Decontamination of heavy-metal polluted soil BY electrokinetic remediation

Kristina Greičiūtė, Saulius Vasarevičius

The electrokinetic method used for remediation of soil contaminated with heavy metals is a new developing technique for cleaning soil from heavy metals and radionuclides. By feeding an electric current through soil one induces movement of charges, which is generated by migration of ions and colloids in water present in pores and by reductive reactions on the surface of electrodes. The aim of the present investigation was to find out whether the electrokinetic soil cleaning method is suitable for cleanup of polluted soils. The method utilizes electrophoresis to drive contaminant species into and through a bed adjacent to a drive electrode. The bed comprises a moderately electrically conductive adsorbent material which is porous and is infused with water or other solvent capable of conducting electrical current. The efficiency of the method depends not so much on soil permeability as on the electric conductivity of water in pores and the distance of movement. The major task and the key goal were to determine suitability of the electrokinetic method for the treatment of soil contaminated with heavy metals (Cr, Cu, Mn, Ni, Zn and Pb). Also, it was important to determine the maximum level of cleanup, dependence on the voltage, the level of soil pollution for which this remediation method would prove to be effective for cleaning soil down to the permitted pollution limits, the type of soil most suitable for this remediation technique, the materials of the electrodes, etc. The electrokinetic method is most effective when cleaning polluted sand and sandy loam. The highest efficiency and smallest residual concentrations were reached when removing manganese and chromium from soil. A higher cleaning efficiency is reached applying 29 V rather than 24 V voltage. However, results were quite similar, therefore to clean soil from heavy metals the standard 24 V voltage could be applied, but the cleaning process should be prolonged.

Key words: soil, pollution, heavy metals, electrokinetic remediation, cleaning efficiency

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1. INTRODUCTION

The electrokinetic method used for remediation of soil contaminated with heavy metals is a new developing technique for cleaning soil from heavy metals and radionuclides (Jacobs, 1994; Rajeshwar, Ibanez, 1997). By feeding an electric current through soil one induces movement of charges, which is generated by migration of ions and colloids in water present in pores and by reductive reactions on the surface of electrodes. Electrodes and soil must be saturated in order to optimize transportation of electrons (Lindgren, Kozak, 1992). Movement of a pure water flow towards the cathode in soils with a negative charge is called electro-osmosis. Electromigration is the movement of ions due to an electric field, and colloid movement towards the anode is called electrophoresis. These mechanisms generate movement of ions of pollutants towards one or another electrode. The direction and velocity of the movement depends on the charge (size and polarity) of the ions and the velocity of the movement of the flow generated by electro-osmosis (Lindgren et. al., 1992). Non-ionic particles move due to the movement of the water flow generated by electro-osmosis. The electrokinetic reme-
Electrokinetic remediation method involves the installation of electrodes into the soil, and a relatively low electrical potential is applied (Lindgren, Kozak, 1992; Lindgren, et. al., 1992; Electrokinetic Remediation, 1991). The very first studies of this technique have shown that electrokinetic remediation is particularly well suited for treatment of clayey soils since such soils conduct the current well (Alshawabken et al., 2003). This is one of the advantages of the method as clayey soil is otherwise difficult to clean, because the metals bind strongly to it. The method has now been tested on soil contaminated with several different metals and has proven particularly well suited for removing copper and chromium. The metal removal efficiency generally depends on how great a percentage of the metal can be transformed into a soluble form and how rapidly (Acar, Alshawabken, 1996; Acar, Alshawabken, 1997; Shapiro, Probstein, 1993). Pollutants accumulated at electrodes can be removed in several ways, including galvanization, precipitation on an electrode, pumping of water accumulated nearby an electrode, etc. The movement direction and scope of pollutants depend on the pollutant concentration (anions versus cations), the type and structure of soil, chemical processes of the interaction and the electric current density in the water of soil pores. The soil moisture level must be above minimum to ensure an efficient cleaning process. Electrokinetic remediation is a method still currently under development. The velocity of pollutant migration depends on moisture content, the size of soil particles, mobility of ions, density of the electric current of water in pores, pollutant concentration and the total ion concentration.

Previous experiments performed by a group of scientists from Vilnius Gediminas Technical University showed that the best results of the application of the electrokinetic method for the cleanup of soil contaminated solely with lead, when lead removed from the soil was not disposed of together with the electrolyte, were achieved in the cleaning of sand – its cleanup efficiency was 73.88% at 18 V and 64.89% at 29 V. The increase of the voltage to 29 V resulted in a nearly double increase in the cleanup efficiency of sandy loam – from 37.47% to 65.04%. The experiment conducted under the conditions described above was very inefficient for treatment of loam contaminated with lead. It was mentioned before that electrokinetic remediation is most suitable for the cleanup of clayey soils, since such soils are good conductors of electric current. Such results confirm the fact that in saturated soils a better cleaning efficiency can be reached in less dense soils. Anyway, more experiments should be done in this field.

Upon the application of the voltage of 29 V for the cleanup of different types of soil (loam, sandy loam and sand), it was noted that after 30–32 hours from the start of the experiment the lead concentrations did not decrease – on the contrary, they went up in the soil part at the cathode. This can be explained by the fact that lead ions have a positive charge (Pb^{2+}), therefore, due to the potential difference, they move towards the cathode and accumulate either in the electrolyte or in the soil nearby. Such results show that if the design of the apparatus fails to ensure removal of accumulated lead, it would be inefficient, because mobile lead ions accumulate next to electrodes and stay in soil if they cannot enter the electrolyte.

It is recommended to improve the design of the apparatus to ensure the sorption of heavy metals removed from soil by zeolite and to introduce an electrolyte supply and removal system.

The aim of the article is to present an improved apparatus for the cleanup of soil contaminated with heavy metals. The main distinguishing feature of the improved apparatus and method is removal of pollutants collected near the electrodes. When secondary pollution effects will be avoided, it will then be possible to evaluate the general features of the proposed apparatus and to decide whether the method and the apparatus are effective. One of the aims of the research was to determine which type of soil and from which metals can be cleaned most effectively, what voltage should be used, etc.

2. METHODS

The tasks of the experiment were: 1) to improve the device for removing heavy metals from polluted soil; 2) to use electrokinetic remediation for the cleaning of three different soil types polluted with six different metals: chromium, copper, manganese, nickel, zinc and lead; 3) to estimate, what voltage is needed to reach the highest cleaning efficiency; 4) to estimate if 48 hours is enough to clean soil by a particular method; 5) to estimate what soil types and metals this method is the best; 6) to estimate the limits of soil pollution level within which electrokinetic remediation is still efficient; 7) to estimate natural zeolite influence on cleaning efficiency, when it is used as an absorbent absorbing heavy metal ions concentrating near the electrodes.

The apparatus was modified as follows: 1. The material of the anode was changed: a stainless steel plate was replaced with a graphite stick 50 mm in diameter. The cathode was not changed and it was stainless steel in the form of a holey plate. The materials of the feeding electrodes – graphite and stainless steel – were chosen taking into account their electrochemical properties. It is very important ensure that the selected electrode material is as inert as possible so that the particles that separate from could not find their way to enter the electrolyte.

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2. The apparatus sections containing the electrodes were filled up with an adsorbent – natural zeolite – up to 1/3 of the soil volume. Its purpose in the apparatus is to sorb heavy metals ions which, affected by the electric current, move towards the electrodes. To collect pollutants at the electrodes, natural zeolite is used. The unique properties of zeolite that determined the choice include resistance to the impact of high temperatures, aggressive media, ionizing radiation, selectivity to large cations of alkaline and some heavy metals. Zeolites are noted for their ion change and adsorption properties. Zeolite of fraction 3 to 5 mm is proposed to be used in the apparatus (Fig. 1).

3. One of the reasons for the poor soil cleanup efficiency is the fact that some of heavy metal ions get into the electrolyte solution, which in its turn is in contact with the soil being treated, and this can hinder the cleaning process. Thus, the electrolyte solution should be continuously changed. Consequently, the apparatus was improved introducing an irrigation-drainage system (Fig. 1).

Prior to the commencement of the experiment, types of the selected soil samples were identified (sand, sandy loam, clay loam), as well as their granulometric composition (i.e. particle amount and ratio per cent), moisture, and the amount of soil organic matter in the samples of each type (Table). Each test was started by artificially contaminating the soil samples. The weight of the samples was 400 g. Samples of the soil of three types were infused with 0.5 l of metal (Cr, Cu, Mn, Ni, Zn, Pb) salt solution and kept for 24 hours. Then the metal salt solution was poured off, the samples were dried, and the starting concentration of the heavy metals was calculated. Upon contamination, the dry soil samples were hydrated to a level of 15% by weight. Out of the 15% of the moisture level, 10% was distilled water and 5% was solution of sodium sulphate to increase the specific conductivity of the soil. The soil near the cathode was hydrated with an acetic acid solution to assist the formation of the acid front and to speed up the migration of lead ions. Sodium sulphate solutions were introduced to the anode and cathode compartments in equal amounts. The level of the electrolytes should be slightly lower than the soil level in the middle compartment. The experiment was conducted with three types of soil: moderately heavy loam, sandy loam and sand. Sandy loam and sand were taken respectively from the tactical fields No 1.
and No 2 of the Gaižniūnai military training area, and loam was taken from an uncontaminated environment not used for industrial activities. The input data of the experiment are given in Table. To determine the dependence of the efficiency of the electrokinetic remediation method on the voltage, the experiment was performed twice: at 29 V and 24 V. Each experiment lasted for 48 h. Samples were taken after the first 24 h following the application of the voltage to the contaminated soil. Two more series of sampling followed after 32 h and after 48 h. The samples were taken separately from each compartment (at the anode and cathode) in order to determine residual pollutant concentrations and areas of more efficient cleanup as accurately as possible.

### 3. RESULTS AND DISCUSSION

After three different types of soil (E1 – moderately heavy loam, E2 – sandy loam and E3 – sand) had been artificially polluted, it was noted that the biggest concentrations of all metals were obtained in sandy loam and the smallest in sand. It could be explained by the content of organic matter in soil, as pollutants can easily bind with the organic matter of soil and easily separate. As is seen in Table, the highest content of organic matter as well as the biggest concentrations of all six metals were estimated in sandy loam.

The concentrations of chromium in sandy loam were 12 times as high as the maximum allowed concentration (MAC) before the 1st and the 2nd test, while in moderately heavy loam they were 10 times bigger than MAC. Chromium concentrations determined in sand before the 1st and the 2nd test were respectively 4 and 4.5 times bigger than MAC.

The decrease of chromium concentrations was very even in all the soil types applying both 24 V and 29 V voltages. The distribution of chromium concentrations in different soil types after finishing the test was the same as at the beginning: the biggest concentration was determined in sandy loam and the smallest – in sand. Applying different voltages, the dynamics of soil cleaning process was almost the same in moderately heavy loam and sandy loam. Concentrations of chromium not exceeding the MAC (100 mg/kg) were obtained only for sand, though the cleaning efficiency was high enough for all types of soil (for moderately heavy loam 85%, for sand 83%, and for sandy loam 80%) (Fig. 2). It cannot be said that chromium compounds are most effectively cleaned from sand, as the initial concentration of chromium in sand was the smallest. It is likely that a sufficient cleaning efficiency could be reached in all soil types, if the duration of cleaning would be longer. Soil cleanup processes in all the soil types studied were more even at 24 V, though soil cleaning efficiency was different during different time intervals. Applying both 24 V and 29 V, the most even and efficient process was reached in the case of E1 soil type – the efficiency was 80%. After application of 24 V, the cleaning efficiency was very similar for all soil types (for moderately heavy loam 85%, for sandy loam and sand 83%) (Fig. 2). Such results of the experiment show that chromium compounds were successfully removed from all the types of soil applying both 24 V and 29 V.

### Table. Properties of soil, used for the experiment and initial heavy metals concentrations

<table>
<thead>
<tr>
<th>Type of soil</th>
<th>Granulometric composition of soil</th>
<th>Amount of organic materials in soil, mg/kg</th>
<th>Metal concentration before test No 1</th>
<th>Metal concentration before test No 2</th>
</tr>
</thead>
<tbody>
<tr>
<td>E1</td>
<td>Moderately heavy loam: sand 37%, silt 39%, clay 24%;</td>
<td>17.5 mg/kg</td>
<td>Cr – 1021.29 mg/kg</td>
<td>Cr – 995.52 mg/kg</td>
</tr>
<tr>
<td>E2</td>
<td>Sandy loam: sand 71%, silt 24%, clay 5%;</td>
<td>34.95 mg/kg</td>
<td>Cr – 1195.92 mg/kg</td>
<td>Cr – 1230.23 mg/kg</td>
</tr>
<tr>
<td>E3</td>
<td>Sand Sand: sand 99%, silt 0,5%, clay 0,5%;</td>
<td>4.6 mg/kg</td>
<td>Cr – 406.40 mg/kg</td>
<td>Cr – 453.67 mg/kg</td>
</tr>
</tbody>
</table>

Lentelė. Eksperimentui panaudoto dirvožemio savybės bei pradinės sunkiųjų metalų koncentracijos
After three different types of soil had been artificially polluted with copper compounds, its highest concentration was found in sandy loam: it was 11 times higher than the MAC. Concentrations of copper in moderately heavy loam (E1) were 750.10 mg/kg before the first and 697.13 mg/kg before the second test, i.e. 7.5 and 7 times exceeded the MAC. The initial copper concentrations were smallest in sand and were twice bigger than the MAC. Results of the study have shown that electrokinetic remediation is not relevant to clean moderately heavy loam polluted with copper (cleaning efficiency was 43% when applying 29 V and 39% 24 V). For sandy loam, 48 hours were not enough to reach a sufficient cleaning efficiency, and the further tendency of copper concentration decrease was still evident. The best results were obtained in the case of sand, as its cleaning efficiency was high (81% applying 29 V and 78% 24 V) and the residual concentrations did not exceed the MAC. Some previous researches have shown that electrokinetic remediation is most suitable to clean clay or loam, because clay is the best conductor as compared with other soil types (Acar, Alshawabeken, 1993; Alshawabkeh et. al., 1999). Some other sources indicate that electrical features of soil are not the main factor influencing the cleaning efficiency, as it strongly depends on the electrical conductivity of soil pore water and the way of pollutant particles movement. Both of these features depend on soil humidity (Probst, 1994). During the cleaning processes soil was humidified using salt solution, and sand has the most coarse grain structure as compared with other types of soil. Such features explain the highest sand cleaning efficiency (Schultz, 1997). The results of the experiment show that electrokinetic remediation is not relevant for cleanup of copper-polluted moderately heavy loam.

The biggest initial concentrations of manganese were determined in sandy loam and were moderately heavy loam. Investigations of manganese were determined in sandy loam and were twice bigger than the MAC. Results of the study have shown that electrokinetic remediation is not relevant to clean moderately heavy loam polluted with manganese (cleaning efficiency was 16% when applying 29 V and 15% 24 V). For sandy loam, 48 hours were not enough to reach a sufficient cleaning efficiency, and the further tendency of copper concentration decrease was still evident. The best results were obtained in the case of sand, as its cleaning efficiency was high (81% applying 29 V and 78% 24 V) and the residual concentrations did not exceed the MAC. Some previous researches have shown that electrokinetic remediation is most suitable to clean clay or loam, because clay is the best conductor as compared with other soil types (Acar, Alshawabeken, 1993; Alshawabkeh et. al., 1999). Some other sources indicate that electrical features of soil are not the main factor influencing the cleaning efficiency, as it strongly depends on the electrical conductivity of soil pore water and the way of pollutant particles movement. Both of these features depend on soil humidity (Probst, 1994). During the cleaning processes soil was humidified using salt solution, and sand has the most coarse grain structure as compared with other types of soil. Such features explain the highest sand cleaning efficiency (Schultz, 1997). The results of the experiment show that electrokinetic remediation is not relevant for cleanup of copper-polluted moderately heavy loam.

<table>
<thead>
<tr>
<th>Soil Type</th>
<th>Voltage (V)</th>
<th>After 24 h</th>
<th>After 32 h</th>
<th>After 48 h</th>
</tr>
</thead>
<tbody>
<tr>
<td>E1</td>
<td>24</td>
<td>32%</td>
<td>37%</td>
<td>39%</td>
</tr>
<tr>
<td>E2</td>
<td>29</td>
<td>42%</td>
<td>42%</td>
<td>35%</td>
</tr>
<tr>
<td>E3</td>
<td>29</td>
<td>60%</td>
<td>60%</td>
<td>55%</td>
</tr>
</tbody>
</table>

Fig. 2. Efficiency of chromium cleaning from soil by electrokinetic remediation

Fig. 3. Efficiency of manganese cleaning from soil by electrokinetic remediation

Fig. 4. Efficiency of zinc cleaning from soil by electrokinetic remediation
respectively 2.3 and 2.5 times bigger than the MAC before the 1st and the 2nd test. The initial manganese concentrations in loam were 1.6 and 1.7 times bigger than the MAC, while in sand they did not exceed the MAC. The cleaning process dynamics was very similar in all the three types of soil applying both 24 V and 29 V voltages: it was most intensive during the first 24 hours. Even though the biggest residual concentration of manganese was determined in moderately heavy loam (762.82 mg/kg), it did not exceed the MAC, as well as residual concentrations in sandy loam and sand. The best result was reached when cleaning manganese from sandy loam – the cleaning efficiency was respectively 96% and 94% after applying 29 V and 24 V (Fig. 3). The least efficiency was reached in the case of moderately heavy loam, and it was 68% after applying 29 V voltage and 64% with 24 V; nevertheless it was enough to reach sufficient soil cleanup (Fig. 3). It can be stated that electrokinetic remediation is undoubtedly suitable to clean soil from manganese, applying both 24 V and 29 V voltages.

Like for the other metals studied, the biggest concentrations of nickel were determined in sandy loam and the smallest in sand. Nickel concentrations in sandy loam were 9 and 8 times bigger than the MAC. In all three types of soil the cleaning process was more or less even applying both voltages; however, no stabilization of the process was reached. It is likely that extension of process duration would ensure better results. Initial concentrations of nickel 5 to 9 times exceeded the MAC (75 mg/kg), but no desirable cleaning effect was reached in any case, i.e. residual concentrations still exceeded the MAC. It is likely that electrokinetic remediation is not suitable for cleanup of soil polluted with nickel compounds. The best cleaning efficiency was reached after applying 29 V to clean sandy loam and loam: the cleaning efficiency was respectively 82% and 71%. Sand cleaning efficiency was worst (49%). When 24 V was applied to loam, sandy loam and sand, cleaning efficiency was respectively 64%, 78% and 47%.

Most intensively zinc was cleaned from all the types of soil during the first 24 h applying both selected voltages. The next 8 h the concentration of zinc in soil was almost constant, and a rapid decrease started only after 32 h of experiment, i.e. when soil was artificially acidified with acetic acid. The initial concentrations of zinc in sandy loam and moderately heavy loam were respectively 4 and 2.4 times higher than the MAC, while zinc concentration in sand was almost the same as the MAC which is 300 mg/kg. The cleaning efficiency of sandy loam and sand was very similar when applying 29 V (respectively 85% and 89%), but the initial concentration in sand was several times smaller than in sandy loam (Fig. 4). Applying 24 V, the highest efficiency was reached in sandy loam (86%) and sand (79%) (Fig. 4). The residual concentrations of zinc in sandy loam did not exceed the MAC, while in sand they were almost equal to the background one. The residual concentrations of zinc in loam equalled the MAC, but it is likely that they could decrease if the time of the experiment would be extended.

Lead is one of the main metals in the composition of different munitions, so it is especially important to determine whether electrokinetic remediation can be efficient when cleaning lead from soil. Unfortunately, the experiment showed the worst results in case of lead cleanup from soil. Like those of the other metals, the biggest concentrations of lead were determined in sandy loam and moderately heavy loam and the smallest in sand. The process of lead cleaning from sandy loam and moderately heavy loam was very even. The concentrations were gradually decreasing during all the experiment. The cleanup of sand was most intensive during the first 24 h, while later the concentrations of lead remained almost the same. The biggest initial concentration as well as the highest cleaning efficiency (68% applying 24 V and 70% applying 29 V) was determined in sand (Fig. 5). This was the only case when concentrations not exceeding the MAC were reached. The cleaning efficiency of sandy loam and moderately heavy loam was very similar apply-
ing both 29 V and 24 V and was in the range of 57%–60% (Fig. 5). The residual concentrations of lead in sandy loam and loam were 2–3 times bigger than the MAC. It can be stated that such a small difference in the voltages doesn’t have a great impact on the cleanup process. A considerably greater value can be ascribed to the duration of the cleaning process and to the specific features of the soil. For an effective cleanup of soil from lead compounds, the conditions of the cleanup process should be optimized.

When cleaning three types of soil polluted with six different metals, in all cases a higher cleaning efficiency was reached in the anode zone, possibly because lead ions have a positive charge (Pb⁺) and due to the potential difference move towards the cathode and accumulate either in the electrolyte or in the soil nearby. It is likely that the extension of the cleanup process duration would enable equalization of concentrations in the cathode and anode sections and the removal of all mobile lead ions would be guaranteed.

Electrokinetic soil remediation is most relevant to clean sandy loam and sand. This might be explained by the fact that cleaning efficiency strongly depends on the electrical conductivity of soil pore water and the way of pollutant particle movement. Sandy loam and sand have a most coarse grain structure as compared with other types of soil, which ensures a better movement of metal ions and thus better results of cleaning. The results of the experiment showed that in all cases the efficiency of soil cleanup was higher 29 V. In this case, processes in soil were more intensive and the desirable effect was reached faster. Anyway, it is possible that extension of cleaning duration at 24 V would give the same results.

4. CONCLUSIONS

1. The electrokinetic method is most effective to remove heavy metals from polluted sand and sandy loam. For these soil types, the highest cleaning efficiency was reached (ranging from approximately 60% to 90% for sandy loam and 70% to 80% for sand, except cases of nickel and lead, as for these metals electrokinetic remediation appeared to be inefficient).

2. The highest efficiency and the lowest residual concentrations were reached for manganese and chromium (70% to 90%).

3. Cleaning efficiency is better applying 29 V rather than 24 V. However, the results were rather similar, therefore to clean soil from heavy metals the standard voltage of 24 V could be applied, but in this case the cleaning process should be prolonged.

References


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SUNKIAISIAIS METALAI UŽTERŠTO DIRVOŽEMIO VALYMAS ELEKTROKINETINIU METODU

Santrauka
Elektrokinetinio metodo panaudojimas sunkiaisiais metalais užteršto dirvožemio valymo nuo sunkiųjų metalų ir radionuklidų technologija. Pro dirvožemį leidžiant elektros srovę vyksta krūvių judėjimas, kurį sukuria jonų ir koloidų, esančių dirvožemio porose, migracija bei redukcinės reakcijos ant elektrodų paviršiaus. Tyrimo tikslas buvo nustatyti, ar elektrokinetinis dirvožemio valymo metodas yra efektyvus ir tinkamas sunkiaisiais metalais (Cr, Cu, Mn, Ni, Zn ir Pb) užterštiems dirvožemiams valyti. Valymo metu teršalių dalelės yra priverčiamos judėti į ir pro sluoksnį, sudarytą iš laidžios sorbuojančios medžiagos, sudrėkintos laidumą didinantį skysčiu. Metodo efektyvumas ne tiek priklauso nuo dirvožemio laidinių savybių, kiek nuo porose esančio vandens elektrinio laidumo ir dalelių judėjimo kelio. Taip pat buvo svarbu nustatyti išvalymo efektyvumą, jo priskaičiavimą nuo įtampos, valymo metodo ribas (kokia užterštumo dirvožemio porose esantis efektyvias, kokiu metodu dirvožemio tipui metodas yra tinkamiausias ir pan). Tyrimo rezultatai rodo, kad metodas efektyviasias, kai valomas sunkiaisiais metalais užterštas smėlis ir priesmėlis. Didžiausias efektyvumas ir mažiausios liekamosios koncentracijos buvo pasiektos valiant is dirvožemio manganą ir chromą. Didesnis efektyvumas visais atvejais buvo pasiekiamas, kai įtampa buvo 29 V, tačiau ir esant 24 V, ir prie 29 V įtampai gauti rezultatai buvo labai panašūs.