

In situ removal of copper from sediments by a galvanic cell

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Abstract

This study dealt with *in situ* removal of copper from sediments through an electrokinetic (EK) process driven by a galvanic cell. Iron (Fe) and carbon (C) were placed separately and connected with a conductive wire. Polluted sediments were put between them and water was filled above the sediments. The galvanic cell was thus formed due to the different electrode potentials of Fe and C. The cell could remove the pollutants in the sediments by electromigration and/or electroosmosis. Results showed that a weak voltage less than 1 V was formed by the galvanic cell. The voltage decreased with the increase of time. A slight increase of sediment pH from the anode (Fe) to the cathode (C) was observed. The presence of supernatant water inhibited the variation of sediment pH because H^+ and OH^- could diffuse into the water. The removal of copper was affected by the sediment pH and the distribution of electrolyte in sediment and supernatant water. Lower pH led to higher removal efficiency. More electrolyte in the sediment and/or less electrolyte in the supernatant water favored the removal of copper. The major removal mechanism was proposed on the basis of the desorption of copper from sediment to pore solution and the subsequent electromigration of copper from the anode to the cathode. The diffusion of copper from sediment to supernatant water was negligible.

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1. Introduction

Pollutants from various sources, including industrial, mining, municipal sewage, agricultural and other activities, have entered into the waters in rivers and lakes. With the flowing of the polluted water, most pollutants such as heavy metals are accumulated in the bottom sediments via adsorption. When the characteristics of supernatant water changed, pollutants in the sediments will be released into water and threaten ecosystem (Seidel et al., 2004). Sediments are long-term source of pollutants and pose potential risks to fish, human and animals (Brenner et al., 2002). For ecological and economic reasons, feasible technologies, which can remove pollutants from contaminated sediments such that the sediments can be restored for beneficial use, are urgently needed.

Most of the current sediment remediation technologies are *ex situ* methods (Mulligan et al., 2001). Generally, the polluted sediments are dredged up and treated on land. These processes need large labor and poses great risk exposures to workers. So, *in situ* technologies are more favorable because they avoid the dredging and transporting process. *In situ* methods include capping, solidification/stabilization, chemical treatment, biological treatment and so on (EPA, 1993). Capping covers contaminated sediments with cleaner sediments with or without lateral walls (Azcue et al., 1998; Stüben et al., 1998). It consumes much clean sediment and pollutants are not removed from the site. Solidification/stabilization and chemical treatment may lead to secondary pollution and it is difficult to ensure the complete mixing of treatment reagents with the contaminated sediments (Varjo et al., 2003; Reitzel et al., 2005). Biological treatment needs special microorganisms and is generally effective for the remediation of organic contaminated sediments (Duba et al., 1996; Dybas et al., 1998; King et al., 2006). Consequently, it is urgent to develop effective and economical *in situ* sediment remediation processes.

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Sediments are clayed, so the methods that treat permeable soils are no longer applicable. Electrokinetic (EK) technology was particularly developed to remediate clayed soils (Acar and Alshawabkeh, 1993; Probststein and Hicks, 1993). EK method would be effective for the remediation of sediments. With the application of a weak electric field, pollutants in clayed soils are removed by electromigration, electroosmosis and electrophoresis (Acar and Alshawabkeh, 1993; Probststein and Hicks, 1993; Virkutyte et al., 2002). The great potential of EK process for the removal of heavy metals and organic compounds from contaminated soils has been proved in the past years (Shapiro and Probststein, 1993; Lageman, 1993; Lageman et al., 2005; Li et al., 1998; Ho et al., 1999; Ko et al., 2000; Virkutyte et al., 2002; Saichek and Reddy, 2003; Zhou et al., 2004, 2006; Yeung and Hsu, 2005). Recently, researchers have used EK technology to treat harbor sediments (Stichnothe et al., 2001; Nyström et al., 2005a, 2005b, 2006). The effective removals of Cu, Zn, Cd and Pb from the harbor sediments have been obtained by *ex situ* electro-dialytic treatment (Nyström et al., 2005a, 2005b, 2006). Till now, there is no report on *in situ* EK treatment of sediments.

In this study, we tentatively used a galvanic cell, instead of power supply, to remove copper from sediments. The schematic diagram of the process is shown in Fig. 1. Iron (Fe) and carbon (C) are placed separately and connected with a conductive wire. Polluted sediments are put between them. Water was filled above the sediments. A weak voltage will be spontaneously formed in the sediment–water system because of the different electrode potentials of Fe and C (Reaction (1), (2) or (3)). Like a battery, the sediment was the inner part of galvanic cell and the wire was the outer part. It could be inferred that pollutants in the sediments can be moved by the electric field. Because the electrode potential of C is more positive than that of Fe, electric current flow from C to Fe in the wire and from Fe to C in the sediment. As a consequence, positive ions in sediment move from Fe to C by electromigration and negative ions from C to Fe.

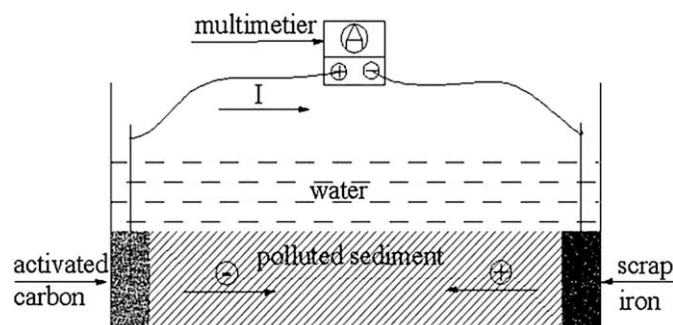
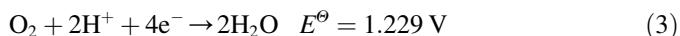
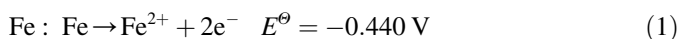


Fig. 1. Schematic setup of the galvanic cell system.

Compared with traditional EK technology, galvanic cell uses the inexpensive materials such as iron and carbon to produce electric field. Hence, the electric energy consumption was greatly reduced. The objectives of the present study were (1) to explore the feasibility of Fe/C galvanic cell to *in situ* remove copper from sediments; (2) to investigate the influence of the characteristics of the supernatant water and bottom sediment on the removal efficiency and (3) to analyze the mechanism of the removal of copper from sediment.

2. Materials and methods

2.1. Chemicals

$\text{CuCl}_2 \cdot 2\text{H}_2\text{O}$ (Chengdu Chemical Reagent Factory, China, $\geq 99.0\%$) was used as the source of copper. Scrap iron was provided by the mechanical factory of Huazhong University of Science and Technology, China. The iron was first washed in boiling NaOH (1%) solution for 10 min to remove oil and then in H_2SO_4 (1%) to remove rust. Finally, it was washed with tap water and deionized water thoroughly. Crustose carbon was purchased from Kexin Chemical Co. Ltd., China. It was washed by tap water and deionized water before use. Deionized water was used in the experiments. All the other reagents used were of analytical grade.

Kaolin is a kind of representative clayed soil (Ko et al., 2000; Saichek and Reddy, 2003; Yuan et al., 2006, 2007a), which has the similar viscosity to sediment. So, kaolin (Shanghai Fengxian Fengcheng Chemical Reagent Factory, China, chemical purity) was used as the simulated sediment. The main characteristics of kaolin have been studied in our previous study (Yuan et al., 2007b). It is clayed, weak acidic (pH 5.75), negatively charged (zero point of charge, ZPC, 3.36) and contains low content of organic matters (0.28%) and low cationic exchangeable capacity (1.71 cmol/100 g).

2.2. Procedures

Copper-contaminated sediments were prepared by adding the CuCl_2 solution to kaolin. The kaolin and the solution were stirred thoroughly. The water content of 50% was made by controlling the addition of CuCl_2 solution. The content of NaCl in sediment was adjusted by adding CuCl_2 solution containing NaCl (as the electrolyte). The final content of copper in the contaminated sediment was determined to be 150 mg/kg dry soil in T1 and T2 and 169 mg/kg in T3–T6 (Table 1). The simulated sediments were then settled for about 24 h. The uniform distribution of copper in the sediments was confirmed by the consistency of atomic absorption spectrophotometer (AAS) analysis of three random samples.

The sediment treatment setup was made of a polypropylene square container (36.5 cm length \times 18.5 cm width \times 21.5 cm depth) as shown in Fig. 1. A certain amount of the simulated sediment was added to the container. The sediment was pressed by a wood pan to remove air bubbles. Crustose carbon and scrap iron were placed at the two sides, respectively. In T1 and T2, 250 g crustose carbon and 600 g scrap iron were used;

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