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The Viscoelastic Characteristics of the Asphalt Concrete Modified with Different Synthetic Waxes Using a Modified Huet- Sayegh Model

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Abstract. The article presents the results of dynamic modulus tests carried on the asphalt concrete (AC16W). The sinusoidal load was applied to the samples in accordance with DTC-CY method. The neat bituminous binder (penetration grade 35/50) was modified by means two synthetic waxes, coming from the Fischer-Tropsch raction, with various molecular weights and softening point temperature results (hard and softer). The relaxation phenomenon in terms of changes in complex modulus and phase angle was evaluated using the modified Huet-Sayegh (2S2P1D). Estimated model parameters pointed out that the addition of the synthetic wax with the high (hard wax) and the low (softer wax) molecular weight raised the stiffness of the bituminous binder in relation to the reference bitumen 35/50. The application of the modified Huet-Sayegh model showed that the presence of the synthetic wax in the bitumen significantly affected the stiffness modulus of considered asphalt concretes. Basing analysis on Cole-Cole diagram it was found significant differences in the viscoelastic behaviour between the reference asphalt concrete and the asphalt concretes with synthetic waxes. In contrast, there were no significant differences between viscoelastic properties of tested asphalt concretes modified, used in the experiment, synthetic waxes. Furthermore, the sensitivity to the loading time of asphalt concretes containing both synthetic waxes was marginal.

Keywords: 2S2P1D model, synthetic wax, dynamic test, asphalt concrete, master curve.

Conference topic: Roads and railways.

Introduction

Bituminous mixtures and recycled base layers with bituminous binders are subject to the phenomena assigned to a viscoelastic model (Marques, Creus 2012; Buczyński, Lech 2015). A load applied to a bituminous mixture causes immediate stress relaxation (Yuqing *et al.* 2013). In thermodynamics, relaxation is the process of decreasing internal energy of micelles in the bitumen (Stefańczyk, Mieczkowski 2008). In this way, it contributes to the reduction of stiffness modulus due to the presence of viscous element in the bitumenviscoelastic model. The stiffness modulus master curve is a helpful tool for characterizing the effects of temperature and loading time onlinear viscoelastic materials. Attempts to develop a reliable model for describing rheological properties of bitumen or bituminous mixtures have been widely reported in the literature. These properties are evaluated through the stiffness modulus and phase angle.The technique known as the time-temperature superposition principle (TTSP) allows shifting temperature levels along the time of loading axis to generate a master curve (Goodrich 1998). The materials to which the TTSP principle can be applied are classified as thermorheologically simple (Airey 2002). Among currently used mechanistic models for rheological changes in bituminous mixtures, the generalized Huet-Sayegh model referred to as 2S2P1D can be successfully applied for establishing the master curve (Yusoff *et al.* 2013). The model, calibrated by the Ecole Nationale des Travaux Publics de l’Etat (ENTPE), is capable of providing correct characterization of the behaviour of bitumen and bituminous mixtures (Olard, Di Benedetto 2003).

Materials and test procedures

The two types of Fisher-Tropsch (F-T) synthetic wax (Iwański, Mazurek 2016; Vaitkus *et al.* 2009) used in the tests varied in the molecular mass (according to material safety data sheet) and the resulting softening point. The wax having lower molecular mass (lower density) was designated by “M”, whereas the wax with the higher molecular mass (Iwański, Mazurek 2011) was designated by “N”. The base bitumen was a neat bitumen denoted as 35/50. Basic rheological and physical properties of the waxes used, declared in the product fiches are compiled in Table 1.

The results obtained for wax type N (Table 1) indicate that this type of crystallite (synthetic wax) will affect the base bitumen rheology more strongly (Iwański, Mazurek 2013). High softening point and low penetration values of the synthetic wax will have a substantial effect on rheological properties of the base bitumen.

Table 1. Basic rheological parameters of synthetic waxes and the modified bitumen 35/50

Parameter	Unit of measure	Value				
		Synthetic wax type M (hard wax)	Synthetic wax type N (soft wax)	Bitumen 35/50	Bitumen 35/50+2.5% M	Bitumen 35/50+2.5% N
Softening point	°C	81	100	52	68	75
Penetration	0.1 mm	9	<1	41	28	35
Flash point	°C	>200	>285	>240	–	–
Kinematic viscosity at 135°C	cSt	9	12	6	4	3
Density at 25°C	Mg/m3	0.9	0.94	0.99	–	–
Molecular mass	g/mol	Approx.1000	< 1000	–	–	–

Mineral mix design

Bituminous mixture designated as AC16W had to meet Polish requirements set out in the WT-2/2010 (WT-2/2010, 2010). The AC16W layer was designed for the traffic category KR3-6 ($ESAL_{100kN} < 2.5 \cdot 10^6 \div 7.3 \cdot 10^6$ axles). The composition of the mineral mixture of the AC16W layer was designed by the limit value curve method, in which critical values were in compliance with the WT-2/2010. Figure 1 shows the AC16W grading curve.

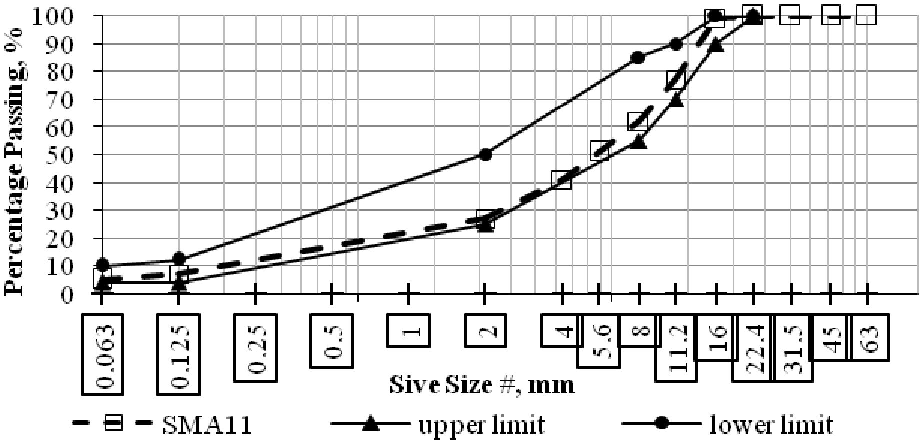

Fig. 1. Particle size distribution in AC16W

In the mineral mixture of AC16W, aggregate fractions 8/16 and 2/8 were made up of limestone, with the 0/4 fraction of dolomite. Limestone powder was used as the filler. To maintain proper adhesion between the bitumen and aggregate, 0.1% of adhesion promoter was used.

Fundamental physical and mechanical properties of the AC16W layer

All mixtures of the asphalt concrete (AC16W) were prepared with the same optimal 4.6% bitumen content. This optimal thershold was determind according to Marshall metodology for the asphalt conctrete with a neat bitumen. It met all reqirements according to polish technical reqirements in WT-2/2010 (WT-2/2010, 2010). Prior to this, the bitumen was modified with 2.5% of M and N waxes at 155 °C for 30 minutes at the blender speed of 400 rpm. All bitumens were modified by the synthetic wax in the amount of 2.5% regardless of their molecular mass. This amout of the synthetic wax for bitumen 35/50 was recommended by authors (Qin *et al.* 2015). The modified bitumen was used to make four samples of asphalt concrete for each modification variant. A total of twelve AC16W samples were produced and compacted in the gyratory compactor to adjust their sizes to the requirements of the DTC-CY procedure (EN 12697-26 Annex D). As a result, the basic physical and mechanical properties of AC16W were determined, as summarized in Table 2.

Table 2. Basic physical and mechanical properties of AC16W

Parameter	Mixture type			Criterion in WT-2/2010	Test Standard
	AC16W-Z	AC16W-M	AC16-N		
Volumetric density ρ_{ssd}, Mg/m^3	2.347	2.346	2.347	–	PN-EN 12697-6
Void spacein the bituminous mixture V_m, % (v/v)	5.7	6.5	6.2	4.0÷7.0	PN-EN 12697-8
Filling the void space with bitumen VFB, %	65.11	62.03	63.14	–	PN-EN 12697-8
Void space in mineral mixture, % (v/v)	16.34	17.01	16.74	–	PN-EN 12697-8
Compaction temperature, °C	145	130	130	–	–

Dynamic modulus in the linear viscoelasticity range

The dynamic modulus of the concrete asphalt (AC16W) was determined by DTC-CY (Direct Tension-Compression Test on Cylindrical Samples) to EN 12697-26 Annex D. Four replications were made of the samples 101 mm in diameter for each case under analysis, while the diameter had to be larger than the four times the maximum grain size in the bituminous mixture. The height (H) to diameter (D) ratio was 1.8. The samples were subjected to constant sinusoidal load with an amplitude of $\varepsilon_o < 25\mu\varepsilon$. The dynamic modulus was determined at 10 °C, 20 °C and 40 °C with loading frequencies of 0.1Hz, 0.3Hz, 1Hz, 3Hz, 10Hz and 20Hz. Dynamic modulus (E*) and phase angle (δ) were obtained from the test results.

The dynamic shear modulus (G*) of the bituminous binder was measured according to EN 14770 (EN 14770:2012). A 25 mm diameter cone-plate set (testing geometry) was used to determine the complex shear modulus and the phase angle performed on all test samples. All tests were carried out at frequency range from 0.1 Hz to 10 Hz and at temperature set-points: 40 °C , 60 °C and 70 °C.

Modified Huet-Sayegh model (2S2P1D)

The 2S2P1D model is a version of the classic Huet-Sayegh model modified by Di Benedetto (Yusoff *et al.* 2013). It is applied to describe linear viscoelastic (LVE) behaviour of a structure, i.e., in the small strain range. The LVE range, where the stiffness modulus does not depend on the applied load levels, has to be determined experimentally.

The model consists of two elastic elements: G_o (representing the static modulus when loading time tends to infinity) and G_o (instantaneous modulus "G_g" when the loading time tends to zero), two parabolic dashpots h and k and a linear dashpot β (defined by zero shear viscosity η_o), t – time of loading, h – exponent changing from 0 to 1 (h = 0 elastic behaviour, h = 1 viscous behaviour). After transformation of the 2S2P1D model in the frequency domain, the function describing the variations in G* in time has the form (1):

$$G^*(\omega) = G_o + \frac{G_g - G_o}{1 + \alpha(i\omega\tau)^{-k} + (i\omega\tau)^{-h} + \alpha(i\omega\beta\tau)^{-1}}, \tag{1}$$

where: $G^*(\omega)$ is the complex shear modulus in the frequency domain, k and h are exponents $0 < k < h < 1$ h changing from 0 to 1 (h = 0 elastic behaviour, h = 1 viscous behaviour), α, β are constants, τ is the characteristic time. Equation (2):

$$\eta_o = (G_g - G_o)\beta\tau, \tag{2}$$

where η_o is the zero shear viscosity, is the basis for calculating dynamic viscosity for long time of loading. This parameter is useful as it characterizes the rate of plastic deformation increase in the material under analysis, and it can be helpful in expressing the sensitivity of bituminous mixtures to rutting.

Estimation of the model parameters required using the non-linear method of least squares for minimization of the objective function at the established initial values. For this purpose, a complex block script was developed in the MathCad program and the solver with implemented Quasi-Newton method was used. Knowing that G* = G' + iG", the parameters in equation (1) were approximated through the search for the objective function minimum.

Finally, to estimate the seven parameters of the model, at least eight values of the complex modulus G* had to be determined at different frequencies. Unlike in the commonly used Burger model, the mechanical parameters of the

2S2P1D model are not dependent on temperature. The only parameter that can be temperature-dependent is the characteristic time, which is a very good solution when we want to use the time-temperature superposition principle (TTSP) in the model and plot master curves describing the change of the complex modulus in time. To determine the TTSP, the following modified equation (3) was used

$$\tau = \tau_o e^{\left(A_1 + T \cdot A_2\right)}, \tag{3}$$

where: T – test temperature, τ_o – initial characteristic time, A_1, A_2 – parameters of the model.

This way of defining the time-temperature superposition (3) is a compromise between the parameter estimation quality and the number of parameters estimated. A more detailed decription on adopted, in the present work, TTSP model can be found in the Yusoff's work (Yusoff 2012). The final stage of parameter approximation in the 2S2P1D model involves the assessment of the quality of the model fit to the values from the experiment. Two goodness-of-fit statistic methods were used: the coefficient of determination R^2 and the mean normalized error MNE (Yusoff *et al.* 2013). As these two measures are not correlated, the high quality of the model has to take account of the high level of the R^2 coefficient and low level of the mean squared normalized error MNE.

Test results

Viscoelastic characteristics of the bitumen

In compliance with the test methodology, thedynamic shear rheometer (PN-EN 14770:2012) was used to determine viscoelastic parameters of the bitumen: the elastic component of the complex modulus (G'), its imaginary part (G") and the phase angle (δ). The results obtained from the method used to estimate the 2S2P1D model parameters (section 3) are summarized in Table 3.

Table 3. Estimated 2S2P1D model parameters for bitumen

Bitumen	2S2P1D model parameters										
	G_o [Pa]	G_g [Pa]	h [–]	k [–]	α [–]	β [–]	τ [–]	A_o [–]	A_1 [–]	R^2 [–]	MNE [%]
35/50	0	$4.42 \cdot 10^7$	0.53	0.35	1.00	196.68	0.001	–0.18	2.94	0.97	9.6
35/50M	0	$4.76 \cdot 10^7$	0.34	0.22	6.06	387.52	0.001	–0.23	6.15	0.97	12.1
35/50N	0	$7.24 \cdot 10^7$	0.35	0.24	9.18	684.05	0.0008	–0.23	5.69	0.94	14.2

Note that the value of "β" in the bitumen containing the N – wax is the highest in all bitumen types. This wax ensures the high level of dynamic viscosity in the modified bitumen for a long time of loading. It is extraordinarily beneficial to use this bitumen in bituminous mixtures that need to have a high resistance to permanent deformation. The same fact was observed as the high level of the complex modulus at low frequencies (Fig. 2b). Attention should be drawn to the change in "h" and "k" parameters. These are the main variables in the 2S2P1D model, responsible for the viscoelastic character of the bitumen expressed with the use of the Cole-Cole plan (Fig. 2a). The decrease in both parameters causes the bitumen to show the predominance of the elastic component of the complex modulus over the viscous component. At the same time, the sensitivity of the bitumen to the loading time will also decrease. Considering the results compiled in Table 3, the bitumen with the synthetic wax, both N and M, considerably affect the rheology of the bitumen changing it to be more elastic relative to neat bitumen 35/50.

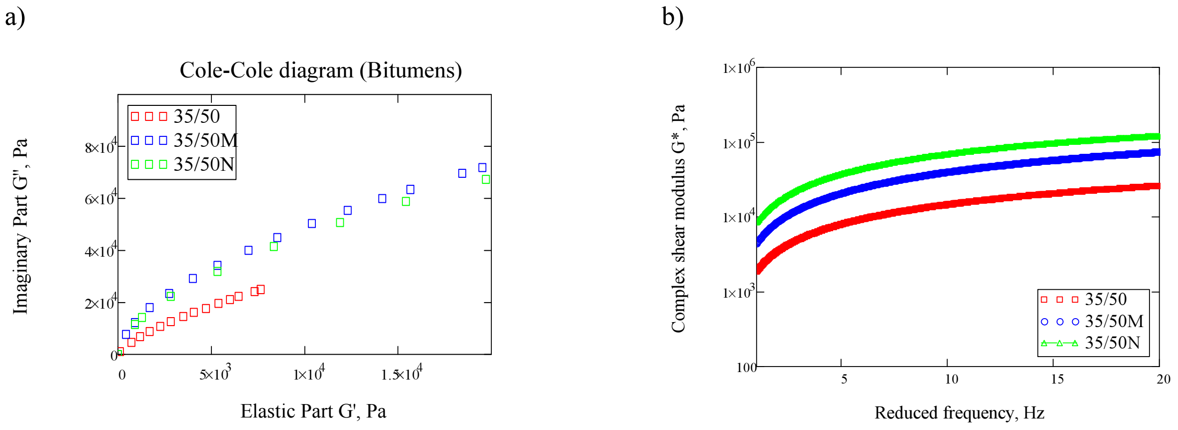

Fig. 2. Bitumen viscoelastic characteristic according to 2S2P1D model; a) Cole-Cole diagram; b) master curve of G* at 60 °C

Evaluation of the simulation results indicates that the N wax causes the highest increase in the bitumen stiffness (high G*). Analysis of the slope of the complex modulus curve by the high quality of parameters “α” (asymptoteat high frequencies) and “β” (asymptote for low frequencies) shown in Table 3 indicates that compared with the reference bitumen 35/50, the sensitivity of bitumen modified with any type of synthetic wax is lower. Consequently, the rate of the stiffness modulus reduction in the wax modified bitumen will decrease, which is of great importance in the case of bituminous mixtures produced at the crossroads.

Viscoelastic characteristic of AC16W

In addition to the evaluation of the effect of synthetic wax on bitumen, the primary analysis concerned the asphalt concrete. The fitting of the changes in the values of the dynamic modulus E* was performer in the same way as in the case of the shear modulus G* of the bitumen. Table 4 compiles the results of fitting the test data to the 2S2P1D model.

Table 4. Estimation of 2S2P1D model parameters for asphalt concrete AC16W

Bitumen used in AC16W	2S2P1D model parameters for bituminous mixtures										
	E_o [MPa]	E_g [MPa]	h [–]	k [–]	α [–]	β [–]	τ [–]	A_o [–]	A_1 [–]	R^2 [–]	MNE [%]
35/50	232.0	$3.41 \cdot 10^4$	0.526	0.263	6.61	358.3	0.1	–0.252	11.22	0.98	13.9
35/50M	120.1	$3.81 \cdot 10^4$	0.398	0.199	4.4	967.8	0.1	–0.275	11.37	0.98	6.17
35/50N	438.4	$6.61 \cdot 10^4$	0.420	0.10	2.9	$3.31 \cdot 10^3$	0.1	–0.266	4.99	0.99	3.99

Compiled results of fitting parameters “k” and “h” of the bitumen and asphalt concrete showed that in the asphalt concrete, parameter “h” characterizing viscous behaviour of the material was comparable with the values obtained for the bitumen. The parameter “k” (responsible for elastic behaviour) decreased considerably in the asphalt concrete due to the presence of the mineral skeleton. The rate of change of “k” increases with the increasing hardness of the wax added and the lowest in the asphalt concrete containing neat bitumen 35/50. It may thus be expected that the asphalt concrete with synthetic wax will be much more elastic than that with ordinary bitumen. This is confirmed by the low level of “α” i.e., elastic asymptote attains a wide range of loading times. Consequently, within a wider range of frequencies, the assumption of an elastic system of layers in the mechanistic methods-based design is met. This parameter also characterizes the inclination of the G* curve for high frequencies (short loading time). And thus, the asphalt concrete for typical loading times of pavements under vehicle load will have the reaction definitely elastic in contrast to the asphalt concrete with bitumen 35/50, where the reduction in stiffness induced by relaxation is more pronounced. Attenuation effect in the asphalt concrete will be marginalized. The use of synthetic wax causes the increase of “β” closely related to the viscosity of the intact concrete asphalt. This increase leads to the low rate of permanent deformation for a long loading time. The increase of “β” in AC16W grows with increasing content of harder synthetic wax in the bitumen. Consequently, the synthetic wax-modified asphalt concrete may be expected to inhibit creep-induced permanent deformation. Figure 3 shows master curves for the stiffness modulus of wax-modified asphalt concrete at the reference temperature of 20 °C, and the Cole-Cole diagram.

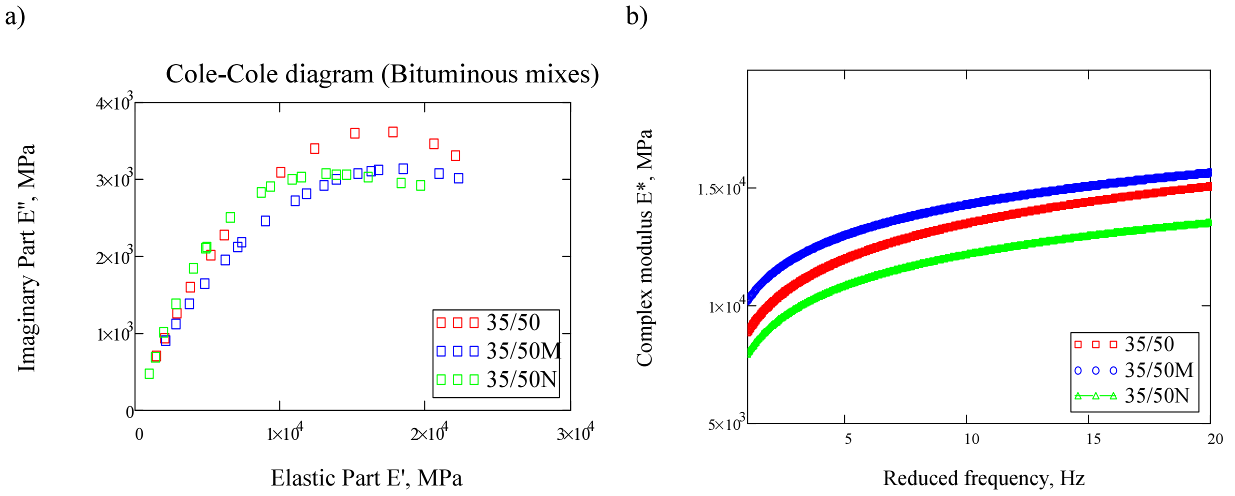

Fig. 3. Asphalt concrete (AC16W) viscoelastic characteristic according to 2S2P1D model;
a) Cole-Cole diagram; b) master curve of E* at 20 °C

The results of the E'-E" relationship of the stiffness modulus with different synthetic waxes in the Cole-Cole diagram (Fig. 3a) are below the results of the E'-E" relationship of the asphalt concrete with ordinary bitumen. This means that asphalt concretes containing synthetic wax will deform much more elastically than the concrete with ordinary bitumen. The change curve for the complex modulus for the asphalt concrete with bitumen 35/50 at the reference temperature is placed much higher than that with bitumen 35/50N (Fig. 3b). The low value of "α" expressing predominance of the asymptote of elasticity, and high "β" makes it more loading time-insensitive. It is also very important that the durability of bituminous mixtures is more dependent on their elastic characteristic than on the maximum complex modulus of stiffness describing the behaviour of the bituminous mixture in a comprehensive way. The complex modulus does not define the rate of elastic recovery of AC16W directly.

It has to be noted that the results had a high coefficient of determination, $R^2 > 0.98$ and low error, MNE < 14% (Table 4) and it was grapically presented in Figure 4b. The error in the asphalt concrete with bitumen 35/50N is the lowest as its behaviour corresponds best to the LVE model.

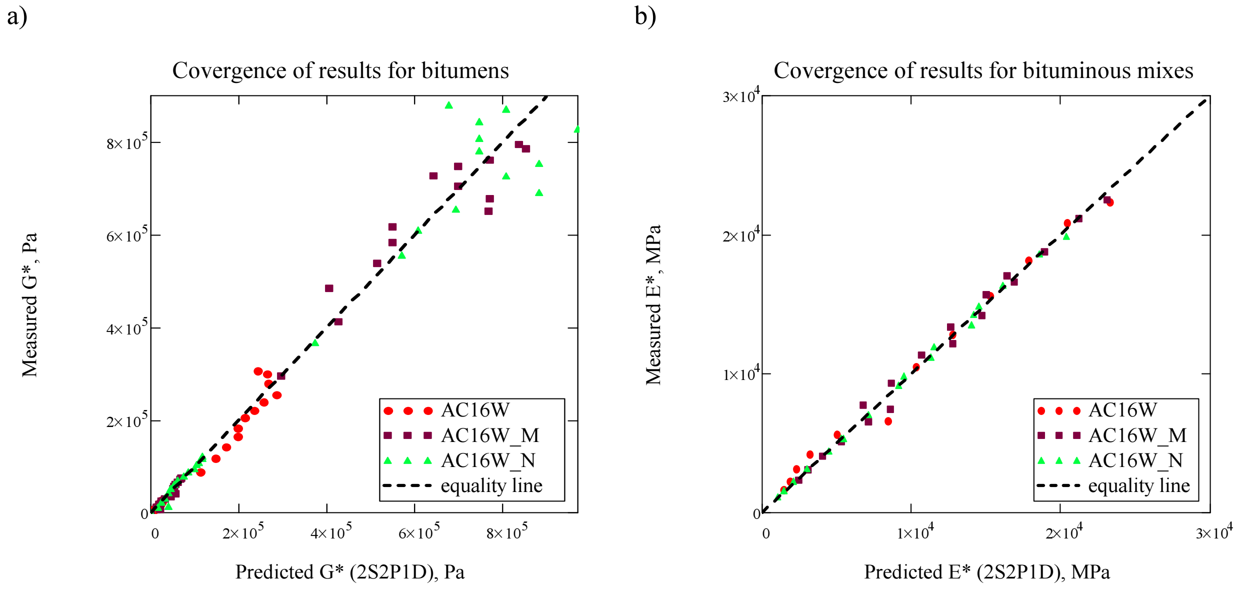

Fig. 4. Measured versus predicted data; a) G* for bitumen, b) E* fo AC16W

Analysis of the quality of fit of the shear modulus G* results for bitumens at 40 °C shows discrepancies between its model and experimental values. This results from the fact that N wax-modified bitumen had a substantial gain in the complex modulus value. The recorded complex modulus value was close to the cone and plate's measurement capability (25 mm). The maximumresidual (MNE) did not exceed 15% for bitumen 35/50N. In all cases, high fitting was obtained, as shown by $R^2 > 0.94$ (Table 3). The dispersion of the results was graphically presented in Figure 4b.

Conclusions

Analysis of the results allowed formulating the following conclusions:

- Application of the 2S2P1D model allows precise prediction of LVE stiffness modulus results and their representation in the form of a master curve;
- The use of synthetic wax with a high molecular mass (hard) has a strong effect on the viscoelastic character of the bitumen and the AC16W mixture made with this bitumen;
- The use of wax with lower molecular mass increases the complex modulus at a slight decrease in the zero shear viscosity at high temperatures/high frequency according to level of parameter "β";
- Synthetic wax increases the zero shear viscosity of a bituminous mixture, thus leading to its higher resistance to permanent deformation;
- Viscoelastic character of the AC16W mixture reflects the viscoelastic character of the bitumen. Increase in elastic predominance in the bitumen (E') rises with the hardness of the wax used;
- The wearing course made with the AC16W mixture and an addition of synthetic waxes regardless of their molecular has a lower sensitivity to time/temperature than in the case ordinary bitumen is used;
- Goodness-of-fit of the experimental results to the 2S2P1D model is higher in AC16W mixtures containing hard synthetic waxes.

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