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THE ASSESSMENT OF INFLUENCE OF STYRENE-BUTADIENE-STYRENE ELASTOMER'S CONTENT ON THE FUNCTIONAL PROPERTIES OF ASPHALT BINDERS

OCENA WPŁYWU ZAWARTOŚCI ELASTOMERU STYREN-BUTADIEN-STYREN NA WŁAŚCIWOŚCI FUNKCJONALNE LEPIECZY ASFALTOWYCH*

This paper discusses the issue of improving the functional properties of road asphalt pavements by modifying bituminous binder with SBS copolymer. The main purpose of the paper is to assess the resistance to permanent deformations and the temperature susceptibility of polymer-modified road asphalt binders, which are most commonly used in the upper layers of road and airport pavements. The bitumens subject to the study originate from various crude oil deposits (Russian and Venezuelan). They were modified in laboratory conditions with a concentrated additive with the known content of the SBS copolymer of 9%. The result was a asphalt binder containing the known percentage of the SBS copolymer of 1.5%, 3.0%, 4.5% and 6%. The rheological properties of the tested bitumens were determined by use of a dynamic shear rheometer (DSR), and with the application of the sinusoidal variable load, in the broad test temperature spectrum (from 40°C to 100°C). The analysis of the values of the dynamic shear modulus $|G^|$ of all the studied bitumens shows that the increase in the content of SBS copolymer in the tested binder increases the value of $|G^*|$, which may result in higher resistance to permanent deformations of road pavements caused by repeated traffic loads, especially in the case of pavements operated at high temperatures. The asphalt mixtures resistance to rutting is one of the basic parameters related to road pavement service-life, affecting both the safety and driving comfort of users.*

Keywords: dynamic shear rheometer (DSR), rutting factor, copolymer SBS, bitumen, complex shear modulus.

Tematyka pracy związana jest z zagadnieniem polepszenia właściwości funkcjonalnych drogowych nawierzchni asfaltowych poprzez modyfikację lepiszcza asfaltowego kopolimerem SBS. Głównym celem pracy jest ocena odporności na odkształcenia trwale oraz wrażliwości na zmiany temperatury asfaltów drogowych modyfikowanych polimerami, które są najczęściej używane w wierzchnich warstwach konstrukcji nawierzchni drogowych i lotniskowych. Przedmiotem badań były asfalty pochodzące z różnych złóż ropy naftowej (rosyjskiej i wenezuelskiej). Asfalty te poddano modyfikacji w warunkach laboratoryjnych z dodatkiem koncentratu o znanej zawartości kopolimeru SBS równej 9%. Otrzymano w ten sposób lepiszcza asfaltowe o znanej zawartości kopolimeru SBS równej 1,5%; 3,0%; 4,5% oraz 6%. Właściwości reologiczne badanych asfaltów oznaczono z użyciem reometru dynamicznego ścinania DSR stosując w testach obciążenie sinusoidalnie zmienne, w szerokim zakresie temperatury pomiarowej (od 40°C do 100°C). Analizując wartości dynamicznego modułu ścinania $|G^|$ wszystkich badanych asfaltów można stwierdzić, iż wzrost zawartości kopolimeru SBS w badanym lepiszczu zwiększa wartość $|G^*|$, co może skutkować większą odpornością na odkształcenia trwale nawierzchni drogowej spowodowane wielokrotnie powtarzającymi się obciążeniami ruchem pojazdów, w szczególności w przypadku nawierzchni eksploatowanej w wysokiej temperaturze. Odporność mieszanek mineralno-asfaltowych (MMA) na powstawanie kolein jest jednym z podstawowych parametrów związanych z eksploatacją nawierzchni drogowych, wpływając zarówno na bezpieczeństwo, jak i komfort jazdy użytkowników.*

Słowa kluczowe: reometr dynamicznego ścinania, wskaźnik odkształcalności, kopolimer SBS, asfalt, dynamiczny moduł ścinania.

1. Introduction

Nowadays, road pavements are subject to ever increasing traffic loads [4]. Taking into account both the construction, as well as maintenance costs [6], it is reasonable to search for solutions to optimise, for example, the composition of materials used for pavement structure. The analysis of the test results presented in [1, 11, 12] shows that one of the key factors increasing pavement rutting is the composition of the asphalt mixture, especially the applied asphalt binder. Therefore, there are studies striving to achieve the best rheological

properties for the applied bitumens processed in the crude oil distillation. These properties can be improved by introducing various modifiers to the binder structure, i.e. polymers [5], crumb rubber from car tyres [6] or natural asphalts [7]. Many scientific papers have analysed the effects of bitumen modification with the most commonly used polymers, including plastomers (e.g. polyethylene, polypropylene, ethylene vinyl acetate [17]), thermoplastic elastomers: SBS (styrene-butadiene-styrene) [1, 16]; SIS (styrene-isoprene-styrene) or mixed modifiers consisting of various polymers [2, 12]. Binders modified with polymers show an improvement in rheological properties com-

(*) Tekst artykułu w polskiej wersji językowej dostępny w elektronicznym wydaniu kwartalnika na stronie www.ein.org.pl

pared to unmodified bitumens [20]. The polymer most widely used in road construction is the block copolymer SBS, which, when added to hot bitumen, increases its volume several times in relation to its initial value [21]. When the concentration of the polymer in a modified bitumen is about 6%, the polymer becomes the dispersion phase and forms a continuous network in the structure of the bitumen. In case of a lower SBS concentration, the polymer network does not have to be continuous. Therefore, it is vital, both for the technical and economical reasons, to set the boundary content of the SBS copolymer in the asphalt binder in which the polymer network can form. Elastomer modified bitumens at operating temperatures are characterised by immediate elasticity (elastic deformation) and delayed (creep) elasticity [19, 21]. The papers [1, 12, 21] present modification methods and benefits of using SBS copolymer to modify binder, i.e. higher softening point, decrease in temperature susceptibility (expansion of the temperature-related viscoelasticity range), increase in cohesion at low temperature, significant improvement to elastic properties (observed e.g. in the elastic recovery test). Improving the rheological properties of binders results in better characteristics of the asphalt mixtures treated with the modified binder [13], that is increase in the resistance to permanent deformation and thermally induced cracking.

Airey in paper [1] analysed bitumens originating from two crude oil deposits (Russian and Venezuelan). The bitumens were modified with SBS copolymer, the concentration of the polymer in the modified bitumen amounting to 3%, 5%, and 7%, respectively. He noticed a significant influence of the polymer on rheological properties of the modified bitumen, i.e. increase of dynamic shear modulus and a higher proportion of the elastic part in the bitumen, especially at high temperature. He has also demonstrated the compatibility problem of the bitumen-polymer system. Airey [1] proved that bitumens with paraffin wax (of the Russian origin), due to higher content of the aromatic group, are better at binding the polymer in the modified bitumen structure. Behnood and Olek [6] performed a comparative analysis of three types of modifiers: SBS copolymer, crumb rubber and polyphosphoric acid. The low-temperature properties were studied with a bending beam rheometer (BBR), while the rheological properties at high temperatures were determined with a dynamic shear rheometer (DSR). At high temperatures, they observed an increase in the value of dynamic shear modulus for all the bitumens tested. Bitumen used in the production of asphalt mixtures, in road pavements, is exposed to ageing processes both during storage, production of asphalt mixtures, transport, paving and operation of the pavement [22]. Ageing phenomenon occurring during the production of asphalt mixtures and paving process [24] is considered to be the most unfavourable danger due to high temperatures. During short-term (technological) ageing occurring in the production and construction process of the asphalt pavement, bitumen is exposed to high temperatures (140 – 200°C) and oxygen in the air. Airey [1] and Sarnowski [19] have demonstrated the problem concerning the ageing of modified bitumen. Modified binders show improved rheological properties in the wide viscoelastic range. The authors observed that once modified bitumen has undergone the process of ageing, it shows a higher proportion of viscous part in relation to the elastic part, which may be caused by partial degradation of the polymer at high temperature, which occurs during the technological processes of coating the aggregate with bitumen, and the transport, paving and compaction of asphalt mixtures [1]. The next stage of ageing takes place during pavement operation (long-term ageing). In this case, binders are exposed to temperatures of up to 60°C in the summer season, with simultaneous exposure to oxygen, sunlight, water and chemicals [16,22]. Bai [6] performed a study on the influence of short-term and long-term ageing on rheological properties of asphalt binder modified with SBS copolymer. The tests were run on three bitumens with different polymer content, that is 3%, 6% and 9%. Based on the tests using the dynamic shear rheometer, Fraass Breaking Point apparatus and penetrometer, they

proved the negative influence of ageing on low-temperature properties of polymer modified bitumen.

The needs for maintenance and repair of the road network are very high. The systematically increasing traffic loads have adverse effect on the condition of the pavements, quickening their degradation [15]. Examining pavement condition encompasses such important means as pavement diagnostics and functional properties testing, i.e. longitudinal evenness, transverse evenness (ruts) [8], friction coefficient, load capacity, etc. [14, 15, 23]. Basic types of asphalt pavement destruction include: rutting, fatigue cracking, and low-temperature cracking [10, 13]. The formation of permanent deformations (ruts) in road pavements is influenced by many factors [13, 14], including the applied aggregate, binder, asphalt mixture, climate conditions, traffic load [9], and the applied pavement structure.

The purpose of the presented research work was to analyse the functional properties of binders modified with SBS copolymer, with special consideration of its temperature susceptibility, as the type of the applied binder is one of the key factors affecting the resistance to permanent deformation (ruts) in asphalt pavements. The resistance to rutting of asphalt mixtures is one of the basic preconditions for proper road pavement operation, affecting both safety and comfort of road users. This paper puts forth an original achievement of applying temperature susceptibility analysis of the modified binders tested at a wide range of temperatures.

2. Properties of the studied binders

Modification of bitumens with SBS copolymer is usually performed in the refineries, and rarely in the road construction companies' installations. Polymer modified bitumen used for the production of asphalt mixture can be obtained by purchasing a ready-made modified binder from a refinery, producing bitumen modified in a special technological installation, or purchasing bitumen with a known mass content of SBS copolymer, e.g. 9%, and mixing it in appropriate proportions with petroleum bitumen (road bitumen) [21].

The research was carried out using 50/70 penetration grade bitumens of similar hardness, expressed as penetration value determined at 25°C (Table 1), produced from crude oil from Venezuela and Russia. In the conducted studies, bitumens were combined with a concentrate of bitumen modified with SBS copolymer (block copolymer with linear structure) with 9% polymer content by composing in the proportions 5:1, 2:1, 1:1, and 1:2 to obtain asphalt binders with SBS copolymer content: 1.5%; 3.0%; 4.5% and 6.0% (in relation to the

Table 1. Basic properties of the tested asphalt binders

Tested material	Properties	$T_{R\&B}$ [°C]	Pen ₂₅ [mm/10]
V50/70		47.4 ± 0.2	66.0 ± 0.8
V1.5%SBS		47.9 ± 0.3	69.5 ± 0.4
V3%SBS		52.8 ± 1.3	71.3 ± 0.8
V4.5%SBS		74.5 ± 2.0	66.4 ± 0.6
V6%SBS		87.9 ± 1.3	66.3 ± 0.8
R50/70		47.8 ± 0.4	69.3 ± 0.3
R1.5%SBS		48.8 ± 0.3	70.3 ± 0.3
R3%SBS		49.5 ± 0.3	71.3 ± 0.4
R4.5%SBS		77.0 ± 1.1	69.5 ± 0.5
R6%SBS		83.8 ± 0.6	71.9 ± 1.0
K9%SBS		100.3 ± 1.4	74.3 ± 1.0

where: $T_{R\&B}$ - softening point determined acc. to PN-EN 1427:2015-08,
Pen₂₅ - penetration in 25°C determined acc. to PN-EN1426:2015-08

weight of the obtained modified bitumen), respectively. The specimens of the asphalt binders have been marked in the paper by the bitumen's origin, and then the percentage content of SBS polymer, e.g.:

- R6%SBS – means bitumen produced from Russian crude oil with a content of 6.0% of SBS copolymer,
- V50/70 – means 50/70 bitumen from Venezuelan crude oil, containing no SBS copolymer,
- K9%SBS – means modified bitumen concentrate containing 9.0% SBS.

Bitumen was analysed both in its initial state and after the process of technological (short-term) ageing simulated with the RTFOT (Rolling Thin Film Oven Test) method according to PN-EN 12607-1:2014.

The analysis of the results presented in Table 1 shows that the asphalt binders were selected so as to obtain bitumens of similar hardness, expressed through penetration determined at 25°C (Pen₂₅ was obtained in the range 66.0 mm/10 to 74.3 mm/10). Therefore, all modified binders subjected to the study can be classified as 45/80 modified bitumens, which are available on the Polish market, although they have different percentages of SBS copolymer content.

3. Purpose and methodology of the studies

The main aim of the study was to evaluate the resistance to permanent deformation and temperature susceptibility of road bitumens modified with SBS copolymer on the basis of tests carried out with dynamic shear rheometer (DSR) of the Physica MCR 101 type. Permanent deformation and sensitivity to temperature changes in the Polish climate zone are of key importance in the use of asphalt pavements.

The tests were performed in compliance with the norm: PN-EN 14770:2012 "Determination of complex shear modulus and Phase Angle - Dynamic Shear Rheometer". They employed two methods involving kinematic (sinusoidal) coercion:

- at different angular frequency ranges from 100 rad/s to 0.1 rad/s, and a constant test temperature of 60°C±0.01°C,
- with a deflection angle amplitude of 10 mrad and variable temperature, i.e. from 100°C to 40°C, and a temperature decrease of 1°C every 1 minute. The test procedure involved an assumption of a constant angular frequency value equal to 10 rad/s.

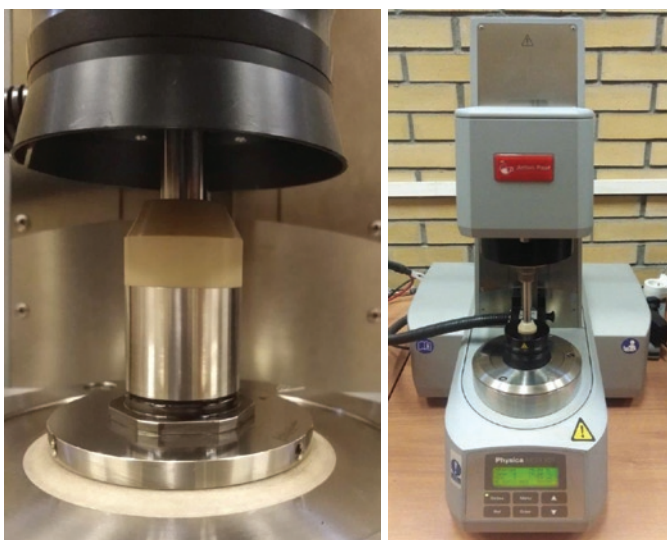


Fig. 1. A side-view of the tested bitumen sample. Fig. 2. Dynamic Shear Rheometer (DSR) of the Physica MCR 101 type

A sample of the asphalt binder was placed between two circular parallel plates with a diameter of Ø25 mm, maintaining the required gap height of 1 mm (Fig. 1).

According to the American Superpave specification, the susceptibility of bitumen binders to permanent deformation in road pavements is determined by the rutting factor, expressed as a value of $|G^*|/\sin \delta$. In this paper, the above mentioned coefficient was determined for bitumens subjected to the RTFOT short-term ageing method, as well as for bitumens not subjected to it.

The analysis also included the values of the Shear Modulus Index (SMI), a measure of the temperature susceptibility of the binders studied, calculated according to the formula [20]:

$$SMI_{T_2/T_1} = \left| \frac{\log \log |G_{T_1}^*| - \log \log |G_{T_2}^*|}{\log(T_1 + 273,15) - \log(T_2 + 273,15)} \right| \quad (1)$$

where:

SMI – Shear Modulus Index

$|G_{T_1}^*|; |G_{T_2}^*|$ – dynamic shear modulus at temperature T_1, T_2 , respectively, [Pa]

$T_1; T_2$ – extreme temperatures of the measurements taken using DSR, where $T_1 > T_2$, [°C]

It was assumed in this paper that $T_1 = 100^\circ\text{C}; T_2 = 40^\circ\text{C}$.

4. Analysis of test results

Figure 3 shows a graph of the relation of the angular frequency to the dynamic shear modulus of the tested bitumen (range from 0.1 rad/s to 100 rad/s). For all tested bitumens, the dynamic shear modulus values increases with the growth of angle frequency. The highest value of $|G^*|$ at an angular frequency of 0.1 rad/s has been observed for the bitumen with the content of SBS copolymer equal to 9%, while the lowest value has been found for the R50/70 bitumen. With an angular frequency of 100 rad/s, the dynamic shear modulus achieves values close to from 23680Pa for K9%SBS to 41010Pa for V50/70 bitumen.

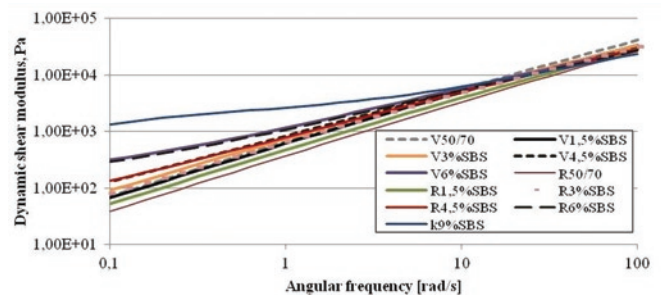


Fig. 3. Graph showing the relation of the angular frequency to the dynamic shear modulus of the bitumens tested at 60°C

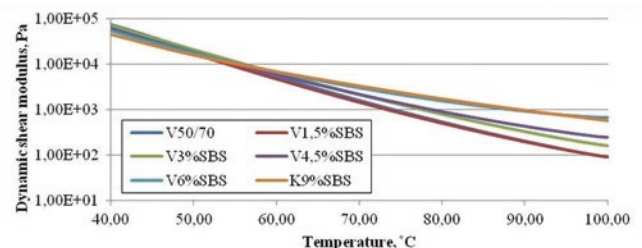


Fig. 4. A graph showing temperature dependence of $|G^*|$ dynamic shear modulus for unaged Venezuelan binders, at a constant angular frequency value of 10 rad/s

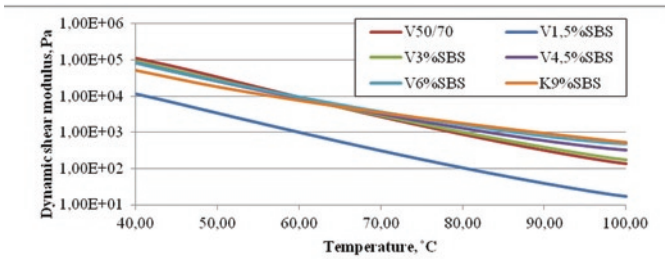


Fig. 5. A graph showing temperature dependence of $|G^*|$ dynamic shear modulus for Venezuelan binders subjected to RTFOT ageing, at a constant angular frequency value of 10 rad/s

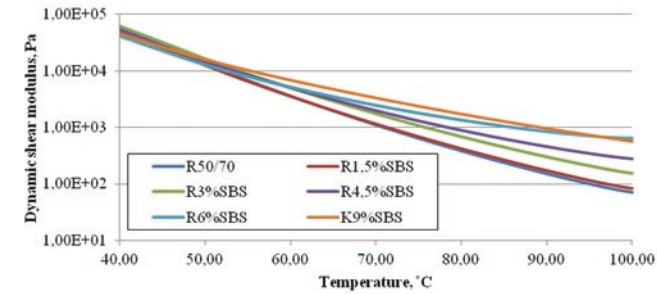


Fig. 6. A graph showing temperature dependence of $|G^*|$ dynamic shear modulus for unaged Russian binders, at a constant angular frequency value of 10 rad/s

Asphalt binders are materials with viscoelastic properties. The analysis of the values of the phase angle δ can be used to evaluate changes in the rheological properties of bitumen across the entire temperature spectrum during the production process, paving and operation of the asphalt pavement. Viscous materials are characterised by the damping factor $\tan \delta \rightarrow \infty$ ($\delta = 90^\circ$), whereas in the case of elastic materials $\tan \delta = 0$ ($\delta = 0^\circ$); viscoelastic materials have a phase angle values of $0^\circ < \delta < 90^\circ$. In Figures 4-7 there are graphs showing temperature dependence of $|G^*|$ dynamic shear modulus of the studied bitumen binders of Venezuelan and Russian origin, both submitted to, and not having been subject to RTFOT ageing. The value of the $|G^*|$ dynamic shear modulus decreases with the increase of the test temperature for all analysed bitumens.

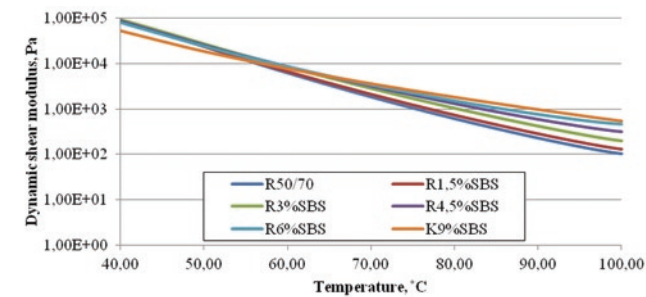


Fig. 7. A graph showing temperature dependence of $|G^*|$ dynamic shear modulus for Russian binders subjected to RTFOT ageing, at a constant angular frequency of 10 rad/s

In view of the high summer temperatures, the elastic component is especially important, which is associated with low $\tan \delta$ values. It was observed that in the case of unmodified bitumens (R and V) and bitumens with a low content of SBS copolymer (up to 3%), there is a regularity that the higher the values of the dynamic shear modulus, the lower the values of the phase angle δ , as shown in the Black diagram (Fig.8.). Based on the graphs showing the relation of the dynamic shear modulus to the phase angle, called Black's graphs, it is possible to perform an analysis of two basic parameters determined in the

dynamic shear rheometer [1,3,20]. In the case of asphalt binders with 6% and 9% SBS copolymer content, small phase angle values are obtained, both at very low and high $|G^*|$ values. The highest variation in phase angle was observed for reference 50/70 penetration grade bitumens and low-modified binders (with up to 3% copolymer content). Their values at high temperatures are close to 90° , so it can be assumed that these binders at the high temperature range have properties similar to those of a viscous liquid. The increase in the content of the SBS copolymer in the bitumen makes the variation in δ values ever smaller. Above 70°C , the δ values are reduced for bitumens with a copolymer content of 4.5%, 6%, and 9% SBS concentrate, which shows the beneficial effect of applying the polymer for modification, since modified binders have a higher share of the elastic part at high temperatures, which may indicate greater resistance to permanent deformation.

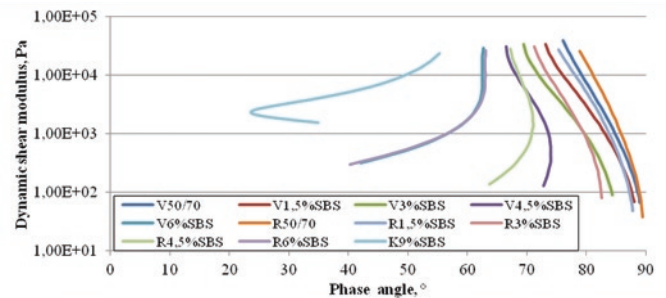


Fig. 8. Black's graph showing the relation of the phase angle to the dynamic shear modulus in the tested asphalt binders

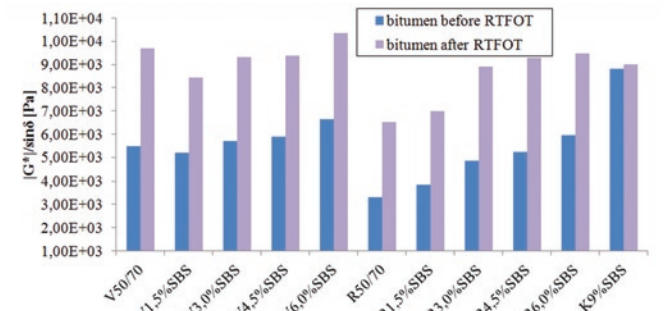


Fig. 9. Values of $|G^*|/\sin \delta$ rutting factor of the asphalt binders tested at 60°C .

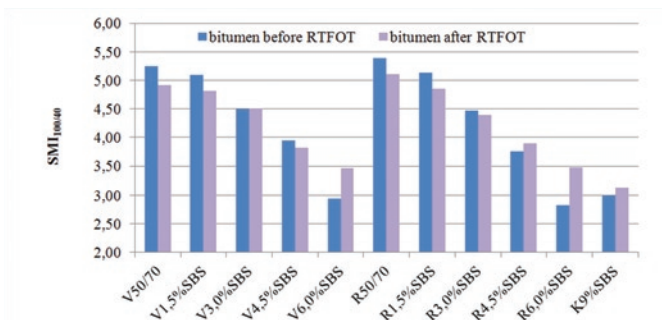


Fig. 10. Shear Modulus Index of the tested bitumen binders

As an effect of conducted tests, the rutting factor was determined (defined as the ratio of the $|G^*|$ dynamic shear modulus to the sinus of the phase angle δ ($|G^*|/\sin \delta$)) at the temperature of 60°C , assumed as the extreme temperature occurring in asphalt pavements in Poland (in which the rutting test for asphalt mixtures according to PN-EN 12697-22:2008 is also performed).

The Superpave specification indicates a relation between the resistance to permanent deformation in bitumen pavements and the

properties of the tested binders determined by the DSR method, with the following requirements:

- $|G^*|/\sin\delta \geq 1,0$ kPa – for unaged bitumen not submitted to ageing
- $|G^*|/\sin\delta \geq 2,2$ kPa – for bitumen subjected to short-term ageing simulated with the RTFOT method.

The results shown in Figure 9 show that all the binders tested meet the above requirements of the Superpave specifications. A higher value of the rutting factor of the bitumen characterised with the resistance to permanent deformation of the asphalt pavement is obtained by a higher value of the dynamic shear modulus $|G^*|$ and a lower value of the phase angle δ .

The lowest value of the rutting factor was observed for unmodified bitumens of Russian origin, both before and after ageing with RTFOT method. Special attention should be paid to the value of $|G^*|/\sin\delta$ of the concentrate containing 9% of SBS copolymer content, since the difference in the rutting factor before and after ageing is only 0.2 kPa; which may indicate that the effect of ageing on the values of the rutting factor is low.

The measure of temperature susceptibility is expressed with the PI penetration index. It can be calculated on the basis of penetration results determined at two various temperatures or by means of an indirect method using values of penetration determined at 25°C and softening point. The above methods make it possible to estimate the penetration index of unmodified bitumens. However, in the case of bitumens modified with elastomers, the results obtained with each method may significantly differ [20]. In this study, the temperature susceptibility was determined on the basis of formula (1). It can be stated that the assumptions for penetration at softening point (800 mm/10) and Fraass breaking point (1.25 mm/10) for bitumen modified with elastomers are not correct [20]. The analysis of the obtained SMI values showed a remarkable influence of the copolymer content on the reduction of

bitumen sensitivity to changes in stiffness to temperature. Bitumen R50/70 proved to be the most sensitive to changes in properties due to temperature changes. The lowest SMI value at the temperature range 100°C – 40°C was achieved for the bitumen with SBS content equal to 6% in the unaged state, while upon RTFOT ageing, the SMI value increased for this group of bitumens (both of Venezuelan and Russian origin), which may indicate partial polymer degradation under the influence of high temperature (163°C) and oxygen.

5. Conclusions

By comparing the values of the dynamic shear modulus of the tested bitumens it can be stated that with the increase in the content of SBS copolymer the value of $|G^*|$ increases, which may indicate a higher resistance of asphalt pavements made with SBS copolymer modified bitumens to deformations caused by repeated shear stress (which illustrates repeated load cycles caused by traffic in real conditions).

With the increase in the content of SBS copolymer in the tested bitumens, the value of phase angle decreases, which results in the improvement of elastic properties of the binders.

SMI (Shear Modulus Index) analysis showed a considerable effect of styrene-butadiene-styrene copolymer (SBS) content on the reduction of the sensitivity of asphalt binders to changes in stiffness at variable temperatures. Reducing the susceptibility to temperature changes illustrates the significantly favourable effect of using SBS copolymer as a modifier of asphalt binders.

The use of binders modified with SBS copolymer in asphalt mixtures improves the functional properties of flexible pavements (which is confirmed by the values of the rutting factor and SMI), thus, improving the operational parameters of road pavements and their durability.

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