Marine Pollution Bulletin 113 (2016) 44–54

Contents lists available at ScienceDirect

Marine Pollution Bulletin

journal homepage: www.elsevier.com/locate/marpolbul

Influence of enhancing electrolytes on the removal efficiency of heavy metals from Gabes marine sediments (Tunisia)

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ARTICLE INFO

Article history:
Received 8 February 2016
Received in revised form 28 June 2016
Accepted 28 June 2016
Available online 12 October 2016

Keywords:
Marine sediments
Heavy metals
Electrokinetic remediation
Enhancing electrolytes

ABSTRACT

This study focused on the feasibility of the treatment of heavy metals-contaminated sediments from Gabes harbor (Tunisia) using enhanced electrokinetic process. It presented a laboratory short-time electrokinetic experiment. The enhancing agents, as citric, acetic acids and sodium dodecyl sulfate (SDS) were used regarding their low environmental hazard. The electrokinetic cell was specially designed in order to elaborate two experiments at the same time. This paper is composed of three parts. The first part introduces the characterization of Gabes sediments. The second part describes the design of laboratory electrokinetic cell and the followed methods. The third part is dedicated to the results analysis. Treatment efficiency revealed that more than 80% of lead was removed from Gabes marine sediments. The reduction of cooper concentration, in sediments after treatment, ranged from 74 to 87%. Despite, the high removal of cadmium that ranged from 58 to 79%, treated sediments presented Cd concentration above the threshold limit.

1. Introduction

Dredging marine sediment is a vital activity for the commercial harbor of Gabes. For instance, the total dredged materials from harbor reached 2.2 Mm^3 in 2009. Environmental issues rise concerning the outcome of these dredged materials. The most common practice for their disposal is to dump them back at sea. The surrounding of Gabes harbor is composed of industries and chemical such as the Tunisian Phosphate Industry. Thus, the harbor activity is characterized by an industrial vocation that essentially assures the transit of chemicals on behalf of neighboring plants. This commercial traffic consists primarily of sulfur and ammonia import and phosphoric acid and phosphate fertilizer export. According to a previous study (Bel Hadj Ali et al., 2014; Missaoui et al., 2015), Gabes sediments present important level of contamination. As a result, the treatment of these materials is a necessity, in order to decontaminate them or to reduce the contamination levels acceptably. Sediments can be divided into two major groups based on the nature of the pollutant. On one hand, there are organic pollutants represented by hydrocarbons and derived products such as PAHs (Polycyclic Aromatic Hydrocarbons), PCBs (Poly Chlorinated Biphenyls), and Tributyltin (TBT). Their behavior depends on parameters such as solubility, volatility, and electronic structure (Yaron and Clavet, 1996). On the other hand, there are inorganic pollutants that include heavy metals such as mercury (Hg), lead (Pb), nickel (Ni), copper (Cu), chromium (Cr), zinc (Zn), and cadmium (Cd) (Caplat et al., 2005; Chatain et al., 2013; Lions et al., 2010). Generally, heavy metals have a longer lifespan than organic pollutants.

For treating metal-contaminated sediments, several remediation techniques are used and may be regrouped in two categories. The first category is biological techniques including biodegradation that consists in the use of microorganisms either to break the organic structure of the pollutant substances or to accelerate its natural decomposition. The biological techniques include also phytoremediation where plants are used to stabilize or extract contaminants from the sediment (Bhargava et al., 2012). Phytoextraction is mostly effective in treating nickel-contaminated and zinc-contaminated soils whereas phytostabilization is suitable for other metals such as chromium, lead and mercury (USEPA, 1997). The second category is Physico-chemical techniques with multiple subcategories such as the Stabilization/Solidification (S/S) and the Separation/Recuperation (S/R) methods. S/S techniques consist in confining the contaminants inside the sediments' matrix by reducing their interactivity with their natural environment. They include vitrification, phosphatation, and hydraulic binding (Chiang et al., 2012; Lafhaj et al., 2007; Tomasevic et al., 2013; Yu et al., 2009). As a complement, thermal techniques can be used as a complement to metal immobilization techniques in order to further stabilize the pollutants(Agostini et al., 2007; Kribi et al., 2012; Lafhaj et al., 2008; Samara et al., 2009). As for S/R techniques, they consist in removing the contaminant from the sediment (Chiavola et al., 2010; Fonti et al., 2014; Goodsir et al., 2013; Masciandaro et al., 2014). The vacuum extraction and electrokinetic remediation belong to this category. It can be noted that other treatment method can combine two different techniques such as coupled electrokinetic and phytoremediation technique. The EK remediation produces

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http://dx.doi.org/10.1016/j.marpolbul.2016.06.102
0025-326X/

the solubilization of metals in the soil and thus ease their absorption by plants (Bhargavi and Sudha, 2015; Mao et al., 2015).

Compared to other techniques, the electrokinetic (EK) treatment is promising for contaminated soils for its high remediation efficiency and time and cost effectiveness (Niroumand et al., 2012). During electrokinetic treatment, the contaminated sediment are placed within a confined area delimited by electrodes. This area defines the electrokinetic cell composed of the contaminated soil, the anodic chamber (anode + anolyte solution) and the cathodic chamber (cathode + catholyte solution). The anolyte and catholyte solutions define the water for ordinary electrokinetic remediation. A direct electric current is then applied to the electrodes. The removal process relies on two mechanisms: electro-osmosis and electro-migration. Electro-osmosis takes effect in one direction that corresponds to the transport of pollutants from anode to cathode. Whereas, electro-migration has two directions as ions move to the oppositely charged electrode. Electro-migration is enhanced by the acid front generated in the anode compartment by electrolysis and migrating towards the cathode while facilitating the heavy metals dissociation and/or desorption. It is inhibited at the same time by the basic front generated by a reduction in the cathode compartment and migrating in the opposite direction while causing the precipitation of the heavy metals (Giannis et al., 2007).

The enhanced electrokinetic techniques, using reagents, presented a way to improve and accelerate the removal process. Different purging solutions can be used in the electrokinetic remediation (Ammami et al., 2015; Giannis and Gidarakos, 2005; Iannelli et al., 2015; Masi et al., 2015). New environment-friendly concept was elaborated, which consists in using other purging solution as electrolytes such as weak acid, chelating agents and surfactants (Hahladakis et al., 2014). In addition, surfactants or weak acids can be used as saturation solution for the sediment before starting the electrokinetic treatment. Reddy and Ala (2006) studied the electrokinetic treatment of heavy metals-contaminated sediments. Two surfactants (5% Igepal CA-720 and 3% Tween 80), a cosolvent (20% n-butylamine), and a cyclodextrin (10% HP-β-CD) were used to enhance the process. As a result, none of the experiments succeeded to effectively remove the heavy metals. Rozas and Castellote (2012) investigated in the electrokinetic remediation using citric, acetic, humic acids and EDTA for the remediation contaminated dredged material. Three years later, Rozas and Castellote (2015) studied the influence of non-ionic surfactants (one bio-surfactant, one chelating agent, and one weak acid) on electrokinetic remediation of marine sediments. The use of sodium dodecyl sulfate (SDS) as an electrolyte and a chelating agent was investigated by researchers and has proven efficient in removing cadmium from contaminated soils (Cameselle et al., 2013; Giannis et al., 2007; Huang et al., 2012; Kaya and Yukselen, 2005; Peng and Tian, 2010; Yuan and Weng, 2006).

This work studies the feasibility of the treatment of heavy metals-contaminated sediments from Gabes harbor (Tunisia) using enhanced electrokinetic process. This study presented a laboratory short-time electrokinetic experiment. The enhancing agents, as citric, acetic acids and SDS was used regarding their low environmental hazard. The electrokinetic cell was specially designed in order to elaborate two experiments at the same time. This paper is composed of three parts. The first part introduces the characterization of Gabes sediments. The second part describes the design of laboratory electrokinetic cell and the followed methods. Finally, the results are presented, analyzed, and compared to the environmental quality assessment thresholds and some recommendations are suggested.

2. Materials and methods

2.1. Materials

Nine samples were collected from the upper sediments layer of the Gabes commercial harbor. The latter is located in the middle of Gabes gulf in the south East of Tunisia (Fig. 1). Physical and chemical characterization were carried out on collected sediments. Chemical results on Gabes sediments, in previous studies, showed high contamination with heavy metals mainly cadmium and zinc (Bel Hadj Ali et al., 2014; Missaoui et al., 2015). In order to study the performance and efficiency of the electrokinetic remediation, the sediment sample containing the higher concentrations of major heavy metals was studied.

2.1.1. Physical characterization

The sediment physical characteristics are reported in Table 1. According to the particle size results, the sediment is a slightly clayey sandy silt. It presented a high fine fraction (<63 μm) about 82.5% of total size distribution. A high natural water content was observed in the sediment. With 17% of organic matter content, Gabes sediment

Fig. 1. Map of the study area in Gabes, Tunisia (Image captured with Google Earth).