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# STUDY ON THE TECHNICAL LIGNIN EFFECT ON THE ROAD BITUMEN PROPERTIES

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**Abstract.** The effect of technical lignin on the characteristics of road bitumen obtained by oxidation of petroleum residues (oxidized bitumen) was examined. Two samples of hydrolysis-type technical lignin, obtained as a by-product of the production of feed yeast, were used. The first sample was technical lignin purified by sieving to remove unprocessed plant material. The second sample was technical lignin, purified by sieving to remove unprocessed plant material and flotation enrichment to remove inorganic components. The effect of adding two samples of technical lignin in different amounts on the main performance characteristics of road bitumen was analyzed. It was concluded that it is advisable to use technical lignin as a relatively inexpensive substitute for much more expensive road oil bitumen. It was also concluded that flotation beneficiation is effective as a method of increasing the purity of technical lignin.

**Keywords:** hydrocarbon raw materials, technical lignin, lignocellulosic biomass, lignin sources, road bitumen, modification.

## 1. Introduction

Despite the rational consumption and continuous development and improvement of technological processes for the extraction, processing, and use of non-renewable energy sources, their global reserves are rapidly declining. That is why more and more industries are now turning to energy efficiency and resource conservation. This is facilitated by the steady increase in the amount of industrial waste and by-products of processing various types of raw materials, including organic ones, which can be used as energy sources. Another positive trend is the steady growth in demand for renewable energy sources, various chemicals, and energy carriers in almost all industries, including the road sector[1].

The key binder in the production of asphalt roads is bitumen, which is derived from a non-renewable source of raw materials – oil. Despite the importance of bitumen as a construction material in many industries, oil refining is primarily focused on the production of fuels, lubricants, and raw materials for the chemical industry. Therefore, as oil reserves are depleting and fuels and lubricants are critical for industry and society, the trend toward reducing bitumen consumption is constantly increasing. A complete rejection of bitumen, in particular road bitumen, is currently impossible due to the lack of a quality alternative. However, many scientific studies are aimed at reducing its consumption by improving its quality with various modifying agents, as well as partial replacement of road bitumen with related materials of organic origin.

Considering its characteristics, resources, and safety for humans, lignin is one of the best products in this category. Today, it is a rather underrated but widely available material, characterized by significant and renewable reserves and used in many industries. Lignin, the second most abundant organic polymer on Earth, is one of the main reasons for the viscoelastic behavior of plants[2]. In particular, this product is derived from plant material. Its content in hardwood averages 27–33 %, in softwood – 18–25 %, and in herbaceous plants – 17–24 %. In industry, lignin is mainly obtained from plants and agricultural processing waste. A significant percentage of this organic component is also obtained from sugar cane pulp, clover, alfalfa, some types of bamboo, and cereals. In addition, lignin raw materials include rice husks, wheat and barley straw, nut shells, as well as corn and potato stalks and leaves left in the fields after harvest. Among the valuable products derived from lignocellulosic biomass are lignin-based polyurethane foam, carbon fibers, phenolic resin, various biomedical applications (hydrogels), *etc*. Meanwhile, a significant amount of lignin-containing materials is still simply burned to generate heat, and the rest remains unused[3, 4].

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Lignocellulosic biomass needs to be pretreated before commercial lignin-based products can be produced. The choice of pretreatment method depends on the source of the lignocellulosic biomass and the purpose of the pretreatment[5]. Among the methods used to extract lignin from biomass are the following:

• Chemical pretreatment, which involves the dissolution of biomass in various solvents, including alkaline (*e. g.*, NaOH, $Ca(OH)_2$, KOH), acidic (*e. g.*, $CH_3COOH$, $H_2SO_4$, $H_3PO_4$), metal-containing, ionic liquids, and sub or supercritical fluids. Chemical pretreatment contributes to the destruction of the lignocellulosic material structure as a result of interaction with inter- and intra-polymer bonds of cellulose, lignin, and hemicellulose, which are components of biomass[6].

• Physical pretreatment. These methods are used to reduce the size of lignocellulosic particles and maximize the surface area. Methods such as milling, microwave irradiation, extrusion, freezing, sonication, and pyrolysis are used for this purpose. Despite their ease of use and lack of formation of inhibitory molecules, these methods can have limitations in achieving adequate lignin fractionation[5, 7].

• Physicochemical treatment, which combines two previous methods and allows to obtain a higher yield of desired products due to a greater degree of biomass processing.

Biological pretreatment is one of the cleanest processes of biomass decomposition. The biological method for the lignocellulosic biomass pretreatment uses various microorganisms, mostly enzymes, bacteria, and fungi, and is conducted under milder conditions compared to previous processes.

Before synthesizing many different types of valuable products from lignin, it is first depolymerized from the biomass and then extracted[8]. After the biomass has been pretreated, the lignin separated from the pretreated biomass is called technical lignin. Technical lignin is divided into the following types[5]:

• Alkali lignin obtained *via* Kraft process. The process of obtaining this product is based on treating biomass with sodium sulfide ($Na_2S$) and sodium hydroxide (NaOH)[9]. Alkali lignin differs from native lignin (*i. e.*, lignin on plants). This type of lignin from different sources (softwood and hardwood) is used to produce both water-soluble and insoluble products. In particular, alkali lignin is used to produce flame retardant, resins, polyurethanes, thermoplastics, composites, carbon fibers, and other water-soluble polymers. In recent years, alkali lignin has become increasingly available to consumers.

• Lignosulfonate, which has been considered an anionic polyelectrolyte soluble in an acid solution, water, and high polar organic solvents[10]. Lignosulfonate is cheaper than other forms of lignin and can be used not only in a pellet form for animal feed (as a source of sodium and calcium) but also as a lubricant for wood pellet production[11]. Moreover, this form of lignin can be used as an adsorbent, coating material, dispersant, polymer additive, *etc*.

• Soda lignin which is obtained from the soda pulping process or soda-anthraquinone production. The soda process is known as sulfur-free, which is a major advantage compared to the kraft process[12]. Soda lignin is mostly used as a raw material for water-insoluble materials such as polyurethane foams, polyolefins, resins, and adsorbents.

• Hydrolyzed lignin. Based on most biorefining concepts, wood can be treated with acid, base, or enzyme to dissolve cellulose and a part of hemicelluloses. Meanwhile, lignin remains undissolved and is usually used as fuel. The low solubility, reactivity, and high viscosity of hydrolyzed lignin limit its use in aqueous media[6]. Hydrolyzed lignin is used as an adhesive, flocculant, dispersant, and flame retardant.

• Organosolv lignin. The principle of organosolv process is based on the extraction of hemicellulose and lignin from biomass using organic solvents, such as methanol, acetone, ethanol, or mixtures of organic solvent and water within the temperature range of 100–250 °C. The main drawback of the organosolv process, however, is the high cost of solvent recovery, the thermal instability of solvents, as well as the high environmental risks raised by using solvents[11]. Organosolv lignin is mostly used as nutrition for animal feed due to its sulfur-free formula. Also, this form of lignin is used as the raw material for the production of mostly insoluble products such as resins, wood adhesives, plasticizers, flame retardants, and some water-soluble products such as adsorbents and dispersants[6].

Lignin has a very complex chemical structure that varies depending on the source and purification process, resulting in the fact that the exact chemical structure of lignin is not fully known[13]. Lignin, a high molecular weight polymer, contains phenolic hydroxyl, carboxyl, benzyl alcohols, and methoxyl groups[14].

Lignin has several advantageous characteristics, including antioxidant and antibacterial properties, biodegradability and compatibility with other substances, outstanding thermal reactivity, and adhesive properties[4, 15, 16].

Given the wide range of lignin properties, many scientific studies describe the possibility of adding it to road bitumen. However, these studies usually use different grades of purified lignin[17–24]. The purpose of the studies described in this article was to determine the effect of unrefined lignin (which, as a by-product of various industries, is stored for a long time in places of accumulation and storage under the influence of external atmospheric factors) on the characteristics of road bitumen obtained by oxidation of petroleum residues.

# 2. Experimental

## 2.1. Materials

For the experimental studies, road bitumen and technical lignin were used as initial materials.

Samples of oxidized road petroleum bitumen of BND 70/100 grade were withdrawn at PJSC "Ukrtatnafta" (Kremenchuk, Ukraine). Its physicochemical characteristics are presented in Table 1.

A sample of technical lignin was taken from a storage facility in the Zaporizhzhia region of Ukraine. It is of the hydrolytic type (obtained using dilute sulfuric acid) and is a by-product of feed yeast production. The collected sample contained a significant amount of various undesirable impurities, primarily unprocessed plant material (Fig. 1A). It also likely contained inorganic components (slaked lime $Ca(OH)_2$ and gypsum sludge) in the form of small light-colored granules or lumps (Fig. 1A) that may have been exposed to lignin during the yeast production process. $Ca(OH)_2$ is used as a neutralizer of sulfuric acid in the hydrolysis process of plant material. Gypsum sludge is a by-product of sulfuric acid neutralization. Therefore, the first stage of work was the preparation of lignin. Using sieves with different mesh sizes, it was classified and cleaned from unwanted impurities. The resulting fraction $\leq 0.14$ mm was then dried to an air-dry state. As a result, a sample of technical lignin (*TL*1) was obtained (Fig. 1B), and its qualitative and quantitative analysis is presented in Table 2.

**Table 1.** Physicochemical characteristics of bitumen

| Index | Unit of measurement | Value |
|---|---|---|
| Penetration at 25 °C | dmm | 78 |
| Softening point | °C | 52.6 |
| Ductility at 25 °C | cm | 58 |
| Elastic recovery at 25 °C | % | 17.5 |
| Solubility in organic solvent | % | 99.95 |
| Adhesion to gravel | mark | 3.5 |
| Adhesion to glass | % | 65 |
| Resistance to hardening at 163 °C (RTFOT method): | | |
| mass change | wt. % | 0.086 |
| softening point after RTFOT | °C | 59.6 |
| penetration at 25 °C after RTFOT | dmm | 39 |
| softening point change | °C | 6.8 |
| retained penetration | % | 50.0 |

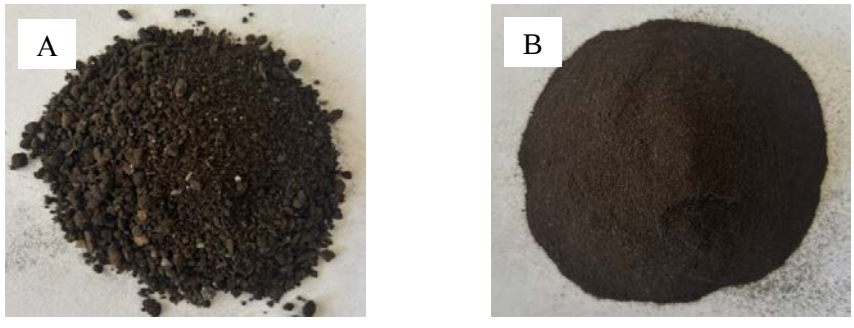


**Fig. 1.** Technical lignin.
A – collected from the place of storage; B – after cleaning from mechanical impurities and drying

**Table 2.** Quantitative and qualitative analysis of technical lignin (*TL*1)

| Index | Unit of measurement | Value |
|---|---|---|
| Moisture content, $W^a$ | wt. % | 2.63 |
| Ash content, $A^d$ | wt. % | 27.45 |
| Yield of volatile substances, $V^d$ | wt. % | 49.74 |
| Total sulfur content, $S_t^d$ | wt. % | 0.71 |
| Carbon content, $C^d$ | wt. % | 42.36 |
| Hydrogen content, $H^d$ | wt. % | 4.26 |
| Nitrogen content, $N^d$ | wt. % | 0.61 |
| Oxygen content, $O_d^d$ | wt. % | 24.61 |

## 2.2. Experimental procedure

To concentrate technical lignin, a modified coal flotation enrichment method[25] was used, adapted to the specific characteristics of the raw material. The procedure was as follows: a pulp was prepared in a porcelain vessel by mixing technical lignin (*TL*1) and water (1:12 w/w). Then flotation reagents (Extraflock 160 as a flocculant and Montanol as a foaming agent) were added to the resulting pulp in the specified amounts. After thorough mixing with a metal spatula and keeping for 5 min, the pulp was poured into a pneumatic-mechanical flotation machine. Flotation was carried out for 30 min, continuously collecting the flotation concentrate (foam) from the surface of the pulp with a spatula into a separate container. To remove moisture, the obtained flotation concentrate was first filtered and then dried at a temperature of 105 °C. As a result, a sample of technical lignin (*TL*2) was obtained, the qualitative and quantitative analysis of which is presented in Table 3.

**Table 3.** Quantitative and qualitative analysis of technical lignin (TL2)

| Index | Unit of measurement | Value |
|---|---|---|
| Moisture content, $W^a$ | wt. % | 3.78 |
| Ash content, $A^d$ | wt. % | 22.83 |
| Yield of volatile substances, $V^d$ | wt. % | 50.82 |
| Total sulfur content, $S_t^d$ | wt. % | 0.93 |
| Carbon content, $C^d$ | wt. % | 46.41 |
| Hydrogen content, $H^d$ | wt. % | 4.64 |
| Nitrogen content, $N^d$ | wt. % | 0.87 |
| Oxygen content, $O_d^d$ | wt. % | 24.22 |

The method for adding technical lignin to road bitumen was as follows: the required amount of bitumen was heated to the specified temperature under stirring; then the necessary amount of lignin was added and maintained for a set time under constant stirring. Based on the comparison of the qualitative indices of the original bitumen and the bitumen-lignin mixture, conclusions were drawn about the effectiveness of using technical lignin as a modifier or substitute for road petroleum bitumen.

## 2.3. Methods of analysis

The main physicochemical characteristics of the original bitumen and bitumen-lignin mixture were determined according to standard methods: softening temperature[26]; penetration[27]; ductility[28]; elastic recovery[29]; adhesion to glass[28] and gravel[30].

To determine the bitumen adhesion to the surface of stone mineral material, granite crushed stone (S1) of the required fractions, selected from natural stone at LLC Novograd-Volyn Stone Crushing Plant, was used.

To study the resistance of bitumen to technological aging, we used the RTFOT method[31], which involves heating samples in a furnace at a temperature of 163 °C and a constant supply of air for 75 min. Under the appropriate conditions, the film of bituminous binder, which is scrolled in cylindrical flasks, is constantly overflowing, which allows activating the processes of technological aging evenly for the entire bitumen sample.

To show the effect of aging on the bitumen properties, the value of retained penetration was calculated using Eq. (1):

$$\text{Retained penetration} = \frac{P^{25}_{\text{AGED}}}{P^{25}_{\text{UNAGED}}} \cdot 100\ \%, \quad (1)$$

where $P^{20}_{\text{AGED}}$ is the needle penetration depth determined at 25 °C for the sample after aging, 0.1 mm; $P^{20}_{\text{UNAGED}}$ is the needle penetration depth determined at 25 °C for the sample before aging, 0.1 mm.

To evaluate the effectiveness of flotation enrichment as a method for increasing the purity of unrefined lignin, the degrees of increase or decrease in the quantitative and qualitative characteristics of the technical lignin sample *TL*2 relative to the *TL*1 sample were calculated:

- Degree of reduction in ash content:

$$A_-^d = \frac{(A^d_{TL1} - A^d_{TL2})}{A^d_{TL1}} \cdot 100, \quad (2)$$

where $A_-^d$ is a degree of reduction in ash content, %; $A^d_{TL1}$ and $A^d_{TL2}$ are values of ash content in the technical lignin samples *TL*1 and *TL*2, respectively, wt. %.

- Degree of increase in total sulfur content:

$$S^d_{t+} = \frac{(S^d_{t,\ TL2} - S^d_{t,TL1})}{S^d_{t,TL1}} \cdot 100, \quad (3)$$

where $S^d_{t+}$ is a degree of increase in total sulfur content, %; $S^d_{t,\ TL1}$ and $S^d_{t,\ TL2}$ are values of total sulfur content in the technical lignin samples *TL*1 and *TL*2, respectively wt. %.

- Degree of increase in carbon content:

$$C^d_+ = \frac{(C^d_{TL2} - C^d_{TL1})}{C^d_{TL1}} \cdot 100, \quad (4)$$

where $C_{+}^{d}$ is a degree of increase in carbon content, %; $C_{TL1}^{d}$ and $C_{TL2}^{d}$ are values of carbon content in the technical lignin samples *TL*1 and *TL*2, respectively wt. %.

- Degree of increase in hydrogen content:

$$H_{+}^{d} = \frac{(H_{TL2}^{d} - H_{TL1}^{d})}{H_{TL1}^{d}} \cdot 100, \quad (5)$$

where $H_{+}^{d}$ is a degree of increase in hydrogen content, %; $H_{TL1}^{d}$ and $H_{TL2}^{d}$ are values of hydrogen content in the technical lignin samples *TL*1 and *TL*2, respectively wt. %.

- Degree of increase in nitrogen content:

$$N_{+}^{d} = \frac{(N_{TL2}^{d} - N_{TL1}^{d})}{N_{TL1}^{d}} \cdot 100, \quad (6)$$

where $N_{+}^{d}$ is a degree of increase in nitrogen content, %; $N_{TL1}^{d}$ and $N_{TL2}^{d}$ are values of nitrogen content in the technical lignin samples *TL*1 and *TL*2, respectively wt. %.

- Degree of reduction in oxygen content:

$$O_{d-}^{d} = \frac{(O_{d,TL1}^{d} - O_{d,TL2}^{d})}{O_{d,TL1}^{d}} \cdot 100, \quad (7)$$

where $O_{d-}^{d}$ is a degree of reduction in oxygen content, %; $O_{d,TL1}^{d}$ and $O_{d,TL2}^{d}$ are values of oxygen content in the technical lignin sample *TL*1 and *TL*2, respectively wt. %.

## 3. Results and Discussion

Table 4 presents the results of adding unrefined lignin (TL1) to road bitumen.

Table 4 shows that the binder quickly loses its performance characteristics when bitumen is combined with unrefined lignin.

This is confirmed by a decrease in penetration from 7.8 to 2.8 mm, an increase in softening point by 15.6 °C, and a decrease in ductility from 58 to 12 cm.

At the same time, bitumen adhesion does not deteriorate. However, this is most likely due to an increase in its viscosity, which is a consequence of prolonged exposure to sufficiently high temperatures, and not to lignin. Based on the results obtained after the aging of the bitumen-lignin mixture, it can also be argued that the effect of lignin on the binder is practically absent, and, in some cases, even worsens the bitumen properties. This is especially true for the change in softening point after RTFOT, which increases to 8.8 °C compared to the original bitumen (6.8 °C) and bitumen mixed without lignin (6.6 °C). At the same time, this can be explained by the negative impact of the high mixing temperature (180 °C), which is critical for bitumen properties. As for the other indicators obtained after aging by the RTFOT method, it can be seen that the values of residual penetration and mass change for sample 2 (without lignin) and samples 3–6 are almost the same. Thus, it can be concluded that it is impossible to use technical lignin, for example, as a modifier of road bitumen to improve its performance properties.

On the other hand, it is evident that bitumen containing 1.0 wt. % TL1 (sample 3) is practically identical in characteristics to Sample 2 (original bitumen mixed under the same conditions but without lignin). Therefore, it can be stated that in small quantities (in this case, 1.0 wt. %), lignin can serve as a relatively inexpensive substitute for significantly more expensive road petroleum bitumen.

It should be noted that technical lignin is not completely soluble in bitumen. Thus, all bitumen-lignin mixtures obtained using TL1 (samples 3–7) were heterogeneous. This is a significant drawback, since international and national standards for bitumen modified with various additives[32, 33] require complete dissolution of the modifiers in the binder. However, as mentioned above, technical lignin is not a modifier for road bitumen. Moreover, in the preparation of asphalt concrete mixtures, various additives, such as mineral powders, are used in small quantities along with bitumen and crushed stone, which also do not dissolve in bitumen. Therefore, it can be assumed that the insolubility of technical lignin in bitumen will not affect the ultimate goal of obtaining high-quality asphalt concrete.

Further, after analyzing the results obtained, the task of the next stage of work was determined, namely, to increase the lignin content in bitumen without deteriorating its performance characteristics. This is likely to be achieved by increasing the degree of purification of technical lignin using the flotation enrichment method.

Thus, the purpose of flotation enrichment was to increase the organic content of technical lignin by separating inorganic inclusions. For this purpose, as mentioned in the previous subsection, the same method was used as for coal, and Tables 2 and 3 present the quantitative and qualitative analysis of technical lignin before and after flotation enrichment (TL1 and TL2, respectively). Based on these data, we calculated the efficiency of flotation enrichment as a method of increasing the purity of technical lignin. The obtained data are presented in Table 5.

As can be seen from the data presented in Table 5, flotation enrichment of technical lignin had the most positive effect on the ash content, carbon content, and hydrogen content. The ash content of lignin after flotation (sample TL2) decreased by almost 17 % compared to lignin before flotation (sample TL1). The amount of carbon and hydrogen increased by almost 10 % each. This indicates that flotation enrichment changed the ratio of the organic part of technical lignin to its inorganic mass in an increasing direction. The oxygen content practically did not

change (the degree of increase in its content in TL2 relative to TL1 is 1.58 %). Along with this, a significant increase in the nitrogen and sulfur content is observed (the degrees of increase in their content in TL2 relative to TL1 are 42.62 % and 30.99 %, respectively). However, it can be assumed that such a sharp increase in the amount of these elements in lignin after enrichment will not change its properties and, accordingly, its impact on road bitumen, since the initial amounts of nitrogen and sulfur in lignin before enrichment were insignificant – 0.61 % and 0.71 wt. %, respectively.

**Table 4.** The influence of technical lignin (TL1) on the physical and chemical characteristics of road bitumen

| Index | Unit of measurement | BND 70/100 | | BND 70/100 + TL1, wt. % | | | | |
|---|---|---|---|---|---|---|---|---|
| | | Original (Sample 1) | After mixing without lignin (Sample 2) | 1.0 (Sample 3) | 1.5 (Sample 4) | 3.0 (Sample 5) | 5.0 (Sample 6) | 7.0 (Sample 7) |
| Penetration at 25 °C | dmm | 78 | 41 | 42 | 38 | 35 | 33 | 28 |
| Softening point | °C | 52.8 | 60.6 | 61.1 | 62.7 | 65.6 | 66.4 | 67.4 |
| Ductility at 25 °C | cm | 58 | 25.5 | 24.0 | 21.0 | 17.5 | 15.5 | 12.0 |
| Elastic recovery at 25 °C | % | 17.5 | 17.0 | 24.0 | 22.5 | 22.0 | 21.0 | 21.5 |
| Adhesion to gravel | mark | 3.5 | 3.5 | 3.5 | 3.5 | 3.5 | 3.5 | 3.5 |
| Homogeneity | – | Yes | Yes | No | No | No | No | No |
| Resistance to hardening at 163 °C (RTFOT method): | | | | | | | | |
| mass change | wt. % | 0.086 | 0.10 | 0.154 | 0.195 | 0.215 | 0.273 | 0.28 |
| softening point after RTFOT | °C | 59.6 | 67.2 | 68.8 | 70.5 | 73.6 | 74.8 | 76.2 |
| penetration at 25 °C after RTFOT | dmm | 39 | 31 | 28 | 26 | 25 | 23 | 22 |
| softening point change | °C | 6.8 | 6.6 | 7.7 | 7.8 | 8.0 | 8.4 | 8.8 |
| retained penetration | % | 50.0 | 75.6 | 66.6 | 68.4 | 71.4 | 69.7 | 78.6 |

* Mixing conditions: temperature 180 °C, time 120 min, intensity of mixing 800 rev/min.

**Table 5.** Effectiveness of flotation enrichment as a method for increasing the purity of technical lignin

| Index | Unit of measurement | Value |
|---|---|---|
| Degree of reduction in ash content | % | 16.83 |
| Degree of increase in carbon content | % | 9.56 |
| Degree of increase in hydrogen content | % | 8.92 |
| Degree of increase in nitrogen content | % | 42.62 |
| Degree of reduction in oxygen content | % | 1.58 |
| Degree of increase in the content of total sulfur | % | 30.99 |

**Table 6.** Effect of technical lignin (TL2) on the physicochemical characteristics of road bitumen

| Index | Unit of measurement | BND 70/100 | | BND 70/100 + TL2, wt. % | | | | |
|---|---|---|---|---|---|---|---|---|
| | | Original (Sample 1) | After mixing without adding lignin (Sample 2) | 1.0 (Sample 8) | 1.5 (Sample 9) | 3.0 (Sample 10) | 5.0 (Sample 11) | 7.0 (Sample 12) |
| Penetration at 25 °C | dmm | 78 | 41 | 46 | 42 | 38 | 35 | 32 |
| Softening point | °C | 52.8 | 60.6 | 61.7 | 62.7 | 66.4 | 67.0 | 67.6 |
| Ductility at 25 °C | cm | 58 | 25.5 | 27.0 | 23.2 | 23.5 | 22.0 | 20.5 |
| Elastic recovery at 25 °C | % | 17.5 | 17.0 | 24.0 | 23.5 | 24.0 | 25.5 | 28.5 |
| Adhesion to gravel | mark | 3.5 | 3.5 | 3.5 | 3.5 | 3.5 | 3.5 | 3.5 |
| Homogeneity | – | Yes | Yes | No | No | No | No | No |
| Resistance to hardening at 163 °C (RTFOT method): | | | | | | | | |
| mass change | wt. % | 0.086 | 0.10 | 0.105 | 0.145 | 0.165 | 0.195 | 0.220 |
| softening point after RTFOT | °C | 59.6 | 67.2 | 69.2 | 69.9 | 72.8 | 73.4 | 74.6 |
| penetration at 25 °C after RTFOT | dmm | 39 | 31 | 31 | 30 | 28 | 27 | 25 |
| softening point change | °C | 6.8 | 6.6 | 7.5 | 7.2 | 6.4 | 6.4 | 7.0 |
| retained penetration | % | 50.0 | 75.6 | 67.4 | 71.4 | 73.7 | 77.1 | 78.1 |

Mixing conditions: temperature 180 °C, time 120 min, mixing intensity 800 rev/min

As mentioned above, the ultimate goal of flotation enrichment of technical lignin was to increase its content in bitumen without compromising the performance characteristics (an increase of above 1.0 wt. %, which was obtained using TL1, Table 4). To confirm or refute this, sample TL2 was added to road bitumen in different amounts under the same conditions as sample TL1. The obtained results are presented in Table 6.

Comparing the results obtained using samples TL1 and TL2, it can be concluded that flotation enrichment allows a slight increase in the content of technical lignin in road bitumen, namely from 1.0 wt. % to 1.5 wt. %. In particular, the main characteristics (penetration, softening point, ductility, and elasticity) of sample 9, for which the TL2 content is 1.5 wt. %, are practically the same as those of sample 3 (TL1 content is 1.0 wt. %). Increasing the TL2 content in bitumen above 1.5 wt. % significantly worsens the binder properties. It should also be noted that samples of technical lignin TL2, like sample TL1, do not completely dissolve in bitumen.

Given the positive effect obtained, technical lignin, purified first by sieving to remove unprocessed plant material and then by flotation enrichment to remove inorganic components, was used for further research. At the same time, when developing a full-fledged technology for using technical lignin as a partial substitute for road bitumen, an essential aspect must be the calculation of the economic feasibility of conducting flotation enrichment.

In addition, when studying the effect of technical lignin on the properties of road bitumen, it is advisable to choose such conditions for combining lignin (after flotation) with bitumen that will minimize the negative impact of the mixing process on the characteristics of the binder. This is likely to be achieved primarily by reducing the mixing temperature. These assumptions will be examined in the next stages of the study.

## 4. Conclusions

It is impossible to use technical lignin as a modifier for road bitumen because it does not completely dissolve in bitumen and leads to the deterioration of its physicochemical properties.

When mixing technical lignin with road bitumen at a temperature of 180 °C for 120 minutes in an amount of 1.0–1.5 wt. %, its negative impact on the main physicochemical characteristics of the binder is minimal. So, it can be used as a relatively inexpensive substitute for significantly more expensive road petroleum bitumen.

Before use (mixing with road bitumen), technical lignin must be purified, first by sieving to remove unprocessed plant material, and then by flotation enrichment to remove inorganic components.

Flotation enrichment of technical lignin most positively affects the ash content, carbon content, and hydrogen content. For the studied lignin, the ash content decreased by 16.83 % as a result of flotation; the amounts of carbon and hydrogen increased by 9.56 % and 8.92 %, respectively. This indicates that flotation enrichment changed the ratio of the organic part of technical lignin to its inorganic mass toward an increase in the former.

Further, the study of the technical lignin effect on the properties of road bitumen, conditions for combining lignin (after flotation) with bitumen will be selected to minimize the negative impact of the mixing process on the characteristics of the binder. In particular, it is necessary to study the effect of temperature and mixing time on the physicochemical characteristics of the bitumen-lignin mixture.

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## ДОСЛІДЖЕННЯ ВПЛИВУ ТЕХНІЧНОГО ЛІГНІНУ НА ВЛАСТИВОСТІ ДОРОЖНЬОГО БІТУМУ

***Анотація.*** *Здійснено дослідження щодо впливу технічного лігніну на характеристики дорожнього бітуму, отриманого окисненням нафтових залишків (окисненого бітуму). Використано два зразки технічного лігніну гідролізного типу, одержаного як нецільовий продукт під час виробництва кормових дріжджів. Перший зразок – технічний лігнін, очищений просіюванням від неперероблено рослинного матеріалу. Другий зразок – технічний лігнін, очищений просіюванням від неперероблено рослинного матеріалу і флотаційним збагаченням від неорганічних компонентів. Проаналізовано вплив додавання різних кількостей двох зразків технічного лігніну на основні експлуатаційні характеристики дорожнього бітуму. Зроблено висновок про доцільність використання технічного лігніну як порівняно дешевого замінника значно дорожчого дорожнього нафтового бітуму. Зроблено висновок про ефективність флотаційного збагачення як методу підвищення чистоти технічного лігніну.*

***Ключові слова:*** *вуглеводнева сировина, технічний лігнін, лігноцелюлозна біомаса, джерела лігніну, дорожні бітуми, модифікація.*