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Electrodialytic remediation of polychlorinated biphenyls contaminated soil with iron nanoparticles and two different surfactants

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ABSTRACT

Polychlorinated biphenyls (PCB) are persistent organic pollutants (POP) that strongly adsorb in soils and sediments. There is a need to develop new and cost-effective solutions for the remediation of PCB contaminated soils. The suspended electrodialytic remediation combined with zero valent iron nanoparticles (nZVI) could be a competitive alternative to the commonly adapted solutions of incineration or landfilling. Surfactants can enhance the PCB desorption, dechlorination, and the contaminated soil cleanup.

In this work, two different surfactants (saponin and Tween 80) were tested to enhance PCB desorption and removal from a soil sampled at a polluted site, in a two-compartment cell where the soil was stirred in a slurry with 1% surfactant, 10 mL of nZVI commercial suspension, and a voltage gradient of 1 V cm^{-1}.

The highest PCB removal was obtained with saponin. Higher chlorinated PCB congeners (penta, hexa, hepta and octachlorobiphenyl) showed removal percentages between 9% and 96%, and the congeners with highest removal were PCB138, PCB153 and PCB180. The use of low level direct current enhanced PCB removal, especially with saponin. Electrodechlorination of PCB with surfactants and nZVI showed encouraging tendencies and a base is thus formed for further optimization towards a new method for remediation of PCB polluted soils.

1. Introduction

Soil contamination with persistent organic pollutants, such as polychlorinated biphenyls (PCB), is an important environmental problem, due to their persistence, chemical stability and strong adsorption to soils that inhibits their extraction and degradation, and also to the risks associated with human health and ecosystems [1,2]. An inclusive state of the art review on the technologies available for PCB contaminated soils and sediments showed the advantages and disadvantages of the existing methods, and highlighted the need to find cost-effective and sustainable alternatives [3]. One possible solution to cope with recalcitrant contaminants such as PCB can be the integration of remediation technologies that, when coupled together (simultaneously or in succession, in the so called "treatment trains"), work in a synergistic manner, minimizing the remediation costs for achieving risk-based endpoints, in a quicker and more efficient way than employing single technologies [4].

Zero valent iron nanoparticles (nZVI) are strong reductants that can dechlorinate PCB in aqueous solutions [5–7], but revealed limited results in soils so far [8,9]. Pd/Fe bimetallic nanoparticles, when combined with electrokinetic remediation (EK), resulted in only 20% PCB removal after 14 days with historically contaminated soil [10]. The electrodialytic remediation of suspended soil in conjunction with nZVI enabled a 83% PCB removal in just 5 days [11].

Electroremediation is a group of evolving technologies that started in the 1990s, now targeting a wide range of contaminants, and have lately incorporated enhancement techniques and the combination with other technologies [12]. The use of surfactants for enhancing electrokinetic remediation of contaminated soil with organochlorines and mixed contaminations was reviewed by Gomes et al. [12] and Cameselle et al. [13]. Surfactants improve solubilization and desorption behavior of hydrophobic organic compounds, increasing their availability in contaminated environments [14,15], which can boost the remediation technologies efficacy. Different surfactants have already been tested, but beyond their desorption properties, they should also be environmental

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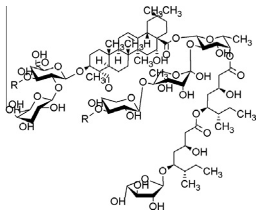
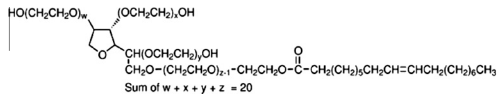
Table 1
Properties of the nonionic surfactants used in the experiments.

Trade name	Molecular weight	Molecular structure	CMC – Critical micelle concentration (mg L^{-1})
Saponin	1650		42.6 [18]
Tween 80	1310		40 [28]

friendly substances [16] and minimize the inhibitory effect of the surfactants on iron nanoparticles reactivity [10,17]. Saponin is a representative non-ionic plant-derived biosurfactant that can efficiently increase desorption and degradation of PCB in contaminated soils [18,19]. Tween 80, a nonionic surfactant used in the food industry, was tested for enhanced EK remediation of dichlorodiphenyltrichloroethane (DDT) [20], hexachlorobenzene [21], perchloroethylene [22], and polycyclic aromatic hydrocarbons (PAH) [23,24]. Both surfactants were also tested simultaneously and individually for the electro-Fenton degradation of phenanthrene in marine sediment [25]. The electrodialytic remediation of PAH with Tween 80 in spiked and contaminated soils was also tested by Lima et al. [26].

In this work, we tested two different surfactants (saponin and Tween 80) to enhance the electroremediation of PCB contaminated soil in the two-compartment electrodialytic (ED) setup developed at the Technical University of Denmark (DTU) [27], in which the soil is suspended and stirred simultaneously in combination with the addition of nZVI. The main objectives were to: (i) compare the effectiveness of the two surfactants for increasing PCB desorption and the subsequent dechlorination; (ii) evaluate the need of using direct electric current in the reactor with suspended soil and nZVI; (iii) assess the potential inhibitory effect of the surfactant in the nZVI reactivity.

2. Materials and methods

2.1. Chemicals and solvents

PCB standards were analytical grade, obtained from Fluka, Sigma–Aldrich (PCB 28, 52, 101, 138, 153, 180 and 209) and Ultra-scientific (PCB 30; PCB 65 and PCB 204). The solvents hexane and acetone were Pestinorm (VWR BDH Prolabo). The surfactants Tween 80 (Sigma Aldrich) and saponin (GPR Rectapur) were lab grade (Table 1). Hydrochloric (37.6%), nitric (65%) and sulfuric (95–97%) acids were trace metal grade. Anhydrous Na_2SO_4, $KMnO_4$, NaCl, and silica gel (silicic acid) were lab grade. Silica gel was cleaned up before use according to the USEPA method 3630C. The water was deionized with a Milli-Q plus system from Millipore (Bedford, MA, USA). A nZVI slurry-stabilized suspension (NANOFER 25S, NANO IRON, s.r.o., Rajhrad, Czeck Republic) was used in the experiments (Table 2).

Table 2
Characterization of the zero valent iron nanoparticles used in the experiments, according to the supplier information.

Product Name	NANOFER 25S (NANO IRON, s.r.o.)
Stabilizer	Polyacrylic acid (PAA)
pH	11–12
Suspension density	1.15–1.25 g cm^{-3} (20 °C)
Average particles size	50 nm
Particle size distribution	20–100 nm
Average surface area	20–25 m^2 g^{-1}
Iron content	80–90 wt.%

2.2. Soil characterization

The contaminated soil used in the experiments was provided by a hazardous waste operator in Portugal and is a mixture of contaminated soils from industrial sites with transformers oils spills. Table 3 presents the physical and chemical characteristics of the soil used in the experiments. The soil was homogenized, air dried and sieved, and only the particles with size <2 mm were used in the experiments.

Table 3
Physical and chemical characteristics of the soil.

Parameter	
Particle size distribution (%)	
Coarse sand (200 < Ø < 2000 μm)	19.1
Fine sand (20 < Ø < 200 μm)	67.3
Silt (2 < Ø < 20 μm)	12.7
Clay (Ø < 2 μm)	0.9
Textural classification	Loamy sand
pH (H_2O)	12.2
Conductivity (mS cm^{-1})	18.76
Exchangeable cations ($cmol_{(c)}$ kg^{-1})	
Ca^{2+}	83.75
Mg^{2+}	3.2
K^{+}	26.88
Na^{+}	9.37
Sum of exchangeable cations ($cmol_{(c)}$ kg^{-1})	123.2
Calcium carbonate (%)	18.0
Organic matter (%)	16.46
Total PCB[a] (μg kg^{-1})	258 ± 24
Metals[b] (mg kg^{-1})	
Al	20,980 ± 590
As	9 ± 2
Cd	0.7 ± 0.1
Cr	52 ± 3
Cu	142 ± 95
Fe	13,162 ± 301
Ni	32 ± 1
Pb	45 ± 3
Zn	2155 ± 40

[a] Sum of PCB28, 30, 52, 65, 101, 138, 153, 180, 204 and 209.
[b] Acid digestion with HNO_3 according to the Danish Standard DS259.