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Mechanism of the Influence of Adhesion Additives on the Bonding Strength Between Alternative Road Bitumen and Mineral Materials

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ABSTRACT: This research investigates the mechanisms by which adhesion additives influence the bonding strength between alternative road bitumen and mineral aggregates. Alternative bitumen was synthesized using oil sludge, gossypol resin, and exhausted methyl diethanolamine (MDEA). Various testing methods, including water emulsion tests, ultrasonic adhesion measurements, and pull-off tests, were employed to evaluate adhesion properties. Experimental results showed that the incorporation of nitrogen- and oxygen-containing functional groups from additives significantly improved adhesion and wetting properties. Gossypol resin exhibited stronger hydrogen bonding with mineral surfaces, enhancing asphalt concrete durability. The use of methyl diethanolamine at concentrations up to 35% further improved adhesion characteristics according to the GOST 11507-74 standard. The study also demonstrated that wetting contact angles decreased as additive content increased, confirming enhanced surface interactions. These findings contribute to the development of sustainable, high-performance road materials and offer new insights into optimizing the interface between alternative bitumen binders and mineral aggregates for improved pavement longevity.

KEY WORDS: Alternative bitumen, adhesion additives, gossypol resin, oil sludge, MDEA, bonding strength, wetting angle

I. INTRODUCTION

The rapid development of transport infrastructure and the increasing demand for high-performance road materials have intensified research into alternative sources of bitumen. Traditionally, road bitumen is produced as a by-product of petroleum refining; however, the depletion of petroleum resources, coupled with growing environmental concerns, has necessitated the search for sustainable and efficient alternatives [1, 2].

Alternative bitumen production focuses on utilizing non-petroleum raw materials such as bio-oils, industrial waste, recycled plastics, and other organic or inorganic precursors. These alternatives aim to reduce the environmental footprint associated with traditional bitumen production, enhance material performance, and support the principles of a circular economy. Moreover, innovations in chemical modification and polymer blending have expanded the possibilities of tailoring the properties of alternative bitumen to meet specific performance requirements, such as improved temperature susceptibility, durability, and resistance to aging [3-5].

Recent studies have explored various approaches to synthesizing alternative road binders, including pyrolysis of biomass, upgrading of heavy oils, and chemical recycling of polymers. Each method offers distinct advantages and challenges in terms of production costs, material properties, and long-term performance under diverse climatic and loading conditions [6-8].

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The development of alternative bitumen is not only an urgent technological task but also a strategic initiative aligned with global sustainability goals. It holds the potential to minimize dependency on fossil fuels, lower greenhouse gas emissions, and create innovative pathways for waste utilization [9].
This paper presents an overview of current methods for obtaining alternative road bitumen, evaluates their key characteristics, and discusses prospects for their practical application in modern road construction [10].

II. SIGNIFICANCE OF THE SYSTEM

The scientific significance of the research results is explained by the development of a scientific basis for the use of alternative road bitumen, produced from residual materials of local oil and gas processing plants and secondary raw materials of the oil-fat industry, for the construction of asphalt concrete pavements.
The practical significance of the research results lies in the production of alternative road bitumen based on oil sludge, gossypol resin, and exhausted methyl diethanolamine (MDEA), as well as in the preparation of high-quality asphalt mixtures using this alternative binder.

III. METHODOLOGY AND DISCUSSION

The adhesion properties of road bitumen ensure the ability to bond with mineral aggregates such as stone (gravel) and sand fillers. These properties are critically important for the long-term performance and durability of road pavements, as they determine the cohesion of asphalt mixtures and their resistance to cracking and moisture-induced damage. Below are the main methods used for determining the adhesion characteristics of road bitumen:
1. "Petrol" Water Emulsion Test (Bred Method).
This method measures the adhesion strength of bitumen to various mineral fillers and is often used to evaluate the moisture resistance of road bitumen.

Procedure:
Sample Preparation: Bitumen is heated to 150°C.
Emulsion Application: A water emulsion containing a specific solvent is added to the bitumen.
Surface Coating: The resulting mixture is applied to a mineral surface (e.g., crushed stone) simulating a road pavement.
Durability Assessment: The resistance of the bitumen-coated mineral surface in water is evaluated.
Adhesion Strength Measurement: The bond strength between the alternative bitumen and the mineral material is measured after freezing and thawing cycles.
Standards: GOST 32533, ASTM D5936.

2. Ultrasonic Adhesion Method.
This method assesses the bonding strength between bitumen and minerals using ultrasonic wave propagation.

Procedure:
Sample Preparation: Asphalt mixture samples are immersed in a water-filled vessel.
Application of Ultrasonic Waves: Ultrasonic waves are directed at the sample.
Detection of Changes: Variations in the amplitude of ultrasonic waves reveal the intensity of adhesion failure.
Standards: This method is less common and is mainly applied in scientific research for detailed adhesion analysis.

3. Adhesion-Tension Test.
This method measures the tensile force required to break the adhesion between alternative road bitumen and mineral aggregates.

Procedure:
Sample Preparation: Mineral surfaces are coated with the alternative bitumen and allowed to cure.
Tensile Testing: Samples are subjected to tension using a universal testing machine until failure occurs.
Results: The test determines the bitumen's ability to maintain adhesion under external mechanical stress.
Standards: ASTM D3625, ISO 18473.

4. Water Immersion Test.
This method evaluates the impact of water on the adhesion strength of bitumen, simulating real operating conditions.

Procedure:
Sample Preparation: Alternative bitumen is applied to mineral materials.

ISSN: 2350-0328
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Water Exposure: Samples are immersed in water and subjected to cyclic temperature changes (freeze-thaw cycles).
Assessment of Adhesion Loss: The degree of adhesion deterioration between the bitumen and mineral aggregates is measured.
Standards: ASTM D3625, EN 13302.
5. Bitumen-Gravel Adhesion Test (Kendall Method).
This method assesses the adhesion properties of alternative road bitumen to different types of crushed stone and fillers.
Procedure:
Sample Preparation: Alternative bitumen and gravel samples are prepared according to established standards.
Force Application: Samples coated with bitumen are subjected to various mechanical forces to determine the bonding strength.
Result: This method accurately evaluates the quality of adhesion between bitumen and crushed stone or other mineral materials.
Standards: ASTM D3625, GOST 33161.
6. Mechanical Pull-Off Test Using a Lab Device.
This method is used to assess the adhesion of alternative road bitumen to pavement surfaces such as asphalt concrete or concrete bases.
Procedure:
Sample Preparation: The surface is coated with a layer of alternative bitumen.
Force Application: Tensile force is applied using a specialized device to detach the samples.
Result: The force required to separate the alternative bitumen from the mineral surface is measured.
Standards: ASTM D4541, ISO 4624.
7. Interfacial Tension Method.
This method is used to determine the bonding forces at the molecular level between alternative road bitumen and fillers.
Procedure:
Sample Preparation: Bonded samples of alternative bitumen and various fillers are prepared.
Tension Measurement: The method measures the molecular adhesion forces between bitumen and mineral surfaces, providing precise information on adhesion strength.
Standards: This method is typically used in scientific research to study interactions at the molecular level.

IV. EXPERIMENTAL RESULTS

It is necessary to study the tautomeric form of gossypol resin, which exists as two tautomeric forms — the lactol (II) and carbonyl (III) structures — corresponding to 2,2-di-(1,6,7-trihydroxy-3-methyl-5-isopropyl-8-aldehydonaphthyl).
The development of complex emulsion-mineral mixtures containing gossypol resin and their application as surface-active agents in road construction have been proposed and substantiated based on analytical results.
The additives studied and currently utilized enhance the surface activity of the bitumen binder, thereby improving its adhesion properties. This process is primarily achieved due to the presence of active functional groups within the molecular structure of the substances used as additives (Table 1).

Table 1
Functional groups of the additives

Additive	Functional Groups
Oil sludge	Aromatic groups, carbon and hydrogen compounds
Gossypol resin	Carbonyl, carboxyl, hydroxyl, $S_{30}N_{32}ON$ groups
Oil sludge + S	Aromatic groups, carbon and hydrogen compounds, sulfide compounds
Gossypol resin + S	Carbonyl, carboxyl, hydroxyl, sulfide compounds, $S_{30}N_{32}ON$ groups

According to the results obtained using standard methods for evaluating the adhesion properties and wetting contact angles of viscous bitumen binders, it was found that the addition of nitrogen-containing gudron (gossypol resin)

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exerts a slightly better effect on the adhesion properties of bitumen compared to additives based on oil sludge, which mainly contain oxygen-bearing functional groups (carbonyl, carboxyl, and hydroxyl groups).

Depending on the quantity of additives introduced into the bitumen, an ideal adhesion to the surface of crushed granite chips and gravel aggregates is achieved. It was observed that increasing the amount of methyl diethanolamine (MDEA) additive up to 35% enhances the adhesion properties according to the "passive adhesion method" described in GOST 11507-74.

The study of adhesion between bitumen and mineral materials showed that adhesion occurs at the interface between the two phases. The theory explaining the adhesion improvement by additives suggests that the adhesion follows the chemical adhesion theory, where polar functional groups within the additives interact with the mineral surfaces. The molecular structure of the investigated bitumen primarily consists of nonpolar long carbon chains and hydrogen-saturated rings, representing saturated hydrocarbons and aliphatic compounds present in volatile oils or paraffin fractions.

When polar functional groups are incorporated into the bitumen structure, they interact with active centers on the surface of mineral aggregates, significantly improving adhesion. The structure of gudron (gossypol resin) contains polar carbonyl groups, small amounts of carboxyl groups, and oxygen-bearing hydroxyl groups. Moreover, nitrogen-containing complex compounds used as adhesion additives enhance the chemical bonding reactions with selected mineral surfaces due to the presence of polar nitrogen functionalities.

At the interface between the bitumen and crushed granite aggregates, acid-base complex reactions occur, leading to the formation of water-insoluble compounds. For instance, calcium carbonate present in marble reacts with the carboxylic acids in the bitumen to form water-insoluble complex salts.

Furthermore, hydroxyl groups on the surface of granite aggregates react with the complex organic acid functional groups in bitumen, forming chemical bonds. When selected adhesion additives are introduced into the bitumen, interactions occur between the hydroxyl groups of the granite surface and the complex compounds in the binder.

It is well known that wetting of the mineral surface by bitumen is one of the key factors for achieving effective adhesion. The degree of wetting is characterized by the contact angle, defined as the inclination of the boundary angle formed when a drop of bitumen contacts the solid surface (Table 2, Figures 1 and 2).

Table 2.
Effect of adhesion additives on the adhesion stability between standard BND 60/90 bitumen grade and alternative mineral materials

Main component name and amount, %	MDEA additive amount, %	Adhesion according to "passive" method, %	Contact angle of bitumen wetting on glass substrate (°)
Standard BND 60/90 bitumen		76	113
GR (95)	5	82	93
GR (90)	10	94	84
GR (85)	15	97	61
OS (95)	5	83	92
OS (90)	10	90	88
OS (85)	15	94	72
OS (80) + S (15)	5	79	106
OS (80) + S (10)	10	82	101
OS (80) + S (5)	15	86	88
GR (80) + S (15)	5	96	73
GR (80) + S (10)	10	92	83
GR (80) + S (5)	15	94	78

Explanation for Table 2: GR-Gossypol resin; OS-Oil sludge; S-Sulfur additive; WCA – Wetting Contact Angle

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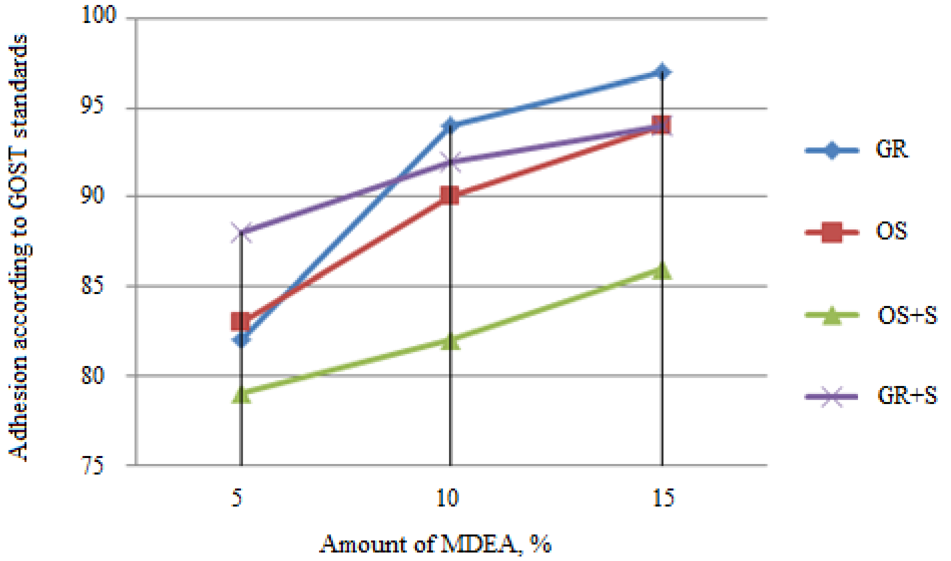

Figure 1. Dependence of the adhesion ability of BND 60/90 bitumen on the amount of additives

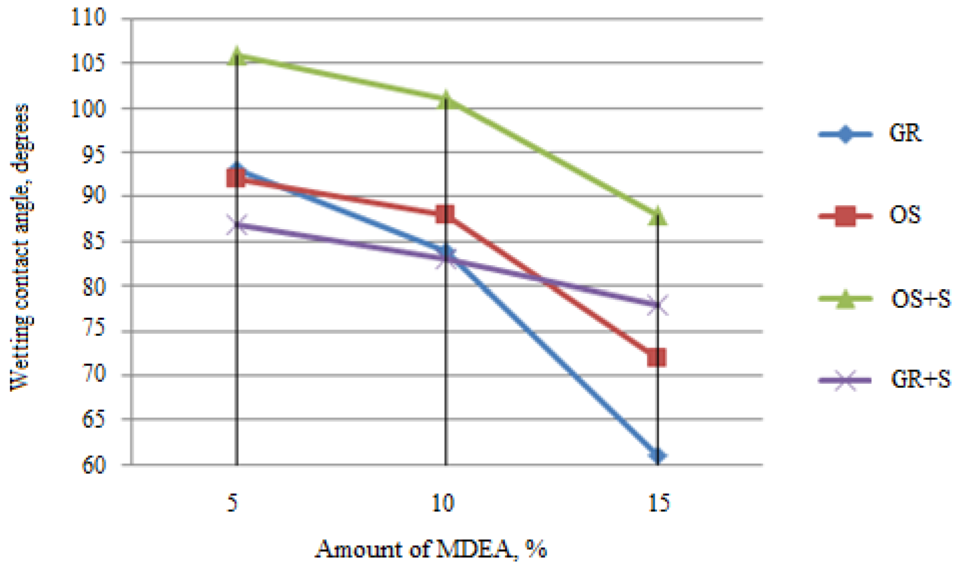

Figure 2. Dependence of the wetting ability of BND 60/90 bitumen on the amount of additives

The wetting contact angle was measured at the "Petroleum Chemistry" laboratory using the OSA-15 optical device. The contact angle of bitumen wetting on glass substrates was determined by utilizing a projection generation unit. After placing a 2.5 mm diameter drop of bitumen on the glass surface at a controlled set temperature, measurements were taken 2 minutes later, allowing the drop to reach equilibrium to ensure the reliability of the wetting contact angle assessment.

To achieve ideal experimental conditions, special attention was paid to simulating the most severe working environments: the complex and variable composition of the glass, the ideal smoothness and density of the substrate were carefully considered. It was established that when glass is used as the substrate in the instrument, the lowest or critical adhesion values are obtained, providing a conservative basis for analysis.

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Vol. 12, Issue 4, April 2025

In practice, based on the experimental results, it was confirmed that under industrial conditions, the adhesion between bitumen binders and actual mineral aggregates is significantly higher than the adhesion measured on glass substrates.

V. CONCLUSION AND FUTURE WORK

Based on the research results, it was observed that particles of gudron (gossypol resin) form hydrogen bonds with metal hydroxides present on the surface of granite gravel and crushed granite aggregates, which contributes to the improvement of asphalt concrete properties in the future. In this process, the hydrocarbon radicals formed on the mineral surface are expected to chemically bond with the organic compounds present in the bitumen binder.

A significant factor enhancing the adhesion process between the viscous bitumen binder and the mineral material surface is the incorporation of various additives into the bitumen, especially substances containing functional groups similar to those in the bitumen structure. Therefore, the adhesion mechanism is strongly influenced by interactions occurring at the molecular scale.

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