



# Citric Acid-Enhanced Electroremediation of Toxic Metal-Contaminated Dredged Sediments: Effect of Open/Closed Orifice Condition, Electric Potential and Surfactant

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## ABSTRACT

Citric acid (CA), a widely used eco-friendly electrolyte, can be employed as an agent for enhancing toxic metal (TM) removal from contaminated dredged sediment using electrokinetic (EK) technology. In this study, dredged harbor sediments co-contaminated by TMs were subjected to enhanced EK treatment using a mixture of chelating agent (CA) and surfactant as an additive in the processing fluids. Several control conditions that may influence the efficiency of TM removal were tested, including open/closed sediment chamber orifices, electric potential gradients (0.5, 1.0, and 1.5 V cm<sup>-1</sup>), and electrolyte surfactant. Tween 20 (4 mmol L<sup>-1</sup>) was used as a surfactant within the electrolyte to investigate the extent of TM removal in sediment with high organic matter content. The results showed that an open orifice led to a greater electro-osmotic flow (EOF) with moderate TM removal. In contrast, a closed orifice with a nonionic surfactant electrolyte allowed the highest removal of TMs from the matrix. Moreover, increasing the electric potential gradient led to a higher EOF under the open orifice condition, but no significant increase in TM removal was observed owing to a higher accumulation of TMs in the middle of the matrix, caused by the opposite direction of EOF and electro-migration of metal-citrate complexes.

**Key Words:** electric potential, electrokinetic remediation, electro-migration, electro-osmotic flow, electro-osmotic permeability, metal-citrate complex

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## INTRODUCTION

Although dredging operations are essential to maintain navigation security in waterways and harbors, economic and environmental issues associated with polluted dredged sediments have been increasing worldwide. Nearly  $5 \times 10^7$  m<sup>3</sup> of sediments are dredged each year from French harbors and channels (Benamar and Baraud, 2011). The disposal of dredged sediments is rigorously regulated by several international conventions (Rozas and Castellote, 2012). In France, the reference values to assess the chemical hazard of dredged sediments to aquatic ecosystems as well as preventive actions to minimize harmful effects related to the disposal of these materials are available in directives and regulations such as GEODE (Groupe d'Etudes et d'Observation sur les Dragages et l'Environnement) guidelines.

Organic contaminants (*i.e.*, polycyclic aromatic hydrocarbons (PAHs), polychlorinated biphenyls, tribu-

tylins, *etc.*) and inorganic contaminants (toxic metals, TMs) bound to dredged sediments present a contamination risk for coastal waters, which in turn affect the ecosystem. For this reason, several approaches, such as thermal treatment, bioremediation, phytoremediation, solidification/stabilization by hydraulic binders and washing treatment, have been widely implemented for mitigating the pollution potential of these materials (Agostini *et al.*, 2007). However, remediation of fine-grained sediments is relatively complex due to their low permeability and great specific surface area of the particles (Alshawabkeh *et al.*, 1999; Genc *et al.*, 2008; Yeung and Gu, 2011).

Electrokinetic (EK) remediation has been demonstrated to be a cost-effective remediation technique for clay-rich soils (Acar and Alshawabkeh, 1993; Gu and Yeung, 2011; Yeung and Gu, 2011; Ammami *et al.*, 2014). Although its effectiveness for accelerating slurry dewatering has been well established for several years, this method is not extensively applied for dredged ma-

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rine sediment remediation (Ammami *et al.*, 2014). The EK remediation process involves electro-osmosis, electro-migration, and electrophoresis, enabling TMs to be removed from contaminated sediments under a direct current electric field (Virkyute *et al.*, 2002). The  $H^+$  and  $OH^-$  generated at the anode and cathode, respectively, move towards the opposite electrode (Giannis *et al.*, 2010). It is well known that acidification of the sediment by  $H^+$  can facilitate desorption and dissolution of TMs (Li *et al.*, 1998). In contrast, the basic front progressing from the cathode toward the anode can increase metal precipitation, which may inhibit the removal capacity of the EK system (Kim B K *et al.*, 2011). Therefore, pH control at the cathode by an acidic additive is needed to prevent metal precipitation and thus enhance contaminant removal (Pazos *et al.*, 2006; Yuan and Chiang, 2008; Gu and Yeung, 2011; Ryu *et al.*, 2011; Ammami *et al.*, 2014).

The use of chelating agents, such as ethylenediaminetetraacetic acid, ethylenediamine-N, N'-disuccinic acid, nitrilotriacetic acid, humic acids, and citric acid (CA), as electrolyte solutions has been investigated. The results showed that they could improve metal removal from sediments (De Giannis *et al.*, 2008; Colacicco *et al.*, 2010; Ammami *et al.*, 2015; Bahemmat *et al.*, 2016; Masi *et al.*, 2016; Song *et al.*, 2016). Among the aforementioned chelating agents, CA can be used as a catholyte solution owing to its buffering effect, biodegradability, and chelating and complexing actions. The CA can form several bonds with toxic metal ions to produce anionic complexes (metal-citrate complexes), which migrate towards the anode (Evangelou *et al.*, 2007; Yoo *et al.*, 2015). In recent studies, several surfactants, such as SDS, Tween 20, Tween 80, Brij35, Poloxamer 407, and Nonidet P40, were tested to enhance EK remediation of PAH-polluted soils and sediments (Saichek and Reddy, 2005; Gu and Yeung, 2011; Alcántara *et al.*, 2012; Hahladakis *et al.*, 2013, 2016; Ammami *et al.*, 2014). In addition, surfactants can increase the efficiency of TM removal by improving TM desorption from the sediment matrix, owing to their ability to lower the surface and interfacial tensions (Ammami *et al.*, 2014; Hahladakis *et al.*, 2014).

Bench-scale EK remediation studies have been widely reported, but there are limited tests using pilot-scale EK remediation treatments (Li *et al.*, 2009). In addition, the bench-scale EK remediation tests were often carried out in a soil/sediment chamber without orifices (in an airtight space) (Puppala *et al.*, 1997; Wang *et al.*, 2005; Zhou *et al.*, 2005; Yuan and Chiang, 2008; Alcántara *et al.*, 2012), whereas pilot-scale EK remediation tests were often performed in an open tank or *in situ* conditions (Kim *et al.*, 2002; Li *et al.*,

2009). Consequently, the influence of open/closed systems on the EK remediation process has not yet been investigated and remains an open research topic.

During EK remediation treatment, sediment acidification induces  $CO_2$  gas release from carbonates [ $CaCO_3 + 2H^+ \rightarrow Ca^{2+} + H_2O + CO_2$ ]. The  $CO_2$  gas accumulates in the matrix and thereby increases the void volume, lowering the amount of aqueous phase (Rutigliano *et al.*, 2008). This process greatly influences electro-osmotic flow (EOF) and electric resistivity during EK treatment and decreases the efficiency of TMs removal.

In this study, EK remediation tests were performed under open and closed orifice conditions using the same device. Different electrical potential gradients were applied, leading to the generation of various electrochemical parameters, which may affect TM removal. Tween 20 was used to increase the solubility of organic matter, which in turn can affect the sorption of toxic metals in the sediment.

## MATERIALS AND METHODS

### *Electrokinetic remediation apparatus*

The experimental apparatus used in this study (Fig. 1) has been described elsewhere (Ammami *et al.*, 2014, 2015; Song *et al.*, 2016). The main part of the experimental apparatus consisted of a poly tetra fluoroethylene (PTFE) cell, including a sediment chamber (diameter 4.9 cm, length 14 cm) and two electrode compartments. A row of three orifices (diameter 1.0 cm) spaced 5 cm apart was added at the upper generatrix of the sediment chamber. The natural sediment was packed in the chamber and graphite electrode plates were placed in each electrode compartment. The electrodes were separated from the sediment by a 0.45- $\mu m$  PTFE filter (Millipore, USA) placed over a screen. The set-up was assembled using four clamping rods sealed by two O-rings. Two peristaltic pumps (KNF Stepdos, Germany) with low flow rate ( $5 \text{ mL h}^{-1}$ ) were used to supply and renew anolyte/catholyte aqueous solutions. A voltage gradient ( $1 \text{ V cm}^{-1}$ ) was continuously applied and the electrical current was periodically measured. At the test end, the sediment was extracted and cut into four slices for analysis. Gas vents were included in the electrode compartments to allow electrolysis gases to escape. During the test, effluents were collected from both electrode compartments by two overflow holes and stored in glass flasks.

### *Sediment sampling and TM analysis*

Sediment samples were collected from the disposal site of a French harbor in Tancarville (Normandy, Fra-

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