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EFFECTS OF MIXING CONDITIONS ON PROPERTIES OF ASPHALT MODIFIED BY CRUMB RUBBER

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Abstract

This work includes the study of mixing conditions on properties of asphalt modified by crumb rubber. For measuring were used five-level, three-factor design of experiment. Determinate were three dependent variable – softening point, penetration at 25°C and dynamic viscosity at 135°C. The monitored variables for mixing were concentration of crumb rubber (wt.%), mixing temperature (°C) and mixing time (min.). The results allowed us to estimate the influence of mixing conditions on modified asphalt samples and also showed that the most suitable criteria is dynamic viscosity.

Keywords: design of experiment; concentration of crumb rubber; mixing temperature and mixing time.

1. Introduction

Rising energy and material costs and increase awareness of the problem of emissions in the production of modified asphalt mixture highlighted the potential benefits of the process of reducing the temperature of asphalt mixture preparation. In this connection, the properties of the asphalt are improving by additives. There are many additives used as asphalt mixes modifiers. Among of these additives the crumb rubber is often used with regards to environmental and material costs. The process of crumb rubber interaction with asphalt affects the composition of asphalt, type of rubber, rubber particle size, temperature, time and energy of mixing [1-4]. The practical application and laboratory monitoring properties of bitumen modified by crumb rubber requiring determine the conditions for preparing such mixtures. The parameters of the preparation of these compositions are the exception of the asphalt modifier comprising an amount of a rubber crumb applied, time and temperature of mixing [5-10]. Publications show that the method of preparing asphalt modified by GD is important for maintaining good repeatability of qualitative parameters of prepared samples. Table I present mixing conditions of crumb rubber with asphalt, in some literature sources.

Table I. Mixing conditions for preparing of asphalt mixture

Crumb rubber		Mixing conditions		Cited from
Particle dimensions [mm]	Used concentration [% m/m]	Temperature °C	Time (min)	
Inexplicit	Inexplicit	160-180	180	[5]
0.4	15- 20	-	-	[6]
0.1 - 0.3	25	200	120	[7]
0 - 0.5	1	170	120	[8]
0 - 0.6	0.5 - 1.5	180	45 - 120	[9]
0.8	15 -20	190 - 200	45	[10]

Overview of the conditions for mixing the crumb rubber in asphalt gives impulse to determine the impact of independent variables to the appropriate values to assess the quality and homogeneity of the resulting mixture. The basic independent variables used in the evaluation of modified bitumen are the softening point, penetration and dynamic viscosity. Selection of appropriate intervals of independent variables is related to the preparation conditions of road mixtures which are determined in qualitative standards.

The aim of the study was verified the influence of conditions on effectiveness of asphalt mixing with crumb rubber and to define suitable criteria for evaluation of rubber-asphalt mixtures. The samples were measurement according to five-level, three-factor design of experiment. The chosen dependent variables were: determination of softening point, penetration at 25°C and dynamic viscosity at 135°C. And the independent variables were concentration of crumb rubber (wt.%), mixing temperature (°C) and mixing time (min.) The results allowed us to estimate the influence of mixing conditions on modified asphalt samples and also showed that the most suitable criteria is dynamic viscosity.

2. Materials and methods

In this study was used asphalt from refinery Slovnaft, a.s, Slovak Republic. Approximately 10 kg of asphalt sample was heated at 135° C for 4 hours. Melted asphalt was mixed and poured to galvanized metal containers. The asphalt samples of 200 g ± 5 g were prepared. The crumb rubber granules (CR) provided by V.O.D.S., a.s., Košice, Slovak Republic were separated by screening. In experiments the fraction with crumb rubber particles about the size 0.841-0.595 mm (20-30 mesh) was used. Equipment IKA-ULTRA-TURRAX® T18 with extension S 18N-19G was used for mixing crumb rubber with bitumen. The operating speed was 15 000 rpm. The samples were melted at temperature 135 ± 1°C and were poured off to test containers. Each sample contained 200 ± 5 g of bitumen and was mixed with crumb rubber. Each dose of crumb rubber was calculated to the actual mass of bitumen. The mixing process was carried out at the appropriate temperature with accuracy ± 3°C, during the relevant period of time with accuracy ± 1 minute. After the mixing process each sample was analyzed according to the European standards for the determination of softening point [11], penetration [12], and viscosity [13]. The penetration measurements were performed on Analis P731 equipment. Penetration measurements were performed according to European standard [12] and three repeated injections were done with each sample. Softening point was measured by using the Petrotest Instrument (SUR). The standard prescribes two parallel measurements. The instrument RV2 HAAKE was used for the measurement of dynamic viscosity. The stator of MV II system was tempered by heating circulator Julabo MA-4. For determination of standard deviation of viscosity measurements the Brookfield standard B200 was used. The average dynamic viscosity 218.3 mPa.s of the standard B200 was calculated from eight experiments and it reached value ±1.3%.

3. Results and discussion

The experiments were planned using 5-level type of basic scheme 2^3, level of reliability α was 0.05 and arm was 1.681. For experiments three measurement factors were selected: τ-time of mixing (min.), t - temperature of mixing (°C) and c – concentration of CR (% wt.) The changes of dynamic viscosity (η) were monitored at 135°C, changes of penetration (P) at 25°C. The determination of softening point (t_{KG}) was performed using the ring and ball method. Asphalt without CR was evaluated with the same tests as asphalt with CR. The results of tests for asphalt without CR are shown in Table II.

Table II. Properties of asphalt without crumb rubber

Viscosity at 135°C (mPa.s)	Penetration at 25°C (p.u.)	Softening point RB (°C)
264	104	44.3

3.1. The design of experiment

Intervals of individual factors have been identified under the terms of the preparation of rubber-asphalt viable experience in the laboratory and in practice. The fair values of factors were converted into coded data according to the relationship 1:

$$x_{i,R} = x_{i,Rs} + I_i * x_{i.k} \qquad (1)$$

where: $x_{i,R}$ is the factual value of the i-th factor encoded by the corresponding level; $x_{i,Rs}$ is the mean factual value of the i-th factor corresponding to the coded layer 0; I_i is the step of the i-th factor; $x_{i,k}$ is corresponding coded level of the i-th factor.

In the table III are listed factual values of the independent variables corresponding to the coded factor in the conditions of the design of experiment.

Table III. Factual and coded values of the independent variables in the design of experiment

Symbol	Factor - variable	-1.681	-1	0	1	1.681
X_1	τ- Time (min.)	10.4	30.5	60.0	89.5	109.6
X_2	t - Temperature (°C)	119.7	132.0	150.0	168.0	180.3
X_3	c – Concentration of CR (wt.%)	2.14	3.30	5.00	6.70	7.86

In accordance with the data in Table III were randomly assigned order made experiments at real values of the independent variables corresponding to the coded values. Table IV shows the coded values of the independent variables, the order and the experimental results obtained in the designed experiment.

Table IV. Coded values of independent variables, order and results of dependent variables obtained in the designed experiment

No. of experiment	Coded variables			Order of experiment	t_{RB} [°C]	P_{25} [0,1mm]	η_{135} [mPa.s]
	X_1	X_2	X_3				
1	-1	-1	-1	4	42.9	94	265.5
2	1	-1	-1	1	45.8	100	276.8
3	-1	1	-1	7	44.9	99	280.0
4	1	1	-1	10	45.8	83	288.1
5	-1	-1	1	13	44.8	97	336.9
6	1	-1	1	16	45.1	96	330.0
7	-1	1	1	18	46.9	76	418.3
8	1	1	1	20	45.1	90	382.4
9	1.681	0	0	2	44.7	92	270.0
10	1.681	0	0	17	46.1	79	348.6
11	0	1.681	0	8	45.3	99	285.6
12	0	1.681	0	11	44.5	87	462.8
13	0	0	1.681	14	41.2	88	255.8
14	0	0	1.681	5	44.4	86	410.0
15	0	0	0	19	44	85	372.5
16	0	0	0	6	46.2	89	377.1
17	0	0	0	9	45.8	99	408.3
18	0	0	0	12	42.3	92	357.0
19	0	0	0	15	44.5	89	403.8
20	0	0	0	3	43.4	86	380.7

Processing of the results of the experiments as described in the literature [14] allowed the calculation of the coefficients of the regression model. Dependences of selected variables (t_{RB}, P_{25}, η_{135}) on factors X_1, X_2, X_3 were obtained in the form:

$$Y = b_0 + b_1X_1 + b_2X_2 + b_3X_3 + b_{11}X_1^2 + b_{12}X_1X_2 + b_{13}X_1X_3 + b_{22}X_2^2 + b_{23}X_2X_3 + b_{33}X_3^2 \quad (2)$$

Where: Y is selected dependent variable (t_{RB}, P_{25}, η_{135}) and independent variables are $X_1 = \tau$, $X_2 = t$, $X_3 = c$.

In the table V. are coefficients b_0, b_i, b_{ii} and b_{ij} for dependences of selected dependent quantities $Y = t_{RB}$, $Y = P_{25}$ a $Y = \eta_{135}$.

Table V. Coefficients b_0, b_i, b_{ii} and b_{ij} of dependences $Y= F(X_1, X_2, X_3)$

	Y = t_{RB} [°C]	Y = P_{25} [mm]	Y = η_{135} [mPa.s]
b_0=	44.341	89.886	383.686
b_1=	0.341	-1.381	7.964
b_2=	0.202	-4.334	33.511
b_3=	0.577	-1.492	45.153
b_{11}=	0.534	-0.860	-29.083
b_{12}=	-0.513	-0.875	-4.025
b_{13}=	-0.663	2.875	-7.775
b_{22}=	0.357	1.795	-6.116
b_{23}=	0.013	-1.875	13.500
b_{33}=	-0.386	-0.329	-20.731

The processing of results acquire of variance values S1 and S2 representing a linear and a quadratic part of the equations (2), wherein in component S2 are also included interaction effects of the individual factors. On the basis of the criterion F is possible to confirm the significance of the contribution of linear and / or quadratic factors. To meet the statistically significant effect of linear and / or quadratic factor must be true:

$$F_{S1} > F_{krit} \quad \text{resp.} \quad F_{S2} > F_{krit} \qquad (3)$$

The output of statistical processing is also a component of variability SLF characterizing the adequacy or inadequacy of the regression equation. In this assay it is verified the adequacy of application of a mathematical model. The criterion for testing the inadequacy of the component SLF is its comparison with critical value. If the relevant criterion is less than the critical value (SLF < SLFkrit) equation sufficiently accurately describes the experimental situation. On the contrary, if this criterion is higher (SLF > SLFkrit) equation is not sufficiently precise. In table VI is a comparison of the mentioned criteria with their critical values.

Table VI Comparison of criteria S_{LF}, F_{S1} and F_{S2} with their critical values

i	$t_{RB} = F(X_1, X_2, X_3)$		$P_{25} = F(X_1, X_2, X_3)$		$\eta_{135} = F(X_1, X_2, X_3)$	
	F_{Si}	F_{krit}	F_{Si}	F_{krit}	F_{Si}	F_{krit}
1	1.035	5.409	4.074	5.409	38.647	5.409
2	1.102	4.950	1.073	4.950	8.302	4.950
SLF	0.492	5.050	2.207	5.050	3.445	5.050

In evaluating of the effect of a particular independent variable (a particular factor) it is useful to compare the absolute values of regression coefficients with the corresponding critical values. If the absolute value of the coefficient (b_0, bi, b_{ii}, b_{ij}) is greater than the corresponding critical value (bk0, bki, bkii, bkij) its influence can be considered as significant and relevant independent variable affects to outputting dependent variable. The critical values of the coefficients of the equations: $Y=F(X_1, X_2, X_3)$ are shown in Table VII.

Table VII. Critical values of coefficients in questions: $Y = F(X_1, X_2, X_3)$

	t_{KG} [°C]	P at 25°C [mm]	η at 135°C [mPa.s]
bk0=	1.539	5.304	20.430
bki=	1.021	3.520	13.558
bkii=	0.995	3.429	13.205
bkij=	1.334	4.598	17.711

3.2. Results evaluation of determination of softening point on the variables X_1, X_2, X_3

Using criteria SLF, S_1 a S_2 are assessed dependence of the softening temperature of asphalt mixture with crumb rubber at selected independent factor. It has been found that the criterion of SLF (Table VI) is less than the critical value (SLF < SLF_{krit}). According to this criterion equation adequately describes the experimental situation. However, a statistically significant impact on both the linear and quadratic components of factors has not been established. This claim is based on comparison of variances S_1, S_2 depending $t_{KG} = F(X_1, X_2, X_3)$ to their critical values. In both cases, the condition statistically significant effect is not satisfied. The coefficients of dependence $t_{KG} = F(X_1, X_2, X_3)$ is shown in Table 5. Dependence built on the coded variables: factor X_1 (mixing time), X_2(mixing temperature) and factor X_3 (concentration of crumb rubber) can be evaluated by the value of the regression coefficients. A comparison of the absolute values of regression coefficients of each factor with the relevant critical values if softening point denies the effect of all three variables (Table V and Table VI).

The failure of experiments cannot be explained by neglecting the impact of external factors on the dependent variable t_{RB}. Independent variable intervals were wide enough to not overlap experimental errors. Method of t_{RB} determination but insufficiently captured the really existing changes in the softening point with changes of factors. Determination of the softening point is likely to be affected by the random occurrence of the crumb rubber particles at the periphery of the sample poured into the ring. The standard EN 1472 gives also the consistency values for results of polymer modified asphalts. This standard provides a different conformity for softening point determination of modified and unmodified asphalts. Increased levels of repeatability are mainly for bitumen having higher softening point. In round five trials of modified bitumen with high softening points was observed average repeatability of 2.2 ° C. Consistency of results for bitumen modified by crumb rubber is not supported by round robin tests. This may be the reason for unreliable results of experiments.

3.3. Results evaluation of determination of penetration at 25°C on the variables X_1, X_2, X_3

In the case of penetration depending on the selected factors, it was found that the criterion SLF is below the critical value (Table VI). The equation P = F adequately describe the experimental situation, but the influence of linear and quadratic components of factors is statistically insignificant. This fact is clear from the comparison values S1 and S2 with critical values.

A comparison of the absolute values of individual regression coefficients in the formula $P_{25} = F(X_1, X_2, X_3)$ with the corresponding critical value will determine the impact of individual factors. The factor b_0 alone is not decisive in the impact assessment. Consideration of other factors showed that only the coefficient b_2 is in absolute value greater than the critical value. To confirm the linear effect of temperature on penetration value that is not enough. It shows the dependence of the measured and calculated under the same penetration encoded factors. The correlation coefficient R^2 obtained by linear regression of the measured and calculated values of penetration was equal to 0.5379. This confirms the discrepancy between the model equations and the experimental determination of penetration.

Evaluation of experiments for penetration as a function of selected factors showed that output variable as irrelevant for the assessment of incorporating crumb rubber in asphalt.

Determination of penetration is not adequately tested for samples gum-asphalt. The accuracies data for determination of penetration listed in European Standard EN 1426 are valid for asphalt without modifiers. In the case of a mixture of bitumen with crumb rubber is determination of the penetration affected by the presence of the crumb rubber particles e.g. accidental contact of the needle with solid particles. Pending the round robin tests is necessary for these mixtures data repeatability and reproducibility of measurements of penetration regarded as merely indicative.

3.4. Results evaluation of determination of viscosity at 135°C on the variables X_1, X_2, X_3

Dependence of dynamic viscosity on selected factors has value of variability components SLF less than the critical value, and therefore equation sufficiently accurately describes the experimental variability situation (Table VI). As well as components that represent linear variability (S1) and the quadratic variability (S2) satisfies the conditions of statistical significance, the value of S1 and S2 are greater than the critical value. This confirms that the studied factors affecting the dependent variable - dynamic viscosity.

The absolute values of the coefficients b_0, b_2, b_3, b_{11}, b_{33} in the equation depending on the dynamic viscosity of the encoded factors are above the corresponding critical values (Table V and Table VII). For values of other coefficients is manifested reverse inequality when compared to the critical values. Therefore, the viscosity is slightly affected by mixing time in linear term, by interaction effects of factors X1X2, X1X3 X2X3 and by temperature in the quadratic term. The equation of dynamic viscosity depending on the selected factors was not recalculated by omitting coefficients with negligible influence. It resulted from a comparison of the quadratic model with the linear model. Experimental measurements of viscosity were compared with the viscosities calculated using the quadratic respectively of the linear model. Regression coefficient *R* of equation $\eta_{\text{experimental}} = F\ (\eta_{\text{calculated}})$ was in quadratic model equal 0.94. For linear model was equal 0.78. It turned out preferably in the equation leave regression coefficients although their impact is negligible.

A comparison of the absolute values of individual coefficients in the linear term indicates the dominant effect of the concentration of crumb rubber and mixing temperature. Negative coefficients in the quadratic term in equation contribute to achieving maximum of the function. This fact was clearly demonstrated during the viscosity of the mixing time.

Formulation of viscosity by equation with three factors provides an illustration of a three-dimensional graph of changes in viscosity as a function of two variables and with the third factor having constant value. Such type of viscosity dependencies on the concentration of crumb rubber in asphalt at three different temperatures and with constant mixing time are in figure 1.

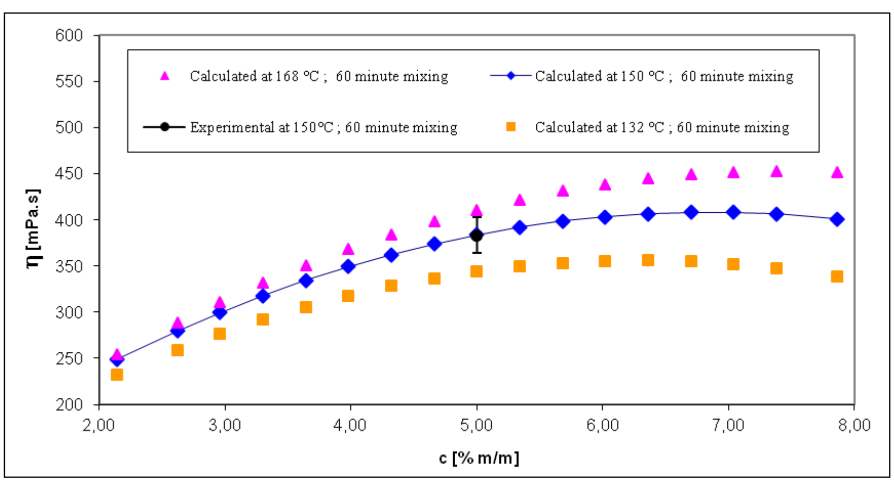

Figure 1. Calculated changes of viscosity depending on concentration of CR, with different mixing temperature and with the same mixing time (60 min.)

A good overview provides a graphical illustration of viscosity calculated for one variable at constant but different values of two variables. Completion the plot by measuring points allows us to assess overlap calculated curves with experiment (Figure 2).

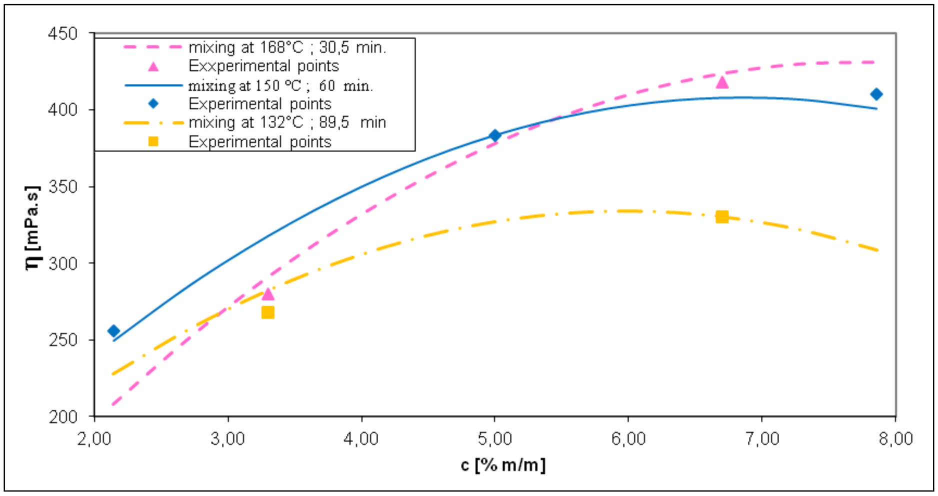

Figure 2. Viscosity as a function of the concentration of crumb rubber in asphalt calculated at different values of temperature and different mixing times in confrontation with data points

The dominance of the concentration effect of crumb rubber and the temperatures at the mixing is illustrated in Figure 3. The low concentration of crumb rubber limits the increase in viscosity. By the high concentration of crumb rubber, the viscosity significantly increases also at lower temperature and the time of stirring.

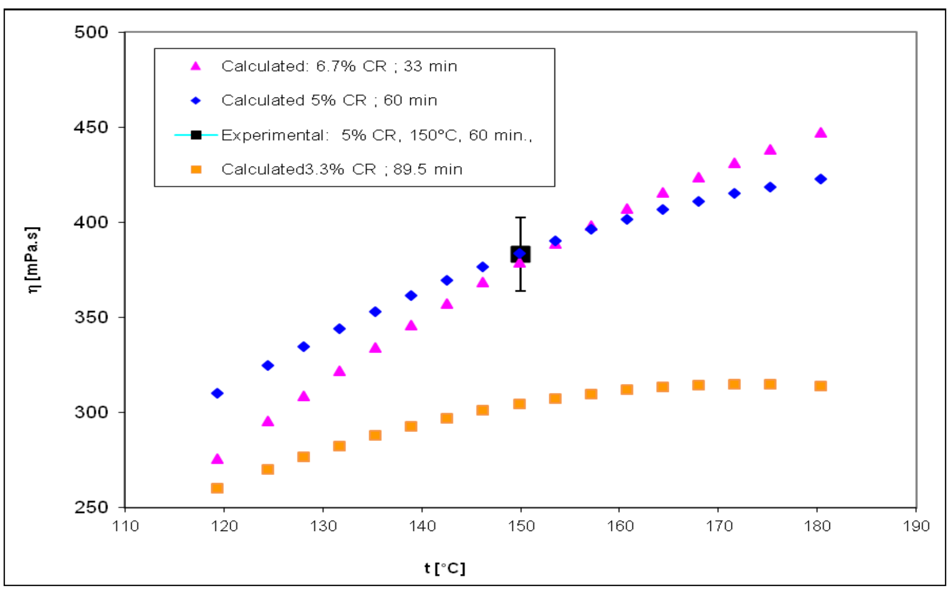

Figure 3. The viscosity as a function of temperature calculated at different values of mixing time and at various concentrations of crumb rubber in asphalt

Although not all the coefficients have significant impact to the independent variable, the equation $\eta_{135} = F(X_1, X_2, X_3)$ satisfactory fitted the function and preferably follows the experimental results. Evaluation of the dependence permits the inference that they were properly determined intervals of factors, factors have a real impact on the independent variable. Dynamic viscosity proved to be a useful parameter for monitoring changes in incorporation of crumb rubber in asphalt.

4. Conclusions

The results obtained by design of experiments have shown that the determination of softening point and penetration are not suitable for assessment of the effect of mixing conditions crumb rubber in asphalt. Absolute values of coefficients in the equations of dependency of output quantities on the selected factors not confirmed real impact.

The absolute values of the coefficients in the equation of dependence of dynamic viscosity on the encoded factors X 1, X 2 and X 3 are greater than the relevant critical values for the case of factor crumb rubber concentration and the factor of temperature at stirring. Comparison of individual coefficients in the linear terms of the equations also showed the dominant effect of the concentration of crumb rubber and mixing temperature to change the viscosity of the samples.

The designed experiment with a dynamic viscosity as dependent variable in the interval selected factors confirmed the appropriateness of viscosity to evaluate the effectiveness of incorporating crumb rubber in asphalt.

References

[1] Mashaan NS, Karim MR Investigating the rheological properties of crumb rubber modified bitumen and its correlation with temperature susceptibility. Materials Research. 2013; 16(1): 116–127.

[2] Shen J, Amirkhanian S, Xiao F, Tang B. Influence of surface area and size of crumb rubber on high temperature properties of crumb rubber modified binders, Construction and Building Materials. 2009; 23(1):304–310.

[3] Abdelrahman MA, Carpenter SH. Mechanism of interaction of asphalt cement with crumb rubber modifier, Transportation Research Record. 1999; 1661: 106–113.

[4] Jeong KD, Lee SJ, Amirkhanian SN, Kim KW. Interaction effects of crumb rubber modified asphalt binders, Construction and Building Materials. 2010; 24(5): 824–831.

[5] Xiang L,Cheng J, Que G. Microstructure and performace of crumb rubber modified asphalt. Construction and Building Materials. 2009; 23(12):3586-3590.

[6] Júnior AFA, Battistelle RA, Bezerra BS, Castro R. Use of scrap tire rubber in place of SBS in modified asphalt as environmentally correct alternative for Brazil. Journal of Cleaner Production. 2012; 33: 236-238.

[7] Tan Y,Guo M, Cao L, Zhang L. Performance optimatization of composite modified asphalt sealant based on rheological behavior. Construction and Building Materials 2013; 47: 799 – 805.

[8] López-Moro FJ, Moro MC, Hernández-Olivarez F, Witoszek-Schultz B, Alonzo-Fernando M. Microscopic analysis of the interaction between crumb rubber and bitumen in asphalt mixtures using the dry process. Construction and Building Materials. 2013; 48: 691-699.

[9] Moreno F, Rubio MC, Martinez-Echevaria MJ. Analysis of digestion time and the crumb rubber percentage in dry-process crumb rubber modified hot bituminous mixex. Construction and Building Materials 2011;25(5): 2323-2334.

[10] Liu H, Chen Z, Wang W, Wang H, Hao P. Investigation of the rheological modifi-cation medification mechanism of crumb rubber modified asphalt (CRMA) contai-ning TOR additive. Construction and Building Materials. 2014; 67, Part B, : 225 – 233.

[11] EN 1427 Bitumen and bituminous binders - Determination of the softening point - Ring and Ball method, 2015.

[12] EN 1426 Bitumen and bituminous binders - Determination of needle penetration

[13] EN 13302. Bitumen and bituminous binders - Determination of dynamic viscosity of bituminous binder using a rotating spindle apparatus, 2010.

[14] Alexy, P., Základy plánovania a vyhodnocovania experimentov. STU, Katedra plastov a kaučuku, 1988.

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