

Minimization of the Negative Environmental Impact of Oil Sludge by Using it in the Production of Bitumen

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Abstract – In this paper, the prevention of negative technogenic impact on the environment of oil sludge by using it as a secondary resource is considered. Oil sludge from various objects of oil fields in Kazakhstan (Mangystau region) has been studied. The possibilities of using oil (after its separation from oil sludge by bioremediation) as a partial substitute for bitumen base in the production of modified bitumen are considered. The main physical and mechanical characteristics of modified bitumen are determined. The results confirm that the modified bitumen prepared with oil sludge and oil separated by bioremediation method meets the requirements for polymer-bitumen binder to Kazakhstan standards and is suitable for the production of modified bitumen in its physico-chemical characteristics.

Keywords – Bioremediation; bitumen; microorganisms; modification; oil sludge; processing; utilization

1. INTRODUCTION

Oil sludge is formed daily at oil refineries, it is estimated that sludge accounts for 30 to 40 % of investment costs and 50 % of operating costs of oil refineries [1]. As a rule, they contain organic and inorganic substances that differ from other refined products, and they are influenced by factors such as the composition of crude oil, the size and age of the refinery [2].

The inorganic part of oil sludge is formed due to the ingress of soil and minerals into them, and includes quartz (52 %), calcite (15 %), halite (16 %), feldspar - albite (7 %), adular (5 %), hematite (5 %). The organic part of the oil sludge is represented by aromatic hydrocarbons and paraffins.

In terms of toxicity, oil sludge is classified as industrial waste of the 3rd hazard class. Oil sludge is one of the sources of toxic heavy metals, such as Fe, Co, V, Ni, Cu, Cr, Mn.

The characteristics of sludge in oil reservoirs vary greatly [3]. About 500 tons of oil sludge are produced annually. According to research, an average of 30 000 tons of oil sludge is formed at each oil refinery in the United States per year [4]. The total amount of oil sludge produced in China is 3 million tons annually [5]. According to the results of research, 450–500 thousand tons of oil sludge are formed at oil refineries in Kazakhstan and CIS countries per year, and their total volume is 7.6 million tons [6]. Additives such as suspending agents,

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dispersing agents and thickeners accumulate in oil sludge, which pose a serious problem for the environment. The use of oil sludge in useful materials, such as organic compounds with low molecular weight and carbon residues, helps to solve the problem of its utilization, as well as its use in cement and asphalt production [7]. Extracting oil from oil sludge is of great importance, and finding ways to avoid the waste of this valuable material has long been on the agenda of universities and research centres.

According to the results of physico-chemical investigations of regional oil sludge, we found that they contain a significant amount of oil products, which are mainly heavy fractions of oil [8]. In addition, it was found that the composition and properties of the organic part of the oil sludge are similar to heavy residues with a high content of resins and asphaltenes, high density [9], [10].

Oil sludge processing methods can be divided into non-destructive and destructive. Non-destructive ones include: controlled open unloading; burial; the use of oily sludge; application of sludge as an organic fertilizer. Destructive ones include: incineration on site or together with household waste; inclusion in cement during its production by wet process; aerobic processing [11].

Another classification of processing methods depending on the initial properties of oil sludge is more complete [12]–[17]. According to this classification, the methods are divided into:

- thermal – burning in open barns, furnaces of various types; physical – mixing and physical separation of oil sludge;
- physico-chemical – the use of specially selected reagents;
- biological – microbiological decomposition of petroleum products directly in storage sites.

One of the most common processes of processing oil-containing waste abroad is the gasification of sludge used in a mixture with other waste. This process is applicable for the processing of liquid, solid and pasty waste [18].

It should be noted that the gasification process can be used for the disposal of waste oil products, sewage treatment plants [18]. The general disadvantages of the thermal method of waste processing are considered to be incomplete combustion of oil products, atmospheric air pollution by combustion products, rather large costs for neutralization and purification of flue gases.

The trend of recent decades is the separate processing of oil sludge in direct dependence on the method of their formation. At the moment, this approach to the current problem quite successfully solves environmental problems and the rational use of oil sludge.

The use of oil sludge as material resources of secondary raw materials is widespread. It is mainly used in road construction as additives to binders that improve the quality of asphalt concrete mixtures by increasing the strength characteristics of the roadbed and reducing the water-absorbing properties of oil sludge due to the significant content of oil products in it can be attributed to secondary material resources. Its use as a raw material is widespread, since a certain environmental and economic effect is achieved.

The authors [19] investigated the process of mixing oil sludge with bitumen for the production of asphalt. According to the research results, it was found that due to a decrease in the stability of bitumen molecules, the ignition temperature decreases, the bitumen resistance decreases. In addition, SBS polymer was used for modification, which gave a positive effect and increased the durability of the samples. The results showed that the durability of asphalt increases with the use of 25–50 % sulphur and 2–7 % styrene butadiene polymer. Therefore, Marshall asphalt samples with a sulphur content of 30 % were prepared using bitumen and oil sludge. The best results were obtained by 50 % of oil sludge in bitumen

and a mixture of 2 % polymers and 15 % of oil sludge with bitumen. The results showed that asphalt resistance increases with the use of 25–50 % sulphur and 2–7 % styrene-butadiene polymer. Therefore, Marshall asphalt samples with a sulphur content of 30 % were prepared from bitumen and oil sludge. 50 % oil sludge in bitumen and a mixture of 2 % polymer and 15 % oil sludge with bitumen gave the best results (30 % sulphur + 50 % oil sludge in bitumen 20 % and a mixture (2 % polymer + 15 % oil sludge in bitumen 83 %). These mixtures are economically advantageous.

The article [20] describes in detail the complex oil-containing waste treatment by applying solar energy. The developed Helio devices are equipped with concentrating elements to extract oil in the purification of dump oil. The questions of practical application of pre-treated oil-contaminated soils and oil sludge to strengthen road surfaces are considered. The results of the experimental study on the production of soil concrete is based on oily dump. The structures of the soil concrete made based on oil-contaminated soils and oil sludge are investigated and relevant proposals are made. The method developed by the authors can be considered as one of the methods for the disposal of oil waste in order to reduce the harmful effects of oil-containing waste on the environment.

Significant oil losses in oil-containing wastes and their adverse impact on the region environmental setting bring about the need to develop an oil-containing wastes treatment technology. To tackle this issue, the authors have set an aim of designing a helio device and creating an oil-containing wastes treatment method based on it to extract oil products. Considering a widespread in the composition and properties of potential oil sludge raw materials and their tendency for either formation of stable emulsions or phase separation, we have conducted in-depth modern physical and chemical studies and defined the need to develop a commercial oil-containing wastes purification method. The authors designed the device, in which oil product hydrocarbons undergo thermal treatment using solar energy. Following oil-containing wastes purification using solar energy, the particulate load in soil does not exceed 6.65–6.79 % and the absolute molecular weight of hydrocarbons approaches that of bitumen. The developed oil-containing wastes purification method solves an important environmental issue of oil-containing wastes recycling, promotes recovery, and prevents degradation of natural complexes, and reduces soil and water pollution [21].

The use of oil fly ash after the recovery of heavy valuable metals was investigated [22]. More specifically, its use, as an adsorbent of dyes from industrial wastewater, was evaluated. Methylene blue was used as a model compound to study the adsorption capacity of the proposed carbonaceous residue from metal recovery treatments. The effects of contact time, initial dye concentration, and adsorbent dose were investigated. The maximum amount of dye was adsorbed after one hour. Moreover, 1–3 g of residues were necessary for the removal of 200–1000 mg dm⁻³ from 0.050 dm³ of contacted solution. The Langmuir isotherm model was in good agreement with the adsorption equilibrium data, indicating a maximum monolayer saturation capacity of approximately 40 mg/g at 25 °C. High abatement efficiencies (up to 99 %) were obtained, and the adsorbed dye was released almost immediately by re-contacting with water. The adsorption capacity was at least four times lower than that of commercially available active carbon. The double treatment of oil fly ash with deionised water and hydrochloric acid allows for the extraction of over 85 % of the vanadium, iron, and nickel content in the ash. However, the negligible or zero cost of solid residues, otherwise disposed in landfills, indicates their potential as a valid alternative. The use of oil fly ash for both recovery of heavy valuable metals and the subsequent removal of dyes from wastewater suggest a zero-waste process.

The oil containing drilling waste is a worldwide environmental problem associated with oil and gas exploration. In Poland, the problem of the drilling waste has become important since

starting of shale gas exploration. The results of thermal treatment of drilling waste from shale gas exploration are presented [23]. It has been shown that organic content vaporized completely at temperature up to 500 °C. The main problem is high content of chloride, sulphate, sodium, potassium, magnesium in the waste and its water leachate. Toxicity tests confirmed that high salinity of the samples pose important risk for environment. Due to the high content of barium, the drilling waste may be utilized in production of cement with high chemical and heat resistance and opaque to X-ray. Thermal treatment process is a viable option for remediation of the drilling waste.

A prospective direction in the treatment of soils from oil is the immobilization of microorganisms on various carriers (peat, vermiculite, etc.). It is known that in most cases a neutral environment with a pH between 6 and 9 is ideal for biodegradation [24]. So, for our saline (weakly alkaline) soils, the introduction of a carrier in the form of limestone-shell screening improves the contact of oil-contaminated soil with microorganisms [25]. The bacterial cells of the drug immobilized on the carrier are not exposed to weathering and remain viable in hot climates.

In [25], a method of bioremediation of oil-contaminated soils with a high paraffin content using the drug 'SHER' with immobilization of local wastes of limestone-shell rock is proposed.

In the studies of Huihui W. *et al.* [26], it was found that during aging of oil sludge, the physico-chemical properties of the resulting soil and its composition change significantly.

A comparative assessment of the destructive activity of Bioden and Ecobak bioregulators developed in Kazakhstan using PetroTreat (Russia) showed that in the process of cleaning soils contaminated with diesel fuel, fuel oil, spent engine oil, PetroTreat efficiency is higher than Ecobak and Bioden. But, at the same time, it was found that the Ecobak turned out to be more effective when cleaning oil-contaminated sandy soils of the Akmolá and Mangystau regions [27].

Microorganisms are potent contributors to maintaining a safe environment, as they are able to degrade organic toxicants. For environmental applications, mostly bacteria are used while fungal strains have received less attention. However, they are able to degrade highly persistent organic contaminants and survive extreme conditions, and may thus be promising organisms. To find new fungal candidates for these applications, twelve soil samples from polycyclic aromatic hydrocarbon (PAH) contaminated sites in Austria were used to isolate fungal strains. A microplate screening method using PAH contaminated soil as inoculant was set up to isolate fungal strains being able to live in presence of toluene, hexadecane, or polychlorinated biphenyl 126. Not many microbial strains are known that degrade these three contaminants, while the PAH contamination acted as selective pressure for the soil microbiota. After obtaining pure cultures, the fungal strains were further screened for their ability to live in the presence of one of the three contaminant substrates. The authors presented the microtiter plate screening method is a cost efficient and quick approach to identify fungal strains for pollutant degradation and results in candidates with a high relevance for bioremediation techniques [28].

Study of biodegradability of organic constituents of the oil/water emulsion obtained from a metal-processing waste stream was carried out in paper [29]. The conducted study showed that oil/water emulsion is non-toxic to bacterial consortia, therefore it was colonized with indigenous microorganisms. Emulsion was treated using a threefold wastewater dilution and employing two variants of bio stimulated aerobic bacterial communities: 1) Bioremediation of uninoculated emulsion by the autochthonous bacteria alone; 2) The wastewater samples inoculated with a ZB-01 microbial consortium which is a source of specialized bacteria for process bioaugmentation. Determination of biodegradation efficiency achieved in a 14-day

test was carried out using two parameters: total load of organic content determined as a chemical oxygen demand (COD) and concentration of high-boiling organic compounds. Both approaches showed significant reduction of the emulsion organic fraction; however, bio augmented with inoculated consortium demonstrated higher biodegradation efficiency. High degradation yields (58 % and 71 %, respectively) obtained for both cases in a 28-day test was confirmed by means of gas chromatography analyses coupled with mass-spectrometric detection (GC-MS). The study demonstrated that oil-based metalworking emulsions can be efficiently treated by biological method under conditions enabling aerobic bacterial proliferation.

We have previously conducted a study of the use of oil sludge in order to obtain modified bitumen with the addition of polymers. The content of oil sludge in polymer-modified bitumen in the range of 15–20 % is considered economically feasible. Since in the tests of polymer-modified bitumen, the optimal content of oil sludge is determined in the range of 20–25 %, their use can be considered economically justified [30], [31].

The purpose of this study is to investigate the effect of oil additives separated from oil sludge by bioremediation on the quality of raw materials in the production of modified bitumen.

The novelty of the research lies in the fact that oil separated from oil sludge by bioremediation is used to obtain road bitumen.

2. MATERIALS AND METHODS

In the work, oil sludge (OS) of the Uzen and Zhetibay deposits of the Mangistau region (Kazakhstan) were used as objects of research. The physico-chemical characteristics of the OS are determined.

To study the physico-chemical characteristics of oil sludge, the following physico-chemical analysis methods were used: to determine the content of mechanical impurities in oil, the method for determining mechanical impurities (GOST 6370) was used, the essence of which is filtering the tested products with preliminary dissolution of slowly filtered products in gasoline or toluene, washing the sediment on the filter with a solvent, followed by drying and by weighing [32]. The water content in oil is made according to GOST 2477 ‘Oil and petroleum products. Method for determining the water content’. The essence of the method consists in heating an oil sample followed by steam condensation and measuring the volume of condensed water entering the trap receiver (hereinafter referred to as the trap) [33]. The content of the organic part (hydrocarbons), mass. % of them: paraffins, asphalt-resinous substances, wt. % and other hydrocarbons are made according to GOST 11851 [34].

According to the results of Table 1, the OS from the Uzen deposits mainly consists of the organic part (hydrocarbons) – 91.49 %, consisting of 16.0 % wt. of paraffins and 35.7 % wt. of asphalt-resinous substances, other hydrocarbons 39.7 % wt., the content of mechanical impurities 0.6 % wt., water is absent. It was also found that mechanical impurity of 37.8 % and water 30.8 % predominate in the composition of the OS from the Zhetibay deposit, the oil content is 31.4 % wt., of which 22.5 % wt. of paraffins and 8.9 % wt. of asphalt-resinous substances.

For further study, samples of OS Uzen were selected because it is dominated by hydrocarbons.

Further, experiments on bioremediation of oil sludge with the biological preparation ‘SHER’ with the immobilization of bacteria with the waste of local limestone-shell rock were carried out for research [24].

The bioremediation experiment was carried out in laboratory conditions using a technology consisting in mixing oil sludge with a biological product and a carrier.

TABLE 1. PHYSICO-CHEMICAL CHARACTERISTICS OF OS

Name of indicator	OS deposits		Test method
	Uzen	Zhetibay	
Water content, wt. %	0.0	30.8	GOST 2477
The content of the organic part (hydrocarbons), wt. %			
of them:			
- paraffins, wt. %	91.49	31.4	GOST 11851
- asphalt-resinous substances, wt. %	16.0	22.5	
- other hydrocarbons, wt. %	35.7	8.9	
	39.7	–	
The content of mechanical impurities, % wt.	8.51	37.8	GOST 6370

During the experiment, constant conditions were maintained in the laboratory: the air temperature was within 20 ± 2 °C, the humidity in the room was 60–70 %.

The results of the experiment on remediation of soils contaminated with paraffin oil using a biological preparation at room temperature showed that at 21 °C, the destruction of oil in 3 days was 88.63 % [25], therefore, the separation of the oil-water emulsion was carried out after 24 hours of settling.

Oil degradation was assessed visually by changing the oil stain and the accumulation of biomass. Calcite CaCO_3 in limestone contributes to a more intense release of the emulsion. At the same time, an oil film on the surface of the medium was not observed. That is, the oil mostly turned into a homogeneous emulsion. At the same time, fine particles precipitated out (no more than 6–9 %).

A laboratory study was also carried out to determine the physico-chemical properties of the emulsion isolated using a separation funnel. The results of the analysis are shown in Table 2.

The emulsion mainly consists of 75.8 % petroleum products [35], water content – 11.5 % [36], sediment content – 12.7 % wt. [35].

TABLE 2. COMPOSITION OF OIL-WATER EMULSION

Name of indicator	Unit of measurement	Sample 1	Sample 2	Sample 3	Test method
Water content	% wt.	11.5	8.4	9.2	GOST 31734-2012
Oil product content	% wt.	75.8	76.6	76.1	GOST 26449.1-85
Precipitation content	% wt.	12.7	15.0	14.7	GOST 31734-2012

Separation of oil from the oil-water emulsion is carried out by the settling method.

Further, the separated oil was added during the preparation of modified bitumen to study its effect on the process.

In the study, bitumen grade ROB 100/130 produced by JV ‘CASPI BITUMEN’ LLP was used for the preparation of modified bitumen. The characteristics of road oil bitumen grade ROB 100/130 are given in Table 3.

To prepare modified bitumen, the ratio of bitumen: oil from OS: modifier (SBS, latex) is experimentally selected. To do this, oil from OS is added to the ROB 100/130 bitumen

mixture in various ratios (OS is preliminarily processed by bioremediation to separate the mechanical part from oil) and polymer.

TABLE 3. CHARACTERISTICS OF ROAD OIL BITUMEN GRADE ROB 100/130

Name of indicator	Actual value	Test method
Ring and ball softening temperature, °C	44.00	ST RK 1227
Penetration at a temperature of 25 °C, 0.1 mm	118.00	ST RK 1226
Penetration at a temperature of 0 °C, 0.1 mm	31.00	
Extensibility at a temperature of 25 °C, cm	>150	ST RK 1374
Fraas brittleness temperature, °C	-22	ST RK 1229
Flash point, °C	280.0	ST RK 1804
Penetration index	-0.6	

The method of experiment and installation is similar to that presented in [31], [37].

The weight of bitumen averaged 200 g. For the analysis, the sample preparation was carried out in the following order. Before carrying out the modification process, the bitumen sample was melted to a mobile state (at a temperature not exceeding 105 °C) and oil was slowly added. Then the polymer was added to bitumen and oil, the mixture was heated with constant stirring, to a homogeneous state. The temperature was maintained in the range of 165–170 °C, the materials were mixed from 60 minutes to 180 minutes, depending on the type of modifier.

As a result of a series of experiments, samples of modified bitumen were obtained from bitumen grade ROB 100/130 and oil from oil-contaminated soil (OCS). Finished highly elastic polymer SBS-01-10 (styrene-butadiene-styrene) and latex of the finished emulsion were used as a modifier. Preparation of modified bitumen from conventional bitumen becomes modified as a result of the addition of artificial polymers to it, improving its properties.

Latex is introduced gradually and evenly as a modifier. The rate of introduction of the modifier depends on how intensively the water will evaporate. At a higher input rate, the water contained in latexes can form a foam layer on the bitumen surface. According to the results of previous studies [38], the dispersion temperature was chosen – 170 °C for 180 minutes (when modifying bitumen for less than 180 minutes, a high-molecular compound does not completely dissolve) and 60 minutes when adding polymer latex.

In order to verify the compliance of the modified bitumen obtained with the requirements of regulatory documentation, a comprehensive analysis of the obtained samples of modified bitumen with the use of oil from OCS and modifier, considering different mixing times, was carried out. For all samples of the modified bitumen obtained, the softening temperature and the penetration depth of the needle were determined.

To establish the conformity of the obtained samples of modified bitumen, the following basic physical and mechanical characteristics were determined: softening temperature, needle penetration depth (penetration), extensibility (ductility) and Fraas brittleness temperature. The softening temperature is determined by the ‘Ring and ball’ method according to ST RK 1227. The penetration is determined by a penetrometer according to ST RK 1226. Extensibility also indirectly characterizes the adhesion of bitumen and is related to the nature of its components. Extensibility was determined by a ductilometer TSKB-974N according to ST RK 1374. The Fraas brittleness temperature was determined on an apparatus for determining the brittleness temperature of bitumen ATX-04.

3. RESULTS AND DISCUSSION

Before we studied the method of utilization of oil sludge with the production of polymer-modified bitumen, the optimal parameters and ratios of ROB100/130:OS:latex = 80:20:5 were established [31].

This article presents the results of a research on the effect of an oil additive, obtained by separation of oil sludge by bioremediation on the quality of raw materials for the production of modified bitumen.

Table 4 shows the physico-mechanical characteristics of the obtained samples of modified bitumen with an SBS modifier and oil extracted from oil sludge.

TABLE 4. PHYSICAL AND MECHANICAL CHARACTERISTICS OF MODIFIED BITUMEN WITH SBS MODIFIER

Name of indicator	Preparation of modified bitumen		
	Ratios	Softening point ring and ball, °C	Penetration at a temperature of 25 °C, 0.1 mm
ROB 100/130:OS:SBS	60:40:5	59.00	40.00
	65:35:5	46.00	96.00
	70:30:5	58.00	100.00
	75:25:5	63.00	86.00
	80:20:5	71.0	87.0
ROB 100/130:O:SBS	60:40:5	44.00	69.00
	65:35:5	51.00	97.00
	70:30:5	64.00	69.00
	75:25:5	63.00	87.00
	80:20:5	65.00	91.00

Note: ROB — road oil bitumen; OS — oil sludge; O (oil separated by a bioregulator from oil sludge), SBS — styrene—butadiene—styrene.

The ratio of bitumen:sludge:modifier and bitumen:oil:modifier was chosen depending on the type of modifier and characteristics of bitumen, as well as previously studied works [25].

One of the most commonly used polymers in bitumen modification and approved for use by Kazakhstan standards is styrene-butadiene-styrene. Polymer SBS, as the best additive to bitumen, was used to prepare samples of modified bitumen in a ratio of 100:5. By changing the percentage ratio (bitumen:oil sludge as a control sample) ‘bitumen: oil’, the optimal ratio of bitumen to oil was selected at a constant value of the modifier. Ring and ball softening point and needle penetration depth tests were carried out on all samples with oil.

In samples of modified bitumen, with a decrease in the oil sludge content, the penetration value increases, but then decreases (Table 4). Polymers adsorb bitumen oils and form a separate dispersed phase, which leads to a decrease in the oil/asphaltene ratio, resulting in an increase in viscosity and an increase in the hardness of the binder [39].

Sulphur and its compounds are used to stabilize the polymer-bitumen dispersion. Reactions occur between the polymer and sulphur, as a result of which new chemical compounds arise, they remain evenly distributed in bitumen due to their lattice structure. Sulphur vulcanization is a chemical process that is widely used in the production of technical rubbers, it makes it possible to improve the stability during storage of some PBBs (for example, modified bitumen SBS) [40]–[45].

It is believed that sulphur works in two directions: chemically crosslinking polymer molecules and chemically binding polymer and bitumen through sulphide and or polysulfide bonds [19]. These chemical interactions are much stronger than physical ones (for example, aggregation of PS blocks into copolymers), and they do not disappear even at fairly high temperatures, which is believed to be very useful for improving stability during storage of PBBs. The crosslinking of polymer molecules leads to the formation of a stable polymer mesh in bitumen; while the connection between polymer and bitumen directly reduces the possibility of separation. In addition to increased stability during storage, some researchers [46]–[49] argued that vulcanization of sulphur can also improve elasticity, deformation resistance and some rheological properties of PBB.

In connection with the above, the effect of changes in the indicators of modified bitumen on the percentage of stabilizer in the initial mixture was studied.

The test results showed that from the obtained samples of polymer-modified bitumen (PMB) with oil sludge from the Uzen deposits at a ratio of 75:25:5 (ROB100/130:OS:SBS) corresponds to the actual parameters of polymer-modified bitumen (PMB) PMB 70/100, such as penetration at a temperature of 25 °C, 0.1 mm is 86 (test method ST RK 1226), the softening point ring and ball is 63 °C (test method ST RK 1227), and with oil (extracted from oil sludge by bioremediation) at a ratio of 70:30:5 (ROB100/130:O:SBS) corresponds to the actual indicators of polymer-modified bitumen PMB 50/70, such as penetration at a temperature of 25 °C, 0.1 mm is 69, the softening point ring and ball is 69 °C (Table 5) and meets the requirements of ST RK 2534-2014.

The optimum bitumen:oil ratio is chosen as 70:30, 75:25 and 80:20. Increasing the ratio of oil added by 30 % or more did not lead to positive results, as the results of the analysis do not meet the requirements of ST RK 2534-2014. And also, according to the research results, it is clear that the addition of oil extracted from oil sludge by bioremediation corresponds to the results of the use of oil sludge. This leads to the conclusion that there is no need to extract oil from oil sludge, which requires additional costs of biological products.

Studies were also conducted on the selection of the optimal bitumen ratio: oil sludge:latex and bitumen:oil:latex (Table 5). The ratio of latex was selected according to the test results.

From the analysis of the research results, it can be seen that a decrease in the ratio of the latex polymer leads to a significant improvement in most indicators, i.e. the less latex, the better the results. In particular, penetration and extensibility are improved at 25 °C – in the range from 35 to more than 150 cm. The softening point ring and ball varies from 39.0 to 71.00 °C. The brittleness temperature is within the normal range.

According to the physico-mechanical characteristics, we conclude that for polymer-modified bitumen with oil sludge content up to 25 % and polymer 1–5 %, therefore, with oil up to 30 % and polymer 1–5 %, the most optimal ratio of all indicators is achieved.

TABLE 5. SELECTION OF THE OPTIMAL RATIO WITH THE LATEX MODIFIER

Name of indicator	Unit of measurement	Preparation of modified bitumen			Test method
		ROB100/130:OS:latex			
Percentage ratio		80:20:5	80:20:3	80:20:1	
Softening point ring and ball, not lower:	°C	71.00	40.0	47.2	ST RK 1227

Penetration at a temperature of 25 °C, not lower:	0.1 mm.	68.00	106.3	102.6	ST RK 1226
Extensibility, not less than: at 25 °C	cm.	142.0	–150	35.5	ST RK 1374
Fraas brittleness temperature, not higher:	°C	–23.7	–23.7	–18.5	ST RK 1229
ROB 100/130:O:latex					
Percentage ratio		80:20:5	80:20:3	80:20:1	ST RK 1227
Softening point ring and ball, not lower:	°C	61.00	48.0	39.0	ST RK 1226
Penetration at a temperature of 25 °C, not lower:	0.1 mm	87.00	101.3	126.0	ST RK 1374
Extensibility, not less than: at 25 °C	cm	>150	>150	42.0	ST RK 1229
Fraas brittleness temperature, not higher:	°C	–22.0	–20.0	–18.0	ST RK 1229

4. CONCLUSIONS

Microbiological processing of oil sludge is aimed at the maximum extraction of oil from oil sludge. Oil extraction allows to significantly reduce the volume of oil sludge waste for the disposal of solid oil sludge residues. Due to this, the issue of increasing the depth of oil refining, which is acute at Kazakhstan refineries, is being solved. This method of oil extraction can be used as a secondary raw material for the preparation of modified bitumen and is characterized by the cheapness of its production. The analysis of the work performed showed that polymer-modified bitumen of various grades, such as PMB 100/130, PMB 70/100, PMB 50/70, PMB 35/50, can be obtained from samples of oil sludge and oil extracted by bioremediation from the oil sludge of the Uzen deposits, and they fully comply with the requirements of Kazakhstan standards ST RK 2534-2014.

This leads to the conclusion that the technological parameters of bitumen are the same in both variants. The use of the proposed method of oil sludge processing is expedient from the point of view of improving the ecology of the environment and is economically justified by obtaining additional quantities of petroleum products.

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REFERENCES

- [1] Park H. M., Kim Y. R., Aargau S., Thiago F., Lute JES. Effects of aggregate structure on hot-mix asphalt rutting performances in low traffic volume local. *Construction and Building Materials* 2009;23:2177–2182. <https://doi.org/10.1016/j.conbuildmat.2008.12.007>
- [2] Ichikawa S., Kai I., Mihara Y. Patent US 4532271. Bituminous paving material and a method for the preparation thereof. 30.07.1985.
- [3] Yildim Y. Polymer modified asphalt binders. *Construction and Building Materials* 2007;21:66–72. <https://doi.org/10.1016/j.conbuildmat.2005.07.007>
- [4] Environmental Protection Agency (EPA). Safe, environmentally acceptable resources recovery from oilrefinery sludge. Washington DC, 1991.
- [5] Wang X. *et al.* Effect of biostimulation on community level physiological profiles of microorganisms in field-scale biopiles composed of aged oil sludge. *Bioresource Technology* 2012;111:308–315. <https://doi.org/10.1016/j.biortech.2012.01.158>
- [6] Lotosh V. E. Pererabotka othodov prirodopol'zovaniya. (Recycling of environmental waste). Ekaterinburg: Poligrafist, 2007. (In Russian).
- [7] Chang C. Y., Shie J. L., Lin P., Wu C. H., Lee D. J., Chang C. F. Major products obtained from the pyrolysis of oil sludge. *Energy Fuels* 2000;14:2145–2156. <https://doi.org/10.1021/ef0000532>
- [8] Solov'yanov A. A. Pererabotka nefeshlamov s ispol'zovaniyem khimicheskikh i biologicheskikh metodov. (Processing of petroleum sludge with chemical and biological methods). *Zashchita okruzhayushchey sredy v neftegazovom komplekse. Environmental protection in the oil and gas industry* 2012;5:30–39. (In Russian).
- [9] Czin G., Luan' M., CHen' T. Perspektivy razvitiya processov pererabotki nefeshlamov. (Prospects for the development of oil sludge processing processes). *Himiya i tekhnologiya topliv i masel* 2011;4:44–54. (In Russian).
- [10] Yagafarova G. G., Barakhnina V. B., Safarov A. K. H., Il'ina E. G., Yagafarov I. R. Bioremediatsiya neftezagryaznennykh pochv. (Bioremediation of oil-contaminated soils). *Materialy 3-go syezda neftepromyshlennikov Rossii. Neftepererabotka i neftekhimiya: problemy i perspektivy.* (Materials of the 3rd meeting of Russian oilmen. Oil refining and petrochemistry: problems and prospects). Ufa, 2001.
- [11] Mazlova Y. A., Meshcheryakov S. V. Problemy utilizatsii nefeshlamov i sposoby ikh pererabotki. M.: Noosfera, 2001.
- [12] Reshetov V. A., Pavlov V. T., Pavlov A. T., Likhachev M. P., Bolotskiy A. N., Turunov D. L., Morkovin V.V. Patent 2193578 RF. Sposob pererabotki nefesoderzhashchikh shlamov Rossii (A method for processing oil-containing sludge in Russia). (Russian).
- [13] Yagafarova G. G. Utilizatsiya ekologicheskikh opasnykh burovykh ostavov. (Disposal of environmentally hazardous drilling residues). *Neftegazovoye delo (Oil and gas business)*. 2006.
- [14] Yagafarova G. G., Akchurina L. R., Fedorova Y. A., Safarov A. K., Moskovets A. V., Yagafarov I. R. Dorozhnyye smesi iz serogo betona. (Gray concrete road mixes). V sb. materialy nauchno-prakticheskoy konferentsii Neftegazpererabotka 2014. (Materials of the scientific-practical conference 'Neftegazpererabotka 2014'). Ufa, 2014. (In Russian).
- [15] Yagafarova G. G., Akchurina L. R., Fedorova Y. A., Yagafarov I. R., Safarov A. K. Povysheniye effektivnoye rekul'tivatsii neftegraznennykh gruntov. (Increasing the effective reclamation of oil-bearing soils). *Bashkirskiy khimicheskij zhurnal. Bashkir Chemical Journal* 2011;18(2):72–74.
- [16] Yagafarova G. G., Fedorova Y. A., Leont'yeva S. V., Safarov A. K., Yagafarov I. R., Lavrenchuk S. M. Aktivatsiya aborigennykh nefteokislyayushchikh mikroorganizmov uchenyye trudy Mezhdunarodnoy nauchno-prakticheskoy konferentsii. (*Proceedings of the International Scientific and Practical Conference*). Kurganskiy Gosudarstvennyy Universitet 2010.
- [17] Yagafarova G. G., *et al.* Poligon dlya utilizatsii nefesoderzhashchikh othodov. (Landfill for the disposal of oily waste). *Bezopasnost' v tekhnosfere* 2010;3:45–47.
- [18] Magid A. B. Biotestirovaniye kak metod opredeleniya toksichnosti neftezagryaznennykh otkhodov (Biotesting as a method for determining the toxicity of oil-contaminated waste). *Neftepererabotka i neftekhimiya* 2011;9:24–27.
- [19] Karami E., Jafari Behbahani T. Upgrading Iranian petroleum sludge using polymers. *Journal of Petroleum Exploration and Production Technology* 2018;8:1319–1324. <https://doi.org/10.1007/s13202-017-0416-1>
- [20] Abdibattayeva M., Bissenov K., Zhubandykova Z., Orynassar R., Tastanova L. Complex Oil-containing Waste Treatment by Applying Solar Energy. *Environmental and Climate Technologies* 2020;24(1):718–739. <https://doi.org/10.2478/rtuct-2020-0045>
- [21] Abdibattayeva M., Bissenov K., Zhubandykova Z., Orynassar R., Tastanova L., Almatova B. Purification of Oil-Containing Waste Using Solar Energy. *Environmental and Climate Technologies* 2021;25(1):161–175. <https://doi.org/10.2478/rtuct-2021-0011>
- [22] Primerano P., Milazzo M. F. Recycling of Oil Fly Ash in The Adsorption of Dyes from Industrial Wastewater. *Ecological Chemistry and Engineering S* 2020;27(2):257–270. <https://doi.org/10.2478/eces-2020-0012>

- [23] Drzewicz P., Nałęcz-Jawecki G., Drobnińska A., Zgadzaj A., Smoliński A., Krzan M., Starzycka A. Toxicological Evaluation of Thermal Treatment of Drilling Waste from Shale Gas Exploration in Poland. *Ecological Chemistry and Engineering S* 2019;26(1):45–57. <https://doi.org/10.1515/eces-2019-0004>
- [24] Patent №35103 RK. Munamen lastanğan topyraqtı bioremediasialay tásılı. /Isanov T.Sh., Kenjetaev G.J., Boranbaeva A.N., Korbakova S.E., Syrlybekqyzy S., Isanov M.Sh., Sherádzanova T.T., Kýandykov B.M. Opublikovano: 04.06.2021. Byul. №35103
- [25] Boranbayeva A *et al.* Remediation of soils contaminated with oil with a biological product with immobilization of bacteria by carriers from local soils. *IOP Conf. Ser.: Earth Environ. Sci.* 1043:012047. Voronezh, Russia. <https://doi.org/10.1088/1755-1315/1043/1/012047>
- [26] Wang H., *et al.* Effects of Aged Oil Sludge on Soil Physicochemical Properties and Fungal Diversity Revealed by High-Throughput Sequencing Analysis. *Archaea* 2018:Article ID 9264259. <https://doi.org/10.1155/2018/9264259>
- [27] Suyunova A. B., Conway V. D. Comparative evaluation of the efficiency of biological products for cleaning-up of the Caspian coast soils polluted with oil products. *International Journal of Pharmaceutical Research* 2018;10:811–815. <https://doi.org/10.31838/ijpr/2018.10.04.140>
- [28] Poyntner C., Prem M., Mann O., Blasiani B., Sterflinger K. Selective screening: isolation of fungal strains from contaminated soils in Austria. *Journal of Land Management, Food and Environment* 2018;68(3):157–169. <https://doi.org/10.1515/boku-2017-0014> (In German).
- [29] Kaszycki P., Petryszak P., Supel P. Bioremediation of A Spent Metalworking Fluid with Auto- And Allochthonous Bacterial Consortia. *Ecological Chemistry and Engineering S* 2015;22(2):285–299. <https://doi.org/10.1515/eces-2015-0017>
- [30] Imanbayev Y., Akkenzheyeva A., Bussurmanova A., Serikbayeva A., Boranbaeva A. Preparation of polymer bitumen binder based on mixtures of petroleum products in the presence of a stabilizer. *Processes* 2021;9(1):182. <https://doi.org/10.3390/pr9010182>
- [31] Boranbayeva A. N., Serikbayeva A. K., Imanbayev E. I., Busurmanova A. S., Akkenzheyeva A. S. Snizheniye nagruzki na okruzhayushchuyu sredu za schet utilizatsii nefteshlamov na dorozhnykh bitumakh. (Reducing the burden on the environment due to the disposal of oil sludge on road bitumen). *Ekologiya promyshlennogo proizvoditelya: mezhotraslevoy nauchno-prakticheskiy zhurnal. (Ecology of an industrial manufacturer: interdisciplinary scientific and practical journal)* 2022;1:117.
- [32] GOST 6370-83. Petroleum, petroleum products and additives. Method for determination of mechanical admixtures.
- [33] GOST 2477. Petroleum and petroleum products. Method for determining the water content.
- [34] GOST 11851-2018. Petroleum. Methods of paraffin determination.
- [35] GOST 31734-2012. Interstate standard. Oil fuels. Method of determination of water and sediment by centrifugation.
- [36] GOST 26449.1-85. 26. Gravimetric method for the determination of petroleum products.
- [37] Airey G. D. Rheological Properties of Styrene Butadiene Styrene Polymer Modified Road Bitumens. *Fuel* 2003;82(14):1709–1719. [https://doi.org/10.1016/S0016-2361\(03\)00146-7](https://doi.org/10.1016/S0016-2361(03)00146-7)
- [38] Akkenzheyeva A. S., Sydykov S. Y. Modifikatsiya bituma s ispol'zovaniyem stirol-butadiyen-stirol'nogo blok-sopolimera. *Yessenov Science Journal* 2018;1(33):71–76.
- [39] Vargas M. A., López N. N., Cruz M. J., Calderas F., Manero O. Viscoelasticity of asphalts modified with SEBS copolymers functionalized with various amounts of maleic anhydride. *Rubber Chem. Technol.* 2009;82:244–270. <https://doi.org/10.5254/1.3548248>
- [40] Wen G., Zhang Y., Zhang Y., Sun K., Fan Y. Rheological characterization of storage-stable SBS-modified asphalts. *Polym Test* 2002;21(3):295–302. [https://doi.org/10.1016/S0142-9418\(01\)00086-1](https://doi.org/10.1016/S0142-9418(01)00086-1)
- [41] Wen G., Zhang Y., Zhang Y., Sun K., Fan Y. Improved properties of SBS modified asphalt with dynamic vulcanization. *Polymer Engineering and Science* 2002;42(5):1070–1081. <https://doi.org/10.1002/pen.11013>
- [42] Chen J. S., Huang C. C. Fundamental characterization of SBS-modified asphalt mixed with sulfur. *Journal of Applied Polymer Science* 2006;103(5):2817–25. <https://doi.org/10.1002/app.24621>
- [43] Zhang F., Yu J., Wu S. Effect of ageing on rheological properties of storage-stable SBS/sulfur-modified asphalts. *Journal of Hazardous Materials* 2010;182(1–3):507–17. <https://doi.org/10.1016/j.jhazmat.2010.06.061>
- [44] Zhang F., Yu J., Han J. Effects of thermal oxidative ageing on dynamic viscosity, TG/DTG, DTA and FTIR of SBS- and SBS/sulfur-modified asphalts. *Construction and Building Materials* 2011;25(1):129–37. <https://doi.org/10.1016/j.conbuildmat.2010.06.048>
- [45] Li Y., Li L., Zhang Y., Zhao S., Xie L., Yao S. Improving the aging resistance of styrene-butadiene-styrene tri-block copolymer and application in polymer modified asphalt. *Journal of Applied Polymer Science* 2010;116(2):754–61. <https://doi.org/10.1002/app.31458>
- [46] Jasso M., Bakos D., MacLeod D., Zanzotto L. Preparation and properties of conventional asphalt modified by physical mixtures of linear SBS and montmorillonite clay. *Construction and Building Materials* 2013;38:759–65. <https://doi.org/10.1016/j.conbuildmat.2012.09.043>
- [47] Zhang B., Xi M., Zhang D., Zhang H., Zhang B. The effect of styrenebutadiene-modification on the characteristics and properties of asphalt. *Construction and Building Materials* 2009;23(10):3112–7. <https://doi.org/10.1016/j.conbuildmat.2009.06.011>

-
- [48] Zhang H., Yu J., Wang H., Xue L. Investigation of microstructures and ultraviolet aging properties of organo-montmorillonite/SBS modified bitumen. *Materials Chemistry and Physics* 2011;129(3):769–76. <https://doi.org/10.1016/j.matchemphys.2011.04.078>
- [49] Lee D. Modification of asphalt and asphalt paving mixtures by sulfur additives. *Ind Eng Chem Prod Res Dev* 1975;14(3):171–7. <https://doi.org/10.1021/i360055a009>