

# Rheological characteristics of ground tire rubber-modified bitumens

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## Abstract

This work deals with the characterisation of the linear viscoelastic properties, closely related to its performance as road paving binder, of ambient ground tire rubber-modified bitumen. Specifically, the influence that rubber particle size exerts on the rheological properties of these binders has been studied. The resulting viscoelastic behaviour has been compared with that shown by unmodified and polymer-modified (SBS) bitumens. In total, an unmodified bitumen, five ground tire rubber-modified and three polymer-modified (SBS) bitumens have been studied. Rubber-modified bitumens show improved viscoelastic characteristics and, therefore, higher viscosity than unmodified binders. Consequently, enhanced resistance to permanent deformation or rutting and low-temperature cracking should be expected in ground tire rubber-modified bitumens. In addition, rubber-modified bitumen (9 wt.%) shows very similar linear viscoelastic properties to SBS-modified bitumen having 3 wt.% SBS at  $-10^{\circ}\text{C}$ , and 7 wt.% SBS at  $75^{\circ}\text{C}$ .

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*Keywords:* Bitumen; Viscoelastic behaviour; Crumb rubber

## 1. Introduction

Bitumen has been widely used as an adhesive material in pavement mixtures, surface dressing, bridge deck waterproofing, overlays and the protection of buildings, for example waterproofing roof and joint and crack seals. This is because asphalt is strong, readily adhesive, highly waterproof and durable [1]. In general, the components of bitumen can be broadly categorised as maltenes and asphaltenes. The viscoelastic properties of bitumen, and consequently its performance as a road paving binder, are dramatically influenced by the ratio between the asphaltene and maltene fractions [2–4].

There are several major distresses of road pavement related to bitumen properties. Since bitumen is a viscoelastic material, its rheological properties are very sensitive to temperature as well as to the rate of loading. With respect to temperature, the most frequent problems of road pavement are rutting, fatigue cracking and thermal cracking [3]. Polymer modification has beneficial effects on bitumen binders and therefore on road pavements: decreased thermal susceptibility and permanent deformation (rutting) and increased resistance to low-temperature cracking, since the binder undergoes a decrease in its effective glass transition temperature [3–8].

A limited number of polymers have been used as modifying agents. However, the high cost of these polymers compared to bitumen means that the amount of polymer needed to improve pavement performance should be as small as possible [9]. This major restriction could be avoided by using recycled polymers, such as granulated tire rubbers.

From an environmental and economic standpoint, the use of ground tire rubber as a bitumen-modifying agent may contribute to solving a waste disposal problem and to improving the quality of road pavements. Two different ground tire rubbers may be obtained depending on the temperature of the process: (A) ambient ground rubber, which is obtained at room temperature and has a sponge-like surface that, consequently, has a large surface area, which increases the reaction rate with bitumen. (B) cryogenically ground rubber, which is obtained at or below the embrittlement temperature of the rubber, and produces clean flat surfaces, which reduce the reaction rate [10].

Different studies have noted the relationship between the rheological characteristics of asphalt binders and field performance [11], illustrating the importance of understanding the rheological behaviour of bitumen. Bitumen is a Newtonian fluid when handled and mixed with mineral aggregates at high temperatures. The linear viscoelastic region describes the resistance of the bitumen to traffic loading (rutting and cracking due to fatigue) [4].

The main objective of this work has been to study the rheology of ambient ground tire rubber-modified bitumen.

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To this end, the effect that particle size exerts on the linear viscoelastic properties and flow behaviour of these binders has been studied. Their rheological characteristics have been compared with those shown by unmodified and polymer-modified (SBS) bitumens.

## 2. Experimental

An unmodified bitumen 60/70 penetration grade, supplied by Construcciones Morales, S.A. (Spain), three SBS-modified (1, 3 and 7 wt.% SBS) bitumens and five ground tire rubber-modified bitumens were tested in this study.

Styrene-butadiene-styrene triblock copolymer (SBS), Kraton D-1101CS, provided by Shell Chemical Company (UK) and ambient ground tire rubber, M35 (Table 1), donated by Alfredo Mesalles, S.A. (Spain) were used as modifying agents. The commercial ground rubber, M35, had a mean particle diameter of 0.63 mm and a wide particle size distribution. Five rubber fractions (0.1, 0.29, 0.35, 0.63 and 0.74 mm mean diameters) were separated by screening from sample M35. The morphology of the rubber particles was observed with a Jeol JSM-5410 scanning electron microscope (20 kV and 7500 $\times$ ), using samples coated with gold to avoid charging. Modified binders were processed for 1.5 h in an open low-shear batch mixer at 180 °C and a rotating speed of 1200 rpm. An unmodified bitumen (60/70) sample was also submitted to these processing conditions.

In order to analyse rubber devulcanization and depolymerization due to processing, rubber-modified bitumen was dissolved in tetrahydrofuran (THF). The insoluble particles (remaining rubber with particles of carbon black, etc.) were removed by filtration using Albet 150 filter papers (3–5  $\mu$ m pore size). THF was removed from the soluble fraction in a vacuum oven at 50 °C. Previous analysis carried out with fresh (non-processed) crumb rubber showed that 11 wt.% of its components were soluble in THF (Tables 1 and 2). Such components would correspond to process oils, plasticizers and other tire rubber additives.

Frequency sweep tests, between 0.01 and 100 rad/s, in the linear viscoelasticity range, were performed in a controlled-stress Haake RS150 rheometer, using profiled plate-and-plate geometries (10 and 20 mm diameter; 1 and 2 mm gap). Stress sweep tests, at the frequency of 1 Hz, were previously carried out on each sample to determine the linear viscoelasticity region. Time-sweep tests, at a constant stress and frequency, were also performed to confirm

that no structural modifications occurred during the time required for each test. Measurements were done in a temperature range between –10 and 75 °C. Flow measurements at 50 °C were conducted in Haake RS150 rheometer, using a profiled plate-and-plate geometry (20 mm diameter; 1 and 2 mm gap).

## 3. Results and discussion

### 3.1. Influence of processing temperature

Prior to studying the influence of processing variables on the viscoelastic properties of modified binders, the effect of processing conditions on the unmodified bitumen used was investigated. Fig. 1 shows the frequency dependence of the viscoelastic functions, storage ( $G'$ ) and loss ( $G''$ ) moduli at –10 °C for bitumen 60/70 and the evolutions of above-mentioned functions when this material was processed at 180 °C and 1200 rpm (bitumen 180). As may be seen, both samples show higher values of the loss modulus in the low-frequency region, compared to the storage modulus. By contrast, higher values of the storage modulus are found at high frequencies. As a consequence, for both samples, the storage and loss moduli undergo a continuous increase with frequency and reach a cross-over point at an intermediate frequency. This cross-over frequency is of the order of 0.3 rad/s for bitumen 180, and 0.7 rad/s for bitumen 60/70. However, the values of the storage modulus at the cross-over frequency are approximately the same in both cases ( $\approx 10^7$  Pa). Most of the bitumen components are of low molecular weight, so that entanglement effects are not significant and, therefore, a continuous transition from the elastic (glassy) to the Newtonian region takes place by decreasing the frequency [4].

As may be seen in Fig. 1, the processing conditions, to which bitumen 180 was submitted, give rise to an increase in both moduli in comparison with bitumen 60/70, although the resulting viscoelastic curves display a similar shape. Hence, thermal treatment favours bitumen hardening or “primary ageing” [12].

Rather different viscoelastic behaviours were found at 75 °C (Fig. 2). Thus, both samples exhibit higher values of the loss modulus than the storage modulus in the experimental frequency window, having  $G'$  and  $G''$  slopes close

Table 1  
Chemical composition for fresh crumb rubber (non-processed)

Material	wt.%
Total rubber hydrocarbon (natural and synthetic rubber)	50 $\pm$ 5
Carbon black	32 $\pm$ 3
THF extractable	11 $\pm$ 3
Ash	4 $\pm$ 2

Table 2  
Remaining insoluble rubber after bitumen processing

Sample (9 wt.%, 180 °C)	Insoluble rubber (wt.%)
0.10 mm	85
0.29 mm	86
0.35 mm	85
0.63 mm	85
0.74 mm	86
Fresh crumb rubber (non-processed)	89

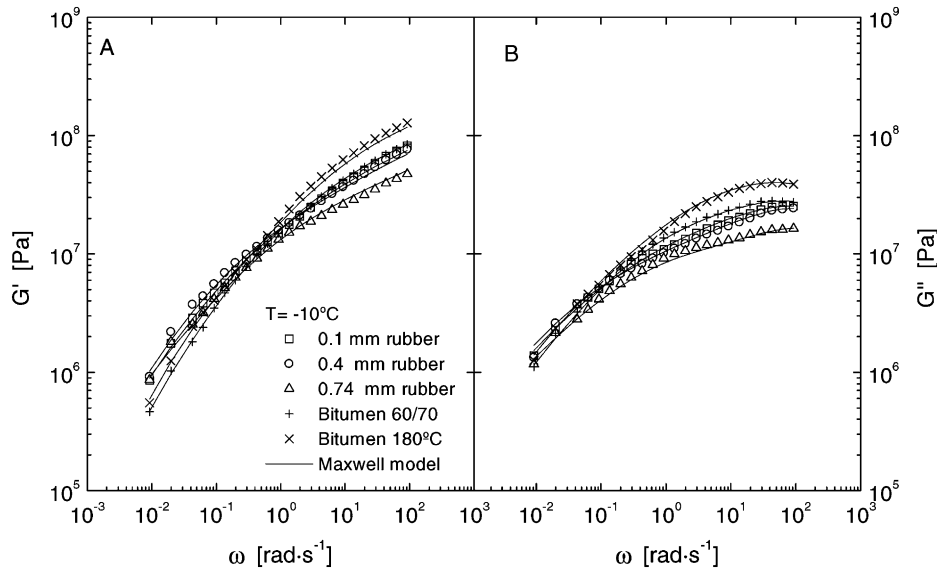


Fig. 1. Evolution of the storage and loss moduli with frequency at  $-10^{\circ}\text{C}$  for different bitumens.

to 2 and 1, respectively, as corresponds to the Newtonian or terminal region of the mechanical spectrum. These results demonstrate the predominantly viscous characteristics of these materials at  $75^{\circ}\text{C}$ . At this temperature, the differences between bitumen 60/70 and bitumen 180 samples are even more important. Thus, the processed sample shows higher values of both moduli in the whole frequency range. Bitumen hardening or primary ageing, as a consequence of processing, results from the partial oxidation of the maltene fraction, leading to an increase in asphaltene concentration [13], in this case from 20.2 to 21.2 wt.% This fact was also confirmed by the increase in zero-shear-limiting viscosity,  $\eta_0$ , found at  $50^{\circ}\text{C}$ , from  $4.6 \times 10^3 \text{ Pa s}$  for bitumen 60/70 to  $5.8 \times 10^3 \text{ Pa s}$  for

bitumen 180. Similar results have been also shown by other authors [14].

An increase in the storage modulus (which contributes to the increase in viscosity) improves asphalt performance at high temperature, such as permanent deformation or rutting [15–17]. On the other hand, such an increase may favour low-temperature cracking, as a result of brittle fracture of the glassy bitumen matrix, which would appear at higher temperatures after processing at  $180^{\circ}\text{C}$  [3–8]. As a consequence, a suitable modifying agent should reduce the values of  $G'$  at low temperatures and increase them at high temperatures [18–21]. In the following sections, the use of ground tire rubber as a modifying agent will be evaluated.

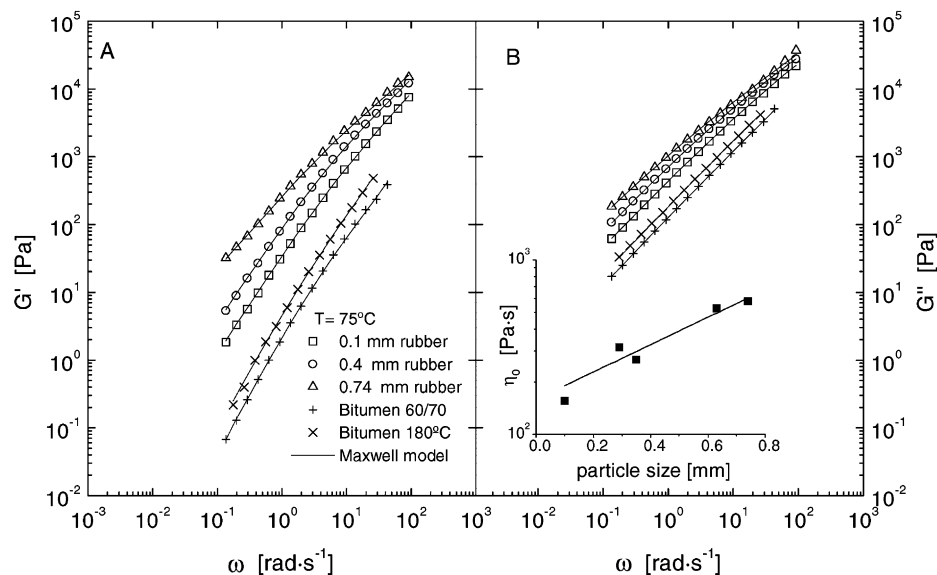


Fig. 2. Evolution of the storage and loss moduli with frequency at  $75^{\circ}\text{C}$  for different bitumens.

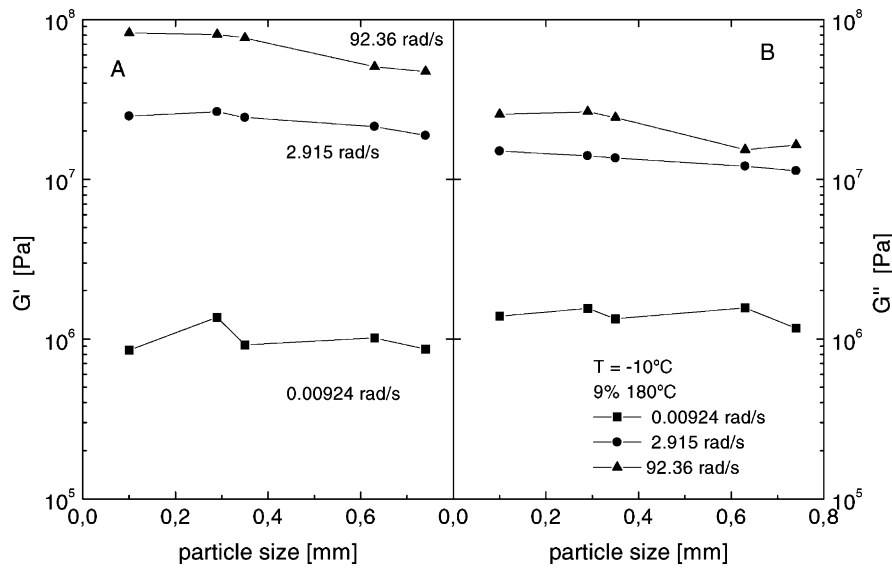


Fig. 3. Evolution of the storage and loss moduli with rubber particle size at three selected frequencies at  $-10^{\circ}\text{C}$ .

### 3.2. Effect of tire rubber addition

Fig. 1 shows the influence of particle size on the linear viscoelastic behaviour at  $-10^{\circ}\text{C}$  of selected ground tire rubber-modified bitumens with the same concentration (9 wt.% rubber). If we compare rubber-modified bitumens and processed bitumen (bitumen 180), it is apparent that rubber addition decreases the values of both moduli at  $-10^{\circ}\text{C}$  in the high-frequency region. Such a reduction is more evident as rubber particle size increases (see Figs. 1 and 3). Thus, a rubber-modified bitumen with a rubber particle size of 0.1 mm shows values slightly lower than bitumen 60/70, whilst a rubber-modified bitumen with a rubber particle size of 0.7 mm shows the minimum values of  $G'$  and  $G''$ . As mentioned before, a decrease in the storage modulus at

low-temperature yields positive effects on the performance of a road asphalt binder, since rubber addition seems to move the glassy region of the mechanical spectrum to a higher frequency (or lower temperature). Hence, thermal cracking would appear at lower temperatures.

At high temperatures (i.e.  $75^{\circ}\text{C}$ ), the rheological behaviour shown by rubber-modified bitumens is similar to that displayed by unmodified binders (Fig. 2). Thus, they show a predominantly viscous behaviour ( $G'' > G'$ ) in the whole frequency range studied, typical of the terminal region of the mechanical spectrum. Rubber addition leads to an increase in both storage and loss moduli, which is more important as particle size becomes larger, showing an exponential increase in both moduli, more apparent at low frequencies (Fig. 4).

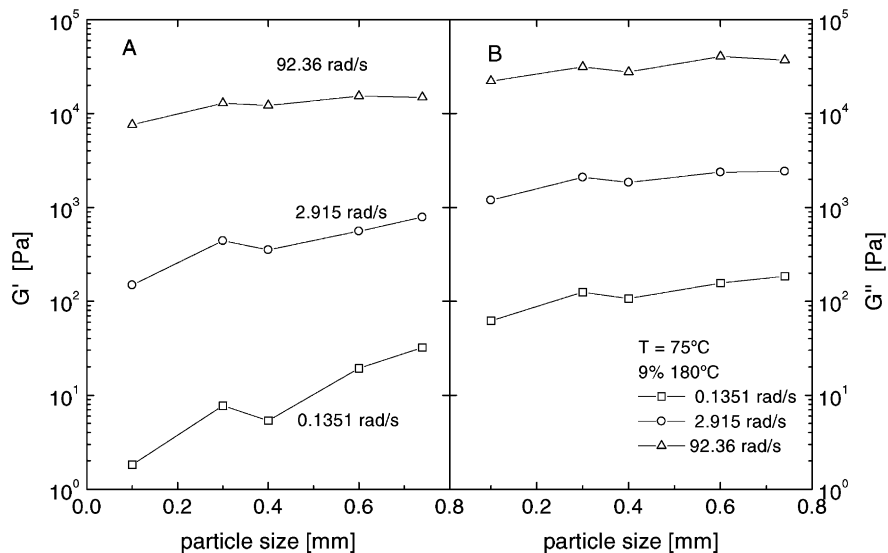


Fig. 4. Evolution of the storage and loss moduli with rubber particle size at three selected frequencies at  $75^{\circ}\text{C}$ .

The dynamic linear viscoelastic behaviour of these systems may be described by a generalised Maxwell model (Figs. 1 and 2):

$$G' = G_e + \sum_{i=1}^N G_i \frac{(\omega\lambda_i)^2}{1 + (\omega\lambda_i)^2} \quad (1)$$

$$G'' = \sum_{i=1}^N G_i \frac{\omega\lambda_i}{1 + (\omega\lambda_i)^2} \quad (2)$$

where  $G_e$  is the elastic modulus.

This model considers a superposition of a series of  $N$  independent relaxation processes, each process having a relaxation time  $\lambda_i$  and a relaxation strength  $G_i$ . The resulting distribution or spectrum of relaxation times may be used to obtain the zero-shear-limiting viscosity of the material,  $\eta_0$ , as follows:

$$\eta_0 = \sum_{i=1}^N G_i \lambda_i \quad (3)$$

where  $N = 15$  for all samples studied.

The evolution of  $\eta_0$  with particle size at 75 °C has been inserted in Fig. 2B. Zero-shear-limiting viscosity at 75 °C undergoes an exponential increase as rubber particle size (PS) does.

$$\eta_0 = 158 e^{1.80 PS} \quad (4)$$

As a result, at high temperatures, rubber-modified bitumens display improved viscoelastic characteristics and, therefore, higher viscosity than unmodified binders. Consequently, enhanced resistance to permanent deformation or rutting should be expected in ground tire rubber-modified bitumens.

### 3.3. Rheology and microstructure

The presence of insoluble rubber particles in the bitumen after processing was confirmed by a filter test. Rubber-modified bitumens were dissolved in tetrahydrofuran (THF), where the only insoluble components are rubber waste consisting of sulphur-cross-linked rubber-containing carbon black [22]. A comparison of the original network (fresh crumb rubber) with that resulting after processing provides information on the devulcanization/depolymerization process. As shown in Table 2, approximately 85 wt.% insoluble rubber particles remain after processing. In comparison with fresh crumb rubber, a slight decrease ( $\approx 4$  wt.%) in insoluble components is detected after mixing at 180 °C, which is not affected by particle size and must be related to a devulcanization/depolymerization process [23]. As a result, the rheological properties of rubber-modified bitumens would depend on both soluble and insoluble components of ground rubber.

As may be deduced from Fig. 5, bitumen modified just by the soluble rubber fraction, obtained by removing the insoluble particles by filtration, shows enhanced viscoelastic characteristics compared with bitumen 180. However, the effect of the insoluble components is more evident, especially at high temperatures, where the terminal or flow region of the mechanical spectrum appears. Hence, at high temperatures the rheological response of the material seems to be mainly affected by the presence of rubber particles.

Rheology of suspensions can be influenced by particle Brownian motion, shape anisotropy, buoyancy and sedimentation forces as well as a variety of interaction forces between particles [24]. However, if the fluid phase surrounding the particles has a very high viscosity (above 1 Pa s), as is the case here (see Fig. 6), then the viscous forces, imposed by

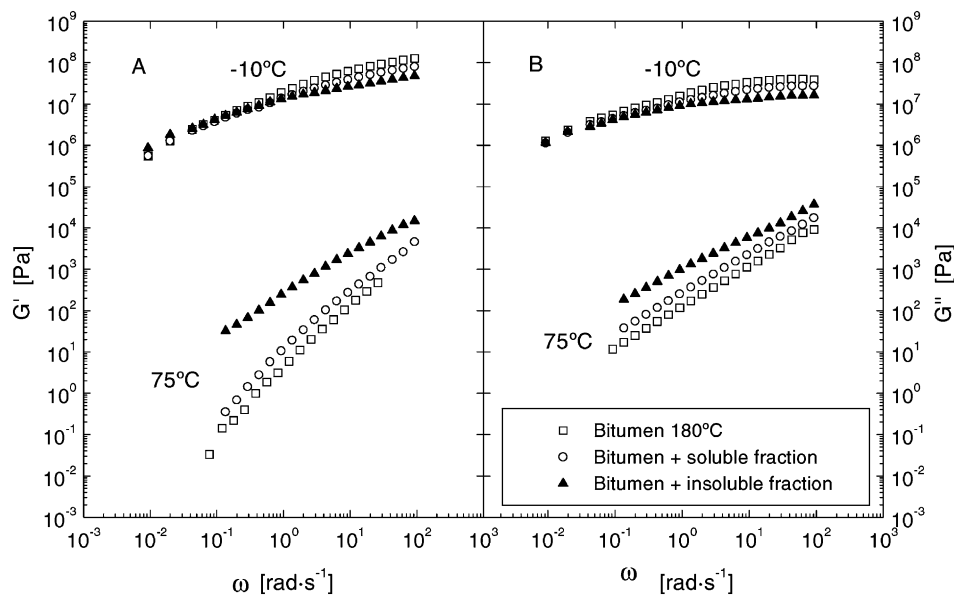


Fig. 5. Effect of soluble and insoluble rubber components on the linear viscoelastic functions of modified bitumens at  $-10$  and  $75^\circ\text{C}$ .

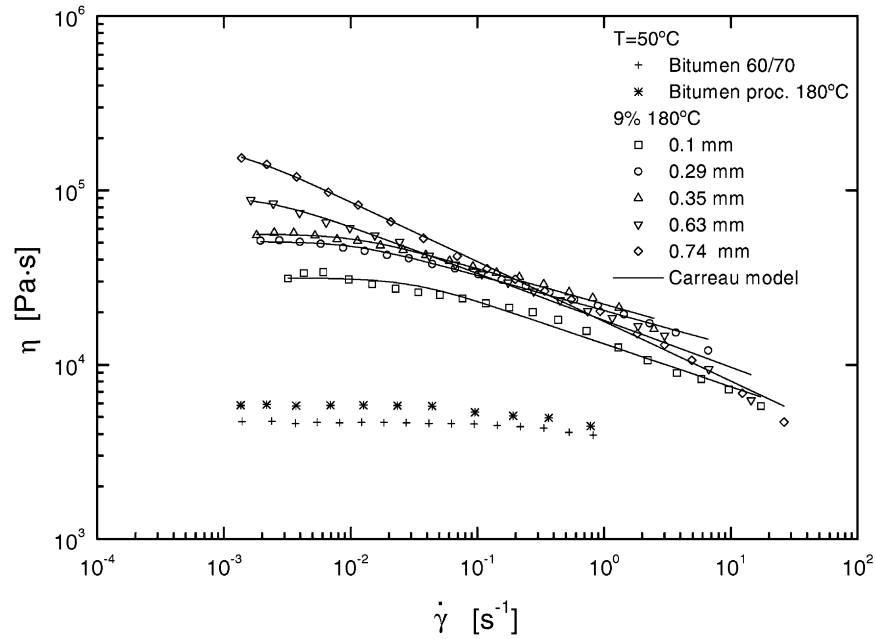


Fig. 6. Influence of particle size on the flow behaviour of ground tire rubber-modified bitumens.

the fluid on the particles upon inception of flow are so large that particle–particle interactions will be negligible. Consequently, no long-range “structures” can build as a result of forces between the particles, and they act as truly inert suspended particles [25]. A fact confirmed by the predominant viscous properties shown by the modified bitumens studied at high temperature (Fig. 2).

In addition to this, it should be expected that the presence of the particles would increase the flow resistance of the material, since the adhesion of the fluid at the particle–liquid interface must increase the viscous dissipation in the liquid [26]. This result may be also seen in Fig. 6, where a remarkable increase in viscosity with rubber addition is shown. Moreover, a modification of the rheological response is found, from a Newtonian behaviour in a wide range of shear rates (bitumen 60/70 and bitumen 180) to a shear-thinning behaviour (rubber-modified bitumens). Thus, the flow curves show a tendency to a constant viscosity at low shear rates,  $\eta_0$ , followed by a power-law decrease in viscosity. As a result, flow behaviour shown by rubber-modified bitumens, is properly described by the Carreau model [27]:

$$\eta = \frac{\eta_0}{(1 + (\lambda_c \dot{\gamma})^2)^s} \quad (5)$$

where  $s$  is a parameter related to the slope of the shear-thinning region and  $\lambda_c$  a characteristic time of the material, defined as  $\lambda_c = 1/\dot{\gamma}_c$ , where  $\dot{\gamma}_c$  is a critical shear rate for the onset of this intermediate region.

Fig. 7 shows the influence of particle size on the Carreau model parameters. As may be seen, both  $\lambda_c$  and  $\eta_0$  undergo

an exponential increase with particle size at 50 °C:

$$\lambda_c = 16.2 e^{5.34 PS} \quad (6)$$

$$\eta_0 = 2.2 \times 10^4 e^{2.62 PS} \quad (7)$$

Taking into account the fact that the solid volume fraction remains almost constant after processing (Table 2), an increase in particle size would lead to a decrease in viscosity, since specific surface area of particles decreases as well. However, particle shape must also be taken into consideration. Thus, different authors found a dependence of the particle aspect ratio (length/diameter) on the viscosity of polymeric fluids [28–30] so that higher aspect ratios lead to higher viscosities [25,30]. Hence, the increase in viscosity observed with increasing particle size would be related to an increase in the aspect ratio of the rubber. This assumption is confirmed in Fig. 8, which shows the rubber particle shape of two samples having different size (0.29 and 0.74 mm). As may be seen, rubber particles are not spherical, furthermore aspect ratio increases with particle size (from an approximate value of 1.2 for sample 0.29 mm to 2.9 for sample 0.74 mm).

The exponential increase in  $\lambda_c$  with particle size (Fig. 7) implies that the critical shear rate, for the onset of the shear-thinning region, related to the progressive orientation of the non-spherical particles along the shear field, decreases. As may be observed in Fig. 7, the slope of power-law region increases with particle size. An interesting technological result may be deduced from this fact. Namely, bitumen containing the largest particle size, which shows enhanced rheological behaviour at rest (in the linear viscoelastic region and at low shear rate), also shows the

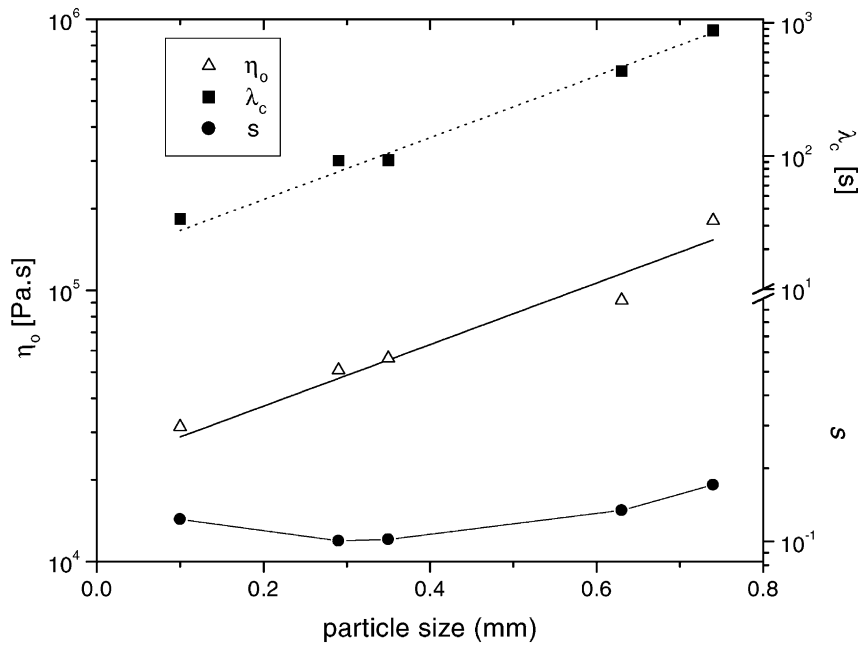


Fig. 7. Influence of particle size on Carreau parameters.

lowest viscosity at high shear rates, where it would be handled and mixed with mineral aggregates at high temperature (around 135 °C). However, undissolved and undispersed crumb rubber in the bitumens may cause a high viscosity in the compaction temperature region (above 115 °C) and sedimentation of the rubber particles during transporting and storage of the binders (163–180 °C) [23,31].

### 3.4. SBS-modified bitumens

Finally, it has been considered quite interesting to compare the rheological behaviours shown by both ground rubber-modified and SBS-modified bitumens, the latter being widely used as asphalt binder (Figs. 9 and 10). Both bitumens show similar linear viscoelastic behaviours and functions at –10 and 75 °C.

If a SBS elastomer is added to bitumen, it absorbs part of the maltene fraction (i.e. saturates and naphthene aromatics) [20], with which the polymer is extended. Consequently, SBS-modified bitumen is a multiphase system formed by a polymer-rich phase and an asphaltene-rich phase [20,32]. The SBS builds a three-dimensional network in the polymer-rich phase with the swollen polystyrene endblocks acting as physical cross-links. Depending on the polymer concentration and the swelling ability of the maltenic fraction, the polymer-rich phase may become the continuous one. It is apparent, from the experimental results obtained (Fig. 9), that the asphaltene-rich phase is the continuous one in the polymer-modified bitumens studied in this work (up to 7 wt.% SBS). Thus, the addition of SBS to the unmodified bitumen only produces an increase in the linear viscoelasticity functions at high temperatures, mainly

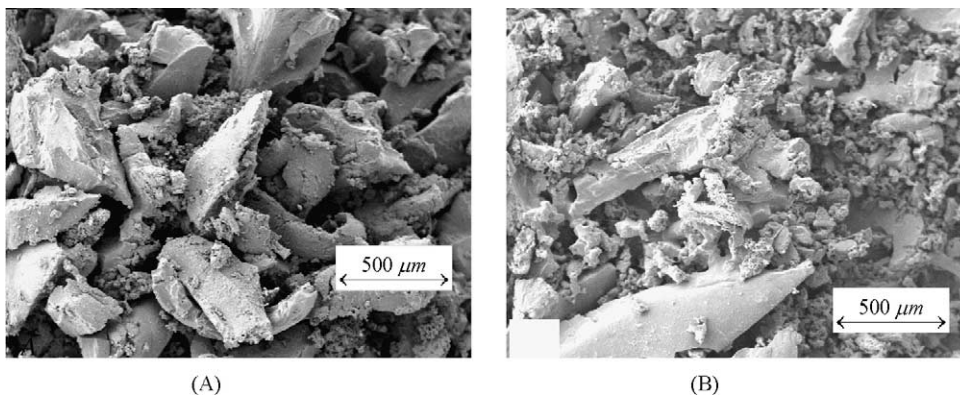


Fig. 8. SEM of two ground samples having different mean particle size: (A) 0.74 mm; (B) 0.29 mm.

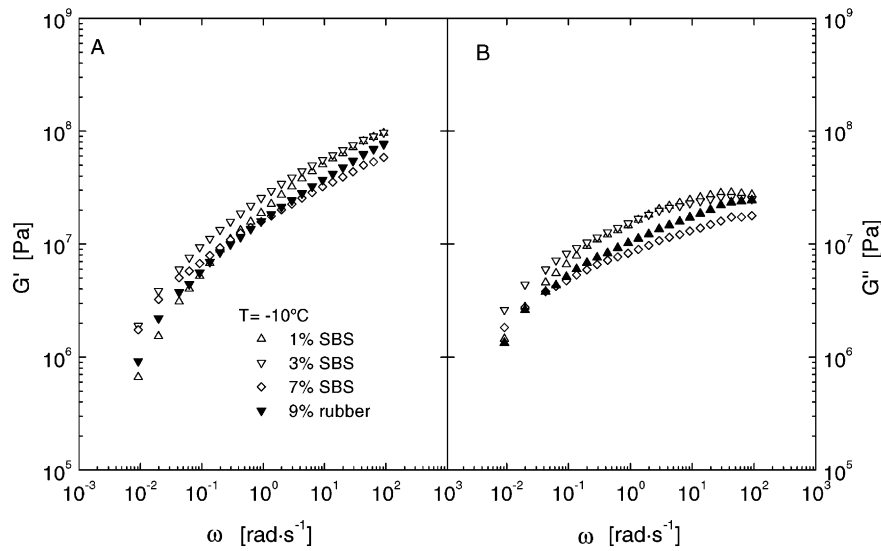


Fig. 9. Evolution of the storage and loss moduli with frequency at  $-10^{\circ}\text{C}$  for rubber-modified and SBS-modified bitumens.

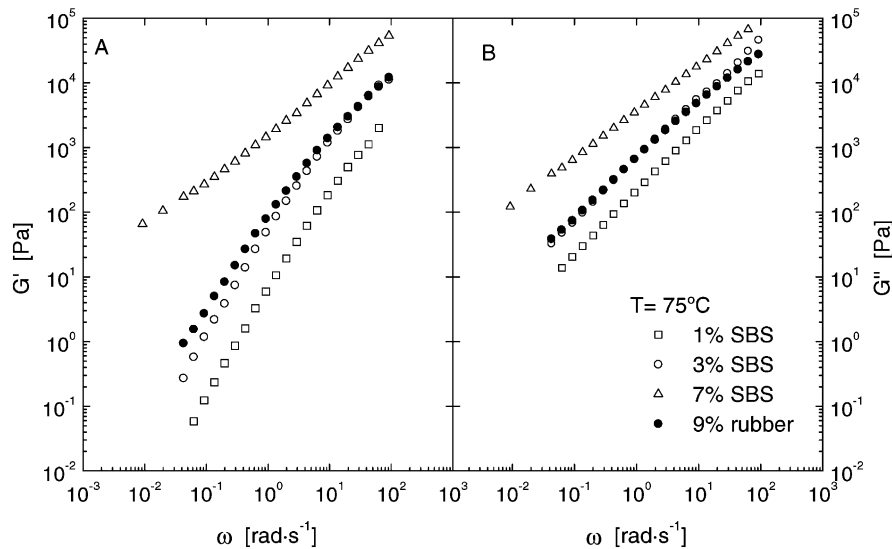


Fig. 10. Evolution of the storage and loss moduli with frequency at  $75^{\circ}\text{C}$  for rubber-modified and SBS-modified bitumens.

in the low-frequency range, and a slight decrease in thermal susceptibility, due to the fact that  $G'$  and  $G''$  decrease at low temperatures. As can be deduced from Figs. 9 and 10, 9 wt.% rubber addition (0.35 mm particle size) to bitumen provides similar viscoelastic properties to those shown by 7 wt.% SBS-modified binder at  $-10^{\circ}\text{C}$ , and by 3 wt.% SBS-modified binder at  $75^{\circ}\text{C}$ .

#### 4. Conclusions

All the modified and unmodified bitumens studied show a continuous transition from the elastic (glassy) to the Newtonian region, which takes place by decreasing the frequency at low temperatures. In contrast, these materials

show predominantly viscous characteristics at  $75^{\circ}\text{C}$ , typical of the terminal region of the mechanical spectrum.

The effect of processing on the unmodified bitumen is to increase both the storage and loss moduli. This fact contributes to the reduction of permanent deformation or rutting but it favours low-temperature cracking, as a result of brittle fracture of the glassy bitumen matrix.

Bitumen modification using ground tire rubber produces a significant increase in both moduli at high temperature. On the contrary, a significant decrease in the values of both linear viscoelasticity functions is observed at low temperature (i.e.  $-10^{\circ}\text{C}$ ). This effect is more important as the rubber particle size increases. As a result, rubber-modified bitumens show improved viscoelastic characteristics and, therefore, higher viscosity than unmodified binders. Consequently,



enhanced resistance to permanent deformation or rutting and low-temperature cracking should be expected in ground tire rubber-modified bitumens.

The addition of SBS to the bitumen only produces an increase in the linear viscoelasticity functions at high temperatures, mainly in the low-frequency range, and a slight decrease in thermal susceptibility, due to the fact that  $G'$  and  $G''$  decrease at low temperatures. Rubber-modified bitumen (9 wt.%) shows very similar linear viscoelastic properties to SBS-modified bitumen having 3 wt.% SBS at  $-10^{\circ}\text{C}$ , and 7 wt.% SBS at  $75^{\circ}\text{C}$ .

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