



Study and Evaluation of the Adhesion Properties of Alternative Road Bitumen to Various Mineral Materials

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ABSTRACT: This study focuses on evaluating the adhesion properties of alternative road bitumen samples prepared from oil sludge, gossypol resin, and waste methyl diethanolamine (MDEA) when combined with different mineral materials such as marble, crushed granite, and granite gravel. Bitumen samples with varying MDEA content were developed and tested according to GOST 11508-74 requirements. The degree of coating of mineral surfaces was measured through boiling tests, and the adhesion performance was assessed visually based on coating percentages. The results demonstrated that the adhesion of bitumen samples improved with increasing MDEA content. Among the mineral aggregates, marble showed the highest adhesion, followed by crushed granite and granite gravel, consistent with the mechanical and electrostatic theories of adhesion. The surface morphology, roughness, and electrochemical properties of the mineral materials significantly influenced the interaction with bitumen. Additional analysis based on chemical bonding and thermodynamic adhesion theories confirmed that strong and stable bonds form at the bitumen–mineral interface, enhancing adhesion performance. The developed alternative bitumen samples exhibited high potential for use in road construction, providing better durability and resistance to environmental conditions compared to conventional petroleum-based bitumen.

KEY WORDS: Alternative road bitumen, oil sludge, gossypol resin, methyl diethanolamine (MDEA), adhesion, mineral materials, asphalt concrete mixtures, mechanical adhesion theory, electrostatic adhesion theory, chemical bonding, surface roughness.

I. INTRODUCTION

Currently, asphalt-bitumen composites based on petroleum-derived road bitumen are widely used for paving automobile roads. The demand for high-quality asphalt mixtures is increasing year by year. Considering the shortage of petroleum-based road bitumen, developing alternative road bitumen formulations that can substitute it has become of significant importance [1].

Globally, special attention is being given to the development of ingredients that shape the structure of alternative road bitumen obtained from petroleum, gas, and chemical industry residues. These efforts aim to improve technological and physico-mechanical properties, enhance resistance to cold and heat, increase elasticity, prolong service life, and reduce production costs [2–4].

In Uzbekistan, significant scientific and practical work is underway to develop new ingredients and modifiers with high-performance characteristics for the production of alternative road bitumen. This involves studying their physicochemical properties, developing technologies to obtain alternative road bitumen based on gossypol resin, oil sludge residues, and ethanolamine group additives, and preparing high-quality asphalt mixtures for use in road construction.

The New Uzbekistan Development Strategy sets important tasks, such as "fully utilizing available opportunities to further develop the potential of local industrial sectors and introducing standards that meet external market and international requirements." In this context, asphalt mixtures produced using alternative road bitumen aim to increase the durability of automobile roads. These asphalt mixtures not only maintain quality during hot and cold seasons but also demonstrate strong resistance to the pressure of heavy transport vehicles [5–7].

Furthermore, ongoing scientific research focuses on creating new ingredients and modifiers to improve the properties of road bitumen, studying their physicochemical properties, developing modification technologies, and evaluating the physicochemical, technological, physico-mechanical, and operational properties of the resulting road construction bitumen for practical application [8–10].



II. SIGNIFICANCE OF THE SYSTEM

The necessity of using up to 9% of waste methyl diethanolamine (MDEA), which is no longer suitable for regeneration, has been substantiated to improve the adhesion properties of bitumen to mineral materials. A high-quality asphalt mixture was prepared and laid using alternative road bitumen obtained based on oil sludge, gossypol resin, and waste methyl diethanolamine (MDEA). The technology for producing alternative road bitumen based on oil sludge, gossypol resin, and waste methyl diethanolamine (MDEA) has been developed.

III. METHODOLOGY AND DISCUSSION

In the dissertation, the adhesion properties of alternative road bitumen samples with mineral materials were evaluated using the following method:

According to GOST 11508-74 requirements, the degree of coating of various mineral material surfaces by alternative road bitumen samples was determined.

Each selected mineral material was mixed with the alternative bitumen samples at a temperature of 150°C, then laid onto 0.25 and 0.5 mm iron meshes and immersed in a bath filled with distilled water. Each sample was boiled for 30, 60, and 90 minutes.

After completing the tests, bitumen floating on the water surface was removed with filter paper, and the mineral aggregates settled at the bottom were weighed.

Each sample was visually evaluated based on the degree of surface coverage.

Visual evaluation was carried out as follows: alternative road bitumen samples should show an index of more than 75% coverage of the surface of the mineral material. If the alternative bitumen sample has a surface coverage of less than 75% of the mineral material, we give a grade of “2” or “unsatisfactory”. If the alternative bitumen sample has a surface coverage of 76% to 85% of the mineral material, we give a grade of “3” or “satisfactory”. If the alternative bitumen sample has a surface coverage of 86% to 95% of the mineral material, we give a grade of “4” or “good”. If the alternative bitumen sample has a surface coverage of more than 96% of the mineral material, we give a grade of “5” or “excellent”.

The following mineral materials were used for preparing asphalt concrete mixtures:

1. Marble stone was used as a mineral material containing CaCO_3 . Marble consists primarily of dolomite (calcium and magnesium carbonate – $\text{CaMg}(\text{CO}_3)_2$) and calcite (calcium carbonate – CaCO_3). It may also contain impurities such as iron oxides, iron sulfides, silicates containing iron, manganese carbonates, and graphite inclusions. Marble was utilized as an analog to crushed limestone, one of the main types of crushed stone used in road construction aside from gravel and crushed granite.

Based on the research results, the following mineral materials were selected to create an adhesion environment with viscous bitumen binder:

According to acid-base conditions:

Crushed granite and gravel formed an acidic environment.

Marble stone formed a basic environment.

According to surface electric charge:

Crushed granite and gravel stones exhibited an electro-negative surface charge.

Marble stones exhibited an electro-positive surface charge.

2. Crushed granite belongs to the class of inorganic materials and is obtained by crushing rocks in mountainous areas. Its average density ranges from 1.6 to 3.2 g/cm³. Crushed granite consists mainly of quartz natural crystals, forming a granular, hard rock texture. In the study, crushed granite stones with sizes of 5–17 mm were used, which are widely applied in road and airfield pavement construction.

3. Granite gravel consists of smooth-surfaced, rounded or egg-shaped stones typically formed in river or marine environments. In the research, granite gravel fractions sized 5–17 mm were used.

IV. EXPERIMENTAL RESULTS

The experimental data on the adhesion properties of bitumen with different mineral material surfaces are presented in **Table 1**.

The compositions of the alternative road bitumen (ARB) samples are as follows:

- **MYB-1 composition:** 50% gossypol resin, 65% oil sludge, 5% MDEA
- **MYB-2 composition:** 50% gossypol resin, 60% oil sludge, 10% MDEA
- **MYB-3 composition:** 50% gossypol resin, 55% oil sludge, 15% MDEA
- **MYB-4 composition:** 50% gossypol resin, 50% oil sludge, 20% MDEA
- **MYB-5 composition:** 50% gossypol resin, 45% oil sludge, 25% MDEA
- **MYB-6 composition:** 50% gossypol resin, 40% oil sludge, 30% MDEA
- **MYB-7 composition:** 50% gossypol resin, 35% oil sludge, 35% MDEA
- **MYB-8 composition:** 50% gossypol resin, 30% oil sludge, 40% MDEA

Table 1

Study and Evaluation of the Adhesion Properties of BND-60/90 Bitumen and Obtained Alternative Road Bitumen Samples with Various Mineral Materials

Additive, %	Mineral Materials					
	Granite stones		Gravel		Marble	
	Coating Degree, %	Score	Coating Degree, %	Score	Coating Degree, %	Score
BND-60/90	76	3	74	2	78	3
MYB-1	82	3	77	3	86	3
MYB-2	86	3	84	3	90	4
MYB-3	92	4	90	4	93	4
MYB-4	89	3	87	3	89	3
MYB-5	91	4	89	3	93	4
MYB-6	93	4	90	4	95	5
MYB-7	96	5	92	4	97	5
MYB-8	92	4	88	3	91	4

Note: Explanation of scores:

- 2 — "unsatisfactory": the binder film covers less than 75% of the surface of the aggregates;
- 3 — "satisfactory": the binder film covers up to 90% of the surface of the aggregates;
- 4 — "good": the binder film covers up to 95% of the surface of the aggregates;
- 5 — "excellent": the binder film covers more than 96% of the surface of the aggregates.

Based on the data presented in **Table 1**, the adhesion graphs shown in **Figure 1** were constructed.

According to the data from **Table 1** and the graph in **Figure 1**, the adhesion properties of bitumen with crushed granite, granite gravel, and marble materials decrease in the following order: **from marble → to crushed granite → to granite gravel**.

This trend suggests that the adhesion of bitumen to alkaline rocks (such as marble) is significantly better than to rocks with an acidic environment (such as granite aggregates).

It is established that the rougher the surface texture of the selected mineral material, the higher its adhesion properties with the bitumen binder.

However, in some materials with a high surface smoothness percentage, the interaction with the bitumen binder is much lower.

For example, crushed granite shows higher adhesion to bitumen compared to granite gravel.

This behavior can be explained based on the **mechanical theory of adhesion**, where the key factors include:

- The roughness and irregularity of the mineral surface enhancing mechanical interlocking,
- The viscoelastic deformation characteristics of the bitumen,
- The energetic absorption mechanism at the bitumen–aggregate interface, leading to increased boundary strength.

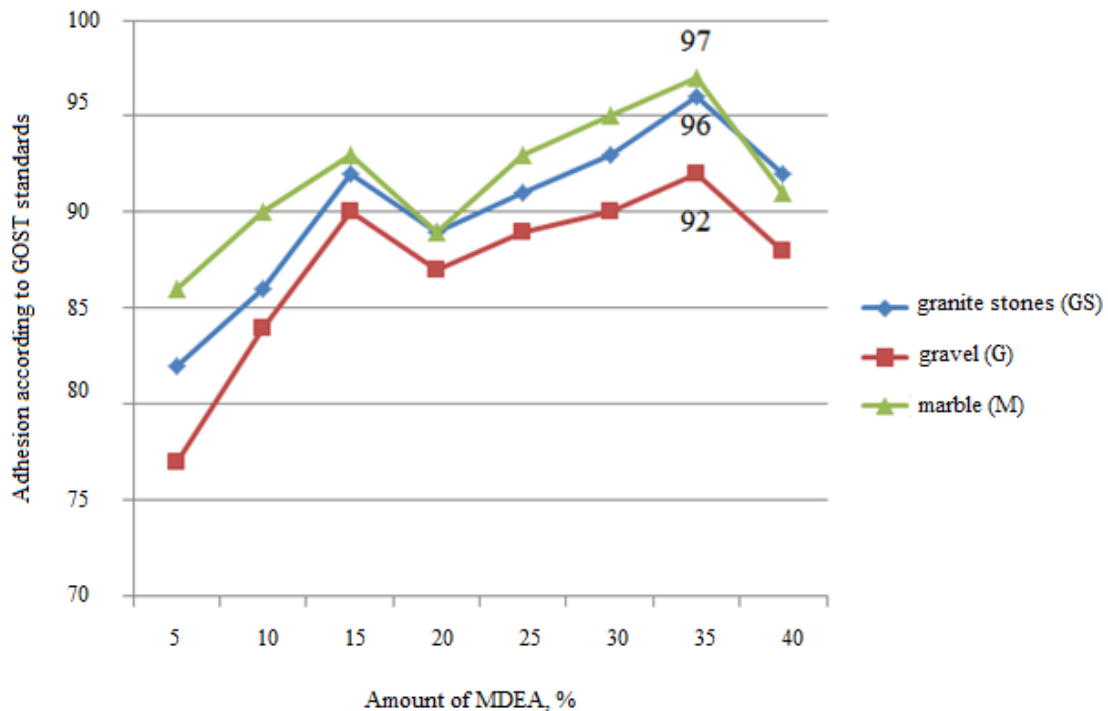


Figure 1. Adhesion of MYB samples with various mineral materials depending on the MDEA content

Considering the surface roughness of the mineral material, the primary role in this process is played by the viscous bitumen binder, since the presence of anionic and cationic polar functional groups in the bitumen composition significantly enhances its adhesion to mineral materials. In addition, the surface roughness of the mineral material increases the contact area, thereby improving adhesion properties.

The interaction between the active centers of the bitumen binder and the mineral material contributes to an increase in the effective contact surface.

According to the data presented in Table 1, the adhesion properties of bitumen are better with mineral materials that have electropositive surface characteristics (such as marble) compared to materials with electronegative surface characteristics (such as granite aggregates).

This phenomenon is explained by the electrostatic theory of adhesion.

V. CONCLUSION AND FUTURE WORK

Based on the research results, the interactions between bitumen and mineral materials were studied using the mechanical and electrostatic theories of adhesion.

Both theories emphasize the importance of the physical properties of crushed granite and granite gravel in understanding adhesion: the surface morphology can promote adhesion and induce selective adsorption, and the nature of the mineral material's surface significantly affects adhesion behavior.



In addition, the chemical bonding theory and thermodynamic theory were also thoroughly investigated, leading to the following conclusions:

According to the chemical bonding theory, stable insoluble complexes are formed at the bitumen–mineral phase interface, providing strong adhesive strength.

Based on the thermodynamic theory, a close contact between the two phases is a necessary condition for achieving good adhesion properties.

Relying on the theoretical analysis of adhesion mechanisms, it can be concluded that factors such as the smoothness, roughness, surface chemisorption properties, and other surface characteristics of mineral materials influence their potential interactions with bitumen.

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