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Article

Evaluation of Rheological Properties of Polymer-Modified Asphalt Binders and Mastics with Organic Additive—Imidazoline

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Abstract: The article presents the results of testing the asphalt binder modified with SBS copolymer (5%) and its mixture with mineral fillers called asphalt mastics. The aim of the research and analysis was to check the possibility of using an organic additive in the form of imidazolines and to assess their impact on the viscoelastic properties of the obtained asphalt mastics. The main advantage of using imidazoline is the increased resistance of asphalt mixtures to low-temperature cracking at the top of the road route and reducing its maintenance costs. Based on the examination of the adhesion of the binder with the addition of various imidazolines to the aggregate, a selection was made from six analyzed subtypes. One of them was selected for further research. The tests were carried out in a dynamic shear rheometer in a wide temperature range from −36 °C to 82 °C, determining the most important parameters of the tested materials, i.e., dynamic shear modulus ($|G^*|$), phase angle (δ) and the non-recoverable creep compliance (J_{nr}). The test results and their analysis confirmed the beneficial effect of imidazoline on the viscoelastic properties of mastics in the low-temperature range, even at the lowest content of 0.2%. Based on the results of the determination of J_{nr} and $|G^*|/\sin\delta$ in the range of high operating temperatures, no significant deterioration in permanent deformation parameters was observed. Moreover, the use of imidazoline reduces technological temperatures during the production of HMA used in the pavement and, therefore, reduces CO_2 emissions into the atmosphere.

Keywords: bitumen; asphalt mastic; imidazoline; sustainable asphalt pavement; rheological properties; dynamic shear rheometer; MSCR; SBS

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1. Introduction

In times of climate crisis and in the face of the problem of growing amounts of industrial waste (including from construction sites), it is extremely important to try to use new solutions aimed at increasing the durability of materials and reducing the negative impact of their production on the environment.

Due to the recent occurrence of extreme temperatures, materials previously used in pavement structures may degrade faster than expected by engineers. This problem has been addressed by scientists in China [1]. Asphalt surface courses are susceptible to low-temperature cracking or rutting due to the significant impact of high and low temperatures on their properties. This is mainly determined by the properties of binders used in asphalt mixtures. Tiansheng et al. predict that by 2050, low-temperature cracks in road surfaces in regions where extremely low temperatures will increasingly occur may increase by 35% [1]. In the United States, Gudipoli et al. predict an increase in the occurrence of fatigue cracking by 2 to 9% in the following years and rutting phenomena by 9 to 40% [2].

The beneficial effect of polymers on improving the rheological properties of asphalt binders, especially on expanding the temperature range of their viscoelasticity, has long been known. The polymers most commonly used to modify road bitumens are elastomers that form a cross-linked structure or mixture with the bitumen. There is a huge diversity among polymer-modified bitumens from different production batches or from different manufacturers. The most commonly used elastomer is the SBS copolymer (styrene–butadiene–styrene) with a two- or three-block or disordered structure (e.g., styrene–butadiene-random (SBR)) [3–5]. SBS elastomers have mechanical properties comparable to volcanic rubber but are only physically cross-linked. This cross-linking may be temporarily destroyed under the influence of high temperatures (above the glass transition temperature of polystyrene, i.e., above 100 °C), but after cooling, the cross-linking is rebuilt (regenerated) [6]. When talking about the modification of bitumen with polymers, the homogeneity of the modified binder must be taken into account [5].

Faster degradation of asphalt pavement increases its maintenance costs. Material standards used in pavement engineering are not adapted to rapid climate changes, which have been a completely underestimated phenomenon until recently [7]. Therefore, scientists propose new additives that use industrial waste (as a form of disposal) or natural substances that are intended to increase the resistance of binders to changes in properties under the influence of extreme temperatures. Motamedi et al. [8] tested mastics—asphalt binders combined with mineral fillers, which are obtained from waste generated during the production of mineral aggregates. Researchers proved that the addition of lime filler reduced rutting, and synthetic polyurethane reduced low-temperature cracking of the asphalt binder. Another possibility is to use the ground remains of asphalt pavements after renovations as an additive in the production of new asphalt mixtures (so-called RAP—Reclaimed Asphalt Pavement) [9]. Yousefi et al. [10] examined the properties of WMA (Warm Mix Asphalt—asphalt mixtures with reduced mixing temperature) with the addition of RAP. Among other things, it increases the stiffness of binders and reduces rutting but increases susceptibility to cracking. Selected additives used in WMA allowed for the addition of large amounts of RAP without negative effects on the mechanical properties of the mixture. Xingyu et al. [11] used an epoxy resin polymer combined with the addition of RAP in asphalt binders. Scientists proved that the polymer content of approximately 40% by weight has a positive effect on their properties at low and high temperatures because the resin creates a reinforcing network in the binder structure.

Imidazoline is an organic chemical compound used in the production of cationic emulsions used in the renovation of asphalt pavements [12,13]. The imidazoline additives added to binders used in the research are wastes from the synthesis by cyclization of diamidoamine [14]. Previous research has shown that lard, oleic and rapeseed imidazolines can be successfully combined with asphalt binders, and they form a homogeneous substance [14]. Moreover, the addition of imidazoline makes it possible to reduce the mixing temperature of the asphalt mixture, thus reducing energy consumption during its production and CO_2 emissions into the atmosphere. However, further research on the use of imidazoline as an additive to asphalt binders was not continued due to deep rutting of the pavement in the case of several percent imidazoline content in the binder. For this reason, it was decided to analyze the influence of imidazoline in the content below 1% on the rheological properties of asphalt binders and mastics.

In this article, the authors examined binders modified with SBS copolymer (styrene–butadiene–styrene), which is one of the most popular copolymers used in asphalt mixtures due to its beneficial effect on the rheological properties of the binder at low and high temperatures [15–18]. Authors of this article examined the effect of the addition of imidazoline (0.2%, 0.4% and 0.6% content) in relation to the binder mass on the properties of binders and asphalt mastics (with lime, granodiorite and basalt fillers) in a dynamic shear rheometer (DSR) enabling the examination of the viscoelastic properties of the binder by determining such parameters as rutting factor ($|G^*|/\sin\delta$) or the non-recoverable creep compliance (J_{nr}) [18]. The influence of the addition of imidazoline to the asphalt binder on the formation of permanent deformations in road pavements was analyzed. Research carried out by [14,17] showed that imidazoline, when its content exceeds 1%, causes an increase in permanent deformations in the pavement.

2. Materials and Methods

2.1. Materials

2.1.1. Asphalt Binders and Mastics

The tests used road bitumen with a penetration grade of 50/70, in accordance with EN 12591, mixed with a styrene–butadiene–styrene copolymer (SBS) highly modified asphalt binder (160/220 penetration grade bitumen was used as a base binder) at a SBS concentration of 9% (referred to as the "concentrate" in the article). By mixing these two binders in appropriate proportions at a temperature of 170 °C, a binder with a concentration of 5% SBS was obtained (55.5% of the mixture was a concentrate). The research employed binder, which is most often used in the production of hot mix asphalt. After mixing, the binders were stored in covered aluminum containers at room temperature so that they did not change their properties. The results of determining the penetration and softening point of binders are presented in Table 1 (where the designation "RII" means the name of rapeseed imidazoline type II).

Table 1. Penetration and softening temperature values of binders.

Bitumen	Penetration (0.1 mm)	$T_{R\&B}$ (°C)
50/70	67.5 ± 0.4	46.6 ± 0.5
5% SBS	71.0 ± 0.8	93.3 ± 0.3
5% SBS + 0.2% RII	70.8 ± 0.5	87.4 ± 0.3
5% SBS + 0.4% RII	67.3 ± 1.7	88.5 ± 0.4
5% SBS + 0.6% RII	69.3 ± 1.7	88.9 ± 0.4

The asphalt binder was then mixed with three mineral fillers sifted through a sieve with a # 0.125 mm mesh (86% of limestone weight has grain size under 0.063 mm, granodiorite 74% and basalt 82%). Limestone, granodiorite and basalt aggregates were selected for the tests due to the difference in their acidity (limestone is considered an alkaline aggregate due to the low percentage of SiO_2 silica, basalt—neutral, and granodiorite aggregate as acidic [14,19,20]). They were mixed to obtain a volume ratio of filler to bitumen (F/B) = 0.60 in accordance with EN 13179-1. The research methodology is presented in Figure 1.

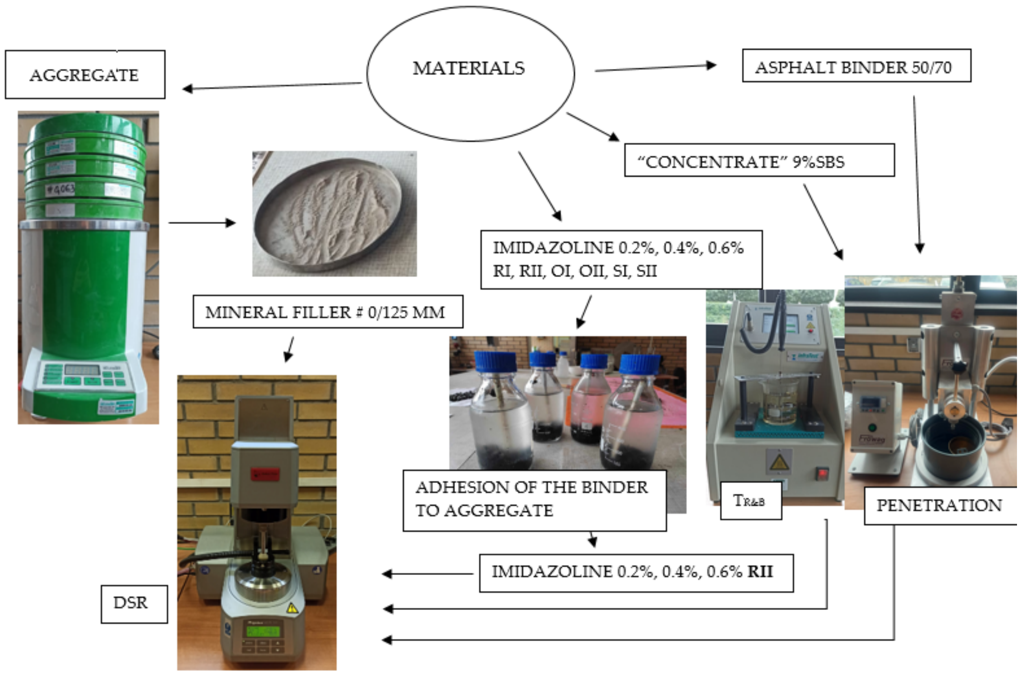


Figure 1. The research methodology.

2.1.2. Imidazolines

Before the research, imidazolines were selected from 6 subtypes: lard imidazoline type I and II (abbreviated in the article as "SI and SII"), rapeseed imidazoline type I and II (RI and RII, respectively) and oleic imidazoline type I and II (OI and OII). They were made based on animal fat, rapeseed oil and oleic acid, respectively. Types I and II differ in their chemical structure, according to Figure 2.

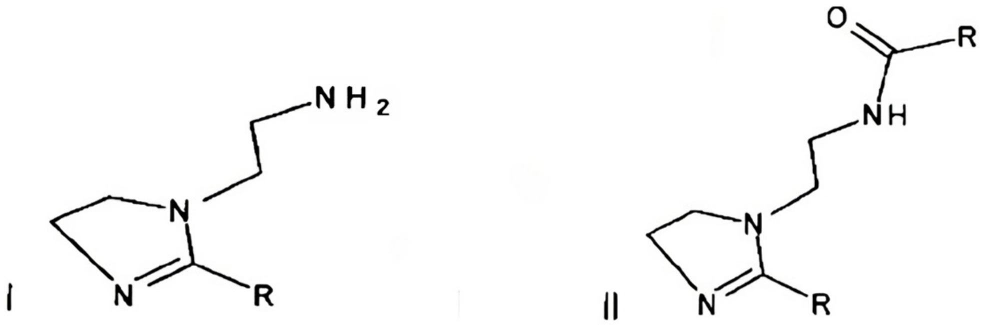


Figure 2. Imidazolines type I and II—chemical structure, where R = C_nH_m, n = 15–20, m = 31–39 [15].

Imidazoline contains from 85% to 100% pure imidazoline with an admixture of up to 10% amidoamines, up to 1% aikilotriamines and up to 10% glycerin. Imidozolines were mixed with the asphalt binder at a temperature of 170 °C using contents of 0.2%, 0.4% and 0.6% of the binder weight.

One type of imidazoline was used in the research: rapeseed type II. The selection was made based on the results of adhesion tests of the asphalt binder (5% SBS + 0.4% imidazoline to limestone, granodiorite and basalt aggregate, in accordance with EN 12697-11). The percentage of grain surrounded by binder was assessed in the graphical software (based on the analysis of the number of pixels between the unsurrounded and surrounded parts of the aggregate as shown in Figure 3—the grain areas not surrounded by binder were manually marked and compared to the number of black pixels signifying the aggregate surrounded by bitumen) after 3, 6 and 24 h of rotating 4 bottles containing tested samples. They were filled with samples of aggregate covered by bitumen and then distilled water with a glass rod, mixing the grains while rotating them in a machine at 40 rpm (Figure 4). After 3, 6 and 24 h, the bottles were emptied, photos of the grains were taken, and then they were refilled with distilled water. Results are presented in Table 2. Rapeseed imidazoline type II was considered to have the most beneficial effect on the adhesion of the binder to the limestone aggregate, type I rapeseed imidazoline on the adhesion to basalt aggregate, and type II oleic imidazoline on the adhesion to granodiorite aggregate. Imidazoline was selected for further research as it offers the biggest improvement in regard to the adhesion of the binder to limestone aggregate commonly used in road construction [20].

Table 2. Percentage of aggregate surface not covered by asphalt binder.

Type of Imidazoline Added to the Asphalt Binder	Area Which Was Not Covered after 3 h of Testing (%)	Area Which Was Not Covered after 6 h of Testing (%)	Area Which Was Not Covered after 24 h of Testing (%)
Limestone Aggregate			
O I	3.3 ± 1.0	5.8 ± 0.2	33.1 ± 13.2
O II	5.3 ± 1.1	10.8 ± 2.6	34.9 ± 6.4
R I	3.4 ± 0.4	5.3 ± 1.8	33.1 ± 6.5
R II	3.7 ± 1.6	8.1 ± 0.3	23.0 ± 7.1
S I	4.5 ± 0.8	14.6 ± 3.3	33.6 ± 4.1
S II	3.8 ± 0.2	6.0 ± 1.3	40.9 ± 3.9

Table 2. *Cont.*

Type of Imidazoline Added to the Asphalt Binder	Area Which Was Not Covered after 3 h of Testing (%)	Area Which Was Not Covered after 6 h of Testing (%)	Area Which Was Not Covered after 24 h of Testing (%)
Basalt Aggregate			
O I	18.9 ± 1.3	27.0 ± 2.0	31.4 ± 4.8
O II	30.9 ± 2.3	37.6 ± 3.6	48.3 ± 1.3
R I	17.6 ± 0.3	29.9 ± 4.1	30.7 ± 2.2
R II	15.0 ± 3.8	36.9 ± 0.3	43.2 ± 8.1
S I	43.3 ± 0.8	43.6 ± 3.3	46.1 ± 2.9
S II	30.2 ± 3.8	40.8 ± 3.3	44.7 ± 1.4
Granodiorite Aggregate			
O I	5.4 ± 1.0	12.0 ± 2.7	25.8 ± 4.4
O II	5.9 ± 0.8	14.1 ± 2.4	23.0 ± 2.5
R I	14.0 ± 3.2	29.8 ± 0.8	33.1 ± 6.5
R II	17.5 ± 2.6	30.1 ± 2.6	34.9 ± 4.4
S I	24.5 ± 5.1	28.4 ± 4.2	32.2 ± 4.3
S II	30.3 ± 0.3	30.5 ± 3.6	41.7 ± 1.6

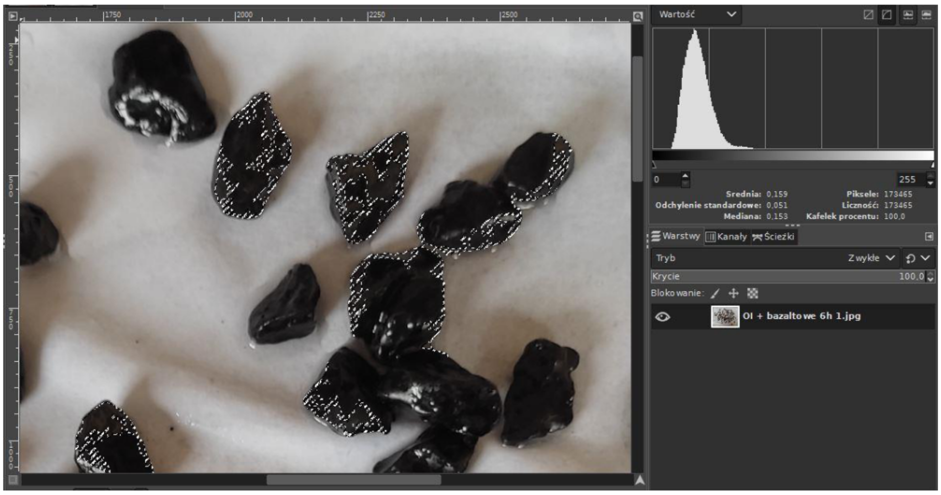


Figure 3. Analysis of aggregate parts which were not fully covered with bitumen in the GIMP 2.10.36 graphical software.

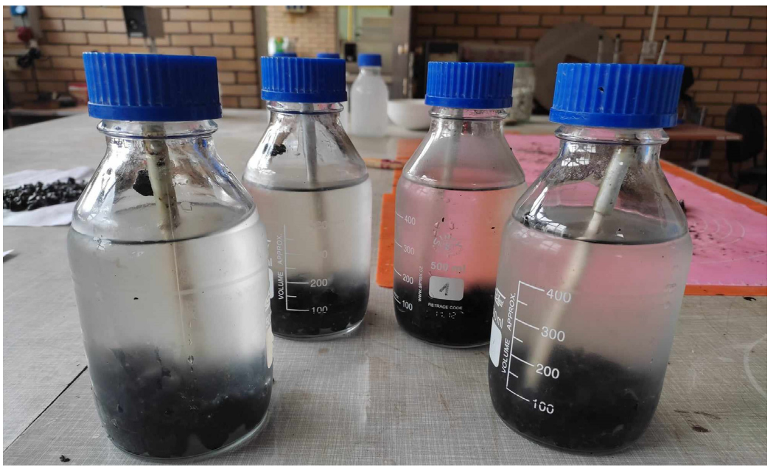


Figure 4. Laboratory bottles filled with distilled water cooled to 5 °C and aggregate covered by asphalt binder with the addition of imidazoline.

2.2. Methods

Tests of asphalt binders and mastics were carried out in a dynamic shear rheometer using 3 research programs:

- The test at intermediate temperatures was performed using a system of two parallel measuring plates with a diameter of 8 mm and a test gap of 1.5 mm. Measurement temperatures ranging from 40 °C to −36 °C were used, with increments of 6 °C. At each temperature, the test was performed at a logarithmically variable vibration frequency value in the range of 100 Hz to 0.1 Hz.
- The test was performed in the high temperature range from 82 °C to 40 °C, with increments of 6 °C. A measuring system of two parallel plates with a diameter of 25 mm and a test gap of 1.0 mm was used. At each temperature, the test was performed at a variable frequency from 100 to 0.1 Hz.
- MSCR (Multiple Stress Creep Recovery) cyclic creep testing with stress relief using a system of parallel plates with a diameter of 25 mm and a measurement gap of 1 mm. A variable temperature was used in the study, i.e., in the range from 82 °C to 58 °C in 6 °C increments. At each of the given temperatures, the test consisted of the following stages: creep cycle (loading the asphalt binder sample for a period of 1 s) and stress relief cycle (relieving the stresses of the asphalt binder sample for a period of 9 s). Figure 5a single cycle lasted 10 s and was repeated 10 times. The test was performed at three different shear stress values of 0.1 kPa, 3.2 kPa and 10 kPa. The test began with the application of the lowest shear stress (0.1 kPa). The sample was subjected to 10 cycles of loading and unloading, then the stress was changed, and a similar test was carried out for each temperature stress level.

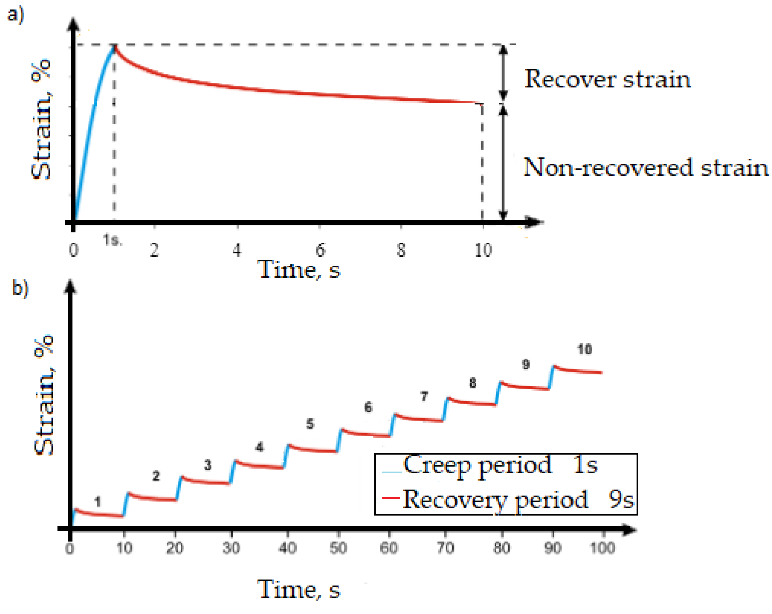
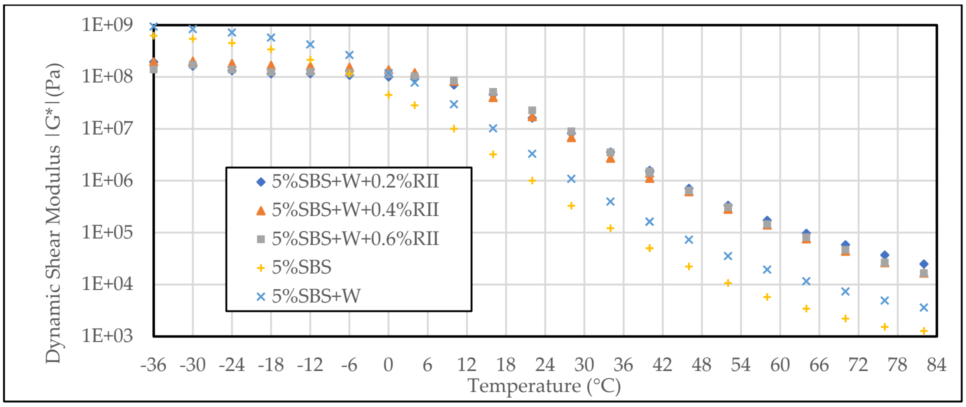


Figure 5. MSCR method, (**a**) single cycle of creep and recovery, (**b**) ten cycles of creep and recovery.

3. Results

The rheological properties of the analyzed asphalt binders and mastics with the addition of imidazoline were determined by tests using a dynamic shear rheometer (DSR). For each tested binder and mastic, four samples were made, and the average value of the tested parameters was calculated, excluding results exceeding the gross error value (Grubbs' test) at the confidence level P_u = 95%. The graphs in Figure 6 show the dependence of the dynamic shear modulus (DSM) on the temperature in the so-called operating temperature range, i.e., from −36 °C to 82 °C, determined with sinusoidal kinematic excitation at a constant angular frequency of ω = 10 rad/s. The rheological properties of the resulting asphalt mastics as a viscoelastic material depend mainly on the temperature and test parameters. Determination of the dynamic shear modulus |G*| and the phase angle δ makes it possible to assess the viscoelastic range of the tested asphalt binders and mastics. In order to avoid the formation of permanent deformations in asphalt pavements at high operating temperatures occurring in the summer, a higher value of the dynamic shear modulus and a smaller phase angle are beneficial, i.e., the tested material should have a larger share of the elastic part [16]. In each analyzed case, an increase in the test temperature causes a decrease in the value of the dynamic shear modulus. The presence of the filler in each case influenced the viscoelastic characteristics of the mastics by stiffening them in relation to the 5% SBS bitumen.



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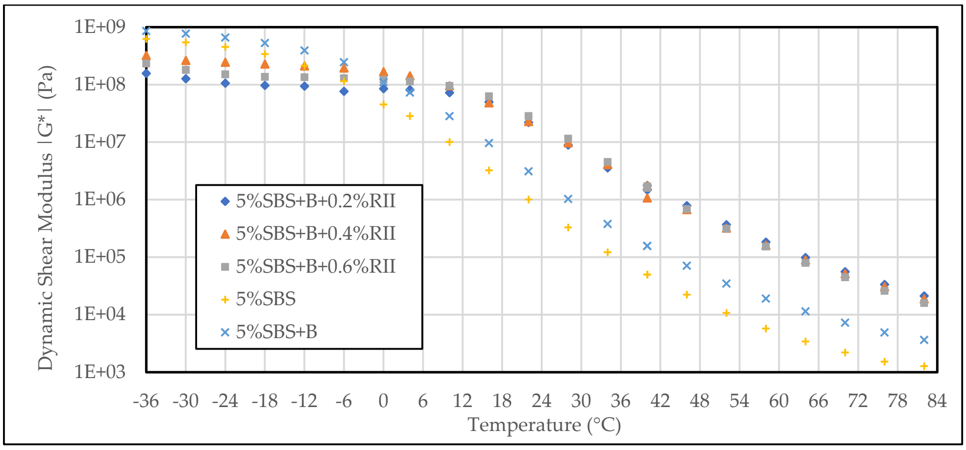

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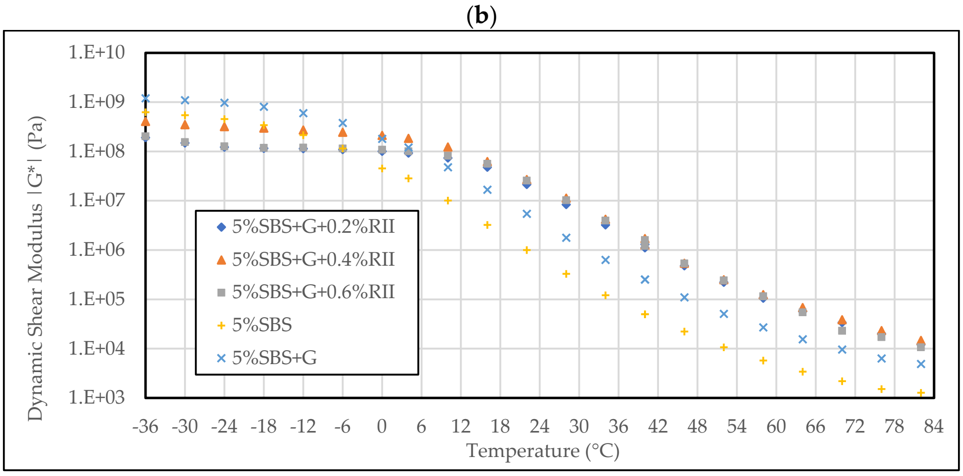


(**c**)

Figure 6. DSM values in the temperature range of −36 °C to 82 °C of binders and mastics, (**a**) with lime filler, (**b**) with basalt filler, (**c**) with granodiorite filler.

One of the noticeable effects of the use of imidazoline is a reduction in the DSM value at negative temperatures and an increase at positive temperatures compared to mastic without the addition of imidazoline. However, differentiating the imidazoline content between 0.2% and 0.6% did not significantly affect the DSM values.

The exact percentage changes in the DSM values compared to mastic without the addition of imidazoline are presented in Table 3.

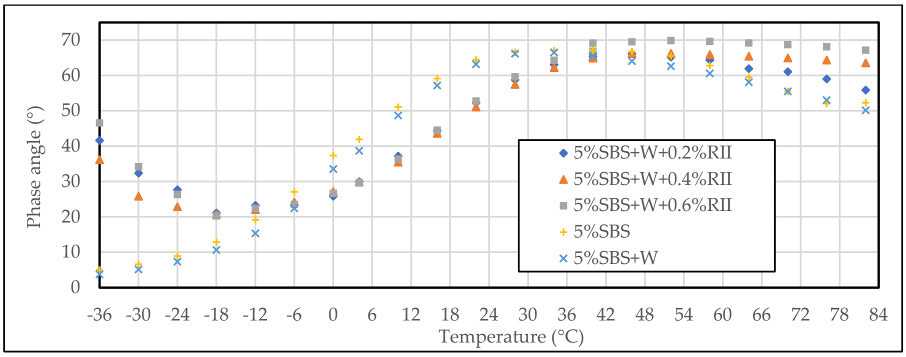
Table 3. Summary of changes in the DSM values compared to mastic without the addition of imidazoline.

DSM Change in Comparison to Mastics without Imidazoline (%) at 82 °C			
Imidazoline	**Limestone**	**Basalt**	**Granodiorite**
0.2% RII	591.49	476.95	147.06
0.4% RII	362.76	412.82	199.71
0.6% RII	359.33	333.60	117.01
DSMChange in Comparison to Mastics without Imidazoline (%) at 0 °C			
0.2% RII	−15.30	−23.47	−44.24
0.4% RII	16.26	50.38	17.57
0.6% RII	−4.71	9.80	−39.78
DSM Change in Comparison to Mastics without Imidazoline (%) at −36 °C			
0.2% RII	−79.28	−81.74	−84.14
0.4% RII	−79.16	−62.76	−66.12
0.6% RII	−85.13	−72.92	−83.10

3.1. Results of the Determination |G| and δ of Asphalt Binders and Mastics Tested at Temperatures from 82 °C to −36 °C*

Figure 7 shows the relationship between phase angle and temperature. There is a noticeable increase in the phase angle value at low temperatures (from −36 °C to −6 °C) compared to mastic without the additive by a maximum of approximately 40° at the lowest temperature tested. Then, at temperatures from −6 °C to 40 °C, the phase angle decreased in relation to the 5% SBS mastic (by a maximum of 13° at 16 °C). At the temperature range from 40 °C to 82 °C the situation was reversed, and the phase angle of the mastic with imidazoline increased its value in relation to the mastic without the addition of imidazoline by a maximum of 20.5° for an imidazoline content of 0.6%.

Figure 8 shows Black's diagram—DSM values as a function of phase angle. In the case of mastics with the addition of imidazoline, for the highest DSM values, the phase angle ranged from 36 to 46°, but then these values began to decrease to approximately 20°. This phenomenon can be associated with the tendency of phase angle graphs (Figure 6) in the range from −36 °C to −6 °C. For DSM with values lower than 100,000 kPa, curve values similar to those of asphalt mastic without the addition of imidazoline were observed. In the case of the lowest DSM values (in the range from 1.28 to 1000 kPa), only mastics with the addition of 0.2% imidazoline showed a beneficial reduction in the phase angle value compared to the results obtained for mastics without imidazoline with the addition of lime filler. For the remaining mastics, the function curves coincided with each other, but it can be noted that for all mastics, the highest phase angle values were obtained for 0.6% imidazoline content in the mastic.



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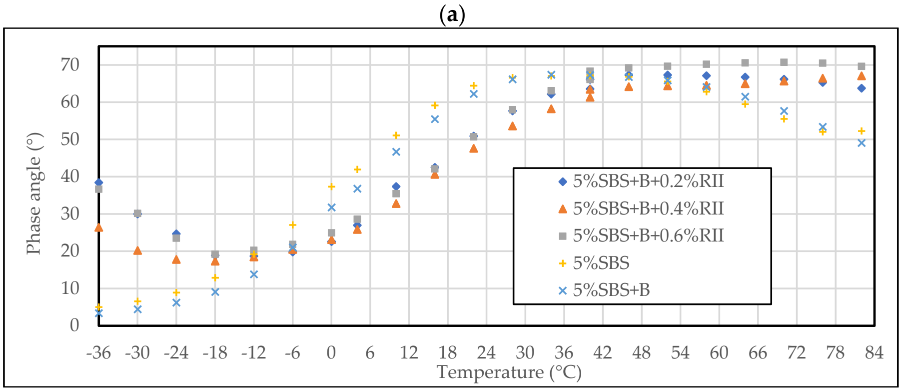

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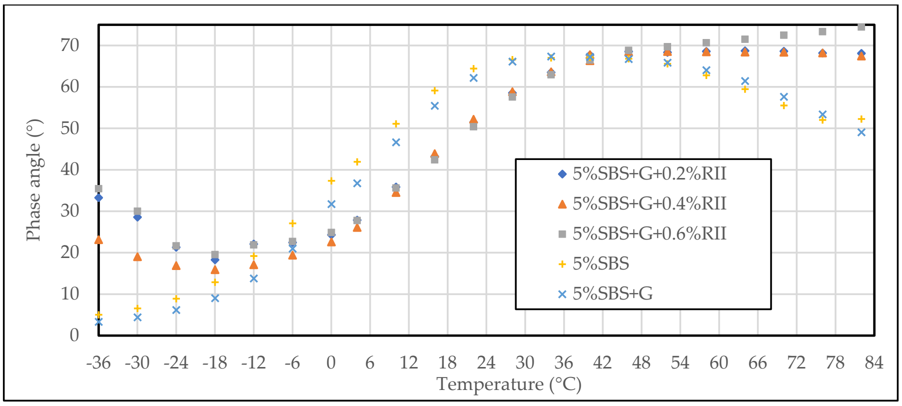

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Figure 7. Phase angle values of asphalt binders and mastics, (**a**) with limestone filler, (**b**) with basalt filler, (**c**) with granodiorite filler.

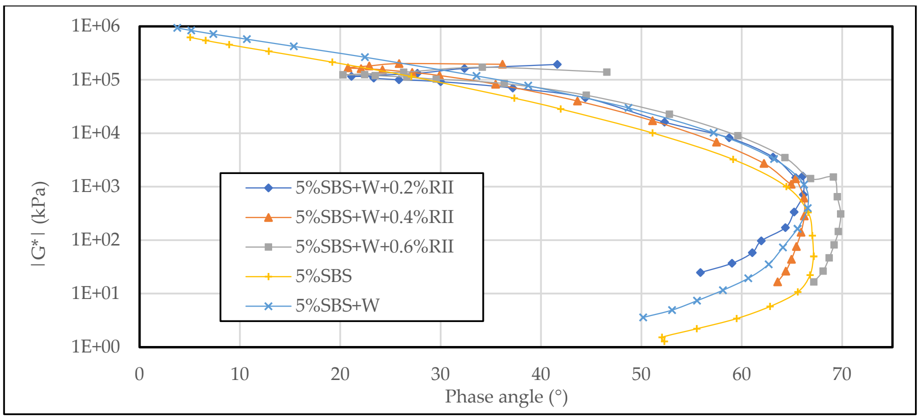

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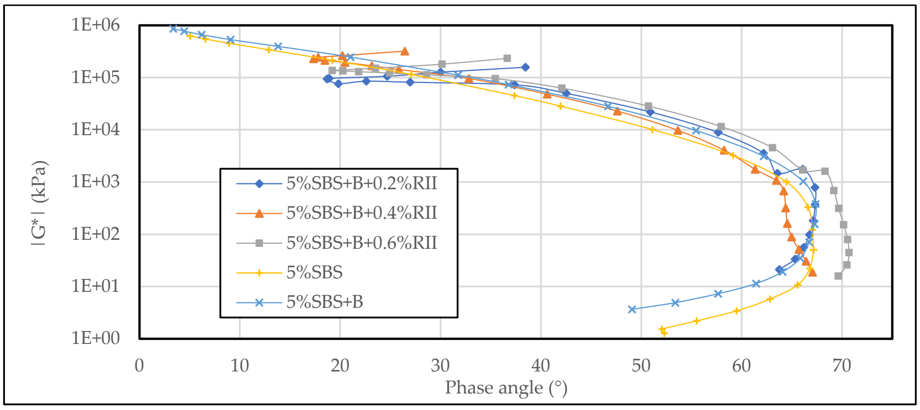
Figure 8. *Cont.*



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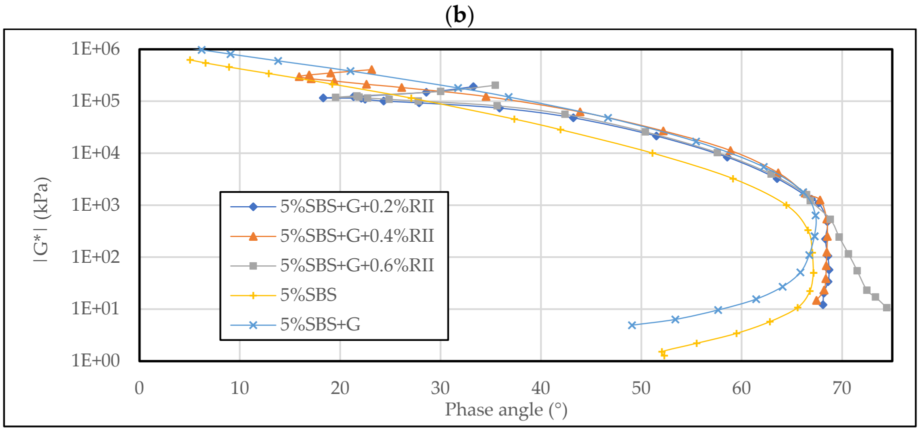

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Figure 8. Black's diagrams of asphalt binders and mastics, (**a**) with limestone filler, (**b**) with basalt filler, (**c**) with granodiorite filler.

3.2. Results of Testing of Asphalt Binders and Mastics Using the Multiple Stress Creep Recovery Method

Figure 9 shows the J_{nr} values for the highest stress values (10 kPa) at 70 °C obtained in the MSCR test. The results are shown in a comparison between bitumen modified with 5% SBS and mastics. A significant difference was observed in the parameter values for the binder compared to all tested mastics. The addition of the filler stiffened the material, as evidenced by J_{nr} values less than 0.001 kPa^{-1}. The addition of imidazoline increased the parameter values, but only slightly (it only reached the value of 0.001 kPa^{-1} for some mastics). Therefore, if the addition of imidazoline in the range below 1% increases the susceptibility of the surface to rutting, this addition should only have a minor effect.

A parameter called the "rutting factor" was also analyzed—expressed as the quotient of DSM to sin δ (Table 4). Its values were presented in comparison with J_{nr}. Rutting factor values increased by several dozen kPa for each of the three types of mastics, which is a significant difference in contrast to the trends observed for the J_{nr} parameter. This can be considered a positive phenomenon, as increased values of this parameter indicate less rutting of the pavement.

Table 4. Summary of the rutting factor and J_{nr} values at 70 °C with a stress value of 10 kPa.

Tested Mastic	J_{nr} (kPa^{-1})	|G*|/sin δ (kPa)
5% SBS + W	0.0006	8.95
5% SBS + W + 0.2 RII	0.0006	66.85
5% SBS + W+ 0.4 RII	0.0009	48.49
5% SBS + W + 0.6 RII	0.0009	61.21
5% SBS + B	0.0003	8.89

Table 4. *Cont.*

Tested Mastic	J_{nr} (kPa^{-1})	|G*|/sin δ (kPa)
5% SBS + B + 0.2 RII	0.0004	51.38
5% SBS + B + 0.4 RII	0.0005	56.27
5% SBS + B + 0.6 RII	0.0006	47.34
5% SBS + G	0.0011	11.42
5% SBS + G + 0.2 RII	0.0012	36.41
5% SBS + G + 0.4 RII	0.0007	41.62
5% SBS + G + 0.6 RII	0.0012	24.22

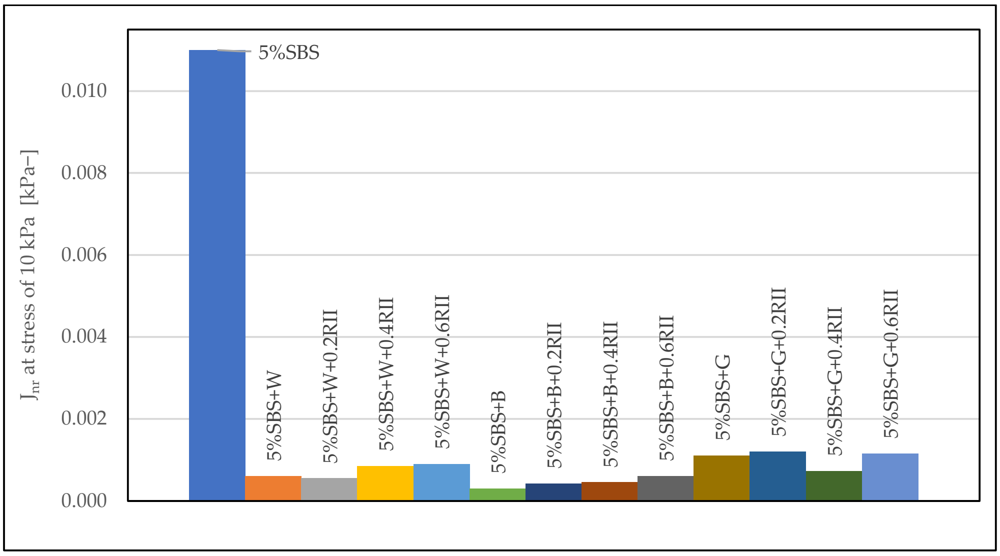

Figure 9. J_{nr} values of mastics at 70 °C and a stress value of 10 kPa.

4. Discussion

Previous research on the use of imidazolines as an addition to asphalt binders [9] proved that the use of higher contents (1–5%) may result in a radical reduction in resistance to rutting. The aim of the research presented in this article was to check the influence of the addition of imidazolines (in the range of 0–0.6% of the binder mass) to asphalt binder modified with SBS copolymer on the properties of mastics. It was assumed that the addition of imidazoline to modified asphalt binder in an amount less than 1% would not significantly increase the susceptibility to rutting of asphalt mixtures made with these binders. Based on the results of the DSM determination, a positive effect of imidazoline on the properties of asphalt binder with fillers was established when comparing its results with the values for mastics without the addition of imidazoline. At negative temperatures, mastics with the addition of imidazoline have lower DSM values, while at positive temperatures, the values increase, which suggests greater resistance of mastics to low-temperature cracking and rutting, respectively. The viscoelastic properties of mastics with the addition of imidazoline, therefore, approach those of an "ideal" binder (Figure 10). However, no significant differences were found between the DSM values and the imidazoline content, which suggests that the use of only 0.2% imidazoline satisfactorily improves the viscoelastic properties of asphalt mastics.

The phase angle values of mastics with the addition of imidazoline in relation to mastics without the addition should be analyzed taking into account three temperature ranges. Only in the temperature range from −6 °C to 40 °C was a decrease in the phase angle value of mastics with the addition of imidazoline found, which is beneficial in the context of road pavement operation, as it is associated with a greater share of the elastic properties of the binder. Taking into account that this temperature range is the most common during the use of the surface (in a moderate climate), imidazoline can be considered an additive that has a positive effect on its resistance to low-temperature cracking. The highest phase angle values at 82 °C were recorded for the imidazoline content of 0.6%, while the lowest for 0.2%, which additionally justifies the use of the lowest of the three imidazoline contents used in the tests as an addition to asphalt binders.

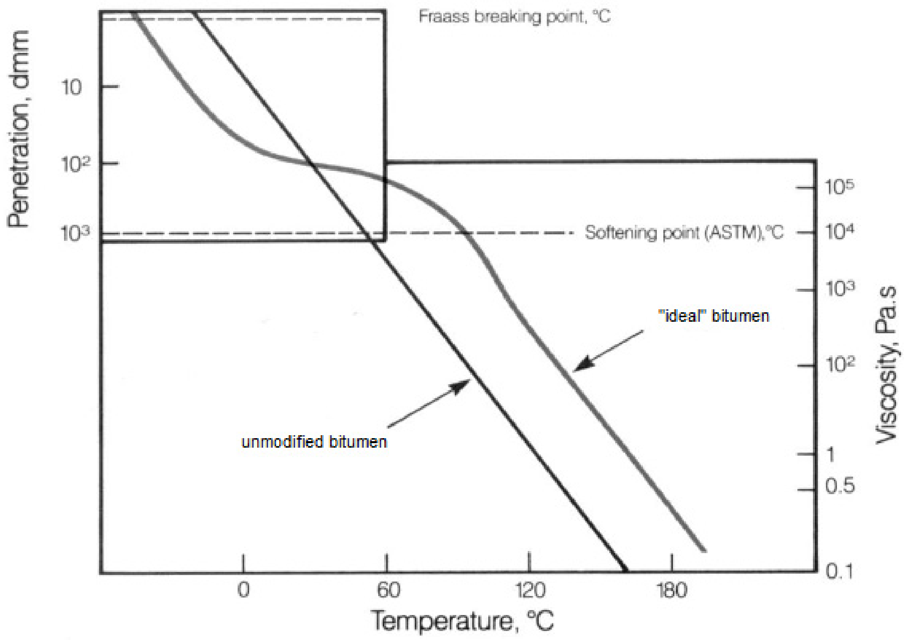

Figure 10. Idealized viscosity–temperature relationship for a polymer-modified bitumen [6].

Black's diagrams confirmed previous observations—the addition of imidazoline greater than 0.2% results in an increase in the phase angle value at the lowest DSM values. A positive tendency was noticed only for the addition of 0.2% imidazoline, where the phase angle decreased compared to limestone mastic without the addition of imidazoline. This is related to the phenomenon of liquefaction of the binder by the addition of imidazoline and limiting its elastic component. This process may have a negative impact on the resistance of asphalt mixture to rutting (especially at high temperatures). However, changes in the course of the curves on Black's diagrams can be observed at the highest DSM values—at negative temperatures, the mastic changes its properties to be more elastic compared to the mastic without the addition, which is a positive phenomenon in terms of the pavement resistance to low-temperature cracking.

One of the main doubts in the use of imidazoline was ensuring sufficient resistance of the surface to rutting—a few percent content of this additive made the material susceptible to permanent deformation at high temperatures. For this reason, the J_{nr} values of mastics and the rutting factor ($|G^*|/\sin\delta$) were determined. The obtained results confirmed that the addition of imidazoline with a content ranging from 0.2 to 0.6% did not significantly affect the values of the J_{nr} parameter—the results were comparable to mastics without the addition. However, an increase in the rutting factor value was noticed for mastics with the addition of imidazoline, which proves that imidazolines, in small percentages, can increase resistance to rutting. Therefore, it can be concluded that there is no high risk of the formation of ruts of increased depth in the pavement after the use of modified bitumen with the addition of imidazoline, even with a content of 0.6%. However, due to the comparable results of other determined parameters (phase angle, DSM), the addition of 0.2% imidazoline was considered sufficient.

Imidazoline derivative acts as a surfactant, improving the interaction between the filler particles and the bitumen matrix. As a result, the filler particles better disaggregate in the bitumen matrix, producing higher particle counts. The increase in the number of particles should contribute to a better structuring of particles with the formation of a gel network

(60 vol. % of particles certainly form it), increasing its stiffness and strength, which should increase the resistance to rutting. In addition, the imidazoline molecules adsorb on the surface of the filler particles and do so instead of the bitumen molecules. Bitumen molecules adsorbed on the surface of the particles have reduced mobility, which increases the glass transition temperature and brittleness of the bitumen matrix. However, the addition of the imidazoline derivative decreases the volume of adsorbed bitumen molecules, which results in an indirect increase in flexibility compared to the imidazoline-free system. In a somewhat similar way, bio-oil acts when added together with mineral filler particles to a bitumen matrix, behaving as a surfactant and improving adhesion between filler andbitumen [20].

5. Conclusions

The addition of imidazoline in the tested mastics significantly reduces technological temperatures in the production of asphalt mixtures, its incorporation and thickening, which significantly reduces CO_2 emissions into the atmosphere. The imidazoline additives added to binders used in the research are wastes from the synthesis by cyclization of diamidoamine [14]. The article analyzes the results of testing mastics made with the 5% SBS modified binder with the imidazoline additive content below 1% (0.2%, 0.4% and 0.6% were used). In order to avoid deterioration of the resistance to permanent deformation in the pavement, it is recommended not to exceed this additive content. It was concluded that

- The use of three different imidazoline contents (ranging from 0.2% to 0.6%) did not significantly affect the differences in the DSM, phase angle and J_{nr} results. For this reason, it is reasonable to consider 0.2% imidazoline in the binder as an additive improving rheological properties.
- The addition of imidazoline 0.2% significantly improved the properties of mastics at low and high temperatures, as suggested by the DSM values (values close to the ideal binder compared to asphalt mastics).
- The most beneficial effect on the elastic properties of the binder (represented by the phase angle values) was found in the temperature range from −6 °C to 40 °C, which is the temperature most frequently occurring in a moderate climate during pavement operation. Due to the increase in the phase angle values at higher temperatures, the most favorable imidazoline content is 0.2%, which may reduce the occurrence of ruts in the pavement.
- At negative temperatures, the phase angle of mastics with the addition of imidazoline increases in comparison to the results obtained on mastic samples without the addition of imidazoline—the mastic becomes more elastic, which increases its resistance to low-temperature cracking. However, it becomes more susceptible to rutting at the highest temperatures used in the tests. Only with the content of 0.2% imidazoline in the binder used in mastic was a reduction in the phase angle value observed for the highest DSM values, in contrast to higher (0.4 and 0.6%) imidazoline contents.
- Due to possible increased rutting of the pavement, the values of the J_{nr} and rutting factor parameters were determined. After comparing results with those obtained for mastic without imidazoline additives, it was not found that the imidazoline content used in the tested binders (even 0.6%) had a significant negative impact on the resistance of mastics to permanent deformation.

In subsequent studies, it is recommended to analyze the effect of imidazoline on the rheological properties of asphalt binders and mastics in a Dynamic Shear Rheometer. Preliminary tests showed that it was impossible to test the mastics in a BBR (Bending Beam Rheometer) because the material was becoming too stiff, and it was impossible to pour the samples into the mold.

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