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ORIGINAL CONTRIBUTION

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The rheology of recycled EVA/LDPE modified bitumen

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Abstract This paper describes linear viscoelasticity, at low and intermediate temperatures, and the flow behaviour, at high temperatures, of polymer modified bitumen (PMB) containing 5 and 9 wt% recycled EVA/LDPE. The relationship between flow behaviour and microstructure of the modified bitumen was also considered, by comparison of experiments carried out in capillary and rotational rheometers and photomicrographs taken using a microscopy system whilst the sample was being sheared. Blends of 60/70 penetration grade bitumen and waste plastic (EVA/LDPE) were processed in an open mixer using a four blade propeller. Rheological tests, differential scanning calorimetry (DSC) and microscopy showed that the bitumen performance was improved by adding the recycled polymer. As a consequence, the use of recycled EVA/LDPE in PMBs can be considered a suitable and interesting alternative from both an environmental and economical point of view. The experimental results also show that pure bitumen has shear-thinning characteristics. The blending of polymer into the bitumen modifies the melt processing characteristics of the blend, whilst the viscoelastic properties of the semi-solid composite are enhanced.

Keywords Modified bitumen · Recycled polymer · Flow behaviour · Linear viscoelasticity · Capillary rheometry

Introduction

Bitumen has different applications, and it is mostly used in the road and paving industry. The performance of such surfaces depends on the binding properties of bitumen, as the bitumen constitutes the continuous matrix and is the major component of the system that deforms under load (Becker et al. 2003). Bitumen may also have application in roofing membranes and in other waterproofing materials (García-Morales et al. 2004a). It is generally assumed that bitumen, produced from crude oil distillation processing, is a colloidal system in which the asphaltenes are dispersed into an oily matrix of the remaining components, the maltenes (Pérez-Lepe et al. 2003), and this colloidal structure serves as a model to describe some of the observed thermo-mechanical properties of bitumen. Other bitumen models have been proposed, such as the SHRP model (Bonemazzi and Giavarini 1999; Redelius 2000).

Extreme in-service temperatures as well as high traffic loadings may cause significant problems in bituminous pavements and roads, among which deformation at high temperature (rutting), thermal cracking at low temperature, load-associated fatigue cracking and ageing are the most common (Becker et al. 2003). For this reason, a significant amount of work focused on the enhancement of the performance of paving materials has been and is being carried out.

Bitumen modification by polymer addition usually improves the mechanical properties of the composite and, therefore, the behaviour of road pavements: thermal susceptibility and rutting can be diminished, whilst the resistance to low temperature cracking may increase, since the binder undergoes a decrease in its effective glass transition temperature (García-Morales et al. 2004a). Polyolefins such as polyethylene (HDPE, LLDPE or LDPE) and copolymers, such as SBS and EVA have been commonly employed (Airey 2003; Blanco et al. 1996; Yousefi 2003).

From an environmental and economical point of view, the addition of waste plastics to the road bitumen is a good way of achieving waste disposal, also taking into account that the cost is an important aspect when selecting a polymer as a modifying agent (García-Morales et al. 2004a, 2004b).

Knowledge of the rheological behaviour of bitumen is necessary (Partal et al. 1999). However, the properties of binders have usually been characterised using standard and/or modified standard test methods, which do not necessarily accurately predict the full bitumen performance. The appearance of the Strategic Highway Research Program (SHRP) protocol favoured the use of the dynamic shear rheometry (DSR) to characterise the mechanical behaviour of polymer modified bitumen over a wide range of temperatures (García-Morales et al. 2004b). Viscoelasticity measurements provide useful information about the resistance of bitumen to traffic loading (rutting and fatigue cracking) in the high in-service temperature region, as well as at low temperature where thermal cracking is likely to happen. In addition, viscous measurements, at higher temperatures, provide information about the processing properties of the bitumen where it is handled and mixed with mineral aggregates, and finally applied on the road.

In this paper the use of a blend of recycled ethylene-vinyl acetate copolymer and low-density polyethylene as a modifying agent for the bituminous binder employed in the construction of road pavements is examined. Different rheological measurements were carried out on neat and modified bitumen over both application and processing temperature ranges. Linear viscoelasticity tests were performed at low and intermediate temperatures (related to bitumen in-service properties) and viscous flow tests were carried out at high temperatures (related to bitumen modification processing and road paving: mixing characteristics, laydown, compaction).

Experimental

Bitumen of penetration grade 60/70, provided by Construcciones Morales S.A. (Spain), was used as a base material for polymer modification. Waste plastic (EVA/LDPE) from agriculture, provided by Egmasa (Spain), was used as the modifying agent. Asphaltene content, determined by the procedure outlined in ASTM D3279, and penetration grade of the base bitumen, as well as some physico-chemical characteristics of the polymer are shown in Table 1.

Blends of bitumen and polymer, at 5 and 9 wt% polymer concentrations, were prepared in an open mixer, using an IKA RW-20 stirring device (Germany). Samples were processed for 6 h, at 180 °C, and at a rotating speed of 1200 rpm. The modified bitumen was compared with neat bitumen and bitumen processed under the above described conditions, which will be named as ‘processed bitumen’ hereafter. At a temperature of 180 °C, both the bitumen and polymer are in the melt phase and a dispersion of polymer drops is formed in the bitumen matrix.

The rheological characterisation of neat and modified bitumen was carried out using four different rheometers, namely, a controlled-strain Rheometrics Scientific ARES rheometer (USA), two controlled-stress Haake RS150 and RS100 rheometers (Germany) and the Cambridge MultiPass rheometer (MPR), a double piston capillary instrument developed by the Department of Chemical Engineering at University of Cambridge (Mackley et al. 1995).

Steady state flow curves at different temperatures and temperature sweeps (1 °C·min^{-1}) at constant shear rate (1 s^{-1}) were carried out with the RS100 rheometer, coupled to the heating system Haake TC501, and using a plate-and-plate geometry (20 and 35 mm diameter, 1 and 2 mm gap). Frequency sweeps, at different temperatures, within the linear viscoelasticity region were performed with the RS150 rheometer, using serrated plate-and-plate geometries (10 and 20 mm diameter, 1 and 2 mm gap). Steady state flow curves at different temperatures and temperature sweeps (1 °C·min^{-1}) in oscillatory mode (1 rad s^{-1} and 1% strain) were accomplished with the ARES rheometer, using a Couette geometry, having the inner and outer cylinders 32 and 34 mm diameter respectively, and a plate-and-plate geometry (25 and 50 mm diameter, 1 and 2 mm gap). The Cambridge MultiPass rheometer was used to perform steady shear experiments in ‘multipass’ mode, at different temperatures, using a 2 mm diameter and 40 mm length capillary, and two packed beds with spheres of different diameters (1 and 2 mm). At least two runs of each test were performed on each instrument.

Table 1 Physico-chemical characteristics of the bitumen and recycled polymer

Bitumen	
Asphaltene content (wt%)	20.00
Penetration grade (1/10 mm)	60–70
Recycled polymer	
EVA/LDPE	2/1
vinyl acetate (wt%)	5
Black carbon (wt.%)	1

DSC measurements were carried out with a TA Instruments Q100 (USA), using 10–20 mg of sample in hermetic aluminium pans, and a heating rate of 10 °C min^{-1}. The sample was purged with nitrogen at a flow rate of 50 cm^{3}min^{-1}.

Optical microscopy was used to study the morphology of the modified bitumen. A Cambridge Shear System 450, manufactured by Linkam Scientific Instruments (UK), coupled to a standard Olympus BH2 microscope was employed with that purpose, which allowed observation of the sample under shear at different temperatures.

Results and discussion

Linear viscoelasticity and flow behaviour

Figure 1 shows oscillatory shear curves for neat and modified bitumen at −10 and 50 °C. As can be observed, the elastic and viscous moduli increase as the temperature decreases, having an elastic modulus close to 10^8 Pa at −10 °C at high frequency. Polymer modification gives rise to a significant increase in the viscoelastic functions, mainly at 50 °C, temperature at which the curves of the linear viscoelasticity functions for modified bitumen show important differences in relation to those for neat and processed bitumen, particularly in the low frequency region. The viscous modulus shows higher values than the elastic modulus in the whole range of frequency, at 50 °C, for 5 wt% modified bitumen, neat and processed bitumen. The 9 wt% modified bitumen exhibits a quite different behaviour. Thus, G′ and G″ values are very similar within the frequency range studied and both curves present the same slope, 0.5 ± 0.001, showing a typical behaviour of a critical gel (De Rosa and Winter 1994).

At low temperature, the addition of polymer to the bitumen does not produce a significant change in the viscoelastic functions in relation to the neat or processed bitumen, although the polymer modification may reduce the bitumen stiffness at high enough frequencies. Thus, modified bitumen shows a decrease in the slope of G′ and, as a consequence, the glassy region of the mechanical spectrum of these materials shifts to higher frequencies or lower temperatures (Martínez-Boza et al. 2000). In addition, the difference between the neat and processed bitumen curves increase at −10 °C, which proves that oxidation undergone by processing appears more significantly at low temperatures. Oxidation is the phenomenon which takes place during the stirring of bitumen at the conditions described in the experimental section, in which bitumen undergoes changes in its chemical composition, in particular, a transformation of aromatics into resins and resins into asphaltenes. It is usually named as ‘primary ageing’, unlike the ‘secondary ageing’ which takes place during the bitumen life-time (Pérez-Lepe et al. 2003).

On the other hand, polymer modification makes the viscoelastic functions increase at intermediate in-service temperature, with the consequent benefit for the bitumen, improving ‘rutting’ resistance.

The flow behaviour of neat and modified bitumen at higher processing temperatures is shown in Fig. 2 and 3, for a wide range of shear rates, by combining rotational and capillary rheometry (Barnes et al. 1989; Macosko 1994; Walters 1975). Flow curves of neat bitumen were obtained, at different temperatures, from 10^{-2}to 10^4 s^{-1},

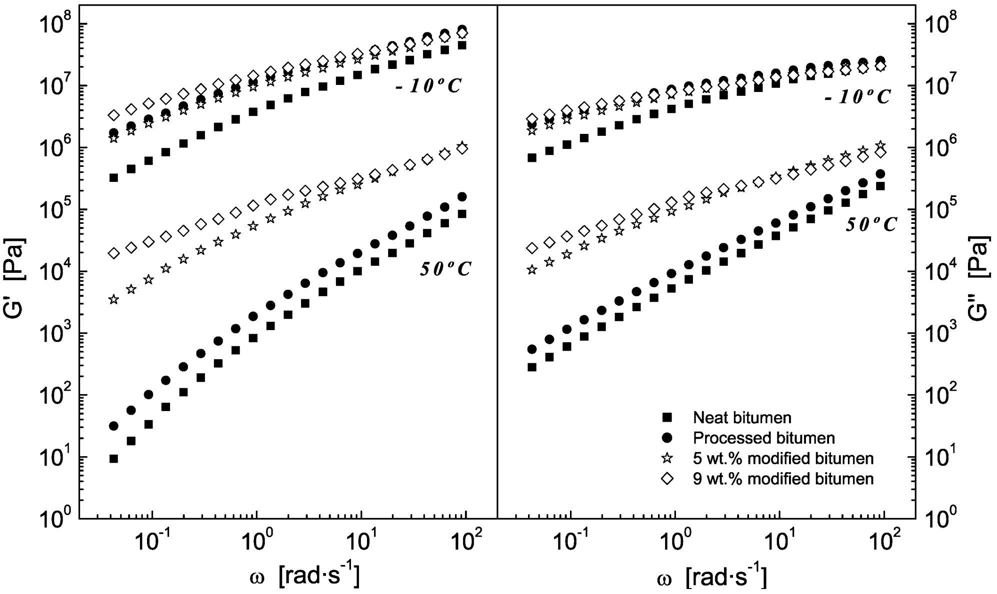


Fig. 1 Evolution of the linear viscoelasticity functions with frequency, at −10 and 50 °C, for neat, processed and polymer modified bitumen (5 and 9 wt%)

Fig. 2 Flow curves of neat bitumen, at different temperatures. Additional plots of MPR bottom piston position and pressure profiles

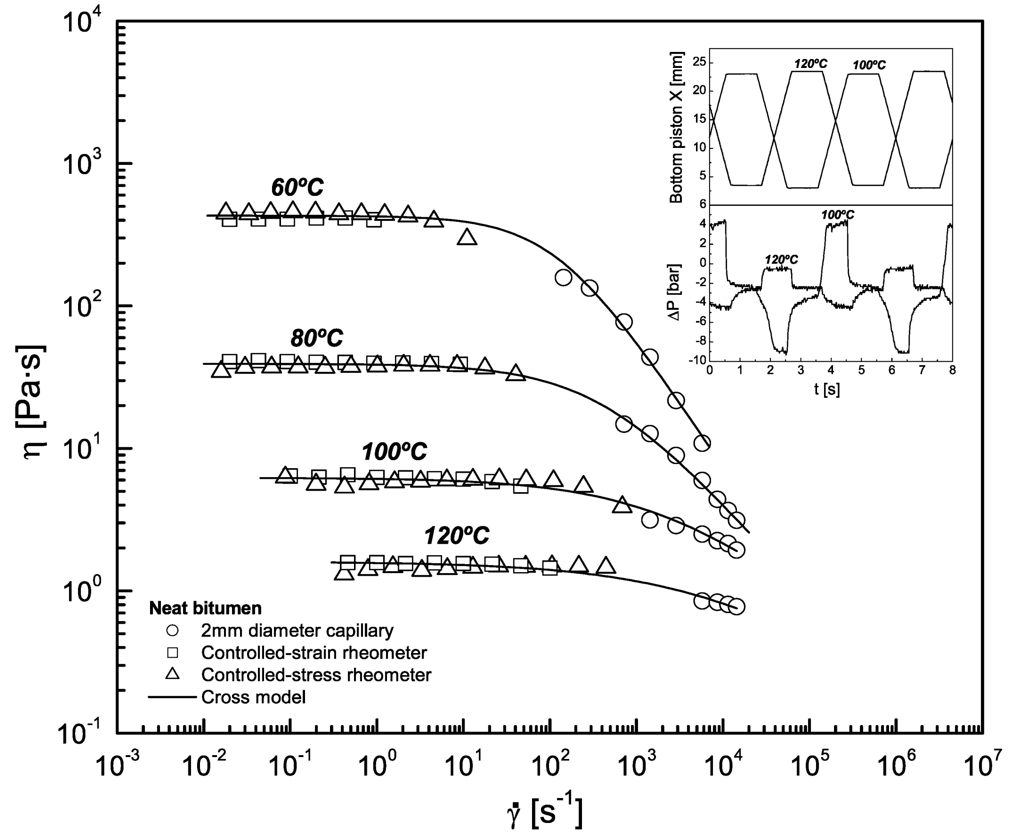


Fig. 3 Flow curves of 5 and 9 wt% polymer modified bitumen, at 100 and 180 °C respectively. Additional plots of the bottom piston position and pressure profiles

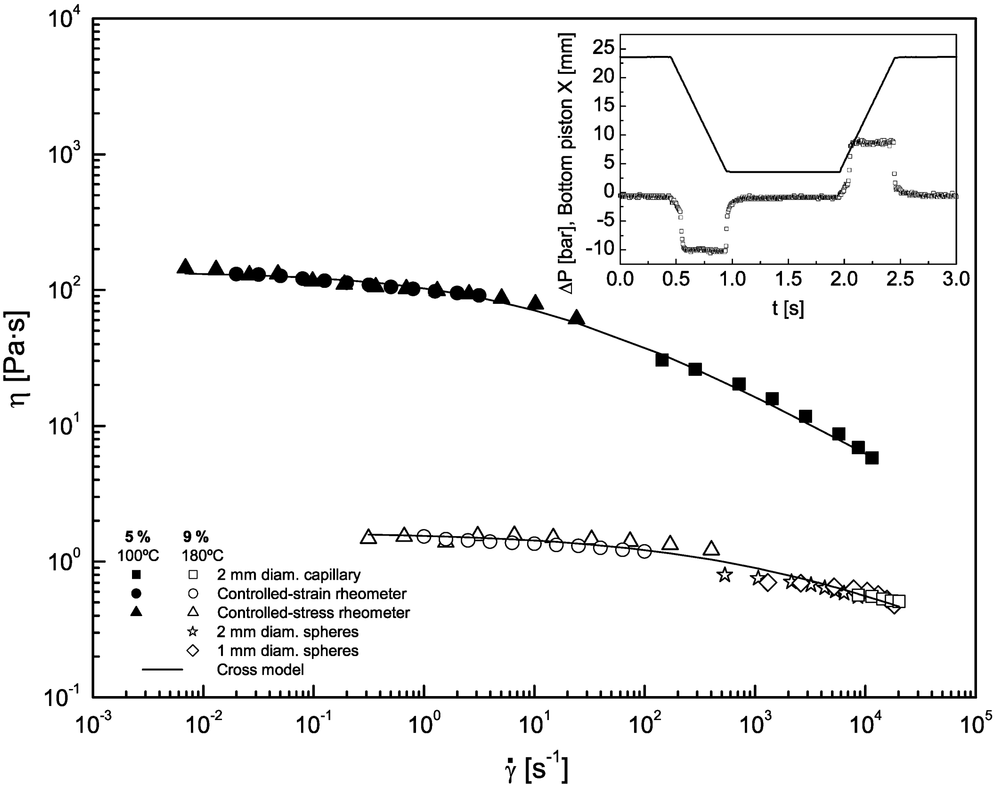


Table 2 Cross model parameters for neat and modified bitumen at different temperatures

	Neat bitumen				Modified bitumen	
	60 °C	80 °C	100 °C	120 °C	5 wt% 100 °C	9 wt% 180 °C
η_0[Pa·s]	430.26	39.29	6.22	1.60	136.66	1.65
λ [s]	8.13×10^{-3}	2.31×10^{-3}	3.60×10^{-4}	9.23×10^{-5}	8.54×10^{-2}	6.10×10^{-4}
p	0.920	0.694	0.500	0.416	0.450	0.370

using controlled-strain and controlled-stress rheometers, for low-medium shear rate values, and a capillary rheometer (MPR) in the high shear rate region (Fig. 2). The bitumen shows a shear-thinning behaviour, with a region corresponding to a Newtonian viscosity at low shear rate, η_0, and a further drop in viscosity for higher shear rates which became more apparent as the temperature was decreased. The flow behaviour shown by the neat bitumen was represented fairly well by the Cross model:

$$\frac{\eta-\eta_\infty}{\eta_0-\eta_\infty} = \frac{1}{1+(\lambda\cdot\dot{\gamma})^p} \qquad (1)$$

where η_0 (Newtonian viscosity), λ (characteristic time) and p (related to the slope in the shear-thinning region) increase as temperatures decreases. η_∞ was considered to be zero for the systems studied. Cross parameters for neat bitumen are presented in Table 2.

It can also be seen that Newtonian viscosity values get closer as the temperature increases. Consequently, the thermal susceptibility of the bitumen is reduced at high temperature, due probably to a simpler structure, as most of the resin stands dissolved into the maltenic matrix. The bitumen behaves as a shear-thinning fluid at high temperature in the high shear rate region, which may be caused by its colloidal structure, with asphaltene cores peptised by a shell of resin. According to the colloidal model, bitumen is a mixture of asphaltenes (black particles) dispersed in an oily medium (maltenes) composed of saturates, aromatics and resins. Part of the resins constitutes a shell around the asphaltene particles, and a temperature-dependent equilibrium between the resins around the asphaltenes and those dissolved in the oily matrix is established (Lesueur et al. 1996).

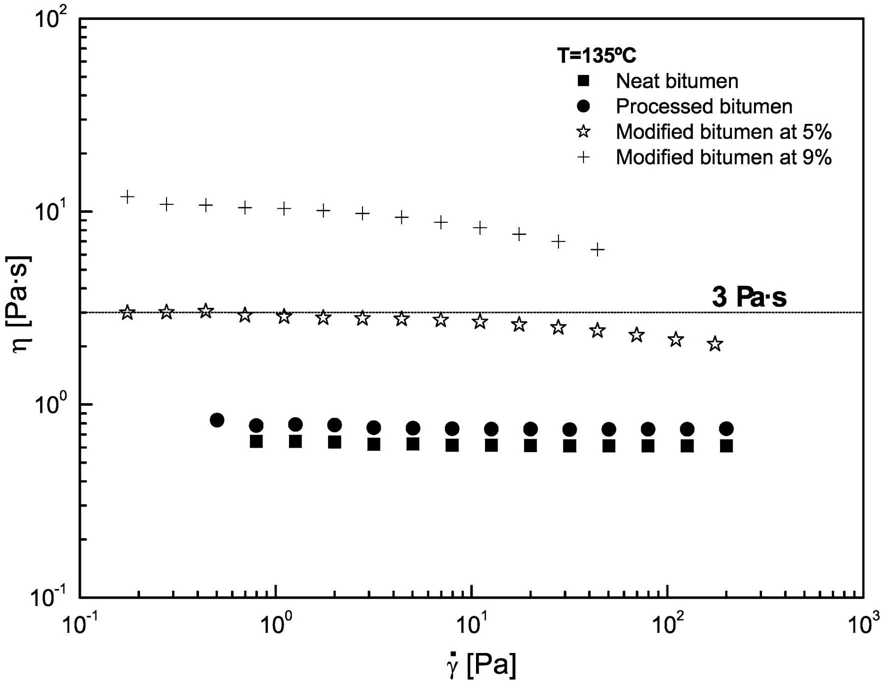
The overall flow behaviour cannot be found by using standard rotational rheometers with parallel plates alone and capillary rheometry, which is able to reach sufficiently high shear rates, should be used for that purpose. In the case of the MPR, two pistons operating in tandem were used to drive the fluid sample through a small capillary insert, and the pressure difference across the capillary measured. Bottom piston positions and differential pressure profiles are included in Fig. 2, for the neat bitumen at 100 and 120 °C and the same conditions of velocity (20 mm s^{-1}), amplitude (10 mm) and idle time (1 s) for the pistons, showing that pressure drop across the capillary becomes different from zero just in the period of time when the pistons move up or down. Moreover, differential pressure is higher at low temperature as the bitumen has a higher viscosity.

High temperature flow curves for modified bitumen, 5 and 9 wt%, in a wide range of shear rates, are shown in Fig. 3, for selected temperatures. Data sets for the 5 and 9 wt% modified bitumen were obtained by using a controlled-strain, a controlled-stress and a capillary rheometer. For the second bitumen, additional viscosity tests were performed by changing the 2 mm diameter capillary for two packed beds with steel spheres of two different diameters (1 and 2 mm) in order to obtain lower shear rates, close to those accomplished by rotational rheometers. The bitumen at high temperature was made to pass through the packed bed, and the pressure at its both sides was measured by pressure transducers (Machac et al. 1998; Chhabra et al. 2001). As can be observed in Fig. 3, the values obtained with the packed bed are in good agreement with those obtained by using a 2 mm capillary. The bottom piston position and pressure differences profiles have been inserted in Fig. 3, for an experience using 2 mm diameter spheres, with 9 wt% modified bitumen.

As for the neat bitumen, the flow curves present a Newtonian region at low-medium shear rates, followed by a drop in viscosity as the shear rate increases, and, therefore, the flow behaviour can be represented by the Cross model (Eq. 1). The values of the parameters η_0, λ and p are shown in Table 2. The effect of polymer addition at high temperature is clearly observed by comparing neat bitumen and 5 wt% modified bitumen at 100 °C (Figs. 2 and 3). Thus, Table 2 shows that the addition of 5 wt% EVA/LDPE increases η_0 one order of magnitude. Furthermore, higher values of λ are obtained and, as a consequence, the material becomes more structured by the polymer addition. On the other hand, it should be noticed that 9 wt% modified bitumen has a value of the low-shear-rate viscosity, at 180 °C, quite similar to that shown by the neat bitumen at 120 °C. However, the modified bitumen is much more structured, as may be deduced from the values of the characteristic time (Table 2).

It is well-known that the viscosity of a modified bitumen should be controlled due to its strong effect on bitumen workability and handling. In that sense, the flow curves of neat, processed and modified bitumen, at 135 °C, are compared in Fig. 4. As was previously pointed out for oscillatory shear experiments, the differences between neat and processed bitumen are smaller

Fig. 4 Flow curves, at 135 °C, of neat, processed and polymer modified bitumen (5 and 9 wt%)



as the temperature increases, showing that the oxidation effects appear more clearly in the low and intermediate temperature region. Figure 1 demonstrates that polymer modification improves the mechanical properties of bitumen at both low and medium in-service temperatures, with an increase in the resistance to permanent deformation and thermal cracking. However, viscosity should be kept low enough at higher handling temperatures of bitumen (Crockford et al. 1995) (laydown and compaction temperatures) in order to allow an easy application on the pavement. AASHTO MP1 (McGennis 1995) requires that the viscosity at 135 °C should be less that 3 Pa s. As can be seen in Fig. 4, a 5 wt% modified bitumen stands below the above-mentioned limit and, thus, it may be properly applied in pavement building. Blends at 9 wt% exceed 3 Pa s and thus, the pumping operations performed during the binder application on the road might result quite difficult. However, blends at 9 wt% might be successfully employed in the manufacture of roofing membranes, which demand higher proportions of polymers (Fawcett and McNally 2000). As a consequence, rheology and microstructure of modified bitumen at 9 wt% are widely studied in this paper.

Modified bitumen microstructure under shear

Figure 5 shows the results obtained during two temperature sweep tests, performed on 9 wt% polymer modified bitumen, carried out in oscillatory and steady-shear mode at the conditions mentioned in the experimental section. The frequency sweep test shows the behaviour of the modified bitumen at very low strain, that is, as the system stands nearly at rest and no orientation effects take place on the dispersed polymer (inset in Fig. 5). Temperature is the only parameter causing a decrease in complex viscosity. In addition a significant drop in viscosity occurs at about 105–110 °C. In order to explain this fact, standard calorimetry on neat bitumen, 9 wt% modified bitumen and recycled EVA were accomplished in a wide range of temperatures (Fig. 6). Two melting peaks appeared during the experiment carried out with the recycled polymer, a larger one at 109 °C and another one, much smaller, at 122 °C. As shown in Table 1, the recycled polymer is a blend of EVA copolymer with a low content in vinyl acetate (peak at 109 °C) and a minor proportion of LDPE (peak at 122 °C). As can be seen, a small peak at about 109 °C, corresponding to polymer melting, appears for the 9 wt% modified bitumen. It is worth pointing out that other experiments carried out on a similar blend of bitumen and polymer (García-Morales et al. 2004a, 2004b) not only gave rise to a larger peak, but it was also shifted to a lower temperature, a fact related to the polymer fraction swollen by bitumen light components.

If a significant shear is applied to the modified bitumen, substantial structural changes are observed in a shear rate sweep experiment (Fig. 5). Very long streaks of polymer orientated in the flow direction, which do not recover their original form after shear, appear at the lowest temperatures (see Fig. 5), close to the melting point of the swollen polymer phase. At higher

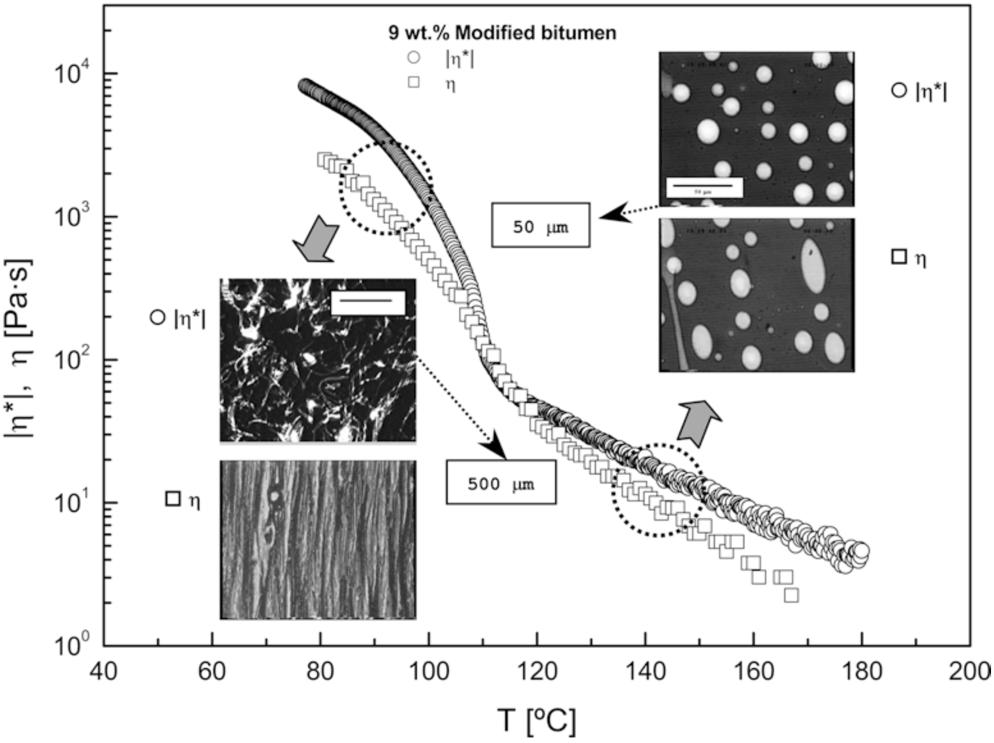


Fig. 5 Evolution of viscosity ($1\ s^{-1}$) and complex viscosity ($1\ rad\ s^{-1}$) with temperature for 9 wt% polymer modified bitumen. Additional photomicrographs of the polymer modified bitumen microstructure for different shear conditions

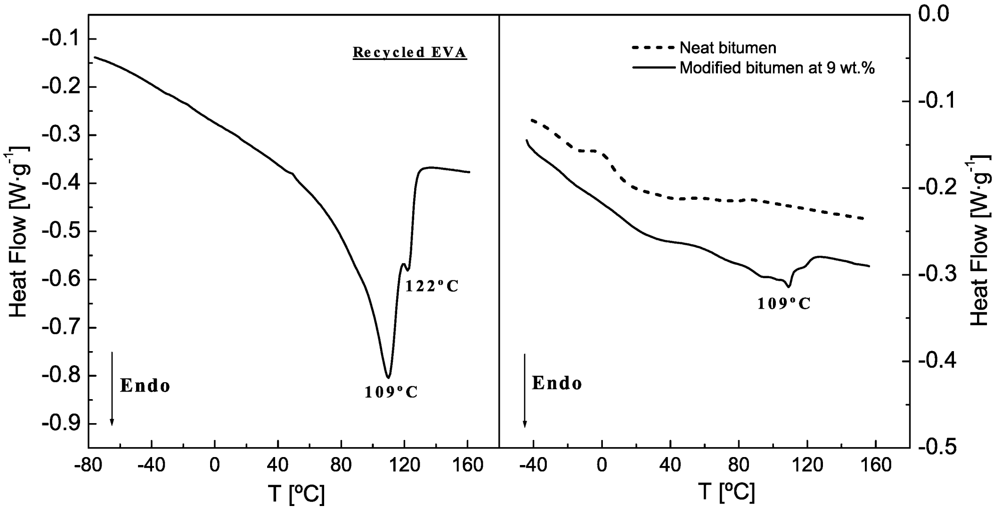


Fig. 6 DSC curves for recycled EVA/LDPE, neat bitumen and polymer modified bitumen (9 wt%)

temperatures and moderate shear, the droplets of polymer, which remained in a spherical shape during the oscillatory mode, tend to slightly deform in the flow direction, adopting ellipsoid shapes which are transformed into spherical droplets after shear stops (inset in Fig. 5). As a consequence of the structural changes in polymer droplets, the resistance to the flow diminishes and lower values in viscosity are obtained, at the same temperature, in steady flow.

The striking decrease in complex viscosity observed above 90 °C can be related to the melting process of the swollen polymer phase. In addition, during steady flow at temperatures close to the polymer melting point, shear makes the polymer drops extend into very long streaks. Thus, the viscosity decreases due to polymer elongation. At higher temperatures and moderate strain rate, the polymer droplets lengthen into ellipsoid particles and recover their spherical shape after shearing (Luciani et al. 1997).

The effect of shear on polymer droplets has been confirmed by other experiments that combine shear flow and sample relaxation. Thus, Fig. 7 illustrates the results

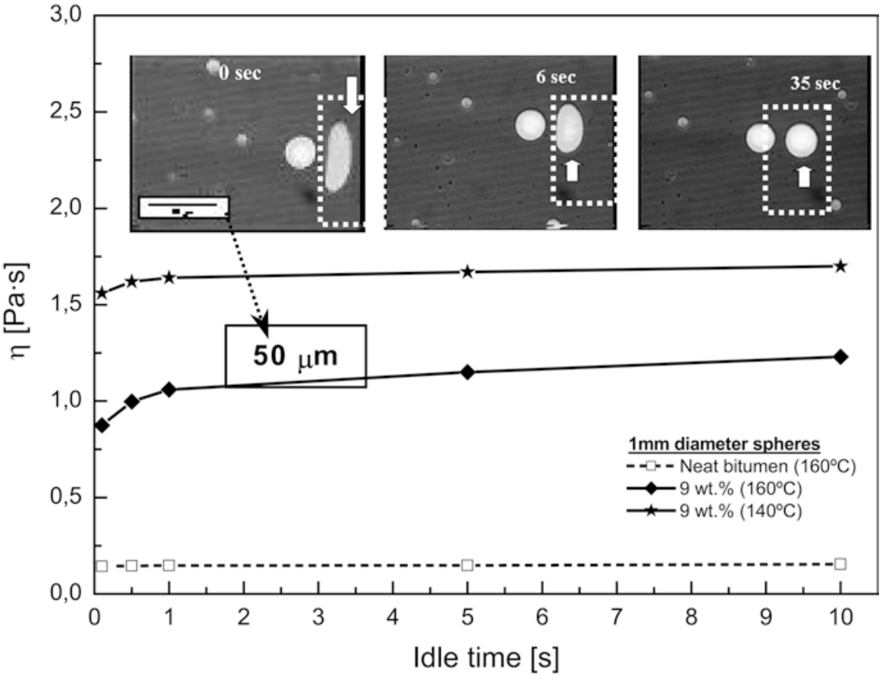


Fig. 7 Evolution of viscosity with pistons idle time, corresponding to MPR experiments, for neat bitumen (160 °C) and 9 wt% polymer modified bitumen, at 140 and 160 °C respectively. Additional photomicrographs of the droplet relaxation process

of an experiment using the MPR rheometer and a packed bed with spheres, aiming to show the way that polymer droplet relaxation may affect the viscosity of the modified bitumen. Pressure drop of bitumen across the packed bed was measured for a group of experiments in which the idle time of pistons between their up and down movements is increased. Although polymer droplets deform passing through the packed bed, the relaxation process may make them recover their original spherical shape if a long enough idle time is considered. As has already been mentioned, spherical droplets give rise to higher viscosity than elongated drops which easily orientate towards the flow direction. Hence, for short idle times, which do not allow polymer drops to take their initial shape back, the viscosity of the modified bitumen is lower. Once the polymer particle has thoroughly returned to its original state, the viscosity value keeps constant although experiments at higher idle times are considered. Plots of viscosity vs idle time, as a function of temperature, are shown in Fig. 7 for experiments carried out, with 1 mm diameter spheres and the same piston velocity (60 mm s^{-1}), on 9 wt% modified bitumen and neat bitumen. It can be seen that an increase in viscosity occurs for modified bitumen as longer idle times are considered. For the case of neat bitumen the viscosity value remains constant for experiments at a fixed piston velocity.

Some photomicrographs showing the relaxation process of a polymer droplet in the modified bitumen have been included in Fig. 7. Previously, shear was applied on the modified bitumen in order to produce droplet deformation. Then shear was stopped and the evolution of the shape for a selected droplet (marked by a short-dashed line) was followed along time. It is worth mentioning that the relaxation time is not the same for all the droplets, but the velocity at which the phenomenon takes place depends on the ratio σ/r_0, where σ is the interfacial tension and r_0 the radius of the droplet (Luciani et al. 1997; Wolf et al. 2000). Hence, for a considered temperature, shorter relaxation times are obtained for the smallest polymer particles.

Conclusions

Polymer modification using recycled EVA/LDPE improves the mechanical properties of bitumen in the temperature region where the material is used as a surface coating. In relation to processing, the 5 wt% modified bitumen has viscosity values, at 135 °C, that allow easy application on the road. On the contrary, the viscosity of the 9 wt% modified bitumen is too large for processing at that temperature. Neat bitumen has normally been considered to be a Newtonian fluid at high temperature. However, a drop in viscosity takes place when experiments are performed at very high shear rate, which were obtained by using capillary rheometry. For a modified bitumen, a significant decrease in the complex viscosity with increasing temperature was observed in oscillatory shear and this was related to the melting process of the swollen polymer phase. Such a decrease in viscosity is less evident when steady shear is applied on the sample, because the modified bitumen microstructure is strongly affected by shear.

Surprisingly, we have found that the bitumen-polymer system is very well suited for optical observation during shear. Providing the optical depth of the bitumen is sufficiently small, good contrast between the "dark" bitumen and the "transparent" polymer can be achieved. This in turn enabled detailed studies to be made on polymer droplet extension and relaxation in an optical shear cell.

The addition of recycled EVA/LDPE to bitumen enhances the materials mechanical properties, as well as providing a useful environmental alternative for the disposal of waste plastics.

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