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FEASIBILITY OF PRODUCING BITUMEN FROM DIFFERENT TYPES OF TAR

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Traditionally, the presence of n-paraffins in tar is known to negatively affect the fragility, adhesion, and internal cohesion of bitumen particles, making paraffinic tar impractical for producing high-quality bitumen. This study investigates the potential enhancement of bitumen properties through the compounding of residue from the distillation of Orkhiv oil with tar. Bitumen samples were prepared via oxidation of the compounded mixture within a temperature range of 210–270^{0}C. The research explores the effects of oxidation duration (ranging from 3 to 12 hours), temperature, and oxidizer consumption on the resulting bitumen properties. Furthermore, the study examines the impact of the mixture ratio of paraffinic tar and residue from Orkhiv oil distillation on bitumen characteristics. Key bitumen properties, including softening point, ductility, penetration, penetration index, plasticity interval, brittleness temperature, and adhesion to glass, were systematically determined. Optimal process conditions were identified, specifically at a temperature of 250^{0}C and an airflow rate of 2.5 min^{-1}. The kinetic parameters of the oxidation process for the tar mixture were studied, leading to the calculation of the reaction rate constant under various influencing factors. The results demonstrate the feasibility of producing bitumen suitable for asphalt concrete coatings with improved performance characteristics.

Keywords: tar, bitumen, oxidized bitumen, compounding, oxidation, reaction rate constants.

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Introduction

Bitumen can be used in many areas of human activity as an insulating and binding material in the construction and repair of roads and airfields, various coatings, etc. Bitumen materials used as coatings have greater strength and are at least 2 or 2.5 times cheaper than cement and other materials [1].

Bitumens belong to materials with well-defined properties such as elasticity, plasticity, and viscosity, which change when the temperature changes. At the same time, physical and chemical properties are practically unrelated to the chemical composition of bitumen components, its microstructure, physical and mechanical properties. Based on a comprehensive study of the physicochemical properties, the chemical composition of bitumen and their components, the study of the supramolecular structure of their surface shows that the quality of the components has no less important influence on the structure and physicochemical properties of bitumen than their ratio [2].

The high quality of road bituminous materials is characterized by their optimal group chemical composition, structure and dispersion of the oil dispersion medium. In Ukraine, bitumen mainly is produced by traditional technology, namely the oxidation of oil residues with air [3]. In the process of oxidation, many oxidizing reactions occur, such as dehydrogenation, dealkylation, oxidative polymerization, cracking and polycondensation. As a result, the components of such residues are compacting.

It is necessary to have an appropriate initial composition of raw materials for the production of high-quality bitumen with the required combination of components; those with particular components have a significant impact on the final product. Significant influence on the quality indicators of oxidized has a bitumen the number of paraffinic-naphthenic hydrocarbons, their optimal content by the raw materials is opined to be 20–22 wt.%, if aromatic hydrocarbons at least 34 wt.%, resins at least 35 wt.%, and asphaltenes no more than 18.5 wt.%.

There are oils with a unique composition (for example, Orkhiv oil, Yabluniv oil, etc.) from which one can directly obtain high-quality bitumen materials of optimal dispersion, both oxidized and residual. However, the resources of such raw materials in Ukraine and abroad are small and cannot meet the modern needs for binding materials. Therefore, the development of bitumen production is related to obtaining compound bitumen with optimal dispersion and structure [1].

The high content of paraffinic carbohydrates is one of the main problems of the low quality of raw materials for domestically produced bitumen. In particular, it affects the bitumen production. The physicochemical manifestation of this is that normal paraffins are sufficiently inert from a chemical point of view. It means that the residues of atmospheric-vacuum distillation of high-paraffin oils are practically not suitable for obtaining high-quality road bitumen by the method of direct oxidation.

n-Paraffins dissolved in oil component bitumen is a solid saturated component that crystallizes at low temperatures, lowering the brittleness temperature of bitumen. It is known, n-paraffin is a crystalline substance, therefore it has no plastic properties and its presence reduces such a property as the fragility of bitumen, as well as reduces adhesion to the mineral filler and internal cohesion between its particles [4].

In Ukrainian state standard DSTU EN 12591, measurement of the brittleness index according to Fraas is established as optional for all EU countries, but it is specified that the content of n-paraffins in bitumen should not exceed 4.5% of the total weight of bitumen. Many European countries set limits on the maximum content of n-paraffins in road bitumen. For instance, Poland, Romania, and Sweden impose a limit of 2% of the mass, while Hungary and Italy allow up to 2.5% of the total mass. These restrictions are particularly important because the climate conditions in these countries are much milder in winter than in Ukraine, where the average temperature in winter is from -3^0C to -15^0C [5].

Components contained in oils, as a rule, make up from 5% to 15% of the mass of road bitumen, depending on its brand. They contain mainly crystalline linear alkanes or n-paraffins, usually in amounts from 0% to 15% mas. The ratio of hydrogen: carbon in these components is close to 2, except that only traces of heteroatoms are present. The average molecular weight of the saturated components is approximately 600 g/mol. They mainly have an aliphatic structure [4].

The effect of paraffin-naphthenic hydrocarbons on the properties of bitumen differs from the effect of aromatic hydrocarbons, resins and asphaltenes. An increase in the concentration of paraffinic-naphthenic hydrocarbons in the tar due to a decrease in aromatic hydrocarbons, resins and asphaltenes contributes to an increase in penetration, but leads to a decrease in elongation and brittleness temperature. The heightened content of paraffinic-naphthenic hydrocarbons, owing to their challenging oxidation, necessitates increased air consumption and oxidation duration while also exerting a negative impact on the structural homogeneity of bitumen and its adhesion to mineral aggregates. However, paraffin-naphthenic compounds, being thinners and plasticizers, improve the viscosity-temperature properties of bitumen.

The brittleness and plasticity of bitumen depend on the content of asphaltenes and resin, therefore, increasing their content of asphaltenes and resin increases the hardness (elasticity) of bitumen, simultaneously increasing the melting point. On the other hand, the oil is partly a solvent for the resin and allows the bitumen to be more elastic. Lowering the molecular weight of oil and resin also increases the plasticity of bitumen [6].

Evdokimova et al. [7] proposed a technology for producing petroleum road bitumen by compounding deeply oxidized bitumen with tar, pre-oxidized to various temperatures, in order to produce binders stable to the processes of thermo-oxidative aging. It was revealed that the use of oxidized tar with a softening point of 360^0C during compounding.

In Europe and North America, the leading manufacturers of road binders obtain them using compounding technology. There is a variety of bitumen compounding methods. Some methods are based at special preparation of the raw material, regulation of its hydrophobicity, acid resistance to obtain the desired product. The others focus on the use of modifiers such as elastomers and other types of polymers. Thus, the organization of technological stages of preparation of raw materials and/or production of the final product, for example, by mixing deep-vacuum distillation residues and deasphaltizate with low-viscosity residues and fractions of oil with low viscosity in defined ratios and under the conditions of distillation and deasphalting. All these methods of compounding include the use of concentrates of polycycloaromatic compounds as one of the most promising components from the point of view of quality and economy [8].

Compounding is considered the most effective method of obtaining and ensuring necessary physicochemical and operational properties of bitumen. In this sense, compounding is an effective way of preparing raw materials at the stage of industrial processing, as it allows regulating the content of tars in raw materials. In addition, the compounding of petroleum binders can occur at the preparation stage of raw materials or at the production stage of finished products [9].

In work [10], conventional road bitumen with penetration grade of 100–130 mm is compounded with tar to produce bitumen with improved low-temperature resistance. Similar studies were also conducted and described elsewhere [5,11,12].

By employing a combination of compounding and oxidation processes, it becomes feasible to attain bitumen with targeted properties.

Thus, it is necessary to develop new technology for obtaining road bitumen from paraffinic and highly paraffinic raw materials, and this requires the search for innovative solutions in the preparation of raw materials and the production of finished products. It is important to note that one of the main tasks in the development of such technology is the minimal modification (changes) of existing technological schemes and the absence of the need for specific equipment for the production of bitumen [11].

The purpose of this research is to produce high-quality bitumen by combining two processes, such as compounding and oxidation from tars, which differ significantly from each other in physical and chemical properties.

Experimental

The preheated tar mixture was loaded into a weighted empty reactor. The empty trap was weighed, the installation was assembled, and then the electric heating of the reactor was turned on. The intensity of heating the reactor was regulated using a voltage regulator.

The start time of the experiment was fixed while the temperature in the reactor was 5^0C lower than the specified one. After that, the air was supplied to it and the intensity of reactor heating was reduced. The air consumption was set by the rheometer with the help of a control valve. In the future, the required temperature in the reactor and the airflow were maintained with the help of a voltage regulator and a control valve.

After the end of the time allotted for oxidation, the air supply to the reactor was stopped, the installation was dismantled and the reactor with bitumen was weighed.

Therefore, the water condensate from the «black solar» was separated into a pre-weighed container. The catcher with «black solar» and the container with water condensate were weighed separately. According to the results of the research, the material balance of the process of obtaining oxidized bitumen was drawn up.

Thus, to prepare a mixture of paraffin tar and tar from Orkhiv oil, each tar was pre-weighed in a necessary amount. The weighted components were heated and mixed until homogeneous. Therefore, the resulting mixture was stowed in the reactor.

Oxidation of high molecular weight compounds was performed on a laboratory installation consisting of a reactor unit, an air supply system, a cooling unit and capture of volatile oxidation products.

The air supply system includes a compressor, a control valve, and a rheometer (for measuring the flow of air supplied to the reactor).

The reactor unit consists of a glass reactor, a furnace for maintaining the required temperature in the reactor, and a voltage regulator. The reactor is equipped with a capillary for supplying air to the reactor under the layer of raw materials, a pocket where a thermocouple is placed to control and regulate the temperature in the reactor, and a nozzle for the removal gaseous products. The thermocouple regulates temperature in the reactor.

The unit for cooling and catching volatile oxidation products consists of a water cooler for cooling and condensing volatile products and a trap in which water condensate and black solar are gathering. The parameters were determined using standard methods mentioned in the Ukrainian state standards DSTU 4044:2019, DSTU ISO 12185:2009, DSTU B V.2.7-81-98, DSTU B V.2.7-81-98, EN 1426:2018, DSTU 8825:2019, DSTU 9169:2021, and DSTU 8859:2019.

Results

The raw material for the production of oxidized petroleum bitumens was tar extracted from eastern Ukrainian paraffinic oils, selected at PJSC «UKRTATNAFTA» (Kremenchuk), the characteristics of which are given in Table 1 (Sample 1).

The residue from the distillation of Orkhiv oil (Table 1, Sample 2) was used as a component introduced into the raw material to improve the performance of the target product.

The oil bitumen produced by such a traditional oxidation method does not meet the requirements of current standards, and the aim of improving its quality is quite urgent. One of the ways to solve this problem is to deliberately influence operational quality indicators, which involves the oxidation of compound raw materials to obtain such bitumens. Therefore, to address the issue of producing oil bitumen with improved quality indicators (properties), the mixing method of paraffinic oil tar with the residue from the distillation of Orkhiv oil was utilized, followed by the oxidation of this mixture. For bitumen production, we added the residue from the distillation of Orkhiv oil was to the tar. To preliminarily estimations the properties of the obtained bitumens, various amounts of Orkhiv oil tar were introduced into the mixture. Table 2 shows the results of the investigations on the properties of the bitumens. The oxidation process occur at a temperature of 250^{0}C, with a volumetric air flow rate of 2.5 min^{-1}, and a process duration of 3 hours.

It was found that with an increase in the content of Orkhiv tar in the mixture, the softening point of the obtained oil bitumen increases to 39^{0}C, penetration decreases to 165×0.1 mm, and ductility increases to 87 cm, while adhesion to glass reaches 25%. Therefore, from the presented results, it can be observed that the mixture with a 50% content of Orkhiv tar exhibits the best ductility and glass adhesion properties, while other indicators remain within the standard range DSTU 4044:2019.

The possibility of quality control of bitumens was investigated based on the variation of process technological parameters of oxidation, namely temperature, air flow rate, and process duration.

However, the main influence factor remains the temperature. It is known that the transformations of high-molecular-weight components occur more intensively at high temperatures. At the same time, cracking and condensation reactions prevail, and the majority of oxygen is carried away with the outgoing process gases, indicating that the oxidation process is of a dehydrogenation nature.

At low temperatures of the process, the limiting reactions are those involving the oil fraction of the tar. The oxidation process shifts towards an increase in the content of oxygen-containing compounds. According to ref. [5], at 125^{0}C, the main condensed di- and polycyclic aromatic hydrocarbons undergo oxidation, and their quantity decreases by half compared to the initial raw material.

The number of dehydrogenation reactions increases at temperatures of 250^{0}C and above. The naphthogenic hydrocarbons active interact with oxygen in the air, causing an increase in the quantity of condensed bicyclic aromatic hydrocarbons. However, the amount of bound oxygen in the bitumen does not increase compared to its content in the initial raw material.

Study [12] demonstrated a significant improvement in the properties of road bitumens obtained at reduced oxidation temperatures. For

Table 1

Characteristics of raw materials

Parameter	Sample 1	Sample 2
softening point, (ring-and-ball method), ^{0}C	32	37
ductility at 25^{0}C, cm	35	>100
penetration at 25^{0}C, mm	29.0	10.6
density at 20^{0}C, kg/m^3	943	998
content of sulfur, wt.%	1.2	6.3

Table 2

Effect of the composition of the tar mixture on the properties of petroleum bitumens

Parameter	Value		
	content of Orkhivtsy tar in the mixture		
	10%	30%	50%
softening point (ring-and-ball method), ^{0}C	35	36	39
ductility at 25^{0}C, cm	50	63	87
penetration at 25^{0}C, mm	23.0	19.1	16.5
index of penetration	–1.91	–2.16	–1.30
brittleness temperature, ^{0}C	–45	–39	–34
plasticity interval, ^{0}C	80	75	73
adhesion to glass, %	14.5	23.2	25

instance, lowering the oxidation process temperature to 220⁰C leads to an increase in penetration depth, a reduction in low-temperature brittleness, and an expansion of the plasticity range of the oxidized bitumens. The properties of the bitumens after heating are enhanced, it can be attributing to the decrease in asphaltene content with an increase in the oil content in the bitumen.

Research of oxidation duration influence on petroleum bitumen properties was conducted in the range from 3 to 12 hours. The oxidation process of the bitumens was carried out at a temperature of 250⁰C and with an air flow rate of 2.5 min^{-1} in a laboratory setup as described above. The results of the investigations on the effect of oxidation duration on the properties of petroleum bitumens are presented in Table 3.

The research results indicate that with an increase in the oxidation duration from 3 to 12 hours, the softening point of the obtained bitumen increases from 39⁰C to 60⁰C, while the penetration of the bitumens decreases from 165×0.1 mm to 60×0.1 mm. It is worth noting that the ductility of the bitumen decreases significantly when the oxidation duration reaches 9 hours, reaching a value of 80 cm. However, a further increase in the oxidation duration (up to 12 hours) does not lead to any additional changes in ductility. This can be explained by the fact that during the initial stages of oxidation, the raw material components transform resins, which are characterized by increased ductility. Subsequent prolonged oxidation results in minor conversion of resins into asphaltenes, and therefore, the ductility remains unchanged.

Increasing the oxidation duration leads to an increase in the penetration index. This parameter increases from −1.30 (at an oxidation duration of 3 hours) to 1.507 (at an oxidation duration of 12 hours). The intensification of the oxidation process is confirmed by the rise in the brittle point temperature of the bitumen over time. However, the plasticity range slightly decreases with the increase in the oxidation duration, while the «glass adhesion» parameter improves indicating enhanced adhesive properties of the bitumen.

The next stage of the research involved determining the influence of the process temperature on the characteristics (indicators) of the obtained bitumens. The oxidation process was conducted for 3 hours within the temperature range of 210−270⁰C, with an air flow rate of 2.5 min^{-1}. The results of the study are presented in Table 4.

The research results indicate that with an increase in the oxidation process temperature, the softening point of the modified bitumens slightly rises from 33⁰C (at an oxidation temperature of 210⁰C) to 47⁰C (at an oxidation temperature of 270⁰C). At the same time, the ductility of the bitumens increases with an increase in the oxidation temperature (from 210⁰C to 250⁰C) from 61 cm to 87 cm. However, further increasing the process temperature leads to a decrease in ductility down to 80 cm. It can be concluded that such a pattern of ductility change with change the process temperature is attributed to the fact that the maximum amount of resins is formed at 250⁰C. Subsequently, as the temperature increases, the resin content decreases while the content of asphaltenes increases, resulting in a reduction in ductility.

As expected, the penetration of the bitumen decreases from 275×0.1 mm (at a process temperature of 210⁰C) to 154×0.1 mm (at 270⁰C). It is worth noting that the most significant reduction in penetration occurs at a temperature of 250⁰C. Since the penetration is 165×0.1 mm at 250⁰C and decreases to 154×0.1 mm at 270⁰C, the optimal oxidation temperature for the tar is 250⁰C. Further increasing the temperature slightly intensifies the oxidation process.

It has been observed that the «adhesion to glass» parameter increases slightly with an increase in the

Table 3

Effect of the duration of oxidation on the properties of petroleum bitumens

Parameter	Value			
	duration of oxidation			
	3 h	6 h	9 h	12 h
softening point (ring-and-ball method), ⁰C	39	45	54	60
ductility at 25⁰C, cm	87	83	80	80
penetration at 25⁰C, mm	16.5	11.8	8.4	6.0
index of penetration	–1.30	–0.25	1.16	1.51
plasticity interval, ⁰C	73	72	73	70
brittleness temperature, ⁰C	–34	–27	–19	–10
adhesion to glass, %	25	33	38	48

oxidation process temperature while the penetration index of the bitumen decreases, along with the temperature at which the bitumen becomes brittle.

The next step of the conducted research was to study the influence of air flow rate, which varied within the range of 2.0–3.0 min^{-1}. The oxidation process occurred for 3 hours at a temperature of 250^0C. The results of the conducted experiments are presented in Table 5.

The experimental findings reveal that increasing the airflow rate leads to higher softening points and ductility of bitumen as determined by the «Ring and Ball» method, while the penetration decreases. Moreover, the «adhesion to glass» property of bitumen improves with higher supplied airflow rates, enhancing their adherence to solid surfaces. However, this increase in airflow rate also results in a higher brittleness temperature of the bitumen, albeit with a slight reduction in the plasticity range.

It is evident from analysis of the data presented in Tables 3, 4 and 5 that by introducing paraffinic tar, residue from the distillation of Orkhiv oil, and modifying process conditions, bitumen with enhanced quality indicators can be obtained. A comparative analysis of bitumen samples obtained under different conditions was performed to assess their compliance with standards in the construction industry. Notably, samples obtained with an oxidation duration of 3 hours did not meet the specified standard values for the studied parameters. However, samples obtained at a process temperature of 250^0C and an oxidation duration of 6 and 9 hours demonstrated compliance with the DSTU 4044:2019 standard, based on the investigated parameters.

To establish the mechanism of the bitumen oxidation reaction under diverse oxidation process conditions (process duration, temperature, and airflow rate), we calculated rate constants of oxidation (K_0) by the following Lockwood's formula:

$$K_0 = \frac{1}{\tau} \ln \frac{t_{spo}}{t_{spr}}, \quad (1)$$

where τ is the duration of the process (s); t_{spo} is the softening temperature of bitumen during oxidation (^{0}C); and t_{spr} is the softening temperature of the initial

Table 4

Effect of oxidation temperature on the properties of petroleum bitumens

Parameter	Value				
	temperature of oxidation				
	210^0C	230^0C	250^0C	260^0C	270^0C
softening point (ring-and-ball method), ^{0}C	33	36	39	43	47
ductility at 25^0C, cm	61	65	87	84	80
penetration at 25^0C, mm	27.5	18.6	16.5	16.0	15.4
index of penetration	–2.30	–2.27	–1.30	0.19	1.42
plasticity interval, ^{0}C	74	78	79	77	81
brittleness temperature, ^{0}C	–41	–42	–40	–39	–38
adhesion to glass, %	15.4	21	25	28	31

Table 5

The influence of consumption air flow on the properties of oxidized petroleum bitumens

Parameter	Value		
	consumption air flow		
	2.0 min^{-1}	2.5 min^{-1}	3.0 min^{-1}
softening point (ring-and-ball method), ^{0}C	36	39	44
ductility at 25^0C, cm	67	87	90
penetration at 25^0C, mm	17.4	16.5	14.8
index of penetration	–2.52	–1.30	0.24
brittleness temperature, ^{0}C	–41	–40	–35
plasticity interval, ^{0}C	81	79	79
adhesion to glass, %	20	25	30

raw material (^{0}C).

The changes in the rate constant of oxidation process depending on the conditions of the experiment are shown in Figs. 1, 2, and 3.

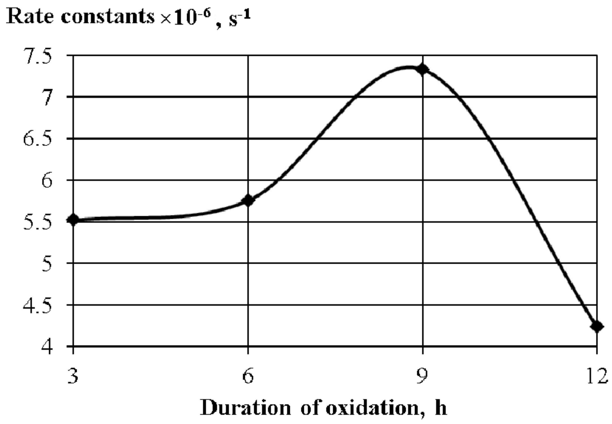

Fig. 1. Change in the rate constant with the duration of the process (T=250^{0}C, consumption air flow 2.5 min^{-1})

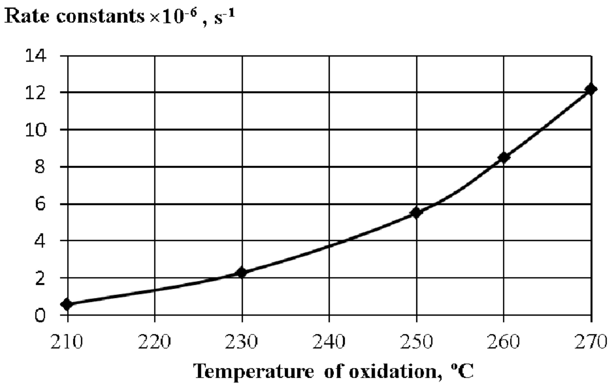

Fig. 2. Change in the rate constant with the temperature of the process (τ=3 h, consumption air flow 2.5 min^{-1})

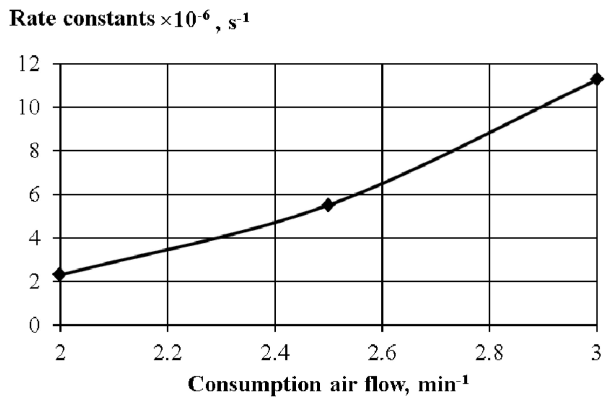

Fig. 3. Change in the rate constant with the air flow rate during the oxidation process (T=250^{0}C, τ=3 h)

For the multi-component system (Fig. 1), which is the tar, the change in the oxidation rate constant with process duration exhibits a complex nature. In the initial stage from 3 to 6 hours, the oxidation rate constants change insignificantly, indicating an induction period in the process. In the subsequent stage from 6 to 9 hours, there is a sharp increase in the oxidation rate constant, which is indicative of significant intensification of the process and, accordingly, considerable transformations in the chemical composition.

Furthermore, continuing the process for up to 12 hours is characterized by a sharp decrease in the oxidation rate constant. This indicates an increase in the viscosity of the reaction mixture and hindered access of oxygen from the air to the reactive hydrocarbons.

Figures 2 and 3 show a significant increase in the reaction rate constant, typical for the oxidation process, with increasing process temperature and increasing air flow rate, respectively.

Thus, the rate constants of the process (Fig. 1) demonstrate the reaction mechanism of bitumen oxidation, namely: the induction period (duration of 3–6 hours), intensive oxidation (duration of 6–9 hours) and inhibition of the process (duration of more than 9 hours). Thus, the optimal time for oxidation to obtain bitumen is 6–9 hours.

Discussion

The research outcomes reveal that as the oxidation duration increases from 3 to 12 hours, the softening temperature of the obtained bitumen rises from 39^{0}C to 60^{0}C, while the penetration of the bitumen reduces from 165×0.1 mm to 60×0.1 mm. Notably, the ductility of the bitumen reaches its maximum value (87 cm) at an oxidation duration of 3 hours. However, with a subsequent increase in the oxidation duration up to 12 hours, the ductility gradually diminishes to 80 cm. This phenomenon can be attributed to the initial stage of oxidation, wherein the conversion of oil components in the raw material yields resins that exhibit increased ductility. Nevertheless, with a further extension of the oxidation duration, the resins transform into asphaltenes, leading to a gradual reduction in ductility.

The experimental results demonstrate a positive correlation between the oxidation duration and the penetration index of bitumen. Specifically, the penetration index rises from −1.3 (at an oxidation duration of 3 hours) to 1.51 (at an oxidation duration of 12 hours). The observed increase in the brittleness temperature of the bitumen with longer oxidation durations confirms the intensification of the oxidation process. Additionally, the plasticity range experiences a slight reduction with increasing oxidation duration. However, a noteworthy improvement is observed in the «adhesion to glass» index, signifying enhanced adhesion properties of the bitumen as the oxidation duration increases.

The research results (Table 4) reveal that as the oxidation temperature increases, the softening temperature of the obtained bitumen shows a slight rise, transitioning from 33^{0}C (at an oxidation temperature of 210^{0}C) to 47^{0}C (at an oxidation temperature of 270^{0}C). Simultaneously, the ductility of the bitumen demonstrates an increasing trend from 61 cm to 87 cm with a rise in the oxidation temperature from 210^{0}C to 250^{0}C. However, further escalation of the oxidation temperature results in a minor decline in ductility, reaching 80 cm. This dependence of ductility on the oxidation temperature arises from the optimal equilibrium of paraffinic-naphthenic hydrocarbons, resins, and asphaltenes formed at 250^{0}C, which leads to the highest ductility. Consequently, at higher temperatures, the resin content diminishes while the amount of asphaltenes increases, contributing to a reduction in ductility.

The penetration of the bitumen displays an anticipated reduction from 275×0.1 mm (at a temperature of 210^{0}C) to 154×0.1 mm (at 270^{0}C). Notably, the most substantial decrease in penetration occurs at a temperature of 210^{0}C. Specifically, the penetration at 210^{0}C is 275×0.1 mm, while at 250^{0}C, it decreases only to 165×0.1 mm. Consequently, the optimal temperature for the tar oxidation process is 250^{0}C, as further elevating the temperature marginally intensifies the oxidation process. Furthermore, it has been observed that the adhesion to glass (indicated by the «glass adhesion» parameter) exhibits a slight increase with higher oxidation temperatures. In contrast, the penetration index of the bitumens decreases with rising oxidation temperatures, as does the temperature at which the bitumen reaches its brittle state.

The experimental findings (Table 5) demonstrate that as the air flow rate increases, the softening temperature of the bitumen, as determined by the «ring and ball» method, along with its ductility, also increase, while the penetration decreases. Moreover, the «adhesion to glass» property of the bitumen improves with higher supplied air flow rates, enhancing their adhesion to solid surfaces. However, this increase in the air flow rate also results in a higher brittleness temperature of the bitumen, albeit with a slight reduction in the plasticity range.

Quality indicators of the obtained bitumen resulting from the conducted research were determined and are presented in Table 5. It has been observed that the softening temperature of the obtained petroleum bitumen increases from 36^{0}C to 47^{0}C, while the penetration decreases from 174×0.1 mm to 148×0.1 mm, with the process duration being a significant influencing factor.

By comparing the results presented in Table 3, Table 4 and Table 5, we can observe that adding paraffinic tar and the residue from the distillation of Orkhovitska crude oil results in bitumen with improved quality indicators. Specifically, the ductility increases from 87 to 90 (at 25^{0}C), the penetration of the bitumen decreases from 148×0.1 mm (at 25^{0}C) to 60×0.1 mm (at 25^{0}C), and softening temperature decreases from 47^{0}C to 44^{0}C.

Conclusions

As a result of the conducted research, it was established the feasibility of producing petroleum bitumen through the oxidation method using paraffinic tar extracted from East Ukrainian paraffinic crude oils.

Besides, the influence of technological factors (temperature, oxidation duration, and air flow rate) on the performance properties of oxidized petroleum bitumen was studied.

Samples of the obtained oxidized petroleum bitumen were analyzed to determine their ductility (at 25^{0}C), penetration (at 25^{0}C), and softening point using the ring and ball method. Based on the analysis results, it was established that it is fundamentally possible to obtain bitumens with improved performance properties by using the residue of paraffinic crude oils with the addition of the residue from the distillation of Orkhiv oil as a feedstock. In conclusion, we have confirmed that by using paraffinic tar and adjusting the oxidation conditions, petroleum bitumens with different predetermined properties can be prepared.

The use of compounding processes involving various types of tars and the oxidation of their mixtures, besides producing bitumens with desired characteristics, partially addresses the issue of comprehensive raw material utilization.

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МОЖЛИВІСТЬ ОДЕРЖАННЯ БІТУМУ З РІЗНИХ ВИДІВ ГУДРОНІВ

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Традиційно було відомо, що присутність n-парафіну в гудроні негативно впливає на крихкість, адгезію та внутрішню когезію частинок бітуму, що робить непрактичним використання парафінової смоли для виробництва високоякісного бітуму. У цьому дослідженні розглядається потенційне покращення властивостей бітуму шляхом змішування залишку від дистиляції орхівської нафти з гудроном. Зразки бітуму одержували шляхом окислення компаундованої суміші в інтервалі температур 210–270^{0}С. У дослідженні вивчали вплив тривалості окислення (від 3 до 12 годин), температури та витрати окислювача на кінцеві властивості бітуму. Окрім того, встановлювали вплив співвідношення суміші парафінової смоли та залишку від перегонки орхівської нафти на характеристики бітуму. Були визначені важливі властивості бітуму, включаючи температуру розм'якшення, пластичність, пенетрацію, індекс пенетрації, інтервал пластичності, температуру крихкості та адгезію до скла.

Ключові слова: гудрон, бітум, окиснений бітум, компаундування, окиснення, константи швидкості.

FEASIBILITY OF PRODUCING BITUMEN FROM DIFFERENT TYPES OF TAR

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Traditionally, the presence of n-paraffins in tar is known to negatively affect the fragility, adhesion, and internal cohesion of bitumen particles, making paraffinic tar impractical for producing high-quality bitumen. This study investigates the potential enhancement of bitumen properties through the compounding of residue from the distillation of Orkhiv oil with tar. Bitumen samples were prepared via oxidation of the compounded mixture within a temperature range of 210–270^{0}C. The research explores the effects of oxidation duration (ranging from 3 to 12 hours), temperature, and oxidizer consumption on the resulting bitumen properties. Furthermore, the study examines the impact of the mixture ratio of paraffinic tar and residue from Orkhiv oil distillation on bitumen characteristics. Key bitumen properties, including softening point, ductility, penetration, penetration index, plasticity interval, brittleness temperature, and adhesion to glass, were systematically determined. Optimal process conditions were identified, specifically at a temperature of 250^{0}C and an airflow rate of 2.5 min^{-1}. The kinetic parameters of the oxidation process for the tar mixture were studied, leading to the calculation of the reaction rate constant under various influencing factors. The results demonstrate the feasibility of producing bitumen suitable for asphalt concrete coatings with improved performance characteristics.

Keywords: tar; bitumen; oxidized bitumen; compounding; oxidation; reaction rate constants.

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