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The assessment of the influence of styrene-butadiene-styrene copolymer on the selected rheological properties characterising durability of modified bitumens used in road pavements

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Abstract. The subject matter of the research pertains to the improvement of rheological properties of petroleum bitumens by their modification with SBS (styrene-butadiene-styrene) copolymer. The authors have determined selected rheological properties characterising the durability of modified bitumens used in road pavements. The bitumens were modified in laboratory conditions with modified bitumen concentrate of a known SBS copolymer content of 9%. The result was a binder containing the known percentage of the SBS copolymer of 3%, 4.5% and 6%. Rheological properties of the tested bitumens were determined by the use of a DSR dynamic shear rheometer (in a wide temperature range from 40°C to 100°C) and a ductilometer at 5°C. DSR was used for performing MSCR test to determine the resistance of the asphalt mixture with the SBS-modified binder to permanent deformations in the high temperature range (from 40°C to 82°C). The comparison of the values of the dynamic shear modulus |G*| of all the bitumens tested shows that with a growing content of the SBS copolymer in the tested binder the value of |G*| increases, which may indicate greater resistance to permanent deformation of the asphalt pavement. The MSCR test has shown that the increased use of the SBS copolymer addition in the bitumen translates to decreasing values of the non-recoverable creep compliance J_{nr}. The SBS copolymer accelerates stress relaxation in the bitumen sample, thus increasing pavement resistance to low-temperature cracks. Furthermore, modification reduces the negative impact of ageing on the properties of the binder, manifested by its stiffening and slowdown of relaxation.

Key words: dynamic shear rheometer (DSR), modified bitumen, dynamic shear modulus, relaxation, Multiple Stress Creep Recovery (MSCR).

1. Introduction

Consistently growing traffic loads and the influence of changing climatic conditions prevailing in Poland shorten the service life of asphalt pavements [1, 2]. Increase in heavy traffic is predominantly caused by high demands posed by passenger and freight transport, which contribute to rapid degradation of road pavements. Newly developed pavements are subject to ever higher requirements related to resistance to permanent deformations at high temperature, resistance to cracking at low temperature, resistance to fatigue under recursively repeated loads and resistance to water and frost [1]. Pavement's service life is largely affected by the asphalt mixture's composition and rheological properties of the applied bitumen binder, i.e., cohesion, viscosity, elasticity, stiffness and adhesion to mineral aggregates [3].

One way of improving binder properties is to modify their structure by introducing appropriate modifiers [4, 5]. At present, polymers are most commonly used for this purpose, especially elastomers, which improve the elastic properties of bitumen, limiting the intensive formation of permanent deformations in asphalt pavements and increasing pavement durability [1, 6, 7]. According to SHRP (Strategic Highway Research Program), the rheological properties of modified bitumen are tested using, among others, BBR (Bending Beam Rheometer), ductilometer featuring the option to measure tensile force and DSR (Dynamic Shear Rheometer) [1, 3, 8].

Modifications with styrene-butadiene-styrene block copolymer have a remarkable influence on rheological and physicochemical properties of the asphalt binder [9–11]. This influence depends also on the binder's copolymer content [12, 13]. Increased content of SBS copolymer (in literature the threshold is set at about 6% [13]) is accompanied by a change in the structure and proportions of the binder components. The modifier in the binder becomes a dominant component and forms a polymer network, which significantly changes the binder's properties [12, 14, 15].

Polymer modification is of key importance in the context of the binder's resistance to degradation due to ageing. During the ageing process not only does the binder undergo oxidation, but it is also subject to gradual decomposition and disappearance of the polymer [14, 16, 17]. Zhang and others [18] proved in their research that regardless of the method of ageing, e.g., RTFOT (Rolling Thin Film Oven Test), PAV (Pressure Aging Vessel) or UV (ultraviolet), an increased content of

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SBS copolymer reduces stiffness of the material and improves its operational properties. A positive influence of SBS on ageing processes was also demonstrated by Wang and others [19], and Zhang and others [20]. The possibilities of improving the properties of binders modified with SBS copolymer are currently being tested. In their research, Rasool and others [21] used a combination of SBS with other modifiers, i.e., crumb rubber (also recycled rubber, which will allow for the disposal of harmful waste). Owing to possible financial savings and environmental protection, the possibility of reapplication of the recycled, aged pavement with SBS (*reclaimed asphalt pavement – RAP*) in asphalt mixtures is also being researched [22–25]. This requires the use of appropriate preparations that enhance the properties of the degraded polymer. There is an ongoing study on the composition of substances intended for this purpose and their quantities needed in order to obtain a bitumen pavement featuring parameters that meet the standard requirements.

The most recent scientific articles describing research on bitumen binders modified with SBS copolymer discuss the issue of rheological properties at high temperature and the influence of varied copolymer content in the binder on its output parameters [4, 12, 26]. However, the research on the influence of this modifier on the properties of the binder at low temperature is still a scientific niche, which is gradually, albeit slowly, filled by occasional scientific publications [22, 27].

The aim of the research described in this article was to assess the influence of styrene-butadiene-styrene copolymer on selected rheological properties of road bitumen 50/70, which have an effect on the durability of the pavement during its lifetime. The study concerning the pavement resistance to permanent deformation was conducted by means of the modern MSCR method, and relaxation was observed by means of the modified tensile test carried out using ductilometer. The paper reflects upon the aspect of short-term ageing simulated by RTFOT method and variable copolymer content in the binder with respect to unmodified 50/70 road bitumen. The research has been conducted using SBS copolymer modified bitumen samples, acquired by combining 50/70 bitumen with a 9% SBS concentrate. Most research works described in literature concern bitumen modified with SBS copolymer, obtained directly from a manufacturer (in such cases, the exact copolymer content in modified bitumen is usually unknown). The chosen binder percentages (3%, 4.5%, 6% and 9% of SBS) were selected in order to highlight changes in the binder properties related to the change of its structure and formation of the polymer network in the bitumen binder.

2. The features of the tested asphalt binders

The tests were carried out on 50/70 penetration grade road bitumen, obtained as a result of distillation of Russian crude oil. The bitumen was modified in laboratory conditions by introducing into its structure a bitumen concentrate modified with SBS copolymer. The concentrate with SBS copolymer content of 9%, obtained from 160/220 penetration grade road bitumen, was combined with 50/70 penetration grade bitumen to arrive at the known SBS copolymer contents in the obtained bitumen, i.e., 3%, 4.5% and 6% of styrene-butadiene-styrene copolymer (in relation to the mass of modified bitumen obtained). Bitumen binders were heated to 140°C (190°C in case of SBS copolymer concentrate). Then they were mixed for 5 min using a laboratory mixer at an angular speed of 400 RPM. In order to obtain the determined contents, the mass proportions between the concentrate and 50/70 road bitumen were maintained as 1:2, 1:1 and 2:1, respectively. All tested binders were conditioned in the same manner. During the test, the heating process was controlled, and an attempt was made to prepare a set of studied samples during one heating cycle of the material. Bitumen was submitted to analysis both in its initial state and after a short-term (technological) ageing process simulated by the RTFOT method according to EN 12607-1:2014 "Bitumen and bituminous binders. Determination of the resistance to hardening under the influence of heat and air – Part 1: RTFOT Method".

Table 1 summarises the results for basic properties of the tested bitumens (penetration and softening point) carried out according to the standards EN 1426:2015-08 and EN 1427:2015-08, respectively.

Table 1
The results for penetration determined at 25°C and softening point ($T_{R\&B}$) of the tested asphalt binders before and after the ageing process

Binders	**Penetration before ageing [mm/10]**	**Penetration after ageing by RTFOT method [mm/10]**	**$T_{R\&B}$ before ageing [°C]**	**$T_{R\&B}$ after ageing by RTFOT method [°C]**
50/70	54.6 ± 0.7	37.1 ± 0.5	50.3 ± 0.5	55.4 ± 0.3
3% SBS	63.7 ± 0.5	42.2 ± 0.9	55.0 ± 0.7	56.4 ± 0.3
4.5% SBS	68.5 ± 0.6	44.4 ± 0.8	83.3 ± 0.8	83.8 ± 4.1
6% SBS	72.3 ± 0.4	51.1 ± 0.6	99.0 ± 2.4	90.7 ± 1.5
9% SBS	79.4 ± 0.8	65.7 ± 0.7	104.5 ± 0.7	101.0 ± 0.9

The expanded standard uncertainty ranges have been determined for direct measurements with a confidence level of 95%. The analysis of the results presented in Table 1 shows that asphalt binders were selected so as to obtain bitumens of similar hardness, expressed through penetration values at 25°C (Pen_{25} was obtained in the range from 54.6 to 79.4 mm/10). Therefore, all modified bitumens can be classified as meeting the standard requirements for PMB 45/80-55 (for bitumen with 3% SBS copolymer content) and PMB 45/80-80 (for binders with 4.5–9% SBS copolymer content), available on the Polish market, although they have different percentages of SBS copolymer. The R&B softening point temperature (50.3°C) of the tested unmodified bitumen allows for its classification, in accordance with EN 12591:2010, as type 35/50 or 50/70 bitumen. On the basis of the penetration test, bitumen has been classified as 50/70 (however, the penetration value was within the lower limits of the standard).

3. Methodology of the studies

The main aim of the study was to assess the influence of SBS elastomer content on rheological properties of bitumens modified with SBS copolymer on the basis of tests carried out with dynamic shear rheometer (DSR) and ductilometer, taking into account technological ageing simulated by RTFOT method.

The tests were conducted using Physica MCR 101 dynamic shear rheometer (DSR) at varying temperature, i.e., from 100°C to 40°C, with temperature being reduced by 1°C every 1 min. The test was carried out in accordance with EN 14770:2012, and the application of kinematic (sinusoidal) excitation with a deflection angle of 10 mrad. A sample of bitumen (weight of 0.500 ± 0.01 g) was placed between two parallel plates, with a diameter of Ø25 mm and a gap height of 1 mm.

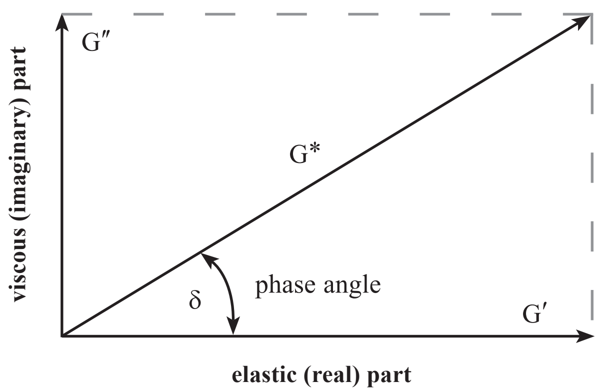

Fig. 1. Graphical interpretation of the complex shear modulus G* (based on [1])

Figure 1 presents in a graphic form the quantities determined with the DSR method under sinusoidal loading, which can be defined as follows [1, 28]:

$$G^* = G' + iG'', \tag{1}$$

where:
- G^* – complex shear modulus equal to τ_{max}/γ_{max} (complex stiffness modulus determined by DSR method under sinusoidal loading), Pa,
- G' – real part of the complex shear modulus, $G' = G^* \cdot \cos(\delta)$, Pa,
- G'' – imaginary part of the complex shear modulus, $G'' = G^* \cdot \sin(\delta)$, Pa,
- i – imaginary unit meeting the equation $i^2 = -1$,
- τ_{max} – maximum value of the shear stress, Pa,
- γ_{max} – maximum value of strain.

$$|G^*| = \sqrt{G'^2 + iG''^2}, \tag{2}$$

where $|G^*|$ – dynamic shear modulus (absolute value of complex shear modulus), Pa,

$$\delta = tg^{-1}\left(\frac{G''}{G'}\right), \tag{3}$$

where δ – phase angle between stress and strain, °.

A complex shear modulus consists of a storage modulus G′, and a loss modulus G″. The former determines the ability to store energy, and the latter to dissipate energy. The complex shear modulus G* is interpreted as the ratio of the maximum shear stress value to the maximum strain value.

The Multiple Stress Creep Recovery (MSCR) test was then performed by the use of a DSR. During the MSCR test, the following phenomena are examined:
- creep of the binder sample – during a 1-second loading
- recovery of the binder sample – during the 9-second unloading time (after relieving the applied load).

The sample was subjected to a constant stress load for 1 second, and then unloaded for another 9 seconds. Ten load/unload cycles were performed at a shear stress value of 3.2 kPa. The test was carried out at eight temperatures ranging from 82°C to 40°C with a value step of 6°C.

During the MSCR test, a deformation change has been recorded in the first and the tenth second of each load/unload cycle. It yields results in form of the irreversible part of the non-recoverable creep compliance J_{nr} (5) and the percent recovery R (4) at the applied stress of 3.2 kPa [29].

$$R = \frac{\sum_1^{10} \varepsilon_r(\tau, N)}{10}, \tag{4}$$

where:
- τ – shear stress, kPa,
- N – number of load/unload cycles,
- ε_1 – strain increase after the first second of the cycle (at the end of the load phase),
- ε_{10} – strain increase after the tenth second of the cycle (at the end of the unload phase).

$$J_{nr} = \frac{\sum_1^{10} J_{nr}(\tau, N)}{10}, \tag{5}$$

where:
- τ – shear stress, kPa,
- N – number of load/unload cycles,
- $J_{nr} = \varepsilon_{10}/\tau$,
- ε_{10} – strain increase after the tenth second of the cycle (at the end of the unload phase).

Out of the results obtained, J_{nr} 3.2 kPa is the most useful parameter for binder classification, as the measure of the binder resistance to permanent deformations.

In the second stage, tests were performed by use of a ductilometer. The tensile test in the ductilometer at 5°C was performed following the instructions of EN 13589:2018-08, with the following modification: upon completion of the tensile procedure, the measuring of tensile force was continued for 20 min to observe relaxation. Brass moulds were filled with different types of the tested binders, then placed in a ductilometer bath and conditioned for 90 min before the start of the test. The test samples were stretched at 50 mm/min until an elongation of 400 mm was achieved. After the tensile test was completed, the force values were recorded for another 20 min in order to plot relaxation curves. The procedure was run for 4 samples of each binder.

Calculations of the deformation work (energy) were performed for all modified bitumens. Work was calculated in accordance with EN 13587:2016-12 as the area under the tensile force diagram in the function of elongation ranging from 200 to 400 mm at 5°C (6).

$$E'_s = E'_{0.4} - E'_{0.2}, \tag{6}$$

where:

τ – shear stress, kPa,

E'_s – deformation work, J,

$E'_{0.4}$ – deformation work needed to obtain an elongation sample equal to 0.4 m, J,

$E'_{0.2}$ – deformation work needed to obtain an elongation sample equal to 0.2 m, J.

4. Analysis of test results

4.1. Tests conducted in the DSR. The tested bitumens are materials with viscoelastic properties. In the case of unmodified 50/70 penetration grade bitumens and the binders with a 3% content of SBS copolymer, the authors observed a regularity that the higher the values of the dynamic shear modulus $|G^*|$, the lower the values of the phase angle δ. In the case of asphalt binders with 6% and 9% SBS copolymer content, small phase angle δ values are obtained, both at very low and high dynamic shear modulus $|G^*|$ values. The highest variation in phase angle δ was observed for 50/70 bitumen and binders with 3% copolymer content. Their values at high temperatures are close to 90° (for 50/70 bitumen), 80° for bitumen with 3% SBS content, presumably featuring properties similar to those of a viscous liquid in the high temperature range. Viscous materials are characterised by the damping factor designated as $\text{tg}\delta \to \infty$ ($\delta = 90°$), and $\text{tg}\delta = 0$ ($\delta = 0°$) for elastic materials; viscoelastic materials have a phase angle with values in the range of $0° < \delta < 90°$. The increase in the content of the SBS copolymer in the bitumen makes the variation in δ values less and less different, regardless of the temperature. As the temperature increases (Fig. 3), the δ value decreases for bitumens with SBS 4.5% and 6% content. An increase in the test temperature in all analysed bitumens causes a decrease in the dynamic shear modulus $|G^*|$, both before and after the technological ageing process (Figs. 2, 4). The greatest difference in dynamic shear modulus $|G^*|$ values was recorded for 50/70 penetration grade bitumen, from 70.51 Pa at 100°C to 57135 Pa at 40°C.

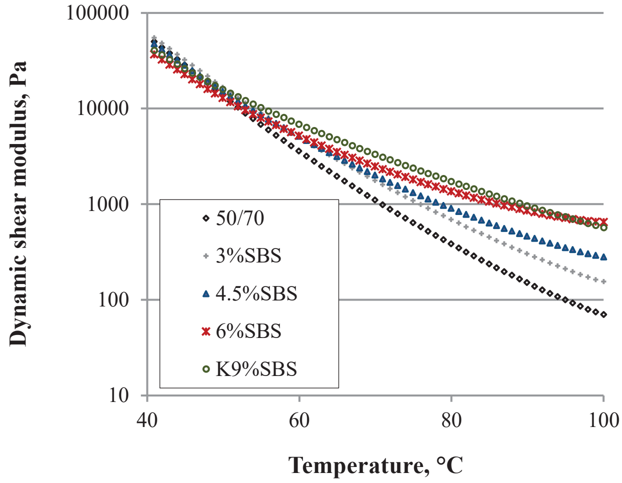

Fig. 2. Temperature dependence of dynamic shear modulus $|G^*|$ for binders not subjected to RTFOT ageing, at a constant angular frequency of 10 rad/s

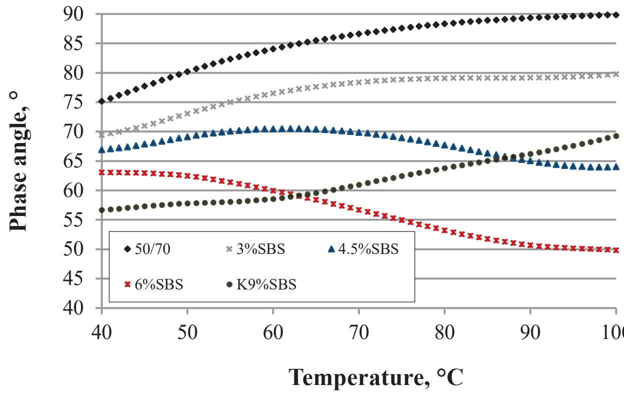

Fig. 3. Temperature dependence of phase angle δ for binders not subjected to RTFOT ageing, at a constant angular frequency of 10 rad/s

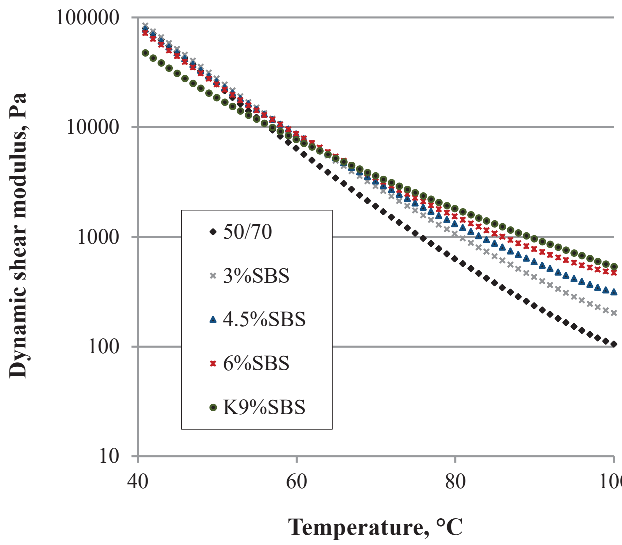

Fig. 4. Temperature dependence of dynamic shear modulus $|G^*|$ for binders subjected to RTFOT ageing, at a constant angular frequency of 10 rad/s

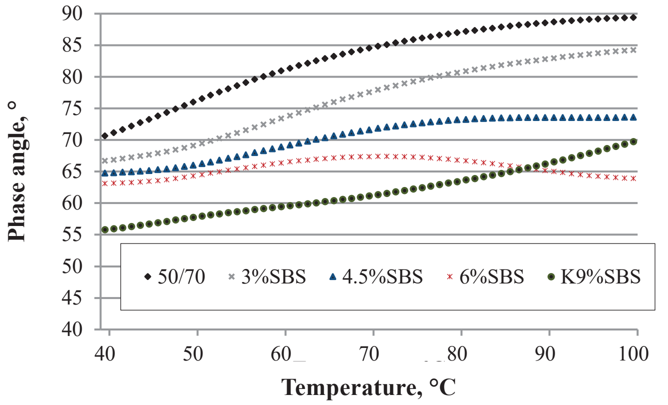

Fig.5. Temperature dependence of phase angle δ for binders subjected to RTFOT ageing, at a constant angular frequency of 10 rad/s

Figures 6 and 7 present Black diagrams for all analysed binders taking into account the RTFOT ageing process. The idea of Black curves is to present the relations of the dynamic shear modulus |G*| in the phase angle δ function on one graph. In the case of 50/70 penetration grade bitumen and the binder containing 3% SBS, increase in |G*| gives a noticeable decrease in the δ angle, while in the case of modified bitumen with a SBS copolymer content of more than 4.5%, a certain regularity can be observed that with an increase in |G*| the δ angle increases until it reaches its maximum at some point, and then continues to decrease. Black curves of the tested asphalt binders make it possible to observe how a higher modifier content in the binder relates to enhanced elastic properties. Only for 50/70 and 3% SBS bitumens does the short-term ageing process reduce the value of the phase angle δ in comparison with bitumens subjected to ageing through stiffening. In the case of highly modified bitumens which have undergone ageing, the δ angle values have increased, which may result from polymer degradation; nevertheless, these values still remain much lower compared to 50/70 bitumen.

In order to better illustrate the share of the viscous and elastic parts in the studied bitumens, the authors have drafted Cole-Cole diagrams, which show the dependence of the G″ loss modulus on the G′ storage modulus (Figs. 8 and 9). In the case of 50/70 bitumen, the proportion of the viscous part

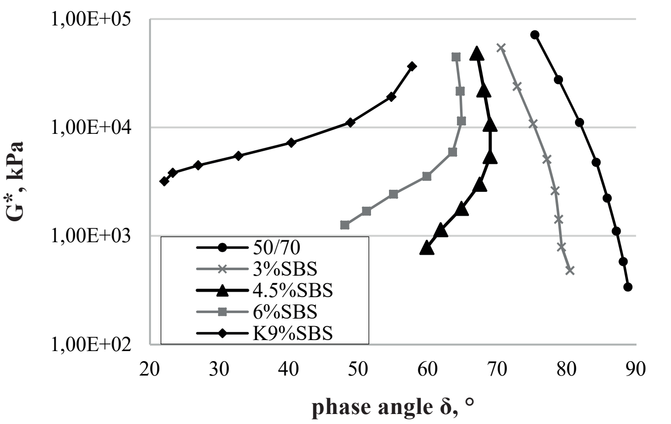

Fig. 6. Black curves of asphalt binders tested at an angular frequency of 10 rad/s in the temperature range of 40–100°C, before ageing by RTFOT method

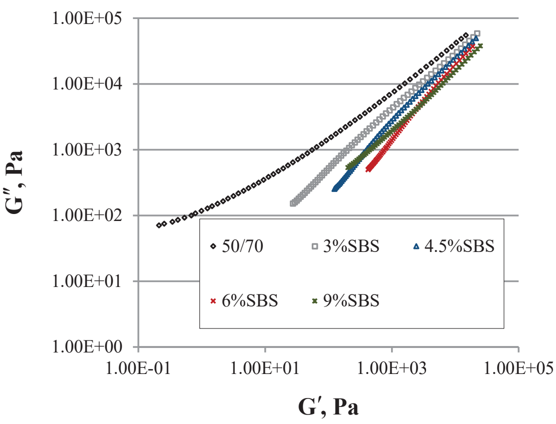

Fig. 8 Cole-Cole diagram of asphalt binders tested at an angular frequency of 10 rad/s in the temperature range of 40–100°C before ageing

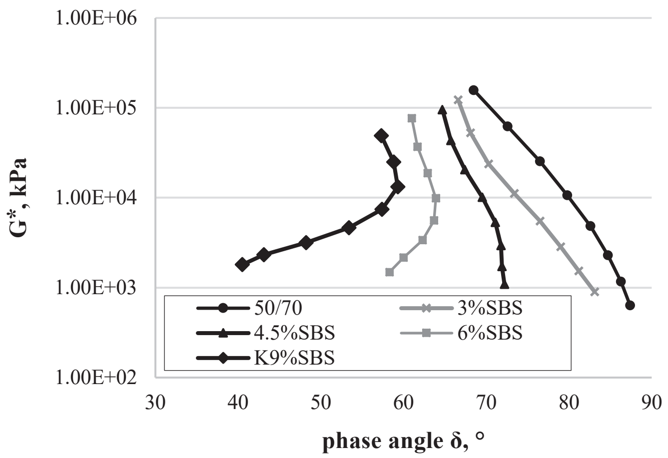

Fig. 7 Black curves of asphalt binders tested at an angular frequency of 10 rad/s in the temperature range of 40–100°C, after being subject to ageing by RTFOT method

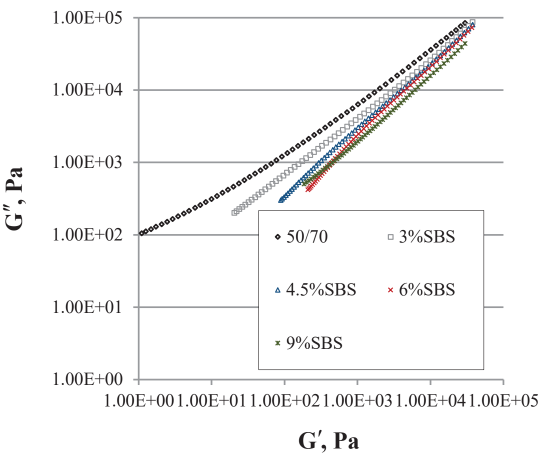

Fig. 9 Cole-Cole diagram of asphalt binders tested at an angular frequency of 10 rad/s in the temperature range of 40–100°C after ageing by RTFOT method

is greater than that of the elastic part (both before and after RTFOT ageing). On the other hand, for binders with a 6% and 9% SBS copolymer content, there is no misalignment between the properties of the elastic and viscous parts in the bitumen (they work within the viscoelastic range). There is a noticeable resistance of bitumens modified with SBS copolymer to the influence of short-term ageing, as the tested materials retain almost constant proportions of the real and imaginary parts, before and after ageing by RTFOT method (they retain constant viscoelastic properties).

Figures 10 and 11 show the relation of mean J_{nr} 3.2 values to temperature. It was observed that with the increasing amount of copolymer addition in the asphalt binder, the J_{nr} 3.2 values decrease. This is a desirable occurrence, because it involves increased resistance to rutting. The differences in values are particularly evident at high temperatures. The value of J_{nr} 3.2 at 82°C before ageing for unmodified 50/70 bitumen amounts to 25.698 kPa^{-1}, and for K9% SBS copolymer concentrate it is 0.113 kPa^{-1}. A decrease in J_{nr} 3.2 value can be noticed in the case of RTFOT ageing for 50/70 road bitumen and modified binders containing 3% SBS and 4.5% SBS. As the binder becomes stiffer in the course of ageing, the values of J_{nr} 3.2 decrease. For highly modified asphalt binders containing 6% SBS and 9% SBS, the opposite trend was observed. The post-ageing values increased – on average by 17% for binders containing 6% SBS, and by 58% for those containing K9 % SBS.

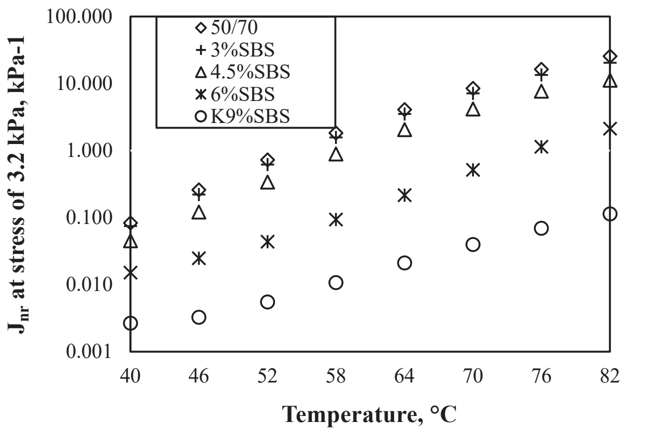

Fig.10. Values of J_{nr} at a present stress of 3.2 kPa as a function of the binder temperature before ageing

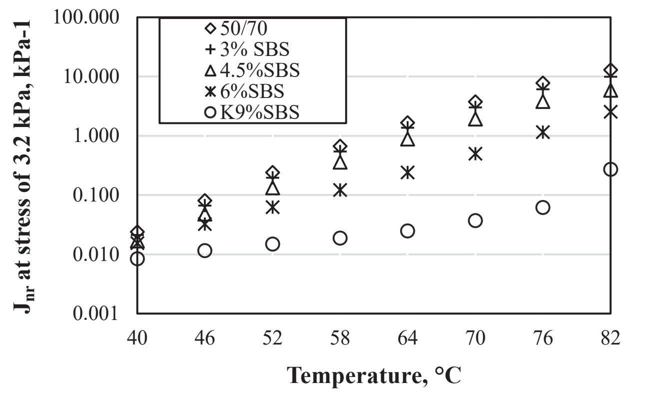

Fig. 11. Values of J_{nr} at a present stress of 3.2 kPa as a function of the binder temperature after RTFOT ageing

On the basis of the results of J_{nr} 3.2 determined at 64°C after RTFOT ageing, the studied binder was classified using AASHTO MP 19. The results have been compiled in Table 2.

Table 2
Designations of the analysed binders according to AASHTO MP 19

Asphalt binders	J_{nr} 3.2 before ageing [kPa^{-1}]	J_{nr} 3.2 after RTFOT ageing [kPa^{-1}]	Designations according to AASHTO MP 19
50/70	4.094	1.664	PG 64-XXH
3% SBS	3.525	1.359	PG 64-XXH
4.5% SBS	2.064	0.886	PG 64-XXV
6% SBS	0.216	0.239	PG 64-XXE
K9% SBS	0.021	0.025	PG 64-XXE

Table 2 shows that only highly modified binders (6% SBS, K9% SBS) can meet the requirements set for asphalt mixtures designed for extremely heavy traffic. Bitumen modification at a concentration of 4.5% SBS meets the requirements of very heavy traffic, and bitumen containing 3% SBS and 50/70 bitumen can carry heavy traffic.

The comparison of mean recovery values of R3.2 (Fig. 12) for 50/70 road bitumen and modified binders 3% SBS and 4.5% SBS shows higher values determined at 40°C after the ageing process. As test temperature increases, bitumen recovery values decrease, even reaching negative values. This phenomenon can be caused by machine error and sample inertia after

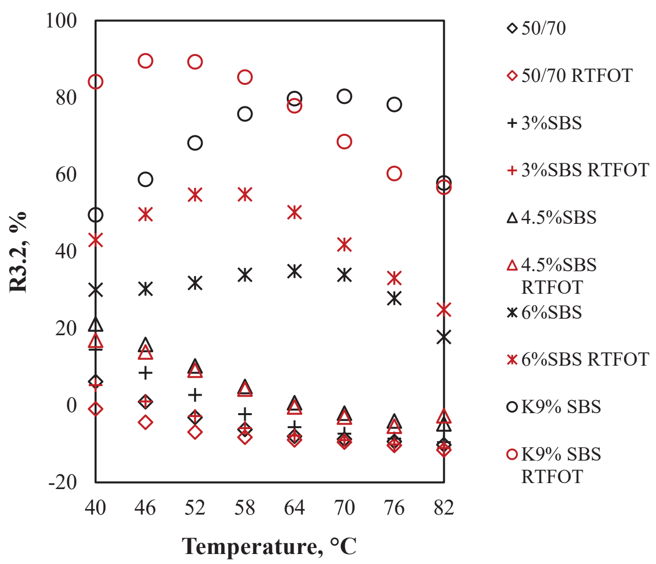

Fig. 12. Compilation of mean percentage values of recovery R3.2 obtained by the MSCR method in a DSR on bitumen samples before and after RTFOT ageing

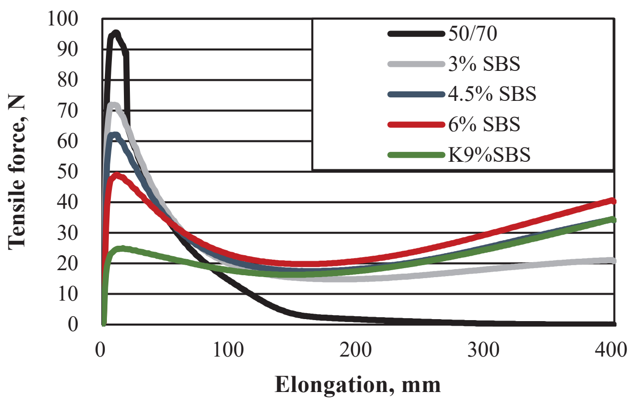

Fig. 13. Mean values of tensile force recorded during the tensile test at the temperature of 5°C

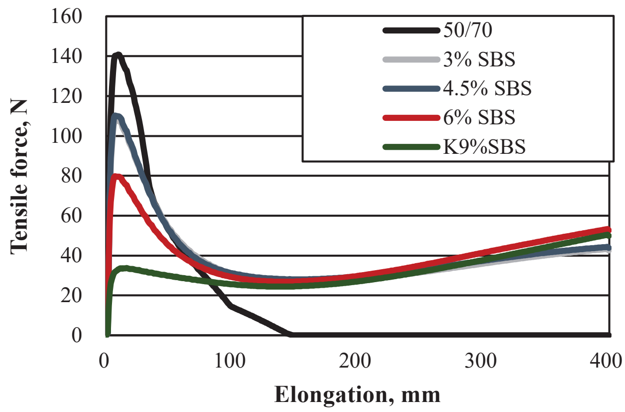

Fig. 14 Mean values of tensile force recorded during the tensile test at the temperature of 5°C for binders submitted to RTFOT ageing

relieving the load for unmodified and low-modified bitumens at high temperature when the binders become more viscous.

The percentage increase of the recovery values for 6% SBS and K9% SBS bitumens is related to the cross-linking of the polymer and not to the stiffening of the binder, hence the differences in the obtained values. According to the specification of the Institute of Seattle Implementation of the MSCR Test and specification: Questions, Clarifications, and Emphasis [29], one may speak of continuous polymer network when the R values range between 20 to 40%. It is formed by the swelling of a copolymer dispersed in the bitumen oil fraction.

Notably, for the results obtained after ageing only modified bitumens with over 6%SBS content have continuous cross-linking (at 82°C it does not exceed 20%). The bitumen containing 4.5% SBS does not meet these lower temperature requirements (at 40 °C it is within 20%). Negative values of the recovery have been obtained for 50/70 bitumen, 3% SBS and 4.5% SBS, in other words bitumen with low modification rate. This occurrence can be explained by the inertia of the measuring device in conjunction with a small recovery value observed during the 9 seconds period after introduction of the load. This can be observed during measurement in high temperatures (76°C and 82°C), where the elasticity properties decline.

4.2. Tensile test in ductilometer. Figures 13 and 14 were obtained from a tensile test performed in accordance with EN 13589:2018-08 standard. The chosen temperature was 5°C. When an elongation of 400 mm was reached, the stretching mechanism was stopped at 50 mm/min, and, for the next 20 min, force at constant elongation was being recorded. During the tests, the 50/70 unmodified binder samples were destroyed (both before and after RTFOT short-term ageing). The highest maximum force value was arrived at for 50/70 bitumen, while the lowest for SBS 9% concentrate – the difference was as much as 74%. For all modified bitumens, a further increase in force values was recorded, which exclusively in the case of the concentrate exceeded the first maximum by about 10 N, indicating a strengthening effect of the modification on the binder**.**

Upon submitting the binders to short-term RTFOT ageing, the values of tensile force increased for all tested binders. For an elongation of 400 mm, the differences between them were 49%, 78%, 76% and 69% for 3% SBS; 4.5% SBS; 6% SBS and 9% SBS, respectively. The modification reduced the negative impact of ageing by stiffening the asphalt binder.

Table 3 presents deformation work values together with measurement uncertainties obtained with ductilometer software with statistical significance $\alpha = 0.05$. The highest values were achieved for binder samples containing 6% SBS, both before and after ageing. The deformation work values were increased by about 2.5 J for aged samples compared to those not subjected to ageing, which is directly related to increased tensile strength of the stiffened bitumen binder. All tested binders meet the requirements of the PN-EN 14023:2011 standard concerning deformation work values.

Table 3
Deformation work values of bitumen binders before and after being submitted to RTFOT ageing

Binders	Mean deformation work value [J] before RTFOT ageing	Mean deformation work value [J] after RTFOT ageing
50/70	–	–
3% SBS	3.60 ± 0.20	7.16 ± 1.03
4.5% SBS	5.10 ± 0.08	7.43 ± 0.75
6% SBS	6.04 ± 0.39	8.26 ± 0.84
9% SBS	4.99 ± 0.14	7.60 ± 0.59

In terms of road pavement resistance to low-temperature cracks, it is most favourable when asphalt layers undergo relaxation as fast as possible. The analysis of Figs. 15 and 16 shows that modification with the SBS copolymer results in a faster stress dissipation in the function of time.

This can be stated despite the fact that the graphs in Figs. 15 and 16 describe force as a function of time. The tests assumed that after the mechanism had been stopped and stretching had been complete, the sample cross-section was not subject to further deformation, so the relaxation curves will have the

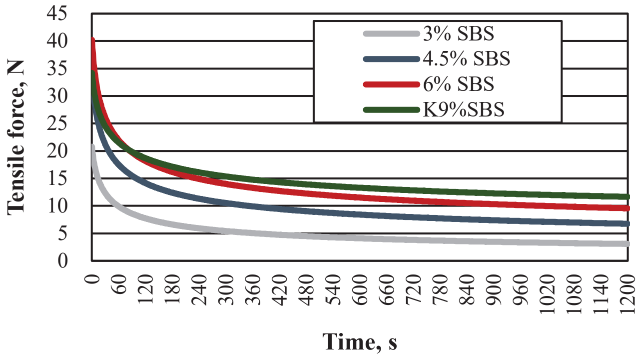

Fig. 15 Relaxation curves of the binders before ageing obtained in the tensile test in a ductilometer at 5°C

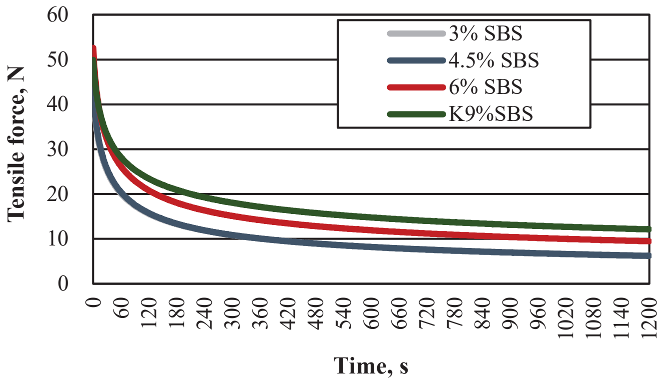

Fig. 16 Relaxation curves of the binders subjected to RTFOT ageing obtained in the tensile test in a ductilometer at 5°C

shape and tendency of force graphs, although calculation of exact stress values is impossible. For unaged samples, the percentage difference in the values of tensile forces between the start and end of the measurement is 85% for 3% SBS, 80% for 4.5% SBS, 76% for 6% SBS, and 66% for 9% SBS. Furthermore, regardless of the copolymer content in the samples, relaxation occurs most rapidly in the first 2–3 min upon the end of stretching, while after 20 min the changes in stress values become negligible.

The review of the obtained results in terms of short-term ageing reveals that it has a negative impact on the relaxation process. It slows down due to hardening and stiffening of the binder [11, 16]. Within 20 min, the force values decreased by 85% for 3% SBS, 86% for 4.5% SBS, 82% for 6% SBS and 77 for 9% SBS. Tensile force curves of binders with 3% SBS and 4.5% SBS content reached very similar values, differing from each other by about 3–5%. However, modification by use of SBS copolymer limits changes in rheological properties of the bitumen binder, most strikingly for SBS concentrate, where the differences between the pre- and post-ageing relaxation curve are minimal. Reducing SBS copolymer content in the bitumen increases the difference between the pre- and post-ageing curves with a negative tendency for binders subjected to RTFOT ageing. In aged binders, stress yields higher values after 20 min, because the test material is stiffer.

5. Conclusions

The results of the tests for selected rheological properties of bitumen modified with SBS copolymer, conducted with a DSR and a ductilometer, can be su mmarised with the following conclusions:

- increase in SBS copolymer content in the studied binder causes an increase in the dynamic shear modulus $|G^*|$. This fact may indicate greater resistance to permanent deformations of the bitumen pavement,
- the MSCR test has shown that increased use of the polymer in the bitumen translates to decreasing values of J_{nr}, which has a direct effect on the increase in resistance to rutting (permanent deformation). In this way, the authors have proven that modifying the binder with SBS copolymer enhances resistance to permanent deformation of the asphalt mixture during its lifetime (thus, increasing pavement durability), which was the purpose of the paper,
- increase in percentage of recovery R is related to the cross-linking of the polymer, and not to the stiffening of the binder. Continuous polymer network is found in tested bitumens with 6% SBS and 9% SBS copolymer content. It is formed when the copolymer dispersed in the bitumen oil fraction undergoes swelling, which supports the conclusion that it is the dispersion phase that has a greater influence on the rheological properties of the binder,
- SBS copolymer modifications accelerate stress-relaxation phenomenon in asphalt layers, which gives reason to believe that they also increase pavement resistance to low-temperature cracking,
- furthermore, modification reduces the negative impact of ageing on the properties of the binder, manifested by its stiffening and slowing down relaxation. The use of SBS copolymer reduces the increase in tensile forces during the tensile test of aged binders compared to binders not subjected to short-term RTFOT ageing.

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