

A review on enhancement techniques of electrokinetic soil remediation

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ABSTRACT: During the last decade, the expansion of urbanization and industrial activities caused serious environmental problems such as soil pollution. Novel technologies are required to remediate soil contamination with toxic contaminants, including heavy metals and organic pollutants. In recent years, there have been increasing attempts to improve soil remediation efficiency. Electrokinetic (EK) is an emerging remediation technology for mixed contaminants, especially in low hydraulic conductivity soils. During EK remediation process, an electric field is applied to the specimen to remove the contaminants from soil by means of electro-osmotic (EO), electro-migration (EM), and electrophoresis (EP) processes. The usual EK technique may have no suitable removal efficiency or contaminants migration. Thus, enhancement techniques or their combination with other technologies are required to overcome EK limitations, increasing its efficiency. The present study reviews seven main enhancement techniques combined with EK; they include using surfactant, chelants, complexing agents, soil pH control, bioremediation, permeable reactive barriers, and ultrasonication. It should be mentioned that, selecting the suitable combination technique highly depends on the soil type and the kind of contaminant.

Keywords: electrokinetic, enhancement techniques, soil remediation, Zeta potential.

INTRODUCTION

Soil contamination is an important environmental issue due to anthropogenic activities and inefficient waste handling techniques (Virkytyte et al., 2002; Gomes et al., 2012; Pedersen et al., 2016; Habibul et al., 2016). Organic and inorganic contaminants such as heavy metals and Organochlorines, present in the soil matrix, which are migrated to the groundwater may lead to harmful negative impacts (Gomes et al., 2012; Yeung et al., 2011; Valdovinos et al., 2016; Mena et al., 2016).

Moreover, contaminated soils have potential adverse effects on plants, animals, and humans. Thus, soil remediation is considered a crucial environmental issue, investigated by many researchers (Darban et al., 2009; Saeedi et al., 2009; Zanjani et al., 2012; Kim et al., 2009; Moghadam et al., 2016; Figueroa et al., 2016; Estabragh et al., 2016; Ng et al., 2015; Masi and Losito, 2015; Li et al., 2014; Jeon et al., 2014; Naidu et al., 2013). The situation worsens when the contaminated soil has low hydraulic permeability and/or complex composition of clay and fine-grain soils (Moghadam et al., 2016; Darban et al.,

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2009; Saeedi et al., 2009; Chen et al., 2013; Guedes et al., 2014; Annamalai et al., 2015). Electrokinetic (EK) remediation is considered an emerging technology that has already proven its value, especially in low permeability contaminated soils (Gomes et al., 2012; Darban et al., 2009; Saeedi et al., 2009; Zanjani et al., 2012; Bahemmat et al., 2016; Ji et al., 2016; Ramírez et al., 2015a; Chirakkara et al., 2015; Mohamed Johar and Embong, 2015; Kim et al., 2015; Shin et al., 2016).

Basically, EK is based on the application of an electric field to the contaminated soil, either by conducting a constant voltage or a direct current (Virkutyte et al., 2002; Amrate et al., 2005; Akretche, 2002; Saeedi et al., 2009; Zanjani et al., 2012; Hassan et al., 2015;

Cameselle, 2015; Ng et al., 2014; Zhou et al., 2014; Zou et al., 2016).

The contaminants migrate through three main phenomena, namely Electro-osmotic (EO), Electro-migration (EM), and Electrophoresis (EP). In an electrolytic cell, EO is the movement of soil moisture from the anode to the cathode. The transportation of ion complexes towards the opposite charge is called EM that usually occurs when ionic inorganic species, such as heavy metal cations, exist. Moreover, charged particles or colloids transport via a phenomenon named EP. Figure 1 depicts mentioned contaminant migration (Virkutyte et al., 2002; Saeedi et al., 2009; Fan et al., 2014; Zhou et al., 2014; Ren et al., 2014).

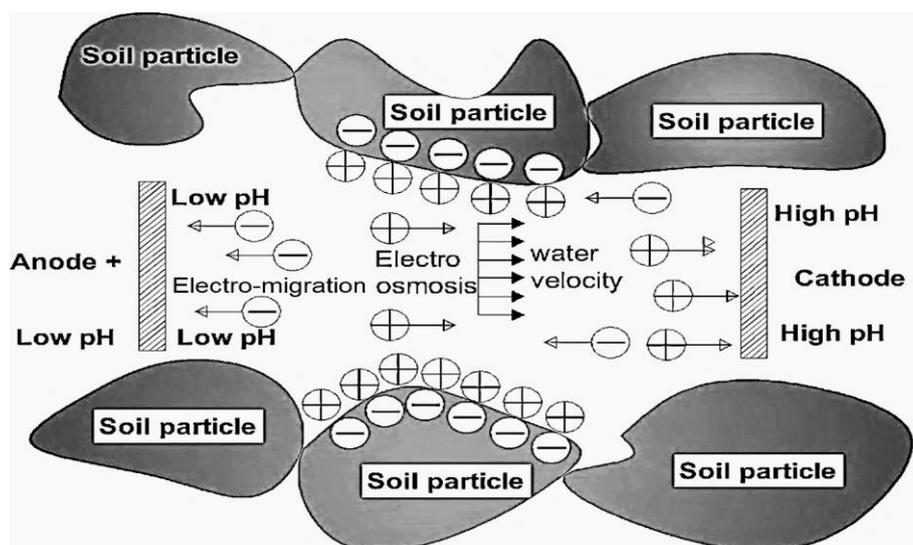
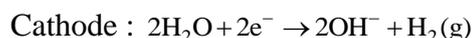


Fig. 1. Electro-osmosis and electro-migration of ions (Moghadam et al., 2016; Acar et al., 1994)

The surface charge of water-saturated clays is typically negative, causing the direction of EO flow from anode to cathode (Virkutyte et al., 2002; Vane and Zang, 1997; Lynch et al., 2007; Alshawabkeh and Acar, 1992; Suzuki et al., 2014). Water electrolysis is considered the dominant electron transfer that affects the EK process significantly (Virkutyte et al., 2002; Amrate and Akretche, 2005; Saeedi et al., 2009; Zanjani et al., 2012; Ramírez et al., 2015b).



The generated H^+ and OH^- ions are transported through the soil sample in opposite directions by EM, advection and diffusion. Due to the counteracting EO flow and greater ionic mobility of H^+ , compared to OH^- , the acid front dominates the chemistry of the soil, except for small sections close to the cathode (Alshawabkeh and Acar, 1992; Acar et al., 1990; Probststein

and Hicks, 1993; Saeedi et al., 2009; Zanjani et al., 2012).

Zeta potential (ζ) is the potential difference between the shearing surface and the bulk fluid (Page and Page, 2002; Moghadam et al., 2016). As the zeta potential (ζ) of soil particle surfaces is lowered, the positive EO volume flow rate increases and advective transport of the organic contaminants happen towards the cathode (Yeung and Gu, 2011). It should be noted that, the more negative the zeta potential (ζ) of the soil surface, the greater the EO flow rate (Kim et al., 2009; Moghadam et al., 2016).

According to Vane and Zang (1997), zeta potential depends on ionic species and strength, pH, type of clay minerals, and temperature. It should be noted that zeta potential is usually negative for water-saturated clays and silts (Virkyute et al., 2002); however, water electrolysis in EK propagates acid and base front from the anode and cathode, probably affecting the soil zeta potential to a large extent (Yeung et al., 1997). Polarity of zeta potential is a crucial issue in soil remediation by means of EK. Once it becomes positive, reverse EO happens that has serious effects on the efficiency of contaminants' removal (Yeung et al., 1997; Virkyute et al., 2002; Saeedi et al., 2009; Zanjani et al., 2012). For example, during metal remediation from clays, soil pH should be maintained low enough to ensure that most of the contaminants exist in the dissolved phase. On the other hand, pH should be high enough to maintain negative zeta potential (Yeung et al., 1997; Virkyute et al., 2002). The simultaneous need for remaining dissolved metal contaminants and maintaining a negative zeta potential is considered the main obstacle in soil remediation, by means of EK process (Virkyute et al., 2002; Yeung et al., 1997; Saeedi et al., 2009; Zanjani et al., 2012).

In general, EK is considered a green remediation technology, used for remediation of soils contaminated by various

types of contaminants such as heavy metals and organic pollutants (Huang et al., 2012; Saichek et al., 2005). The capability of EK to remediate low permeability soil, in situ remediation, and short remediation time are believed to be the main advantages of EK process; however, poor solubility, weak desorption capacity, and low removal efficiency of non-polar contaminants comprise its main disadvantages (Huang et al., 2012). Thus, some enhancement techniques or combined technologies should be applied to overcome these limitations and increase EK remediation efficiency.

The enhancement techniques are applied to EK: in order to:

1. Solubilize or mobilize the contaminants;
2. Control or adjust soil pH in the suitable range for EK;
3. Transform, breakdown, or destroy the contaminants (Yeung et al., 2011).

Thus, to overcome EK limitations and its disadvantages some enhancement techniques such as surfactant, chelants, complexing agents, soil pH control, bioremediation, permeable reactive barriers, and ultrasonication have been provided for combination with EK so that soil remediation efficiency is increased (Yeung et al., 2011; Gomes et al., 2012; Virkyute et al., 2002; Saeedi et al., 2009; Zanjani et al., 2012; Moghadam et al., 2016).

ENHANCED EK

Surfactants and Cosolvents

Surfactants can act as adhesives, wetting, foaming, or flocculating the agents, detergents, penetrants, dispersants, and de-emulsifiers to decrease the surface tension of a liquid (Mulligan et al., 2001; Yeung et al., 2011). Therefore, the main functions of surfactants are surface tension reduction, critical micelle concentration, solubility enhancement, foaming capacity, and wetting ability (Mulligan et al., 2001; Yeung et al., 2011). Three main types of surfactants include:

1. Cationic surfactants, such as CTAC;
 2. Anionic surfactants, such as sodium dodecyl sulfonate (SDS);
 3. Non-ionic surfactants, such as Tween 80 (Huang et al., 2012; Yeung et al., 2011).
- Moreover, biological surfactants (biosurfactants) such as β -cyclodextrin are categorized as natural surfactants.

It should be noted that water hardness, sorption of surfactants onto solid phase, inactivation of surfactants, and difficulties in recovering the surfactants are considered to be main limitations of using surfactants in order to enhance EK remediation (Harwell et al., 1992; Yeung et al., 2011). Thus, to overcome these limitations, such factors as effectiveness of surfactants in EK process,

toxicity and biodegradability of the surfactant and its byproducts, ability to be recovered and recycled, and functionality of the surfactant at different pHs should be taken into consideration (Yeung et al., 2011; Park et al., 2007; Yuan and Weng, 2004).

Usually, due to high toxicity of the cationic surfactants and opposite migration direction of the anionic ones, compared to EO in EK process, they are selected as surfactant to enhance EK remediation less frequently. However, nonionic or biological surfactants are often selected due to their less toxicity and environmental-friendliness (Yeung et al., 2011; Huang et al., 2012). Figure 2 demonstrates the schematic diagram of EK, enhanced by surfactant.

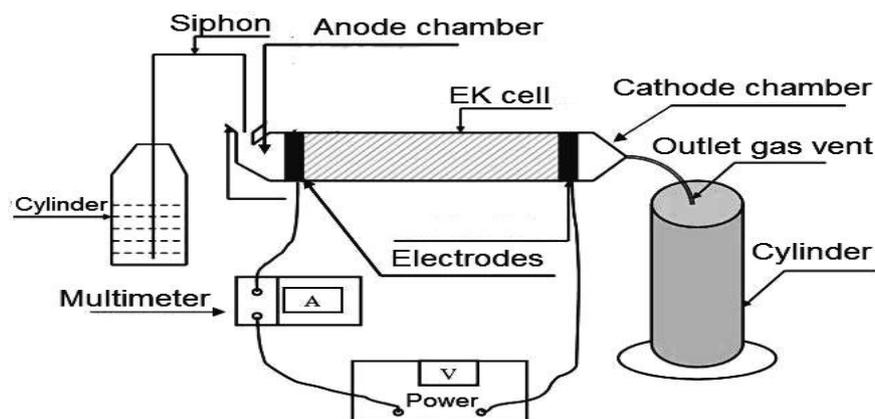


Fig. 2. EK remediation enhanced by surfactant (Yuan et al., 2006; Huang et al., 2012)

Cosolvent such as methanol, ethanol, propanol, acetone, tetrahydrofuran, and butylamine are used to enhance aqueous solubility of organic pollutants including alkanes, halogenated hydrocarbons, benzenes, phenols, polychlorinated biphenyl, and Polycyclic Aromatic Hydrocarbons (PAH), better transporting them with EO flows (Yeung et al., 2011; Huang et al., 2012; Li et al., 2000; Maturi and Reddy, 2008; Saichek and Reddy, 2003; Wan et al., 2009; Yuan et al., 2007).

Chelants

The formation of more than two separate bonds between a bi-dental or multi-dental ligand, and an individual ion or metal atom

is known as chelation (Yeung et al., 2011). Therefore, chelation can desorb toxic metals from soil surface, using formation of strong water-soluble complexes (Yeung et al., 2011). Chelating agents such as ethylenediaminetetraacetic acid (EDTA), (diethylenetriamine) pentaacetic acid (DTPA), and citric acid have been frequently used in EK processes (Virikutyt et al., 2002; Yeung et al., 2011; Darban et al., 2009; Kiyae et al., 2013).

The main functions of chelants are solubilizing of sorbed contaminants from soil surface and decreasing the zeta potential of soil surfaces (Gu et al., 2009; Yeung et al., 2011). As aforementioned, decrease in the zeta potential causes EO flow rate to

increase, facilitating the advective transportation of the contaminants (Yeung et al., 2011; Virkutyt et al., 2002).

Remediation of contaminated gold mining sediment by mercury was studied by Darban et al. (2009), using EK enhanced by EDTA as chelate. The removal efficiency from using this technique turned out to be up to 70% (Darban et al., 2009).

Complexing agents

Complexing agents are chemicals which form only a single bond between the ion or metal atom (coordination complexes) that differs from complexing agents (Yeung et al., 2011). Complexing agents such as Γ^- , Cl^- , NH_3^- , and OH^- are used in the EK process to form soluble metal complexes (Yeung et al., 2011). For example, many researchers believe that mercury could be efficiently extracted by iodide-enhanced EK remediation as soluble complex HgI_4^{2-} (Shen et al., 2009; Reddy et al., 2003; Cox et al., 1996). Acetic acid (CH_3COOH) is another complexing agent, used to enhance EK remediation of Cu-contaminated soil (Zhou et al., 2006). Moreover, Cyclodextrins, which is nontoxic and biodegradable, is frequently used to enhance the EK process to remediate contaminated soil (Maturi and Reddy, 2006; Wan et al., 2011; Li et al., 2010).

Soil pH control

Water Electrolysis generates H^+ and O_2 at the anode and OH^- and H_2 gas at the cathode. The migration of H^+ and OH^- lead to acidic and basic condition in the soil, near the anode and cathode. The adverse impacts of the development of acid/base front may be controlled through two main methods, namely electrode conditioning and ion exchange membrane (Yeung et al., 2011; Virkutyt et al., 2002).

In electrode conditioning, weak acids such as CH_3COOH (Genc et al., 2009) and citric acid (Gidarakos and Giannis, 2006) may be used to neutralize the generated hydroxyl ions during EK remediation. It should be mentioned that improper use of

some acids such as HCl may lead to the presence of toxic byproducts, posing a health hazard (Yeung et al., 2011).

Ion exchange membrane is the other enhancement technique of reservoir conditioning (Yeung et al., 2011). Cation and anion exchange membrane allow only cations and anions to pass through. Thus, installation of cation exchange membrane between the cathode and specimen can hinder the migration of the generated OH^- ions as well as metal hydroxides precipitation (Yeung et al., 2011). In this technique the precipitation of metal contaminants occur on the surface of the membrane or into the catholyte, leading to the membrane fouling and the increase in flow resistance (Yeung et al., 2011; Shen et al., 2007; Lee et al., 2002).

EK combined with bioremediation (Bio-EK)

Bioremediation uses microorganisms to degrade hazardous contaminants (especially organic pollutants) under a suitable condition (Yeung et al., 2011). However, in cases such as low hydraulic conductivity soils, it is difficult to supply microorganism and the required electron acceptors or nutrients to the hazardous contaminants (Yeung et al., 2011). Thus, EK combined with bioremediation is designed to activate or supply microorganisms and electron acceptors, providing nutrients for the contaminants through EO flow to promote the reproduction, growth, and metabolism of the microorganisms (van Cauwenberghe, 1997; Virkutyt et al., 2002; Yeung et al., 2011).

Some examples of combination of EK and bioremediation to remediate contaminated soils include the injection of EK-using *Pseudomonas* into diesel-contaminated soil (Lee and Lee, 2001), *Mycobacterium frederiksbergense* into the contaminated clayey soil to PAH (Wick et al., 2004), and *Acidithiobacillus thiooxidans* (sulfur-oxidizing bacteria) into contaminated soil, by means of toxic metals including Cd, As, Zn, Pb, and Cu (Lee et al., 2011).

EK combined with Permeable Reactive Barriers (PRB)

A Permeable Reactive Barrier (PRB) is an innovative technology, made of reactive material placed across the flow path of a dissolved phase contaminant plume, to degrade or remove contaminants (Saeedi et al., 2009; Palmer, 2001; Yeung et al., 2011). When EK is combined with PRB, the EO

flow plays an important role to remove or degrade dissolved contaminants (Fig. 3). It should be noted that the sorption of contaminants on surface of PRBs material or degradation of contaminants largely depend on the pH. Thus, the pH gradient during EK process may affect removal efficiency of contaminants, using this combination technique (Yeung et al., 2011).

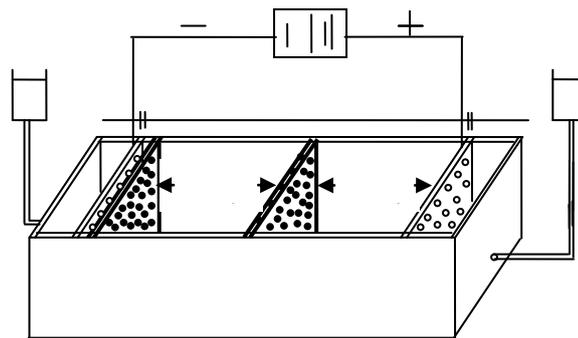


Fig. 3. Electrokinetic coupled with PRB (Saeedi et al., 2009)

The zero-valent iron (ZVI) is frequently used as a reactive material in PRB, combined with EK. The feasibility of EK remediation coupled with ZVI PRB to remediate nitrate-contaminated soil was investigated by Chew and Zhang (1998). TCE Degradation by means of EK, coupled with ZVI PRB was investigated by Moon et al. (2005). They found that the rate of reductive dechlorination of TCE improved up to six times more than ZVI PRB alone. The removal mechanism of Cr^{+6} , using EK combined with ZVI PRB, was studied by Cang et al. (2009), who obtained a removal efficiency up to 72% when they placed ZVI PRB between the electrodes and contaminated soils. In addition to ZVI and atomizing slag (Chung and Lee, 2007), carbon active (Saeedi et al., 2009) was used as reactive material, combined with EK, to remediate contaminated soils.

EK combined with ultrasonication

Ultrasonication is based on the cavitation (bubble formation) to increase kinetic energy, temperature, volume, and pressure

of soil pore water (Gomes et al., 2012; Yeung et al., 2011; Chung and Kamon, 2005). It can destroy organic pollutants such as PCBs and PAH by increasing molecular motion of the contaminants, inducing mobilization, and disintegrating sorbed contaminants on soil surfaces (Gomes et al., 2012; Yeung et al., 2011).

Remediation of contaminated clayey soil by Pb and phenanthrene, using EK combined with ultrasonication, was studied by Chung and Kamon (2005). They found that the combination of EK and ultrasonication improved remediation efficiency, compared to EK only. Remediation of spiked kaolin by HCB, phenanthrene, and fluoranthene was investigated by Pham et al. (2009) who used EK coupled with ultrasonication. Their results revealed higher EO flow, current and efficiency, in comparison with EK only.

CONCLUSION

EK remediation is an innovative and promising remediation technology especially in fine-grained contaminated soils. However, due to some of its limitations and

disadvantages, several enhancement techniques are provided to overcome the limitations and increase EK remediation efficiency. Thus, this paper has reviewed some of the main enhancement techniques to improve EK remediation effectiveness. Overall, it can be concluded that surfactants, chelates, and complexing agents can enhance the solubility of contaminants using different functions to improve removal efficiency in EK process. Moreover, soil pH and zeta potential can control the EO direction flow and removal efficiency. The combination of EK with bioremediation can specifically enhance degradation of organic pollutants in fine-grained soils. For EK, coupled with PRB, it could be a very effective technique, when contaminants are soluble and transported through the EO flow. Moreover, the degradation of ionic and nonionic contaminants, using EK combined with ultrasonication has promising efficiency and research prospects. In general, selecting the best combination technique to increase EK efficiency depends on the contaminant types (OP, Heavy metals, etc.) and soil type. For example, combination of EK and bioremediation can be very effective for the soil, contaminated with PAH. EK combined with surfactant, chelant, and complexing agents can be very effective to remediate soil contaminated with heavy metals. Moreover, EK combined with ultrasonication is considered a suitable remediation technique for both ionic and nonionic contaminants. Moreover, it should be noted that controlling the soil pH in EK, combined with other techniques, may increase removal efficiency. In some cases, soil pretreatment by means of weak acids may increase the EK efficiency. Moreover, EK, combined with other techniques including fenton and phytoremediation, may be effective tools to improve EK efficiency.

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