

An Electrokinetic Process Coupled Activated Carbon Barrier for Nickel Removal from Kaolinite

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Abstract

Electrokinetic (EK) remediation coupled with activated carbon barrier was evaluated to remove nickel (500 mg/kg) from kaolinite. Laboratory experiments were performed by applying a constant voltage to create electric field strength of 1 or 1.25 V/cm for 3 days. Findings showed that the barrier filled with activated carbon could prevent the formation of reverse electro-osmotic flow, which had an adverse effect on the Ni(II) removal. Application of activated carbon barrier into EK process resulted in an increase of Ni migration from 11 to 47%.

Keywords: activated carbon; barriers; electrokinetic; nickel; soil remediation

1. Introduction

Anthropogenic activities, improper hazardous waste management and handling techniques in the past have made heavy metals, organic compounds, and other hazardous materials pollution in sites, creating an enormous adverse impact on the quality of groundwater, soil, and associated ecosystems (Virkutyte et al., 2002; Nouri et al., 2009). Electrokinetic (EK) remediation process is one of the emerging technologies to remediate metal-contaminated soils with low hydraulic permeability (Virkutyte and Sillanpaa, 2007; Chung and Lee, 2007; Reddy and Saichek, 2003; Zhu et al., 2009). Basically, this technique is based upon the application of an electric field to the soil, sludge, and sediments, either by conducting a direct current or a constant voltage (Virkutyte et al., 2002; Amrate et al., 2005; Akretche, 2002). The application exploits two main transport processes of ions in solution: electro-osmosis and electro-migration (Lynch et al., 2007; Alshawabkeh and Acar, 1992). Wide range of parameters such as ionic concentration, pH condition, and zeta potential affect electro-osmotic (EO) flow during the transport of dissolved contaminants (Lynch et al., 2007). Due to the negative surface charge of water-saturated clays, the direction of EO flow is from anode to cathode (Virkutyte et al., 2002; Vane and Zang, 1997). Electrolysis has great effect on EK process (Virkutyte et al., 2002; Amrate *et al.*, 2005):

Anode:

$$H_2O \rightarrow 2H^+ + 1/2 O_2(g) + 2e^-$$
 (1)

Cathode:

$$2H_2O + 2e^- \rightarrow 2OH^- + H_2(g)$$
 (2)

Counteracting EO flow and higher ionic mobility of H^+ which is about 1.76 times that of OH⁻, make the advance of base front slower than the advance of the acid front. Consequently, the chemistry across the specimen is dominated by acid front, except for small sections close to the cathode (Acar *et al.*, 1990; Alshawabkeh and Acar. 1992; Probstein and Hicks, 1993).

As recommended by Sims (1990), both kaolinite and clay have low hydraulic conductivity, reducing redox potential, slightly alkaline pH, which is suitable for the remediation of heavy metal using EK process. In addition, the low acid-base buffering capacity of kaolinite also contributes to the higher metal removal efficiency (Hamed *et al.*, 1991; Hicks and Tondorf, 1994).

The electrokinetic can be used in combination with other cleanup techniques (Chung and Lee, 2007; Lynch *et al.*, 2007). Ground water remediation using permeable reactive barriers (PRB) is a technology developed in the early 1990s (Simon and Meggyes, 2000). PRBs, when contains reactive substances in the barrier, could reduce contaminants in groundwater. The advantages of this technique mostly are in-situ operation and effectiveness for a variety of contaminants (Milligan *et al.*, 2001). EK process coupled with PRB (EK/PRB) is an innovative soil remediation technique developed recently. Shariatmadari *et al.* (2009) investigated the application of nano zero-valent iron (nZVI) as PRB coupled with EK to remediate Cr(VI)-contaminated soil. They reported an 88% of Cr reduction after the soil received the EK/PRB treatment. The effectiveness of incorporating zero-valent iron (ZVI) into EK to remediate hyper-Cr(VI) contaminated clay is also reported (Weng *et al.*, 2007). Moreover, Chand and Cheng (2006), reported high arsenic removal of using EK coupled with PRB in soil matrix. In addition, Yuan and Chiang (2008) applied the same approach to remediate perchlorethylene contaminated soil.

Activated carbon (AC), due to its high efficiency/ cost ratio, is the most common adsorbent used in water and wastewater treatment processes (Hasar, 2003; Kadirvelu et al., 2001; Erdogan et al., 2005). Previous studies on the EK removal of Ni from soils mostly exhibited low removal efficiencies (Maturi and Reddy, 2006). Al-Hamdan and Reddy (2008) found that a portion of nickel precipitated in kaolinite which had adverse effect on nickel removal. They concluded that increasing test duration made more nickel migration toward cathode, thus improved removal efficiency. A pervious study Reddy and Chinthamreddy (1999) showed that negligible amount of nickel (less than 1%) migrated to cathode compartment and most of it remained in the soil accumulated near the cathode. They found that using reducing agents like sulfide could affect the metal migration. Kim et al. (2008) used nitric acid to enhance nickel removal in an EK process. They found that pretreatment of soil by mixing contaminated soil with HNO₃ could increase nickel removal. In general the nickel removal in conventional EK process was not promising. Some researchers claimed that an occurrence of reverse EO in heavy metals removal during EK process exhibited an adverse effect on metal migration efficiency (Kim et al., 2008; Genc et al., 2008; Weng et al., 2007). The method to avoid this reverse EO remains suspended issue. As reported by Lynch et al. (2007), ion concentration and sorption capacity of the soil may affect EO flow direction to some extent. Because the AC has high metal adsorption capacity, it seems that the use of AC as a barrier can increase the metal removal efficiency as the metals pass through the barrier. In the present study, the applicability of an EK coupled with AC barrier in prevention the formation of reverse EO flow and probable improvement in remediation of nickel contaminated kaolinite was investigated.

2.1. Materials and electrokinetic experiments set up

Kaolinite used in the experiments was obtained from Marand clay company, Tabriz, Iran. Approximately 98% of the kaolin has particles size smaller than 20 μm. The pH of the kaolin slurry, prepared from kaolin mixed with distilled water in the ratio of 1:1 (w/w), was 8.2. The kaolin was artificially contaminated by nickel nitrate solution to achieve a Ni contamination of 500 mg/kg. The contaminated kaolin was left at room temperature, 25°C, for 12 h before it was packed into the reactor cell (Fig. 1) to attain equilibrium. The granular activated carbon was purchased from Merck Company. The maximum adsorption capacity of this kaolinite was 2.71 mg/g for Ni(II). The specific surface area of AC was measured by N2 absorption isotherm using a Gemini 2375 Micromeritics instrument by Brunauer-Emmett-Teller (BET) method. Specific surface area and bulk density of AC were 891m²/g and 2.106 cm³/g, respectively. Yavuz et al. (2003) have obtained an adsorption capacity of 2.8 mg/g for Ni(II) on kaolinite. In the present study the maximum adsorption capacity was 5.42 mg/g for Ni(II) onto activated carbons. Laboratory scale electrokinetic experiments were conducted in a rectangular $30 \times 12 \times 10$ cm (length, height, and width, respectively) plexiglass container, the length of the soil specimen was 15 cm (Fig. 1). KNO₃, 0.05 M was chosen as a conductive solution allowing current to pass through soil and to facilitate the ions migration (Virkutyte and Sillanpaa, 2007). The barrier containing activated carbon was either placed in the electrode compartment, the soil beside the cathode and the soil in the middle of the sample in different experiments.

A direct current (DC) power supply was used to generate constant electric gradient of 1 or 1.25 V/ cm for 3 days. The electrode plates used in this study is perforated stainless steel electrodes (A316) and was made by Foolad Mobarakeh Sepahan, Iran. The current fluctuations were monitored during the experiments. Electrode plates (0.6 mm thicknesses and 12×10 cm length) were placed directly into the soil (Fig. 1). New electrodes were used for each test to avoid any cross-contamination between the tests. The testing box and valves were soaked in a dilute hydrochloric acid solution for 24 h, and then rinsed with distilled water before starting the experiment. Also, all experiments were performed in duplicate to ensure the results accuracy. Voltage applied, test durations and the AC barrier locations in different experiments in this study are presented in Table 1.

2. Materials and methods

3. Analyses



Figure 1. Schematic representation of electrokinetic cell, PRB thickness is 0.5 cm

Test number	Voltage applied (V/cm)	Test duration (days)	s) Barrier's location	
1	1	3	No barrier	
2	1	3	in the electrodes compartment	
3	1	3	in the soil beside the cathode	
4	1	3	in the soil at the middle of the sample	
5	1.25	3	No barrier	
6	1.25	3	in the electrodes compartment	
7	1.25	3	in the soil beside the cathode	
8	1.25	3	in the soil at the middle of the sample	

The soil specimen was sectioned into five after the completion of each test and dried at 42 °C to a constant weight. After homogenizing, samples from each portion were acid digested according to U.S.EPA 3050B method (EPA, 1992). The Ni concentration was measured using an atomic absorption spectrometry (Buck Scientific 210VP). The pH value of the soil samples was measured at the end of the experiments, using a pH-meter. The amount of Ni adsorbed by AC barrier was also measured at the end of each test.

4. Results and discussion

4.1. Variations in anode, cathode and soil pH

In the tests without AC barrier and tests with the barrier in the electrodes compartment (tests 1, 2, 5 and 6) pH value of the anode and cathode chambers significantly changed. However, in tests 3, 4, 7 and 8 (with barrier in the soil specimen), the pH of the anode chamber did not change significantly. Fig. 2 shows the pH profile along the cell at the end of each test. In the

experiments without barrier (Tests 1 and 5), pH profile became acidic near the anode sections and became highly basic near the cathode sections. However, in the experiments with barrier (Tests 2, 3, 4, 6, 7, and 8) the soil pH did not change significantly.

When electricity is applied to the soil, electrolysis of water takes place at the electrodes where oxygen and hydrogen ions are produced at the anode and hydrogen and hydroxide ions are produced at the cathode (Eqs. 1 and 2). Therefore, in the experiments without barrier (Tests 1 and 5), the anode and cathode chambers pH became respectively highly acidic and basic regarding the production of ions. Moreover, the development of acid and base fronts along the soil can also lower soil pH. Figure 2a shows the decrease in soil pH near the anode and increase it near the cathode. In the experiments with barrier in the electrode compartment (Tests 2 and 6), the acid and base front could not develop successfully along the soil profile; therefore, the soil pH did not change significantly. The reason may be related to water electrolysis half-reaction interruption by the presence of AC in the reservoirs. The other affecting



Figure 2(a). Distribution of pH after electrokinetic treatment, where x is a distance from anode and L is a length of soil at the end of experiments No. 1, 2, 5, and 6



Figure 2(b). Distribution of pH after electrokinetic treatment, where x is a distance from anode and L is a length of soil at the end of experiments No. 3, 4, 7, and 8.

factor may also be the buffering capacity of kaolinite. The anode chamber pH did not change significantly during the Tests of 3, 4, 7, and 8. In these experiments, acid front could not develop in the system. Since the activated carbon is a conductor of electricity, placing a layer of activated carbon in the soil is equivalent to placing additional electrode. When the carbon layer is placed in the middle of the soil sample, it functions as a cathode for the anode at the end and as an anode for the cathode at another end. However, using AC barrier may interrupted electrolysis. During the EK experiments Ni ions precipitate by accepting electrons and hydrogen ions produced by electron release during water electrolysis. These half-reactions are in balanced. By using AC barrier, it seems that nickel ions preferably adsorb onto AC rather than precipitation. Therefore the adsorption tendency of Ni may disturb precipitation. It seems that in such systems, pH value changes in cathode and anode may be affected by other competitive reactions like adsorption, although the effect of carbon

active barrier on anode pH is not well understood and should be further investigated.

4.2. Electroosmotic transport

Fig. 3 shows the accumulation of EO flow collected during the experiments. In the Tests of 1 and 5 (tests without barrier), the EO flow was reversed at the end of the second day and the remediation process was interfered. However, in the experiments of carbon barrier (Tests 2, 3, 4, 6, 7, and 8) reverse EO flow did not occur and the direction of EO was all toward the cathode.

The surface charge of hydrolyzed clay particles is typically negative. Electroosmotic flow moved from the anode to cathode due to the existence of negative charge surface of the clay particles. Since the pH_{zpc} value of kaolinite is 4.6 (Stumm and Morgan, 1996), it is expected that the direction of EO flow would be toward cathode at soil pH > pH_{zpc}. As described by



Figure 3(a). Variation of the electroosmotic flow during experiments No. 1, 2, 5, and 6



Figure 3(b). Variation of the electroosmotic flow during tests No. 3, 4, 7, and 8.

Weng *et al.* (2007) reported that developing reverse EO during chromium contaminated clayey soil remediation, mainly attributed to anions originated from the soil containing high Cr(VI) concentration and faster migration of the anions than cations. It seems that in the present study the reverse EO has occurred due to the presence of high amount of nitrate ions. Hence, the reverse EO was found at the end of the second day (Fig. 3). However, in other experiments when activated carbon barrier with high sorption capacity (5.42 mg/g) was used, probably due to the increase in sorption capacity (Virkutyte *et al.*, 2002) the reverse EO did not happen (Fig. 3)

4.3. Nickel migration

Fig. 5 shows the Ni concentration, normalized with respect to the initial Ni concentration of the soil (500 mg/kg), in each of the five sections of the soil sample at the end of the tests. As shown, the Ni migrated from the anode and accumulated in the sections near the cathode. When an electrical gradient of 1.25 V/cm was applied, the nickel migration from the anode toward the cathode was more significant than the one with 1 V/cm.

When the electrodes compartment and the middle of the soil sample contain barrier (Tests 2, 4, 6 and 8), the migration of Ni(II) was less than the other tests. When barrier was placed in the soil beside the cathode, Ni migration was higher than the experiments without barrier. When the Ni concentration after EK experiment in each segment is less than initial concentration, the remediation has been done successfully. Therefore, the more soil length was remediated, the more performance was achieved. To cover the soil sample length and Ni migration as performance index, the amount of Ni migration from 60% of soil sample length has been selected to compare the performance of EK in all experiments. For example, the Nickel migration in 60% of soil length in Test 7 was about 47%. However, the nickel migration in the Test 5 was about 36%. Pervious works on nickel removal from kaolinte without enhancements reported low nickel removal (Kim et al., 2008; Maturi and Reddy, 2006). Less than 1% nickel removal reported by Chinthamreddy (1999) in basic EK as well as with application of reducing agents. According to Al-Hamdan and Reddy (2008), when kaolin was loaded with Ni²⁺ some parts of nickel precipitate as Ni(OH)₂. The amount of precipitation

depends on soil pH. It seems that the relatively low nickel removal was resulted from nickel precipitation due to high initial soil pH (8.2) in the present study. Applying a constant voltage of more than 1 or 1.25 V/ cm to the system would generate stronger acid front and consequently increase nickel removal efficiency.

Changes in the pH profiles in the soil had a great influence on Ni migration. Indeed, experiments conducted in the present study showed that pH has a significant impact on Ni migration in the soil. Due to the low pH at the anode, there was high concentration of Ni found close to the anode. However, when the normalized distance from the anode reached 0.5, Ni concentration significantly increased (Fig. 4). For example, about 80% of Ni migrated from the first section toward the other sections in Tests 5 and 7.

High pH value indicated that some OH⁻ ions were reaching the soil and possibly interfering with the mobility of nickel ions. The high pH situation was favorable to the formation of Ni(OH)₂ near the cathode sections and therefore decreased the soil conductivity. Due to this fact, the high Ni concentration was found near the cathode sections. When barrier was installed in the middle of the soil and placed in the electrodes' compartment, resulted in not significant changing in soil pH and a gap which carbon barrier made it between soil length, Ni migration was less than the other experiments (Fig. 4).

4.4. Mass balance

An approximate mass balance was calculated for Ni in the EK system in comparison to Ni in the spiked soils (Table 2). Saichek and Reddy (2003) suggested that several discrepancies in the mass balance of an EK system could be attributed to the detection limits in the chemical analyses, contaminant adsorption to the electrokinetic cell walls and sample bottles. There is significant amount of Ni retained in the barrier indicating that carbon active could adsorb Ni. Moreover, applying higher voltage would result in increasing Ni adsorption by carbon active barrier.

5. Conclusions

In this study, application of AC barrier to remove nickel from contaminated kaolinite was investigated. Laboratory scale experiments showed that the use of activated carbon barrier could hinder reverse electroosmotic flow and could adsorb migrating nickel. Barrier



Figure 4(a). Normalized Ni concentration at the end of electrokinetic experiments Tests 1, 2, 5, and 6



Figure 4(b). Normalized Ni concentration at the end of electrokinetic experiments Tests 3, 4, 7, and 8

Experiment no.	Barrier's location	Initial Segment No.					- Catho	Activated	
		total amount	1	2	3	4	5	dereservoir (g)	carbon adsorption (g)
1	No barrier	1.2388	0.1133	0.2757	0.2815	0.3422	0.3457	0.00523	
2	in the electrodes compartment	1.2388	0.2394	0.2827	0.3119	0.3294	0.3247	0.00081	0.0135
3	in the soil beside the cathode	1.2388	0.2313	0.3002	0.2733	0.3294	0.3481	0.00096	0.0064
4	into the soil in the middle of the sample	1.2388	0.1243	0.3250	0.2385	0.2709	0.3905	0.00251	0.0090
5	No barrier	1.2388	0.0600	0.1925	0.3810	0.3932	0.4006	0.00601	
6	into the electrodes compartment	1.2388	0.2425	0.3114	0.3520	0.3615	0.4013	0.00091	0.0274
7	into the soil beside the cathode	1.2388	0.0716	0.2236	0.2385	0.3790	0.3932	0.00184	0.0235
8	into the soil in the middle of the sample	1.2388	0.1756	0.3277	0.1851	0.3506	0.3790	0.00385	0.0556

Table 2. Mass balance calculated for laboratory scale experiments (g)

location is an important factor determining Ni migration in the EK process. In general, the Ni migration in the experiments with AC-PRB was higher than experiments without barrier. When barrier was installed in the soil close to cathode, more Ni migration occurred during electrokinetic process than those experiments without barrier. In this case, when 1.25 V/cm was applied in the presence of barrier, the Ni removal was about 47% without occurrence of reverse EO. However, further investigations need to be conducted to find the effect of carbon active barrier on anode pH. Effect of carbon active barrier on Ni removal during EK process in lower Ni concentrations in soils also needs to be investigated.

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