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Review Article

Prospects on integrated electrokinetic systems for decontamination of soil polluted with organic contaminants

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María Ángeles Sanromán

Abstract
Overall, investigations about the utilization of electrokinetic technology alone or in combination with other processes have attracted particular attention in recent years for remediation of soils contaminated with heavy metals and organic compounds. This fact is due to its peculiar benefits together with its capability of operating in a fine and low-permeability matrix. This review aimed to ascertain the most recent developments on the commonly proposed integrated technologies (electrokinetic soil washing, electrokinetics coupled with permeable reactive barriers, electrokinetic-advanced oxidation processes, and bioelectrokinetic remediation), by evaluating the gaps, challenges, and trends of these systems in the last years. Special attention is paid to the current approaches for overcoming the main bottlenecks of electrokinetics concerning scale-up and reduction of electric energy consumption by integration of renewable energies.

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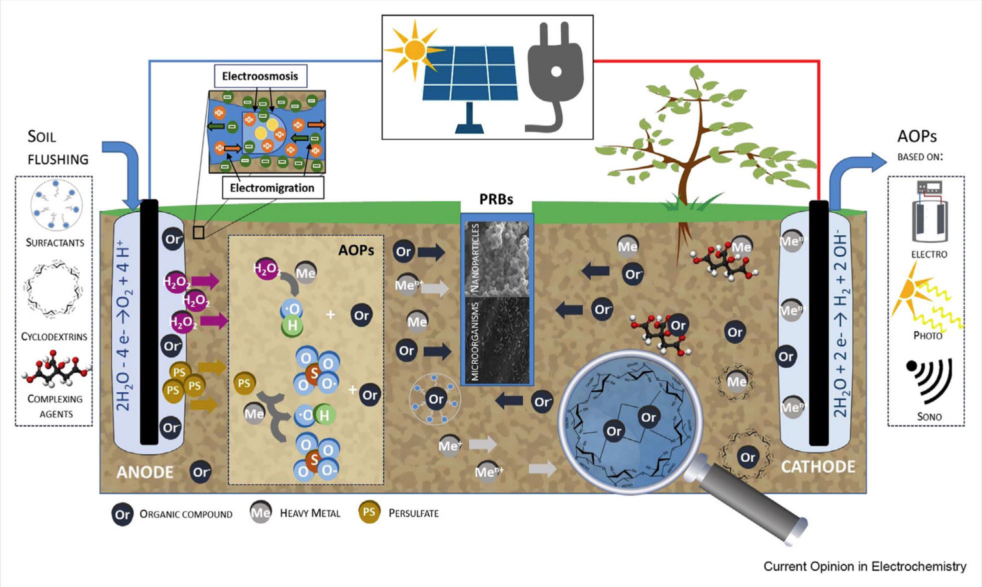
Soil washing, Permeable reactive barriers, Advanced oxidation processes, Bioremediation, Scale-up.

Introduction

Environmental decontamination is a noteworthy issue in the current context of gradual environmental awareness and tightening of related legislation. It is crucial to have appropriate green low-cost methodologies for restoration of soil, sediments, and wastewater. In this regard, the combination of processes can be an attractive possibility because the synergetic effect between them may enhance the elimination levels achieved individually. In the recovery of polluted soils and sediments, the application of electrokinetic (EK) remediation has been proposed as a feasible alternative. EK remediation is a widely recommended technique for the treatment of low permeable soils polluted with heavy metals and organic compounds. It is based on an electrochemical process in which the electric current is used for extracting species from the soil. Usually, a direct low-intensity current or a gradient of low potential is applied to a couple of inert electrodes that are inserted in the soil, as per the scheme shown in Figure 1. As a consequence of the applied potential gradient, the pollutants are transported within the electrical field toward the anode or cathode. Interstitial water or fluid introduced from the exterior, termed process fluid, is used as the conductive means. Availability of an open flow between the electrodes allows the process fluid to circulate inside and out of the pores of the solid medium. The transport of chemical species induced in a porous medium during electrochemical treatment is mainly based on EK processes. The specific mechanisms in which mobilization of the pollutants takes place are electromigration (mobilization of pollutants in ionic form in favor of the electrical field), electro-osmosis (movement of the liquid in relation to the solid surfaces of the electrical field, normally in the direction from the anode to cathode), and electrophoresis (displacement of charged colloidal particles in suspension) [1**,2].

Organic compounds are transported through the soil by electromigration when they are electrically charged and by electro-osmosis if they are not ionic (Figure 1). As shown in Figure 2, it has been determined that by applying an electric field, transport of an organic compound can be achieved, specifically a soluble dye, from the soil to the anode chamber (electromigration) or cathode (electro-osmosis) depending on whether the

Figure 1



Current Opinion in Electrochemistry

Scheme of the EK and different EK-based systems, including main transport mechanisms and proposed *ex situ* AOP treatments. AOP, advanced oxidation process; EK, electrokinetic; PRB, permeable reactive barrier.

dye is anionic (Figure 2a) or neutral (Figure 2b), respectively [2].

Although research demonstrates high removal efficiencies for EK processes, several bottlenecks arise in their practical application at real scale owing to the limitation of electro-osmotic flow, pH jump, low solubility of pollutants, soil heterogeneity, and so on. In this sense, it is interesting to favor electro-osmotic flow, which is influenced by the current intensity, pH, the type and concentration of ionic species, and the type of soil. Typically, the range of pollutant recovery hinges on the pH profile of the soil that affects the solubility and the mobility of the present species. The pH changes happen at diverse EK periods and in different soil positions ascribable to the generation of acid and basic fronts in the soil after water electrolysis. To reduce its negative effect, smoothing the pH jump into the soil, several strategies have been proposed such as pH control or polarity reversal [1**,2]. This review reports recent studies focused on an integrated set of techniques to enhance the EK efficiency in *in situ* and *ex situ* soil decontamination by its combination with soil washing, stabilization, thermal remediation, advanced oxidation processes (AOPs), or bioremediation.

EK soil washing

EK soil washing is considered a promising technology for removing pollutants from solid matrixes by their solubilization in an aqueous medium and displacement through the soil by the action of the electric field [3]. However, in the case of hydrophobic organic pollutants, low water solubility and neutrality of the molecules reduce transport capability out of the soil, requiring addition of extracting or solubilizing agents to washing solution or flushing fluid to solubilize the pollutants from the soil and enhance the remediation process. The commonly selected solubilizing agents are surfactants (synthetic surfactants and biosurfactants), cyclodextrins, humic acids, organic solvents, chelants, vegetable oils, and organic cosolvents [4**].

Addition of these agents, being used alone or mixed is linked to contaminants present in the soil. Thus, Fardin et al. [5*] tested anionic and nonionic surfactants, sodium dodecyl sulfate (SDS), and Tween 80, respectively, at different concentrations to remediate kerosene-contaminated soils, determining that simultaneous addition of both surfactants allowed a more homogeneous kerosene distribution, showing substantial removal levels (around 65%) than EK processes (40%). Another nonionic surfactant typically used to dissolve and mobilize pesticides as organochlorine from the soil is Triton X-100 [6], and several studies reveal a maximum pollutant recovery from the soil when a concentration of 10% of surfactant was used [6,7]. Saberi et al. [8] evaluated Tween 80 and Brij 35 as nonionic surfactants and ethylenediaminetetraacetic acid as the chelating agent, concluding that it is possible that remotion of heavy metals such as Pb, Zn, and Ni in parallel with polycyclic aromatic hydrocarbons (PAHs) from a low-permeability spiked soil, attaining the best results (61%) when Brij 35 was added. Previously, Ammami et al. [9**] studied heavy metal and PAH removal from an aged model polluted sediment. In their study, nitric and citric acids were used to favor the removal of the metal, avoiding the formation of an alkaline front, while the surfactant SDS or Tween 20 was added to solubilize and mobilize PAHs. The best results were achieved when nitric acid was used, and similar results were attained when a more eco-friendly mixture of Tween 20 and citric acid was selected as flushing flow, whereas SDS mixed with citric acid was not a suitable aspirant.

Nowadays, synthetic surfactants have been replaced by natural eco-friendly solubilizing agents such as rhamnolipids or saponin. Thus, decontamination of pyrene-contaminated soil is feasible by augmenting the concentration of rhamnolipids (from 0.5 up to 2 g/L), with a positive effect when the system is operated with pH control. Another alternative is the use of cyclodextrins. They are cyclic oligosaccharides of glucopyranose units linked by α-(1,4) bonds capable of forming complexes with contaminants (organic or inorganic) (Figure 1), including them into their hydrophilic shell and a nonpolar toroidal-shaped cavity in the center [10]. The most commonly used cyclodextrins are hydroxypropyl-β-cyclodextrin and methyl-β-cyclodextrin (MCD), which are mainly applied in remediation of soils polluted with PAHs [11]. Recently, Zhang et al. [12*] evaluated the increase of solubility and mobility of decabromodiphenyl ether (BDE209) in a low-permeability soil by addition of hydroxypropyl-β-cyclodextrin. However, the main limitation of these agents is their price, which increases the operating costs of EK soil washing. To overcome this problem, the recovery of cyclodextrins or the *in situ* generation of β-cyclodextrin in soil from starch has been proposed [10,13].

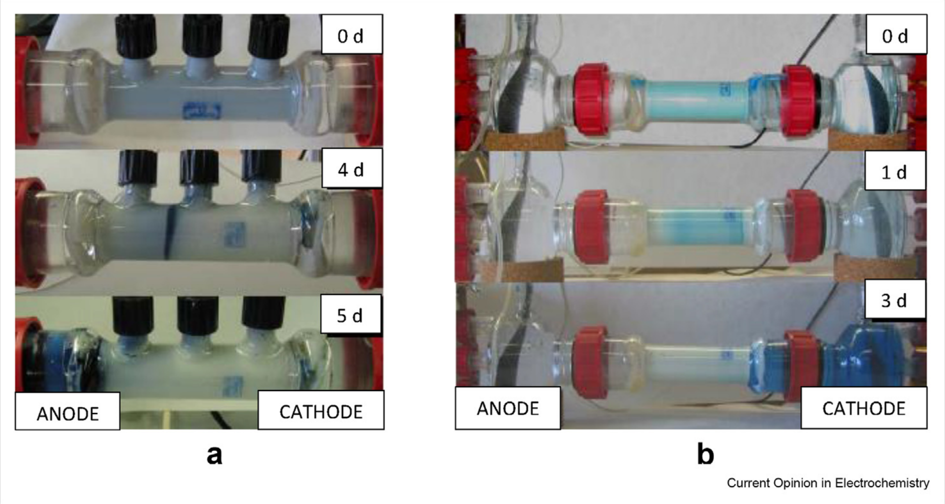
EKs coupled with permeable reactive barriers

This integrated technology is based on the location of a permeable reactive barrier (PRB) perpendicular to the flow of pollutants from the anode to cathode in which they may be degraded or sequestered [14**]. This technology has been vastly applied in remediation of soil polluted with heavy metals [15]. In this passive technology, the key factor is the selection of reactive materials. In the literature, several materials are reported for the construction of the PRB such as elemental metals, adsorbents with high surface area, ion-exchange resins, or microorganisms [16]; however, as summarized in Table 1, the most widely used reactive materials in EKs coupled with the PRB for decontamination of polluted organic soils are iron-based materials and activated carbons. From the results of these studies, it can be concluded that this technique could be considered as a rapid treatment that reduces the riskiness of pollutants present in the soil, converting them into less hazardous species or fixing them in the reactive material used. Nevertheless, to improve the behavior of this PRB, several factors such as soil pH or the solubility of pollutants must be considered. Therefore, several solubilizing agents were added, and enhancement strategies as polarity reversal were applied to control the pH in the soil and improve the electro-osmotic flow (Table 1).

Electrokinetic-AOPs

In the last decades, AOPs have been considered one of the most useful treatments for removal of hazardous and recalcitrant compounds. These processes are based on

Figure 2



EK evolution on the remediation process of kaolinite polluted with different dyes: **(a)** anionic dye (Reactive Black 5); (**b**) neutral dye (lissamine green) [2]. EK, electrokinetic.

Table 1

Representative examples of application of EKs coupled with the PRB in removal of organic contaminants in soil.

Soil	Pollutants	Processing fluid	PRB	Highest removal	Observations	Reference
Kaolinite	Cu and phenantrene	Acetic acid and Brij30	Recyclable food scrap ash	83.86% 90.17%	1 V/cm	[17]
Spiked natural soil of a quarry located in Mora (Toledo, Spain)	Lindane	Sodium dodecyl sulfate	Nanoparticles of zero-valent iron (ZVI) Granular particles of ZVI Granular activated carbon	51% 68% 80%	Soil volume 139.6 dm^3 1 V/cm	[18**]
Artificially contaminated soil	Phenanthrene 2,4,6-trichlorophenol	–	Iron and activated carbon	69.86% 71.53%	1 V/cm	[19]
BDE209-spiked soil	Decabromodiphenyl ether (BDE209)	Hydroxypropyl-β-cyclodextrin, sodium dodecyl sulfate, and humic acid	ZVI	28–30%	2 V/cm	[20**]
Kaolinite	Atrazine and oxyfluorfen	Sodium dodecyl sulfate	Granular activated carbon	90%	Polarity reversal 1 V/cm	[21**]
Silty loam soil sampled near an aluminum smelting plant located in Xinan County (China)	Fluorine	Distilled water and NaOH solution as the catholyte and anolyte, respectively	Modified activated alumina by calcium chloride	76%	2 V/cm	[22]
Clay soil from a farmland in Northern Iran	Perchloroethylene	Triton X-100	Microscaled ZVI	80%	Polarity reversal 1 V/cm	[23]
Spiked soil	Diesel	Tap water	Acclimated diesel degrading microorganisms supported on gravel	26.8%	Polarity reversal 1 V/cm	[24**]

EK, electrokinetic; PRB, permeable reactive barrier.

the formation of strong oxidants with the potential to degrade and mineralize organic pollutants [25]. Several examples have been reported in the literature based on *in situ* generation of hydroxyl radicals (HO•) and sulfate radicals ($SO_4\bullet^-$) that are highly reactive intermediates capable of reducing or eliminating a wide range of pollutants. $SO_4\bullet^-$ could be produced from persulfate (PS) via several activation paths: heat, transition metals, ultraviolet light, and electrochemical techniques [26*]. Thus, through activation of PS or H_2O_2, the radicals are generated by the following reactions:

$$H_2O_2 + M^{n+} \rightarrow HO\cdot + HO^- + M^{(n+1)} \tag{Eq (1)}$$

$$S_2O_8^{2-} + M^{n+} \rightarrow SO_4^{\cdot -} + M^{(n+1)} + SO_4^{2-} \tag{Eq (2)}$$

$$2S_2O_8^{2-} + Fe^0 \rightarrow 2SO_4^{\cdot -} + Fe^{2+} + 2SO_4^{2-} \tag{Eq (3)}$$

$$S_2O_8^{2-} + e^- \rightarrow SO_4^{2-} + SO_4^{\cdot -} \tag{Eq (4)}$$

As previously mentioned, by means of the electric field, the oxidant or reactants can be transported by electromigration and electro-osmotic flow into the soil between the anode and cathode chambers, and they can react with transition metals added to or present in the soil [27**]. EK-Fenton process is a promising approach owing to the generated radicals possessing oxidizing properties and a great capacity for degradation of a wide range of organic pollutants such as pesticides, dyes, ionic liquids, or hydrocarbons [28**,29**,30**]. Recently, Paixão et al. [31**] investigated decontamination of soils spiked with petroleum via EK process using for the Fenton reaction H_2O_2 in the electrode chambers and using iron electrodes. The control of soil pH and the supplementation with citric acid were performed to favor oxidative reactions in the soil. In other studies, addition of external nanocatalysts such as magnetite iron oxide (Fe_3O_4) can be carried out for enhancing soil remediation [32]. Generally, the control of pH in electrode chambers and supplement of solubilizing agents and electrolytes can improve the EK-Fenton process because these strategies boost the solubility of pollutants and the yield of oxidation reactions [33].

In case of PS activation, production of SO_4^{2-} ion, which can be transported by electromigration, increases the electric current [34*]. Removal efficiency of these kinds of oxidative processes can be enhanced by integration of several strategies such as pH control or by supplementation with facilitating compounds. In line with the theory, addition of citric acid and MCD in anolyte solution during EK-PS remediation of a spiked soil with Cu and the flame retardant BDE-209 improved the process. Therefore, by addition of these facilitating compounds, a significant removal of both pollutants was achieved owing to the generation of citrate—metal and MCD—metal complexes, increasing the electro-osmotic flow and electric current [34*]. Availability of organochlorine pesticides can be enhanced by addition of surfactants such as Tween 80 and N,N-dicarboxymethyl glutamic acid tetrasodium, upgrading the oxidation reactions in the material under EK-PS remediation [35]. Similarly, addition of Tween 80 increased pyrene removal efficiency by dissolving metal ions in the soil, in which no significant difference was observed between the voltages of 1 and 1.5 V/cm, reaching complete removal efficiency when PS was added at a concentration of 100 mg/kg [36].

The effect of electrode configuration on EK processes has also been evaluated in several studies using an equal and unequal number of anodes and cathodes, named as one-dimensional (1D) and two-dimensional (2D) electrode configurations [37**]. Several 2D electrode configurations with linear rows of electrodes and surrounding electrode arrangements have been reported to improve removal efficiency of organic pollutants [38,39]. Xu et al. [37**] evaluated several 2D surrounding electrode configurations around a cathode (triangular, square, and hexagonal) and compared with conventional 1D configuration using Ti/RuO_2 alloy sheets as electrodes. In all the configurations tested, the transport of the oxidant toward the cathode was confirmed by regular addition of $Na_2S_2O_8$ solution in the anode reservoir. Hexagonal electrode configuration provided higher PAH removal efficiency (40.9%) than other 2D and 1D configurations (maximum = 31.6%), whereas energy consumption was the lowest (182.9 kWh/t) [37**]. Another alternative to enhance this treatment is inclusion of ion-exchange membranes in the cathode chamber to reduce the negative effect of the redox reaction between PS and reductive H_2 and redundant electrons and adverse effects on soil properties [40**].

Lately, another strategy that has gained growing interest among the scientific community is the sequential treatment via EK soil washing, followed by *ex situ* AOPs (Figure 1). In this case, it is possible to extract organic pollutants from the soil by EK processes using extracting agents, and the total removal of these target pollutants and high mineralization is achieved by *ex situ* degradation by electrochemical, photoelectrochemical, and sonoelectrochemical techniques using boron doped diamond (BDD) anodes [41*,42].

Bioelectrokinetic remediation

Although *in situ* bioremediation is a cost-effective treatment, the long treatment time required to clean

Table 2

Studies reported in the last year on removal of soil polluted with organic contaminants by BEK remediation.

Soil	Pollutants	Microorganisms	Observations	Reference
Spiked soil located in Serkkadu (Tamil Nadu, India)	Diesel	*Staphylococcus epidermidis* EVR4	Biosurfactant and catabolic enzyme production	[46]
Spiked clayey soil supported by Millas Hijos Ceramics (Toledo, Spain)	2,4-Dichlorophenoxyacetic acid	*Rhodococcus ruber* and *Ochrobactrum anthropi* isolated from the oil refinery wastewater treatment plant (Puertollano, Spain)	Polarity reversal. Electrolytes simulate groundwater: inorganic medium with Na_2SO_4, $NaHCO_3$, and $NaNO_3$	[47**]
Artificial petroleum–contaminated soil	Oil from the Yanchang Oilfield (China)	Petroleum-degrading strains isolated from oil-polluted soil: *Delftia* sp., *Marinobacter* sp., *Bacillus* sp.	Total petroleum hydrocarbons, soil temperature, and electric conductivity; SO_4^{2-}, Cl^-, and K^+ explain the changes in the microbial community at 0–21 d and pH and NO_3^- at 63–98 d	[48**]
Spiked soil from Pretoria (South Africa)	Engine oil	*Pseudomonas aeruginosa* PA1 from tank sludge in South Africa by selective enrichment to obtain hydrocarbon degraders	Correlate the relationship between voltage, electrode distance, and biosurfactants on the efficacy of decontamination. Pulsating electrode configurations	[49,50]
Oil samples collected from a petroleum-polluted area of Muntenia (Romania)	Petroleum	Two indigenous *Pseudomonas* sp.	Polarity reversal 1 V/cm. Negative impact on bacterial abundance at short treatment time. Promising results by integration of bioremediation treatment downstream of EK process	[51]
Spiked soil collected from Thiruvalluvar University campus, Serkadu, Vellore district (India)	Crude oil	*Bacillus subtilis* AS2, *Bacillus licheniformis* AS3, and *Bacillus velezensis* AS4	Biosurfactant production and addition. Degraded higher-molecular-weight hydrocarbons (C_8 to C_{28})	[52]
Spiked soil collected in an organic tomato farm in São Nicolau, Santarém (Portugal)	10 different emerging contaminants: 17β-estradiol, sulfamethoxazole, bisphenol A, ibuprofen, 17α-ethinylestradiol, oxybenzone, diclofenac, triclosan, caffeine, and carbamazepine	Biotic and abiotic tests	pH alterations decreased bioremediation efficiency and inhibited electrodegradation near the cathode	[53*]
Spiked soil collected from University of Mining and Technology, Xuzhou (China)	Petroleum resins	Microbial consortium: *Rhizobium* sp., *Arthrobacter globiformis*, *Clavibacter xyli*, *Curtobacterium flaccumfaciens*, *Bacillus subtilis*, *Pseudomonas aeruginosa*, and *Bacillus* sp.	Contribution of inoculated microbes was higher than that of indigenous counterparts	[54]

BEK, bioelectrokinetic.

up the soil or sediments is a drawback of this technique. This fact could be due to several aspects that limit the interaction among microorganisms, nutrients, and pollutants. The application of electrochemical techniques increases the temperature inside the soil. It can improve the supply of nutrients and promote production of natural biosurfactants that increase the solubility of these pollutants and concurrently mobilize the degradation products through the soil [43*]. In addition, the application of EK processes to soils allows mobilization of metals, which are usually toxic for microorganisms, toward electrode chambers by electromigration, increasing the effectiveness of bioremediation technique [44]. Thus, bioelectrokinetic (BEK) remediation is more seamless than EK technique owing to addition of microorganisms [45,46*]]. In Table 2, several recent studies on BEK remediation demonstrated great potential of this integrated technique. The increase of biosurfactant concentration has a great influence on oil decontamination [55], and in other strains such as *Staphylococcus epidermidis* EVR4, a synergistic effect with catabolic enzymes produced (dehydrogenase, catalase, and cytochrome C oxidase) by this bacterium was detected [46*]. Besides, several factors such as pH or concentration of pollutants and inoculum are correlated with degradation of pollutants present in the soil. In this context, strategies such as polarity reversal could contribute to maintaining correct soil pH values [47**], or a new strategy using the anode and cathode in the same compartment at each end of the soil could stabilize the pH and facilitate distribution of nutrients across the soil during BEK remediation [56]. Thus, recent innovations in BEK remediation will render possible efficient remediation of low-permeable polluted soil.

Scale-up of EK processes

Numerous examples of EK studies performed on small-scale laboratory devices have been detailed in the literature; however, scarce field-scale research studies have been reported. This is a significant drawback and one of the most important bottlenecks that EK process needs to overcome. The knowledge of scaling up of process is vital for its implementation on a real scale. In this sense, in this section, several recent examples are summarized to understand the scale-up process that is essential for applying EK process at the field scale. Benamar et al. [57**] investigated at two scales (0.4 kg and 40 kg) the use of an eco-friendly mixture of Tween 20 and citric acid as a promising multidecontamination strategy for dredged sediment [9**]. They stated that the electric field on a large scale could not fully control transport phenomena, and the effect of sediment heterogeneity and inertial effects reduce the effectiveness of EK processes. Several setup scales, laboratory, bench, pilot, and prototype (from 1×10^{-4} up to 21.76 m^3), have been evaluated, providing useful guidance for developing a scale-up of EK processes [58**].

Energy consumption is the cornerstone of EK process scale-up. To promote the use of these technologies in the field scale, it is necessary to reduce the energy cost by using renewable energy sources. In this line, the EK process powered by solar photovoltaic panels has been evaluated both on laboratory and pilot scales for remediation of soils polluted with organic and heavy metals/toxic anionic polluted soils [59**]. Hassan et al. [43**] used solar panels to generate renewable energy in a semipilot BEK process (L400 mm × W300 mm × H180 mm) for diesel fuel removal using novel bacterial strains with an electrode configuration to stabilize soil pH and water content. In these tests, the produced current depended on the weather conditions at solar time. Thus, voltages reached 30 V at 10:00 AM, keeping constant until 3:00 PM and decreasing to 0 V at sunset. Nevertheless, the solar panel can generate sufficient direct current for BEK remediation.

Concluding remarks

A comprehensive review of the latest research studies on integrated EK processes for removal of organic pollutants present in the soil has been conducted. The most common technologies, EK soil washing, EKs coupled with the PRB, EK-AOPs, and BEK remediation, were discussed. Generally, use of strategies such as control of soil pH and addition of solubilizing agents to enhance solubility of heavy metals and organic pollutants improves all the integrated EK processes. However, the vast variety of pollutants and the different characteristics of contaminated soils requested a preliminary evaluation to determine the best operational conditions. Finally, it is envisaged that this review aims to summarize the latest trends in EK remediation of soil, considering the reduction of energy cost by application of renewable energy sources and new approaches concerning its scale-up.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this article.

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