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Influence of polymer modification on asphalt binder dynamic and steady flow viscosities

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HIGHLIGHTS

- Oscillatory analysis and steady state viscosity measurements were performed on PMBs.
- Polymer nature and content strongly influence PMBs rheological properties.
- The applicability of the Cox–Merz rule for modified bitumens was evaluated.
- Cross and Carreau models were suitable for low polymer modified bitumen.

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ABSTRACT

Asphalt pavement performance such as rutting, crack initiation and propagation as well as fatigue behaviour are substantially affected by the rheological properties of the bitumen. In this sense, the use of polymer modification in road paving applications has been growing rapidly over the last decade as it allows significant enhancements in bitumen properties with consequent improvement in road service life. In fact, the use of polymer modified bitumens (PMBs) leads to pavements characterized by higher resistance to rutting and thermal cracking and lower fatigue damage, stripping and thermal susceptibility. This paper presents a laboratory investigation concerning the effect of polymer modification on the flow behaviour of bitumens. Two different polymers, an elastomer and a plastomer, were used as bitumen modifying agents at three different percentages (2%, 4% and 6% by bitumen weight). Oscillatory mechanical analysis as well as viscosity measurements under steady state conditions were performed taking into account different testing parameters such as temperature, loading frequency and shear rate. The results confirm that the rheological properties of PMBs are strongly influenced by polymer nature and polymer content. The bitumen viscosity on the dynamic domain was combined with that in the steady-state domain, confirming the applicability of the Cox–Merz relationship for the plain bitumen and the PMBs with low polymer content. Finally, the Cross and the Carreau models were found to be suitable to fit the steady state and the dynamic results in order to determine the viscosity function of the investigated bitumens.

1. Introduction

It has been well established that the rheological properties of the bitumen substantially affect the asphalt pavement performance [1]. Since bitumens for road paving applications experience a variety of thermo-mechanical states during their service life, it results extremely important to investigate their rheological properties under different temperature and loading conditions. Most pavement distresses, such as rutting at high temperatures and crack initiation and propagation at low temperatures, can be attributed not only to traffic loads but also to the thermal susceptibility of asphalt binders [2,3]. In order to improve asphalt mixture performance, the bitumen properties are often enhanced by means of polymer modification. Polymers are traditionally used to decrease the temperature susceptibility of the bitumen by increasing its stiffness at high service temperatures as well as reducing its stiffness at low service temperatures [4,5]. This leads to enhanced pavements having higher resistance to rutting and thermal cracking and lower fatigue damage, stripping and thermal susceptibility [6,7].

The polymers that are commonly used for bitumen modification can be divided into two main categories: elastomers and plastomers. Elastomers are characterized by high elastic response having

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the ability of resisting permanent deformations by stretching and recovering their initial shape [4], whereas plastomers contribute to form a tough, three-dimensional network to resist deformation [4] when blended with asphalt binder.

The influence of polymer modification on bitumen properties can be evaluated through the investigation of rheological properties such as viscosity, complex modulus and phase angle that are highly influenced by the presence of a polymer network in the bituminous matrix. In particular, viscosity affects mixing, laying and compaction characteristics of asphalt mixtures as well as pavement performance and can be evaluated in steady state conditions by means of Brookfield viscosimeter or Dynamic Shear Rheometer (DSR). On the other hand, complex modulus and phase angle, that were measured by applying sinusoidal loads at various loading frequency and temperature ranges, allow a proper evaluation of the visco-elastic properties of the bitumens. The oscillatory measurements can be further employed to obtain the complex viscosity and the correlation between the dynamic and the steady state condition can be defined [8] through the Cox–Merz relationship [2,8,9]. The Cox–Merz relationship is empirical and establishes a correspondence between the steady-state viscosity at a specific shear rate ($\dot{\gamma}$) and the magnitude of the complex viscosity at an angular frequency (ω) equal to the considered shear rate, as shown in Eq. (1):

$$|\eta^*(\omega)| = \eta(\dot{\gamma})|_{\omega=\dot{\gamma}} \tag{1}$$

This paper presents a laboratory investigation concerning the effect of polymer modification on the flow behaviour of a plain bitumen. Two different polymers were used as bitumen modifying agents at different percentages, in order to evaluate the effect of both polymer type and content on the rheological characteristics of asphalt binders. The effect of polymer modification was evaluated through oscillatory mechanical (i.e. dynamic) analysis and viscosity measurements under steady state conditions.

2. Materials

A 70/100 penetration-grade bitumen from an Italian oil refinery was selected as plain bitumen.

Two polymers, an elastomer and a plastomer, were selected as modifying agents for the production of different modified bitumens (PMBs). A radial styrene–butadiene–styrene (SBS) polymer, which contains 30% styrene and has a density of 0.94 g/cm^3, and a polyolefin (PO) polymer, having a density of 0.94 g/cm^3, were added to the plain bitumen at three different percentages (2%, 4% and 6% by bitumen weight) with the aim to investigate the effect of polymer type as well as polymer content on the rheological properties of the plain bitumen. Table 1 shows the physical properties of the plain bitumen and the polymer modified bitumens.

All modified bitumens were produced in the laboratory using a ROSS high-shear mixer, operating at a rotation speed of 3000 rpm at 180 °C. Initially, 700 g of bitumen contained in a 1000 ml cylindrical can were heated to fluid conditions. Up on reaching 180 °C, each type of polymer was added slowly to the bitumen in order to prevent any polymer aggregation during the mixing process. Subsequently, dicumyl peroxide in granules was added as cross-linker dosed at 0.3% by polymer weight. Mixing was then continued at 180 °C for 3 h. After mixing, the sample was removed from the can, poured into small containers, cooled to room temperature, cover with an aluminium foil and stored for not more than 7 days. Before testing, the stored material was heated once in order to produce testing specimens.

Table 1
Physical properties of the plain bitumen and the polymer modified bitumens.

Materials	Penetration @25 °C 0.1 mm (EN 1426)	Softening point °C (EN 1427)	Dynamic viscosity @135 °C Pa∗s (EN 12595)
Plain	72	47.7	0.31
PMB_SBS2	58	50.4	0.55
PMB_SBS4	54	69.5	0.91
PMB_SBS6	44	94.6	1.40
PMB_PO2	64	52.4	0.54
PMB_PO4	53	55.8	0.78
PMB_PO6	41	96.6	1.00

3. Test program and procedures

Rheological properties of plain bitumen and polymer modified bitumens were studied according to the experimental program shown in Table 2.

In a preliminary phase, complex modulus G^* and phase angle δ were investigated with a Dynamic Shear Rheometer (DSR), performing frequency sweep tests over a range from 1 to 100 rad/s under isothermal conditions, in the temperature range from 4 to 82 °C with step of 6 °C. A plate–plate geometry was adopted with a plate diameter of 8 mm and a gap equal to 2 mm, from 4 to 34 °C and a plate diameter of 25 mm and a gap equal to 1 mm, from 34 to 82 °C. Frequency sweep tests were conducted in control strain within the linear viscoelastic range of the materials by applying a strain amplitude of 0.5%.

In the second phase, the flow behaviour was evaluated in terms of dynamic viscosity measurements, in order to investigate the influence of polymer modification on both temperature susceptibility and shear rate dependency. In particular, bitumen viscosity η was measured in two different test configurations:

(1) Coaxial cylinders with Brookfield device from 90 to 180 °C and shear rate of 10 s^{-1}.
(2) Steady state with DSR, over a shear rate range from 0.001 to 1000 s^{-1}, at a temperature of 58 °C.

Moreover, the complex viscosity η^* was determined from the frequency sweep tests performed over a wide range of temperatures (4–120 °C). Specially, it was calculated as the ratio between the norm of the complex modulus and the corresponding angular frequency, according to Eq. (2):

$$|\eta^*(\omega)| = \frac{|G^*|}{\omega} \tag{2}$$

Different devices and test configurations were used to investigate viscosity with the objective of extending the range of tested temperatures and loading frequencies as well as comparing the viscosity measured under rotational and oscillatory conditions. In fact, the rheological properties evaluated under dynamic conditions can be related to those measured under steady state conditions within specific ranges of shear rate and frequency, according to the Cox–Merz relationship (Eq. (1)).

4. Results and analysis

4.1. Preliminary dynamic characterization

The isochronal plots of complex shear modulus G^* vs. temperature at a loading frequency of 1 rad/s are shown in Fig. 1 for all the studied materials. As expected, as the amount of polymer content increases, modified bitumens show an increase in G^* at all temperatures and a slight decrease in temperature susceptibility for temperatures equal or higher than 40 °C with respect to the plain bitumen, mainly for the highest polymer content. In particular, PMB_PO6 shows a sharp decrease in the slope of the complex modulus isochronal at high temperatures with the establishment of a plateau region which is indicative of a dominant polymer network [4,10]. Moreover, at low temperatures the effect of polymer results not very evident regardless of polymer type and content because of the high stiffness of the plain bitumen that does not allow the polymer network to influence noticeably the rheological properties of the bitumen [4].