STUDY OF THE EFFICIENCY OF EXTRACTION OF OIL PRODUCTS FROM OIL-CONTAMINATED SOILS

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Soils impregnated with oil undergo different transformations depending on the amount and composition of the pollutant. This refers to changes in both the structure of the soil and the component composition of oil. Fluctuating ratios of oil-soil mixtures, especially long-term exposure to the environment, have a significant synergistic effect on these changes. These changes are important to consider when developing effective technologies for the rehabilitation of oil-contaminated soils. In the presented paper, the results of the extraction cleaning method of soil samples with a high content of oil fractions, which were exposed to natural influences for a long time in natural conditions, are discussed. The dependence of the extraction efficiency of oil-contaminated soils on the percentage of extractants, their various ratios, temperature, and the duration of mechanical treatment was studied. Processing of experimental data by static methods formed the basis of a mathematical model of the process based on the algorithm of extreme gradient boosting. The obtained model with optimal hyperparameters was used for forecasting the training data. The totality of the obtained experimental data allowed us to establish the most significant factors affecting the efficiency of oil-contaminated soil cleaning.

Keywords: *oil-contaminated soils, extraction, environment, cleaning of oil-contaminated soils, gradient boosting algorithm.*

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Introduction

The soil is a multiphase, high-reaction system, within which involves complex processes of creation, migration, destruction of various organic and inorganic compounds [1]. Soil perform vital ecological functions, providing vitality natural and anthropogenic systems [2,3]. Pollution with petroleum hydrocarbons leads to change basic properties of the soil (physical, chemical, morphological, biological), causing a violation or inhibition of primary functions of the soil cover and influence on general ecosystem processes on local, regional and global levels [4]. Oil is a multi-component substance consisting of many different ones connections. Toxicity of one component it can be neutralized by the presence others [5]. The most inhibiting impact on soil biogenicity aromatic hydrocarbons, smallest paraffin [6]. Long-term oil pollution time inhibits the activity of oxidative - reductive enzymes. Even through 10 years after oil pollution, low activity of dehydrogenase, peroxidase, polyphenoloxidase is revealed [7].

A significant amount of work is devoted to the study and development of the specified problem methods of cleaning soil from petroleum products, however, most of the methods considered in them have adverse consequences for the environment are expensive [8,9]. In the last 30 years , researches in this direction have shown that one of the most promising and ecologically safe methods of impacting oil-contaminated soils with the

aim of their utilization is the method of physical and chemical impact [10]. In the course of developing the technological utilization of oil-contaminated soils, based on their washing with an aqueous detergent solution of surface-active substances (SAS) with the participation of an organic solvent, theoretically based methods of mechanical, chemical, and physical-chemical effects on oil-contaminated soils were used [11]. The nature of oil- contaminated soil and the properties of oil pollution, the nature of "residual oil" on the surface of soil particles in the form of a strong film of the boundary layer of active components (acids, phenols, resins, asphaltenes, etc.), "obsolescence" of pollution [12] were taken into account. In the course of earlier studies [13-15], it was established that the displacement of oil by water-washing solution from the surface of particles of oil-contaminated soil is a complex process that depends on: the granulometric composition of the mineral solid phase of the soil, especially on the amount of clay fraction in it; structure of pore space and degree of dispersion of aggregates and microaggregates; surface properties and chemical composition of oil and its washing water and a number of other factors. The aim of this work was to study the efficiency of extraction of petroleum products from soils long-contaminated with petroleum hydrocarbons, to assess the influence of technological parameters of the cleaning process and to construct a predictive model of the degree of cleaning based on the Extreme Gradient Boosting (XGBoost) algorithm. Methods of mathematical modeling applied in petrochemistry have been reported previously [16]. The approach proposed in the present study is applied for the first time to petroleum products that have become environmental contaminants due to various reasons.

Materials and methods

For research, samples were taken from different areas on the territory of the oil industry enterprise "Balakhani Oil Company". In order to study the efficiency of the extraction of oil products from highly oil-contaminated samples, we carried out extraction processes using various solvents: benzene, n-hexane and toluene. To do this, the samples were placed in a flask, one of the solvents was added, and they were stirred for 3 hours in a reflux flask at a temperature of 21-22 ^{0} C. The extraction was repeated three times for each sample. Then the extracts were combined and the solvent was evaporated. They determined the content of the dry residue of the components. It was established that the degree of impregnation of oil in soils is quantitatively different. It was interesting to study the process of washing oil-contaminated soils using oil-removing reagents. Water- detergent resin and toluene were used as reagents. The study of the interphase surface tension of toluene solutions of residual oil isolated from contaminated soil at the interface with distilled water was carried out at a temperature of 20 ± 1 ^{0} C. The interphase surface tension of toluene solutions is also studied in oil with a mass fraction of oil of 25% at the boundaries with aqueous solutions of sodium hydroxide and non-ionic SAS OP-10 at a temperature of 20±1 ^{0} C. In the conducted experiments, the study of surface properties was carried out on systems consisting of two immiscible liquid phases: distilled water (aqueous solutions of aqueous sodium bicarbonate solution the surfactant OP-10) and toluene containing residual oil. To predict the degree of cleaning using input parameters, an effective implementation of the gradient boosting algorithm XGBoost (Extreme Gradient Boosting) was applied.

Experimental results

For the qualitative and quantitative characterization of the degree of pollution of petroleum products samples, we conducted the following studies. We took 5 samples of each sample, placed them in porcelain crucibles and calcined them at 100, 200, 300, 400 and 500 ^{0} C. First of all, we obtained data on the humidity of samples of these soils, then the evaporation of hydrocarbons with different boiling temperatures occurred. In practice, we studied

the content of volatile components. The data are shown in figure 1.

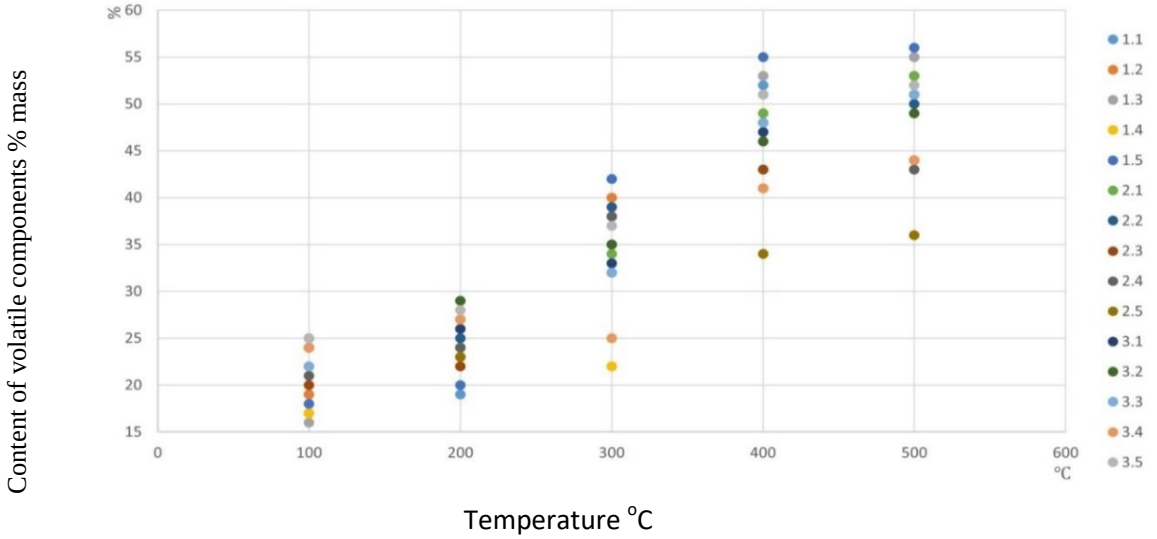


Figure 1. Content of volatile components % mass

Five parallel samples were analyzed from each sample. They studied the content of volatile components of the samples depending on their water and hydrocarbon nature. As can be seen from figures 1 and 2 a, b, c, g, the content of volatile components at 100 ^{0} C is at the same level and is characterized mainly by hidden absorption of water.

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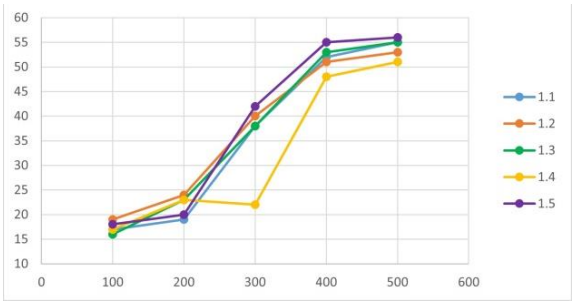


Fig. 2a Content of dry residue

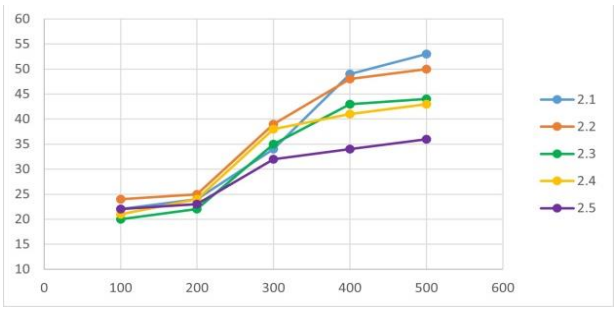


Fig. 2b Content of dry residue to the first sample to the second sample

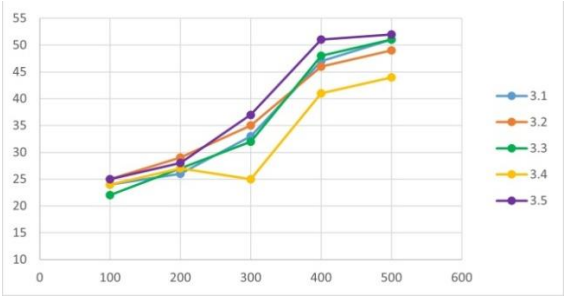


Fig. 2c Dry residue content of the third sample

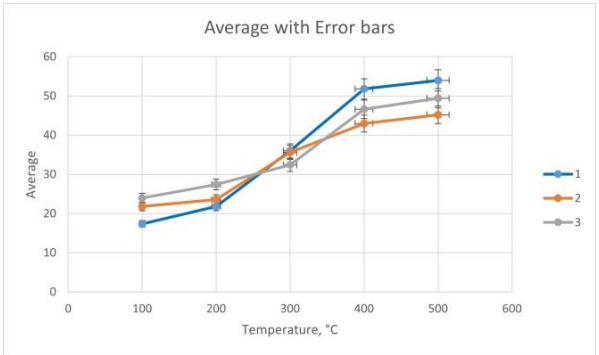


Fig. 2d Generalized results of samples according to contentdry residue sample

The presence of a small amount of free hydrocarbon components, which agrees well with the chromatographic analysis of the sample (Fig. 3).

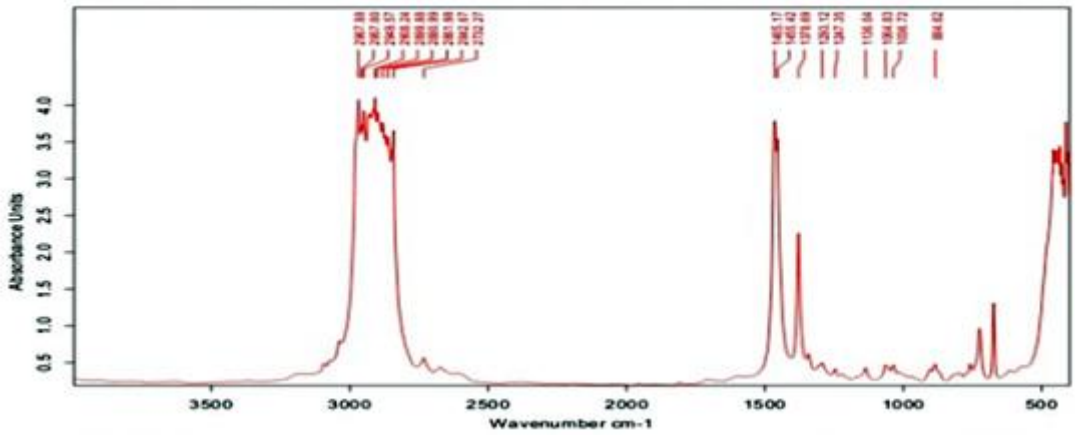


Figure 3. IR spectrum of the hydrocarbon composition of the extract.

The IR spectrum of the hydrocarbon copmposition of the extract was interpreted using artificial intelligence,and the following results were obtained:

1. 2925 and 2855 cm^{-1} — valence vibrations of C–H (CH_2 and CH_3), aliphatic hydrocarbons (saturated chains), typical for petroleum, oil, diesel.
2. ~1460 and 1375 cm^{-1} - CH_2/CH_3 deformation vibrations , also indicate hydrocarbon nature.
3. ~1710 cm^{-1} — carbonyl group C=O, possible presence of oxidation products (aldehydes, ketones, acids).
4. 1100–1000 cm^{-1} — possibly, C–O oscillations (ethers, alcohols — by-products of petroleum decomposition).
5. ~880–720 cm^{-1} — possible aryl rings or substituted alkanes.

Thus, this spectrum confirms the presence of:
- aliphatic hydrocarbons (petroleum products),
- oxidized derivatives (possibly from aging/biodegradation),
- traces of oxygen-containing compounds.

The obtained results unequivocally confirm a rather complex transformation of petroleum components in the microbiological environment of soils underthe influence of technological environmental factors.

When separating the oil fraction from the composition of oil-contaminated soil using various solvents: benzene, n-hexane and toluene, The influence of temperature, amount and variety of solvents, intensity of mixing on the efficiency of extraction of oil products from samples of oil-contaminated soil taken from the vicinity of active wells is studied. The obtained results are shown in figures 4 a, b, c, d.

Figure. 4. it is clear that the use of extractants is quite strongly different according to the degree of solubility of petroleum products contained in the selected sample. The obtained results show that the best indicators are observed when using toluene. Toluene was included in the study as a reference aromatic solvent due to its high solubility when applied to petroleum hydrocarbons. Comparing the extraction results with toluene and benzene and n-hexane allows us to assess the influence of the solvent on the efficiency of petroleum product extraction. For the deepening of technological aspects and the mechanism of washing oil products, adsorbed on the surface of soil components, they studied the influence of an alkaline environment in the presence of SAS. In the process of cleaning the oil-contaminated soil, as an organic solvent, the model solvent ritel toluene (pure for analysis) was used. The study of the interphase surface tension of toluene solutions of residual oils isolated from contaminated soil at the interface with distilled water was carried out at a temperature of 20 ± 1^{0}

C. The interfacial surface tension of toluene solutions of the studied oils with a mass fraction of 25% oil at the boundaries with aqueous solutions of sodium hydroxide and non-ionic SAS OP-10 at a temperature of 20±1 ^{0} C was also monitored. In the conducted experiments, the study of surface properties was carried out on systems consisting of two immiscible liquid phases - distilled water (aqueous solutions of aqueous sodium bicarbonate solution and SAS OP-10 and toluene road of "residual oil". It is known that the presence of interphase surface tension at the oil-water interface depends on the content of SAW in the oil. Study of interphase surface tension the aqueous solutions of this residual oil with a mass content of 25% at the border with the aqueous solution of SAS and NaOH were carried out at a temperature of 20 ± 1 ^{0} C [17]. It was established that when changing the amount of SAS OP-10 from 0.0 to 0.10 % by mass. in the composition of the aqueous solution, the value of the surface tension decreases from 10.3 mN/m to the value of 5.3 mN/m, i.e. in 1.9 times. The change in the interfacial surface tension of toluene solutions of the studied oil from the concentration of hydroxy and sodium in an aqueous solution of 0.0 to 0.6 % by mass. at 20 ^{0} C, it showed that this indicator decreased from 10.30 mN/m to 0.80 mN/m.

The sum of these results allowed us to establish that the use of an organic solvent, nonionic surfactant, and alkali in the composition of an aqueous washing solution during the cleaning of oil-contaminated soil have a positive effect on reducing surface tension, lead to an increase in the molecular surface processes of the impact of oil on the soil, and increase the efficiency of their cleaning.

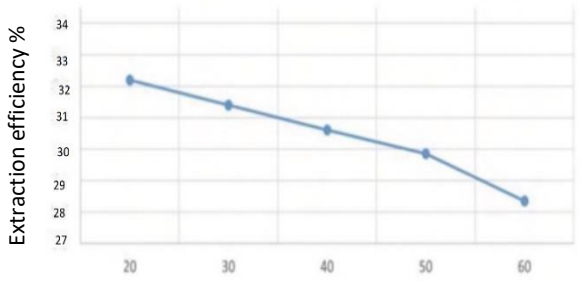


Fig. 4a Dependence on the efficiency of the process extraction from temperature sample

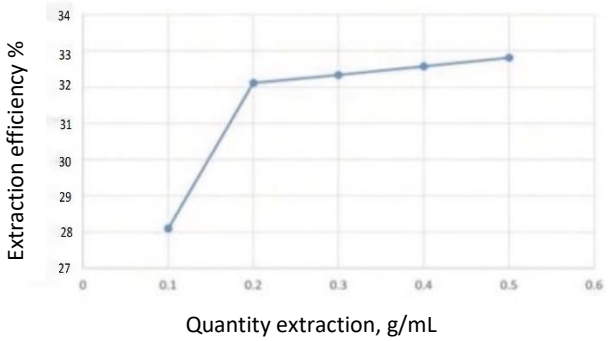


Fig. 4b Dependence on the efficiency extraction from the amount of solvent

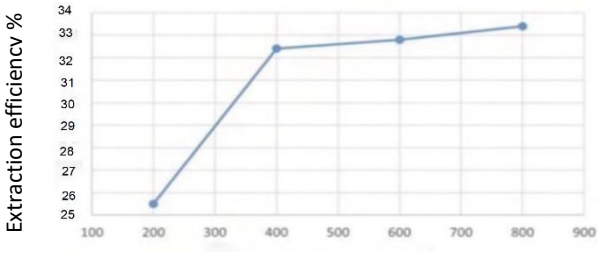


Fig. 4c Dependence of efficiency extraction sample

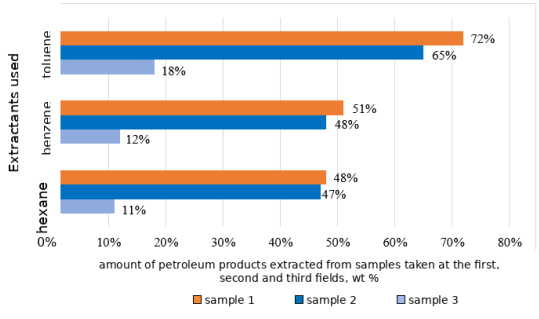


Fig. 4d Amount of petroleum products extracted from the intensity of mixing. by various extractants %, mass.

Mathematical modeling

Based on the obtained results, it should be noted that by diluting oil with an organic solvent, which covers the surface of mineral particles of oil - contaminated soil, its surface activity can be increased due to the formation of an optimal concentration of surfactants included in its composition, and, in particular, free - compounds not associated with intermolecular interactions both in its volume and at the interface between the phases of the oil - water washing solution. The active components of the oil-contaminant layer SAS of mineral particles of the soil (resins, asphaltenes, acids, etc.) are located on their surface in the form of a durable film of the boundary layer. The effectiveness of washing mineral particles depends on the breakthrough of such a film by the current of the aqueous washing solution.

The high efficiency of mass exchange in highly concentrated dispersed systems during the interaction of the dispersed phase with the dispersion medium in the technology of cleaning oil-contaminated soil [18] is ensured by the methods of physical-chemical and mechanical impact on the aggregated particles of the solid phase. For this, it is necessary to produce dispersion, spraying of oil-contaminated aggregates of dusty clay particles and destruction of aggregates consisting of grains of oil-contaminated natural elementary sand particles. At the same time, the degree of destruction of the structure [19,20] should be - considered as a criterion for the effectiveness of the impact on dispersed systems, and the - ultimate destruction as the optimal level of these effects to achieve high mass transfer efficiency. For the effective use of the results of the above experiments, the developed technology for washing oil-contaminated soil provides for the mechanical destruction of the oil film when mixing the mineral particles of the soil with their further contact with an aqueous washing solution containing an organic solvent [21-23].

The totality of the obtained experimental data allowed us to establish the most significant factors affecting the efficiency of oil-contaminated soil cleaning and the interval of variation of each of them: X_1 – the content of alkali in the washing solution, NaOH %; X_2 – SAS content in the washing solution, %; X_3 – duration of mechanical impact, min.; X_4 – the content of the organic solvent in the washing solution, %; Y – degree of cleaning of oil-contaminated soil, %. These parameters were used in the preparation of the experiment planning matrix (Table 1).

For to study the dependence of the efficiency of cleaning oil-contaminated soils on the specified factors, in particular, on the percentage of components in the aqueous solution and the duration of mechanical exposure, a regression equation was compiled: $Y = 0.04717 + 1037.4017X_1 + 162.441X_2 + 0.5872X_3 + 11.9882X_4$ [18]. The interpretation of the model parameters is as follows: an increase of X_1 by 1 unit leads to an average increase in Y of 1037.402 units; an increase of X_2 by 1 unit leads to an average increase in Y of 162.441 units; an increase of X_3 by 1 unit leads to an average increase in Y of 0.587 units; an increase of X_4 by 1 unit leads to an average increase in Y of 11.988 units. Based on the maximum weight coefficient, we conclude that factor X_1 (the amount of alkali) has the greatest influence on the degree of purification (Y). It is also established that one or more model parameters are not statistically significant.

To predict the degree of cleanup of oil-contaminated soil, the Extreme Gradient Boosting (XGBoost) algorithm, an advanced gradient boosting method over decision trees, was used. The algorithm generates an ensemble of sequentially trained trees, each of which minimizes the errors of previous models. The advantages of XGBoost include high forecasting accuracy, robustness to overfitting, the ability to assess the significance of factors, and efficient operation with limited datasets. Model hyperparameters were optimized using a random search method.

Table 1. Matrix of planning and results of experiments

Variable levels		Coded				Working matrix				Output parameter	
		X_1	X_2	X_3	X_4	NaOH, % wt.	SAS, % wt.	Mechanical treatment time, min	Solvent, % wt.	Yes , % degree of cleaning	
Base: 0		0	0	0	0	0.03	0.05	10	0.25		
Interval variation		Δ	Δ	Δ	Δ	0.03	0.05	10	0.25		
top: +1		+	+	+	+	0.06	0.1	20	0.5	Exp.	Calc
lower: -1		-	-	-	-	0	0	0	0		
Experiences:	1	+	+	+	+	0.06	0.1	20	0.5	96	87.10
	2	+	+	+	-	0.06	0.1	20	0	89	86.15
	3	+	+	-	+	0.06	0.1	0	0.5	81	81 .40
	4	+	+	-	-	0.06	0.1	0	0	77	77 .11
	5	+	-	+	+	0.06	0	20	0.5	72	7 0.24
	6	+	-	+	-	0.06	0	20	0	71	69.99
	7	+	-	-	+	0.06	0	0	0.5	70	68.69
	8	+	-	-	-	0.06	0	0	0	68	6 7.85
	9	-	+	+	+	0	0.1	20	0.5	43	28 .07
	10	-	+	+	-	0	0.1	20	0	28	28 .02
	11	-	+	-	+	0	0.1	0	0.5	16	15.86
	12	-	+	-	-	0	0.1	0	0	10	1 4.96
	13	-	-	+	+	0	0	20	0.5	13	13 .76
	14	-	-	+	-	0	0	20	0	10	1 3.76
	15	-	-	-	+	0	0	0	0.5	8	11.88
	16	-	-	-	-	0	0	0	0	0	11.88
	17	0	0	0	0	0.03	0.05	10	0.25	54	53. 56
	18	0	0	0	0	0.03	0.05	10	0.25	54	53. 56
	19	0	0	0	0	0.03	0.05	10	0.25	53	53. 56

To predict the degree of cleaning using input parameters, an effective implementation of the gradient boosting algorithm XGBoost (Extreme Gradient Boosting) was applied. This method of machine ensemble learning is characterized by a high speed of operation, and the models built on its basis have good performance. The random search algorithm is applied to optimize the hyperparameter used model. The obtained model with optimal hyperparameters was used for forecasting the training data. The results of the conducted experiments and calculated parameters are shown in Figure 5 a, b, c, d. and 6.

To construct graphs in three-dimensional space, the program "3D_graph" To test the adequacy of the obtained mathematical model and optimization conditions, a series of parallel experiments were conducted was written in python.

Discussion

Standard python library functions are used in the program. Source data entered into the text of the program in the form of arrays. The result of the program is displayed on the laptop monitor and saved as a graphic file on the disk. A number of parallel experiments were conducted to verify the adequacy of the work of the obtained mathematical model and optimization conditions. From the three-dimensional graph, it can be concluded that the applied mathematical model can, in principle, describe the process of extraction cleaning of oil-contaminated soils.

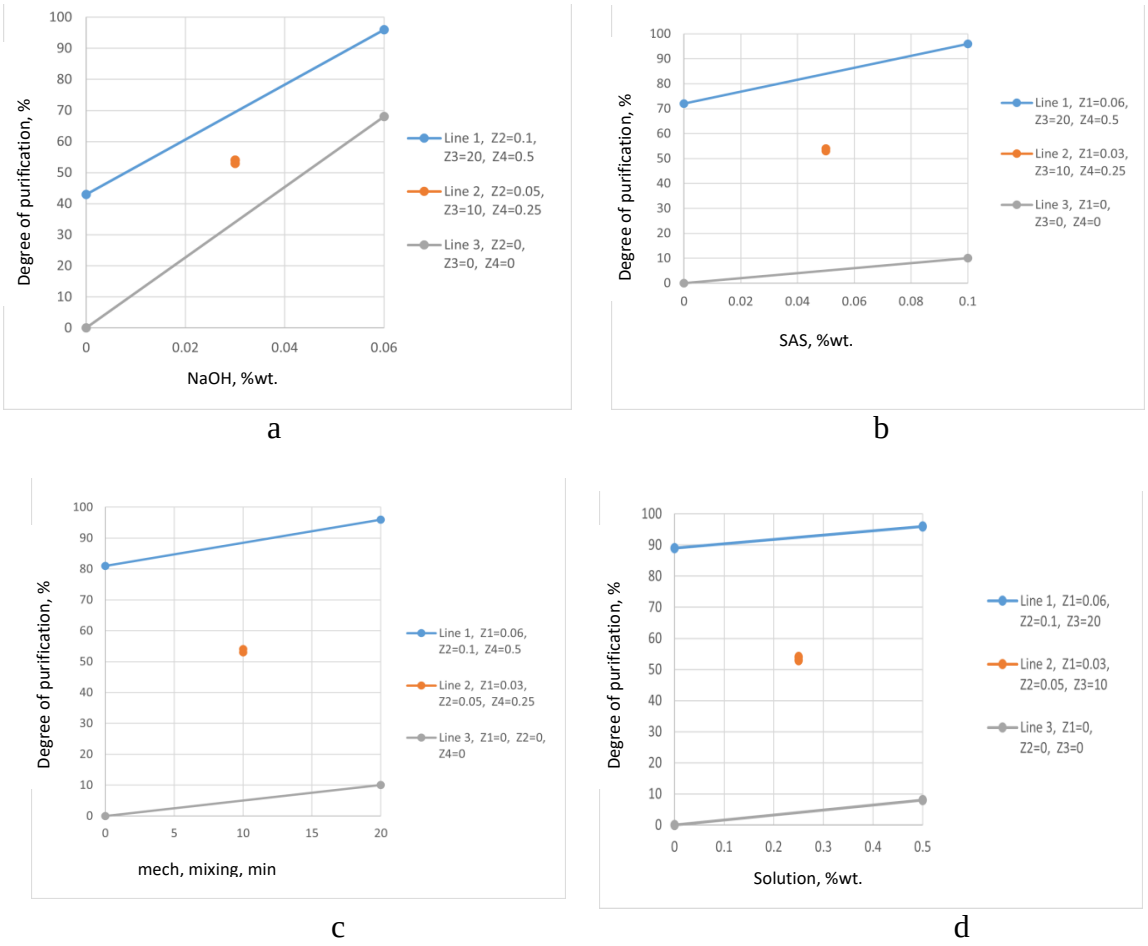


Figure 5. Graphs of the dependence of purification degree on various parameters
a – dependence of the purification degree on alkali,
b – dependence of the purification degree on surfactant,
c – dependence of the purification degree on mechanical treatment,
d – dependence of the purification degree on solvent.

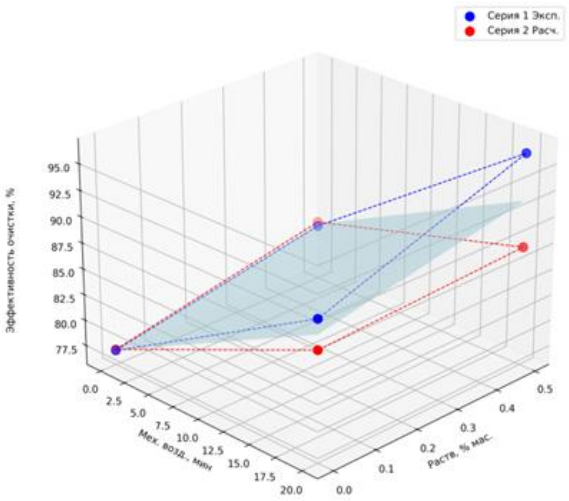


Figure 6. 3D plot of experimental data with regression surface.

However, the difference between experimental results and calculations does not take into account the synergistic effect of alkali and SAM used in the extraction process in the applied

model. This emphasizes the necessity of conducting larger-scale experiments to identify the extraction coefficient , characterizing the synergetic effect, and refining the equation of the mathematical model.

Conclusion

The conducted studies have shown that the efficiency of oil product extraction from oil-contaminated soils depends significantly on the nature of the solvent, the alkali and surfactant content of the washing solution, and the duration of mechanical action. The highest degree of oil product extraction was achieved using toluene. It was established that the alkali content is the most significant factor in the cleaning process. The developed model, based on the XG Boost algorithm, demonstrated the ability to predict the degree of cleaning of oil-contaminated soils and can be used to optimize processing modes.

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NEFTLƏ ÇİRKLƏNMİŞ TORPAQLARDAN NEFT MƏHSULLARININ ÇIXARILMASI SƏMƏRƏLİYİNİN ÖYRƏNİLMƏSİ

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Neftlə hopdurulmuş torpaqlar çirkləndiricinin miqdarından və tərkibindən asılı olaraq müxtəlif çevrilmələrə məruz qalır. Bu, həm torpaq strukturunun, həm də neftin komponent tərkibinin dəyişməsi ilə əlaqədardır. Neft–torpaq qarışıqlarının dəyişkən nisbətləri, xüsusilə də ətraf mühitin uzunmüddətli təsiri bu dəyişikliklərə əhəmiyyətli sinergetik təsir göstərir. Bu dəyişikliklərin nəzərə alınması neftlə çirklənmiş torpaqların reabilitasiyası üçün effektiv texnologiyaların hazırlanmasında mühüm əhəmiyyət daşıyır. Təqdim olunan işdə təbii şəraitdə uzun müddət ərzində ətraf mühit amillərinin təsirinə məruz qalmış, yüksək miqdarda neft fraksiyaları olan torpaq nümunələrinin ekstraksiya üsulu ilə təmizlənməsi üzrə tədqiqat nəticələri müzakirə olunur. Neftlə çirklənmiş torpaq-qruntların təmizlənmə effektivliyinin müxtəlif amillərdən, xüsusilə komponentlərin faiz payından və mexaniki təsirin müddətindən asılılığını öyrənmək üçün texnoloji parametrlərin təsirinin qiymətləndirilməsi meyarı kimi təmizlənmə effektivliyindən istifadə edilmişdir. Eksperimental məlumatların statistik metodlarla işlənməsi ekstremal gradient boosting alqoritminə əsaslanan prosesin riyazi modelinin qurulmasına zəmin yaratmışdır. Optimal hiperparametrlərlə əldə olunan model öyrədilmiş məlumatların proqnozlaşdırılması üçün istifadə edilmişdir. Alınmış eksperimental məlumatların məcmusu neftlə çirklənmiş torpağın təmizlənmə effektivliyinə təsir edən ən mühüm amilləri müəyyən etməyə imkan vermişdir.

Açar sözlər: neftlə çirklənmiş torpaqlar, ekstraksiya, ətraf mühit, neftlə çirklənmiş torpaq-qruntların təmizlənməsi, gradient boosting alqoritmi.