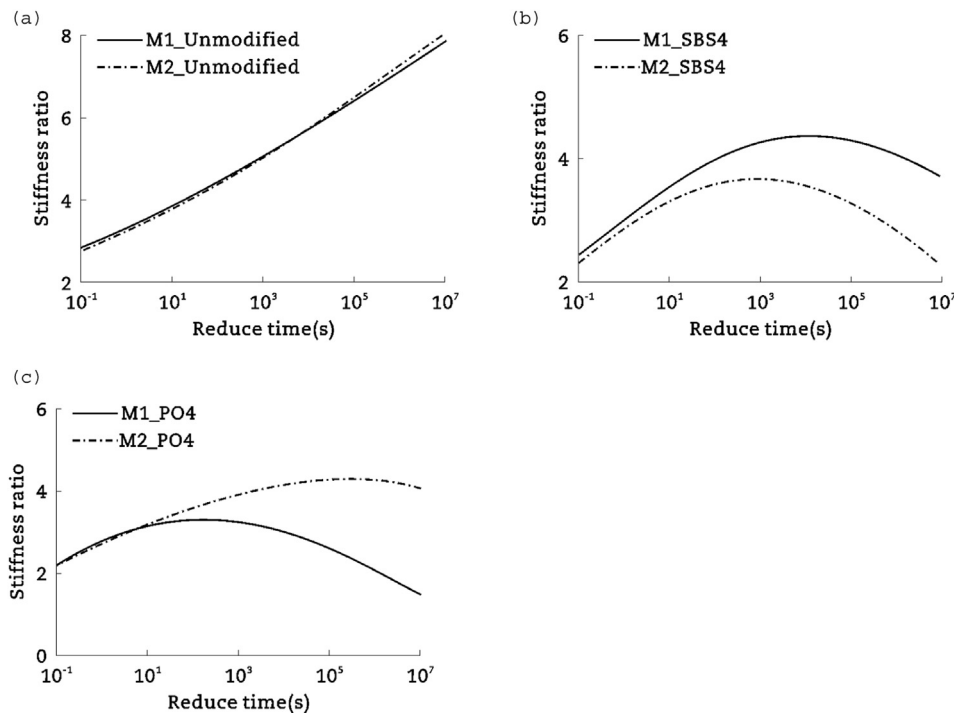


Fig. 1 – Stiffness ratio of mastic to bitumen at a reference temperature of $-12\text{ }^{\circ}\text{C}$. (a) Unmodified mastics. (b) SBS modified mastics. (c) PO modified mastics.

the authors in terms of the fatigue performance of mastics (Frigio et al., 2015). In particular, the filler type affected the fatigue performance of the mastics differently depending on the PMBs considered, suggesting that the rheological response is dependent on the filler/polymer type.

3.2. Rutting resistance

MSCR tests aim to quantify the non-linear performance of materials in order to predict their rutting behaviour. In particular, these tests directly measure the amount of residual strain within a specimen from the stress applied after repeated creep and recovery steps. Hence, the non-recoverable creep compliance (J_{nr}) refers to the ratio between accumulated strain and applied stress and is selected as an indicator of rutting resistance. Moreover, stress dependency is measured by running tests at different stress levels. According to the standard (ASTM, 2010), a stress level of 0.1 kPa in the linear region is applied to investigate the expected behaviour, and a stress level of 3.2 kPa is chosen in the non-linear region. In addition, mastics are also tested at the higher stress level of 10 kPa according to NCHRP (2011).

For each applied stress (τ) and each of the 10 recovery phases (N), the $J_{nr}(\tau, N)$ is calculated by dividing the residual strain post-recovery phase by the stress applied during creep loading. For each applied stress, the average of the non-recoverable creep compliance $J_{nr}(\tau)$ is calculated as the mean of 10 values $J_{nr}(\tau, N)$. Fig. 2 plots $J_{nr}(\tau)$ values measured at 0.1 kPa as a function of temperature for all tested materials.

As expected, J_{nr} increases as the temperature increases for both bitumens and mastics. This is due to more viscous behaviour during the bituminous phase at higher temperatures, which results in higher permanent strain of the material under stress. However, all materials show similar temperature susceptibility, as no significant difference in

strain behaviour (i.e. J_{nr} trend) due to a temperature increase is observed regardless of the filler and/or polymer type.

The experimental data highlight the contribution of the polymer addition within the bitumen. The presence of both polymer types improves the resistance of bitumen to permanent deformations, especially in the case of PO modified bitumen with respect to the neat bitumen.

As far as mastics are concerned, J_{nr} values were found to be significantly lower when compared to the corresponding bitumens over the testing temperature range. This confirms the stiffening effect in reducing the susceptibility of the mastics to permanent deformation. With mastics, polymers improve their resistance to permanent deformations as the J_{nr} of polymer-modified mastics is significantly lower than that of unmodified mastics. However, no clear distinction can be made among the compliance curves of polymer-modified mastics, thus revealing a similar anti-rutting potential.

In order to evaluate the stiffening effect of mineral fillers on both neat and polymer-modified bitumens, the J_{nr} ratio was considered. J_{nr} ratio is defined as the ratio between the creep compliance J_{nr} of the mastic and the respective bitumen at the same stress level. Table 6 summarizes the J_{nr} ratios calculated for all mastics. For each mastic tested, the J_{nr} ratio does not significantly change with the temperature, implying that the stiffening effect of the filler is not temperature dependent. In other words, the presence of filler does not change the temperature sensitivity of bitumen. Even the stress level applied does not seem to affect the J_{nr} ratio of the unmodified mastics, indicating that the filler's stiffening effect is not stress dependent. For polymer-modified mastics, the stress dependency of the materials, in terms of a change in the J_{nr} ratio, is affected by the filler type. Indeed, mastics including the basalt filler (M1) are proved to be less stress dependent than mastics containing limestone (M2), for which a different stiffening effect can be observed as the

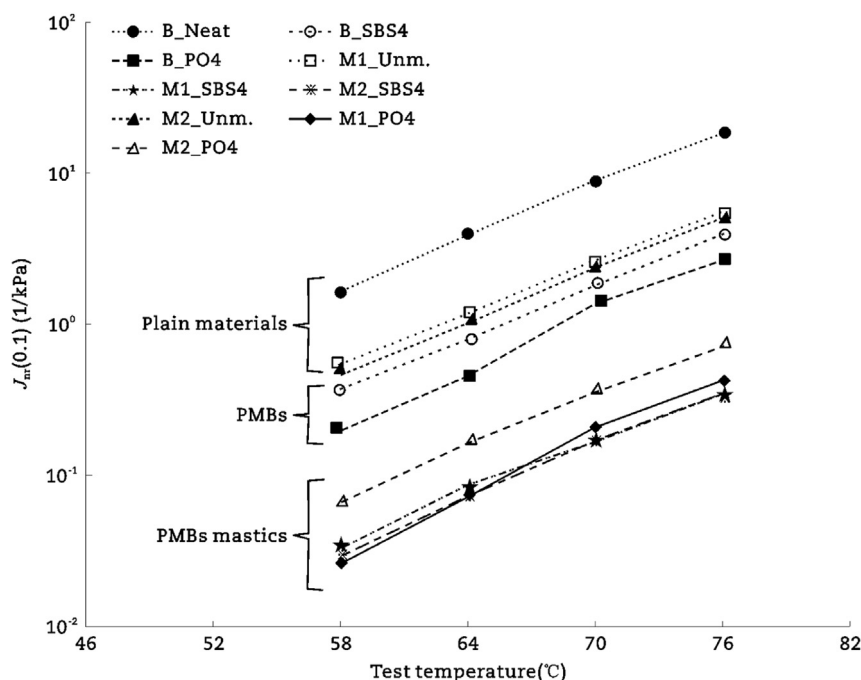


Fig. 2 – Non-recoverable creep compliance at 0.1 kPa for bitumens and mastics.

τ (kPa)	T (°C)	M1_Unmodified	M1_SBS4	M1_PO4	M2_Unmodified	M2_SBS4	M2_PO4
0.1	58	0.33	0.09	0.14	0.30	0.07	0.35
	64	0.30	0.11	0.16	0.27	0.09	0.36
	70	0.30	0.09	0.15	0.28	0.08	0.26
	76	0.30	0.09	0.16	0.30	0.08	0.28
3.2	58	0.30	0.12	0.12	0.27	0.13	0.19
	64	0.31	0.11	0.11	0.28	0.19	0.18
	70	0.31	0.11	0.11	0.28	0.21	0.18
	76	0.31	0.11	0.11	0.28	0.23	0.18

applied stress level changes. In particular, the stiffening effect of M2_SBS4 increased as the stress level increased, whereas it decreased as the stress level increased for M2_PO4. This outcome confirms previous results, suggesting that a physical–chemical interaction occurs between the filler and polymers.

The non-recoverable creep compliance does not take into account the creep phase of testing, as it considers only the residual strain at the end of the recovery phase. For a complete comparison of materials, J_{nr} should be considered in respect to the total creep compliance (J_{TOT}) evaluated immediately before load removal (i.e. at the end of the creep phase) (Santagata et al., 2013). As constant stress is applied, J_{nr}/J_{TOT} coincides with the ratio between the residual strain and accumulated strain at the end of the creep phase. If the material is unable to recover any deformation and the strain

measured at the end of the creep phase remains the same at the end of the recovery phase, the J_{nr}/J_{TOT} will be equal to 1. On the contrary, if the material is totally elastic and able to recover all the accumulated deformation, the J_{nr}/J_{TOT} will be equal to 0.

The results, in terms of J_{nr}/J_{TOT} percentages, are reported in Fig. 3 and Fig. 4 as functions of the temperature and stress level applied, respectively. Both neat bitumen and unmodified mastics show J_{nr}/J_{TOT} values close or equal to 100% at any testing temperature and/or stress state, thereby highlighting their inefficiency to recover deformation and resulting in very low rutting resistance. On the contrary, polymer-modified materials demonstrate an improved resistance to permanent deformation, indicating that the presence of polymers leads to a more elastic response with the ability to recover after a creep phase. This is especially

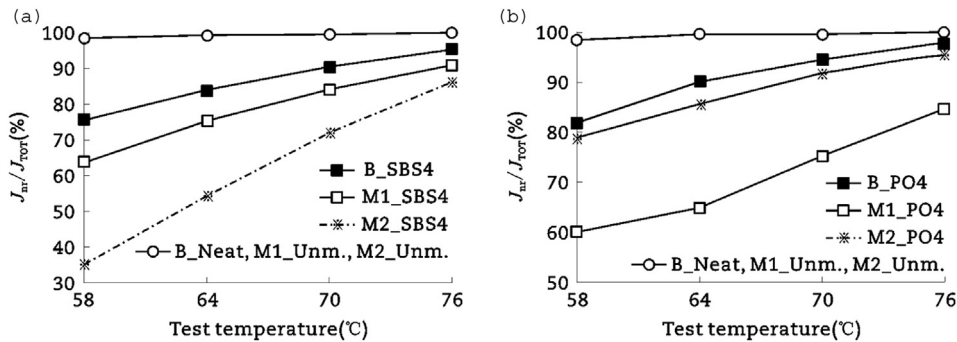


Fig. 3 – Plot of J_{nr}/J_{TOT} values vs. test temperatures at 3.2 kPa. (a) Unmodified and SBS modified materials. (b) Unmodified and PO modified materials.

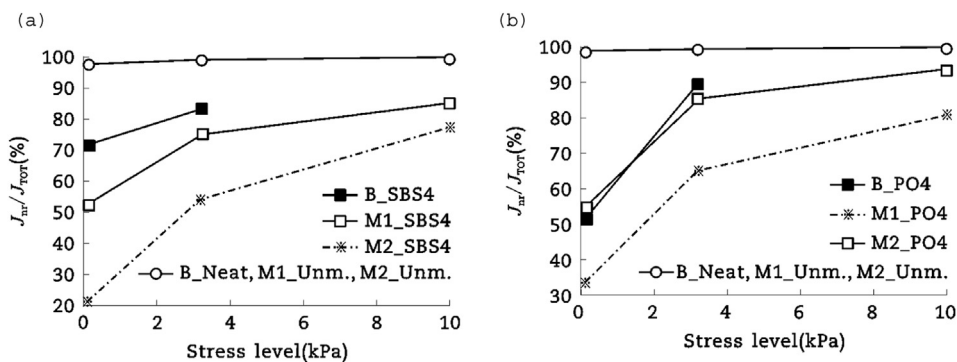


Fig. 4 – Plot of J_{nr}/J_{TOT} values vs. stress levels at 64 °C. (a) Unmodified and SBS modified materials. (b) Unmodified and PO modified materials.

true for SBS modified materials. The capability is both temperature and stress dependent, and tends to decrease as the temperature or stress level increases.

One may observe that the response of both unmodified mastics is similar to that of neat bitumen, revealing that the presence of filler does not improve the elastic response as expected. On the other hand, filler addition significantly improves the anti-rutting potential and shows the more resilient behaviour of polymer-modified mastics for a specific temperature or stress level, as indicated by their lower J_{nr}/J_{TOT} when compared with PMBs. However, the effect of filler type is different depending on the PMBs considered. In particular, the polymer-modified mastic with SBS and limestone filler (M2) shows lower J_{nr}/J_{TOT} values but more marked temperature susceptibility. On the contrary, PO modification appears to have a higher interaction with basalt filler (M1), as observed by the lower J_{nr}/J_{TOT} values of the M1_PO4 mastic compared with the M2_PO4 mastic. In addition, M2_PO4 behaves very similarly to the corresponding bitumen B_PO4, denoting a decrease in the mastic's resiliency response as limestone filler is added. Fig. 4 represents the stress effect on the J_{nr} ratio at 64 °C. The elastic response of polymer-modified mastics shows a similar stress dependency (decrease in J_{nr} ratio with increase in stress), though the effect of a specific stress level is different and highly dependent on the combination of filler/polymer type. In fact, the addition of limestone filler respectively improves and penalizes the elastic response of SBS and PO PMBs, whereas basalt filler produces the opposite result. This evidence highlights the possibility that there is an interaction between the filler and type of polymer-modified bitumen and that its effect on rutting and elastic behaviour is different. These results confirm previous findings regarding the low temperature behaviour of mastics.

According to the conceptual model proposed by Faheem and Bahia (2009), interactions between the filler particle and bitumen take place within the concentrated region, whereas the stiffening effect in the diluted region is due exclusively to the filler concentration. Interestingly, the overall findings confirm the conceptual model hypothesis from a previous study (Frigio et al., 2015) for neat bitumen in terms of rutting and low temperature behaviour, as well as fatigue response. However, some interactions clearly occur at both low and high temperatures, as in case with PMBs, despite the filler concentration being selected within the diluted region. These results highlight the need to develop analytical approaches suitable for polymer-modified bituminous mastics.

4. Conclusions

This study presents a rheological characterization of bitumens and mastics at low temperatures by means of BBR tests and at high service temperatures by means of MSCR tests. 1 neat and 2 polymer-modified bitumens, as well as 2 fillers characterized by a different mineralogy, were selected to produce bituminous mastics.

The following conclusions can be drawn from the BBR results.

- (1) The stiffening effect of fillers is more significant in the neat bitumens as compared with PMBs. Furthermore, filler addition generally causes a decrease in the relaxation capability of the mastics.
- (2) An analysis of the stiffness ratio master curves shows that filler addition results in an increase in stiffness at higher loading times (i.e. higher temperatures) for unmodified mastics. In contrast, filler addition causes the opposite reaction in polymer-modified mastics, thereby suggesting that the filler increases the viscous response at higher temperatures.
- (3) For unmodified mastics, increases in stiffness are similar regardless of filler type, whereas the degree of stiffening is clearly dependent on the filler mineralogy and type of polymer for polymer-modified mastics. This suggests that a physical–chemical interaction between the filler and bitumen occurs and that the interaction level is highly dependent on the type of polymer modification.

Considering the MSCR results, the following conclusions can be drawn.

- (1) The mastics generally show a reduced susceptibility to permanent deformation at any testing temperature and stress level as compared with corresponding bitumens, thereby confirming the stiffening role of the filler.
- (2) Filler addition does not affect temperature susceptibility, which is evaluated as J_{nr} changes with temperature increases, for either unmodified or polymer-modified materials.
- (3) Filler addition enhances the elastic properties of polymer-modified mastics at any temperature and stress level. The improvement in resiliency response was clearly dependent on the combination of filler/polymer type within the mastic, confirming the potential presence of a physical–chemical interaction between the filler and bitumen.

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