



Application of biosurfactants and periodic voltage gradient for enhanced electrokinetic remediation of metals and PAHs in dredged marine sediments



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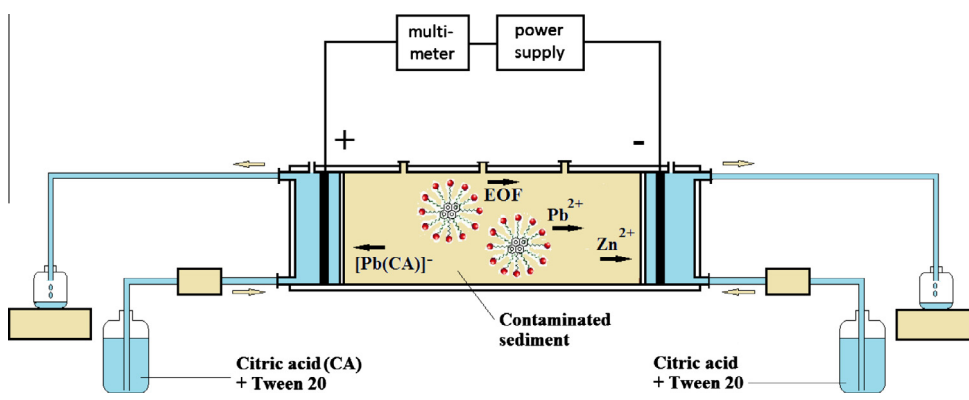
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HIGHLIGHTS

- Improving electrokinetic remediation of metals and PAHs in dredged harbor sediments.
- Increasing citric acid concentration, associated to Tween20 surfactant, is favorable.
- Applying a periodic voltage gradient with low concentration of additives is effective.
- Rhamnolipid and viscosin-like biosurfactants give promising results as additives.

GRAPHICAL ABSTRACT



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ABSTRACT

Dredged harbor sediment co-contaminated by heavy metals and polycyclic aromatic hydrocarbons (PAHs) was subjected to enhanced electrokinetic treatments, using a mixture of a chelating agent (citric acid CA) and a surfactant as additives in the processing fluids. We tested various operating conditions (at 1 V cm^{-1}): different CA concentrations, applying a periodic voltage gradient, pre-conditioning the sediment with the additives, and replacing the synthetic surfactant Tween 20 (TW20) by biosurfactants. Increasing the CA concentration was favorable for both metal and PAH removal. Applying a periodic voltage gradient associated to a low concentration of CA and TW20 provided the best results for Zn, Cd and Pb removal and also for removal of the 16 priority PAHs. Promising results were obtained with solutions containing rhamnolipids (0.028%) and a viscosin-like biosurfactant produced by *Pseudomonas fluorescens* Pfa7B (0.025%), associated to a periodic voltage gradient. Although the rhamnolipid and the viscosin-like compounds involved a higher electrical current than TW20, metals were less removed from the sediment. The electroosmotic flow was lower when we used biosurfactants, hence a less effective effect on PAH removal.

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1. Introduction

Management of dredged sediment is of great concern for maintaining harbor activities (Benamar and Baraud, 2011). In France, chemical guidance values are provided to assess the chemical

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hazard following the disposal of dredged sediment into environmental waters, as recommended by the OSPAR convention directives or French regulations such as GEODE guidelines (14/06/2000). Thus the restricted disposal of dredged sediment into the marine environment has led managers to treat it ashore. However, storage on disposal sites is not a sustainable solution because of the risk of contaminants transferring to the environment. Reuse of dredged sediment in construction materials in civil engineering or habitat restoration can be proposed (Yozzo et al., 2004; Wang et al., 2012), but such sediment management requires treatments to reduce their contamination level.

In contaminated dredged sediment, inorganic metals (Peng et al., 2009) and persistent organic pollutants such as polycyclic aromatic hydrocarbons (PAHs) (Sprovieri et al., 2007) are of great concern regarding their toxicological hazard: they bioaccumulate in food chains, and induce higher mortality rates, growth reduction or disturbance of reproductive processes in marine organisms (Roberts, 2012). Powerful techniques must be developed in order to remove them from sediment, such as thermal desorption, sediment washing or flushing, solvent extraction, bioremediation (Jones et al., 2001; Libralato et al., 2008; Beolchini et al., 2009). However these technologies fail when low permeable soil or sediment is encountered.

Electrokinetic (EK) remediation is a technology that has received attention as a practical *in-situ* and *ex-situ* remediation technique for low permeable clay-rich soil or fine-grained sediment. Briefly, when a low voltage gradient is applied to the sediment *via* embedded electrodes, charged pollutants electromigrate towards the electrode of the opposite charge (Ryu et al., 2011). The electric field induces an electro-osmotic flow, and solubilized neutral pollutants can be also transported with the pore fluid (Cameselle and Reddy, 2012). Successful laboratory-scale EK tests have been reported for sediments or soils contaminated by metals or PAHs (Giannis et al., 2009; Pazos et al., 2010). Simultaneous EK removal of both metals and PAHs has been investigated lately (Maturi and Reddy, 2008; Colacicco et al., 2010; Ammami et al., 2013).

Enhanced EK technology is highly dependent on the type of processing solution used. The main reaction, inherent to the EK process, is water electrolysis. It generates H^+ at the anode side and OH^- at the cathode side, and thus causes metals to precipitate near the cathode (Nogueira et al., 2007). Therefore acids are suitable candidates near the cathode as they enhance their solubilization and transport to the cathode. Adding complexing agents can also convert soil-bound metals into soluble complexes and enhance their removal (Gidarakos and Giannis, 2006). Concerning PAHs, enhancing agents that allow their better desorption from sediment particles and solubilization in the pore fluid can be used to achieve satisfactory results. Various surfactants, cyclodextrins or cosolvents have been tested (Park et al., 2007; Maturi et al., 2009; Hahladakis et al., 2013).

Mechanisms of EK PAH removal by synthetic surfactants have been investigated, but little research has been carried out using biosurfactants (Alcantara et al., 2009; Chang et al., 2009; Gonzini et al., 2010). Biosurfactants are biological surface active agents produced by certain yeasts or bacteria. They were used to clean soils polluted by organics, especially rhamnolipids produced by *Pseudomonas aeruginosa* (Bordas et al., 2005; Lai et al., 2009). Other classes of biosurfactants, such as lipopeptides or bioemulsans have also been tested for applications in soil or sediment remediation (Mulligan et al., 1999; Dahrazma and Mulligan, 2007; Juwarkar et al., 2007; Franzetti et al., 2009).

We conducted bench-scale electrokinetic tests to understand the effect of different operating parameters on the removal, in one stage, of mixed contaminants from dredged sediment, *i.e.* five metals (Cd, Cr, Cu, Pb, Zn) and the 16 priority PAHs defined by the

US-EPA. We tested the amount of chelating agent (citric acid), the nature of the surfactant (synthetic Tween 20 or biological surfactants: rhamnolipids and, for the first time, a cyclolipopeptide) and sediment pre-conditioning. Moreover, little is known about the benefits of the application of a periodic electric potential on the EK surfactant-enhanced removal of PAHs (Saichek and Reddy, 2005). So we also assessed the combination of enhancing agents, never tested together before, with the application of a periodic voltage gradient.

2. Materials and methods

2.1. Materials

2.1.1. Chemicals

Acetonitrile, methylene chloride, toluene and acetone were provided by VWR (Fontenay sous Bois, France). Nitric acid (65%), hydrochloric acid (37%), citric acid (CA) and Tween 20 (TW20) were obtained from Fisher Scientific (Illkirch, France). A standard mixture of 16 priority PAHs, perdeuterated phenanthrene, cadmium nitrate, chromium nitrate, copper sulfate, zinc nitrate and lead nitrate were provided by Sigma–Aldrich (St Quentin-Fallavier, France).

2.1.2. Sediment sampling

The dredged sediment was collected from the disposal site of a French harbor in Normandy, localized near important urban centers and industrialized areas. Its properties are presented in Table 1. pH was measured according to the NF ISO 10390 standard, electrical conductivity according to the NF ISO 11265 standard and moisture content was obtained in accordance with the French NF P 94-050 standard. Organic matter content was measured in accordance with the NF EN 12879 standard. Particle size distribution was determined using a laser particle-sizing device (Malvern Instruments, Malvern, UK).

2.1.3. Biosurfactants

The rhamnolipid biosurfactant (90% purity) was obtained from AGAE Technologies (Corvallis, USA). The other biosurfactant was our own production: *Pseudomonas fluorescens* PfA7B strain was grown on culture plates and its biosurfactant production was recovered after 14 d of incubation, as described in a previous paper (Groboillot et al., 2011). The biosurfactant, a mixture of two viscosin-like cyclolipopeptides (viscosin and massetolide E), was purified and characterized, as already described (Portet-Koltalo et al., 2013). The critical micellar concentrations (CMCs) of the biosurfactants were determined by surface tension measurements using the pendant drop method (Tensiometer DSA30, Krüss, Germany).

2.2. Electrokinetic tests

The experimental EK setup is described in a previous paper (Ammami et al., 2013). Briefly, the natural sediment was packed into a Teflon PTFE chamber. Graphite electrode plates were placed in each electrode compartment, separated from the sediment by porous glass papers. Two pumps filled the electrode reservoirs with the aqueous processing fluids (10 mL h^{-1}). A voltage gradient (1 V cm^{-1}) was applied continuously or periodically (5-d-on/2-d-off cycles) and the electrical current was periodically measured. At the end of each treatment, the sediment was extracted and cut into four slices for analysis.

Experiments 1 to 3 consisted in increasing CA concentrations while keeping the TW20 concentration constant (Table 2). Experiments 4 and 5 consisted in the same operating conditions as experiment 1, but experiment 4 was conducted under a periodic electric potential and experiment 5 with pre-conditioned sediment. In this latter case, the sediment was previously air-dried for a week and

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