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Electrokinetic remediation of oil-contaminated soils

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This investigation was undertaken to determine the factors influencing electrokinetic remediation of soils from petroleum pollutants. The remediation method was applied in two versions: (i) static and (ii) flowing, when a sample was washed with leaching solution. It was found that all the soils studied can be purified using this technique. It was also observed that the mineral and grain-size composition of soils, their properties, and other parameters affect the remediation efficiency. The static and flowing versions of the remediation method removed 25–75% and 90–95% of the petroleum pollutants, respectively from the soils under study.

Keywords: Remediation, soil, oil and petroleum pollution, electrokinetic method, electrochemical degradation and lixiviation, static and flowing version.

Introduction

Oil is one of the most common toxic pollutants of the environment. Most of the soils are polluted with oil and petroleum products at traffic accidents and because of oil spills during oil production and transportation, and therefore development of efficient methods of soil remediation from oil is a pressing problem. Electrokinetic remediation is one of such methods. This method is widely applied for soil remediation from metals, chlororganic and other toxic substances.^[1-3,7] Our earlier studies have shown high efficiency of the electrokinetic method for oil-contaminated soil treatment.^[4–6,8,9] However, its application for soil remediation from oil requires further investigations, since some factors influencing the efficiency of this process are poorly studied. We have undertaken therefore this study, which is aimed at finding out the effects of some factors influencing electrokinetic remediation of soils of various origins from petroleum pollutants.

Materials and methods

The laboratory investigation was performed in electroosmotic cells of two types: type one (type i) envisaged a static version of the experiment (Fig. 1) and type two (type ii) envisaged a flowing version simulating sample washing and electrochemical leaching out the pollutant (Fig. 2). The test procedure at the static version (type i) included the following. A prepared soil sample of disturbed or natural structure with a known amount of introduced oil and preset moisture content was placed into an electroosmotic device and linked up to direct current source. This started the process of electrochemical remediation of the sample, which lasted from 6–8 hours to several days. At that, the electroosmotic filtrate was collected. After completing the cleaning, the current was disconnected, the cell was dismantled, the specimen was cut into equal portions along the way of the electroosmotic filtration, and the final concentrations of oil and moisture were measured in each of the portions.

These results were used then for plotting graphs of redistribution of oil concentrations in the sample from the anode toward the cathode, and these graphs showed the remediation degree in the soil specimen.

The test procedure at the flowing version (type ii) was the same with the only difference that a vessel with a "washing" fluid (anolyte) was attached from the side of the anode to the open-type cell; the fluid (catholyte) enriched in leached out toxicant (oil) was collected from the specimen from the side of the anode (Fig. 2). In this case, the remediation was performed under stationary conditions of electroosmotic filtration, and therefore the moisture content in the specimen did not change and the electroosmosis rate was constant.

We used soils, which ranged in their grain-size composition from sand to loam, as study targets. A total of 13 kinds of the most common soils of various origins, mineral composition, and dispersivity were examined (Table 1). We examined soils of six genetic soil types that are the most widespread in the European part of Russia: from peat

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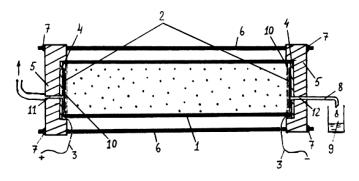


Fig. 1. The laboratory set-up for the electrokinetic soil remediation in the static version, type i: 1–plexiglass tube, 2–platinum electrodes, 3–conductors, 4–rubber linings, 5–rests, 6–pulling together bolts, 7–nuts, 8–branch pipe, 9–drain-pipe for water, 10–paper filters, 11–12–apertures for an output of gas and a liquid component–a filtrate.

and forest to sod-podzol and chernozem types of various grain-size varieties. We examined soil specimens of natural structure collected from different genetic horizons, which allowed approximating the laboratory tests to reality in the maximum degree. The selection of these soil types was caused by the fact that the examined soils are the principal types of the most common soils that are subject in the highest degree to technogenic pollution in the Russian territory.

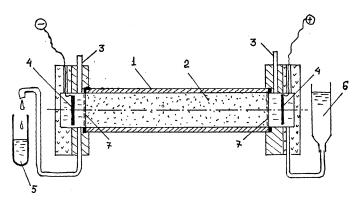


Fig. 2. The laboratory set-up for the electrochemical soil remediation in the flowing version, type ii: 1–plexiglass tube, 2–soil, 3–drain-pipe for gas, 4–platinum electrodes, 5–filtrate, 6–vessel for water, 7–paper filter.

We applied X-ray diffractometry for determining the chemical and mineral composition and salinity of all the examined soils. We also examined the structural and textural specific features of the soils: grain-size and microaggregate compositions, their macro- and microstructures (we applied an electron microscope for examining the latter) as well as sets of their physical and physicochemical properties including adsorption parameters by using common and standard techniques.

 Table 1. The granulometric composition of investigated soils.

		Maintenance of fractions, %								
Soil type	Horizons*	1,0– 0,5	0,5– 0,25	0,25– 0,10	0,1– 0,05	0,05– 0,01	0,01– 0,005	0,005– 0,001	<0,001	Soil
Chernozem meadow	A_1	5	8	20	17	12	10	8	20	Loam average
Peat	A_1	16	13	12	34	6	5	5	9	Sandy loam
Sod-podzol	A_1	6	9	4	7	55	5	6	8	Sandy loam
Grey wood	A_1	6	9	4	7	55	5	6	8	Sandy loam
Dark grey wood	A_1	4	6	19	20	24	5	8	24	Loam easy
Chernozem typical	A_1	1	6	3	45	17	8	9	11	Loam easy
Chernozem ordinary	A_1	6	10	9	5	50	7	13	10	Loam easy
	\mathbf{B}_1	0	0	2	4	61	6	13	10	Loam easy
	$\mathbf{B}_{\mathbf{k}}$	0	0	5	3	65	6	8	12	Loam easy
Sod-podzol	A_1	2	5	5	4	55	5	16	9	Loam average
-	$A_1 A_2$	1	1	5	4	54	5	18	9	Loam average
	B_1	1	1	4	3	52	18	6	15	Loam average
Light grey wood	A_1	2	2	20	20	36	2	5	3	Sandy loam
	$A_1 A_2$	2	1	25	33	27	4	4	3	Sandy loam
	B ₁	2	2	27	23	35	2	3	4	Sandy loam
Alluvial-grass	A_1	6	9	4	7	55	5	6	8	Sandy loam
Chernozem leached	A_1	5	8	12	17	12	18	8	20	Loam heavy
Chernozem southern	A_1	6	5	2	4	19	10	8	44	Clay easy
	\mathbf{B}_1	4	4	1	2	19	10	7	42	Clay easy
Podzol	A ₁	1	1	10	9	43	7	13	16	Loam average
	A_2		1	9	1	46	2	8	33	Loam heavy
	$\overline{A_2} B_1$		1	5	4	42	2	8	38	Loam heavy

*Horizons: A-humus, B-illuvial.

Oil		Physical prope	erties	Fractional composition					
	Density (g/sm ³)	Molecular weight	Viscosity, MPa/s (20°C)	Beginning of boiling (°C)	Up to 150° C	Up to 200° C	Up to 250° C		
Tarasovsk's	0.814	4.0	190	58	20	29	43		
Usinsk's	0.854	7.5	205	64	10	27	40		
Mixture of the crude oil	0.822	71	209	98	2.5	5	12		
Yaregsk's	0.944	86.3	452	140	0.3	4	11		

Table 2. The characteristics of investigated oils.

We used oils from the Tarasovsksky, Usinsky and Yaregsky oil fields, as well as miscellaneous oil, which was a mixture of different oils from West Siberian oil fields, as oil pollutants (Table 2).

Results

Remediation at the static version

We performed a broad set of experiments examining factors that influence the possibility and efficiency of electrochemical remediation of various soils from oil and petroleum products, and the following results were obtained as a result of thoroughly analyzing the factors that affect efficiency of electrokinetic remediation of soils from oil.

We proved that the examined soils can be cleaned from oil by means of electrochemical methods. This provides remediation efficiency of 25–75% and even higher at the static version (type i) depending upon various internal and external factors. Oil travels in the direct current field as small droplets along with the electroosmotic filtrate from the anode to the cathode drawn into the water stream at the expense of viscous friction. A portion of oil travels in the same direction in dissolved state due to its dissolution at an elevated pH value that originates in the cathode zone. The influence of chemical and mineral composition of soils upon the degree of their electrokinetic remediation is realized through their parameters of the double electric layer (DEL). The higher is the physicochemical activity of certain clay minerals at the given DEL, the higher is the soil remediation from oil.

The remediation efficiency grows with increasing the soil dispersivity, and the optimum proportion of grain-size particles of the >0.01-mm fraction in sandy–argillaceous soils is 65–75%. A rise in the soil porosity (or a decline in its density) also enhances efficiency of electrokinetic soil remediation from oil.

A decrease in the physicochemical activity of soils: plasticity index (I_P), hydrophylic properties (K_h), and colloidal activity (K_a) also results in a decline in the intensity of electrochemical migration of oil and, hence, in a decrease of soil purification degree from hydrocarbons.

The performed experiments allowed determining the optimum ratio between solid and liquid phases in soils for electrochemical removal of oil from soils. The moisture content in soils close to that of free soil swelling and the water-oil ratio equal to 2 : 1 and higher are optimal for electrochemical oil migration in water-saturated soils. The proportion of removed oil sharply decreases at decline of moisture content in the soil and at increasing oil proportion in it, and consequently the efficiency of soil remediation from oil drops.

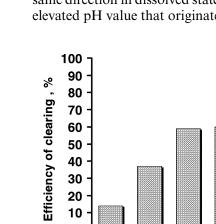


Fig. 3. The influence of "water-oil" ratio in the sod-podsolic soil on efficiency of its clearing (in the static version).

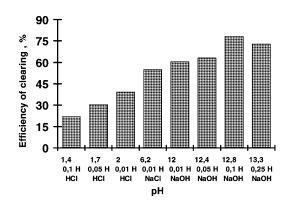


Fig. 4. The influence of composition and pH pore solution of sodpodsolic soil on the efficiency of its clearing from oil (in the static version).

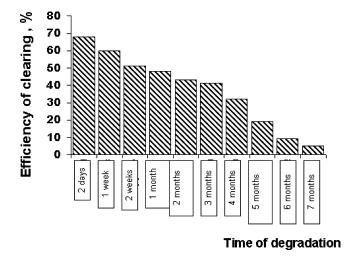


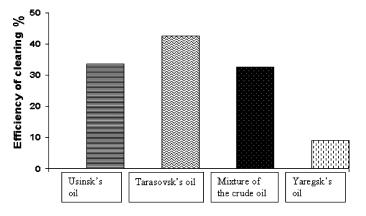
Fig. 5. The efficiency of remediation of the sod-podsolic soil from oil in a cathodic zone for different periods of oil degradation (in the static version).

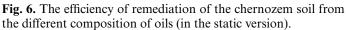
The influence of the water-oil ratio upon the degree of soil purification is shown in Figure 3.

The influence of composition and pH of the pore solution in soils upon the efficiency of their remediation was examined using hydrochloric acid (HCl), neutral salt (NaCl), and sodium alkali (NaOH). The results of these experiments are shown in Figure 4.

We also examined the influence of the degradation process (deterioration with time) of oil upon its electrochemical removal from soils (Fig. 5). It was found that the older is the oil contamination of a soil, the less efficient will be the method, and in this case, we have beforehand to convert the deteriorated oil into a more mobile state (possibly, by adding chemicals, surfactants, solvents, etc.). This remediation method is not efficient in soils with oil contamination older than 6 months.

We also examined the influence of the chemical composition and properties of oils upon the efficiency soil remediation from oil (Fig. 6), and found that the efficiency





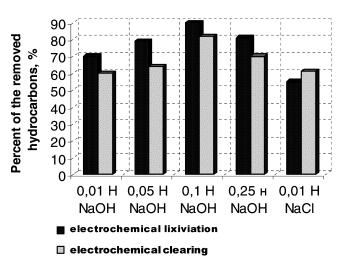


Fig. 7. The efficiency of electrokinetic remediation and lixiviation of oil from the sod-podsolic soil by the various extractants (in the flowing version).

of electrochemical soil remediation from oil pollutants declines with increasing the initial density of oils and abundance in them of pyrobitumens and resins; no such regularity was found with paraffins. The electrokinetic remediation of soils is more efficient from high-gravity oils and less efficient, from heavy oils.

Remediation at the flowing version

Soil remediation from petroleum pollutants is the most efficient at the flowing conditions (type ii), i.e., at electrochemical leaching of hydrocarbons (Fig. 7). The soil remediation efficiency made in this case 90–95% and even more. Tests aimed at selecting a leaching agent showed that water solution of sodium alkali at a concentration of 0.1–0.2 N is the most efficient agent. The tests revealed that oil dissolves under action of the alkali (NaOH) used as anolyte not only in the cathode zone but also completely throughout the interelectrode space, which facilitates fuller oil remediation from petroleum products.

Conclusions

The above-stated data allow drawing the following conclusions:

(i) Electrochemical technique is a highly efficient method of soils remediation from various petroleum pollutants.

(ii) This technique showed the following advantages compared to other methods: (a) a possibility of cleaning soils and underlying grounds directly in the rock mass down to a considerable depth; (b) high remediation degree and efficiency; (c) wide range of petroleum products that can be removed from soils by using this technique; (d) comparatively low cost of this remediation technique (compared to bioremediation and other methods); (e) relatively high rate of the remediation process; and (f) a possibility of efficiently integrating this technique with other methods.

(iii) The prospects of the technique development determine a necessity of more thorough investigation of individual factors that affect the remediation process and a selection of new leaching agents as well as development of flow charts for its efficient integration with other remediation techniques; this will be followed by passing to pilot tests.

Acknowledgment

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