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ПОРІВНЯЛЬНІ ДОСЛІДЖЕННЯ АДГЕЗІЙНИХ ВЛАСТИВОСТЕЙ ДОРОЖНЬОГО БІТУМУ, МОДИФІКОВАНОГО ДОБАВКАМИ СЕРІЇ «АДБІТ»

COMPARATIVE RESEARCH OF THE ADHESIVE PROPERTIES OF ROAD BITUMEN MODIFIED WITH ADDITIVES OF THE "ADBIT" SERIES

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Abstract: The paper presents the results of a comparative study of the influence of the cationic surface-active additive (surfactant) Adbit-R and the energy-saving additive Adbit-N on the adhesive properties of petroleum road bitumen in terms of adhesion of bitumen films to the surface of granite crushed stone grains and glass plates and the adhesion activity index. It is shown that among the studied additives, the surfactant Adbit-R provides road bitumen with the highest values of the studied indicators. In terms of adhesion of bitumen films to the surface of glass plates, the additive Adbit-R exceeds the energy-saving additive Adbit-N (at its concentration in the bitumen composition of 0.4% and 0.5% by mass), respectively, by 2.5 times and 2.0 times. By adding 0.4% and 0.5% of the energy-saving additive Adbit-N to the base bitumen, its adhesion to the surface of glass plates increases, respectively, by 3.0 and 3.7 times, compared to the original bitumen without the additive. It was found that the adhesion activity of bitumen increases with increasing concentration of the energy-saving additive Adbit-N in its composition.

Keywords: highway (motor road), road pavement, surface course, durability, asphalt concrete, long-term water resistance, road bitumen, adhesion, surface properties, surfactant, energy-saving additive, adhesion activity, adhesion index of bitumen to a mineral surface.

Introduction. One of the primary modes of failure in the asphalt concrete layer of highway pavements is the detachment of individual aggregate particles from the surface, which, during continued service, results in gradual wear and the formation of localized potholes. This type of failure is most pronounced during periods of significant moisture penetration and water saturation of the pavement due to precipitation. The process of pothole formation in asphalt concrete pavements is more pronounced on highways with high traffic intensity and speed, particularly during periods of frequent alternating service temperatures. The specified types of asphalt concrete pavement distress significantly reduce the overall durability of the road structure.

The primary cause of pothole formation in the pavement layer is the insufficient resistance of bitumen films on the surface of mineral aggregate particles to the stripping action of water. The intensive reduction in the adhesion strength of bitumen films to the mineral surface in the presence of water is attributed both to the relatively low surface activity of the mineral material and to the inadequate adhesive properties of the organic binder. Therefore, to improve the long-term water resistance and frost resistance of asphalt concrete, various technological methods are commonly employed, aimed at enhancing both the surface activity of the mineral components of the asphalt mixture and the adhesive activity of road bitumen.

Literature review. In the technology of asphalt concrete mixture production, there are three main approaches to improving the long-term water resistance of asphalt concrete. The first is aimed at increasing the surface activity of the mineral components of asphalt concrete (Kosmin, 1980; Kim & Tsoi, 1989; Kovalev, 2002). The second involves regulating the molecular–surface properties of road bitumen as a component of asphalt concrete (Kuchma, 1980; Zinchenko, 1974; Ageieva & Tytar, 1985). The third consists in simultaneously enhancing both the molecular–surface properties of bitumen and the surface activity of the mineral constituents (Ageieva, Fomenko, & Poyasnyk, 1989; Shrestha, 2004).

Numerous studies have established that, in order to improve the adhesion strength between bitumen and the surface of mineral aggregates in asphalt concrete, the aggregate surface is activated by various methods (physicochemical, mechanical, thermal, electrophysical, etc.). In asphalt concrete technology, the most widely applied practical method is the activation, or hydrophobization, of mineral filler surfaces (Kosmin, 1973) during their production, most often by grinding. For surface activation of mineral filler, different organic substances are traditionally used, characterized by good compatibility with road bitumen as the binder component of asphalt concrete. Petroleum pitch, fuel oil, mixtures of pitch and fuel oil, polymers or polymer production residues, and surfactants (either individually or in combination with other organic substances) are commonly employed for this purpose. Another known method involves activating the mineral fraction of asphalt concrete mixtures derived from acidic rocks by using aqueous suspensions of inorganic activators such as magnesia or hydrated lime, and cement. These activators contain multivalent cations (Al^{3+}, Fe^{3+}), which saturate the surface of sand particles, thereby reducing or even reversing their surface charge. To enhance the long-term water resistance of asphalt concrete, it has also been proposed to activate the mineral fraction of the mixture with aqueous surfactant solutions directly in the asphalt mixer prior to combining with bitumen (Poyasnyk, 1996). Studies (Shrestha, 2004) have shown that activation of mineral materials from acidic rocks with aqueous solutions of ferric chloride increases the adhesion index of bitumen to their surface and, consequently, improves the long-term water resistance and frost resistance of asphalt concrete. Based on investigations of the chemical composition and electro-surface properties of stone materials of different origins (Zhdanyuk & Kondrashchenko, 2000; Zhdaniuk, Shilenko, & Schevchenko, 2000), it was concluded that sufficiently high resistance of structured bitumen films to the stripping action of water, as well as high long-term water resistance of asphalt concrete, can be achieved even without prior surface activation. For this purpose, it is recommended to use mineral components from acidic rocks with the highest possible combined content of $CaO + MgO + Al_2O_3 + Fe_2O_3$ and/or with the largest possible surface area exhibiting predominantly positive or electro-neutral surface charge.

Research and practical experience have confirmed that the enhancement of the adhesive activity of road bitumen with respect to the surface of mineral aggregates in asphalt concrete can be achieved through direct modification, both at the stage of bitumen preparation and during its use in asphalt concrete mixture production. Studies (Novitskaya, Izmailova, & Matskevich, 1985; Kovalev, 1993) have focused on regulating the adhesive activity and properties of road bitumen during its preparation by means of petroleum feedstock

oxidation technology. To improve the adhesive and physico-mechanical properties of oxidized road bitumen, it has been proposed to introduce additives of aromatic hydrocarbons of various origins into the oxidized petroleum pitch (Chalokhyan & Volodin, 1980; Farberov, Lavrukhin, & Mikrin, 1983), to intensify the oxidation process through ultrasonic activation of the feedstock, and to employ ultraviolet radiation for the activation of air supplied to the oxidation apparatus, among other methods.

To improve the adhesive properties of viscous road bitumen during asphalt concrete mixture production, it has been proposed to apply foaming techniques or heating using microwave (UHF) energy, as well as to perform ultrasonic activation of hot bitumen at the inlet to the asphalt mixer. These technological measures contribute to enhancing the adhesion of road bitumen to the surface of the mineral components of asphalt concrete (Vendrikhovski, 1995; Zinchenko, 1974).

At present, in Ukraine, the most widely applied technology for enhancing the adhesive activity of road bitumen involves the use of cationic surfactant additives (Kuchma & Babinets, 1975; Fomenko & Holovko, 1980; Kudriavtseva-Valdes, 2010). The addition of surfactants directly into bitumen increases the resistance of bitumen films to the stripping action of water on the surface of mineral aggregates in asphalt concrete (Kyrylova & Chetaiev, 1973). The improvement of bitumen adhesion to the surface of mineral aggregates through the incorporation of various adhesion additives has been established in studies (Solomentsev, Krut, & Malyar, 1999; Pechenyi, 1967; Shemonayeva, Gokhman, Pankov, & Latysheva, 1986). It has been established that the optimal concentration of surfactants in road bitumen depends not only on the efficiency of the surfactant itself and the initial adhesive activity of the base bitumen, but also on the chemical and mineralogical composition and surface activity of the mineral material in contact with the bitumen. When selecting surfactants to improve the long-term water resistance and frost resistance of asphalt concrete, process engineers traditionally perform comparative evaluations of their ability to increase the adhesion index of bitumen films to mineral surfaces (Korolov, 1975; Holovko, 1978). Currently, most manufacturers of both adhesion-promoting and energy-saving additives declare their capacity to improve the adhesion of bitumen to mineral aggregates in asphalt concrete, as well as their contribution to energy efficiency. However, it is well known that, according to their primary functional purpose, surfactants and energy-saving additives belong to different classes of modifying agents. Evidently, they differ in composition, structure, and properties, and therefore should influence the enhancement of the adhesive activity of road bitumen in different ways.

The aim of this study is a comparative research of the effectiveness of cationic surfactant additives and energy-saving additives of the Adbit series in enhancing the adhesive properties of road bitumen.

Materials and methods. For the experiments, viscous petroleum road bitumen grade BND 70/100 was selected, as it is the most widely used in the production of hot asphalt concrete mixtures in Ukraine. The main physical and mechanical properties of the studied bitumen are presented in Table 1. As modifying additives, an adhesion-promoting amine-containing surfactant Adbit-R and an energy-saving additive Adbit-N, both produced by the company BARKOR, were used. According to the manufacturer, the adhesion additive Adbit-R is characterized by the following properties: density — 1.01 g/cm³, conditional viscosity measured with a VZ-246 viscometer (orifice diameter 6 mm) at 20 °C — 240 s, amine number — 42.2 mg HCl per 1 g of product, solidification temperature — 0 °C. The energy-saving additive Adbit-N has the following properties: density — 0.96 g/cm³, conditional viscosity measured with a VZ-246 viscometer (orifice diameter 6 mm) at 20 °C — 150 s, amine number — 26.2 mg HCl per 1 g of product, solidification temperature — −4 °C.

For modification of the base bitumen, the content of the adhesion additive Adbit-R was set at 0.3%, while the energy-saving additive Adbit-N was used at 0.4% and 0.5% by weight of bitumen, in accordance with the manufacturer's recommendations. Modification was carried out by adding the investigated additives to the bitumen at a temperature of 150 °C under continuous stirring. The adhesive properties of the investigated base bitumen and the bitumen modified with the studied additives were evaluated by the experimentally determined adhesion index of the bitumen film to the surface of granite aggregate particles according to method (DSTU, 2018), and to the surface of glass plates under the stripping action of water according to method (DSTU, 2021). The adhesion activity index of the bitumen containing the surfactant additive Adbit-R and the energy-saving additive Adbit-N was determined by a calculation method as the ratio of the increase in the

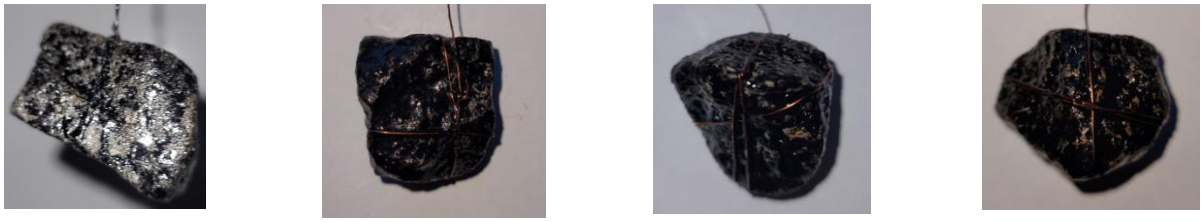
adhesion index of bitumen to the surface of glass plates to the content of the modifying additive in its composition, in accordance with (VBN, 2003).

Results and discussion. Based on the results of comparative studies of the principal properties of the base bitumen and the bitumen modified with the investigated additives, it can be stated that the needle penetration depth is a moderately sensitive parameter to the content of both energy-saving and adhesion additives in the bitumen (increasing by 13.7 % with the addition of 0.4 % and by 16.4 % with the addition of 0.5 % of Adbit-N, and by 6.9 % with the addition of 0.3 % of Adbit-R). The softening point is also only slightly affected by the content of modifying additives in the bitumen. Its reduction is less than 1 °C, which does not exceed the permissible limits of measurement reproducibility.

The results of the study on the effect of modifying additives in bitumen on its adhesion index to the surface of granite aggregate particles according to method (DSTU, 2018), as well as to the surface of glass plates according to method (DSTU, 2021), are presented in Table 1. From the representative samples of granite aggregate shown in Fig. 1 (photographic documentation taken after determining the adhesion index of the investigated bitumen to its surface), it is evident that the studied additives improve the resistance of bitumen films to the stripping action of water. It was established that the largest surface area remaining uncovered by the bitumen film after boiling in water is characteristic of the base bitumen without modifying additives. Analysis of the results presented in Table 1 indicates that the addition of 0.3% of the adhesion additive Adbit-R to the base bitumen increases its adhesion index to granite aggregate particles by 80%. In the case of incorporating 0.4% of the energy-saving additive Adbit-N into the base bitumen, the adhesion index of the bitumen film increases significantly less—by 28%. Increasing the content of Adbit-N to 0.5% in the base bitumen results in a 48% increase in the adhesion index of bitumen films. Evidently, the lower adhesion index of bitumen films modified with the energy-saving additive Adbit-N to granite aggregate particles is caused by the lower amine content in its composition, as indicated by the 38% lower amine number compared to the adhesion additive Adbit-R.

Table 1 − Properties of petroleum road bitumen
Таблиця 1 − Властивості нафтового дорожнього бітуму

No	Name and content of additive in bitumen	Properties of bitumen					
		Penetration at 25 °C, mm^{-1}	Softening point, °C	Ductility at 25 °C, cm	Bitumen adhesion index to the surface of crushed stone particles, points	Bitumen adhesion index to the surface of glass plates, %	Adhesion activity index
1	Base bitumen BND 70/100	73	47.0	> 100	2.5	12.3	-
2	Bitumen BND 70/100+0.4 % Adbit-N	83	46.5	> 100	3.2	37.2	62.3
3	Bitumen BND 70/100+0.5 % Adbit-N	85	46.0	> 100	3.7	46.0	67.4
4	Bitumen BND 70/100+0.3 % Adbit-R	78	46.0	> 100	4.5	94.0	272.3

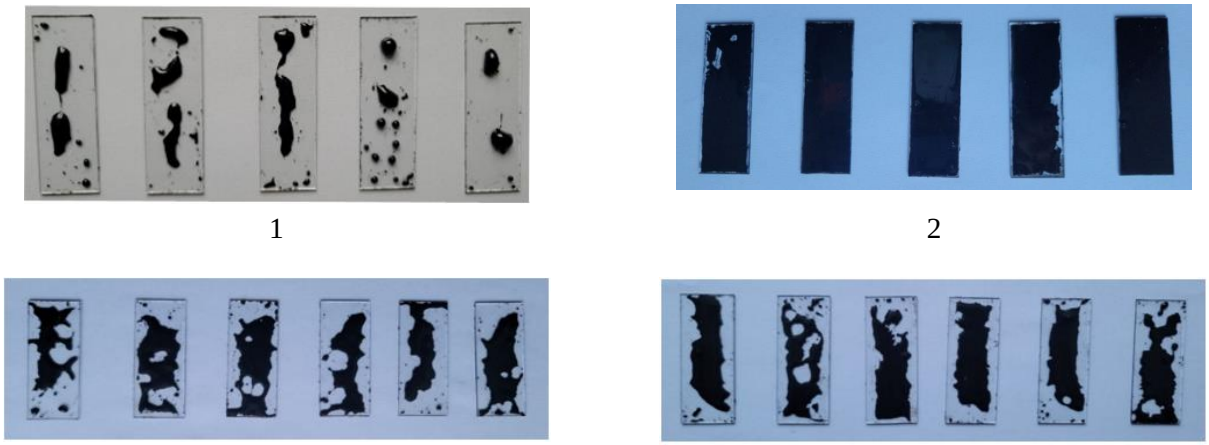


1 2 3 4

Figure 1 – General view of representative samples of granite crushed stone after testing: 1 (base bitumen BND 70/100), 2 (bitumen with 0.3 % Adbit-R), 3 (bitumen with 0.4 % Adbit-N), 4 (bitumen with 0.5 % Adbit-N)

Рисунок 1 – Загальний вигляд репрезентативних зразків гранітного щебеню після випробувань: 1 (бітум на основі БНД 70/100), 2 (бітум з 0,3% Adbit-R), 3 (бітум з 0,4% Adbit-N), 4 (бітум з 0,5% Adbit- N)

The general appearance of the samples after determining the adhesion index of the investigated bitumen films to the surface of glass plates is shown in Fig. 2.



1 2

3 4

Figure 2 – General view of samples after testing: 1 (base bitumen BND 70/100), 2 (bitumen with 0.3 % Adbit-R), 3 (bitumen with 0.4 % Adbit-N), 4 (bitumen with 0.5 % Adbit-N)

Рисунок 2 – Загальний вигляд зразків після випробувань: 1 (базовий бітум BND 70/100), 2 (бітум з 0,3% Adbit-R), 3 (бітум з 0,4% Adbit-N), 4 (бітум з 0,5% Adbit-N)

From the experimental results presented in Table 1 and Fig. 2, it is evident that the addition of the surfactant Adbit-R and the energy-saving additive Adbit-N to bitumen grade BND 70/100 increases the resistance of its films to the stripping action of water on the surface of glass plates, which is consistent with the results of determining the adhesion index of bitumen modified with the investigated additives to granite aggregate particles. The base bitumen exhibits the lowest adhesion index to glass plate surfaces (average value only 12.3%). The addition of 0.3% of the cationic surfactant Adbit-R to the base bitumen leads to the greatest increase in adhesion to glass plates (94%). Incorporation of the energy-saving additive Adbit-N results in a significantly smaller increase in adhesion compared to Adbit-R. At 0.4% content of Adbit-N, the adhesion

index rises to 37.2%, while increasing its content to 0.5% further raises the adhesion index to 46%. Analysis of the experimental data shows that bitumen with 0.3% Adbit-R, 0.4% Adbit-N, and 0.5% Adbit-N exceed the base bitumen without additives in adhesion to glass plates by factors of 7.6, 3.0, and 3.7, respectively. At the same time, bitumen with Adbit-R surpasses bitumen with 0.4% and 0.5% Adbit-N in adhesion to glass plates by factors of 2.5 and 2.0, respectively. Comparative analysis of the bitumen modified with the investigated additives by the adhesion activity index (Table 1) indicates that the highest value is characteristic of bitumen with the cationic surfactant Adbit-R, while the lowest value corresponds to bitumen with 0.4% Adbit-N. The data in Table 1 also show that with increasing content of Adbit-N in the base bitumen, its adhesion activity index rises. According to this index, bitumen with Adbit-R exceeds bitumen with 0.4% and 0.5% Adbit-N by factors of 4.4 and 4.0, respectively. Meanwhile, bitumen with 0.5% Adbit-N exceeds that with 0.4% Adbit-N by only 8.2%.

Conclusions. It has been experimentally confirmed that the addition of the surfactant Adbit-R to petroleum road bitumen, which by its primary functional purpose is intended to enhance its adhesive properties, significantly increases the adhesion index of bitumen films to the surface of granite aggregate particles and glass plates, as well as the adhesion activity index, which are the main standardized criteria of its effectiveness. It has been established that the addition of the energy-saving additive Adbit-N to petroleum road bitumen, which by its primary functional purpose is intended to reduce energy consumption during the preparation of asphalt concrete mixtures and improve their compactability, also contributes to a certain increase in its adhesive properties. It has been shown that with increasing concentration of the energy-saving additive in bitumen, its adhesive properties improve slightly. However, in terms of the adhesion activity index, bitumen with 0.5% Adbit-N is significantly inferior to bitumen with 0.3% of the cationic surfactant Adbit-R (the adhesion activity index is four times lower). Evidently, the lower level of adhesion activity of Adbit-N compared to Adbit-R may be compensated by the simultaneous addition of both investigated additives to bitumen. In the case of combined addition, the concentration of Adbit-R should be determined taking into account the adhesion index of the base bitumen containing Adbit-N with respect to mineral surfaces. The cationic surfactant may not be required if the base bitumen with the energy-saving additive already exhibits high initial adhesion activity and if the indicators of long-term water resistance and frost resistance of asphalt concrete produced on its basis meet the requirements of current national standards.

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ПОРІВНЯЛЬНІ ДОСЛІДЖЕННЯ АДГЕЗІЙНИХ ВЛАСТИВОСТЕЙ ДОРОЖНЬОГО БІТУМУ, МОДИФІКОВАНОГО ДОБАВКАМИ СЕРІЇ «АДБІТ»

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Анотація. В роботі наведено результати порівняльного дослідження впливу катіоноактивної поверхнево-активної добавки (ПАР) Адбіт-Р та енергозберігаючої добавки Адбіт-Н на адгезійні властивості нафтового дорожнього бітуму за показниками зчеплюваності плівок бітуму з поверхнею зерен гранітного щебеню і скляних пластин та показником адгезійної активності. Показано, що серед досліджуваних добавок ПАР Адбіт-Р забезпечує дорожньому бітуму найбільші значення досліджених показників. За показниками зчеплюваності плівок бітуму з поверхнею скляних пластин добавка Адбіт-Р перевищує енергозберігаючу добавку Адбіт-Н (за її концентрації у складі бітуму 0,4 % та 0,5 % за масою), відповідно, у 2,5 раза та 2,0 раза. За додавання до складу вихідного бітуму 0,4 % та 0,5 % енергозберігаючої добавки Адбіт-Н показник його зчеплюваності з поверхнею скляних пластин зростає, відповідно, у 3,0 та 3,7 раза, порівняно з вихідним бітумом без добавки. Встановлено, що показник адгезійної активності бітуму зростає зі збільшенням концентрації енергозберігаючої добавки Адбіт-Н у його складі.

Ключові слова: автомобільна дорога, дорожній одяг, шар покриття, довговічність, асфальтобетон, тривала водостійкість, дорожній бітум, адгезія, поверхневі властивості, поверхнево-активна речовина, енергозберігаюча добавка, адгезійна активність, показник зчеплюваності бітуму з мінеральною поверхнею.

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