Petroleum and Coal

Article **Open Access**

ANALYSIS OF RHEOLOGICAL PROPERTIES OF BITUMEN IN RELATION TO PHYSICAL PROPERTIES

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Received December 18, 2018; Accepted February 3, 2019

Abstract

The bituminous binders used in road constructions are characterized by empirical tests as penetration, softening point and breaking point without characterizing the elastic-plastic response. This now appears to be inadequate mainly in the case of polymer modified bitumen. The objective of this work was to investigate the rheological properties of non-modified and polymer modified bitumen binders in dynamic shear rheometer by complex shear modulus and phase angle. The complex shear modulus values of non-modified bitumen are lower than the polymer modified bitumen at higher temperatures throughout the frequency range. The phase angle values of non-modified bitumen are significantly higher over the temperature and frequency test range. This is caused by lower elasticity compared to polymer-modified bitumen. Comparison of penetration results with complex shear modulus at 25°C showed that the bitumen is harder, the penetration is lower, and the complex shear modulus is higher.

Keywords: *bitumen binder; rheology; complex modulus; phase angle; empirical properties.*

1. Introduction

Bitumen is one of the basic building materials, which is mainly used in road construction. Research works on bituminous binders, and asphalt mixtures were based on the assumption that bitumen can be characterized and sorted by empirical tests such as softening point, penetration or break point. These tests determine the stiffness and consistency of the binder without characterizing the elastic-plastic response. Especially in the case of polymers modified bitumen [1], these tests cannot adequately describe the complex rheological behavior of bitumen binders.

The development of the use of bitumen binders in road construction also leads to the development of new test methods that best describe their physical properties. One of the most important features is the behavior of bituminous binders at low temperatures, where there is a risk of breakage of the road with frost cracks. Further detection of high-temperature resistance where there is a risk of permanent deformations. This corresponds to the high-temperature sensitivity of the bituminous binder, which can be described by the viscosity dependence on the temperature as noted in [2-5]. Last but not least, it is important to evaluate the fatigue properties of the material or to characterize the resistance to loss of cohesion. The development of new testing methods according to [6], functional tests, have focused on characterizing the visco-elastic behavior of bitumen to better prediction of binders behaviour in asphalt mixtures, especially in critical conditions of failure - rutting, low temperature cracking and fatigue cracking in asphalt pavement roads. These methods include characterization in the dynamic shear rheometer DSR, the rotary viscometer or the bending beam rheometer BBR.

2. Bitumen rheology

At present, the most commonly used method of fundamental rheological testing of bitumen properties is dynamic mechanical analysis usually by means oscillatory-type testing. Rheological tests, compared to classical empirical tests, allow the properties of materials to be evaluated against loads, temperature intervals, frequencies and time at different loads. It allows to investigate the dynamic development of materials over time and to model the deformation properties of substances that are influenced by external forces. The basic methods of rheological measurement can be divided according to the load mode as rotation tests, oscillatory tests, creep tests, relaxation tests.

Bitumen binders behave in part as an elastic material (the deformation after loading is reversible, at a very low temperature) and partly as a viscous fluid (deformation due to load is irreversible, at high temperature). The dynamic mechanical analysis (DMA) of rheological properties allows characterization of the viscous and elastic behavior of bitumen binder at service temperatures. The sample of the binder is located between the two plates, the bottom plate is firm, and the upper one oscillates over the sample and creates shear stress. DMA is performed on a fresh binder, bitumen after short-term aging RTFOT and bitumen after long-term aging PAV using dynamic shear rheometer (DSR). The DSR test allows quantifying both elastic and viscous properties. This enables the bitumen binder to be characterized in the temperature range during the road pavement using in service.

At the University of Žilina, the rheological properties are measured on the Physica MCR 301 oscillatory rheometer with convection heating device CTD 450 by Frequency Sweep (FS) test method. FS test method uses a parallel plate system (PP), a set of two parallel plates, the bottom plate is stationary, upper is shear, performing the oscillatory motion. The distance between the plates (shear interval) h is exactly defined (Figure 1). The FS test is performed at a constant temperature, and for the viscoelastic materials, it is controlled by shear strain in the form of a sinusoidal oscillation function:

$$\gamma(t) = \gamma_A . sin\omega t \qquad (1)$$

Testing the viscoelastic materials, the resulting stress and presetting the strain are not in phase, the strain delays behind the stress by a phase angle. If the oscillatory shear is sinusoidal, following [7-8] the shear stress τ is expressed:

$$\tau(t) = \tau_A . \sin(\omega t + \delta) = \tau_A . (cos\omega t + i . sin\omega t) \qquad (2)$$

where τ_A is the stress amplitude (Pa), ω is the angular frequency (rad/s), $\omega = 2\pi f$, t is the time (s) and δ is the phase angle of the measured material response specified in degrees.

The complex shear modulus $G*$ expressing the resistance of the bitumen binder to deformation when subjected to repeated stress [7], is calculated using the equations:

$$G^* = \frac{\tau(t)}{\gamma(t)} \qquad (3)$$

Parameters resulting from oscillation processes are written in the complex form:

$$|G^*| = \sqrt{(G')^2 + (G'')^2} \qquad (4)$$

The storage modulus G' represents deformation energy stored by the sample. After unloading the stored energy acts reversible deformation process and the material behaves elastically. The loss modulus G'' is the amount of deformation energy consumed by the sample during the shear process. A part of this energy heats the sample, there are frictional forces between the molecules, frictional heat occurs, and the residue is released as heat to the environment. The sample with the high loss modulus exhibits irreversible deformation and indication of the relative size of the reversible and irreversible deformation and the expression of the displacement or the delay between the applied shear load and the resultant shear conversion. The complex shear modulus or resistance to deformation of bitumen increases with the addition of polymers [8]. The character of the cross-linked polymers (elastomer example used in polymer modified bitumen) is more or less flexible, soft or relatively rigid solid.

The values of parameter $G*$ and δ are dependent on temperature and load frequency of bitumen binder. At high temperatures, the binders behave as viscous liquids without the ability to return to their original shape. In this case, the viscous component prevails, with the phase angle *δ* approaching 90°. At low temperatures, bitumen behaves as elastic materials that return from deformation completely. This property is represented by the elastic component, and the phase angle δ is approaching 0°.

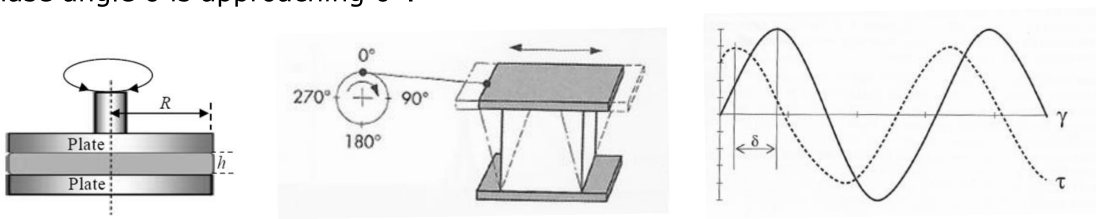

Figure 1. FS test [7]

A bitumen is considered as a liquid with high viscosity. Highly viscous materials usually have a higher temperature dependence compared to low-viscosity fluids. When heating non-crystalizing materials, a transition takes place from a rigid, solid consistency with *G´ > G″.* In this case, the glass transition temperature is reached at the maximum of the *G″* curve and the melting temperature occurs at the intersection of curves of *G´* and *G″* [7]. The temperature susceptibility of binder decreases significantly with the polymer modification which makes binder more suitable to use in extreme climatic regions as mentioned in [9] and [10].

3. Experimental program

The scope of the research study was to evaluate the rheology properties of various bitumen binders in relation to physical properties, in terms of showing a better behavioral characterization as well as differences between unmodified and modified bitumen. Bitumen modified by the addition of macromolecular substances such as synthetic polymers, change their thermo-viscous and elasto-viscous properties. Polymers reduce the temperature sensitivity of bitumen (harder at high summer temperatures, reduce brittleness at low temperatures), extend the plasticity range to 80°C, increase elasticity, aging resistance, extend service life. The qualitative level is related to the type of modifier and its quantity in bitumen (usually 2 to 12 %), the type of bitumen and its chemical composition, the technological process especially the way of homogenization.

The experimental study covered paving grade bitumen of 50/70 and 35/50 gradation and polymer modified bitumen PMB 45/80-75 and PMB 25/55-60. The purpose of the research related to analyzing of rheological properties in relation to empirical properties was divided into two stages. First, the physical properties of bitumen binders, that are the most frequent indicators of their behavior in practice were determined by empirical tests including the needle penetration at 25°C according to EN 1426 and a softening point (the Ring and Ball Test) according to EN 1427. The measured results of the samples are shown in the following Table 1.

Table 1. Bitumen physical properties

	Penetration at 25°C [0.1mm]	Softening point [°C]	Dynamic viscosity at 135°C [mPa.s]	Dynamic viscosity at 165°C [mPa.s]
35/50;A11	44.8	56.2	855.0	203.75
50/70;A1	64.1	49.8	470.44	118.22
50/70;A4	61.0	48.0	516.31	126.3
50/70;A7	54.3	51.0	500.08	128.31
PMB 45/80-75;M2	54.6	80.8	2236.0	553.75
PMB 45/80-75;M3	74.3	75.4	1572.5	383.75
PMB 45/80-75;M4	60.5	78.4	1825.0	417.5
PMB 25/55-60;M11	35.8	62.0	2725.0	585.0

Second, the rheological properties were determined by dynamic-mechanical analysis according to EN 14770. Using the dynamic shear rheometer (Figure 2), the values of the complex shear modulus and the phase angle were determined under the selected test conditions:

Mode of loading: controlled-strain, amplitude γ_A = 5%

Temperatures: range 25 to 80°C

Frequencies: range 0,1 to 10 Hz.

Bitumen binder sample is sandwiched between two circular parallel plates (PP system) with 25 mm diameter and distant from each other to a thickness of 1 mm. The lower plate is fixed while upper plate oscillates back and forth across the sample at a different frequency to create shearing action. This measuring method [7] enables simultaneous monitoring of rheological parameters *G′*, *G″* and η* in the chosen interval of angular frequencies.

Figure 2. Dynamic shear rheometer Physica MCR301 (left), SW for the test and results (right)

4. Results and analysis

The method of "FS test" allows observing the rheological parameters (η*, *G′, G″*) depending on the angular frequency ω at a constant temperature. If the intersection of the curves *G′, G″* can be observed, the molecular weight changes (length and branching of hydrocarbon chains) and degradation of the material can be assessed. The test data record at a certain temperature and for each angular frequency contains values of parameters storage modulus, loss modulus, complex viscosity, shear stress, shear rate, and torque.

The rheological data for the tested bitumen are presented as standard in the form of master curves of complex modulus *G** and phase angle δ at a reference temperature. Master curves represent the viscoelastic behavior of bitumen binder at a given temperature for a range of frequencies, bigger than that tested. It is based on the principle of the time-temperature superposition system. The experimental curves can be superimposed by shifting all curves to one selected as the base curve [7]. Temperature dependence of this shift is expressed by the function $\log a_T(T)$. Temperature shift factor a_T can be expressed by any standard method. In this study the Williams-Landel-Ferry equation according to [11] and [12] was used:

$$\log a_T = \frac{-C_1(T-T_{ref})}{C_2+(T-T_{ref})} \qquad (5)$$

where *T* is temperature, T_{ref} is the reference temperature, C_1 and C_2 are taken as constants.

This method is found in [12] to be applicable to bitumens. The results of tested bitumen are shown in Figure 3 and 4.

Samples of the same grade bitumen exhibit similar values of the complex modulus as seen in both non-modified and modified bitumen binders. At all test temperatures in the observed range of angular frequencies, the highest values of the complex shear modulus were determined in the case of harder bitumen, namely samples PMB 25/55-60; M11 and 35/50; A11; and the lowest values 50/70 paving grade bitumen samples. At higher test temperatures (e.g., 70 and 80°C), the shear modulus values of 35/50 bitumen get closer to the values of PMB 45/80-75 bitumen samples.

At higher load frequencies and pavement service temperature, the shear modulus values of paving grade bitumen are higher than PMB 45/80-75 bitumen samples. From Figure 3 and

Figure 4 it can be noted that the change in bitumen stiffness with a change of loading frequency is more markedly in the case of paving grade bitumen compared to the modified bitumen. That is also confirmed by the values of the loss and storage modulus. The values of the loss modulus of non-modified bitumen are significantly higher than the storage modulus at all tested frequencies and temperatures. The polymer component in modified bitumen, mainly at higher temperatures, increases the storage modulus, acts the reversible deformation process and the material behaves elastic.

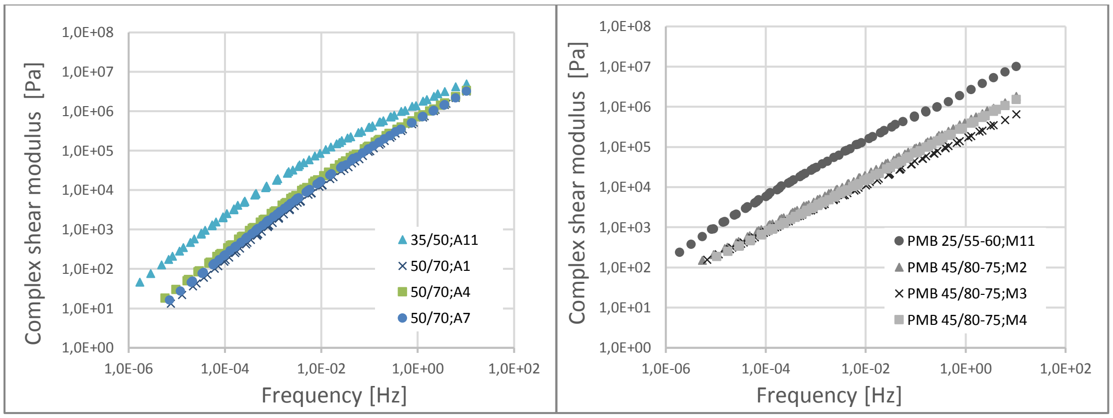

Figure 3. Complex modulus of non-modified (left) and modified (right) bitumen samples at 25°C reference temperature

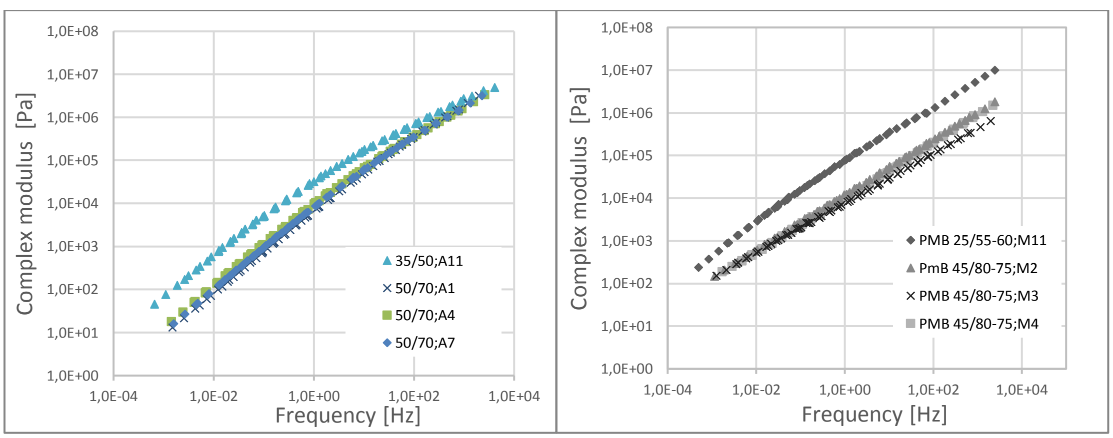

Figure 4. Complex modulus of non-modified (left) and modified (right) bitumen samples at 50°C reference temperature

Through the master curves, it has been shown that the same grade bitumen (paving grade and modified bitumen) have approximately the same values of the complex shear modulus across the frequency range. The master curves of the paving grade bitumen modulus are parallel. Lower penetration bitumen have higher values of the complex shear modulus; the master curve is shifted to higher values. Higher values of the complex modulus indicate that the binder has greater resistance to deformations at repeated loads.

The phase angle δ determines the time lag between the induced shear strain and the required shear stress in a controlled strain test. Phase angle δ can be described as an indicator of the relative magnitude of reversible and irreversible deformation. By comparing the values of phase angle (Figure 5), it can be stated that the phase angle of paving grade bitumen is in the range of 70° to 90°, while for modified bitumen it is from 50° to 70°. The phase angle of modified bitumen is significantly lower; the bitumen behaves elastic because of the polymer component in the bitumen.

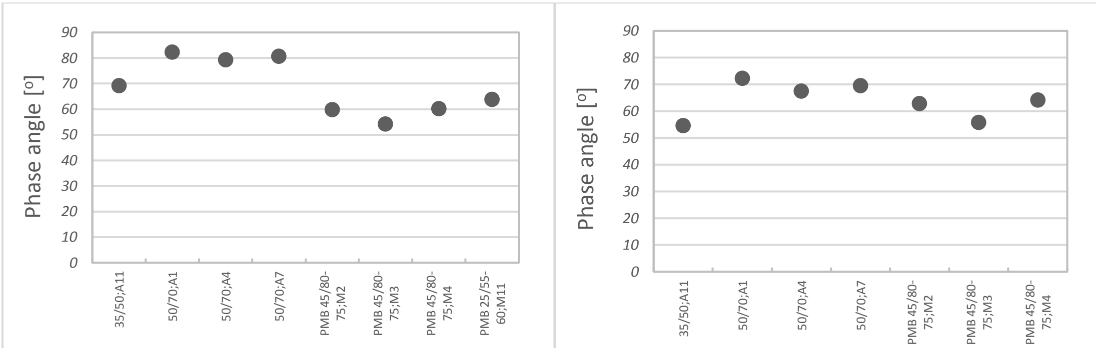

Figure 5. Values of phase angle from the DSR test at 50°C (left) and 25°C (right)

The results of rheological measurements of bitumen binders were compared with the basic properties determined by the empirical tests. The bitumen stiffness at 25°C represented by the penetration was compared with a stiffness modulus set at 25°C and a load frequency of 1.59 Hz, based upon an equivalent vehicle speed of 80 km.h^{-1} (Figure 6). Comparison of penetration results with complex shear modulus values at 25 °C showed that the harder bitumen is, the lower penetration and the higher complex shear modulus are. The relationship between the stiffness as penetration and complex modulus (DSR at 25 °C) was expressed by [13] as *logG*=2.923-1.9*logPen* and confirmed according to [14] dependence between $G^*\sin\delta$ at 25 °C and penetration.

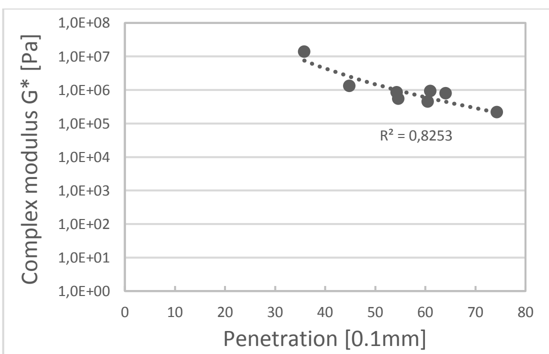

Figure 6. Relation between the penetration at 25°C and complex modulus at 25°C and frequency 1.59 Hz

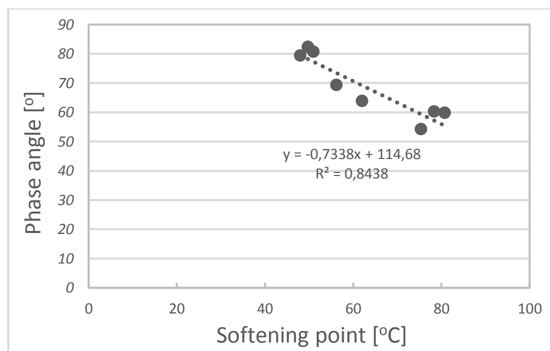

Figure 7. Relation between the softening point and phase angle at 50°C and frequency 1.59 Hz

With increasing temperatures (50°C or 60°C) the complex modulus values decrease. This is a representation of the loading of bitumen in the summer season. The modified bitumen PMB 25/55-60 shows the highest value at both temperatures (50°C or 60°C), next bitumen 35/50, and the other bitumen show significantly lower values. It is evident that the harder bitumen in terms of penetration has higher values of the complex shear modulus. The modulus is reduced by increasing penetration. Relatively high dependence has been shown in Figure 7 between the softening point and phase angle.

5. Conclusion

The main objective of this work was to investigate the rheological properties of bitumen in DSR. The obtained data are graphically presented as isochrones, isotherms, and master curves of the complex shear modulus and phase shift angle. Using the complex shear modulus G^* and the phase angle δ, it is possible to characterize the viscoelastic behavior of bitumen at different temperatures and load frequencies.

By comparing the values of tested paving grade (35/50, 50/70) and modified (PMB 45/80-75, PMB 25/55-60) bitumen, it was determined that the complex shear modulus values of non-modified bitumen are lower than those of the polymer modified bitumen at higher temperatures throughout the frequency range. The phase angle values of non-modified bitumen are significantly higher over the temperature and frequency test range. This is caused by lower elasticity compared to polymer-modified bitumen.

Comparison of penetration results with complex shear modulus at 25 °C showed that the harder bitumen is, the lower penetration and the higher complex shear modulus are. This dependence applies to both paving grade and modified bitumen.

Acknowledgment

The research was supported partially by Scientific Grant Agency of Ministry of Education, Science and Sport of Slovak Republic and Slovak Academy of Science grant VEGA No. 1/0300/17 Research of performance related and rheological properties of bituminous binders.

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