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TEMPERATURE DEPENDENCE OF THE DYNAMIC VISCOSITY OF ASPHALT - POLYMER MIXTURES

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Abstract

The present work dealt with the influence of polymer additives upon the reological properties of asphalt. Six sample mixtures of 400 g asphalt with 16 g polymers (Kraton D1116, SBS rubber Europren Sol T 6205; rubber crumb 0820A; ethylene-vinylacetate - EVA Elvax 350; polyethylene - PE Bralen SA200 and atactic polypropylene -APP) were prepared and their dynamic viscosity measured using IKA-ULTRA-TURRAX® T 18 in the temperature range 103°C to 137°C. Dynamic viscosity of asphalt and different modified mixtures was experimentally determined and then calculated using three mathematical models (exponential function, polynomial function and modified Walter's equation). The relative difference between the experimental and calculated values of dynamic viscosity was determined and then used as applicability (suitability) criterion of the above mathematical models. Modified Walter's equation has been found suitable to calculate the dynamic viscosity at delimited temperatures.

Keywords: asphalt; polymers; viscosity; temperature dependence.

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1. Introduction

The viscosity of liquid hydrocarbon mixtures plays an important role in technological practice. It is used in the calculation of heat transfer coefficients, pressure drops, hydraulic properties and reological and tribological evaluations. Often the viscosity of medium and high viscosity of mixtures must be adjusted at particular temperature. Liquid viscosity is one of most difficult properties to calculate with accuracy. No single method is satisfactory for all temperatures and viscosity ranges. The comparison of experimental results with calculated values of hydrocarbon fraction viscosity at particular temperatures may help to select a suitable method.

Asphalt binders, including polymer-modified binders (PMBs), are normally handled and applied at elevated temperatures. The recommended temperature of application will be one at which the binder is sufficiently fluid to enable pumping, spraying, or mixing with aggregate. The maximum viscosity of a binder at its recommended application temperature is commonly specified to ensure that normal handling and application is possible with standard equipment.

In order to formulate the dependence of asphalt-polymer mixtures viscosity on temperature, a number of relations were proposed. The logarithmic values of dynamic or kinematic viscosity that constitute the temperature exponential function are used most frequently [1,2.,3]:

$$\log(\mu + c) = a(t + 273{,}15)^b \qquad (1)$$

where t is the temperature [grad], μ is the dynamic viscosity [mPa s], *a* and *b* are the correction coefficient for the variation of viscosity with temperature, *c* is the viscosity correlation factor.

Coefficients *a* and *b* can be determined from the experimental viscosities of the hydrocarbon compounds or mixtures at temperatures above softening point. Walter used equation (2) for kinematic viscosity [4] and Mehrotra [5] for dynamic viscosity. Both equations are used in logarithmic form with various values of the *c* coefficient. In Walter's equation, the *c* value is in the interval from 0.6 up to 0.8, with smaller values for higher viscosty.

$$\log(\log(\upsilon + c)) = a + b\log(t + 273{,}15) \qquad (2)$$

where t is temperature [grad], υ *is* kinematic viscosity [$mm^2\ s^{-1}$], a and b are straight line coefficients, c = 0,6 for higher viscosity. The straight line and consequently its coefficients are determined starting from two known viscosity values.

2. Experimental

The following device was used for the purposes of mixing asphalt and polymers samples: IKA-ULTRA-TURRAX® T 18 basic (motor and stand) with dispergation adapter S 18N-19G. Samples were mixed in container with asphalt (400 g) and polymer (16 g). The asphalt was heated to the temperature of 150°C. The above temperature was maintained during 1 hour of mixing, using the heater Schott. During the mixing the temperature was kept within the range of ± 3 °C. Mixing speed was set to level 4 (15500 rev./minute).

Homogenization of asphalt N4326 with polymers was performed under the same conditions, with the following substances. Six mixtures have been prepared, of asphalt N4326 with the following substances: 4% SBS rubber EUROPREN SOL T 6205; 4% rubber crumb 0820A; 4% ethylene-vinylacetate (EVA) ELVAX 350; 4% polyethylene (PE) BRALEN SA200; and 4% atactic polypropylene (APP). The following values have been established after the homogenization: softening point; penetration; and breaking point. Table 1 includes the results of the measurements:

Table 1. Characteristics of samples

Sample	Polymers	Softening point [°C]	Penetration at 25°C [0.1mm]	Fraass breaking point [°C]
1	SBS rubber Kraton D1116	76	88	-15
2	SBS rubber Europren Sol T 6205	72	94	-21
3	Rubber crumb 0820A	46	87	-10
4	Ethylene-vinylacetate Elvax 350	53	92	-10
5	Polyethylene (PE) Bralen SA200	47	86	-12
6	Atactic olypropylene (APP)	55	60	-5
7	Asphalt N4326 without polymer	44	122	-13

The RV2 HAAKE device measured the dynamic viscosity. We used the measurement MV-I system with the stator diameter of 21mm, a rotor diameter of 20.04 mm and a height of 60 mm [6]. For tempering we used the cryostat MK70 WEB WLW Prüfgeräte Werk.

3. Results and discussion

3.1. Dependence of sample viscosity on temperature

Dynamic viscosity was measured under the conditions that have been defined in the experimental part of this paper. The measurements were performed in the temperature range 95 – 142°C. Shear stress was measured within this temperature range, at pre-set shear gradient. The rotor speed revolutions change from 1 rpm to 512 rpm. The maximum pre-set revolutions were limited by the magnitude of the shear stress that was the subject of the measurement. The measuring had to be stopped after the revolutions being set the deflection of the pointer exceeded the range of the recorder scale. The viscosity of samples was established on the basis of dependence of shear stress on shear gradient. Table 2 shows the results of the measurements.

At various temperatures the viscosities of asphalt and asphalt mixtures with polymers get hyperbolic dependence. Table 2 and Figure 1 indicates different influence of polymers to the viscosity of mixtures.

As to extent of influence, specific polymers may be classified into three groups. The SBS rubbers had the maximum impact on the growth of viscosity. The viscosity of those systems remarkably differed from that of the original asphalt. Polymer additives Elvax, Bralen, and atactic PP had approximately the same impact. The viscosity of asphalt N4326 differed only minimally from that of its mixture with rubber crumb. The minimum difference of the viscosities indicates that the rubber crumb does not penetrate into the homogenous asphalt phase during the mixing and that it acts as a filler rather than asphalt properties modifier.

Table 2. Dynamic viscosity of samples at different temperatures.

Sample 1		Sample 2		Sample 3		Sample 4		Sample 5		Sample 6			
Asphalt N4326		Asphalt N4326		Asphalt N4326		Asphalt N4326		Asphalt N4326		Asphalt N4326		Asphalt N4326	
4%-Kraton		4%Europren		4%Rubber		4% Elvax		4%Bralen		Atactic-PP		Original	
t [°C]	μ [mPa.s]	t [°C]	μ [mPa.s]	t [°C]	μ [mPa.s]	t [°C]	μ [mPa.s]	t [°C]	μ [mPa.s]	t [°C]	μ [mPa.s]	t [°C]	μ [mPa.s]
103.0	3786	103.0	2381	103.2	1504	103.5	1779	102,8	1945	103.5	1877	100.5	1817
107.0	3059	107.5	1897	106.0	1269	108.0	1472	108.0	1456	106.0	1653	105.0	1285
111.0	2468	112.0	1688	111.0	944	112.5	1112	112.0	1179	110.5	1314	109.0	1019
117.0	1810	116.0	1507	115.0	740	116.5	883	116.5	909	111.6	1252	113.0	799
122.0	1354	121.5	1219	119.5	586	122.0	671	121.0	728	117.8	851	117.0	630
125.5	1159	125.5	1074	124.5	466	127.5	529	127.0	526	122.4	652	120.0	529
129.5	957	129.5	872	130.0	336	131.5	448	132.0	404	129.5	462	127.5	354
133.0	817	134.5	750	133.6	292	132.9	430	134.7	362	134.8	370	131.0	300
137.5	701	138.5	652	135.5	260	137.5	373	137.5	322	141.4	255	134.0	249

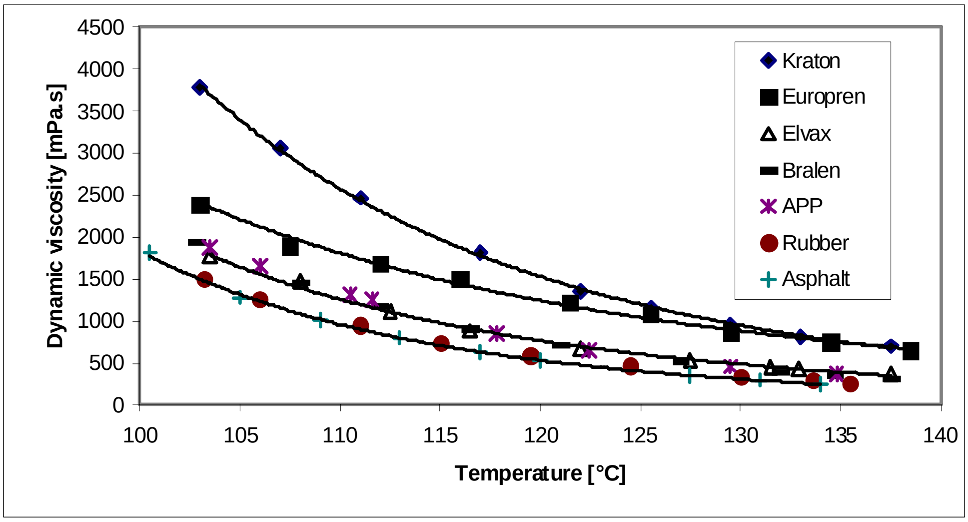

Figure.1 Dependence of sample viscosity on temperature

The determination of asphalt viscosities at higher temperatures is connected with the problem of fluctuating measurement temperatures. Therefore, measurement at stabilised system temperature was advantageous. Calculation of sample viscosity at required temperature necessitates knowledge of dependence of the sample viscosity on temperature $\mu = f(t)$. Equations that are described in literature can be used for the calculation. Such a calculation requires application of known values of viscosities at two temperatures. Various methods were used to compare the adequacy of the sample viscosity calculation at various temperatures:

a) exponential dependence $\mu = a.t^b$ (3)

b) polynomial of the third degree $\mu = A_0 + A_1.t + A_2.t^2 + A_3.t^3$ (4)

c) Equation (2) for higher values of dynamic viscosity

$$\log(\log(\mu + 0{,}6)) = A + B.\log(t + 273{,}15) \quad (5)$$

3.2 Comparison of experimentally measured asphalt viscosities with those calculated from the exponential function.

By regression analysis coefficients *a*, *b* were obtained from viscosities measured at various temperatures (Table 1) for the exponential equation $\mu = a.t^b$. (Table 3).

Table 3. Coefficients of Equation (3), comparison of calculated and experimental viscosities.

	Asphalt N4326						
	4%-Kraton	4%Europren2	4%Rubber	4% Elvax	4%Bralen	Atact-PP	Asphalt N4326
a	4.0513E+15	1.1520E+12	1.1796E+16	5.5891E+14	8.3885E+15	1.6440E+16	6.6937E+16
b	-5.9748	-4.3122	-6.4036	-5.7053	-6,2754	-6,4152	-6,7813
R^2	0.9964	0.9996	0.9995	0.9996	0.9995	0.998	0.9994
T [°C]	122.0	121,5	119.5	122.0	121.0	122.4	120,0
μ *-calc*	1387	1181	588	698	713	664	532
μ *-exp*	1354	1219	586	671	728	652	529
$\lvert\Delta\mu\rvert$	33	37	2	27	15	12	3
r [%]	2,44	3,04	0.34	4.02	2,06	1.84	0,57

r is the relative uncertainty of the experimental value of dynamic viscosity

Equations gained were used to calculate viscosities of individual samples at the temperature around 120°C. The calculated viscosities were compared with the experimental result that had been gathered at the same temperatures as the calculated ones. The difference $\lvert\Delta\mu\rvert$ between the experimental viscosities and the calculated ones was evaluated and expressed as percentage of the basis. The value of the experimentally established viscosity at the given temperature was taken as the basis. In calculations with exponential equation, the maximal difference between the experimental viscosity and the calculated one was 4.02%.

3.3. Comparison of experimentally measured asphalt viscosities with those calculated from polynomial function $\mu = A_0 + A_1.t + A_2.t^2 + A_3.t^3$.

Coefficients A_0, A_1, A_2 and A_3 obtained by linear regression from the viscosities measured at different temperatures (Table 1), for the polynomial equation (4) are given in Table 4.

Table 4. Coefficients of equation (4) and comparison of calculated viscosities with experimental values.

	Asphalt N4326						
	4%-Kraton	4%Europren	4%Rubber	4% Elvax	4%Bralen	Atact-PP	Asphalt N4326
A_0	105772	49364	69352	43925	60628	49697	99448
A_1	-2145.5	-1015.9	-1533	-849.1	-1262.9	-988.39	-2291.3
A_2	14711	7.2756	11.4670	5.4867	8.9144	6.6394	17.802
A_3	-0.0339	-0.0179	-0.0289	-0.0117	-0.0212	-0.0150	-0.0465
R^2	0.9999	0.9948	0.9995	0.9985	0.9998	0.9993	0.9987
t [°C]	122.0	121.5	119.5	122.0	121.0	122.4	120
μ - calc	1422	1231	593	753	776	681	589
μ - exp	1354	1219	586	671	728	652	529
$\lvert\Delta\mu)\rvert$	32	12	7	82	48	29	60
r [%]	2,36	0,98	1,19	12,22	6,59	4,45	11,34

Evaluation of the applicability of polynomial function to calculate the viscosity of samples at 120°C has been realized under the same conditions as exponential dependence. The difference between calculated and experimental values of viscosity applying polynomial function is relatively higher with comparison to the difference found with exponential one. When polynomial function is applied results, the percentage differences between the experimental viscosity and the calculated one moved within the range of < 0.98% to 12.22%>.

3.4 Comparison of experimentally measured asphalt viscosities with calculated ones after Walter equation $\log(\log(\mu + 0,6)) = A + B.\log(t + 273,15)$

Walter equation allows for calculating viscosity at a chosen temperature t_3 by interpolation from two experimental values of viscosities that have been measured at two temperatures t_1 and t_2. The impact of the width of the temperature interval t_1 and t_2 on the precision of the value of the viscosity calculated was the subject of verification of adequacy of use of Walter equation. In the Visual Basic program, two experimental points (t_1 , μ_1) and (t_2 , μ_2) were transformed to the values (t_1+273,15 , W_1) and (t_2+273,15 , W_2). Solution of the equations W_1 = A + B.log (t_1+273,15); W_2 = A + B.log (t_2+273,15) led to calculations of the coefficients A and B. The known coefficients were the basis of calculation of Walter function W_3 = A + B.log(t_3+273,15). The calculated value W_3 was transformed to the viscosity value μ_3 for the temperature value t_3, that is within the temperatures range < t_1; t_2 >. Table 5 indicates the results of comparison of the experimental values of viscosities with the calculated ones, from the temperatures interval < t_1; t_2 >, Δt = 10°C.

Tab. 5. Calculation of viscosity at the temperature t_3 by interpolation from two experimental values of viscosities measured at two temperatures t_1 and t_2 differing by 10°C.

	Asphalt N4326 4%-Kraton		Asphalt N4326 4%Europren		Asphalt N4326 4%Rubber		Asphalt N4326 4% Elvax		Asphalt N4326 4%Bralen		Asphalt N4326 4% Atact-PP		Asphalt N4326 Original	
	t [°C]	*μ* [mPa.s]	*t* [°C]	*μ* [mPa.s]	*t* [°C]	*μ* [mPa.s]	*t* [°C]	*μ* [mPa.s]	*t* [°C]	*μ* [mPa.s]	*t* [°C]	*μ* [mPa.s]	*t* [°C]	*μ* [mPa.s]
Point 1 exper.	117.0	1810	116.0	1507	115.0	740	116.5	882.7	116.5	908.6	117.8	908.2	117.0	630.1
Point 2 exper.	125.5	1159	125.5	1074	124.5	466	127.5	529	127.0	526	129.5	514	127.5	354
Point 3 exper.	122.0	1354	121.5	1218	119.5	587	122.0	671	121.0	728	121.0	717	120.5	529
Point 3 calcul.	122.0	1387	121.5	1235	119.5	591	122.0	649	121.0	714	121.0	720	120.5	516
│Δμ)│		33		17		4		22		14		3		13
r [%]		2.4		1.4		0.8		1.1		2.0		0.4		2.5

The evaluation of the differences between the experimental and calculated viscosities was performed in the same way as in the previous cases. For Walter equation, percentage differences between the experimental viscosity and the calculated one were within the range from <0.4% to 2.5%>. When viscosity at the chosen temperature (t_3 ≅120°C) was calculated while wider temperature range was used (Δt =20°C), the differences between the calculated viscosity values and the experimental ones increased. The difference range was from 0.6% to 7.1%.

4. Conclusions

Measurements at the temperatures from 100°C to 142°C have shown that all polymer additives in the mixture with asphalt increase the viscosity of the original asphalt.

As to the magnitude of influence, specific polymers may be classified into three groups. The SBS rubbers have had the greatest impact on the viscosity growth. The viscosity of that system remarkably differed from the viscosity of the original asphalt. The polymer additives Elvax, Bralen, Atactic PP have demostrated lower impact. The viscosity of asphalt N4326 differs only minimally from its mixture with rubber crumb. The minimum difference between the viscosities indicates that the rubber crumb (d ≈ 1mm) does not have the properties of a modifier, but it acts as asphalt filler.

The precision of dynamic viscosity measurements for asphalt is given by repeatability in the standard method [6]. According to this standard method, the difference between the two test results obtained by the same operator with the same apparatus under constant operation conditions on identical test material would, in long run, in normal and correct operation of the test method, exceed 5% only in one case out of twenty.

From this point of view it is possible to consider suitability of the dependence μ = f(t), gained from experiments, for calculations of dynamic viscosity at a chosen temperature.

Evaluation of the measurements of dynamic viscosity by means of exponencial dependence $\mu = a.t^b$ has shown acceptable agreement between the experimental and calculated dynamic viscosities. The drawback of this procedure seems to be a rather great number of experimental measurements taken at various temperatures.

For polynomial dependence $\mu = A_0 + A_1.t + A_2.t^2 + A_3.t^3$ the differences between the experimental and calculated values seem to be the greatest. In some cases the values of relative uncertainty of measurements are greater than what the normative method allows for.

Expressed by Walter equation, the dependence of viscosity of the modified asphalts on temperature has shown just small differences from <0.4% to 2.5%> between the experimental viscosity and the calculated one. The Walter equation is adequate for calculations of viscosities at certain temperatures. Interpolation in the range of Δt = 10°C leads to the best fit of the calculated viscosities to the experimental results. Interpolation in a wider temperature range (Δt = 20°C) has brought worse results, but still such results that have met the requirement of reproducibility of dynamic viscosity measurements, in agreement with standardised method.

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