Comparative study of the effect of long-term ageing on the behaviour of bitumen and mastics with mineral fillers

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Abstract

This study aims to evaluate the effect of mineral fillers on bitumen ageing. Two different bitumens and four mastics were investigated in the unaged and long-term aged states, based on different properties (consistency, rheology, fatigue resistance and ductility). Mastics stiffened less due to ageing treatment than bitumens, especially with granite filler. However, the results of the performance tests were not definitive regarding the effect of the filler. Aged bitumen showed greater fatigue resistance and higher specific energy of ductile fracture than unaged bitumen, whereas the mastics showed minor variations in the specific energy of ductile fracture with ageing treatment, which is indicative of less ageing, but the fatigue resistance decreased significantly in mastics with one of the bitumens.

Keywords: Bituminous mastic; Filler-bitumen interaction; Ageing; Rheological properties; Fatigue testing; Ductility fracture.

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1. Introduction

The ageing of bitumen is considered a very important phenomenon contributing to asphalt pavement deterioration. Over time, bitumen undergoes several chemical changes that affect its physical, rheological and mechanical properties, and subsequently the pavement performance [1, 2]. Resistance to these changes is commonly referred to as durability and it is dependent on the crude source and refinery process. The most basic change registered in bitumen is hardening, which means the viscosity increases over time. An aged bitumen deforms less upon loading but it is also less able to relax stresses, which is especially important in the low- and intermediate-temperature range, thus weakening the overall performance [3].

The most important cause of bitumen ageing is the reaction with atmospheric oxygen (oxidation) [4]. During the bituminous mixture's fabrication by the (standard) hot method, the high temperature combined with large exposed surface area of bitumen promotes/increases oxidation reactions. Afterwards, oxidation continues throughout the pavement's service life, although at a considerably slower rate due to the moderate temperatures and limited access to oxygen in the pavement. Petersen [1] presents an exhaustive review of the chemical and physicochemical aspects of bitumen oxidation.

However, the bitumen is not isolated in the bituminous mixture, making it possible, therefore, that the chemical reactions occurring in the ageing process are catalysed or "slowed down" due to interactions between bitumen molecules and aggregate particles/filler particles [5–7]. In the 1970s, it was reported that bitumen extracted from field aged bituminous mixtures containing hydrated lime had lower viscosity than the bitumen extracted from mixtures with other mineral fillers [8]. It was then explained that the lower increase of asphaltenes in bitumen with ageing in presence of hydrated lime was because of the strong acid-base reactions established at the surface of hydrated lime particles [9]. However, in addition to the complexity of the bitumen chemistry, the evaluation of the bitumen extracted from bituminous mixture can be also misleading due to contamination (solvents and/or filler) [10].

The importance of the bitumen-aggregate interaction has long been acknowledged, especially with the finest particles (filler), for the behaviour of bituminous mixture [11–13]. Therefore, more recently, some researchers investigated the filler effect on bitumen ageing from the rheological properties of bituminous mastics. Based on the characterization of the shear complex modulus and phase angle with the Dynamic Shear Rheometer (DSR), in [14–16] it was concluded that, regardless of the filler type, bituminous mastics harden less than bitumens under the same ageing conditions. Xie et al. [17] also reported this for ageing induced by ultraviolet radiation. On the contrary, Moraes [18] determined the glass transition temperature of bitumens and mastics with a dilatometric system and found that though the filler incorporation improved the low temperature behaviour, the evolution trend with increasing ageing was similar in bitumens and mastics. It was noted in the literature review that a wide range of fillers and concentrations, with variable stiffening effects on bitumen, were tested, which complicates a comprehensive discussion of research studies. Furthermore, most of these studies did not investigate the effect of ageing on different performance-related properties of bituminous mastics.

2. Objectives and scope

The main objective of this research was to evaluate the effect of traditional mineral fillers on bitumen ageing. To this end, the properties of different bitumens and bituminous mastics were compared in unaged and aged states. The materials were aged in the laboratory with the Pressure Ageing Vessel to simulate the long-term in-field oxidative ageing state. Two 50/70 paving grade bitumens with different ageing susceptibility and two mineral fillers (limestone and granite) were used in the study. The bitumens and bituminous mastics were characterized in terms of their conventional properties (consistency), rheological behaviour in the intermediate and high temperature range, ductility and fatigue resistance. Hence, this provides a wide-ranging description of the effect of ageing on the performance of mastics with different mineral fillers.

3. Materials and experiments

3.1 Materials

Two bituminous binders and two fillers were used in this study. The bitumens (B1 and B2) are of the same paving grade (50/70) but have different chemical compositions (SARA fractions listed in Table 1) and structure, and rheological behaviour. Thus, these binders were selected because the physical properties and chemical composition of filler and bitumen are relevant to the bitumen's ageing [5–7].

The SARA fractions were determined before [19], using a 2-step method. First, the n-heptane insoluble bitumen fraction was separated following ASTM D6560 [20] to quantify the asphaltenes. Then, the n-heptane soluble compounds (maltenes: saturates, aromatics and resins), after evaporation of the solvent, were quantified with Thin Layer Chromatography - Flame Ionization Detection (TLC-FID).

The content of asphaltenes, which are known for their role in bitumen viscosity and ageing sensitivity [21], is higher in B1. Furthermore, bitumen B1 has less aromatics than B2 but the content of saturates compensates, at least partially, the content of the less polar fractions. The proportion of resins is similar in both bitumens. However, previous research showed that besides chemical composition, based on the relative proportion of these four fractions, the compatibility and interactions between functional groups are of upmost importance for the bitumen's physical properties [1]. For example, Redelius and Soenen [22] reported that the effect of asphaltene content on bitumen viscosity is not clear when this factor is considered in isolation, whereas when the bitumen viscosity is normalized to the viscosity of maltene, there is a good correlation.

The fillers (limestone L and granite G) are both natural and were obtained from grinding natural rock to dust. Filler L is a commercial filler obtained from crushed limestone, whereas filler G was collected in the dust collection system of an asphalt plant that works with granite aggregates. The main filler properties are listed in Table 2. The filler gradation is similar but the Rigden voids (test method defined in [23]) are significantly different. It has been demonstrated before [24–27] that the filler stiffening effect is strongly related to the voids in compacted filler (Rigden voids). Rigden [11] reported that with the continuous increase of the filler concentration the mastic behaviour changed from a viscous system to a plastic system at the concentration determined by the voids in compacted filler. It was latter theorized that in a filler-bitumen mixture the Rigden voids represent the volume of bitumen used in filling the voids within the mineral structure, and it is the extra bitumen that looses the mineral structure and gives flowability to the mixture. Therefore, the larger the Rigden voids are the more bitumen is required to attain the same flowability of the mixture. This is further supported by the results of the absorbing test [28] on these bitumens (listed in Table 2). In this test, similar to the experiments of Rigden, filler is continuously added to a bitumen sample (15 g) up to the point that the mastic becomes crumbly, and the test result corresponds to the total filler mass added (normalized to a particle density of 2.65 Mg/m^3). The amount of filler G that can be mixed in the bitumen was about 35% lower than when filler L is used instead. In addition, the bitumen did not affect the absorbing test results despite the differences in chemical composition.

Table 1 – Bitumen properties

Bitumen		**Penetration**	**Softening Point**	**Chemical composition** [%]				**Density**
Code	Grade	[0.1 mm]	[ºC]	Saturates	Aromatics	Resins	Asphaltenes	[kg/m^3]
B1	50/70	54	50.4	7.4	48.7	29.4	14.5	1025.9
B2	50/70	49	48.2	3.1	58.5	28.8	9.6	1039.4

Note - SARA fractions: Saturates, Aromatics, Resins and Asphaltenes

Table 2 – Filler properties

Filler		**Particle density**	**Gradation (% passing)**			**Rigden voids**	**Absorbing capacity** [g]	
Code	Origin	[Mg/m^3]	#2.0 mm	#0.125 mm	#0.063 mm	[%]	B1	B2
L	Limestone	2.71	100.0	99.0	83.5	26	55	56
G	Granite	2.57	100.0	93.1	81.5	42	35	37

3.2 Laboratory experiments

3.2.1 Fabrication of mastics

The bituminous mastics were fabricated with a filler volumetric concentration of 30% using the two fillers and the two bitumens. The filler concentration in dense bituminous mixtures varies roughly between 10% and 50% [29], therefore the mid-point of this interval was adopted for the experiments. Volumetric proportion, instead of mass proportion, was chosen because the fillers had different particle densities.

The fillers were dried in an oven at 150 ºC for several hours and then cooled to room temperature in a drying desiccator for 1h. Then, the fillers were stored in the desiccator. To fabricate each mastic, the bitumen and filler were preheated at 150 ºC for 60-90 min, and then the filler was manually added at constant rate for 20 min during mechanical agitation at 400 rpm. To ensure good filler distribution in the bitumen the agitation was continued for an additional 10 minutes. Finally, the mastic container was cooled to room temperature.

3.2.2 Ageing of bitumens and mastics

The bitumen and mastic samples were aged with the Pressure Ageing Vessel (PAV) (Model 9300, Prentex) for 25 h under the conditions defined in EN 14769 [30]. This conditioning procedure was proposed to simulate the long-term (several years) in-field oxidative ageing in the laboratory for 20 h by imposing high temperature (100 ºC) combined with air pressure (2.1 MPa). The equivalent in terms of the number of years simulated by PAV ageing depends on the in-situ environmental conditions and bituminous mixture properties [31]. The protocol [30] defines adding 50 g of bitumen to each test plate. To ensure similar ageing processes were induced in bitumen and mastic samples, the mass of mastic added to the plates was determined to have similar thickness to the bitumen samples (approximately 3.2 mm). This means 71 g to 74 g depending on the filler's particle density.

The duration of ageing (25 h) was chosen to simulate the short- and long-term ageing effect on binders and mastics based on the work of Migliori and Corté [32]. They concluded from the study of 4 unmodified bitumens that the differences in consistency, complex modulus and asphaltenes content evolution were not significant between ageing for 25h in PAV and ageing with the Rolling Thin Film Oven Test (RTFOT), followed by 20h in PAV. The most commonly used ageing procedure is to simulate first the short-term ageing with the RTFOT, and then the long-term ageing with the PAV for 20 h. However, there is a concern that RTFOT ageing will be different in bitumen and mastic because of their differences in viscosity [9]. In the RTFOT [33] it is supposed that a thin moving film of bitumen is exposed to the hot air to simulate the asphalt plant conditions, but in the case of high viscosity bitumens (hard paving grades and modified binders) and of mastics these conditions will not be created. Hence, homogeneous ageing of the bulk material does not occur. In addition, the same procedure should be used with the bitumen and the mastics.

3.2.3 Consistency at intermediate and high temperatures

The consistency of bitumen and mastics at intermediate and high temperatures was evaluated, respectively, from the needle penetration (Pen) at 25 ºC, as described in the EN 1426 standard [34], and the softening point by the ring and ball method ($T_{R\&B}$) described in the EN 1427 standard [35]. Current bitumen specifications used in Europe [36,37] consider these two properties for the evaluation of bitumen ageing resistance, setting limits for the retained penetration value and the increase in the softening point.

3.2.4 Viscoelastic rheological characterization

The viscoelastic rheological behaviour in the linear region of the bituminous materials was performed with a rotational rheometer (Gemini 2000, Bohlin Instruments), by imposing oscillatory shear loading onto a disk-shaped specimen in varied conditions of frequency and temperature. Testing was performed following the protocol of the European standard EN 14770 [38]. Two test procedures were used, strain sweep and frequency sweep. First, strain sweep was imposed at a specific frequency (0.1 rad/s, 1 rad/s and 100 rad/s) and temperature (20 ºC, 40 ºC, 50 ºC and 60 ºC) to determine the linear viscoelastic region. Then, the frequency sweep (0.1 rad/s to 100 rad/s) was performed to characterize the viscoelastic behaviour. The shear stress level imposed at each temperature was chosen from previous strain sweep test results. The sizes (diameter/height) of the specimens were 8 mm / 2 mm at 20 ºC and 40 ºC, and 25 mm / 1 mm at 40ºC, 50 ºC and 60 ºC. The rheological behaviour of materials was evaluated from the shear modulus (absolute G*, storage G' and loss G'') and phase angle (δ).

3.2.5 Multiple Stress Creep and Recovery test

The multiple stress creep and recovery (MSCR) tests were performed to characterize the elastic response and stress dependence of bituminous materials. The test evaluates the sensitivity to permanent deformation of bituminous binders, which is expected to decrease with ageing. The test was performed following the protocol defined in the European standard EN 16659 [39], with the same rotational rheometer used for the viscoelastic rheological characterization. This means that 20 consecutive cycles of shear loading (1 s) and rest (9 s) were imposed on a 25 mm / 1 mm (diameter/height) specimen. During the first 10 cycles the shear stress was 0.1 kPa, and afterwards 3.2 kPa. The test was performed at 60 ºC. The evolution in stress and strain was analysed per load level. The main test variables used in the analysis were the average percentage recovery ($\%R_L$) at load level L,

$$\%R_L = \frac{1}{10} \cdot \sum_{N=1}^{10} 100 \cdot (\varepsilon_1^N - \varepsilon_{10}^N)/\varepsilon_1^N \tag{1}$$

and the average non-recoverable creep compliance ($J_{nr\,L}$) at load level L,

$$J_{nr\,L} = \frac{1}{10} \cdot \sum_{N=1}^{10} 100 \cdot \varepsilon_{10}^{N}/L \tag{2}$$

where, ε_{1}^{N} and ε_{10}^{N} are the strain at the end of creep (1 s) and rest (10 s) in cycle N.

3.2.6 Fatigue resistance - Linear Amplitude Sweep test

The Linear Amplitude Sweep (LAS) test is a recent method, using a rotational rheometer, developed to evaluate the fatigue resistance of bituminous materials [40]. Different from the traditional time sweep test (TSS), the load/strain oscillatory amplitude is increased during the test course to induce the specimen's failure in a short time (approximately 30 min). In the TSS protocol, testing a single specimen may take several hours, and testing is required at several load amplitudes to obtain the failure law. The LAS test has been implemented before with binders and mastics [41–44].

The LAS test results were analysed based on the concepts of the Viscoelastic Continuum Damage (VECD) mechanics. More information about the theoretical concepts of VECD can be found in [40,44]. The analysis procedure is briefly described as follows. The progress of damage in material is described as:

$$\frac{dD}{dt} = \left(-\frac{\partial W^{R}}{\partial D}\right)^{\alpha} \tag{3}$$

where, D is the damage intensity; t is the time; α is a constant; and W^R is the pseudo strain energy density

$$W^{R} = \frac{1}{2} \cdot C \cdot (\gamma_i^{R})^2 \tag{4}$$

where, γ_i^R is the pseudo-strain amplitude in cycle i

$$\gamma^{R} = G^{*} \cdot \gamma_i \cdot sin(w \cdot t + \delta) \tag{5}$$

and C is the material integrity parameter

$$C = \frac{\tau_i}{\gamma_i^{R}} \tag{6}$$

where, G^* and δ are the complex modulus and the phase angle of the undamaged material, respectively; σ_i and γ_i are the stress and strain amplitude in cycle *i; w* is the angular frequency.

The constant α is determined from the maximum slope (*m*) of the *log (G'(w))* versus *log (w)* as

$$\alpha = 1 + \frac{1}{m} \tag{7}$$

The damage intensity at time *t* (*D(t)*) is obtained by solving Eq. (3) numerically as:

$$D(t) \cong \sum_{i=1}^{N} \left[\frac{1}{2} \cdot (\gamma_i^{R})^2 \cdot (C_{i-1} - C_i)\right]^{\frac{\alpha}{1+\alpha}} (t_i - t_{i-1})^{\frac{1}{1+\alpha}} \tag{8}$$

Then, the progress of the integrity versus damage intensity is modelled with:

$$C(t) = 1 - C_1[D(t)]^{C_2} \tag{9}$$

where, C_1 and C_2 are the model-fitting coefficients. Then, Eq. (7) is introduced in Eq. (2) and differentiated in order to *D*. The obtained equation is introduced in Eq. (3), and the differential equation solved. Finally, *t* is substituted by *N/f* and the equation rearranged as:

$$N = \frac{f \cdot D_f{}^K}{K \cdot \left(\frac{1}{2} \cdot C_1 \cdot C_2\right)^{\alpha}} \cdot G^{*-2\alpha} \cdot \gamma^{-2\alpha} \tag{10}$$

$$K = 1 + \alpha \cdot (1 - C_2) \tag{11}$$

where, D_f is the damage intensity at failure, which was defined as the peak in the shear stress as proposed by Johnson [40].

In this study, the test procedure of a single specimen is initiated with a frequency sweep (0.2 Hz to 30 Hz) at a small shear strain amplitude (0.1%) to determine slope *m*. Then, the strain amplitude was increased linearly from 0.1% to 30% in 3000 cycles. The oscillatory frequency was 10 Hz during the second part of the test. The test temperature was 25 ºC with all binders and mastics.

3.2.7 Ductility - Double Edge Notched Tension Test

The Double Edge Notched Tension (DENT) test evaluates the resistance to ductile failure of bituminous materials. The bituminous specimen is tensioned in a ductilimeter at a constant elongation rate until fracture or the maximum defined elongation is reached. To ensure the material has a ductile behaviour, the test temperature is usually set between 0 ºC and 25 ºC depending on the bitumen hardness. The test name originates from the specific shape of the specimen that is moulded with a notch on both sides of the central section (see Figure 1) to reduce its extensional flow ability. According to the essential work of the fracture approach [45], in a tension test the total energy expended in ductile failure is divided into two components that differ in the zone where energy is expended. The essential energy is expended in the fracture zone, whereas the non-essential part is expended away from the failure zone. Ductility, plasticity and tearing deformation of the material away from the fracture zone are considered non-essential energy, and it should not be considered for the evaluation of the material’s fracture resistance. In general, the larger the cross-section of the specimen, the higher the proportion of non-essential energy in the total deformation work. Hence, different size specimens are tested to estimate the material’s specific essential fracture resistance.

The DENT test was performed following the testing protocol and the results analysis procedure defined in the LS-299 specification [46]. Specimens with three different ligament lengths (5 mm, 10 mm and 15 mm), conditioned in a water bath at 20 ºC, were tensioned at 50 mm/min until failure. The test temperature was selected so that all materials, including aged mastics, would behave in a ductile manner and the tension load would not exceed the load cell limit of the ductilimeter. The tension load (P) and displacement (d) were registered during the test.

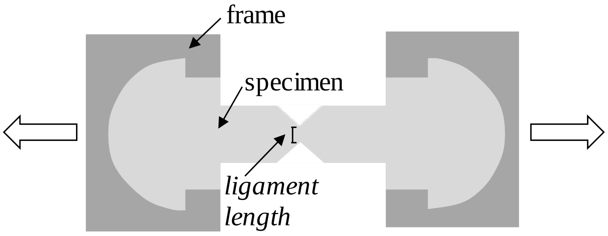

Figure 1 – DENT test specimen

For a test specimen with ligament length *l*, the total work of fracture (W_t) expended in the specimen's failure corresponds to the area under the *P-d* curve, which was numerically determined using the trapezoidal integration rule. In addition, the specific work of fracture (w_t), i.e. deformation work per unit surface area, is:

$$w_t = \frac{W_t}{B \cdot l} \tag{12}$$

where *B* is the specimen thickness (m); and *l* is the ligament length (m).

Based on the method of the essential work of fracture [45], it is assumed that w_t varies with the ligament length, as:

$$w_t = w_e + \beta \cdot w_p \cdot l \tag{13}$$

where w_e is the specific essential work of fracture (J/m^2); w_p is the specific plastic work of facture (J/m^2); β is a constant related to the specimen's shape and size. Hence, w_e is the mechanical property that expresses the material's resistance to ductile failure, which does not depend on the fracture's zone shape and size. This linear model is therefore fitted to the w_t-l results, and w_e corresponds to the intercept of the model.

In addition, to evaluate the bitumen's ductile failure resistance, LS-299 [46] also proposes using the critical Crack Tip Opening Displacement (*CTOD*), which is determined as:

$$CTOD = \frac{w_e}{\frac{P_{5mm}^{peak}}{B \cdot l}} \tag{14}$$

where, P_{5mm}^{peak} is the peak load for the specimen with the (shortest) ligament length of 5 mm. It is suggested that this indicates the fracture toughness to a pre-existing crack, and previous research [47] reported that the *CTOD* value correlates well with the fatigue resistance. The higher the *CTOD* value, the greater the fatigue resistance is.

4. Results and discussion

The effect of PAV ageing (25 h) on the behaviour of the bitumens (B1 and B2) and bituminous mastics (B1-L, B1-G, B2-L and B2-G), was evaluated by means of the results of different test methods. The experimental results are presented and discussed in the following sections.

4.1 Consistency at intermediate and high temperatures

The consistency of bituminous materials at intermediate and high temperatures were evaluated from the needle penetration and softening point values. Figure 2 shows the average tests values, and the variation with ageing (retained penetration and increase of softening point). Current European specifications for paving grade bitumens [36] set only hardening limits for the bitumens after short-term ageing, which for a bitumen class 50/70 is a minimum of 50% of retained penetration and a maximum of 9 ºC/11 ºC of increase in softening point, depending on the severity level chosen. Currently, long-term ageing limits have not yet been defined in Europe. The materials had, as expected, lower penetration and higher softening points after ageing and/or filler incorporation. Ageing had an expressively greater effect on bitumen B1. The retained penetration and increase of the softening point values of B1 were 46% and 15 ºC, respectively, whereas for B2 the results were 57% and 8 ºC. These results show clearly the difference in ageing resistance of the two bitumens. Even after long-term ageing treatment, bitumen B2 complies with the European specification limits [36] (50% and 11 ºC) defined for short-term ageing. As described by Petersen [1], oxidation ageing varies with the chemical composition of bitumen. The relative reactivity of the chemical groups with oxygen was determined as 1:7:32:40 for saturates, aromatics, resins and asphaltenes, respectively. B1 had a higher percentage of asphaltenes (see Table 1). Other research pointed out the importance of the aromatics as precursors of resins or asphaltenes [48,49], which were in higher quantity in B2. However, the durability and performance of bitumen on oxidative mechanisms depends not only on the changes in the chemical composition but also on the balance between chemical groups [1].

In the unaged state, the mastics had a significantly higher consistency than the bitumens. The penetration reduced by approximately 45% and the softening point increased by 8-13 ºC. Filler G had a greater effect than filler L on the softening point, which as referred before is related to the higher Rigden voids of G. These values fall in the range of values reported in the literature [24,27,50], and using the well-known PRADO model developed in Belgium [27] it would be predicted 7 ºC and 11 ºC for fillers L and G, respectively. In addition, the effect of the bitumen was not meaningful (1 ºC variation) which is in line with the results of the absorbing test reported before in section 3.1.

After 25 h in the PAV, mastics aged less than bitumens. The reduction in ageing was roughly 10% in retained penetration and 2 ºC in softening point. The effect of the bitumen is clear, but this is not true of the filler type. Mastics with bitumen B1 aged more than mastics with B2, in line with the trend observed for the base bitumens. Contrarily, the results obtained for mastics with the same bitumen and different fillers were similar.

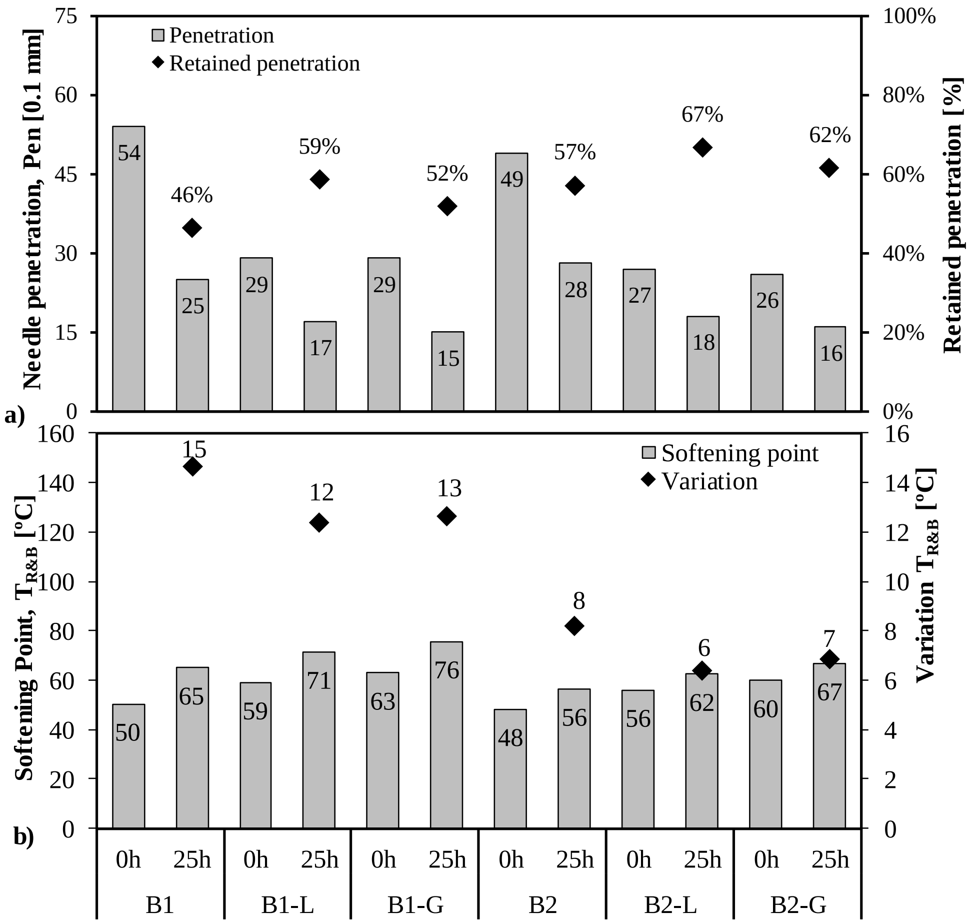

Figure 2 – Needle penetration (a) and softening point (b) test results.

4.2 Rheological behaviour

The viscoelastic behaviour of binders and mastics is illustrated with the Black curves shown in Figure 3. The two bitumens were of the same paving grade but had significantly different rheological behaviour, which is related to the differences in chemical composition (see Table 1) and structure. Thus, previous investigation to these bitumens with atomic force microscopy showed that bitumen B1 had larger "bee-structures" (catana phase) [51]. The rheological behaviour is traditionally related to the colloidal model of bitumen, which is determined by the chemical composition and interactions established between functional groups [52], and the colloidal stability decreases with ageing [53]. The ratio of the aromatics to asphaltenes content decreases with ageing which implies a growing gel character of the bitumen. Thus, it is observed that the time-temperature superposition principle (TTSP) is not fully applicable to aged bitumen B1, which is related to a more complex structure. TTSP is also not fully applicable to mastics of bitumen B1.

In addition, it can be seen that filler and ageing affected the black curves of bitumen differently. The incorporation of filler in bitumen stiffens the mixture and the Black curve moves to the right and slightly downwards. On the other hand, the bituminous materials are stiffened due to ageing, but the Black curves move mostly downwards. This occurs because the phase angle is reduced significantly with ageing, which is in agreement with previous research [14].

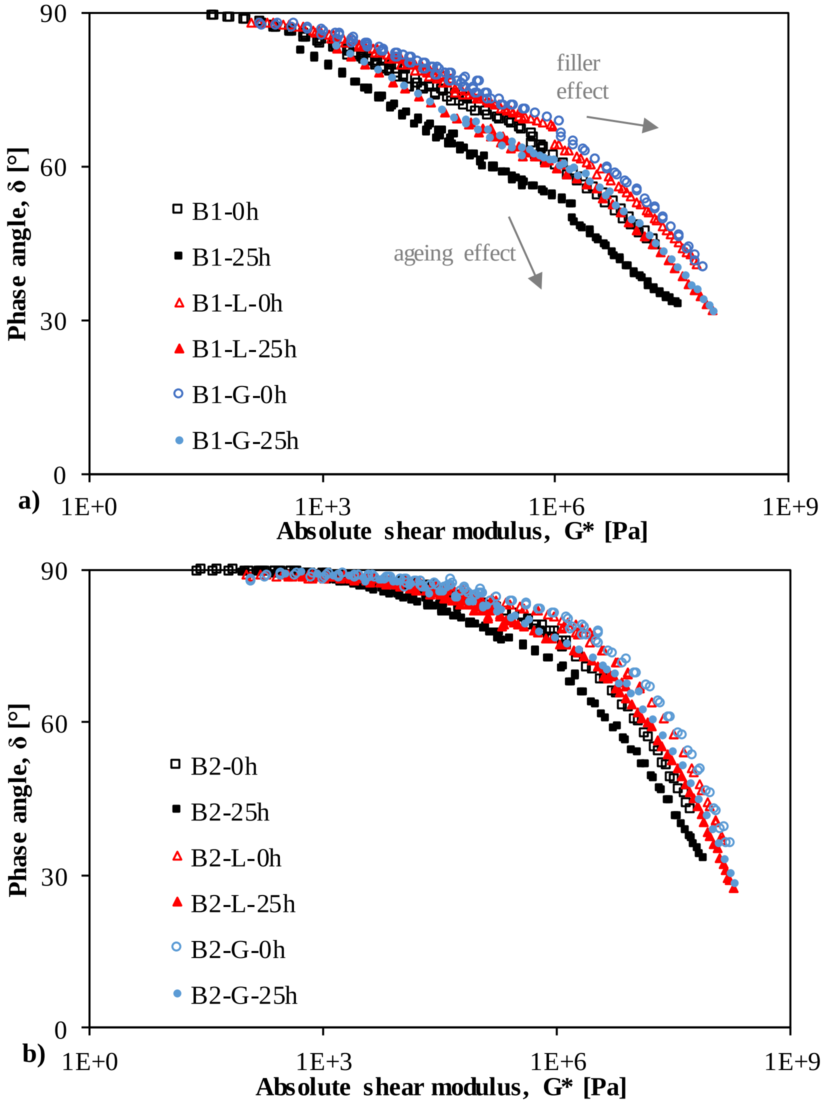

Figure 3 – Black curves (δ-G*): a) bitumen B1; b) bitumen B2.

To better understand these effects, the storage (G') and loss (G'') moduli (at 20 ºC and 60 ºC) versus the frequency (0.01-100 rad/s), are plotted in Figure 4. In simple terms, the G' and G'' represent the elastic and viscous components of complex shear modulus, respectively. As for the results of unaged bitumen, both modulus components increased with filler incorporation and ageing over the complete frequency range. The ageing effect was greater at the lower frequencies, whereas the filler effect did not vary with the load frequency. This means that the behaviour of the aged materials is less dependent on load frequency. Thus, this results in the shift and flattening of the complex shear modulus master curve after PAV ageing was reported before [31]. Furthermore, this rheological change was also observed for aged mastics, irrespective of the filler type. Moraes [18] found a similar trend for the complex shear modulus with various bitumens and fillers, including hydrated lime, which many researchers [54] point out has a special ability to mitigate ageing in bitumen.

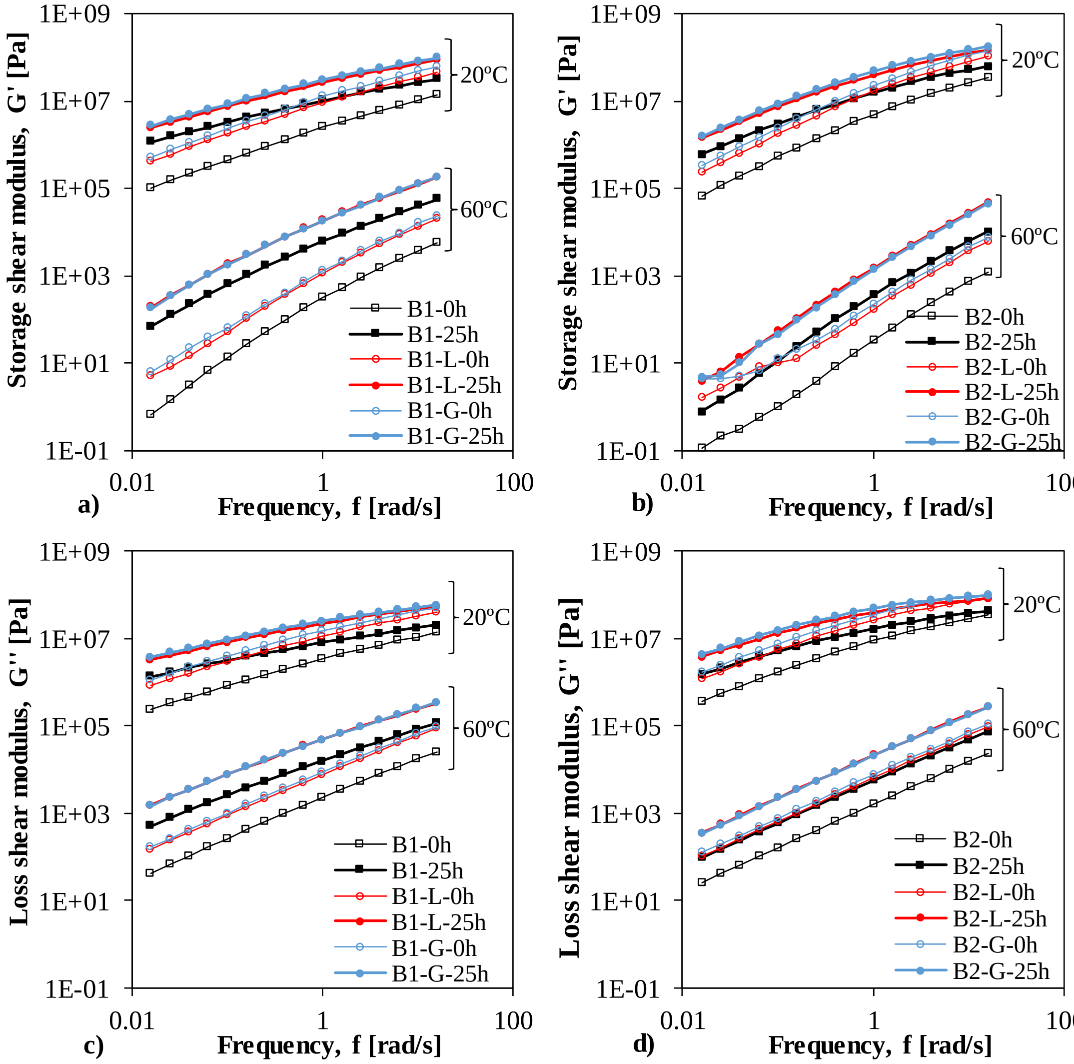

Figure 4 – Storage (G’) and loss (G’’) moduli versus frequency at 20ºC and 60ºC: a) G’, bitumen B1; b) G’, bitumen B2; c) G’’, bitumen B1; d) G’’, bitumen B2.

In addition, the variations in the storage modulus were greater than in the loss modulus. This means that mastics and bitumens became more elastic after ageing. The increase of the elastic component of deformation under load was especially significant for the high temperature conditions. This is explained by the fact that for the range of measured phase angle values (> 50º) a decrease of the phase angle results in an increase of cos (δ) and a decrease of sin (δ). As commented before in the analysis of Figure 3, ageing leads to a significant decrease of the phase angle, whereas the impact of the filler was minimal. Therefore, despite the decrease of δ with ageing, the G’’ modulus (G’’=G*·sin(δ)) increased because of the significant increase in G* value.

Finally, the effect of ageing on the moduli (G*, G´and G´´) is summarized in Figure 5. The figure shows the ageing index, determined as G_{25h}/G_{0h}, for two different test conditions (20 ºC / 10 Hz and 60 ºC / 1.6 Hz). The first test condition is representative of normal in-service circumstances (intermediate temperature and average traffic speed) and the other of the critical combination of high temperature and slow traffic. The results presented show a clear reduction of the ageing index, regardless of the shear modulus considered, with the incorporation of mineral filler in bitumen. The stiffness increase due to ageing was more notorious in bitumens than in mastics, and granite contributed more to ageing reduction than limestone. This trend is better perceived from the results at 60 ºC / 10 Hz, which agree with previous research [18]. Furthermore, the storage modulus is more sensitive to ageing than the absolute and loss moduli. G' index values were higher than G* and G'' index values and showed a larger variation among materials. This rheological change results in a reduction with ageing of the proportion of the delayed-elastic and viscous deformation of the total deformation.

In addition, the ageing indexes were significantly higher for B1 because, as reported by Weigel and Stephan [21], the phase angle reduction with ageing is proportional to the asphaltenes content of unaged bitumen (see Table 1).

To investigate the effects of ageing on deformation recovery properties of bitumen and mastics, MSCR tests were undertaken. The results of the tests at 60 ºC are shown in Figure 6. The vertical bars represent the average values of percentage recovery and non-recoverable compliance. The points represent the coefficient of variation between the unaged and aged states. The coefficient of variation is equivalent to an ageing index, though it is noted that for the recovery variable the determined value reduces with ageing. As defined in the MSCR test protocol, the loading scheme was repeated with each specimen at two stress levels, 0.1 kPa and 3.2 kPa, without interruption between the two load levels.

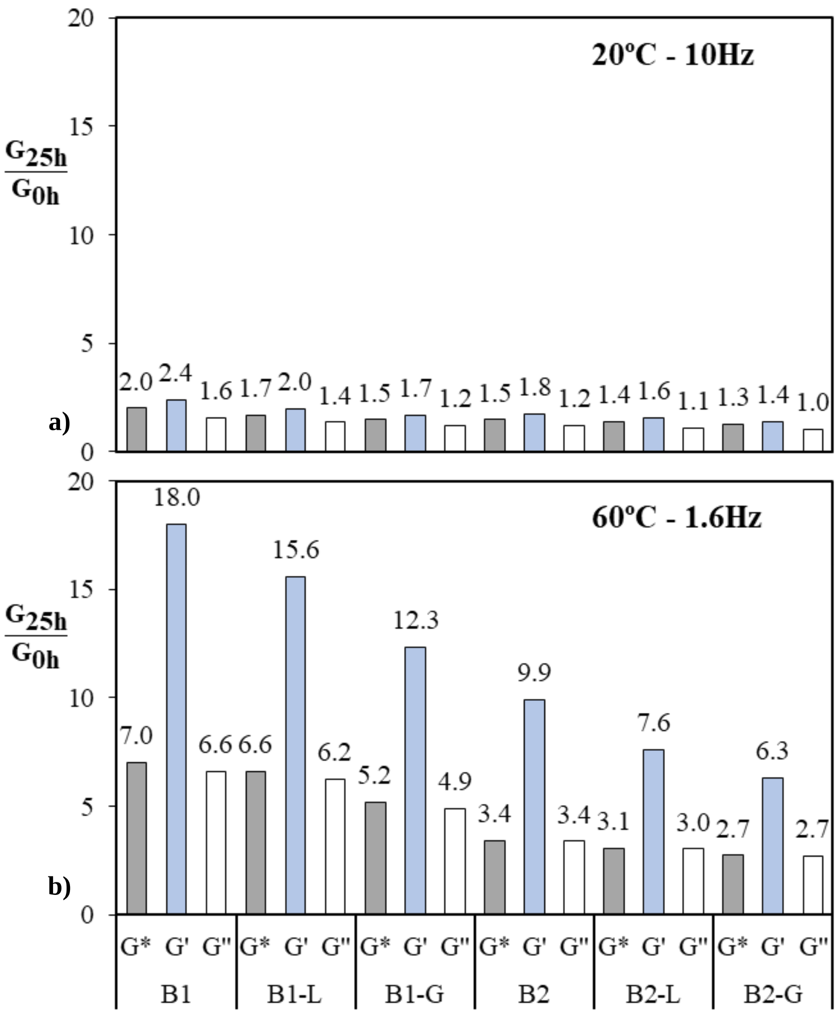

Figure 5 – Shear moduli (G*, G’ and G’’) increase with ageing: a) 20ºC and 10Hz; b) 60ºC and 1.6 Hz.

As expected, aged materials had greater elastic recovery compared to the unaged materials. This means that aged materials were more resistant to permanent deformation. However, the deformation recovery decreased with the increase in applied shear stress, though this trend was not seen for the non-recoverable compliance. However, both test quantities were affected by filler incorporation in bitumen. The elastic recovery increased, and the non-recoverable compliance decreased. In addition, bituminous mastics were less sensitive to ageing. The coefficient of variation of elastic recovery before and after ageing was reduced tremendously by the effect of mineral filler, especially with granite filler. This trend was confirmed with both stress levels. Contrary to this, the coefficient of variation of compliance decreased only slightly in mastics. Finally, the MSCR test results showed a better ageing resistance of bitumen B2 and mastics with this bitumen, which agrees with the results of linear viscoelastic rheological characterization.

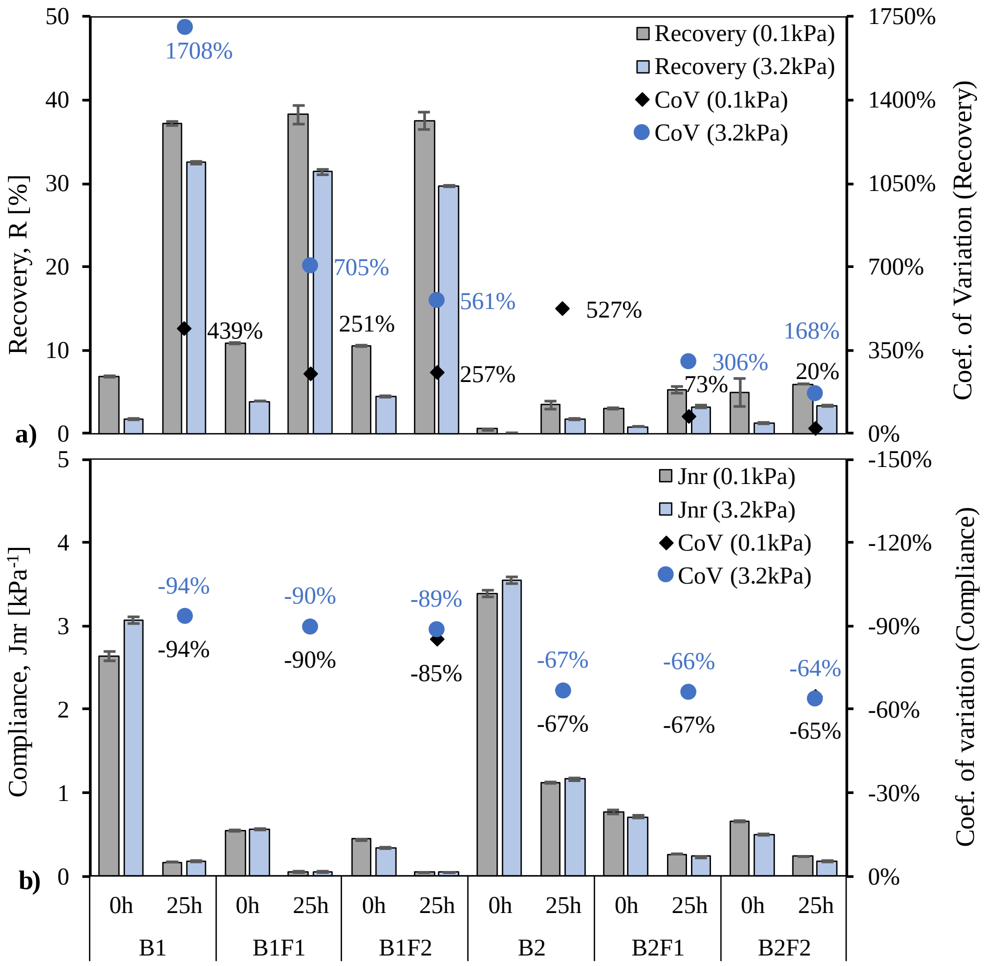

Figure 6 – MSCRT test results: a) recovery; b) non-recoverable creep compliance.

4.3 Fatigue resistance

The LAS test protocol was adopted for the evaluation of fatigue resistance of studied bituminous materials. The test results were analysed following the VECD methodology described in Section 3. Figure 7 presents the LAS test results.

The evolution of shear stress with increasing applied strain in specimens is plotted in Figure 7 a) and b) for bitumen B1 and bitumen B2, respectively. In general, the stress increased with applied strain up to the point where it became nearly stable, and afterwards the stress decreased sharply. The transition to this latter phase has been related with sample failure [40]. The bitumen type affected the shape of the τ-γ curve, the size of the region of nearly constant stress being the most important. In general, mastics and aged materials attained higher stress levels at the peak due to the increased stiffness but the strain at the stress peak decreased. These results are in agreement with previous research [42]. However, bitumen B1 and related mastics showed a strong ability to hold large deformations before extensive internal cracking occurred. In addition, for these materials, despite the stiffening due to ageing, there was not a substantial decrease in the strain value at peak stress, and in the case of the bitumen the plateau region was even extended. As for the filler type effect, mastics with granite filler attained higher values of maximum shear stress in the lower strain values.

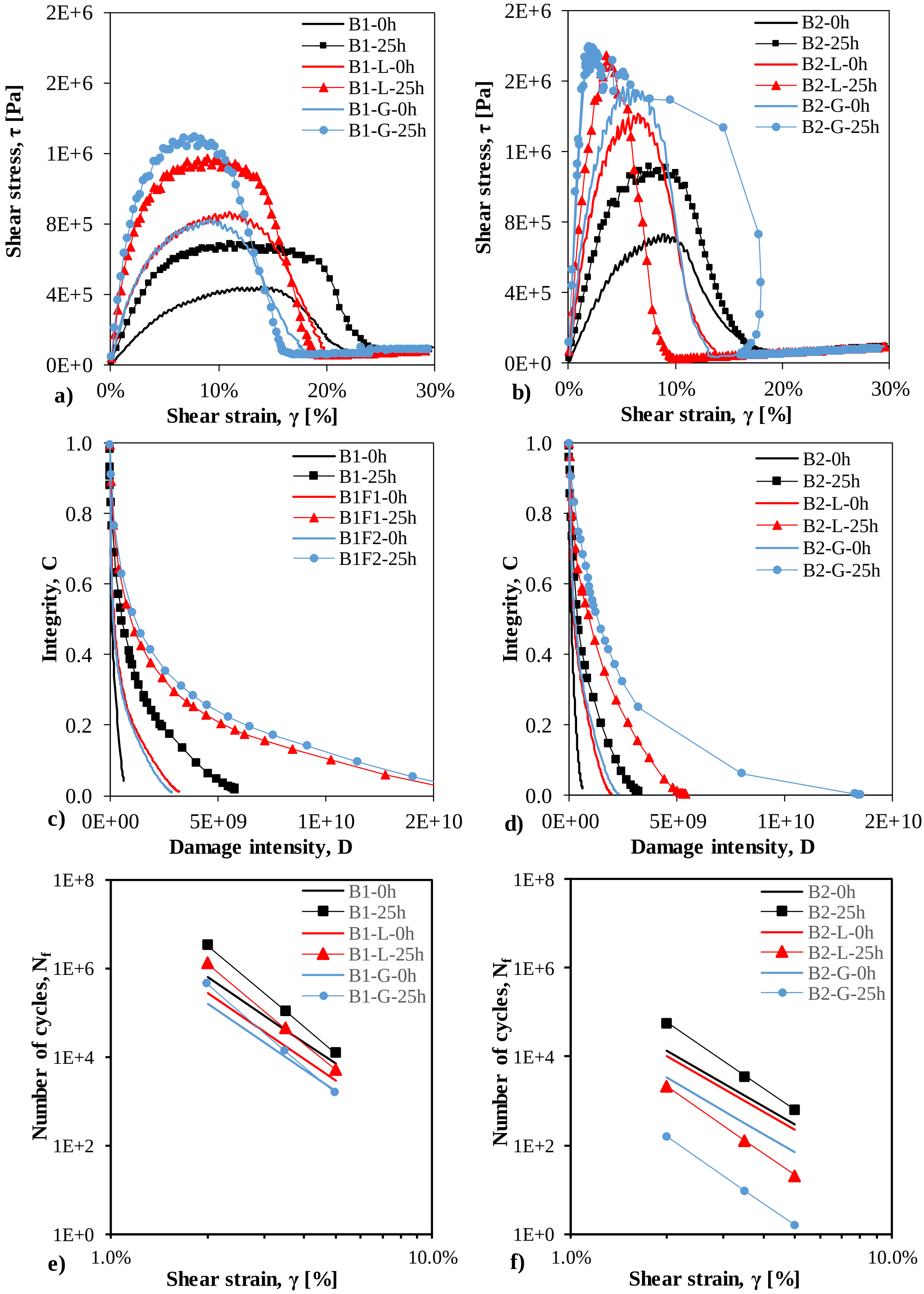

Figure 7 – LAS test results: a) τ-γ, bitumen B1; b) τ-γ, bitumen B2; c) C-D, bitumen B1; d) C-D, bitumen B2; e) N_f-γ, bitumen B1; f) N_f,-γ bitumen B2.

Figure 7 c) and d) show the C-D curves of materials for bitumen B1 and bitumen B2, respectively. According to the VECD approach, the damage growth rate is related to the change in the energy potential and the amount of damage (Eq. (3)). Quantity C, determined by Eq. (6), represents the state of integrity of the material, and it assumes 1.0 at the beginning of the test (undamaged state) and 0 (or close) at the end. Quantity D, determined by Eq. (8), referred to as damage intensity, represents the amount of work required to create a certain reduction of integrity in the material. The variation in C-D curves was similar for both bitumens, and for the same reduction in the material's integrity the required damage intensity was higher in aged materials and mastics. This occurred because these materials are stiffer. As in the LAS protocol the strain amplitude is increased during the test the stiffer materials have higher stress values for the same deformation (at least before failure). D increases with the pseudo-strain amplitude (Eq. (5)), which is equivalent to the stress value of the undamaged material, and the C decrease rate.

However, although bitumen B2 and related mastics were stiffer than equivalent materials, with B1, the reduction of C was achieved with less damage intensity. Bitumen B2 showed less ability to hold increased levels of deformation than bitumen B1 which is seen by the faster decrease in shear stress after reaching the peak value (see Figures 7a and 7b). Therefore, the higher values of pseudo-strain amplitude in B2 are counterbalanced by the faster C reduction, which results in lower D values at the same C level.

The power law model fitted to C-D results was then used to derive the fatigue laws shown in Figure 7 e) and f). To this end, the specimen's failure was considered to be when the shear stress reached the maximum value. It is observed that the fatigue resistance was highly affected by the bitumen type, and with some surprising effects. The estimated number of cycles to failure was higher for bitumen B1 and related mastics, confirming the above-mentioned differences in the stress evolution with applied shear strain. However, unexpectedly, aged mastic with B1 showed greater fatigue resistance than in the unaged state. This trend was also seen for bitumen B2 but not for the related mastics. Under continuous cycle loading in the DSR, the specimen experiences cohesive failure when radial microcracks, formed along the outer edge, progress into the centre, thereby reducing the specimen's effective section. If the specimen's stiffness is too low, flow can occur at the edge, and if the stiffness is too high, failure can occur at the specimen-to-plate interface. Thus, Safaei et. al. [43] indicated a target stiffness of between 12 and 60 MPa. In the tests performed at 25 ºC, unaged bitumen B1 was softer than this and some aged mastics stiffer. However, edge flow and adhesion failure were not detected. These results suggest that, with ageing, the increase in stiffness and of the elastic behaviour (with the decrease of phase angle) was not accompanied by a considerable reduction of ductility. This is even more surprising considering that bitumen B1 was the least ageing-resistant bitumen, as shown before from the variation in conventional properties and linear viscoelastic properties. The relation between the chemical composition and the rheological behaviour of bitumen is very complex, and as stated by Petersen [1] the bitumen's reaction to ageing is very dependent on the compatibility of functional groups, and not only on its proportion. Differently, mastics with bitumen B2 suffered a significant reduction of fatigue resistance after PAV treatment.

In addition, the fatigue resistance of mastics with granite filler was lower than that of mastics with limestone filler. Granite has a greater stiffening effect than limestone, as expected from the comparison of the Rigden voids of the two fillers. Hence, in the literature, the filler stiffening effect is explained by the solid filling of soft medium (bitumen) and the ability of the filler to fixate bitumen near its particles. The more free bitumen exists in the mastic, the more likely the mastic is to deform before cracking. Hence, mastic B2-G was significantly more affected by ageing than B2-L.

4.4 Ductility

The ductility behaviour of bitumens and mastics was evaluated by using the double-edge notched tension test. The effect of ageing and filler incorporation on the force-displacement curve of specimens with equal ligament length is illustrated in Figure 8 a) and b) for the bitumens B1 and B2, respectively. Both effects induced a decrease of the elongation at failure and the increase of the peak load. A similar pattern of P-d curves for the materials with the two different bitumens can also be seen. However, toughness of materials with bitumen B2 were higher because they hold greater deformation at failure.

In this test, several notched specimens with different ligament lengths (notch size) are tested to determine the essential (and non-essential) ductile fracture energy. The application of the essential work of fracture methodology requires that [45]: (1) P-d curves obtained with ligament lengths are similar in shape; (2) ligament length is fully yielded before the initiation of cracking; (3) the volume of plastic zone is proportional to the square of the ligament length. The P-d curves of mastics with the three ligament lengths tested were plotted in Figure 8 c) and d). A similar curve shape was obtained for a single material tested, and the work of fracture (area under the curve), the peak load value and elongation at fracture increased with the ligament length. Based on the analysis of Hashemi [55], the peak load point is representative of the ligament region yielding, and the remaining work of fracture is obtained with necking and/or tearing of the ligament region. Thus, the small load drop immediately following the peak point seen in some P-d curves (e.g. Figure 8a B1-L-25h) is indicative of necking. It was therefore concluded that, in general, testing results complied with the essential work of fracture methodology.

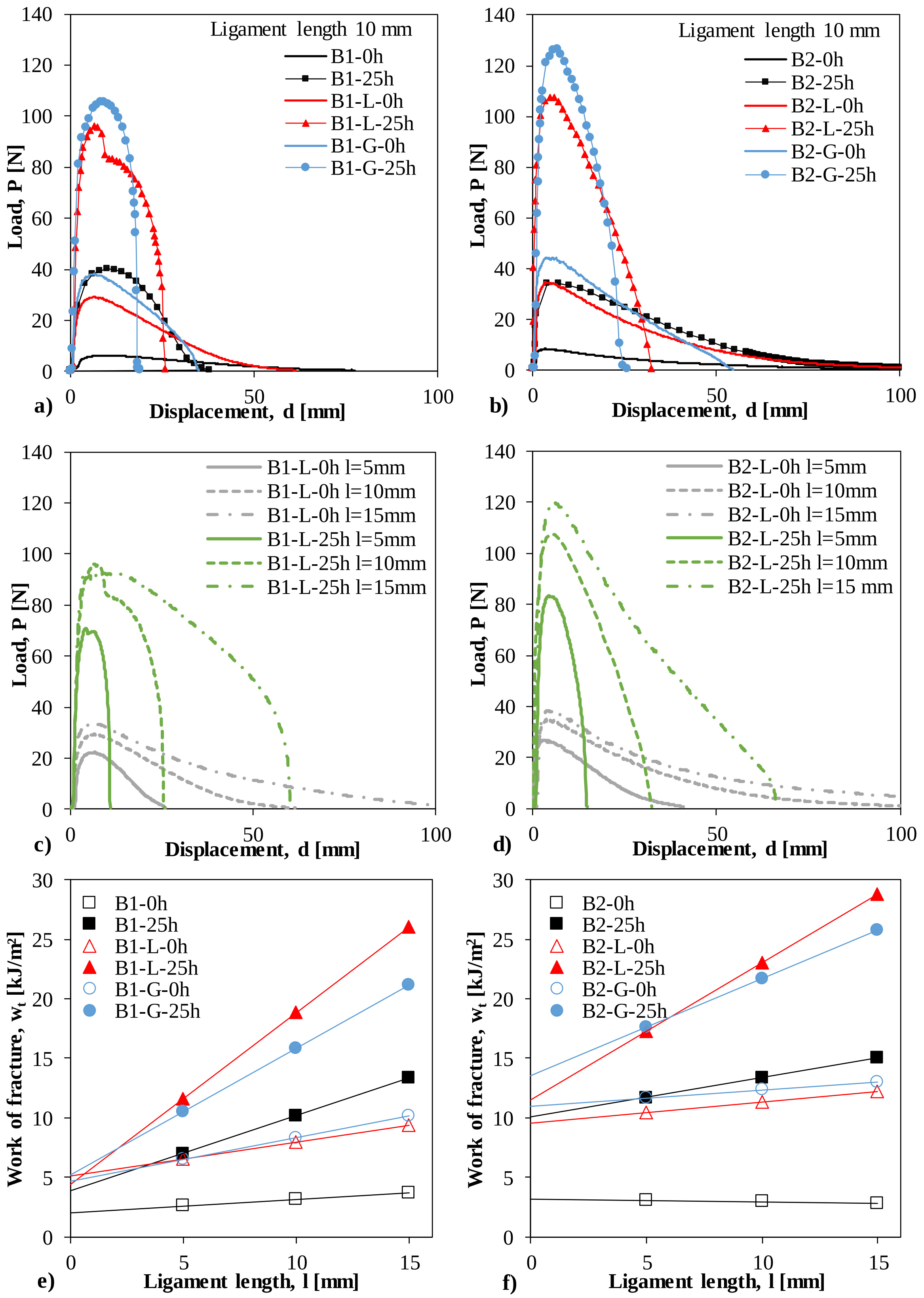

Figure 8 – DENT test results: a) P-d, bitumen B1 and different ageing levels; b) P-d, bitumen B2 and different ageing levels; c) P-d, bitumen B1 and different ligament lengths; d) P-d, bitumen B2 and different ligament lengths; e) w_t-l, bitumen 1; f) w_t-l, bitumen B2.

Figure 8 e) and f) present the variation of the work of fracture with the ligament length for the bitumen and mastics, using the bitumens B1 and B2, respectively. With the exception of the unaged bitumen B2 results, the work of facture increased significantly with the ligament length. Thus, the results fitted very well with the linear model (Eq. (13)). Only unaged bitumen B2 results did not comply with the methodology requirements, which would most likely be resolved by reducing the size of specimens, despite being less practical.

The intercept of the fitted linear model represents the essential work of fracture (w_e) of the material, which is independent of the specimen size, and the slope ($\beta \cdot w_p$) represents the effect of specimen size on measured toughness. Both ageing and filler incorporation increased the $\beta \cdot w_p$ value, which obviously occurs because plastic flow outside of the fracture zone requires more energy as the materials stiffens. In addition, the results of mastics with different fillers were close, although aged mastics with filler L mobilized more volume outside the fracture zone that contributed to specimen toughness. Thus, all specimens with filler L had longer elongations at failure than equivalent specimens with filler G.

The w_e value of bitumens increased substantially with ageing. At the test temperature adopted (20 ºC) the binders were soft, and so the specimens achieved long extensions due to plastic flow distributed over a large volume of the specimen. Different conclusions were reached for the w_e value of mastics with different bitumens. In bitumen B1, all mastics had approximately the same w_e value, while it increased after ageing for mastics with bitumen B2. For the latter, mastics with filler G had slightly higher w_e values than mastics with filler L in both ageing states.

In addition, in the literature [56] it is indicated that there is a good relation between the CTOD variable (determined from w_e and the test results of shortest ligament, Eq. (14)) and the fatigue resistance. Hence, Figure 9 plots the CTOD values versus the number of cycles to failure (at shear strain of 3.5%) estimated with the fatigue laws from LAS tests. A good relation is seen between the ductile fracture behaviour and the fatigue resistance for the materials with bitumen B2, whereas for bitumen B1 the two tests results are not related.

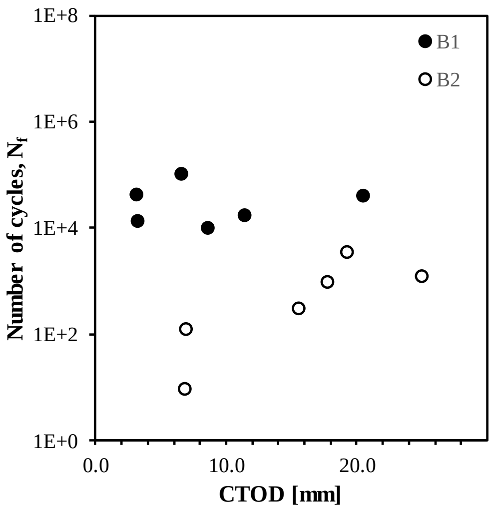

Figure 9 – Number of cycles to failure (LAS fatigue law, γ = 3.5%) versus CTOD.

4.5 Global assessment

Figure 10 compares the behaviour of materials before and after PAV ageing using a radar chart type. The axes properties (the same amplitude for the 6 plots) were defined so that the distance from the chart centre is expected to increase with ageing. From this plot, and supported by previous results and discussions presented, Table 3 summarizes the qualitative assessment of the filler effect on bitumen ageing. In the analysis, the effect of the incorporation of filler in mastic and of the type of filler was separated. For the filler incorporation, a positive mark means that mastics aged less than bitumen under the same conditions, and for the filler type, a positive mark means that mastics with limestone were less affected by ageing than mastics with granite.

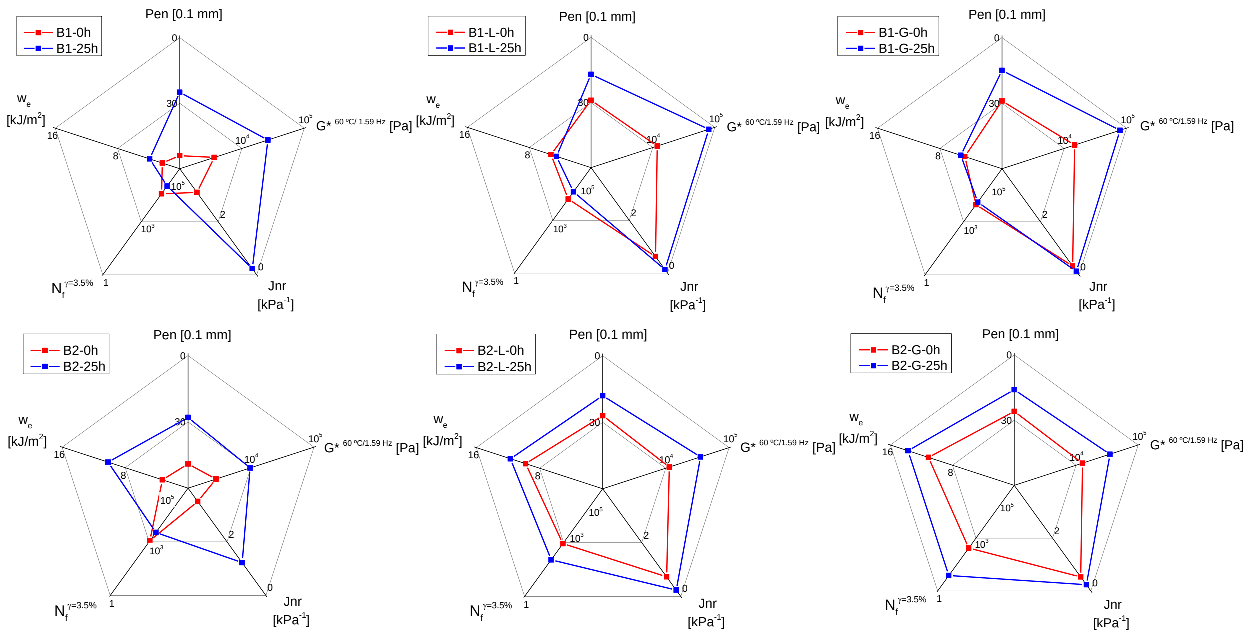

Figure 10 – Assessment of the filler effect on bitumen ageing.

Table 3 – Assessment of the filler effect on bitumen ageing

Property	Filler incorporation	Filler type (Limestone vs Granite)
Conventional		
Pen	+	=
$T_{R\&B}$	+	=
Linear viscoelastic		
G^*-δ relation	=	=
G'-f and G''-f relation	=	=
G_{25h}/G_{0h}	+	–
Rutting		
R	+	–
Jnr	=	=
Fatigue Resistance		
N_f-γ	inconclusive	+
Ductility		
w_e	+	=
$\beta \cdot w_p$	+	–
CTOD	inconclusive	=

Note: assessment marks (+ positive; = similar; – negative)

Overall, these results show that filler had a beneficial effect on bitumen ageing. The bituminous mastics had a significantly lower change in properties than bitumens when subjected to the same ageing conditions. In the literature, two main mechanisms are pointed to this effect: (1) the most polar compounds of bitumen in the fractions of asphaltenes and resins can be adsorbed by the aggregate's surface [5–7], and therefore they are less prone to oxidation reactions; (2) the filler particles in mastic form barriers to oxygen diffusion through the specimen which leads to less oxidation reactions [57].

The mastics hardened less than bitumens but the ageing related changes to the rheological behaviour were not significantly affected. Thus, the fatigue resistance and ductility testing did not show a clear beneficial effect of filler for the two bitumens. It should be noted that, for the testing conditions used, the performance of binders in these tests was improved, which makes the analysis of the filler effect more complex. In addition, the analysis did not show a clear, definitive benefit of one of these fillers over the other. Mastics with granite filler hardened less due to the ageing treatment but the fatigue resistance was more affected than mastics with limestone.

These conclusions are supported on the comparative study of the rheological and performance properties of bitumens and mastics, however the chemical changes in bitumen were not analysed. Hence, it is considered important to investigate in future the physicochemical mechanisms of the filler-bitumen interaction with these fillers and the evolution in chemical composition of bitumen with ageing.

5. Conclusions

The chemical and physical properties of bitumen change during bituminous mixture production and in service, which is commonly referred to as ageing. In the literature, it is stated that the resistance to ageing depends on the bitumen's chemical composition and the functional groups properties and compatibility. However, there is not a broad consensus on the (positive/negative) influence of filler on these changes. The research described in this paper was aimed to evaluate the effect of standard mineral fillers on bitumen ageing. To this end, two different bitumens and four mastics were studied in the unaged and aged states. The ageing treatment was induced using the Pressure Ageing Vessel to simulate the ageing state after several years in service. The mineral fillers used in this study were limestone and granite. The bituminous materials were characterized by their conventional properties, the viscoelastic properties, the fatigue resistance and the ductility properties.

Globally, the factors that determined the mastics' properties were primarily the bitumen and filler properties, and then ageing. The consistency of bitumen and mastics increased significantly due to ageing, although the mastics stiffened to a lesser extent than bitumen. Thus, in comparison to bitumen, the retained penetration increased up to 10%, the softening point variation reduced up to 3 ºC and the increase of complex shear moduli reduced up to 30%. In addition, this trend was seen with both bitumens, which, due to the different chemical composition, had significantly different ageing resistance.

In addition, the rheological behaviour of the materials became more elastic with ageing. Hence, aged materials showed higher deformation recovery with cyclic loading. This occurred because the phase angle decreased over the tested temperature-frequency range. The storage shear modulus proved to be more sensitive to the ageing and filler effect than the loss and absolute shear moduli, especially in the high temperature range. Thus, mastics were less affected by these changes than bitumens. On the other hand, the effect of loading frequency on the complex shear modulus also reduced with ageing, but this change was not affected by the filler.

Contrary to this, the performance tests were not definitive regarding the effect of filler on bitumen ageing. Due to the stiffening, aged materials were less able to hold large deformations levels despite a large increase in required deformation energy/work in some cases. Hence, aged bitumens showed greater fatigue resistance and higher specific energy of ductile fracture that unaged bitumens. Mastics showed minor variations in the specific energy of ductile fracture with ageing treatment, which is indicative of less ageing, but the fatigue resistance decreased significantly with one of the bitumens.

Regarding the effect of the two fillers, granite induced larger stiffening in mastics than limestone, but the rheological behaviour of mastics was less affected by ageing. Differences in the stiffness ageing indexes reached 20%. Granite had larger Rigden voids, which is indicative of the amount of bitumen fixated by filler particles, and therefore more bitumen could have been protected from ageing inducing mechanisms. However, differences in consistency between mastics were not significant. Both mastics showed similar behaviour in the tension test for the ductility fracture analysis, although mastics with granite showed lower fatigue resistance.

From these research outcomes, it is considered that further studies are required. On the one hand, the performance tests may have been influenced by the test temperature (20/25 ºC), and therefore additional mechanical testing should be performed at lower temperatures. On the other hand, the chemical changes in bitumen/mastics due to ageing were not analysed in this study, and the fillers were not characterized in terms of the chemical composition. Therefore, it is recommended to characterize in detail the fillers (chemical composition, particles surface with SEM – Scanning Electron Microscopy) and the chemical composition evolution of bitumen/mastics due to ageing with FTIR – Fourier-transform Infrared Spectroscopy.

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