

THE EFFECT OF DIFFERENT TYPES OF MODIFIERS ON THE PROPERTIES OF BITUMENS AND ASPHALT CONCRETE BASED ON THEM

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Abstract. Highways are extremely important for the development of the national economy and the renovation of infrastructure under wartime conditions. The main material for road pavement construction is asphalt concrete, which includes petroleum bitumen. In Ukraine, the only producer of this material is the Kremenchuk Oil Refinery (PJSC "Ukratnafta"), which necessitates the import of bitumen.

The research paper investigates the effect of various modifiers (polymeric, energy-saving, adhesive and wax additives), as well as their complex modification, on the bitumen performance of Kremenchuk Oil Refinery. The experimental evaluation of bitumen physical and mechanical properties after its modification was carried out, namely: needle penetration depth (penetration) at 25⁰ C, ring and ball softening temperature, and tensile strength. For polymer additives, their effect on bitumen elasticity was also determined. In addition, the effect of modifiers on the quality of the asphalt mixture was investigated: average density, water saturation, tensile strength at 20⁰ C and 50⁰ C, and water resistance coefficient using the rapid method.

The research results demonstrated that the modifiers have a similar effect on bitumen performance, namely, they reduce the penetration and increase the softening point, but with a different degree. All the modifiers, except for the energy-saving Veleton additive, also had a positive effect on the quality of asphalt concrete. This effect of the Veleton additive is probably due to the unusual temperature regime for bitumen modification and asphalt mix preparation, which requires additional research. Polymer additives (in particular, Elvaloy) demonstrated the greatest efficiency, significantly improving the performance of asphalt concrete.

The findings can be used to optimize asphalt mixtures, which will help to increase the durability of the road surface and effectively restore Ukraine's transport infrastructure.

Keywords: petroleum road bitumen, polymer, synthetic wax, compressive strength, water saturation, asphalt mix.

Introduction. The great stretch and fork of highways is the key to the development of the national economy. Unfortunately, the road sector of Ukraine is in a long-term crisis situation, characterized by the insufficient transport and operational condition of a significant part of the road stretch and critical deterioration of the road infrastructure. However, wartime road construction is an integral part of both defensive and humanitarian strategies.

The most common road surface is asphalt concrete. The main binder for the production of asphalt concrete mixtures is petroleum road bitumen. The content of this binder in the mixture is usually 4.5–7.0 % weight. Today, there is only one manufacturer of petroleum road bitumen in Ukraine – the Kremenchuk Oil Refinery (PJSC Ukratnafta). It is not able to meet all the country's demand for this component, so most road construction companies import bitumen. The use of domestic bitumen is a priority for Ukrainian manufacturers, both in terms of cost and delivery time. In order to improve the bitumen characteristics and, as a result, asphalt mixtures, there are modification techniques with various additives. Improving the asphalt quality results in better work performance and pavement durability. Complex additives are widely used, which can significantly improve the heat-resistant, rheological properties of modified bitumen and their adhesion to the

surface of mineral materials [1]. Therefore, the task of improving the bitumen properties of Kremenchuk Oil Refinery due to the use of modern modifiers available on the domestic market is relevant.

Analysis of researches and publications. Today, about 80 % of paved roads in the world have a top layer of bitumen-based materials, mainly asphalt concrete, which provides maximum comfort and efficiency of vehicle and passenger traffic [2].

The use of road bitumen and the operation of pavements based on it causes a number of problems, the most urgent of which are insufficiently high heat resistance and adhesion characteristics of bitumen (even if they meet the requirements of regulatory documents) [3]. Since the bitumen used in Ukraine is produced from light, highly paraffinic oil, such bitumen should be considered as modified [4].

Asphalt pavements must provide maximum standardized strength and shear resistance, resist fatigue failure, and be resistant to daily and seasonal temperature cycles throughout their service life. Today, one of the most effective technological methods to improve the quality of asphalt mixtures during their production and to provide for the preservation of the physical and mechanical characteristics of asphalt concrete unchanged during its service life is the use of polymer-modified bitumen [5].

According to the research of many scientists, polymer additives, adhesive, synthetic and natural waxes are used to modify bitumen [5-10].

Modification with polymers can achieve a positive effect on the high and low temperature characteristics and viscoelastic qualities of asphalt concrete [6]. Polymers also improve mechanical properties, increase resistance to permanent deformation, and resistance to low-temperature cracking. The most commonly used additives are copolymers, such as styrene-butadiene-styrene (SBS), ethylene vinyl acetate (EVA), styrene-ethylene-butylene-styrene (SEBS), and others. The widespread use of these types of polymers for modification is due to their thermoplastic behavior at higher temperatures and the ability to form a polymer network during cooling [7].

To obtain polymer-modified bitumen (PMB), a number of imported additives are used, such as "Elvaloy 4170" manufactured by "DuPont" Chemical Corporation, which is a copolymer of ethylene with butyl acrylate and glycidyl methacrylate, thermoplastic elastomers such as styrene-butadiene-styrene SBS "Kraton D" by "Kraton Polimers", synthetic latexes of the "Butonal NS" series by "BASF", etc.

Bitumen has insufficient cohesion (adhesion) with most stone materials used in road construction. This causes a sharp drop in the water resistance of asphalt concrete and the formation of damage on the pavement. Therefore, to improve the bitumen adhesion to stone materials and increase the water resistance of asphalt concrete, adhesive additives are added to the binder [9].

Waxes are used to modify bitumen. For example, Asphaltan A and Asphaltan B (Romonta), Licomont® (Clariant) and Sasobit® (Sasol), as well as various Biomer waxes produced by Deurex® AG (Elsteraue, Germany). Biomer waxes consist of various proportions of natural paraffin waxes, sugar cane wax and their fatty acids. Waxes are usually used as additives that change the viscosity and increase the softening point of bitumen [10].

Most often in Ukraine, adhesive and polymer additives are used separately or in combination [3, 5, 6]. The main disadvantage that slows down the increase in the production of bitumen modified with thermoplastic elastomers is their high cost (1.5–2.5 times higher than the cost of non-modified bitumen). Therefore, it is important to search for inexpensive substances that would improve the performance characteristics of bitumen, primarily adhesion [3]. In addition, an option to reduce the cost of modified bitumen is a combined modification by reducing the amount of polymer and adding either wax or adhesive additives, which cost less than the polymer.

The literature review made it possible to conclude that today there is a wide range of modifiers of petroleum road bitumen designed to improve its characteristics, and, accordingly, the characteristics of the finished asphalt mix and asphalt concrete. Determining the optimal combination of additives to provide advanced asphalt concrete performance is an urgent task that is important for the post-war reconstruction of the country, considering the destruction of the roads throughout

Ukraine.

The purpose of the study is to determine the effect of various modifiers and their complex on the bitumen and asphalt mixtures quality indicators.

Materials and methods of research. The basis was bitumen of BND 70/100 grade, supplied to the contractor in Odesa region from the Kremenchuk Oil Refinery. Its main characteristics are shown in Table 1.

Table 1 – Key indicators of BND 70/100 bitumen from Kremenchuk Refinery

Indicator	Value
Penetration depth of the needle (penetration) at 25° C, 0.1 mm	73
Softening point, °C	46.7
Tensile strength (ductility) at temperature 25° C, cm	77

The following modifiers were used:

- AVE 18 complex additive is a special additive based on a polymer mixture [11]. It is added to bitumen and requires thorough mixing at the temperature 165–175° C for 2–4 hours;
- Veleton energy-saving additive [11] has high adhesive properties, is very quickly mixed in bitumen;
- synthetic wax with the energy-saving effect [11] – a mixture of granules with a size 3 to 5 mm, mass fraction of the dust fraction up to 1 %, with a fusion point of 110° C;
- polymer ELVALOY [12] – a copolymer of ethylene-butyl acrylate-glycidyl methacrylate, with a fusion point 72° C.

Despite the manufacturers' recommendations, the same modifier amount was added to the bitumen, namely 1.5 % of the bitumen weight, to observe how the same modifier amount changes the characteristics of the original bitumen.

Each modified bitumen was used to produce a fine-grained dense asphalt mix, type A, grade I, in accordance with State Standard of Ukraine BB.2.7-319:2016 [13].

Research results. After the modification with 4 different additives in the same amount (1.5 %), the tests were carried out (according to [14-17]) and the results are given in Table 2.

Table 2 – Characteristics of the original non-modified bitumen and bitumen when modified with the additives in the quantity of 1.5 %

Name of the indicators	BND 70/100 + modifying additive, % wt					Requirements	
	Non-modified bitumen	polymeric Elvaloy	energy-saving additive Veleton	complex Veleton AVE 18	synthetic wax Veleton	State Standard 9116:2021 for the BMW brand 50/70-60	SOE 42.1 - 37641918-068:2017 for the BMW brand 40/60
		1.5%					
Homogeneity	No clots or additive particles					No clots or additive particles	
Needle penetration depth (penetration), $m \cdot 10^{-4}$ (0.1 mm) at 25 ⁰ C	73	64	57	48	53	51-70	40-60
The softening point of the ring and ball, ⁰ C	46.7	54.9	49.6	51.6	53.5	not less than 60	not less than 62
Tensile strength (ductility) $m \cdot 10^{-2}$ at 25 ⁰ C	77	54	54	45	80	not less than 6	not less than 20
Elasticity, %	-	40	-	20	-	not less than 55	-

The test results show that for the modified bitumen, the penetration and tensile strength decreased (Fig. 1), and the softening point increased (Fig. 2). In the studies of Juraj Šrámek, Matúš Kozel, Ľuboš Remek, and Ján Mikolaj, the same trend was observed when adding the SBS polymer [18]. The researchers L. Talakh, O. Koreva, O. Shymchuk, O. Andriychuk, O. Uzhegova, S. Drobyshynets, when considering the Kraton D and Calpren polymers, as well as the synthetic wax Sasobit, also found that the penetration decreased [19], and the softening temperature behind the ring and ball increased [20].

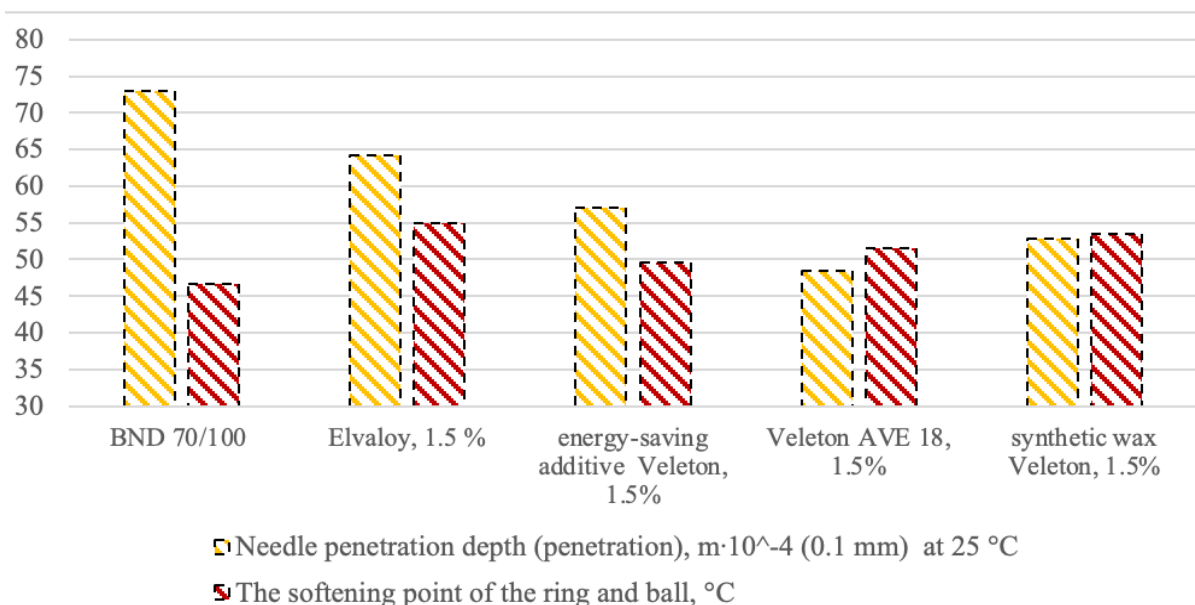


Fig. 1. Effect of the modifiers on the needle penetration depth (penetration) and softening temperature

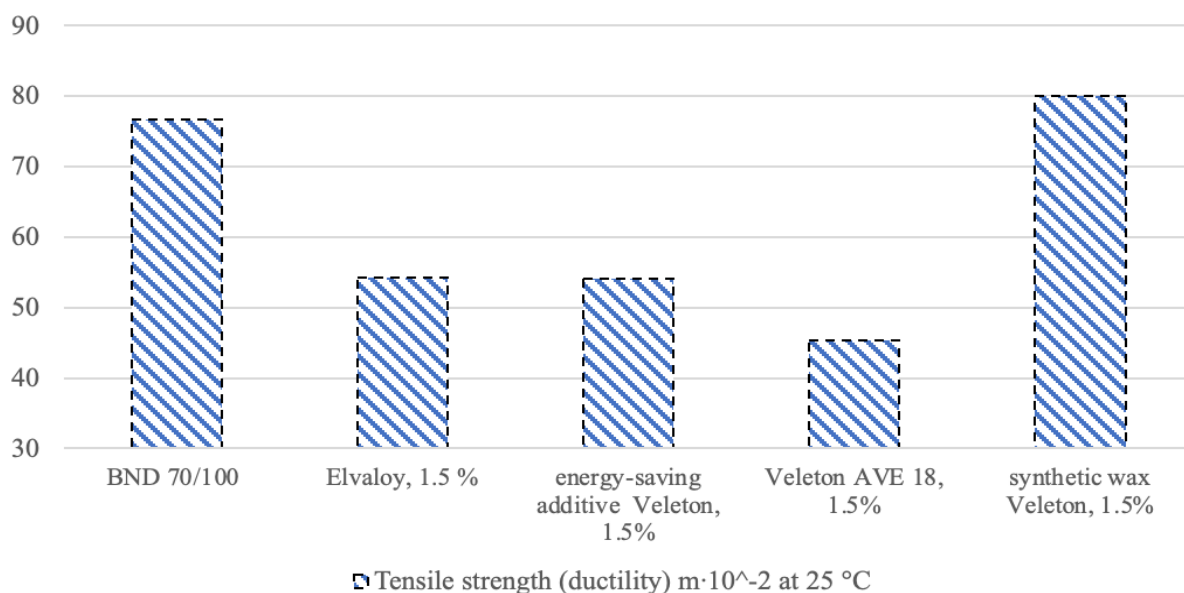


Fig. 2. Effect of modifiers on the tensile strength (ductility) at 25° C

After conducting the modification with all four additives separately, the effect of the complex modification [21] on the bitumen properties was determined. Thus, synthetic wax in the amount of 0.5 % and polymer in the amount of 1.5 % from the bitumen weight were added to the bitumen. The modification with the polymer lasted 80 minutes at the temperature 170–180° C, and with the wax 30 minutes at the temperature 150–160° C. The effect of the order of modifiers addition was also considered (Table 3).

Table 3 – Test results of the original bitumen and its modification with the combination of additives

Name of the indicators	BND 70/100 + modifying additive, % wt						Requirements	
	Non-modified bitumen	polymeric	complex	complexes			State Standard 9116:2021 for the BMW brand 50/70-60	State Standard 9133:2021 for BMKP50/70-65 grade
		Elvaloy	Veleton AVE 18	Elvaloy+ Wax Veleton	Veleton AVE 18+ wax Veleton	wax Veleton + Veleton AVE 18		
Homogeneity	No clots or additive particles							
Needle penetration depth (penetration), m·10 ⁻⁴ (0.1 mm) at 25 ⁰ C	73	64	48	54	46	52	51-70	51-70
The softening point of the ring and ball, °C	46.7	54.9	51.6	55.4	54.2	54.7	not less than 60	not less than 60
Tensile strength (ductility) m·10 ⁻² at 25 ⁰ C	77	54	45	31	48	34	not less than 6	not less than 6
Elasticity, %	-	40	20	31	20	16	not less than 55	-

Analyzing the test results, we can say that the addition of synthetic wax increases the softening point of bitumen, while reducing the penetration and tensile strength. Changing the order of adding modifying additives showed that it also has a certain effect on the penetration rate, while the softening point remains almost unchanged. As for the elasticity, it can be concluded that none of the conducted studies showed the compliance with the regulatory documentation, but this is explained by the percentage of polymer addition, since the polymer was added less than the manufacturer's recommendations. The results are summarized in Table 3 and illustrated in Figs. 3–4.

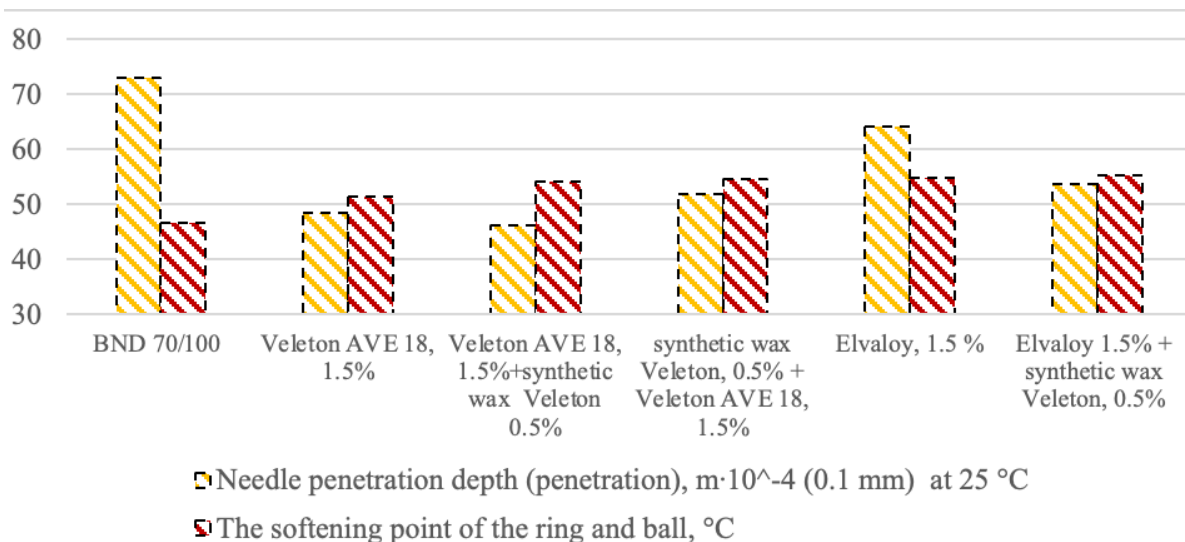


Fig. 3. Effect of modifiers on the needle penetration depth (penetration) and softening temperature by ring and ball

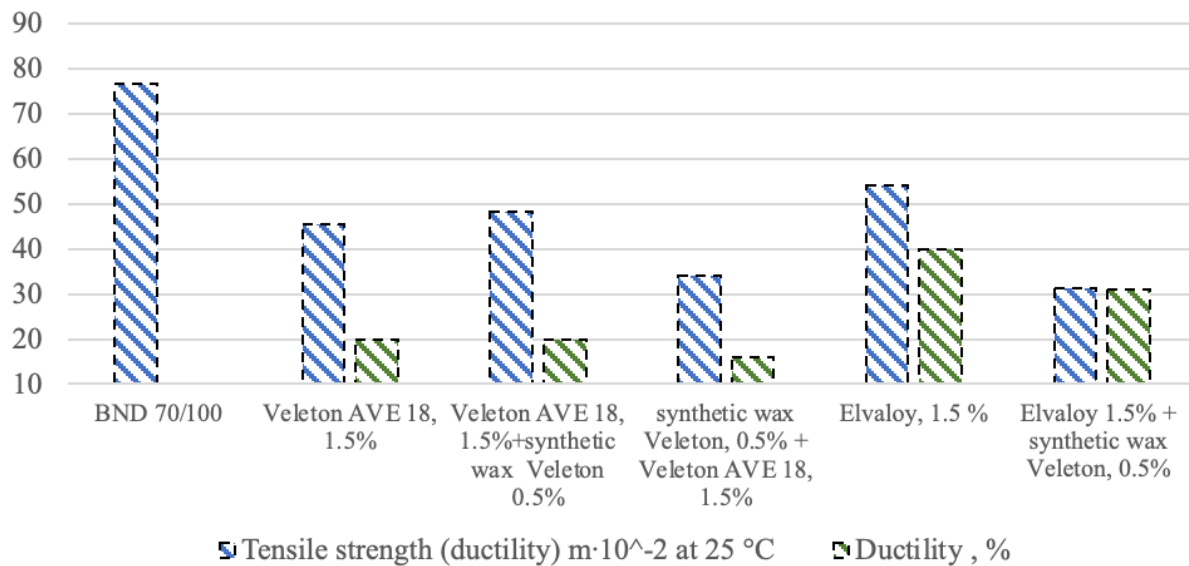


Fig. 4. Effect of modifiers on elasticity and ductility at 25°C

Hot dense fine-grained asphalt mix type A grade I, the composition of which is shown in Table 4, was made from modified bitumen samples, and the following indicators were determined: average density, water saturation, compressive strength at 20° C and 50° C; short-term water resistance coefficient [22, 23].

Table 4 – Composition of the asphalt mixture for the research

Asphalt concrete component	Material content in the mixture at 100%, % w
Crushed stone fraction 10-20 mm	22.8
Crushed stone fraction 5-10 mm	20.9
Screening fraction 0–5 mm	41.9
Aggregates made of secondary rock	9.5
BND 70/100 bitumen (without additives or modified)	4.9

The temperature conditions for the production and compaction of asphalt mixtures without and with various modifiers are shown in Table 5.

Table 5 – Main temperature conditions of manufacturing and compaction of ASG.DR.SH.A.NP.I

Binder	Modifier	Temperature, °C			
		Components of ASG.DR.SH.A.N.I		ASG.D.S.A.NP.I	
		bitumen binder	crushed stone material	mixing	compaction
BND 70/100, PJSC “Ukratnafta”	-	155	170	165	155
	Elvaloy	180	180	175	165
	Veleton. AVE 18	180	180	175	165
	Energy-saving Veleton	160	180	170	160
	Wax Veleton	155	170	165	155
	Wax + Veleton. AVE 18	165	180	170	160
	Wax + Elvaloy	165	180	170	160

The testing results of the physical and mechanical characteristics of asphalt concrete are shown in Table 6 and Figures 5–9.

Table 6 – Testing results of the physical and mechanical characteristics of asphalt concrete made from the original bitumen and its modification with additives in the amount of 1.5 %.

Name of indicators	BND 70/100 + modifying additive, % wt					Requirements	
	Non-modified	polymeric	energy-saving	complex	wax	STATE STANDARDS RD B.2.7-119:201	STATE STANDARDS D 8959:2019
		Elvaloy	Veleton	Veleton. AVE 18	Veleton		
		1.5 %					
Average density, t/m ³	2.40	2.42	2.41	2.41	2.40	not standardized	
Water saturation, % by volume	3.6	2.3	2.7	1.9	2.7	not more than 4.0	
Compressive strength, MPa, at 20° C	5.1	5.9	5.0	5.3	5.5	not more than 2.8	not more than 3.7
Compressive strength, MPa, at temperature 50° C	1.6	2.6	1.3	1.8	1.9	not less than 1.3	not less than 1.8
Water resistance coefficient by the express method	0.95	0.94	0.93	0.98	0.92	not standardized	

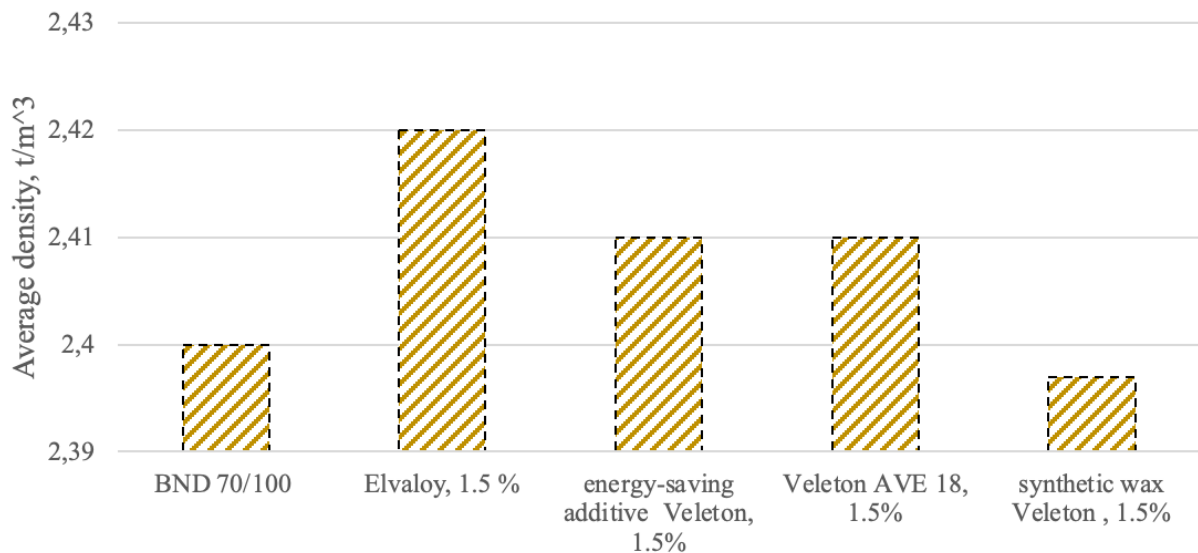


Fig. 5. Effect of modifiers on the average density of asphalt concrete

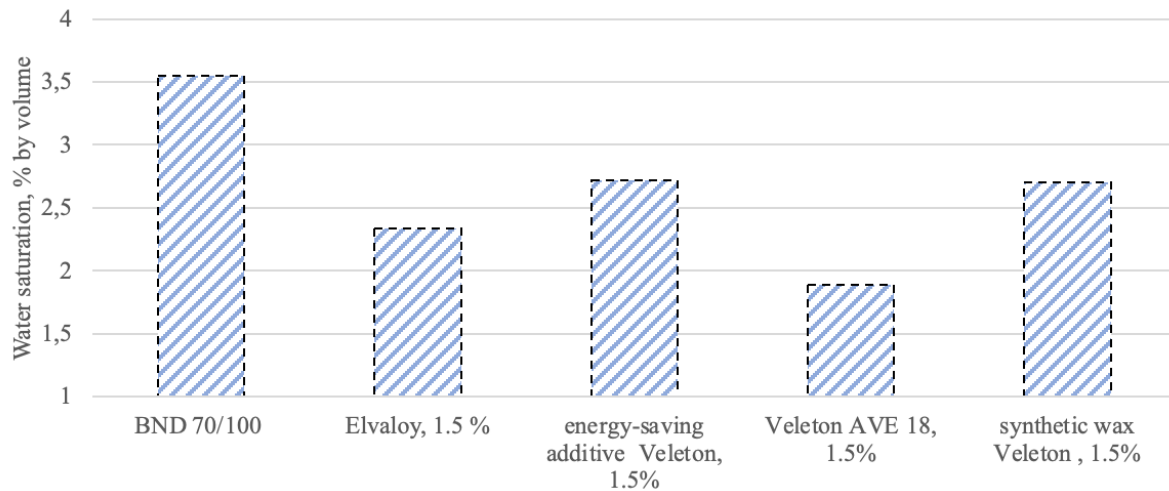


Fig. 6. Effect of modifiers on water saturation of asphalt concrete

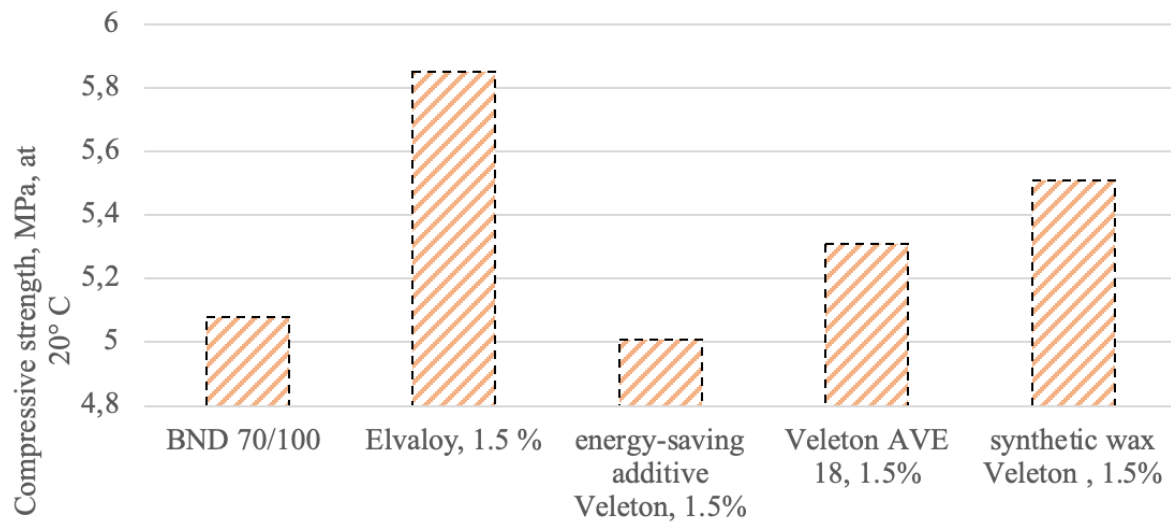


Fig. 7. Effect of modifiers on the compressive strength of asphalt concrete at 20° C

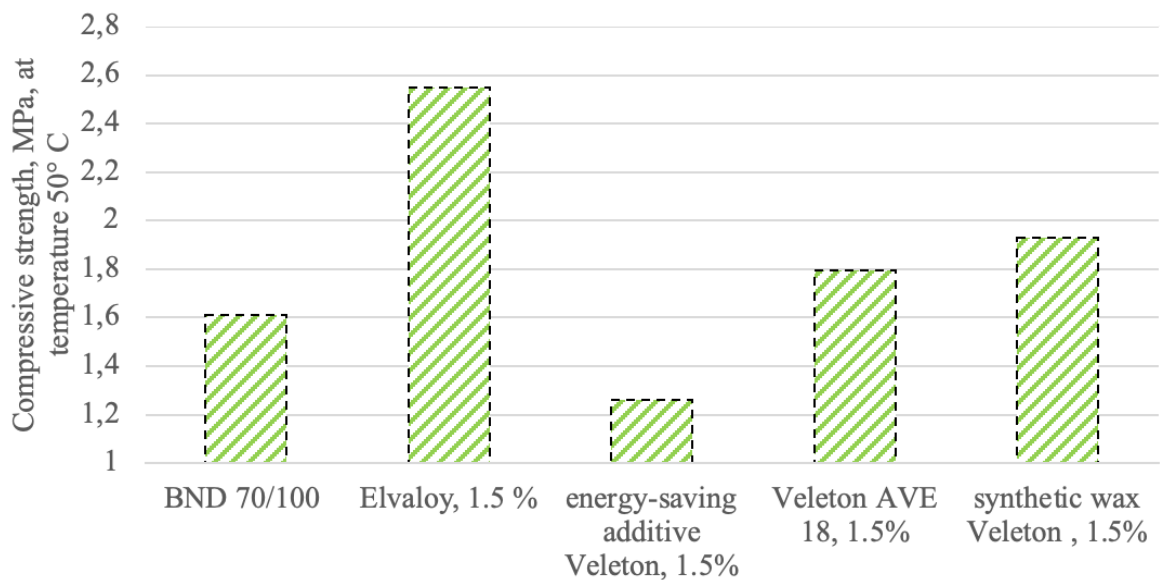


Fig. 8. Effect of modifiers on the asphalt concrete compressive strength at the temperature of 50° C

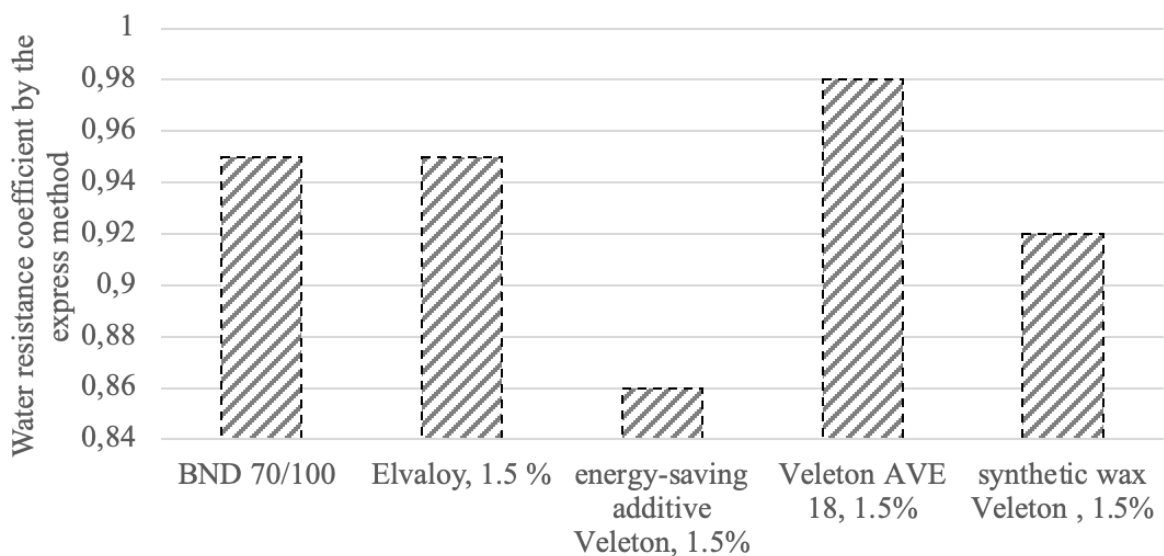


Fig. 9. Effect of modifiers on the water resistance coefficient of asphalt concrete using the express method

According to the test results, it can be concluded that, except for the energy-saving additive VELETON, all the additives have a positive effect on the characteristics of asphalt concrete, namely, decreasing of the water saturation percentage in terms of volume, increasing of the compressive strength at the temperature 20° C and at the temperature 50° C. V. Zhdaniuk, O. Volovyk, O. Makarchev in their study of the effect of synthetic waxes Licomont BS 100 and CCBit 113 AD [24], as well as S. Chuguenko in his study of the polymer Kraton D1101, synthetic wax Licomont BS 100, and cationic surfactant Wetfix BE [25] on the properties of road asphalt concrete, obtained a similar effect on both the properties of bitumen and the characteristics of the asphalt mixture. Such an effect of bitumen with the energy-saving additive VELETON is probably explained by the suboptimal temperature conditions for the preparation and compaction of the asphalt mixture. In fact, the temperature of the mixture was 160–180° C, and the compaction was 150–160° C. Moreover, the bitumen was heated to the temperature 155–165° C, and the stone material 170–180° C. The results of the studies of characteristics changes of asphalt concrete on bitumen with a combined modification are shown in Table 7.

Thus, there was the effect that in most cases, synthetic wax in the combination with a polymer showed worse results than the modification with a polymer additive only: decreasing of the compressive strength, increasing of the water saturation. This may also be due to the temperature of the asphalt mixture components, which is higher than usual for polymer-modified bitumen, namely modified bitumen 170–180° C, stone material 180–190° C, mixture temperature 160–170° C, and when compacted 150–160° C.

Table 7 – Test results of asphalt mixtures on bitumen with the complex modification

Name of indicator	BND 70/100 + modifying additive, % wt						Requirement	
	Non-modified bitume	polymeric	complex	complex			STATE STANDARD B V.2.7-119:2011	STATE STANDARD 8959:2019
		Elvaloy	Elvaloy+ Bick	Veleton. AVE 18	Bick + Veleton. AVE 18	Veleton. AVE18 +Bick		
		1.5 %	1.5 % + 0.5%	1.5%	0.5%+ 1.5%	1.5 %		
Average density, t/m ³	2.40	2.42	2.41	2.41	2.40	2.39	not standardized	
Water saturation, % by volume	3.6	2.3	2.4	1.9	3.8	3.2	not more than 4.0	
Compressive strength, MPa, at 20 °	5.1	5.9	5.2	5.3	4.4	5.1	not less than 2.8	not less than 3.7
Compressive strength, MPa, at 50 °C	1.6	2.6	2.4	1.8	2.3	1.6	not less than 1.3	not less than 1.8
Water resistance coefficient by the express method	0.95	0.94	0.98	0.98	0.95	0.97	not standardized	

Conclusions and prospects for further research.

1. The use of the analyzed modifying additives for bitumen (complex additive AVE 18 [11], energy-saving additive Veleton [11], synthetic wax with energy-saving effect [11], polymer ELVALOY [12]) has the same type of the effects, but in varied ways, namely, decreasing of the penetration and tensile strength and increasing of the softening point.

2. The most effective among the analyzed additives, both in terms of simplifying of the modification process (72° C fusion temperature) and in terms of the modified bitumen, was the ELVALOY polymer. Also, the use of this additive increases the compressive strength of asphalt concrete at the temperature 20° C by 15 % and at the temperature 50° C by 58 %. However, the

market cost of the additives varies, which should be considered when making technical decisions in real practice.

3. The use of only the energy-saving additive VELETON leads to the reduction of the compressive strength at both 50° C and 20° C, so further research is needed to determine the cause of such results.

4. In the future, it is planned to continue investigating the effect of modifying additives on the properties of bitumen and asphalt concrete, both not considered in this work and considered under other conditions, both proportional and temperature.

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**ВПЛИВ РІЗНИХ ТИПІВ МОДИФІКАТОРІВ НА ВЛАСТИВОСТІ БІТУМІВ
ТА АСФАЛЬТОБЕТОНІВ НА ЇХ ОСНОВІ**¹**Ткач С.В.,**

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Анотація. Автомобільні дороги мають критично важливе значення для розвитку національної економіки та відновлення інфраструктури в умовах воєнного часу. Основним матеріалом для будівництва дорожнього покриття є асфальтобетон, до складу якого входить бітум нафтовий дорожній. В Україні єдиним виробником цього матеріалу є Кременчуцький НПЗ (ПАТ «Укртатнафта»), що зумовлює необхідність імпорту бітуму.

У роботі досліджено вплив різних модифікаторів (полімерних, енергозберігаючих, адгезійних та воскових добавок), а також їх комплексна модифікація на характеристики бітуму Кременчуцького НПЗ. Проведено експериментальну оцінку змін фізико-механічних властивостей бітуму після його модифікації, а саме: глибини проникнення голки (пенетрації) за температури 25 °С, температури розм'якшення за кільцем та кулею, розтяжності. Для полімерних добавок також визначався їх вплив на еластичність бітуму. Крім того досліджено вплив модифікаторів на якість асфальтобетонної суміші: середню густину, водонасичення, границю міцності при температурі 20 °С та 50 °С, коефіцієнт водостійкості за експрес-методом.

Результати дослідження показали, що модифікатори мають схожий вплив на характеристики бітуму, а саме дозволяють досягнути зниження пенетрації та підвищення температури розм'якшення, проте в різній мірі. Всі модифікатори окрім енергозберігаючої добавки Veleton також позитивно впливали на показники якості асфальтобетону. Такий ефект від застосування добавки Veleton вірогідно пов'язаний з неприцятанним для неї температурним режимом модифікації бітуму та приготування асфальтобетонної суміші, що потребує додаткових досліджень. Найбільшу ефективність продемонстрували полімерні добавки (зокрема Elvaloy), які значно покращують експлуатаційні показники асфальтобетону.

Отримані результати можуть бути використані для оптимізації складів асфальтобетонних сумішей, що сприятиме підвищенню довговічності дорожнього покриття та ефективному відновленню транспортної інфраструктури України.

Ключові слова: бітум нафтовий дорожній, полімер, синтетичний віск, міцність при стиску, водонасичення, асфальтобетонна суміш.

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