INVESTIGATION OF COHERENCE BETWEEN EMPIRICAL AND RHEOLOGICAL PROPERTIES OF BITUMENS WITH DYNAMIC SHEAR RHEOMETER TESTS

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ABSTRACT

The paper shows correlation between penetration index and rheological parameters tested with dynamic shear rheometer(DSR) for paving grade bitumens at 10 Hz and + 20°C. A linear correlation exists for paving grade bitumens and a second order correlation is given for modified bitumens between bitumen stiffness determined from van der Poel monograph and complex modulus derived from DSR tests at + 20°C and 0.1-30 Hz frequency.
Test data of Multiple Stress Creep Recovery Test at 60 °C were fitted to Burger's model parameters.The four parameters of Burger's model are illustrated versus ten creep-recovery cycles of MSCR test. The average relative errorof fit is favourable for paving grade bitumens the error is larger for modified bitumens. Results of testing zero-shear viscosity in creep mode show extended creep time is needed for reaching steady state flow. This difference is presented on diagrams of viscosity versus shear stress and shear rate for paving grade and modified bitumens. ZSV values were also determined with creep and recovery tests in LVE-range. The ZSV values are close to ones obtained with creep test for paving grade bitumens but large differences were found between respective results of modified bitumens.

Keywords: Zero-shear viscosity, polimer modified bitumen, creep, viscosity, rheology

1. INTRODUCTION

The bitumen rheological properties become increasing attention as they more directly connected to fundamental properties of bituminous mixtures. At the same time some traditional empirical parameters of bitumens like needle penetration or ring and ball softening point are specified and still used in practice thus it is worthwhile to seek relationships between parameters of these approaches. Two sections of this paper are devoted to show coherence of penetration index and stiffness modulus with rheological parameters of paving grade and modified bitumens. In the SHRP Superpave specifications the performance behaviour of bitumen was defined with viscoelastic properties measured with dynamic shear rheometer. During revision of SHRP Superpave specification the multiple stress creep recovery (MSCR) test was proposed [1]. It was attempted to fit the multiple stress creep recovery (MSCR) test data to Burgers model to reveal intrinsic rheological parameters. The zero-shear viscosity (ZSV) determination with creep tests needs longer time and problems with repeatability for highly modified bitumens were discussed earlier [3]. To reduce test time another approach for determination of ZSV was studied making one cycle creep and recovery test in LVE-range.

2. BITUMENS AND TEST PROGRAMME

2.1 Tested bitumens

The tested bitumens are listed in Table 1 where the needle penetration at + 25°C, the Ring and Ball softening point and the penetration index is also given. The Pmb-s are modified with elastomers, the chemically stabilized rubber bitumen (RB) signed as KSGB. The bitumen samples were taken at asphalt plants.

Table 1: Summary of tested bitumens and performed tests

Sample name	Product sign	*G** 10 Hz @+20°C	*ZSV* in creep test @+60 °C	*ZSV* in creep and recovery test @+60°C	*RTFOT+ MSCR* @+60 °C	Needle penetration @25 °C [0.1 mm]	R&B Softening point [°C]	Penetration Index PI
RB	KSGB	+	+		+	78	55,5	1,28936
C-003/08	PmB 30/60 S	+				48	77,0	3,79403
2842/09	PmB 25/55-65	+	+		+	48	66,2	2,08300
SW 2008	PmB	+				56	59,5	1,22011
SW 2009	PmB 25/55-65	+				36	69,2	1,88818
C-005/08	B 50/70	+				61	49,4	-0,88282
2033/09	B 35/50	+				45	55,3	-0,20410
A/07	B 50/70	+				50	52,3	-0,64064
C-004/08	B 35/50	+				41	54,6	-0,56477
1645/09	PmB 10/40-65	+				24	70,8	1,26747
C-087/08	PmB 10/40-65	+				22	70,0	0,97263
C-034/08	B15	+				16	81,0	1,91925
1864/09	B 50/70	+				51	52,3	-0,59442
2448/09	PmB 10/40-65	+	+		+	24	67,0	0,66496
B 10/1	B 10	+				11	88,3	2,14312
B 130/1	B 130	+				87	43,8	-1,57939
B 15/1	B 15	+				14	75,0	0,88154
B 15/2	B 15	+				11	79,3	1,05270
B 15/3	B 15	+	+		+	17	70,5	0,57057
B 160/220/1	B 160/220	+				173	39,0	-1,11152
B 25/1	B 25	+				24	61,3	-0,31517
B 35/50/2	B 35/50	+	+		+	35	56,0	-0,60142
B 50/70/2	B 50/70	+				44	51,0	-1,23417
B 50/70/3	B 50/70	+	+		+	56	51,0	-0,68602
B 70/100/1	B 70/100	+				73	47,3	-0,99455
Sfb 5-JR-50	Sfb 5-JR-50	+				53	70,5	3,06962
B 50/70-4	B 50/70	+	+		+	60	48,8	-1,08023

2.2 The test programme

The needle penetration at +25 °C the Ring &Ball softening point and the penetration index was determined for all bitumens. The stiffness moduli $S_{bitpen\text{-}R\&B}$ of bitumens were determined using van der Poel monograph's computer code Shell Bands [5].

The complex shear modulus *G**, storage modulus *G*', loss modulus *G*'', G*/sinδ, complex viscosity *η**, real part of complex viscosity *η*', imaginary part of complex viscosity *η*″ and phase angle *δ* were determined with DSR measurements in CSD oscillation mode in LVE-range with γ =0,05%, at 10 Hz and at +20 °C on pure samples.

The zero-shear viscosity was measured with two different test mode with creep tests as well as with creep and recovery test. In creep mode the ZSV was determined at +60°C on pure samples, according to CEN/TS 15325:2008 [7]. The test consisted two steps: in Step 1 stress sweep was executed to select creep test stress and in Step 2 the creep test was performed with selected stress during a sufficiently long time to reach steady state flow.

Determination of ZSV was also made with creep and recovery test in LVE-range on pure samples at + 60°C. For this approach at first stress was selected with stress sweep (Step 1) according to technical specification CEN/TS 15325:2008 but in Step 2 one cycle creep and recovery test was performed and the ZSV-value was calculated from Burgers model.

This test was performed in two variants: with 60 min creep phase and 15 min recovery phase and with 240 min creep phase and 15 min recovery phase.

The MSCR tests were performed according to *AASHTO TP 70-07* test method [8]. Before this test to simulate plant aging RTFOT tests were made on bitumen samples. Each cycle of MSCR test consisted a creep phase under constant stress τ_0 during 1s time interval ($t<t_1$), than the stress is removed at time $t=t_1$ and a recovery phase at τ_0=0 with 9s duration ($t\geq t_1$). The test was performed with 10 cycles at 0,1 kPa, 3,2 kPa and 6,4 kPa stress levels sequentially at + 60°C.

3. RESULTS AND DISCUSSION

3.1 Relationships between penetration index and fundamental properties

One of the most widely used empirical parameter of the bitumen is the penetration index (*PI*) developed by Pfeiffer and van Doormal based on penetration and R&B softening point [4]. Correlation was searched between the penetration index and the *G**, *G*', *G*'', G*/sinδ, stiffness modulus $S_{bitpen\text{-}R\&B}$, *η**, *η*', *η*″, *δ*. It was found that very weak or no correlation exists between penetration index and the above mentioned parameters for modified bitumens (R^2=0.20-0.40) but a good correlation was detected for paving grade bitumens between *PI* and G* (R^2=0.8804), *G*'(R^2=0.9026), *G*/sin* δ (R^2=0.9091), *η** (R^2=0.8810), *η'* (R^2=0.7931), and *η*″(R^2=0.9029), (see Figure 1, Figure 2 and Table2).

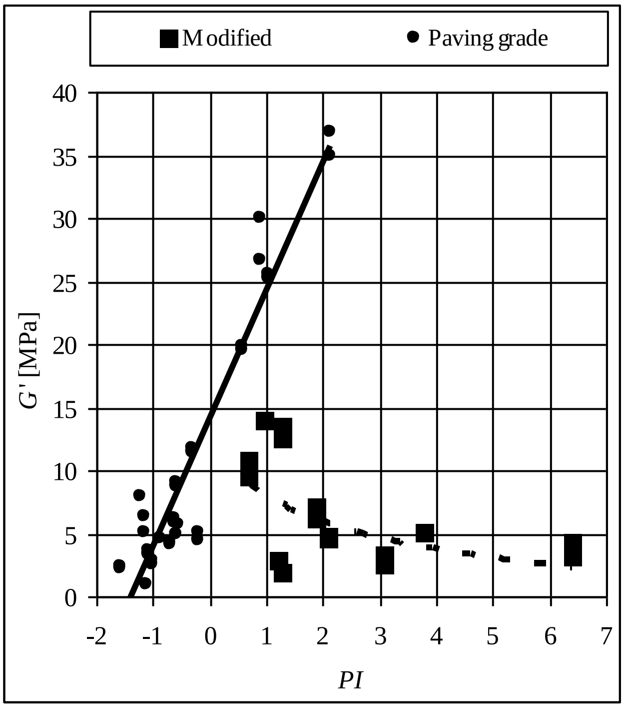


Figure 1: Correlation between penetration index (*PI*) and storage modulus (*G*') of paving grade and modified bitumens at 10 Hz and at +20°C

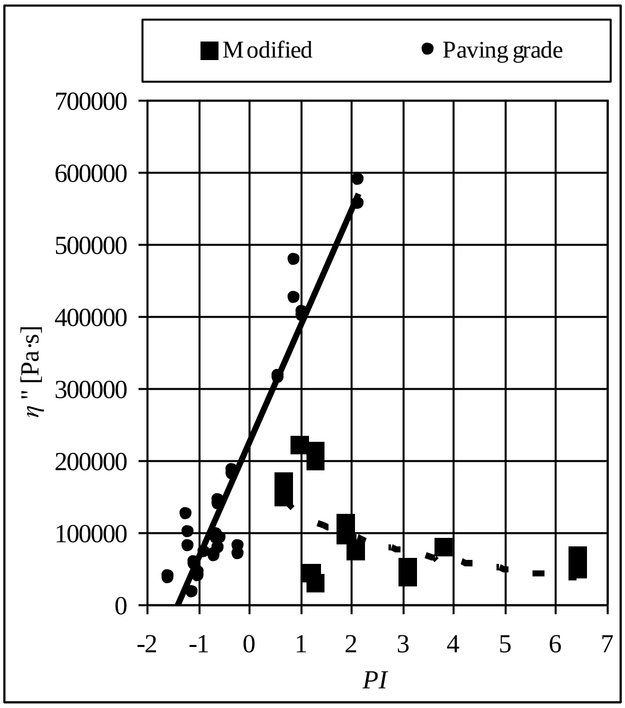


Figure 2: Correlation between imaginary part of complex viscosity (*η*") and penetration index (*PI*) for paving grade and modified bitumens at 10 Hz and at +20 °C

Table 2: Correlation between rheological parameters measured with DSR and penetration index for different paving grade bitumens at 10 Hz and +20°C

Parameter	Correlation equation	R^2
Complex viscosity [Pa·s]	$\eta^* = 175441*(PI)+280694$	0.8810
Imaginary part of complex viscosity [Pa·s]	$\eta'' = 161447*(PI)+223310$	0.9029
Real part of complex viscosity [Pa·s]	$\eta' = 77538*(PI)+164623$	0.7931
$G^*/\sin\delta$	$G^*/\sin\delta = 23.63*(PI)+31.13$	0.9091
Storage modulus [MPa]	$G' = 10.13*(PI)+14.03$	0.9026
Loss modulus [MPa]	$G'' = 4.871*(PI)+10.345$	0.7935
Complex modulus [MPa]	$G^*=11.019*(PI)+17.643$	0.8804
Phase angle [°]	$\delta = 43.575-9.5459*(PI)$	0.7812

3.2 Correlation between complex modulus and stiffness modulus

Correlation was established between bitumens stiffness moduli $S_{bitpen-R\&B}$ calculated from empirical properties and complex moduli E^*_{bitDSR} determined with DSR-measurements at T=+20°C in frequency range of f=0.1-30 Hz. For paving grade bitumens a second order polynomial regression shows good correlation (R^2=0.9284-0.9701) as follows:

$$Y = a + b * X + c * X^2 \qquad \ldots (1)$$

For modified bitumens the linear correlation is (R^2=0.8712-0.9483):

$$Y = a + b * X \qquad \ldots (2)$$

where:
a, b ,c are the coefficients in equations (1) and (2), summarized in Table 3;
$Y=S_{bitpen-R\&B}$ is the stiffness modulus determined with van der Poel nomograph applying Shell-Bands computer code with input of needle penetration and $R\&B$ softening point [5];
$X=E^*_{bitDSR}$ is the complex modulus derived from DSR measurements.
It may be assumed that bitumen is incompressible (Poisson ratio=0.50), than it's complex modulus E^*_{bitDSR} can be calculated using it's measured complex shear modulus G^* as:

$$E^*_{bitDSR} = 3 \cdot G^* \qquad \ldots (3)$$

Regression relationships were determined by frequency separately for groups of paving grade and modified bitumens, than Figure 3 and Figure 4 were built assembling curves calculated with own regression equation for each frequency.

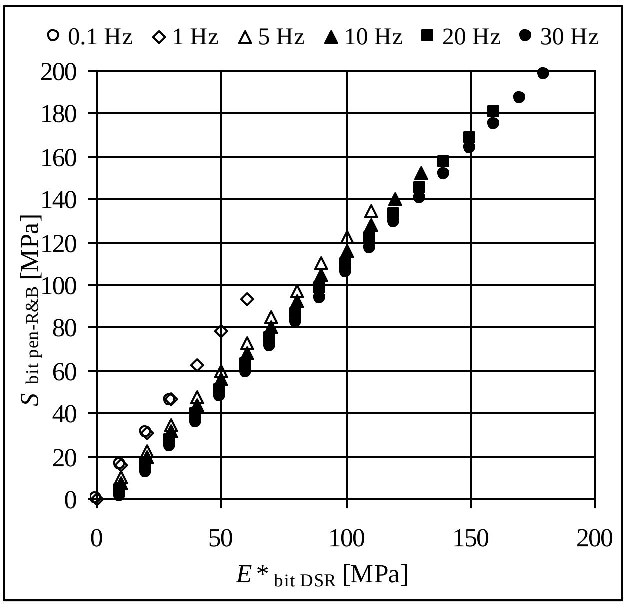


Figure 3: Stiffness determined from van der Poel nomograph ($S_{bitpen-R\&B}$) versus complex modulus (E^*_{bitDSR}) at +20 °C derived from DSR tests at frequencies of 0.1-30 Hz for modified bitumens

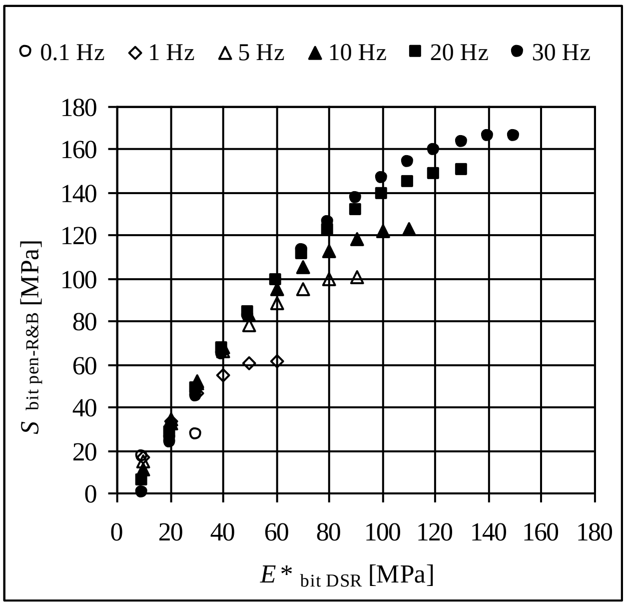


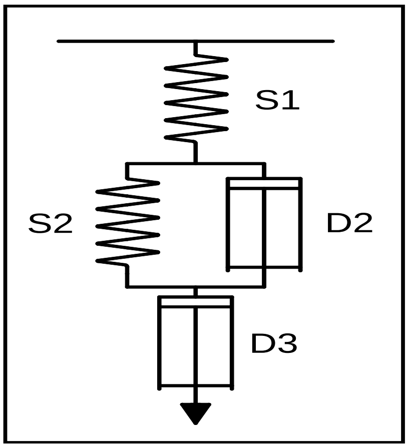
Figure 4: Stiffness determined from van der Poel nomograph ($S_{bitpen-R\&B}$) versus complex modulus (E^*_{bitDSR}) at + 20 °C derived from DSR tests at frequencies of 0.1-30 Hz for paving grade bitumens

Table 3: Coefficients of regression equations (1) and (2) for paving grade and modified bitumens

Test frequency f, [Hz]	Modified bitumens			Paving grade bitumens			
	a	b	R^2	a	b	c	R^2
0.1	+0.2780	1.5280	0.9483	-1.0630	2.1510	-0.041	0.9701
1	-0.1829	1.3570	0.9135	-3.4572	2.2660	-0.019	0.9642
5	-2.4196	1.2488	0.8984	-8.0358	2.3910	-0.013	0.9549
10	-4.6527	1.2100	0.8892	-12.415	2.4640	-0.011	0.9520
20	-8.0640	1.1768	0.8803	-18.935	2.5199	-0.0094	0.9393
30	-10.632	1.1604	0.8712	-24.609	2.5776	-0.0087	0.9284

3.3 Parameters of Burgers model from multiple stress creep recovery tests

The viscoelastic Burgers model is shown on Figure 5 consisting a serial connection of the Maxwell model (spring S1, dashpot D3) and of the Kelvin-Voigt model (spring S2 and dashpot D2). The G_1 and G_2 shear moduli relate to spring elements S1 and S2 the shear viscosities η_2 and η_3 correspond to dashpots D2 and D3 respectively. The strain ε(t) in time t expressed under constant shear stress τ_0 for the creep phase ($t<t_1$) and for the recovery phase ($t\geq t_1$) separately. For determining the parameters of Burgers model a method presented by Li You and Zhanping You was applied where the constitutive equation (4) of this model was converted into two linear equations and the creep-recovery data were fitted with linear equations [2].



$$\varepsilon(t) = \begin{cases} \dfrac{\tau_0}{G_1} + \dfrac{\tau_0 \cdot t}{\eta_3} + \dfrac{\tau_0}{G_2} \cdot \left(1 - e^{-\frac{G_2}{\eta_2} \cdot t}\right) & (t < t_1) \\ \dfrac{\tau_0 \cdot t_1}{\eta_3} + \dfrac{\tau_0}{G_2} \cdot \left(1 - e^{-\frac{G_2}{\eta_2} \cdot t}\right) \cdot e^{-\frac{G_2}{\eta_2} \cdot (t - t_1)} & (t \geq t_1) \end{cases} \quad \ldots(4)$$

Figure 5: The Burgers model

The average relative error ($\overline{ARE}$ %) of the regression model of the whole MSCR test at a given stress level:

$$\overline{ARE}\% = \frac{1}{N} \cdot \sum_{j=1}^{N} \frac{100}{n} \cdot \sum_{i=1}^{n} \frac{|\hat{\gamma}_i - \gamma_i|}{\hat{\gamma}_i} \quad \ldots(5)$$

where:

$\hat{\gamma}_i$ = predicted shear strain by the regression model at ith measuring point;
γ_i = mesured shear strain at ith measuring point;
n = number of measuring points over a creep-recovery cycle ($1\leq i\leq$n);
N = number of creep-recovery cycles of MSCR test, ($1\leq j\leq N$), (here N=10).

Results of regression analysis are summarized in Table 4. The coefficients of variation were mainly largest for G_1 (corresponding to spring S1). The extent of relative error depends on the bitumen type and on the stress level as given in Table 5. The largest relative errors arisen at lowest stress level independently of bitumen type and larger errors occurred for modified bitumens and hard bitumen. The decrease of relative error may be explained with applying stress levels successively without interruption between stress levels. The average relative error of the regression model was favourable for paving grade bitumens (0.51%-4.40%), some larger error occured at modified and hard bitumens (6.7-16.7%). Figures 8-11 illustrate the variation of four parameters of Burgers model at 3.2 kPa stress level during ten creep and recovery cycles for PmB 10/40-65 and a B50/70 paving grade bitumens. Comparing these two bitumens the effect of modification appears mostly on the increasing value of shear viscosity η_3 (corresponds to dashpot D3).

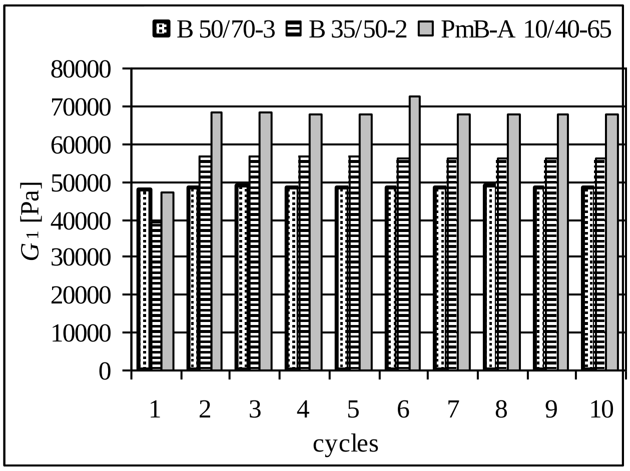


Figure 8: Variation of s shear modulus G_1 during the creep-recovery cycles of different bitumens

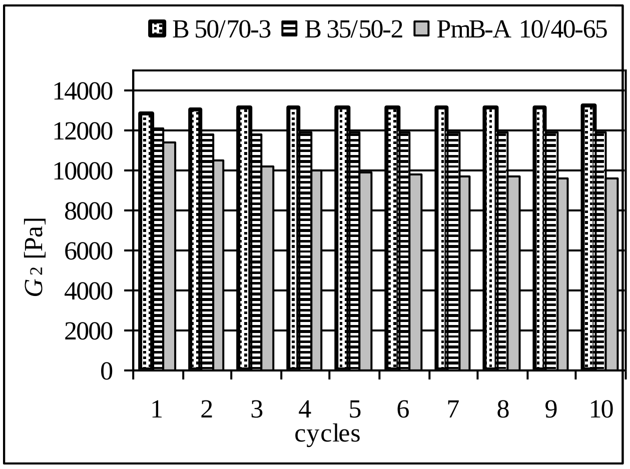


Figure 10: Variation of shear modulus G_2 during the creep-recovery cycles of different bitumens

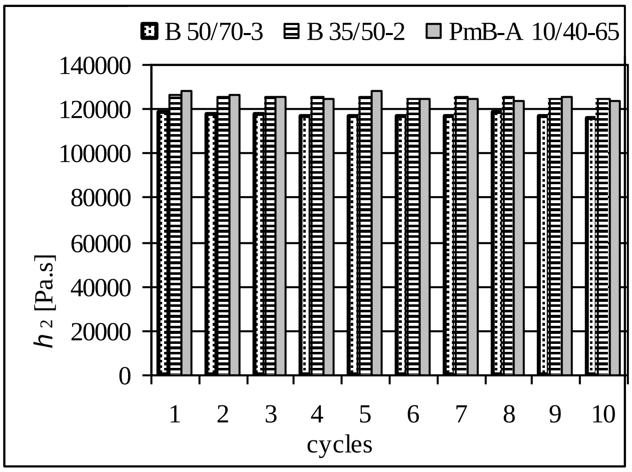


Figure 9: Variation of shear viscosity η_2 during the creep-recovery cycles of different bitumens

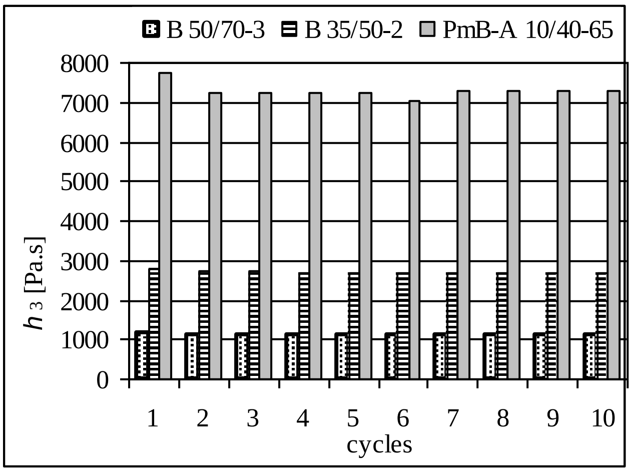


Figure 11: Variation of shear viscosity η_3 during the creep-recovery cycles of different bitumens

Table 4: Parameters of Burgers model at 3.2 kPa stress levels at +60°C of paving grade and modified bitumens

Type and parameter of bitumen		G_1 [Pa]	G_2 [Pa]	η_2 [Pa·s]	η_3 [Pa·s]	*ARE* [%]
Chemically stabilized rubber bitumen	Average value	15364	3554	1296	36623	6.70
	Standard deviation	1527	28	42	327	
	Coefficient of variation, %	9.94	0.79	3.25	0.89	
PmB 10/40-65	Average value	71316	10028	7365	125598	16.51
	Standard deviation	17905	533	203	1402	
	Coefficient of variation, %	25.11	5.32	2.76	1.12	
PmB 25/55-65	Average value	38122	8505	2269	92651	4.40
	Standard deviation	3492	152	22	619	
	Coefficient of variation, %	9.16	1.78	0.96	0.67	
B 35/50 (2)	Average value	54653	11896	2713	125380	3.82
	Standard deviation	5153	65	31	417	
	Coefficient of variation, %	9.43	0.55	1.15	0.33	
B 50/70 (3)	Average value	44946	13072	1146	116856	1.49
	Standard deviation	13729	111	25	848	
	Coefficient of variation, %	0.46	0.85	0.10	0.73	
B 50/70 (4)	Average value	40759	16638	737	137489	0.51
	Standard deviation	4442	127	6	2551	
	Coefficient of variation, %	10.90	0.76	0.83	1.86	
B 15 (3)	Average value	183575	36062	27181	422704	16.72
	Standard deviation	17480	1488	703	3113	
	Coefficient of variation, %	9.52	4.13	2.58	0.74	

Table 5: Average relative error of fitting Burgers model to MSCR test data, [%]

Stress level [kPa]	Bitumen type						
	KSGB	PmB 10/40-65	Pmb 25/55-65	B 35/50 (2)	B 50/70 (3)	B 50/70 (4)	B 15 (3)
0.1	12.6	16.9	5.9	5.1	2.7	2.2	17.1
3.2	6.7	16.5	4.4	3.8	1.5	0.5	16.7
6.4	1.8	9.1	1.3	1.0	0.2	0.1	9.4

3.4 The zero-shear viscosity in creep mode

It was experienced that stress selection for linear range was possible definitely for paving grade bitumens B50/70 for 1 hour test time in Step 2. For modified binders the steady state could only be reached after much longer test time (8-12 hours) and the calculation of steady state viscosity (*SSV)* over last 15 minutes of each hour resulted different values. This was typical for PmB 25/55-65 binder tested at τ=10 Pa. Figure 12 and Figure 13 illustrate the stress sweep and the shear rate dependency of tested bitumens. The viscosities have been calculated over last 15 minutes (Δt) of each hour by formula (6) as in [7]:

$$\eta_i = \frac{\Delta t}{\Delta J} = \frac{900}{(J_{end} - J_{15min-before-end})} \qquad \ldots(6)$$

where:

η_i = the steady state viscosity (*SSV*);

ΔJ = the creep compliance over last 15 minutes.

Table 6 shows that viscosities of PmB 25/55-65 calculated by (6) are very different even between 8-18 hours of testing time. The steady state has been reached between 780-1080 minutes. Therefore the determination of *SSV* has been determined with following steps:

– building the strain curve versus time and determining the linear section on the curve;

– calculation of *ZSV* for this linear section with formula:

$$\eta_0 = \frac{\Delta t}{J_1 - J_0} \qquad \ldots(6)$$

where:

Δt = the testing time in LVE-range;

η_0 = the steady state viscosity accepted for ZSV;

J_1 = the creep compliance at the end of linear section;

J_0 = the creep compliance at the beginning of linear section.

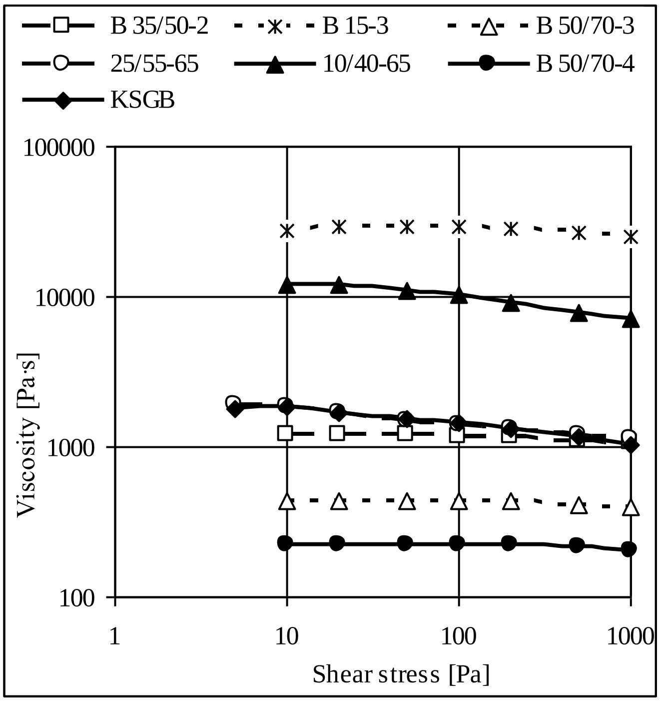


Figure 12: Viscosity versus shear stress for paving grade and modified bitumens at +60 °C

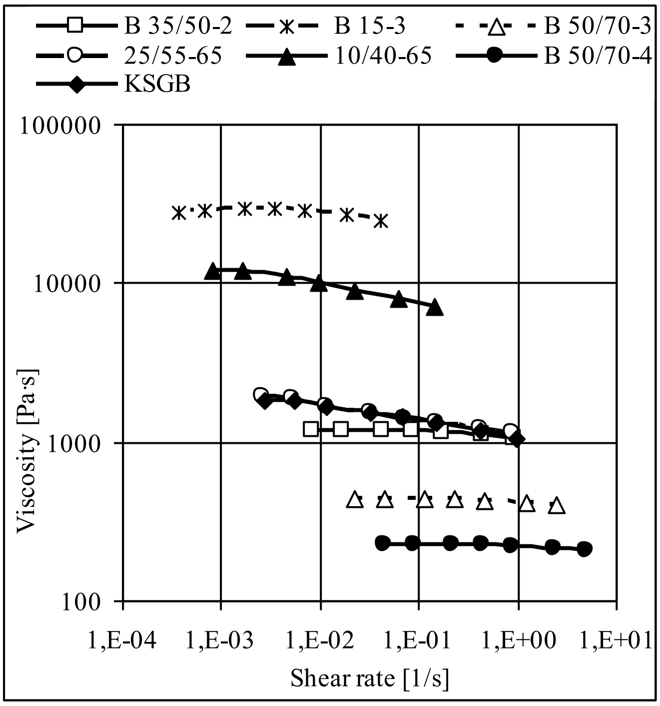


Figure 13: Viscosity versus shear rate for paving grade and modified bitumens at + 60°C

Table 6: Linear intervals and steady state viscosities of paving grade and modified bitumens at + 60 °C

Bitumen type and sample No.	B 50/70 (3)	B 50/70 (4)	B 15 (3)	B 35/50 (2)	PmB 25/55-65	PmB 10/40-65
Test stress, τ [Pa]	50	20	50	50	10	10
Linear interval time [min]	1-720	1-720	100-720	80-1427	780-1080	400-960
SSV, [Pa s]	415	236	34897	1427	4000	12973

3.5 Zero-shear viscosity in creep and recovery mode

The zero-shear viscosity η_0 corresponds to constant of dashpot D3 (Figure 4). The ZSV values were calculated from Burgers model equations with method indicated in section 3.4 [2]. The creep recovery tests were performed with two different creep time (60 min and 240 min) but the recovery time was in both variants 15 min. As can be seen from Table 7 the applied creep time affects the ZSV values on 4.3%-18%. While ZSV values of two paving grade bitumens (B 50/70 and B 35/50) calculated from two different test modes deviates from each other on 1.4%-7.2% this difference is unacceptable large in case of two modified bitumens.

Table 7: Zero-shear viscosities of paving grade and modified bitumens at + 60 °C

Bitumen type	Creep time in creep and recovery mode [min]	Test stress [Pa]	ZSV in creep-recovery mode [Pa·s]	ZSV in creep mode [Pa·s]
PmB 10-40/65	60	10	9091	12973
	240		8696	
PmB 25/55-65	60	10	1433	4000
	240		1692	
B 35/50 (2)	60	50	1342	1242
	240		1225	
B 50/70 (3)	60	50	470	415
	240		447	

4. CONCLUSIONS

The penetration index of paving grade bitumens is in good correlation with rheological parameters (G^*, G', $G^*/\sin\delta$, η^*, η', η'') determined with DSR measurements at 10 Hz and at + 20 °C. These parameters of modified bitumens and hard bitumens indicated nonlinear weak correlation relationships with penetration index.
A second order polynomial correlation exists between bitumen stiffness $S_{\text{bitpen-R\&B}}$ determined from van der Poel nomograph and complex modulus E^*_{bitDSR} derived from DSR tests at 0.1-30 Hz and at +20 °C for different paving grade binders. A linear regression was found between same parameters at same conditions for different modified bitumens. In both cases the coefficient of correlation decreases with increasing test frequency.
Viscosities versus shear stress and shear rate of PmB 25/55-65 modified bitumen and rubber bitumen (KSGB) are close together.
The MSCR test data can be fitted to rheological parameters of Burgers model with acceptable low relative error for paving grade bitumens. The relative error is higher for modified bitumens and hard bitumens. It was shown that the modification increases of shear viscosity η_3 corresponding to dashpot D3 of the Burgers model.
The testing of zero-shear viscosity was investigated in creep mode using method CEN/TS 15325:2008 method. The test stress τ_0= 5-10 Pa was selected for tested modified binders for creep test, with extended 13-18 hours test time. The zero-shear viscosity can be determined with regression analysis using Burgers model from creep and recovery test data. The creep and recovery test can be performed in shorter time than the creep test. ZSV values obtained from creep tests and from creep and recovery tests are close together for paving grade bitumens but larger differences were found for modified bitumens.

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[8] Multiple Stress Creep Recovery (MSCR) test of Asphalt Binder Using a Dynamic Shear Rheometer (DSR), *AASHTO Designation: TP 70-07, June, (2007)*