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EVALUATION OF THE POSSIBILITY OF REDUCTION OF HIGHLY MODIFIED BINDERS TECHNOLOGICAL TEMPERATURES

M. PUCULEK¹, A. LIPHARDT², P. RADZISZEWSKI,³

The article presents results of research on the possibility of reduction of technological temperatures of highly modified binders by using as an additive a fluxing agent of plant origin, developed at the Warsaw University of Technology. The work presents the results of dynamic viscosity tests within the temperature range of 90°C to 180°C for original binders and binders aged using the RTFOT method, modified by adding a fluxing agent of plant origin. On the basis of dynamic viscosity test results, process temperatures for production and compaction of mineral-asphalt mixes with the binding agents being analyzed. Moreover, characteristics of binders were assessed, which define their behavior under high technological temperatures on the basis of multiple stress creep recovery (MSCR) tests.

The research results obtained indicate that it is possible to reduce the process temperatures of mineral-asphalt mixes with highly modified binders by liquefying the binder with an additive of plant origin while retaining high resistance to permanent deformations. The MSCR test results prove that mineral-asphalt mixes containing binders highly modified with additives of plant origin can meet the requirements for extremely high traffic loads.

Keywords: highly modified binder, process temperatures, dynamic viscosity, MSCR test, Bio-flux

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1. INTRODUCTION

Highly modified asphalt binders used for construction of structural layers of flexible road surfaces require use of high process temperatures in the process of production, integration and compacting of mineral-asphalt mixes made using such binders. Experience in use of highly modified binders indicates that process temperatures required for binders of this kind are higher in comparison with traditional polymer-asphalts. Application of high temperatures, particularly in the course of production of the mineral-asphalt mix, results in the risk of excessive aging of the binder and may lead to degradation of its polymer content. At present, there is a number of methods used to reduce the process temperatures, including application of chemical and organic additives, waxes and the frothing process [1]. Particularly worth attention are additives included in the category of the so-called green technologies, such as vegetable oil esters.

Application of additives that reduce asphalt viscosity within the process temperature ranges also requires an assessment of impact of these additives on characteristics of the binding agent within the range of their technological temperatures. It is recommended that additives that are used cause no deterioration of operating parameters of the binder, such as resistance to permanent deformation, fatigue or low-temperature cracking.

Highly modified asphalts, the so-called HiMA, are a special type of polymer-asphalts. In most cases, SBS type polymers (styrene-butadiene-styrene) are used in the amount of 2÷4%. In highly modified asphalts, special elastomers are used, and their content exceeds the limit of 6% m/m. The polymer phase, unlike in the case of traditional polymer-asphalts, constitutes the continuous phase in the highly modified binder. As a result, a continuous polymer network is developed in the binder, which substantially changes the functional features of asphalt and the mineral-asphalt mix layers, at the same time causing no significant problems in production and integration [2].

Highly modified asphalts have above-standard characteristics, such as: very good resistance to rutting [3], resistance to water and frost, resistance to cracking under low temperatures [4], high fatigue strength [5, 6, 7]. These asphalts are dedicated in particular for surfaces characterized by increased traffic loads, substructures of high fatigue strength and asphalt layers characterized by high resistance to low temperatures. They are also very useful in construction of long-lived road surfaces, designed to last 30 to 50 years [8].

Nowadays, the dominant road construction technology is that of hot-laid asphalt surfaces [9, 10]. Moreover, promotion of environmental protection, reduced consumption of natural resources

and energy, as well as elimination of emission of harmful compounds to the atmosphere has become popular all over the world. At the same time, surfaces should be durable and modern. Therefore, it is necessary to introduce innovative and environment-friendly road construction technologies [11].

In order to reduce asphalt viscosity, and thus reduce the process technologies for production and decomposition of mineral and asphalt mixes, liquefied asphalts are used. Liquefied asphalt is obtained by mixing asphalt with a petrochemical or carbochemical solvent, usually containing a substantial quantity of aromatic hydrocarbons. Hardening of asphalt in the surface results from evaporation of the volatile solvent, which pollutes the air and contributes to the greenhouse effect [12]. Production of environment-friendly liquefied asphalts is possible thanks to use of liquefiers of plant origin. It has been proven that vegetable oils and fatty acid esters of these oils can be used as substitutes for aromatic solvents used in the conventional liquefied asphalts. In this case, hardening of asphalts liquefied with oils and/or fatty acid esters takes place through oxidizing polymerization, that is, crosslinking of unsaturated fatty acids as a result of reactions with oxygen and the catalyst. This reaction is catalyzed by metal salts, most of all, organic salts of cobalt, manganese and zircon. As a result, we obtain eco-friendly liquefiers, which increase their viscosity over time to reach the value similar to viscosity of asphalt while reducing the output viscosity, which enables mixing of asphalt with aggregate at a reduced temperature [12]. Bioflux is one of these liquefiers. This liquefier has been patented at the Warsaw University of Technology as the product of oxidization of fatty acid ester of rapeseed oil in the presence of a metallic catalyst and an organic peroxide. Salts of organic acids of cobalt are used as the metallic catalyst, and the polymerization promoter is cumene peroxide. Combined with asphalt, Bioflux reduces its viscosity, which allows for reduction of process temperatures and production of a mineral-asphalt mix in a warm environment [13, 14].

Research has been conducted at the Warsaw University of Technology in order to assess the impact of Bioflux on rheological properties of binders. On the basis of the results, it was found that even small quantities of Bioflux (1.25% and 2.5%) reduced the consistency of asphalt, while greater quantities (3.25% and 5.0%) liquefy the binder. It was found that adding of Bioflux in the quantity of 5.0% results in reduction of the softening temperature by 13–15°C and results in a substantial decrease in the shear modulus value [14]. Depending on the needs, Bioflux is added in the quantity of 1% to 5%.

In research on traditional polymer-asphalts, it was shown that application of vegetable liquefiers resulted in reduction of process temperatures and enhanced workability of mineral-asphalt mixes [14]. No research has been conducted so far with regard to reduction of process temperatures of highly modified binders using liquefying additives of plant additives.

The article presents the research results concerning the possibility of reduction of process temperatures of asphalt PMB 45/80-80 by adding Bioflux. Asphalt PMB 45/80-80 is among the most widely used binders in road construction among all highly modified binders produced in Poland. High temperature properties of these binders were also assessed due to the risk of excessive reduction of binder rigidity with the liquefying additive under high temperatures.

2. MATERIALS AND TESTING METHODS

2.1 MATERIALS

Research was conducted using a binder belonging to the category of highly modified asphalts 45/80-80, and, for the sake of comparison, a polymer-asphalt 45/80-55. The compared binder was selected to have a similar hardness range. The basic properties of asphalt binders being analyzed have been presented in table 1.

Table 1. Main characteristics of the tested binders

Binder property	PMB 45/80-80	PMB 45/80-55
Penetration in 25°C [0,1mm]	75	71
Softening point [°C]	95,5	57,8
Elastic recovery [%]	96	76

Under laboratory conditions, Bioflux was obtained by mixing methyl esters of fatty acids of rapeseed oil with the cobalt catalyst, with the addition of cumene hydroperoxide at the ratio of 98.8% of ester of rapeseed oil fatty acids, 1.0% of cumene peroxide and 0.1% of cobalt. Afterwards, the mix was subjected to oxidation with oxygen from the air at the temperature of 20°C. Oxidation took place in a reactor (fig. 1), which ensures a large area of contact between the gas and the liquid raw material. Oxidation of Bioflux takes 2 hours at the air flow rate of 500 [l/kg·h].



Fig. 1. Ester oxidation reactor

2.2 TESTING METHODS

The binders were modified with Bioflux after heating them for 1 hour to the temperature making it possible to mix it with the additive (160°C PMB 45/80-55 and 180°C 45/80-80). The weighted portion of Bioflux was added to the container with heated asphalt using a syringe and then mixed for 60s. Afterwards, test samples were prepared using such modified binder. Binders were modified by adding 1%, 2%, 3%, 4% m/m of Bioflux, and reference samples were also prepared, containing no additive.

Due to requirements related to the research procedures applied, the binders analyzed were also subjected to process aging using the RTFOT method, and the aging process took place after the binders had been modified by adding Bioflux.

Both of the asphalt binders analyzed were subjected to assessment of dynamic viscosity using a rotational Brookfield viscometer with a measurement system of coaxial cylinders. Dynamic viscosity was measured at 90°C, 110°C, 135°C, 160°C and 180°C, both for original binders and those aged using the RTFOT method.

Due to the existing risk of deterioration of operating properties under high temperatures due to liquefying of binders and the associated reduction of resistance of the surface to permanent deformation, it was also necessary to conduct an assessment of operational properties under high temperatures. For aged binders, a multiple stress creep recovery test (MSCR) was conducted at the temperature of 60°C, which served as a basis for determination of the percentage recovery R and the flexibility module J_{nr} at shearing stress of 0.1 and 3.2 kPa.

Process temperatures were determined by assuming the optimum viscosity levels, at which it is possible to properly surround the aggregate particles with binder, amounting to $0.2 \text{ Pa}\cdot\text{s}$, as well as proper compaction of the mineral-asphalt mix within the range from $2 \text{ Pa}\cdot\text{s}$ to $20 \text{ Pa}\cdot\text{s}$ and pumping over of the binder, amounting to $2 \text{ Pa}\cdot\text{s}$. It should be noted that the binder pumping and mix production temperatures were determined for non-aged binders, while the initial and final compaction temperatures were determined for binders aged using the RTFOT method.

3. RESULTS

3.1. DYNAMIC VISCOSITY AND TECHNOLOGICAL TEMPERATURES

Fig. 2 presents the selected results of dynamic viscosity tests for highly modified asphalt PMB 45/80-80 - original and aged using the RTFOT method with varying content of Bioflux additive.

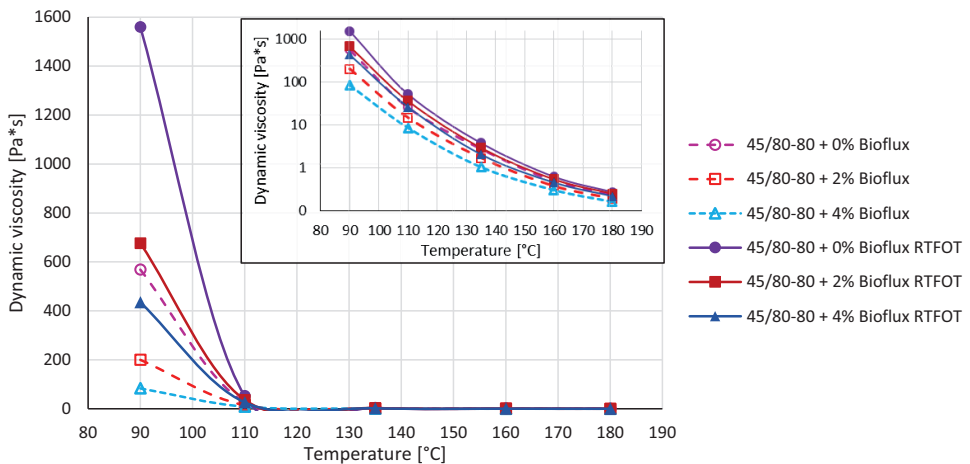


Fig. 2. Dynamic viscosity of original and RTFOT aging PMB 45/80-80 binders

The results of tests presented in fig. 2 indicate that adding of Bioflux results in high advantageous reduction of dynamic viscosity of aged and non-aged highly-modified asphalt within the lower range of process temperatures of 90°C - 110°C . Reduction in viscosity under higher temperatures above 110°C is characterized by lesser dynamics, but is significant from the perspective of impact of adding Bioflux on process temperatures of production of mineral-asphalt mix containing this binder.

Fig. 3 presents the average viscosity reduction as a percentage for the asphalts analyzed upon modification of Bioflux content by 1%, characterizing sensitivity of the binder being analyzed to changes in Bioflux content. It was determined as the ratio of asphalt viscosity containing x-1% of Bioflux to Bioflux content of x% at the specified test temperature.

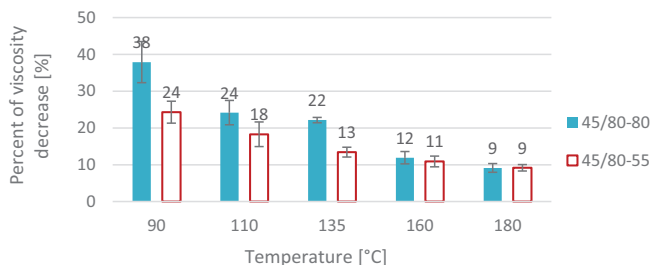


Fig. 3. Percent of dynamic viscosity decrease

Table 2 presents the results of statistical analyzes of the impact of binder type on viscosity decrease. In the statistical analyzes the following levels of significance of the impact of variables was used:

- $p \leq 0,01$ – very significant impact of variable,
- $0,01 < p < 0,05$ – significant impact of variable,
- $0,05 < 0,1 <$ – less significant impact of variable,
- $p > 0,1$ – insignificant impact of variable.

Table 2. Results of statistical analyzes of the impact of binder type on viscosity decrease percent

Temperature	Dependent variable	Qualitative factor – binder type	
		p	significance
90	Percent of viscosity decrease [%]	0,000	very significant
110		0,001	very significant
135		0,000	very significant
160		0,073	less significant
180		0,392	insignificant

On the basis of results presented in fig. 3 and table 2, it can be stated that the most significant differences in sensitivity to changes in additive content between the polymer-asphalt PMB 45/80-55 and highly modified asphalt PMB 45/80-80 have been observed in the lower process temperature

range of $90^{\circ}\text{C}\div 135^{\circ}\text{C}$. In the higher process temperature range of $160^{\circ}\text{C}\div 180^{\circ}\text{C}$, both binders show similar sensitivity to changes in Bioflux content and the differences are not statistically significant.

Fig. 4 presents a breakdown of mix production temperatures for both binders analyzed at five Bioflux content values.

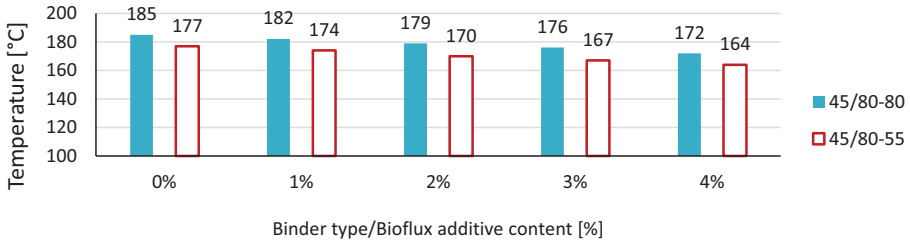


Fig. 4. Asphalt mixture mixing temperature

Both in the case of polymer-asphalt 45/80-55 and highly-modified asphalt 45/80-80, it was found that increasing of Bioflux content by 1% resulted in reduction of enclosing temperature on the average by about 3°C . Achievement by highly-modified asphalt PMB 45/80-80 of the enclosing temperature equivalent to polymer asphalt PMB 45/80-55 without the additive requires using Bioflux at the level of approximately 3% m/m. Research has shown that the enclosing temperature for a highly modified binder at all Bioflux content values is about $8\text{-}10^{\circ}\text{C}$ higher in comparison with the traditional polymer asphalt.

Fig. 5 presents a breakdown of binder pumping temperatures for both binders analyzed at five Bioflux content values.

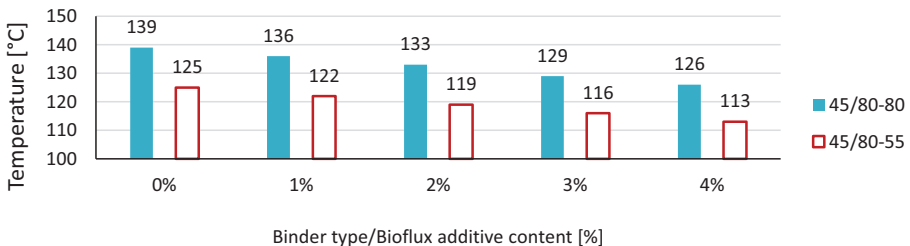


Fig. 5. Binder pumping temperature

On the basis of results presented in fig. 5, it was found that increase in the Bioflux content by 1% results in reduction of the pumping temperature on the average by about 3°C both in the case of

binder PMB 45/80-80 and PMB 45/80-55. Achievement by highly modified asphalt of the pumping temperature equivalent to that of the traditional polymer asphalt without the additive requires using of about 4% m/m of Bioflux. The process temperatures determined for binder PMB 45/80-80 are higher on the average by 13-14°C in comparison with binder PMB 45/80-55.

Fig. 6 presents a breakdown of initial compaction temperatures for both binders analyzed at five Bioflux content values.

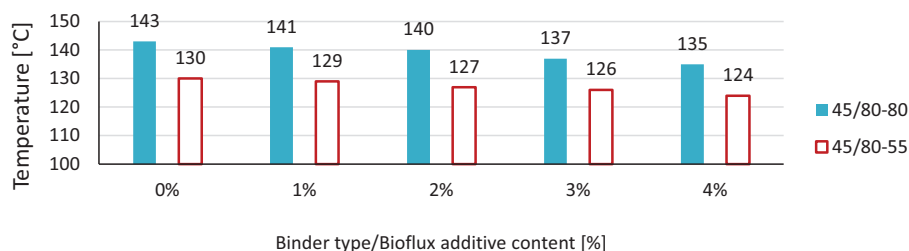


Fig. 6. Compaction start temperature

In relation to the initial compaction temperature of the mix containing polymer-asphalt 45/80-55, it was found that increasing of Bioflux content by 1% resulted in temperature reduction on the average by about 1-2°C, and in the case of highly-modified asphalt, by 1-3°C on the average. Research showed that the initial compaction temperature for the highly modified binder in the case of all Bioflux content values is higher by approximately 11-13°C in comparison with the traditional polymer-asphalt.

Fig. 7 presents a breakdown of final mix compaction temperatures for both binders analyzed at five Bioflux content values.

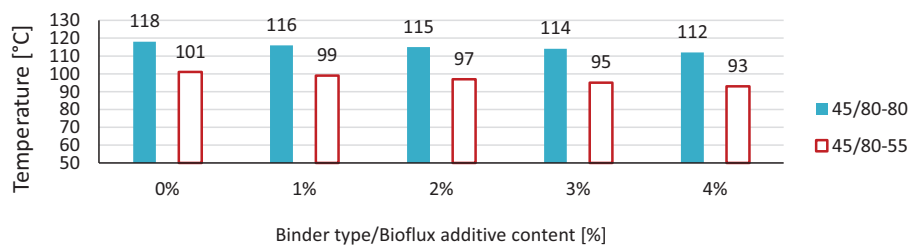


Fig. 7. End of compaction temperature

In relation to the final compaction temperature of the mix containing polymer-asphalt 45/80-55, it was found that increasing of Bioflux content by 1% resulted in temperature reduction on the average by about 1-2°C, and in the case of highly-modified asphalt, by 1°C on the average. Research showed that as for the final compaction temperature, highly modified binder was characterized by somewhat lower sensitivity to viscosity reduction in comparison with the traditional polymer-asphalt. Research has shown that the final compaction temperature for a highly modified binder at all Bioflux content values is about 17-19°C higher in comparison with the traditional polymer asphalt.

Summing up, it can be stated that the differences in process temperatures between the binders analyzed are greater at lower temperatures and lesser at higher temperatures, respectively; at the same time, at higher temperatures, effectiveness of the additive in reduction of process temperatures was higher, which is particularly significant in the context of the process of production of the mineral-asphalt mix.

It should be taken into account that a special SBS polymer is used for production of highly modified asphalts, which behaves differently from typical polymers; therefore, experience from trial sections should be taken into account in the assessment of process temperatures.

2.2. MULTIPLE STRESS CREEP RECOVERY TEST

Fig. 8 presents the results of the multiple stress creep recovery tests, the percentage recovery $R_{3,2}$ fig. 8a and the elasticity modulus J_{nr} fig. 8a, determined at shear stress of 3.2 kPa.

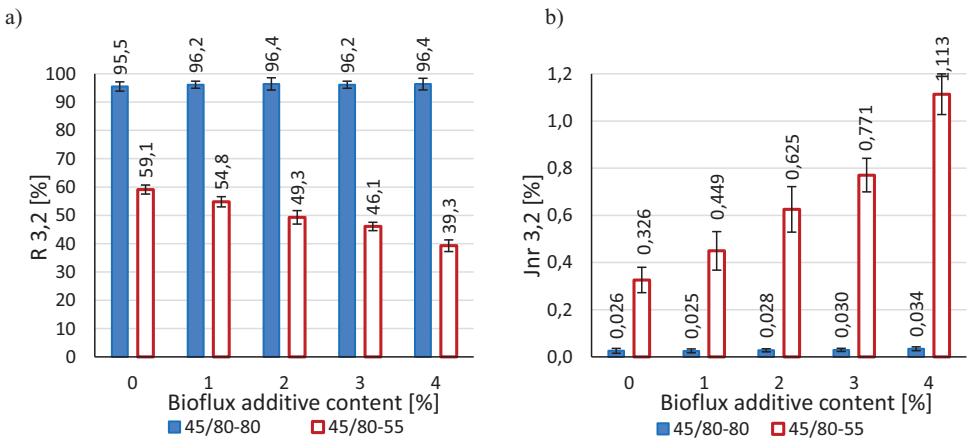


Fig. 8. Comparison of MSCR test results: a) J_{nr} , b) R

Table 3 presents the results of statistical analyzes of the impact of Bioflux content on $R_{3,2}$ and $Jnr_{3,2}$ parameters.

Table 3 Results of statistical analyzes of the impact of Bioflux content on R and Jnr parameters

Binder type	Dependent variable	Qualitative factor – Bioflux content	
		p	significance
45/80-80	$R_{3,2}$	0,781	insignificant
45/80-55		0,000	very significant
45/80-80	$Jnr_{3,2}$	0,633	insignificant
45/80-55		0,000	very significant

Analysis of results presented in fig. 8.a. and table 3 has shown that application of Bioflux additive in the amount of 1% to 4% does not cause a statistically significant change of the percentage recovery R in highly-modified asphalt PMB 45/80-80, and the received values of the R parameter are close to the same level of approximately 96%. In the case of polymer-asphalt PMB 45/80-55, on the other hand, there is a statistically significant decrease in the value of parameter R as the Bioflux content increases. Also in the case of the elasticity modulus Jnr (fig. 8.b.), no significant impact of increase of the liquefier content on this parameter was found for highly modified asphalt. On the other hand, in the case of polymer-asphalt PMB 45/80-55, there is a significant increase in the value of the elasticity modulus as the Bioflux content increases. The differences indicated in behavior of the binders analyzed should be associated with the continuous polymer phase, present in the asphalt, which is not broken despite application of the liquefying agent, ensuring the proper level of elasticity and resistance to accumulation of permanent deformations unlike the traditional polymer-asphalt, in which the polymer does not generate a continuous phase.

As a summary, it can be stated that application of Bioflux additive in a highly modified binder PMB 45/80-80 does not lead to a risk of deterioration of its elasticity or excessive accumulation of permanent deformations.

5. CONCLUSIONS

The research conducted has shown that highly-modified asphalt PMB 45/80-80 is characterized by substantially higher viscosity in relation to polymer-asphalt of comparable hardness PMB 45/80-55, particularly within the temperature range of 90°C – 110°C. This may lead to the necessity to apply higher process temperatures, creating a risk of excessive binder rigidity and

degradation of polymer, moreover, causing difficulties in the course of technological processes. It has been shown that application of Bioflux liquefying agent allows to a certain extent for reduction of viscosity, and thus of process temperatures of highly modified asphalt. At the same time, application of Bioflux has not led to deterioration of properties of highly modified asphalt within the range of high technological temperatures, typical for the MSCR test.

The following general conclusions have been drawn:

- addition of 3% of Bioflux allows for reduction of the temperature for preparation of the mix with highly modified asphalt to the level equivalent to production of the mix containing traditional polymer asphalts;
- the most significant differences in sensitivity to changes in additive content between the polymer-asphalt PMB 45/80-55 and highly modified asphalt PMB 45/80-80 have been observed in the lower process temperature
- in the case of highly-modified asphalt, the MSCR test did not show any negative effect of addition of Bioflux in the amount of 1% to 4% m/m upon R and Jnr parameters, contradictory to polymer-asphalt PMB 45/80-55, for which statistically significant deterioration of elasticity was observed, as well as increased susceptibility to permanent deformations as the Bioflux content increased;
- in the lower process temperature range, the highly modified binder is characterized by beneficially higher dynamics in viscosity change as a function of test temperature in comparison with the traditional polymer-asphalt, which leads to the conclusion that mineral-asphalt mixes will be characterized by better workability;

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OCENA MOŻLIWOŚCI OBNIŻENIA TEMPERATUR TECHNOLOGICZNYCH LEPISZCZY WYSOKMODYFIKOWANYCH

Keywords: highly modified binder, technological temperatures, dynamic viscosity, MSCR test, Bio-flux (słowa kluczowe: lepiszczce wysokomodyfikowane, temperatury technologiczne, lepkość dynamiczna, badanie MSCR, Bio-flux)

SUMMARY:

Lepiszczta asfaltowe wysokomodyfikowane wykorzystywane budowy do warstw konstrukcyjnych podatnych nawierzchni drogowych wymagają stosowania wysokich temperatur technologicznych w trakcie procesów wytwarzania, wbudowania i zagęszczania mieszanek mineralno-asfaltowych wytworzonych z ich użyciem. Doświadczenia w stosowaniu lepiszczki wysokomodyfikowanych wskazują, że temperatury technologiczne wymagane dla tego rodzaju lepiszczki są nawet o 20°C wyższe niż dla tradycyjnych polimeroasfaltów. Stosowanie wysokich temperatur, szczególnie w trakcie procesu wytwarzania mieszanki mineralno-asfaltowej stwarza ryzyko nadmiernego postarzenia lepiszczki jak również może prowadzić do degradacji zawartego w nim polimeru. Obecnie istnieje szereg metod umożliwiających obniżenie temperatur technologicznych m.in. stosowanie dodatków chemicznych, organicznych, wosków oraz procesu spieniania. Stosowanie dodatków obniżających lepkość asfaltów w zakresie temperatur technologicznych wymaga także oceny wpływu zastosowania tych dodatków na właściwości lepiszczki w zakresie temperatur eksploatacyjnych. Wskazane jest stosowanie dodatków niepowodujących pogorszenia parametrów eksploatacyjnych lepiszczki takich jak odporność na deformacje trwałe, zmęczenie czy spękania niskotemperaturowe.

W artykule przedstawiono wyniki badań nad możliwością obniżenia temperatur technologicznych lepiszczki wysokomodyfikowanej poprzez zastosowanie opracowanego w Politechnice Warszawskiej dodatku upłynniającego pochodzenia roślinnego. W pracy przedstawiono wyniki badań lepkości dynamicznej w zakresie temperatur od 90°C do 180°C dla lepiszczki oryginalnych oraz starzonych metodą RTFOT zmodyfikowanych dodatkiem upłynniającym pochodzenia roślinnego. Na podstawie wyników badania lepkości dynamicznej wyznaczone zostały temperatury technologiczne procesów wytwarzania oraz zagęszczania mieszanek mineralno-asfaltowych z analizowanymi lepiszczkami. Temperatury technologiczne wyznaczono przyjmując optymalne poziomy lepkości, przy których możliwe jest prawidłowe otoczenie lepiszczem ziaren kruszywa, wynoszący 0,2 Pa·s a także prawidłowe zagęszczenie mieszanki mineralno-asfaltowej w zakresie od 2 Pa·s do 20 Pa·s. Ponad to dokonano oceny właściwości lepiszczki charakteryzujących ich zachowanie w wysokich temperaturach eksploatacyjnych na podstawie badań cyklicznego pełzania z odprężeniem (MSCR).

Uzyskane wyniki badań wskazują, że jest możliwe obniżenie temperatur technologicznych mieszanek mineralno-asfaltowych z lepiszczem wysokomodyfikowanym poprzez upłynnienie lepiszczki dodatkiem pochodzenia roślinnego przy jednoczesnym zachowaniu wysokiej odporności na deformacje trwałe. Wyniki badania MSCR dowodzą, że zastosowanie dodatku upłynniającego - Biofluxu, do lepiszczki wysokomodyfikowanych nie powoduje pogorszenia ich właściwości wysokotemperaturowych charakteryzowanych procentowym nawrotem r oraz modulem podatności J_{nr} .

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