Electrokinetic Remediation of Sand Contaminated with Copper and Chromium

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Abstract: The electrokinetic remediation of copper and chromium from artificially contaminated sand has been studied. In order to achieve higher remediation efficiencies Selective Cation Membrane and nylon mesh with filter paper membranes, pumping out and refilling of the conductive solution as well as anode system has been used in this study. All those methods help to prevent anions from the cathodic compartment from penetrating into the anodic and the soil system and do not allow to impair the remediation process. In addition, several anode system helps to reduce remediation time as at the same time to raise the efficiency of the overall remediation process. 10 laboratory experiments were conducted to remove copper (II), chromium (III) and chromium (VI) from the artificially contaminated lake sand. Obtained results show that efficiencies higher than 90% may be achieved.

Keywords: remediation, soil, copper, chromium, membrane, anode system

1 Introduction

During the past decades, several solutions of efficient contaminants removal from soils were investigated and it is strongly believed that they will help to solve groundwater as well as soil pollution.

Electrokinetic soil remediation method is an emerging technology, which has attracted increased interest among scientists and governmental officials in the last decade, due to several promising laboratory and pilot scale studies and experiments. This method aims to remove heavy metal and organic as well as radioactive contaminants from low permeability contaminated soils under the influence of an applied direct current.

According to conducted experiments and pilot scale studies, metals such as lead, chromium, cadmium, copper, uranium, mercury and zinc as well as polychlorinated biphenyls, phenols, chlorophenols, toluene, trichlorethane and acetic acid are suitable for electrokinetic remediation and recovery (1 — 5).

Upon contaminant migration to the electrodes, they may be removed by electroplating, precipitation / co-precipitation, pumping near the electrode, or complexing with ion exchange resins.

Electrodes inert to anodic dissolution should be used during the remediation process. Most suitable electrodes used for research purposes include graphite, platinum, gold and silver. The pH of the soil should be maintained low enough to keep all contaminants in dissolved phase.

2 Experimental part

If there are appropriate pH conditions, heavy metals are likely to be adsorbed onto the negatively charged soil particles. Main sorption mechanisms include adsorption and / or ion exchange.

Contaminant removal efficiencies are highly dependent on soil moisture content, soil grain size, ionic mobility, pore water amount, current density, and contaminant concentration (6 — 11).

A set of several experiments were conducted in Oulu University in June 2001 — December 2002. The aim of the experiments was to construct an ideal electrokinetic cell and to achieve maximum removal
efficiencies in removing copper and chromium from selected soils — pure washed and Kuivasjärvi lake sand.

3 Materials

2 plastic containers with three lines of holes cut in the connecting walls; membranes; lake sand; 0.01 M KNO₃; copper nitrate; distilled water; pH meter designed for soils and pH meter designed or solutions; electric power supplies; DC/AC meters; graphite electrodes.

4 Results

Experiments were performed using electrokinetic cell with Selective Cation or nylon mesh and filter paper membranes and without adding any additional chemicals, i.e. any additional enhancement agents. Kuivasjärvi lake sand was artificially contaminated with copper nitrate in experiments 1 — 7, with kalium bichromate in experiments 8 — 9 and chromium nitrate in experiment 10. Contaminants were evenly distributed along the soil matrix and placed into the plastic container, i.e. electrokinetic cell.

Experiments 1 — 3 and 8 — 10 were conducted with one graphite anode and one graphite cathode and Selective Cation membrane, experiments 4 — 7 were carried out with graphite anode system made of 3 different geometrically placed electrodes and one cathode as well as with Nylon mesh and filter paper membrane. Distance between electrodes in 1 — 7 and 10 experiments was 27 cm and in 8 and 9 — 28cm and 28.5 cm respectively. Length of solution in all experiments was 27 cm.

Table 1 presents removal efficiencies of 10 experiments.

<table>
<thead>
<tr>
<th>Experiment No</th>
<th>Contaminant removed</th>
<th>Duration of experiment (h)</th>
<th>Initial concentration (mg/kg)</th>
<th>Concentration left in the sand</th>
<th>Removal efficiency (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Cu (II)</td>
<td>8</td>
<td>300</td>
<td>77.2</td>
<td>74</td>
</tr>
<tr>
<td>2</td>
<td>Cu (II)</td>
<td>8</td>
<td>300</td>
<td>22.87</td>
<td>92</td>
</tr>
<tr>
<td>3</td>
<td>Cu (II)</td>
<td>40</td>
<td>300</td>
<td>70.3</td>
<td>77</td>
</tr>
<tr>
<td>4</td>
<td>Cu (II)</td>
<td>26</td>
<td>300</td>
<td>44.4</td>
<td>85</td>
</tr>
<tr>
<td>5</td>
<td>Cu (II)</td>
<td>26</td>
<td>300</td>
<td>74.8</td>
<td>75</td>
</tr>
<tr>
<td>6</td>
<td>Cu (II)</td>
<td>26</td>
<td>300</td>
<td>21.5</td>
<td>93</td>
</tr>
<tr>
<td>7</td>
<td>Cu (II)</td>
<td>26</td>
<td>300</td>
<td>51</td>
<td>83</td>
</tr>
<tr>
<td>8</td>
<td>Cr (VI)</td>
<td>167</td>
<td>300</td>
<td>5.75</td>
<td>98</td>
</tr>
<tr>
<td>9</td>
<td>Cr (VI) and Cu (II)</td>
<td>191</td>
<td>300</td>
<td>23.45</td>
<td>92</td>
</tr>
<tr>
<td>10</td>
<td>Cr (III)</td>
<td>32</td>
<td>300</td>
<td>10.55</td>
<td>96</td>
</tr>
</tbody>
</table>

Removal efficiencies in experiments 1, 3 and 5 are not as high as in other experiments. It may be due to the fact that after some time the solution gets saturated and the excess ions return back to the soil. Another reason is that when the pH drops too low, back diffusion may take place.

In experiments 8 and 9 enhancement agent, i.e. acetic acid and pumping as well as refilling of the conductive solution was introduced. Removal efficiencies in experiments 8 and 9 are considerably higher than in the rest of experiments. When the solution gets saturated, i.e. more and more ions migrate from the sand into the solution, concentration of ions may become too high and excess ions return back to the soil. It is possible to imply that refilling of conductive solution, i.e. removal of excess ions helps to raise the overall efficiencies of the process. Very high removal efficiencies were obtained in the experiment.
where chromium was simultaneously removed with copper. It would be possible to state that those two ions enhance each other removal from the sand, however, this statement needs to be further investigated.

In experiments 4 and 7, two graphite anodes were used in the process. The distance between those two anodes was 3 cm and 2 cm. The removal efficiency is considerably high, which may be due to the enhanced anodes ability to remove contaminants from higher area.

Experiment 5 was conducted with five anode system and obtained removal efficiency was 75%. Despite the enhanced electrode area, the removal efficiency was not very high in comparison to other experiments. Low obtained efficiency may be explained that every anode is blocking one another field of influence and contaminant ions remain in the soil and do not migrate to the cathode.

In experiment 6, three anodes were used to assess the remediation efficiency of the process. The removal efficiency was 93%, which indicates that three anodes lined up uniformly helps to build a higher electrode surface area and hence, helps to raise the overall efficiency of the process.

**pH values in certain measurement point in the selected experiment**

pH distribution was measured in all 10 experiments, however data is presented only from 6 experiment. It represents the whole experimental results obtained in this study and show pH distribution for Cu (II) ionic specie. Figure 1 presents the pH measurement point A in the soil.

![Fig.1 pH measurement point A in the contaminated sand (view of electrokinetic cell from above)](image)

It is necessary constantly monitor pH changes in the electrokinetic cell because heavy metal species are very sensitive to pH changes, which affect their solubility and removal potentials. As some metal species favour low and other favour high pH environment, pH should be maintained according to the chosen metal specie characteristics.

Figure 2 presents the pH distribution in the soil in the experiment 6, where three anodes were used for the remediation process.

![Fig.2 pH distribution in the measurement point A in the soil during 6th experiment](image)

In this experiment, three anode system was introduced into the electrokinetic remediation system and copper ions were aimed to remove from sand. The initial pH of the system was 7.62 and the pH after 26 hours of remediation was 3.60. Acidic media in the measurement point A in the soil indicates that copper ion mobility has not been impaired and hence remediation of 93% has been reached. It is important to indicate that no any additional enhancement agents were added to the electrokinetic system. In addition, Selective Cation Membrane has been replaced by nylon mesh and filter paper membrane.

5 Conclusions

According to obtained results it is possible to conclude that the proposed electrokinetic cell and its design has demonstrated the ability to remove heavy metals from artificially contaminated sand under the
applied direct electric current. The usage of either Selective Cation or nylon mesh and filter paper membrane stops OH ions from penetrating into soil and raising the pH of the system, hence do not allow to impair the removal of metal species from the soil. The addition of enhancement solutions and employment of membrane helps to avoid sharp pH jump in the system thus raising the remediation efficiencies of the treated contaminants. Pumping out and the refilling of the conductive solution have considerably raised the remediation efficiency.

The usage of the anode system instead of one anode and one cathode has demonstrated the potential to improve the efficiency of the remediation process. Nevertheless, the layout and the importance of the anode system should be further investigated and improved.

References