



Technical Note

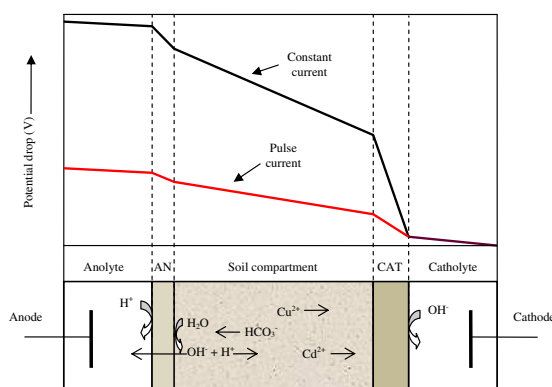
Electrodialytic soil remediation enhanced by low frequency pulse current – Overall chronopotentiometric measurement

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HIGHLIGHTS

- ▶ The pulse current could decrease the energy input during the EDR process.
- ▶ The detailed mechanism was given by chronopotentiometric study.
- ▶ The distribution of the energy consumption in EDR cells was demonstrated.
- ▶ The rate controlling step of EDR process was determined.
- ▶ Polarization of the membranes and clay particles were characterized.

GRAPHICAL ABSTRACT



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ABSTRACT

The effect of low frequency pulse current on decreasing the polarization and energy consumption during the process of electrochemical soil remediation was investigated in the present work. The results indicated that the transportation of cations through the cation exchange membrane was the rate controlling step both in constant and pulse current experiments, thus responsible for the major energy consumption. After 180 h, a decrease in both the initial ohmic resistance in each pulse cycle and the resistance caused by concentration polarization of the anion exchange membrane were seen in the pulse current experiment compared to the constant current experiment. At the cation exchange membrane, only the resistance caused by concentration polarization decreased. In the soil compartment, an average of +60 mV overpotential caused by the polarization of the electric double layer of the clay particles was obtained from the Nernstian behavior simulation of the relaxation process, which was significantly lower than the ohmic voltage drop induced by pore fluid resistance. Therefore, the ohmic polarization was the major contributor to the energy consumption in the soil compartment and diminished by pulse current.

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1. Introduction

The energy consumption of the electrochemically based remediation techniques is an important factor influencing costs and

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thus the applicability (Acar and Alshwabkeh, 1996; Reddy and Saichek, 2004; Kornilovich et al., 2005; Zhou et al., 2006; Ryu et al., 2010). Polarization, as an inevitable process, is a key issue as it is responsible for extra and nonproductive energy consumption. In the electrochemical soil remediation (EDR) system, it mainly includes the polarization of electrodes, membranes and clay particles in the soil.

The electrode polarization could be controlled by electrochemical polarization and/or concentration polarization, depending on the different rates of the electrode reactions and mass transport processes (Bard and Faulkner, 2001). However, in EDR the electrodes are placed in compartments separated from the soil by ion exchange membranes and electrolytes are circulated in the electrode compartments (Ottosen et al., 1997), which reduce the electrode polarization. Concentration polarization occurs in all membrane separation processes owing to differences in the transport numbers of ions in the solution and in the ion exchange membrane (Strathmann, 2010). Passing an electric current through an ion exchange membrane, the ionic concentration on the desalting surface of the membrane is decreased due to concentration polarization, and reduced to zero at the limiting current density. In this circumstance, there are no more ionic species available to carry the current and the voltage drop across the boundary layer increases drastically resulting in a higher energy consumption and generation of water dissociation. In the water saturated soil system, the most active part interacting with the external electric field is the clay particles (Sposito, 1989). The induced polarization is related to the electrodynamic effects on the electrical double layer of the clay particles due to the inherent negatively charged surface and non-conductive bulk (i.e. the solid part of the clay). During application of a direct current to the saturated soil system, excess charge will be accumulated on one side of the clay particles and depleted on the other, giving rise to a concentration polarization (Nischang et al., 2007; Mishchuk, 2010). Due to the incompatibility between the conductivity of the clay particles with low surface conductivity and the surrounding electrolyte solution (i.e. pore fluid) with high ionic conductivity, induced space charge will probably occur with a potential difference across the interface layer, similar to the charging of the ionic double layer at the electrode–electrolyte solution interface (Bard and Faulkner, 2001; Pamukcu et al., 2004). Considering the connection between clay particles and pore fluid as in series, the total voltage drop between two monitoring electrodes across the soil compartment in the EDR system under an external applied electric field can be expressed as:

$$V_t = (V_{eq} + V_R) + \eta \quad (1)$$

According to the expression of the voltage drop in the electrolysis between anode and cathode (Bard and Faulkner, 2001; Pamukcu, 2009), where V_{eq} is the voltage drop under equilibrium state, V_R the ohmic voltage drop induced by the pore fluid, and η is the overpotential.

Pulse current as an approach to decrease the energy input in an electrodynamic desalination process, was investigated by Mishchuk et al. (2001). The basic principle they applied was to keep the characteristic time of a pulse (at the interval magnitude of s) shorter than the time required for building up a concentration polarization layer near the membrane (transition time) in order to avoid water dissociation and nonproductive energy loss even at a current density above the limiting value (i.e. overlimiting current density). However, in EDR, the water dissociation at the anion exchange membrane at the overlimiting current density is of great importance and must necessarily be exceeded, since the resulting acidic front developing in the soil is a key factor for mobilization of the heavy metals. Therefore, the present work is focused on use of low frequency pulsed current (at the interval magnitude of h) to decrease the energy input compared to constant current during EDR. The detailed mechanism was given by chronopotentiometric analysis. Chronopotentiometry is an electrochemical characterization method measuring the electric potential response of a system to an imposed current. Usually, it is used to measure the polarization kinetics of an irreversible electrode process (Pletcher et al., 2006). The method allows a direct access to the voltage contribu-

tions in the different stages of the EDR process, because the dynamic voltage response over time can be analyzed.

2. Materials and methods

2.1. Soil sample

The experimental soil was contaminated by copper and cadmium, sampled from a pile of excavated soil. The soil was air-dried at room temperature, and gently crushed by hand in a mortar and passed through a 2 mm mesh sieve before the experiments.

Some chemical and mineralogical properties of the soil are presented in Table 1. Soil pH and conductivity were measured using a Radiometer pH electrode and conductivity electrode, respectively. The content of organic matter was found as a loss of ignition after 1 h at 550 °C. The carbonate content was determined volumetrically by the Scheibler-method when reacting 3 g of soil with 20 mL of 10% HCl. Grain size distribution was determined by wet-sieving approximately 100 g natural wet soil with 0.002 M $\text{Na}_4\text{P}_2\text{O}_7$ and the mineral composition of the bulk soil as well as the clay fraction was revealed by the XRD-analysis (Jensen et al., 2006).

2.2. Preparation of experimental setup

A schematic diagram of the experimental setup is shown in Fig. 1. The electrodynamic cell was made from polymethyl methacrylate. The cell had an internal diameter of 8 cm. The length of each cell compartment was 5 cm. The ion exchange membranes were from Ionics (anion exchange membrane AR204 SZRA B02249C and cation exchange membrane CR67 HUY N12116B). Platinum coated electrodes from Permascand were used as working electrode in compartments I and III. Between the two working electrodes, four monitoring electrodes (platinum coated electrodes) were used to monitor the voltage drop of three different parts of the cell, see Fig. 1. The soil (370 g) with adjusted water content of 23% was packed in compartment II. After assembly, the entire setup was placed on a slowly agitating table for 1 h to ensure a good distribution of soil particles.

2.3. Procedure

One constant current experiment and one pulse current experiment were made. In each of the electrode compartments 500 mL 0.01 M NaNO_3 adjusted to pH 2 with HNO_3 as electrolyte was circulated by pumps. After 24 h, 40 mA corresponding to a current density of 0.8 mA cm^{-2} was applied in both experiments by a power supply (Agilent E3612A). The voltage drop between anode and monitoring electrodes inserted in the different parts of the EDR cells as a function of time was automatically recorded by data-logger (Agilent 34970A) with a rate of once per 5 s. In the pulse current experiment, the pulse current was accomplished by a power supply timer instrument (Joel TE102), and the program

Table 1
Chemical and mineralogical properties of the soil sample.

pH	7.6
Conductivity	2.1 mS cm^{-1}
Carbonate content	7.1%
Organic matter	6.5%
Grain size distribution	
Sand	24%
Silt	61%
Clay	15%
Clay mineral composition	Kaolinite, Illite and Smectite

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