



# Performance of electroremediation in real contaminated sediments using a big cell, periodic voltage and innovative surfactants



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## HIGHLIGHTS

- Innovative surfactants, periodic voltage and a big electrokinetic (EK) cell were used.
- The EK efficiency in removing PAHs and toxic metals from sediments was assessed.
- Maximum PAHs removal efficacy was obtained by the use of Nonidet P40.
- Arsenic, nickel and chromium obtained the highest removal percentages.
- Periodic application of voltage favoured the efficiency of the overall EK process.

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## ABSTRACT

The present work focused on evaluating the electrokinetic (EK) treatment of real contaminated sediments with toxic metals and polycyclic aromatic hydrocarbons (PAHs), using a big laboratory EK cell, periodic voltage and recently tested non-ionic surfactants. The results indicated that the “day on-night off” application mode of voltage, in conjunction with the selected solubilising agents, favoured the overall EK process. Arsenic, nickel and chromium exhibited the highest removal percentages, obtaining 83%, 67% and 63%, respectively, while zinc and lead attained 54% and 41% at the maximum. Furthermore, in the experiments where the non-ionic surfactants were introduced in the electrolyte chambers, there was a major uniform removal of PAHs from the entire sediment across the EK cell, indicating the high solubilisation capacity of the enhancing agents. Essentially, transport and in some cases removal of PAHs (particularly from sections adjacent to the electrolyte compartments) also occurred in the unenhanced EK run, mainly due their negative charge, their potential weak bonds to the soil matrix and to the periodic application of voltage. Maximum removal was obtained by the use of Nonidet P40 where app. 1/3 (ca. 6498 µg out of 20145 µg) of the total initial amount of PAHs were removed from the cell.

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## 1. Introduction

Electrokinetic (EK) remediation (also known as “electroremediation”) is one of the many techniques that can be employed for the decontamination of polluted sites (involving soils/sediments) from various inorganic (toxic metals) and/or organic (polycyclic aromatic hydrocarbons, PAHs) pollutants.

EK relies mainly on the application of direct electric current to a confined contaminated area by the simultaneous use of electrolyte solutions known as “anolyte” and “catholyte” (introduced in the anodic and cathodic chambers, respectively). The contaminants are, in turn, removed by a variety of electrochemically occurring phenomena (water oxidation and reduction, heat transfer) and transport mechanisms-processes (electromigration, electroosmosis, and electrophoresis) which act either individually or synergistically [1–3].

However, there are various other key points that have been reported to contribute in promoting and favouring the removal of the contaminants that are present in a soil/sediment sample, whether it is real or spiked. Electrodes material and configuration [4–8], use of solubilizing agents e.g. chelating agents, surfactants, cyclodextrins, co-solvents as well as granulometry of the

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soil/sediment sample [9–25], control and effect of operating parameters e.g. pH, electroosmotic flow (EOF), current density, voltage, etc. [26–31] are some of the most important factors affecting the overall efficiency of the EK process.

Considering its numerous applications both in the laboratory [32–36] as well as any attempt of pilot scale or scaling up [4,5,37–40], EK appears to be one of the most sufficient and cost-effective alternatives for treating, even, real sediments (dredged or surficial) which often come with the simultaneous presence of multiple contaminants which interact with the constituents of solid matrices, thereby creating other compounds that limit and/or incommode even more the EK process. However, when all contaminants and/or substances are dissolved, they are, then, easily transported (flushed) into the electrolyte chambers, thus obtaining their desirable removal from the polluted soil/sediment sample.

The majority of the EK remediation studies that exist in literature report, mostly, the remediation of surrogate spiked soil/sediment samples, by the use of small EK cells, with dimensions of sediment compartment  $\varnothing 5$  cm  $\times$  L10 or L20 cm and of electrolyte chamber  $\varnothing 5$  cm  $\times$  L5 cm, with the capacity of receiving approximately 250–500 g of soil/sediment (dry matter). In addition, most of the experiments reported in these studies are performed under the application of a constant voltage gradient, using ordinary and widely tested solubilizing agents, such as chelates (e.g. EDTA, EDDS, PDA, DTPA, LED3A, citric and acetic acid) [41,42] as well as surfactants (e.g. Tween 80, Igepal CA-720, SDS, Triton X 100, APG, Calfax 16L-35) [11,43–45] some of which are finally found inadequate of achieving a sufficient removal of contaminants.

The present work focused on the investigation of the overall performance of EK process in treating real surficial sediments, contaminated by toxic metals and PAHs. Specifications on the procedure included: (a) the use of an EK cell almost ten times bigger in capacity than the commonly used ones in laboratories, (b) periodic application of voltage (day on-night off mode), a tactic that has been implemented only a couple of times in the past, [27,29] and (c) the use of newly introduced (in the EK technology) non-ionic surfactants (commercially known as Nonidet P40 and Poloxamer 407) that have successfully been implemented only in the previous works of the same team of authors [12,13]; hence further investigation in a bigger scale and under the aforementioned experimental conditions is required. Furthermore, non-ionic surfactants are commonly used in electroremediation of soils/sediments due to their high solubilisation capacity, biodegradability and low potency to both terrestrial and aquatic organisms. In addition, these non-ionic surfactants achieved relatively high removal percentages of both organic (PAHs) and inorganic (toxic metals) contaminants, in comparison to previous EK studies and choices of solubilizing agents reported in literature. Nevertheless, EDTA was actually used in one of this work's experiments as one of the best chelating agents, so as to facilitate even more the metal extraction from the soil matrix and compare its effectiveness to the one of surfactants. Finally, acetic acid was introduced in the cathode chamber of the EK cell in all runs to depolarize the hydroxyl ions generated by various cathode electrolytic reduction processes. Further details on the experiments and on the advantages of using acetic acid are given in Section 2.2.

## 2. Materials and methods

### 2.1. Physicochemical properties of sediment samples and analytical methods used

Surficial sediments ( $n=4$ ) were collected from four different, but adjacent, spots of Elefsis Bay, Athens, Greece, by the use of a Petersen zinc-plated grab sampler. They were, then, homogenized and stored at freezing conditions till the final sample reached the

**Table 1**  
Physicochemical properties of the sediment.

Physical Properties	Value
Moisture (%)	24.5
pH	8.05
Redox (mV)	85
Electrical conductivity ( $\text{mS cm}^{-1}$ )	13.88
Organic matter (%)	5.86
Specific gravity	2.01
Cation exchange capacity ( $\text{meq } 100\text{gr}^{-1}$ )	1.76
Main minerals	Percentages (%)
Quartz	30
Calcite	48
Dolomite	6
Mica	5
Chlorite	4
Feldspars	4
Hematite	2
Kaolinite	1
Particle size distribution	Percentages (%)
Sand	81
Silt	9
Clay	0
USCS classification	Sand (S)
Initial toxic metal content	Concentration ( $\text{mg kg}^{-1}$ dw, average of three replicates)
Cr	31.79
Ni	18.17
Cu	59.11
Zn	218.27
As	4.29
Pb	80.42
Initial PAH content	Concentration ( $\text{ng g}^{-1}$ dw, average of three replicates)
Naphthalene	70
Acenaphthylene	<50 <sup>a</sup>
Acenaphthene	<50 <sup>a</sup>
Fluorene	80
Phenanthrene	660
Anthracene	<50 <sup>a</sup>
Fluoranthene	1100
Pyrene	1150
Benzo(a)anthracene	790
Chrysene	540
Benzo(b)fluoranthene	770
Benzo(k)fluoranthene	240
Benzo(a)pyrene	460
Dibenzo(a,h)anthracene	90
Benzo(ghi)perylene	330
Indeno(1,2,3-cd)pyrene	360
Sum PAHs	6700

"<" below detection limit.

<sup>a</sup> Measurements below detection limit were taken as half the limit.

laboratories of Technical University of Crete (TUC). The homogenized sample was, in turn, air-dried for 72 h and sieved to remove the 2-mm oversize fraction. The dry matter was then chemically analysed for determining the total metal and PAH content. The physicochemical properties of the sediment are presented in Table 1. Redox potential (ORP) and pH were calculated according to ASTM D4972, by the use of a Crison pH-meter. Moisture, organic matter and electrical conductivity (EC) were determined according to ASTM D2974, specific gravity using method ASTM D854-92 and cation exchange capacity (CEC) using EPA 9081. A refractometer (Siemens 5000) was used for X-ray diffraction to determine the main minerals in the sediment, while a grain size analysis was performed to classify it according to the Unified Soil Classification System (USCS). The sediment was, in turn, characterized according to ASTM D2488 as sand (S).

The content of six selected toxic metals was determined after acid digestion of the homogenized sediment sample according to EPA method 3051A and metal concentrations were measured in

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