



Treatment of *ex-situ* soil-washing fluids polluted with petroleum by anodic oxidation, photolysis, sonolysis and combined approaches



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HIGHLIGHTS

- SAWS is an efficient approach to depollute petroleum contaminated soils.
- Depollution intensification is attained by US/BDD-electrolysis and photo/BDD-electrolysis.
- Significant differences in efficiency between single and coupled treatment process.
- Oxidants activation by US/BDD-electrolysis and photo/BDD-electrolysis is attained.

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ABSTRACT

In this research, the treatment of soil spiked with petroleum was studied using a surfactant-aided soil-washing (SASW) process followed by sonolysis (US), photolysis and boron doped diamond electrolysis (BDD-electrolysis) for washing liquid treatment. Results clearly demonstrate that SASW is a very efficient approach in the treatment of soil, removing completely the petrochemical compounds by using dosages about 5 g of extracting surfactant (sodium dodecyl sulfate (SDS)) per kg of soil. The main characteristics of the effluents produced in this soil remediation technology as well as the efficiency of the treatment (US, photolysis and BDD-electrolysis) depend on the dosage of SDS. Depollution of the effluents (degradation and mineralization of the organic matter) is related to the reduction in size of micelles formed by SDS and petroleum, and it depends on the treatment used. US and photolysis were inefficient decontamination processes, while BDD-electrolysis favors the complete depletion of micelles. However, the intensification of the efficiency was attained by synergic degradation effects when UV light irradiation and US were coupled with BDD-electrolysis, US/BDD-electrolysis and photo/BDD-electrolysis, respectively. Sulfate (coming from SDS) ions play an important role during the BDD-electrolysis, US/BDD-electrolysis and photo/BDD-electrolysis because persulfate and persulfate radicals are produced (by sulfate activation applying US or photolysis), improving the efficiency of the processes.

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1. Introduction

Recently, an increasing interest in the remediation of soil and water polluted with petroleum has been attained [1–3]. Petroleum refining industries converts crude oil into more than 2500 petroleum products including gasoline, kerosene, aviation fuel, diesel fuel, lubricating oils, etc. World oil demand is expected to rise to 107 thousand barrels per day over the next two decades, and oil will account for 32% of the world's energy supply by 2030 [4].

Nevertheless, the petrochemical activities by industries have generated serious environmental problems in water and soil ecosystems. In the former, the petrochemical wastewaters generated contains many chemicals, such as benzene, volatile phenol, sulfides, ammonia, suspended solids, cyanides, nitrogen compounds and heavy metals. Meanwhile, hydrophobic organic compounds (HOCs) such as petroleum hydrocarbons, polychlorobiphenyls (PCBs) and polycyclic aromatic hydrocarbons (PAH) are extremely difficult to remove from the later [5]. The persistence of HOCs in soils is a matter of significant public, scientific and regulatory concerns because of their potential toxicity, mutagenicity, carcinogenicity and ability to be bioaccumulated in the food chain [5,6].

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Most of them are persistent in the natural environment due to their slow degradation by natural attenuation or by (photo) chemical/biological processes [5,7,8].

Low solubility in water, high octanol/water partition coefficient, a high organic carbon/water partition coefficient and volatility influence on the accumulation, mobility and availability of HOCs in the soil [8,9] as well as the efficiency of their removal during the treatment. Therefore, several soil treatments have been developed in the last years, however, these processes have at least one important drawback such as high costs (thermal treatments), high perturbation of the soil texture (thermal treatments), low efficiency (pump and treat), long treatment time requirements (biodegradation processes), or selectivity toward target pollutants (volatile organic compounds for venting, hydrophilic organic compounds for pump and treat) [5]. In this frame, more efficient, less expensive and ecofriendly approaches were technologically advanced in the last years [5,10]. Soil washing (*ex-situ* process) or soil flushing (*in-situ* process) procedures were developed using extracting agents (surfactants, biosurfactants, cyclodextrins (CDs), cosolvents) in order to enhance the solubility, desorption and biodegradation of soil pollutants [5]. After that, a treatment stage of the oil washing/soil flushing solutions is carried out [10–19].

Nevertheless, the effluents obtained after the soil depollution are difficult to be treated due to the higher organic load (pollutants and extracting agents) and generally, a partial decontamination is attained [5]. For this reason, the main objectives of the present research are to evaluate the efficacy of single technologies (BDD-electrolysis, sonolysis (US) and photolysis) to depollute soil washing fluid obtained from petroleum decontamination of soils by surfactant-aided soil washing (SASW) [14–20] as well as the study of the intensification of the efficiency on the degradation of this effluent by coupling US and photolysis with BDD-electrolysis (US/BDD-electrolysis and photo/BDD-electrolysis).

2. Materials and methods

2.1. Chemicals

Petroleum (American Petroleum Institute (API) gravity >30, it consists basically of alkanes, and approximately 15–25% of cycloalkanes) and hexane were obtained from Sigma–Aldrich (Spain). Sodium dodecyl sulfate (SDS) and sodium hydrogen carbonate (NaHCO₃) were obtained from Panreac. Deionized water (Millipore Milli-Q system) was used to prepare all solutions.

2.2. Preparation of simulated soil

Spiking soil samples with organic compounds is the most frequently contamination method used in the lab-scale studies [5,21], for this reason, a model soil was chosen and it was polluted with petroleum. The model soil used in this work was kaolinite. Samples of polluted soil were prepared by dissolving a known amount of petroleum in hexane and then mixing this petroleum/hexane solution with soil. The spiked clay was aerated for 24 h to evaporate the hexane and, in this way; the petroleum was homogeneously distributed on the clay surface. The resulting petroleum concentration in the soil was around 1000 mg/kg-soil. Kaolinite was provided by Manuel Riesgo Chemical Products (Madrid, Spain), and it is not reactive and has low hydraulic conductivity, low cation exchange capacity and no organic content, and for this reason, it is frequently used as a model of low-permeability soils. Properties of this synthetic clay soil were provided by the commercial supplier, and are detailed in Table 1.

2.3. Analytical techniques

The total organic carbon (TOC) concentration was monitored using a Multi N/C 3100 Analytik Jena analyzer. The organic removal was monitored through the COD content during the treatment by single technologies (BDD-electrolysis, US and photolysis) and coupled electrochemical approaches (US/BDD-electrolysis and photo/BDD-electrolysis) using a HACH DR2000 analyzer. Zeta potential (ζ -potential) was also measured for the clarified liquid using a Zetasizer Nano ZS (Malvern, UK). Measurements of pH were carried out with an InoLab WTW pH-meter. The particle size was monitored during the treatment approaches with a Mastersizerhydro 2000SM (Malvern). The anions (SO₄²⁻, Cl⁻, ClO⁻ and ClO₃⁻) present in the target wastewater were characterized using ion chromatography by means of a Shimadzu LC-20A system [18].

2.4. Ex-situ soil washing procedure

Ex-situ soil washing [5] with surfactant fluid solution was carried out in a stirred tank operated in batch mode. The tank volume was 1 dm³. Low-permeability soil (1000 g) polluted with 1000 mg petroleum per kg of soil and 0.8 dm³ of solubilizing agent (containing deionized water, 500 mg/dm³ of NaHCO₃, and different concentrations of SDS surfactant ranging from 100 to 5000 mg/dm³) were mixed in the reactor for 6 h at a stirring rate of 120 rpm. Thus, the same tank then acted as a settler (during 24 h) to separate the soil from the effluent. These effluents consisted of complex aqueous mixtures of petroleum, surfactant and sodium hydrogen carbonate, with emulsified micro drops and soluble species. As it will be discussed below, only two kinds of effluents will be treated by BDD-electrolysis, US, photolysis, US/BDD-electrolysis and photo/BDD-electrolysis technologies and their characteristics are reported in Table 2.

2.5. Bulk treatment of the soil washing effluents by BDD-electrolysis, photolysis, US, US/BDD-electrolysis and photo/BDD-electrolysis

Three different treatment approaches were employed as follows: BDD-electrolysis (application of electric current), photolysis (UV irradiation) and US (ultrasound irradiation). Ultrasound or photocatalysis processes combined with BDD-electrolysis are considered as emerging and interesting integrated technologies with clear advantages over conventional technology for removing organic pollutants from water [17,18,22–24]. Photolysis and photo/BDD-electrolysis were performed by using a UV lamp VL-215MC (Vilber Lourmat), $\lambda = 254$ nm, intensity of 930 W/cm² and energy 4.43–6.20 eV irradiating 15 W directly to the quartz cover. [17]. US and US/BDD-electrolysis were carried out by employing an ultrasound generator UP200S (Hielscher Ultrasonics GmbH, Germany) equipped with a titanium glass horn of 40 mm diameter, length 100 mm, emitting 24 kHz and maximum ultrasonic power 200 W [18]. Electrochemical oxidation experiments were carried out in a bench-scale plant with a single-compartment

Table 1
Properties of kaolinite: a low permeable model soil.

Mineralogy	Particle size distribution		
Kaolinite	100%	Gravel	0%
Fe ₂ O ₃	0.58%	Sand	4%
TiO ₂	0.27%	Silt	18%
CaO	0.10%	Clay	78%
K ₂ O	0.75%	Specific gravity	2.6%
SiO ₂	52.35%	Hydraulic conductivity	1 × 10 ⁻⁸ (cm/s)
Al ₂ O ₃	34.50%	Organic content (%)	0%
Others	11.42%	pH	4.9

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