Modification of Petroleum Bitumen with Secondary Polyethylene in the Presence of Vermiculite

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Received: November 20, 2023
Accepted: February 1, 2024
DOI: 10.22146/ijc.90881

Abstract: This article presents studies on the modification of petroleum bitumen with polymer waste in the presence of vermiculite. An increase in temperature leads to an increase in the interaction of components, partial breakage of polyethylene and bitumen macromolecules, and the formation of radical-free valences. As a result, fragments of polyethylene and bitumen molecules react with each other, and the formation of qualitatively new structural formations occurs via the strong connection of polar and amorphous bitumen with nonpolar structurally viscous polyethylene. Domestically produced Kulantau vermiculite was used to ensure stable adhesion over a wide temperature range and increase the specific surface area, which acquired additional energy, resulting in an increase in the degree of adhesion to bitumen and increased durability of the binder with improved rheological characteristics. Because of the increased service life of road asphalt pavements, the use of the latter results in a considerable decrease in the cost of polymer-bitumen binders. Based on improved binder formulations, asphalt concrete exhibits excellent water resistance and strength at 50 °C (4.7 to 5.0 MPa). In summary, the modification of bitumen with polymer waste and vermiculite offers a promising avenue for improving the performance and longevity of asphalt pavements.

Keywords: adhesion; asphaltenes; elasticity; polymer-bitumen binders; road industry

INTRODUCTION

Higher traffic volume and temperature have an impact on road performance. Several strategies are being used worldwide to improve the quality of asphalt binder. One of the classic methods is the addition of polymers to bitumen to change its characteristics [1-2]. It is well known that the most important condition for obtaining a high-quality asphalt concrete mixture is good adhesion of bitumen to mineral materials, which is ensured, first, by good enveloping and chemical interaction of the
active components of bitumen with the mineral filler [1-3]. The quality of bitumen, which is a key factor in the production of various types of asphalt concrete mixtures, ultimately determines the quality of the asphalt road surface and its service life. The obvious disadvantages of bitumen were thermal sensitivity, low elasticity, cracking at low temperature, and a tendency to dry out, which eliminated manufacturing through oxidation. Improving the quality of bitumen and asphalt concrete is a top priority in all national-wide road industries [4]. Because the quality of asphalt concrete as a thermoplastic material is dictated by the properties of bitumen, the quality of bitumen greatly influences the quality and service life of road asphalt concrete pavements [5]. Polymer-modified bitumen is more resistant to thermal cracking and rutting. Polymers such as polyethylene, ethyl vinyl acetate, polypropylene, and styrene-butadiene-styrene have been shown to improve the engineering qualities of bitumen. These polymers are reported to improve bitumen qualities such as fatigue strength, thermal cracking resistance, and rutting. Polymers can be incorporated into bitumen via chemical reactions or mechanical mixing [4-5].

In recent years, for various reasons, the quality of road bitumen has noticeably deteriorated. However, various modifying additives for bitumen and polymer-bitumen binders have been developed and actively used to improve the characteristics of asphalt concrete pavements. Bitumen modification is the process of introducing third-party substances, which changes the final physical and mechanical properties of bitumen [6-7]. The most commonly used bitumen modifiers, improving the engineering qualities of bitumen, include high-molecular weight compounds of the following types: block copolymers of divinyl and styrene, thermoplastics (polyethylene, polystyrene, epoxy resin, and ethylene vinyl acetate), elastomers (synthetic rubbers), thermosets (carbamide, polyester, and other synthetic polymers) [8-10]. Polymer-bitumen binders are new road-building materials that are superior in many characteristics to conventional petroleum-based road bitumen and perform a binding function in the production of asphalt mixtures for paving, reconstructing, and constructing roads [11].

Cost-effective modifiers are available and inexpensive. With a shortage of organic raw materials, polymer waste has become a rich material, a key energy resource, and an important economic factor. The largest part of the polymer waste used (secondary polymer raw materials) consists of polyethylene, polypropylene, polyethylene terephthalate, polyvinyl chloride, polystyrene, and rubber crumbs [12-14]. All these products have partially lost their properties and are out of use but have retained their technological properties, which ensures the possibility of their reuse in the form of raw materials and that must be reprocessed. Secondary polymer raw materials can save primary raw materials such as petroleum, while also addressing environmental concerns by preventing the burial or burning of plastic waste. This practice requires additional capital investments and causes pollution [13].

The disposal of polymer materials is a significant environmental concern due to their resistance to physical, chemical, and biological destruction and the gradual release of toxic compounds. Recycling waste polymer materials is critical for conserving natural resources, minimizing greenhouse gas emissions, and reducing energy use [12,14].

The development of binders based on petroleum bitumen, modified with polymer additives and other compounds, activated and stabilized components, is a promising path for improving the rheological properties of bitumen binders. This approach ensures an increase in the strength and deformation characteristics of asphalt concrete and prolongs the service life of road surfaces [15-16]. The introduction of even a small amount of polymer components into bitumen helps to improve its thermoplastic properties, wear resistance at low temperatures and increase the service life of the coating. Distributed in the bitumen environment, the polymer components form a spatial network, providing elastic properties that classical bitumen does not possess [6].

In connection with the above, research aimed at reducing the cost of polymer-bitumen binders (PBB) using less expensive polymer modifiers is of interest. The main factor in the development of binders is the
compatibility of the polymer components with each other and with the components of bitumen. The use of secondary polymers as a bitumen modifier will make it possible to obtain a polymer-bitumen binder, which, compared to conventional bitumen, will have a wider operating temperature range, and elastic properties [17]. Because bitumen binder has insufficient tensile strength, the practice of operating road pavements using petroleum road bitumen (PRB) suggests that coating degradation starts in the first year of operation. The primary causes of asphalt concrete deterioration on road surfaces are traffic patterns and weather, which age the bitumen and cause it to peel and crack. Several works have shown that the quality and performance characteristics of materials based on petroleum bitumen can be improved by modification with polymer synthetic materials [17-18]. Compared with conventional road bitumen, PBB increases the service life of the pavement by 2–3 times, increasing the resistance of the road surface to deformation under high loads, reducing the likelihood of rutting, and allowing road construction work to be carried out at a temperature of −10 °C. Polymer bitumen is characterized by a wide range of operating temperatures, the difference between the softening point and the temperature. Therefore, the study of bitumen with polymer additives has been of constant interest to researchers in recent decades [19].

Bitumen comprises a complex combination of high-molecular weight hydrocarbons derived from petroleum, such as the naphthenic \((C_nH_{2n})\), aromatic \((C_nH_{2n+6})\), and paraffin \((C_nH_{2n+2})\), together with its derivatives that contain complex metal compounds, oxygen, sulfur, and nitrogen. The main chemical composition of bitumen is carbon (70–87%), hydrogen (8–12%), sulfur (0.5–7%), oxygen (0.2–12%), and nitrogen (0–2%) [1-2]. The distribution of the molecular weights of the bitumen components obeys the law of normal distribution, which is also reflected in the nature of the changes in the rheological properties of bitumen in the plasticity range.

The structure of bitumen should be considered a solution of high-molecular weight compounds of petroleum origin (asphaltenes and solid resins similar in structure) in a medium of petroleum oils (and fusible resins close to them). The three main components are as follows: lyophobic particles (asphaltenes), lyophilic substances (resins) surrounding the lyophobic particles and preventing their fusion, and the oil phase in which they are dispersed (Fig. 1) [6].

Thus, according to modern concepts [1-2], all roads and construction bitumen should be considered as solutions of high-molecular weight compounds of petroleum origin – asphaltenes and resins (solid resins) similar in structure and properties in a medium of petroleum oils and resins similar in structure (fusible resins). The high molecular weight compound (HMC) solution theory states that bitumen can exist in various thermodynamic states contingent on external factors (temperature, pressure), going through all phases sequentially from true solutions (at high technological temperatures) to colloidal solutions of supramolecular structures associated with asphaltenes and resins to plastic and finally solids. At temperatures below the softening temperature, the development of processes of association of structural units, the formation of transition-type structures, and the emergence of structures with a high degree of supramolecular formations (associates) occur. In this case, the structure of a plastic amorphous body is formed. At temperatures below the brittle point, bitumen is a solid, where, along with amorphous substances in a glassy metastable form, crystalline substances may be present, as well as a certain number of substances (oils and low-melting resins) that have not

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**Fig 1.** The main components of the bitumen structure
yet lost their plasticity. These processes of changing the structural states of bitumen are thermodynamically reversible, and for each bitumen, they occur within certain temperature limits associated with the plasticity range of the material. During the transition from a liquid (viscous) state to a plastic state and then to a solid state, due to the development of structural formation processes and the growth of supramolecular structures (associates), an increase in deviation from the Newtonian type of flow, characterized by a flow index, is observed. Moreover, the proportion of elastic deformation increases. Asphaltenes are one of the main components of oil and determine many of its physical and chemical parameters. The asphaltene molecules and the heteroatoms in the asphaltene dimer are shown in Fig. 2.

Bitumen can be modified with polymers to create a better-performing binder [18]. This can increase heat and frost resistance, chemical resistance, ductility, and elasticity of composite coatings [17]. Russian scientists have explored using polymers in asphalt concrete mixtures, including recycled polyethylene. Ragni et al. [19] conducted experimental studies on the physical and mechanical properties of low-density and high-density polyethylene. Recycled low-density polyethylene (LDPE) and Kulantau vermiculite were purchased from AVENUE LLP, Kazakhstan. LDPE is crushed to a crushed/fluff size using special equipment [20-23]. The physical and mechanical properties of LPDE are given in Table 2.

Vermiculite is a clay mineral with a crystalline structure. In nature, vermiculite is formed as a result of the hydration of magnesium-iron micas-biotite and phlogopite. In Kazakhstan, the most promising vermiculite

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**EXPERIMENTAL SECTION**

**Materials**

Bitumen BND 70/100, industrial oil I-20Ai, and Kulantau vermiculite were obtained from the Qazaq bitum LLP, Kazakhstan with a chemical formula of $\text{C}_{17}\text{H}_{19}\text{NO}_3$ (7,8-didehydro-4,5-epoxy-17-methylmorphinan-3,6-diol). Table 1 presents the physical and mechanical properties of bitumen. Recycled low-density polyethylene (LDPE) and Kulantau vermiculite were purchased from AVENUE LLP, Kazakhstan. LDPE is crushed to a crushed/fluff size using special equipment [20-23]. The physical and mechanical properties of LPDE are given in Table 2.

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**Table 1. Physical and mechanical properties of the BND 70/100 bitumen**

<table>
<thead>
<tr>
<th>Indicator</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Needle penetration depth, 0.1 mm:</td>
<td></td>
</tr>
<tr>
<td>At 25 °C</td>
<td>75</td>
</tr>
<tr>
<td>At 0 °C</td>
<td>22</td>
</tr>
<tr>
<td>Softening temperature along the ring and ball, °C</td>
<td>48</td>
</tr>
<tr>
<td>Tensile strength at 25 °C, cm</td>
<td>115</td>
</tr>
<tr>
<td>Brittleness temperature, °C</td>
<td>−20</td>
</tr>
<tr>
<td>Flash point, °C</td>
<td>240</td>
</tr>
</tbody>
</table>

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**Fig 2. Asphaltenes and heteroatoms in the asphaltene dimer**
deposits are Kulantau, Iirsu and Zhylandy [24-25]. The general molecular formula of trioctahedral vermiculite is 
\((\text{Mg, Ca})_3(\text{OH})_2(\text{Si}_{8-x}\text{Al}_x)(\text{Mg, Fe})_6\text{O}_{20}y\text{H}_2\text{O}\), and the characterization of vermiculite is shown in Table 3.

**Instrumentation**

The mechanical properties of the modified bitumen with polymer waste were determined following American Society for Testing and Material (ASTM), which is ASTM D882-91 [26]. The tensile strength, elasticity, and modulus of elasticity of the films were measured using an MTS 10/M tensile testing machine with a crosshead speed of 50 mm/min. The tensile characteristics of the mixed films were evaluated. A minimum of four readings were averaged, and a 1-kN load cell was used. All the measurements were conducted using a universal testing machine (Shimadzu AGS-10kNG). The viscosity was measured by ISO 12058-1 at 25 °C [27]. According to ASTM, C170-90, the compressive strength of the samples was measured using a Type LPM 600 M1 SEIDNER hydraulic testing machine (Germany) [28]. The bending tests were performed on an MTS Criterion C4 (MTS Systems GmbH, Germany). The determination of water absorption was performed as per the specifications of BS 1881: Part 122 [29].

**Procedure**

In a laboratory setting, PBB was prepared in a cylindrical metal mixing container with a minimum capacity of 1 L. The bitumen was heated in the mixer using an electric stove. The temperature in the mixer was controlled with a thermometer [22]. The PBB components were mixed using a mechanical paddle mixer, the speed of which was regulated using a laboratory transformer. The required amount of bitumen was loaded into a metal container. At 150–160 °C, recycled polyethylene was introduced into the molten bitumen in portions. First, the mixture was stirred until complete wetting and uniform distribution of the polymer were achieved. Then, intensive stirring of the mixture was started with simultaneous gradual heating to 190–200 °C and Kulantau vermiculite was introduced. Stirring was carried out for 30–40 min. After carrying out the modification process and determining the homogeneity of the mixture, the resulting PBB samples were tested in several studies to determine their physical and mechanical properties (determining the depth of penetration of the needle at 0 and 25 °C, elongation at 25 °C, softening temperature along the ring and ball, elasticity, and adhesion to sand) [23].

Polymer-bitumen binders were manufactured using a laboratory setup that allowed for the mixing of liquids with volumes ranging from 1.0–1.5 L and stirrer shaft speeds ranging from 40–350 rpm. A 3.5–4.0% polymer waste (LDPE), was placed into molten bitumen grade BND 70/100 (76.5–82% of the total volume) at a temperature of 150–160 °C, by a total mass of industrial oil I-20Ai and Kulantau vermiculite of 4.0–5.0%. The mixture was heated at 5 °C/min.

### Table 2. Physical and mechanical properties of primary and secondary LDPE fibers

<table>
<thead>
<tr>
<th>Indicator</th>
<th>Primary polyethylene</th>
<th>Secondary polyethylene</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Low-density</td>
<td>High-density</td>
</tr>
<tr>
<td>Tensile strength</td>
<td>15</td>
<td>22–44</td>
</tr>
<tr>
<td>Elongation at break</td>
<td>580–750</td>
<td>400–550</td>
</tr>
</tbody>
</table>

### Table 3. Qualitative characteristics of vermiculite from the Kulantau deposit

<table>
<thead>
<tr>
<th>Sample No.</th>
<th>Humidity (%)</th>
<th>Degree of hydration (%)</th>
<th>Volumetric weight (kg/m³)</th>
<th>Vermiculite content (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>6.2</td>
<td>65</td>
<td>178</td>
<td>30</td>
</tr>
<tr>
<td>2</td>
<td>9.0</td>
<td>70</td>
<td>200</td>
<td>35</td>
</tr>
<tr>
<td>3</td>
<td>3.7</td>
<td>60</td>
<td>130</td>
<td>28</td>
</tr>
<tr>
<td>4</td>
<td>3.1</td>
<td>75</td>
<td>140</td>
<td>26</td>
</tr>
</tbody>
</table>
RESULTS AND DISCUSSION

At different concentrations of the secondary polymer (1, 2, 3, 4, and 5% by weight of bitumen), the alteration in the characteristics of the polymer-bitumen binder was examined (Fig. 3-5). High-temperature homogenization is characterized by the partial breakage of polyethylene and bitumen macromolecules and the formation of radical-free valences, as a result of which fragments of polyethylene and bitumen molecules react with each other. An intermolecular adhesive force between structural components in a dispersion media is produced during bitumen modification. High temperatures and continuous stirring are required for the combination process to occur, after which a homogenous system is obtained. The structural stability of PBB is a crucial factor that inhibits further bitumen and polymer separation [6,30].

The chemical interaction of bitumen components with the polymer matrix occurs. A new structure is formed with a strong connection of polar and amorphous bitumen with nonpolar structurally viscous polyethylene. Microscopic studies have established that

Fig 3. Dependence of the melting temperature of PBB on the content of LDPE polymer

Fig 4. Dependence of the tensile strength of PBB on the content of LDPE polymer
in the structure of a binder obtained at 280 °C, two continuous phases (polyethylene and bitumen) are formed and intertwined with one another – a “mesh within a mesh” structure. At 220–240 °C, no such microstructure was observed. Analysis of the data obtained shows that even with a LDPE content in bitumen of 2–3%, a spatial structural network is formed. The formation of a spatial structural network can be traced by characteristic inflections (changes in slope) of the characteristic dependences of the properties of the polymer-bitumen organic binder. The specificity of the new structure that formed manifested as increased plasticity at low temperatures (Fig. 3), decreased tensile strength at 25 °C (Fig. 4), and increased elasticity (Fig. 5).

Table 1 outlines the physical and mechanical properties of the BND 70/100 bitumen, presenting key indicators and their respective values. At 25 °C, the needle penetration depth is recorded as 75 mm, indicating the consistency or hardness of the bitumen at this temperature. A higher penetration depth suggests softer bitumen. At 0 °C, the needle penetration depth is 22 mm, reflecting the change in consistency as the temperature decreases. A lower penetration depth at a lower temperature suggests increased hardness or stiffness. The softening temperature, along the ring and ball apparatus, is 48 °C. This parameter is crucial as it indicates the temperature at which the bitumen softens and begins to deform under a specified load. A higher softening temperature implies better high-temperature performance.

The tensile strength of the bitumen at 25 °C is recorded as 115 cm. Tensile strength measures the material’s ability to withstand a stretching force. A higher tensile strength suggests better resistance to deformation under tension. The brittleness temperature, recorded at −20 °C, indicates the temperature at which the bitumen becomes brittle and susceptible to cracking. A lower brittleness temperature is generally desirable for improved low-temperature performance. The flash point of the bitumen is measured at 240 °C. This is the temperature at which the bitumen emits vapors that can ignite in the presence of an open flame. A higher flash point is generally considered safer. In summary, the presented physical and mechanical properties provide a comprehensive overview of the BND 70/100 bitumen. The combination of these properties helps in assessing the bitumen’s suitability for various applications, including its performance at different temperatures, resistance to deformation, and safety considerations related to its flammability.

Table 2 presents the physical and mechanical properties of primary and secondary LDPE fibers, along with their high-density counterparts. The tensile strength of primary LDPE fiber is noted as 15 MPa.
Tensile strength represents the maximum stress a material can withstand while being stretched or pulled before necking, which is the point of irreversible deformation. The tensile strength of secondary LDPE fiber ranges between 9–13 MPa. The variation suggests different strengths within this range, which may be attributed to variations in the manufacturing process or material quality. For primary HDPE fibers, the tensile strength ranges from 22–44 MPa, indicating a significantly higher strength compared to both primary and secondary polyethylene fibers. The elongation at break for primary low-density fiber is in the range of 580–750%. Elongation at break measures the ability of the material to deform before rupture. A higher percentage suggests greater flexibility and stretchability. The elongation at break for secondary high-density fiber ranges from 110–240%. This range indicates a different level of stretchability compared to primary low-density fibers, with potentially different applications. The elongation at break for secondary low-density is noted as 300–280%. Despite being low-density, the elongation at break is still substantial, suggesting a balance between strength and flexibility. The tensile strength and elongation at break properties are crucial for understanding the performance of polyethylene fibers in various applications. Secondary polyethylene fiber exhibits a lower tensile strength range compared to primary polyethylene fiber, suggesting potential differences in the manufacturing process or recycled material characteristics. In summary, Table 2 provides valuable insights into the mechanical properties of different types of polyethylene fibers, aiding in a material selection based on the desired balance of strength and flexibility for specific applications.

Table 3 outlines the qualitative characteristics of vermiculite from the Kulantau deposit, providing information on various parameters for different samples. The humidity content in the vermiculite samples ranges from 3.1 to 9.0%. Humidity is a crucial parameter as it indicates the amount of water present in the vermiculite. Lower humidity levels suggest drier vermiculite, which can be desirable for certain applications. The degree of hydration varies from 60 to 75%. This parameter reflects the extent to which vermiculite has absorbed water. Higher values indicate a greater degree of hydration, which can impact the material's properties, such as its expansion characteristics. The volumetric weight of the vermiculite samples ranges from 130 to 200 kg/m³. Volumetric weight is a measure of the mass per unit volume, and it provides insights into the density of the vermiculite. Higher volumetric weights may indicate denser and potentially more compacted vermiculite. The vermiculite content in the samples ranges from 26 to 35%. This parameter represents the proportion of vermiculite within the overall composition. Higher vermiculite content is generally desirable for applications where the unique properties of vermiculite, such as its ability to expand when heated, are important. The variations in humidity levels suggest differences in the moisture content of the vermiculite samples. Lower humidity levels (e.g., sample 4) may be advantageous for applications where dry vermiculite is preferred. The degree of hydration indicates the extent to which vermiculite has absorbed water. This parameter is important for understanding how hydrated vermiculite is, as it can influence its physical and thermal properties. Volumetric weight is an essential parameter for assessing the density of vermiculite. Higher volumetric weights may suggest denser vermiculite, potentially impacting its handling and application characteristics. The vermiculite content is crucial, as it directly influences the performance of vermiculite in various applications. Higher vermiculite content is generally associated with improved insulating and lightweight properties. In summary, Table 3 provides valuable information about the qualitative characteristics of vermiculite from the Kulantau deposit, offering insights into its moisture content, degree of hydration, density, and vermiculite content. These parameters are critical for selecting and optimizing the use of vermiculite in specific applications, such as construction, horticulture, and insulation.

Based on the data obtained, it was established that as the content of recycled LDPE in the mixture increased, the physical and mechanical properties of the modified bitumen changed. The resulting polymer-
bitumen binders exhibited changes in physical and mechanical properties, such as penetration at 0 and 25 °C, tensile strength at 25 °C, and elastic and softening temperatures compared to those of the original road bitumen. When determining the adhesion characteristics of PBB, all the samples were maintained according to a control sample number 2 for adhesion to sand.

The findings of investigations into the physicochemical characteristics of modified polymer-bitumen binders (MPBB) and road bitumen are shown in Table 4. Additionally, Table 4 illustrates how the use of a polymer composition results in enhanced resistance to temperature-dependent loads at high temperatures, which is explained by the presence of a polymer network; it also increases resistance to temperature-dependent cracking, aging resistance, hardness, and elasticity.

It is known that thermoplastics modify bitumen, creating a rigid spatial network that resists deformation. A continuous spatial network of thermoplastics is formed when the content of the thermoplastic in the binder is about 5–6%. Thermoplastics increase cohesion, heat resistance, elasticity, and adhesive properties, and reduce the fragility of the binder at low temperatures. Most thermoplastics dissolve well in bitumen at temperatures of 150–170 °C [6]. These polymers have high strength but are not sufficiently elastic or crack-resistant at low negative temperatures; some polymers tend to increase in strength with age.

One way to improve the performance characteristics of an asphalt road surface is by adding various adhesive additives to bitumen or PBB binders. Locally produced Kulantau vermiculite was utilized to guarantee stable adhesion over a broad temperature range and increase the specific surface area, which results in increased energy consumption, increasing the degree of adhesion to bitumen and increasing the durability of the binder, which has improved rheological characteristics. The latter results in considerable cost savings because it prolongs the life of the asphalt road surface.

The choice of vermiculite is explained by its low price compared to other clays. In addition, vermiculite is characterized by a high adsorption capacity and effective absorption of products resulting from the oxidation of hydrocarbons, for example, resins, oxygen-containing compounds, and other hetero-organic products. Porous vermiculite passes hydrocarbons with straight chains through its pores and retains cyclic hydrocarbons and hydrocarbons with a branched molecular structure, thereby depleting the bitumen film with oil components and enriching it with asphaltenes and resins, which is accompanied by hardening of the bitumen film.

Adhesive additives improve the adhesion of road bitumen and PBB to mineral filler, slow the aging process of bitumen at high temperatures, increase the water corrosion resistance, frost, and heat resistance of the road surface, and allow the extension of the road construction season. As a result of a series of industrial tests of MPBB using polymer wastes and Kulantau vermiculite to create road surfaces of the required quality.

Table 4 provides a detailed comparison of the physico-mechanical properties of BND 70/100 bitumen and MPBB obtained using polymer wastes and vermiculite. At 50 °C, the ultimate compressive strength for MPBB is significantly higher (5.0 MPa) compared to BND 70/100 bitumen (1.6 MPa), while at 25 and 0 °C, MPBB also shows improved compressive strength compared to BND 70/100 bitumen. At both 25 and 0 °C, MPBB demonstrates comparable tensile strength to BND 70/100 bitumen. However, at 25 °C, BND 70/100 bitumen has a slightly higher tensile strength. The penetration value for MPBB is 100, suggesting that it maintains similar penetration characteristics compared to BND 70/100 bitumen. MPBB shows lower water saturation (4.0%) than BND 70/100 bitumen (5.0%), indicating improved water resistance. The water resistance coefficient for both materials is 1.00, indicating that they have similar resistance to water. Additionally, MPBB has a higher water resistance coefficient (1.00) for long-term water saturation compared to BND 70/100 bitumen (0.89), suggesting better long-term water resistance. MPBB demonstrates higher ultimate bending strength at both 25 and −25 °C compared to BND 70/100 bitumen. Moreover, MPBB exhibits higher viscosity values at both 25 and 0 °C compared to BND 70/100 bitumen, indicating potential changes in flow behavior.
Table 4. Physicomechanical properties of BND 70/100 bitumen and polymer-bitumen binders (MPBB) obtained using polymer wastes and vermiculite

<table>
<thead>
<tr>
<th>Indicators</th>
<th>BND 70/100</th>
<th>MPBB</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ultimate compressive strength, MPa, At a temperature:</td>
<td></td>
<td></td>
</tr>
<tr>
<td>50 °C</td>
<td>1.6</td>
<td>5.0</td>
</tr>
<tr>
<td>25 °C</td>
<td>2.0</td>
<td>2.5</td>
</tr>
<tr>
<td>0 °C</td>
<td>10.8</td>
<td>12.8</td>
</tr>
<tr>
<td>Tensile strength, cm, not less:</td>
<td></td>
<td></td>
</tr>
<tr>
<td>25 °C</td>
<td>30</td>
<td>25</td>
</tr>
<tr>
<td>0 °C</td>
<td>25</td>
<td>19</td>
</tr>
<tr>
<td>Penetration</td>
<td>100</td>
<td>-</td>
</tr>
<tr>
<td>Water saturation, % by volume</td>
<td>5.0</td>
<td>4.0</td>
</tr>
<tr>
<td>Water resistance coefficient</td>
<td>1</td>
<td>1</td>
</tr>
<tr>
<td>Water resistance coefficient for long-term water saturation</td>
<td>0.89</td>
<td>1.00</td>
</tr>
<tr>
<td>Ultimate bending strength, MPa, at a temperature:</td>
<td></td>
<td></td>
</tr>
<tr>
<td>25 °C</td>
<td>2.0</td>
<td>3.5</td>
</tr>
<tr>
<td>~25 °C</td>
<td>4.0</td>
<td>5.8</td>
</tr>
<tr>
<td>Viscosity, MPa, at a temperature:</td>
<td></td>
<td></td>
</tr>
<tr>
<td>25 °C</td>
<td>1.41×10^{10}</td>
<td>2.69×10^{10}</td>
</tr>
<tr>
<td>0 °C</td>
<td>1.03×10^{11}</td>
<td>3.89×10^{10}</td>
</tr>
<tr>
<td>Plasticity at a temperature:</td>
<td></td>
<td></td>
</tr>
<tr>
<td>25 °C</td>
<td>0.381</td>
<td>0.356</td>
</tr>
<tr>
<td>0 °C</td>
<td>0.333</td>
<td>0.321</td>
</tr>
<tr>
<td>Modulus of elasticity, MPa, at a temperature:</td>
<td></td>
<td></td>
</tr>
<tr>
<td>25 °C</td>
<td>2305</td>
<td>2500</td>
</tr>
<tr>
<td>0 °C</td>
<td>2935</td>
<td>3605</td>
</tr>
</tbody>
</table>

Plasticity values show a slight decrease in MPBB at both temperatures compared to BND 70/100 bitumen. MPBB generally shows comparable or slightly higher modulus of elasticity values at both temperatures compared to BND 70/100 bitumen. The use of polymer wastes and vermiculite in MPBB results in improved mechanical properties, such as compressive strength, tensile strength, and ultimate bending strength, compared to conventional BND 70/100 bitumen. MPBB demonstrates enhanced water resistance, as evidenced by lower water saturation and higher water resistance coefficients, especially in long-term water saturation scenarios. The increased viscosity of MPBB may influence its application, potentially offering advantages in specific construction scenarios. Modulus of elasticity values suggest that MPBB maintains or exceeds the elastic properties of conventional bitumen. In summary, the data in Table 4 indicates that the incorporation of polymer wastes and vermiculite into bitumen leads to improvements in various physico-mechanical properties, making MPBB a potentially advantageous material for certain applications in construction and road engineering.

Asphalt concrete based on new binder compositions has high strength at 50 °C (from 4.7 to 5.0 MPa) and water resistance. As shown in Table 4, MPBB have increased heat and crack resistance. A decrease in the penetration and tensile strength of MPBB with the introduction of a secondary polymer indicates the formation of a spatial polymer network. The resulting PBB has a structure different from bitumen [31].

**CONCLUSION**

An increase in temperature leads to the activation of interactions between components and the formation of qualitatively new structural formations. High-temperature homogenization is characterized by the
partial breakage of polyethylene and bitumen macromolecules and the formation of radical-free valences, as a result of which fragments of polyethylene and bitumen molecules react with each other. Utilizing domestically produced Kulantau vermiculite and recycled polyethylene, a comparative analysis of the mechanical and physical characteristics revealed that the modified polymer-bitumen binder offers stable adhesion over a wide temperature range, an increase in the specific surface area that provides additional energy capacity, and a greater degree of adhesion to bitumen. The selective diffusion of oils and resins into materials causes a change in the group composition of bitumen because of the interaction of the bitumen with a material that has a high specific surface area and a fine-porous structure. This alteration of the properties of the bitumen layers on the particle surfaces results in the formation of long-lasting bitumen films on the grain surfaces. Selectivity, a large pore capacity, and high activation phase efficiency are characteristics of vermiculite. The availability and relative affordability of enlarged Kulantau vermiculite produced domestically also influence its choice. These findings verify that the modified polymer-bitumen binder satisfies the Kazakhstan criteria for polymer-bitumen binder and is appropriate for modified bitumen production based on its physical and chemical properties.

■ ACKNOWLEDGMENTS

These studies were carried out with the support of the Science Committee of the Ministry of Science and Higher Education of the Republic of Kazakhstan (grant AP19679034 “Development of technology for producing bitumen materials for the road industry with the disposal of polymer wastes in the technological process”).

■ CONFLICT OF INTEREST

The authors declare no conflicts of interest.

■ AUTHOR CONTRIBUTIONS

Syrmanova Kulash wrote the draft of the manuscript, Kaldybekova Zhanat, Agabekova Aktolkyn, Baizhanova Sholpan, and Tuleuov Rassul conducted the experiment and calculations, Khaladun Al Azzam, El-Sayed Negim, and Bounoua Nadia participated in designing, enhancing the discussion, and revising the manuscript. All authors agreed to the final version of this manuscript.

■ REFERENCES


