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Research Article

Bitumen-Biopolymer Materials Modified with Polylactic Acid with Improved Physical and Chemical Properties

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With the continuous expansion of the global automobile fleet, there is an escalating demand to enhance and maintain current road infrastructure. Given the information provided, there will be a growing demand for bitumen, a key raw material used in the manufacturing of asphalt. Bitumen may account for up to 60% of the total usage in asphalt production. This study aims to determine the effect of different content of polylactic acid (PLA) on the change in the chemical and physical properties of biopolymer bitumen during its modification. This study was carried out by using a sample of petroleum road bitumen from CASPI BITUM (Kazakhstan) and a sample of PLA from Zhejiang Hisun (China). As a part of the research, the change of quality indicators of biopolymer bitumen when adding 4%–10% of PLA to it has been established. The results showed that the values of the average molecular weight and average molar mass increased with increasing the content of PLA in biopolymer bitumen. In particular, when the PLA content in biopolymer bitumen increased up to 10%, the average molecular weight of the biopolymer bitumen increased from 1,263 to 2,759 Mw and the average molar mass increased from 1,215 to 1,395 Mn. It was shown that increasing the PLA content in biopolymer bitumen from 0% to 10% leads to an increase in the softening temperature from 47 to 70°C or ~49%. It was found that all examined samples of biopolymer bitumen are characterized by increased plasticity at 25°C (>100 cm). It has been established that the addition of 8% PLA to bitumen allows one to obtain a biopolymer bitumen of optimal quality. The results obtained can be used to produce road biopolymer bitumen.

1. Introduction

Due to the ever-increasing car fleet in the world, there is a growing need to increase and repair existing roads. According to Statista [1], the market for asphalt in the world is expected to grow by almost 28% from 2018 to 2027 to reach $2.48 billion.

Considering the above, the need for one of the main raw materials for asphalt production (bitumen) will increase, consumption of which in asphalt production can be up to 60%. Bitumen is a product of crude oil processing and contains several hydrocarbons characterized by high molecular weight [2]. Due to its rheological and adhesive characteristics, as well as its impermeability, it has found wide application in road construction [3, 4]. During its service life, bitumen is subjected to thermal, mechanical, and weather (rain, snow, hail, etc.) loads, which leads to a change in its quality, namely cracking and deformation [5, 6, 7]. Researchers should, in accordance with this, develop such formulations of road bitumen in order to maximally resist the negative factors affecting it [8, 9]. The work [10] shows that biodegradable materials are widely used in various industries, and humic acids and polylactic acid (PLA) can serve as raw materials for biodegradable materials. It is widely known that to increase the performance properties, bitumens are modified with various additives, in particular, nanomaterials [11, 12], geopolymers [5, 13, 14, 15], waste glass and plastic [16], rubber [17], and phenol–cresol–formaldehyde resins [18]. The results of previous

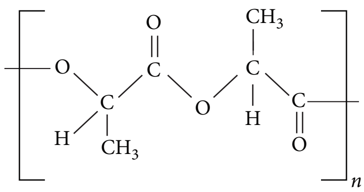

Figure 1: Polylactic acid.

studies showed that the addition of a modifier improved the characteristics of bitumen in terms of elasticity and stiffness and reduced the residual deformation of the road surface as a result of environmental effects and repetitive traffic loads [19, 20, 21].

The works [22, 23] demonstrated convincing evidence of the increase of physical and mechanical characteristics of bitumen by adding different types of polymers. According to studies, the modification of bitumen with the addition of SBP (styrene–butadiene polymers) improves the technical properties of bitumen, reducing temperature dependence, and increasing elasticity. There is increased stiffness at high temperatures, resistance to cracking at low temperatures, moisture resistance, and increased fatigue life [17, 24].

There is now a rising worldwide interest, especially in Kazakhstan, in using nanomaterials for the construction of road surfaces. This is due to the discovery that asphalt concretes, when augmented with nanomaterials, may demonstrate exceptional performance in terms of enduring heavier traffic loads and varying climatic conditions. The research in [25] describes the results of modifying conventional asphalt bitumens using carbon nanopowder.

The work [26] investigated the effect of molecular weight, tactility, and functional side groups of polypropylenes on the characteristics of polymer-modified bitumen (PMB) with regard to four types of polymers and three methods of polymer modification (5%, 10%, and 16% polymer addition).

This study investigates the potential of PLA (Figure 1) as a modifier. PLA belongs to biodegradable polymers for a full biodegradation process that has to be ensured by hydrolysis processes due to elevated temperature, moisture, acids, or bases [27, 28].

This study aims to determine the effect of different content of PLA on the change in the chemical and physical properties of biopolymer bitumen during its modification. To achieve the goal of this work, it is planned to perform the following tasks:

(1) To determine the quality indicators of bitumen and PLA for research, as well as develop a technique for adding PLA to bitumen in the amount of 4%, 6%, 8%, and 10%.

(2) To determine the impact of the addition of PLA on such indicators of bitumen quality as molecular weight, molar mass, penetration, softening temperature, ductility, and stability during storage according to previously developed methods.

(3) To determine the optimal amount of PLA additive to bitumen in terms of the best ratio of its qualitative characteristics, as well as offer a possible mechanism of interaction between PLA and bitumen.

Table 1: Physical properties of the bitumen used.

Parameter	Unit of measure	Value
Depth of needle penetration at 25°C	mm/10	82
Softening point by ring and ball	°C	47
Ductility at 25°C (min.)	cm	100
Elasticity	%	18
Brittleness temperature	°C	−29

Table 2: Quality parameters of polylactic acid.

Parameter	Measurement unit	Value
Impact strength	kJ $(m)^2$	12.9
Tensile strength	MPa	94.4
Relative elongation	%	34
Flexural strength	MPa	102.5
Melt flow Index	g/10 min	3
Melting temperature	°C	177
Molecular weight	g (mol)	90,000
Molecular weight distribution	g (mol)	80,000–100,000

2. Materials and Methods

2.1. Materials. A sample of petroleum road bitumen from CASPI BITUM (Kazakhstan) was used for the study. Table 1 summarizes the physical properties of the bitumen used in this study. Bitumen is a highly viscous, black, and sticky substance that is widely used in road construction, waterproofing, and other applications. The physical properties of bitumen affect its performance and suitability for different purposes.

Table 1 shows that the bitumen used in this study has a moderate depth of needle penetration at 25°C, indicating a medium-hard grade of bitumen. It also has a relatively low softening point by ring and ball, suggesting a low resistance to flow at elevated temperatures. The ductility and elasticity of the bitumen are high, implying a good plasticity and viscoelasticity. The brittleness temperature of the bitumen is low, indicating a low susceptibility to cracking or shattering at low temperatures. In conclusion, Table 1 reveals that the bitumen used in this study has a balanced set of physical properties that make it suitable for the intended application. However, the data in Table 1 are based on laboratory measurements and may not reflect the actual behavior of the bitumen in the field. Therefore, further tests and validations are needed to confirm the reliability and accuracy of the data.

A sample of PLA from Zhejiang Hisun (China) was used for the study. The quality parameters of PLA used for the study are shown in Table 2.

Below we briefly describe the research methods and the results obtained.

2.1.1. Impact Strength. The amount of energy absorbed by a material when it is subjected to an impact load. It indicates the toughness and durability of the material. The impact strength of PLA is 12.9 kJ/m^2, which is lower than some other plastics, such as ABS or PC.

Table 3: Molecular properties of PLA-modified bitumen.

PLA (%)	Retention time (min)	Average molecular weight (Mw)	Average molar mass (Mn)
0	26.073	1,263	1,215
4	26.087	1,341	1,232
6	26.104	1,466	1,291
8	26.127	2,171	1,317
10	26.144	2,759	1,395

2.1.2. Tensile Strength. The maximum stress that a material can withstand before breaking when it is stretched. It indicates the strength and stiffness of the material. The tensile strength of PLA is 94.4 MPa, which is comparable to PS or PET.

2.1.3. Relative Elongation. The percentage of increase in length that a material undergoes when it is stretched until it breaks. It indicates the ductility and flexibility of the material. The relative elongation of PLA is 34%, which is higher than some other plastics, such as PP or PE.

2.1.4. Flexural Strength. The maximum stress that a material can withstand before breaking when it is bent. It indicates the resistance and rigidity of the material. The flexural strength of PLA is 102.5 MPa, which is higher than some other plastics, such as PVC or LDPE.

2.1.5. Melt Flow Index. The rate of flow of a melted material through a standard die under a specified pressure and temperature. It indicates the viscosity and processability of the material. The melt flow index of PLA is 3 g/10 min, which is lower than some other plastics, such as HDPE or PS.

2.1.6. Melting Temperature. The temperature at which a solid material becomes a liquid. It indicates the thermal stability and heat resistance of the material. The melting temperature of PLA is 177°C, which is higher than some other plastics, such as PET or PA.

2.1.7. Molecular Weight. The mass of 1 mol of a polymer molecule based on the number and weight of its repeating units. It indicates the size and complexity of the polymer chain. The molecular weight of PLA is 90,000 g/mol, which is lower than some other plastics, such as PC or PEEK.

2.1.8. Molecular Weight Distribution. The range of molecular weights of the polymer molecules in a sample. It indicates the uniformity and consistency of the polymer chain. The molecular weight distribution of PLA is 80,000–100,000 g/mol, which is narrower than some other plastics, such as PE or PMMA.

3. Preparation of Modified Bitumen

Bitumen samples weighing ~200 g were placed in 1,000 cm^3 aluminum containers, after which they were heated in an oven to a temperature of ~150°C. We used five samples; each analysis was determined by five replicate determinations. To obtain final concentrations of PLA, we dissolved bitumen samples in chloroform. We ensured the complete evaporation of chloroform used in the production of the modified bitumen and addressed the impact of residual chloroform on the bitumen's properties. Concentrations of 4%, 6%, 8%, and 10% PLA were chosen based on the results of the work [29], which summarized the results of using wax additives when modifying bitumen. Next, the prepared PLA was mixed with heated bitumen in a laboratory mixing machine. The prepared modified bitumen will later be used for research.

4. Results and Discussion

The results of gel permeation chromatography (retention time, average molecular weight, and average molar mass) of the studied bitumen samples are shown in Table 3.

Table 3 shows that as the PLA percentage increases, the retention time, the average molecular weight, and the average molar mass also increase. This means that the PLA molecules interact with the bitumen molecules and form larger and more complex structures. This can affect the viscosity, elasticity, and stability of the modified bitumen. According to a previous study, PLA modification can enhance the chemical and physical properties of bitumen, such as consistency, ductility, and thermal resistance. However, the optimal PLA percentage and the compatibility of PLA with different types of bitumen may vary depending on the application and the environmental conditions. Analyzing the data shown in Table 3, one can conclude that when the PLA content increased to 10%, the average molecular weight of the modified bitumen increased from 1,263 to 2,759 Mw, the average molar mass increased from 1,215 to 1,395 Mn.

The obtained studies are consistent with the results of work [30], in which the properties of bitumen modified by polyethylene and polyethylene glycol with different molecular weights were studied.

Below are mathematical Equations (1)–(3) that allow you to predict the values of retention time (RT), average molecular weight (AMW), and average molar mass (AMM) depending on the PLA content:

$$\mathrm{RT} = 0.0004\,\mathrm{PLA}^2 + 0.0044\,\mathrm{PLA} + 25.072;\ R^2 = 0.9937, \tag{1}$$

$$\mathrm{AMW} = 26.776\,\mathrm{PLA}^2 - 72.025\,\mathrm{PLA} + 1267.5;\ R^2 = 0.9852, \tag{2}$$

$$\mathrm{AMM} = 1.8293\,\mathrm{PLA}^2 + 3.4008\,\mathrm{PLA} + 1213.7;\ R^2 = 0.9811. \tag{3}$$

The increase in the molecular weight of mixtures occurs both due to the introduction of a component with a

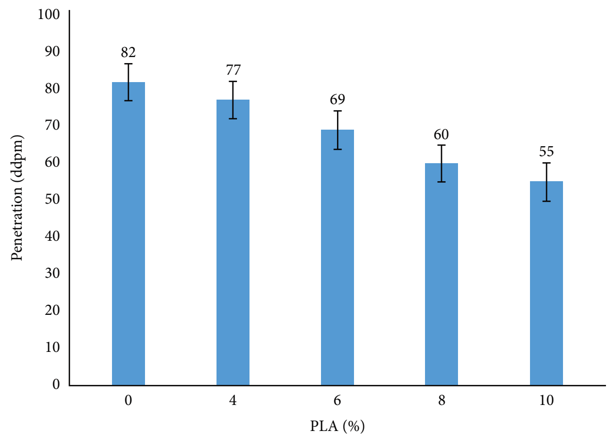

Figure 2: Penetration values.

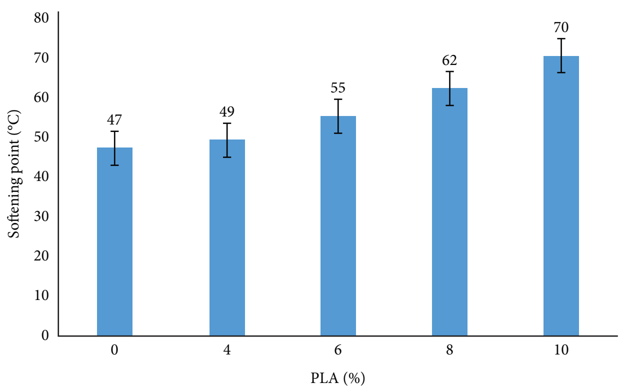

Figure 3: Softening point values.

molecular weight much greater than that of bitumen, and due to the intensification of chemical reactions between PLA and bitumen. Analyzing the data shown in Figure 2, we can come to the conclusion that an increase in the content of PLA leads to a decrease in the penetration value. In particular, the introduction of PLAin the bitumen composition in an amount of 4% leads to a decrease in the penetration value from 82 to 77 ddpm compared to the control sample; increasing the PLA content to 6% leads to a decrease in the penetration value to 69 ddpm; increasing the PLA content to 8% leads to a decrease in the penetration value to 60 ddpm; and increasing the PLA content to 10% leads to a decrease in the penetration value to 55 ddpm.

The histogram of the effect of the PLA additive amount on the modified bitumen softening point is shown in Figure 3. Analyzing the above correlation, one can conclude that increasing the PLA content in bitumen from 0% to 10% leads to an increase in the softening point from 47 to 70°C or ~49%. The ductility of all samples was >100 cm. Table 4 shows the results of determining the stability of modified bitumen during its storage.

From the data presented in Table 4, it can be concluded that all the tested bitumen samples are characterized by increased ductility at 25°C (>100 cm). The elevated concentration of PLA in bitumen leads to an augmentation in the quantity of asphaltenes in the altered binders, hence enhancing the rigidity characteristics of the changed bitumen. Consequently, the modified binder's vulnerability to temperature fluctuations is reduced. Bitumen, especially bitumen treated with PLA, has improved performance at elevated temperatures as a result of reduced sensitivity [16, 17]. The findings from previous studies [12, 14, 31, 32] support the notion that modified bitumen exhibits greater resistance to elevated temperatures compared to regular bitumen. The quality indicators of biopolymer bitumen are comparable with previously obtained results [5, 13, 17, 32, 33]. Based on the findings of research [34], modified bitumen is deemed stable if the interval in softening temperature is below 2.5°C. If the modifier does not dissolve in the modified bitumen when stored at elevated temperatures, it leads to phase separation and the formation of a nonuniform mixture. According to the literature [35, 36], bitumen that has limited stability during storage is not suitable for many purposes.

Table 4: Ductility and stability of bitumen during storage.

PLA (%)	Stability during storage (°C)		
	Top	Bottom	Difference
0	—	—	—
4	51	52	1
6	59	61	10
8	60	62	2
10	69	73	4

The research investigates the process of modifying bitumen by combining a polymer with it, which includes both physically mixing the polymer with bitumen and chemically interacting the two substances. This process has been documented in references [19, 37, 38]. When preparing the modified bitumen at a temperature of 180°C, it is important to take into account the possible deterioration of the bitumen, especially if the modification process lasts for up to 2 hr.

Figure 4 presents reactions between bitumen and PLA [39, 40]. During the processing step, the reaction or chemical compatibility leads to the synthesis of copolymers in the same location where they are being processed [38]. The identification of this process offers essential comprehension of the chemical interplay between PLA and bitumen, possibly facilitating the exploration of cutting-edge and technologically sophisticated modifiers.

Kazakhstan has an extreme continental climate characterized by lengthy, scorching summers, and frigid winters. The winter in the northern region of the nation is characterized by a prolonged and frigid climate. In Nur-Sultan, temperatures may sometimes plummet to as low as −52°C, although sporadic spells of milder weather can elevate temperatures to 5°C. According to this knowledge, there are no other ways for identifying additives that might enhance the performance properties of bitumen.

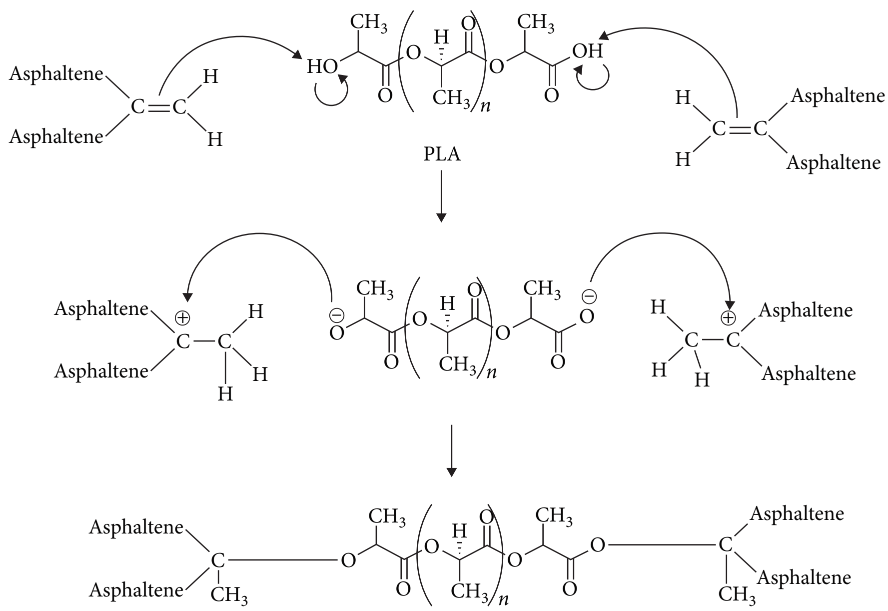

FIGURE 4: Reaction mechanism between PLA and bitumen.

5. Conclusions

The research investigated the impact of incorporating several ratios (4%, 6%, 8%, and 10%) of PLA into the bitumen manufactured by a company in Kazakhstan. Research has shown that augmenting the quantity of PLA in bitumen leads to a corresponding augmentation in the average molecular weight and average molar mass. The results of the penetration test indicate a strong association, indicating that the inclusion of 4% PLA results in a substantial 18.5% increase in penetration compared to the original bitumen. However, as the concentration of PLA reaches greater levels (8%–10%), the degree of penetration decreases significantly. The inclusion of 4% PLA results in a little improvement in penetration (from 65 to 77 ddpm), maybe owing to the limited dispersion properties of the modified bitumen. An elevation in the concentration of PLA in bitumen results in a reduction in penetration rather than an augmentation (from 77 to 55 ddpm). The devaluation may be attributed to the increased dispersion of PLA inside the bitumen. Furthermore, the incorporation of higher concentrations of PLA (8% and 10%) into bitumen results in decreased penetration values in comparison to untreated bitumen. This suggests that the PLA modifier enhances the rigidity of the bitumen. The experiment showed that increasing the content of PLA in bitumen from 0% to 10% resulted in a substantial increase in the softening temperature from 47 to 70°C, suggesting an estimated spike of around 49%. All the bitumen samples assessed exhibited enhanced flexibility at a temperature of 25°C, surpassing a measurement of 100 cm. Empirical evidence has shown that the addition of polymers, such as PLA, may enhance the heat resistance of bitumens and inhibit the separation of distinct phases. The insufficient ability of bitumen to withstand high temperatures, which has been enhanced by adding a 10% amount of PLA, might be attributed to the reduced compatibility between PLA and bitumen. This study has provided a theoretical explanation for the interaction between PLA and bitumen. Multiple studies consistently demonstrate that the most dependable indicators of quality are associated with modified bitumen, particularly when it includes 8% PLA. The findings of the study may be used to alter bitumen PLA and provide a foundation for future investigations into the application of changed PLA bitumen in the particular climatic circumstances of Kazakhstan.

Data Availability

Data will be available upon request.

Conflicts of Interest

The authors declare that there are no conflicts of interest regarding the publication of this paper.

Authors' Contributions

Assel Jexembayeva contributed to conceptualization, validation, investigation, data curation, writing—original draft, visualization, and writing—review and editing. Marat Konkanov contributed to methodology, formal analysis, investigation, data curation, visualization, and writing—review and editing. Larisa Mamedova contributed to conceptualization, methodology, resources, writing—original draft, and writing—review and editing. Lyazat Aruova contributed to validation, formal analysis, resources, writing—original draft, and writing—review and editing.

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