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REVIEW PAPER

A review of combinations of electrokinetic applications

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Abstract Anthropogenic activities contaminate many lands and underground waters with dangerous materials. Although polluted soils occupy small parts of the land, the risk they pose to plants, animals, humans, and groundwater is too high. Remediation technologies have been used for many years in order to mitigate pollution or remove pollutants from soils. However, there are some deficiencies in the remediation in complex site conditions such as low permeability and complex composition of some clays or heterogeneous subsurface conditions. Electrokinetic is an effective method in which electrodes are embedded in polluted soil, usually vertically but in some cases horizontally, and a low direct current voltage gradient is applied between the electrodes. The electric gradient initiates movement of contaminants by electromigration (charged chemical movement), electroosmosis (movement of fluid), electrolysis (chemical reactions due to the electric field), and diffusion. However, sites that are contaminated with heavy metals or mixed contaminants (e.g. a combination of organic compounds with heavy metals and/or radionuclides) are difficult to remediate. There is no technology that can achieve the best results, but combining electrokinetic with other remediation methods, such as bioremediation and geosynthetics, promises to be the most effective method so far. This review focuses on the factors that affect electrokinetic remediation and the state-of-the-art methods that can be combined with electrokinetic.

Keywords Electrokinetic · Soil pollution · Remediation · Contaminant · Electrically conductive geosynthetic

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Introduction

There are many lands that are contaminated by anthropogenic activities. In some cases, harmful substances such as heavy metals or dangerous organic compounds exist in the soil matrix and underground waters. About 63 % of the land on the national priority list (NPL) of the USA (from a total of 1200 sites) is contaminated by toxic and risky heavy metals. Among the toxic heavy metals, lead, chromium, and cadmium are most commonly found at NPL sites, respectively (Consultant 1996). Although polluted soils occupy only a small part of the lands, the risk to plants, animals, humans, and groundwater is too high.

The situation is worse when there is a polluted site with low permeability and/or complex composition of some clays with heterogeneous subsurface conditions. However, researches aiming to remediate, mitigate, or stop the propagation of harmful materials have been carried out over the past 30 years. Heavy metals or metalloids including lead (Pb), mercury (Hg), arsenic (As), copper (Cu), zinc (Zn), chromium (Cr), cadmium (Cd), strontium (Sr), iron (Fe), manganese (Mn), tin (Sn), nickel (Ni), caesium (Cs), and uranium (U) are considered as most pollutants that can contaminate soil and groundwater because of their mobility and solubility. Figure 1 provides an overview of the contaminants affecting the groundwater and soil in European countries as reported in 2011 (Van Liedekerke et al. 2014).

Because of some deficiencies in conventional treatment methods, new remediation techniques are needed to remove hazardous materials from fine content soils efficiently. Although soil washing and stabilization or solidification have been used to eliminate risky heavy metals from silt or sandy soil effectively, these methods are not efficient for fine-grained soils (Ko et al. 2005).

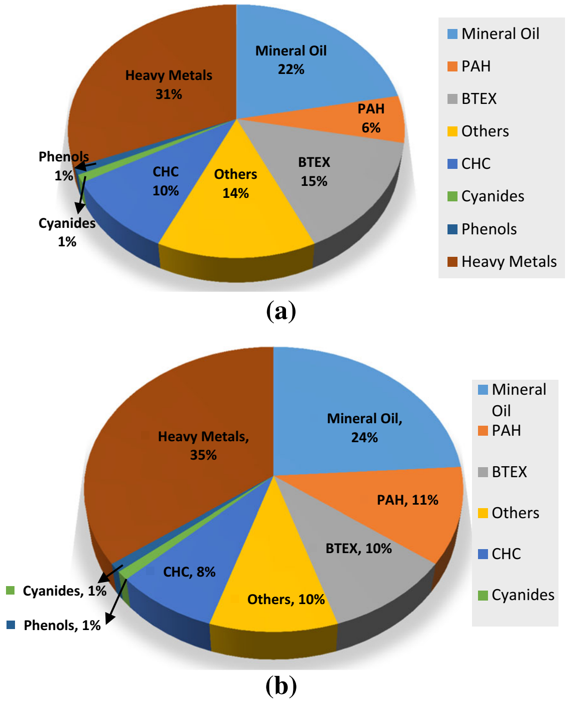

Fig. 1 Overview of contaminants affecting **a** groundwater and **b** soil in European countries (Van Liedekerke et al. 2014)

Selection of the best method for remediation depends on many factors, such as soil and sediment characteristics, amount of pollutants (concentrations), future use of contaminated lands, purpose of remediation, the allowable amount of contaminants in the medium, type of pollutant, available methods, economic conditions, and time to remediate. Electrokinetic remediation is an innovative method in which electrodes are embedded in a polluted soil, usually vertically but in some cases horizontally, and a low direct current (DC) voltage gradient is applied between them. An electric gradient initiates the movement of contaminants by electromigration (charged chemical movement), electro-osmosis (movement of fluid), electrolysis (chemical reactions due to an electric field) (Mulligan et al. 2001), and diffusion (movement of the ionic species in the soil solution caused by concentration gradients formed by the electrically induced mass transport). It must be noted that as the ionic mobility of a species is much higher than its diffusion coefficient, diffusion is often ignored when studying electrokinetic (Acar and Alshawabkeh 1993). Figure 2 shows a conceptual representation of the mentioned movements.

Reddy (2013) pointed out some of the advantages of electrokinetic remediation in comparison with conventional remediation methods: first, the simplicity of the method; second, safety, because in electrokinetic the operator and people in nearby areas are not exposed to contaminants; third, the fact that this method can be used in many contaminated environments and conditions; in other words, electrokinetic can be used for sediments, soils, groundwater, and sludges (which is particularly appropriate for low-permeability soils like clays and heterogeneous soil deposits within the vadose zone, where other treatment methods are not effective or are expensive); fourth, a wide range of contaminants such as metals and metalloids, organic compounds, and radionuclides or a combination of these contaminants can be remediated; fifth, the flexibility of electrokinetic, as it can be used as an in situ or ex situ treatment system and can be easily combined with traditional remediation technologies such as bioremediation; and finally, the cost-effectiveness of this method, which requires almost low electrical energy (compared to other thermal technologies), leading to a lower overall cost that ranges from \$20 to \$225 per cubic yard depending on the type of soil and other site-specific conditions.

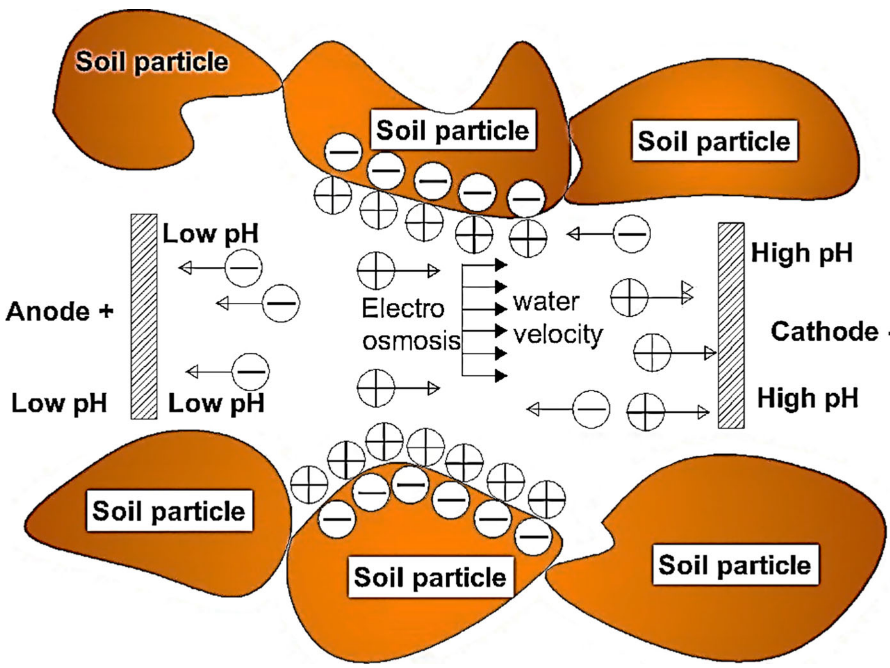

Fig. 2 Conceptual of movements in electrokinetic

Although the method has limited practical applications due to disadvantages such as low removal efficiency for non-polar organic pollutants (OPs), weak desorption capacity, and poor solubility, the disadvantages of a single electrokinetic technique, such as the long remediation time and lower removal efficiency of pollutants, could be enhanced by combining electrokinetic techniques (Huang et al. 2012).

Many studies have been conducted to improve the electrokinetic removal efficiency, using, for example, surfactants, enhancement solutions, pH control, desorbing agents, and pulse and alternating currents, but most of these researches were done at bench scale and may not be applicable to full-scale soil remediation. Table 1 shows a timeline of full-scale applications and the main pilot studies of electrokinetics.

On the other hand, a combination of treatment techniques, when acting in a synergistic manner, will minimize the cost of achieving risk-based endpoints (Rao et al. 2002). A combination of techniques or treatment trains is carried out in succession or concurrently to improve remediation in a quicker and more efficient and cost-effective way (Gómez et al. 2009).

Recent developments in electrokinetic consist of a combination of phytoremediation, electrokinetic-enhanced bio-augmentation for remediation of clays contaminated by chlorinated solvents (Mao et al. 2012), coupling electrokinetic and nanoparticles (Gomes 2014), hydraulic flushing and electrokinetic for removal of PAH and heavy metal simultaneously (Reddy et al. 2010), hydraulic pressure injection of electrolyte to enhance the efficiency of the remediation of pentachlorophenol (PCP)-contaminated soil (Huang et al. 2013), remediation of hexachlorobenzene (HCB)-contaminated soil by electrochemical Fenton oxidation (Oonnittan et al. 2009a, b), and coupling of cosolvents or surfactants with oxidants for enhancement of dense non-aqueous phase liquid (DNAPL) removal (Dugan et al. 2010).

Effects of soil pH on remediation process

The control of soil pH using different methods is a common alternative to improve the removal efficiency of contaminants in the electrokinetic process (Baek et al. 2009; Kim et al. 2009a; Zhou et al. 2004), but soil pH variations affect the zeta potential (ξ) of the soil surface and consequently the electro-osmotic flow changes, because it is highly related to the surface charge of the soil or zeta potential (Baek et al. 2009; Kim et al. 2009a). Zeta potential is the potential difference between the shearing surface (the plane at

Table 1 Timeline of full-scale applications and the main pilot studies of electrokinetics

Application	Year
Remove excess salts from alkali soil in India	1936
Reverse the seepage flow direction and stabilize a long railroad cut (Salsgitter, Germany)	1939
Desalination of concrete, Federal Highway Administration, USA	1976
First electro-reclamation pilot project, former paint factory in Groningen, the Netherlands	1987
Electro-bioreclamation pilot project (former industrial site with diesel fuel and aromatic) at Vorden, the Netherlands	1993
Injection of chemical conditioners, electrokinetic INS, US Army Waterways Experiment Station, Vicksburg, Mississippi	1994
In situ remediation of uranium-contaminated soil, Oak Ridge K25 Facility, Oak Ridge, Tennessee, USA	1995
Pilot project Lasagna, Paducah site (contaminated with TCE), Kentucky, USA	1995
Electrokinetic demonstration at the unlined chromic acid pit, Sandia National Laboratory, USA	1997
Field-scale demonstration of chromium and copper remediation, Point Mugu, California, USA	2004
Pilot-scale electrochemical cleanup of lead-contaminated soils in a firing range, USA	2005
Pilot-scale application in a rice field near a zinc refinery plant located at Jangghang, South Korea	2011

which the diffuse double layer at the surface of the soil particles can slip past the charged soil surface) and the bulk liquid (Page and Page 2002). In other words, the more negative the zeta potential of the soil surface, the more electro-osmotic flow takes place (Kim et al. 2009b). However, studies have shown that electrochemical processes are very complicated and may change according to the site geochemistry. Induced electric potential leads to electrolysis of water content and usually produces H^+ ions and O_2 gas at the anode and OH^- ions and H_2 gas at the cathode. H^+ ions usually move towards the cathode, OH^- ions move towards the anode, and in some cases gases vaporize into the atmosphere. Consequently, depending on the extent of migration of H^+ and OH^- ions, pH change occurs within the soil (Reddy 2013). For example, a lower soil pH near the anode causes desorption and solubility of cationic (negatively charged) metals, such as nickel, lead, and cadmium, enhancing their electromigration towards the cathode. However, the higher pH around the cathode is the reason why these metals adsorb or precipitate, slowing down electromigration and removal at the cathode (Reddy 2013). If the direction of electro-osmotic flow is in the direction of the cathode, then elimination of cationic metal might be improved, but the removal may be decreased when it is in the opposite direction (Kim et al. 2009b). In many cases, buffer solutions have been used to maintain the pH at the electrodes (Mulligan et al. 2001). The ions of metals and metalloids can be eliminated by precipitation or co-precipitation and electroplating at the electrodes. Other methods include recovering the metals by pumping the waste to the surface or ion exchange resins (Smith and Brauning 1995). In most cases, there are high-pH (basic) conditions near the cathode and low-pH (acidic) conditions near the anode (Reddy 2013).

If a pH control solution is not used, because of soil water electrolysis during the process, the soil pH usually decreases to 2–3 in the soil section near the anode and, if uncontrolled, increases to 8–12 in the soil section near the cathode in a low buffering soil (Zhou et al. 2005). The latter causes metal hydroxide precipitation in the soil close to the cathode, and consequently metal removal efficiency is greatly reduced. For this reason, enhancement methods such as conditioning of the catholyte pH (Bonilla et al. 2000; Lee and Yang 2000), adding enhancing chemical reagents to improve metal solubility (Sah and Chen 1998; Yang and Lin 1998; Reddy and Chinthamreddy 2003; Zhou et al. 2004), using ion-selective membrane to exclude OH-migration from the cathode chamber into the soil (Li et al. 1998), and applying sulphur bacteria in the soil column (Maini et al. 2000) have been explored and examined. Kimet al. (2009b) pointed out that removal of zinc and nickel from polluted soil increased with decreasing pH of the extraction solution and that nitric acid removed these materials from the soil very effectively. Also, pretreatment of the soil with acidic solution improved desorption of zinc and nickel, and catholyte conditioning with this solution was very efficient in maintaining the overall soil pH across the electrokinetic cell. They mentioned that the catholyte conditioning and pretreatment method improved the removal of zinc and nickel by up to 41 and 40 % after 4 weeks of operation, respectively. However, the mentioned co-electrokinetic methods are used only for a specific pollutant and condition.

Combination of bioremediation and electrokinetic

Electrokinetic efficiency is an important factor that has been considered by many researchers. Also, more complex sites with various pollutants need innovative and combined remediation techniques. A new emerging in situ hybrid technology has been proposed to increase the mobility and the possibilities of interaction among micro-organisms, pollutants, and nutrients in the soil. This technique is called electrokinetic-enhanced bioremediation or electro-bioremediation (Wick et al. 2007) and uses synergistic effects of bioremediation and electrokinetic in the remediation of organic contaminants. In fact, bioremediation is an efficient, low-cost technology based upon the degradation of pollutants by micro-organisms (Mena et al. 2015). Organic compounds and pollutants can be consumed by micro-organisms to increase their reproduction rate and growth (Kim et al. 2005; Niqui-Arroyo and Ortega-Calvo 2007).

Although it is slower than other physicochemical techniques and is always subject to the ability of the micro-organisms to use the pollutants as a substrate (Ramírez et al. 2014), the biological technique can not only degrade contaminants into less toxic products and oxidize them into carbon dioxide and ultimately water, but also change the mobility of the pollutants and make them settle in a certain place (Huang et al. 2012). The main problem in carrying out remediation of clays using this combination method is the need to maintain optimal conditions for microbial degradation. In other words, factors like sources of energy and carbon, electron acceptors, the presence of appropriate micro-organisms, nutrients, concentration of pollutants, combination of organic pollutants, metal ions, and appropriate environmental conditions such as pH, moisture, and temperature all affect the efficiency of micro-organisms (Ramírez et al. 2014; Schmidt et al. 2007; Xu et al. 2010; Lahlou et al. 2000; Cunningham et al. 2001). The main advantages of electrokinetic-enhanced bioremediation are that it increases the biological pollutant remediation rate through the electrokinetic transport phenomena (Mena et al. 2012; Lear et al. 2007). Transportation of micro-organisms to increase the rate of the biological degradation process is called electrophoresis (Mena et al. 2011). In cold climate areas, the heating produced by high ohmic drops when an electric field is applied to a soil increases the rate of bioremediation processes (Suni et al. 2007). In another novel use, the coupling of electrokinetic soil flushing (EKSF) technology with a biological degradation system through the use of bio-PRBs (permeable reactive barriers) or bio-barriers is suggested for treatment of diesel-polluted clay soil (kaolinite) (Mena et al. 2015).

EKSF consists of the use of a flushing fluid to extract pollutants from soil, efficiently combining the different electrokinetic mass transport processes (electro-osmosis, electromigration, and electrophoresis) and also taking advantage of other processes, such as water electrolysis and ohmic heating, which develop when an electric field is applied to a soil (López-Vizcaíno et al. 2011a, b; Alcántara et al. 2010).

The enhanced mass transport that is attained by this method is very effective for remediation of pollution during bioremediation, and coupling of these methods is more effective than the use of either single treatment alone (Dong et al. 2013; Li et al. 2010; Wick et al. 2007). The main benefit of this coupling is that pollutants are degraded *in situ* by the micro-organisms and a final treatment of the flushing solution is not needed. However, because of some differences between the conditions required for this coupling (severe conditions with high pH and temperature gradients for EKSF and mild conditions with good distribution of nutrients for the bioremediation method), careful assessment is needed; otherwise the expected result will not be obtained (Mena et al. 2015). Also, special attention should be paid to the application of large electric fields, which could result in an antagonistic combination if insufficient attention is paid to the operation conditions (Mena et al. 2011).

Mena et al. (2015) pointed out that by combining EKSF with bio-PRB technology, during short periods (2 weeks), a diesel removal rate of 30 % and energy consumption below 15 % are achieved for kaolinite. Nutrients and SDS (sodium dodecyl sulphate) are efficiently transported in combined bio-PRB/EKSF technology by electromigration and by electro-osmotic processes. Diesel is also transported, although the extent of the transport is not high enough to attain a significant removal by these processes. The pH and lack of nutrients are the two key factors needed to improve this technology, in the first case because extreme pH values cause the death of micro-organisms, and in the second case because lack of nutrients limits the growth of micro-organisms and hence the remediation process. Bio-transformations under aerobic conditions are more energetically favourable than the use of alternative electron acceptors, such as nitrate or sulphate (Spence et al. 2005). However, there are few studies about the influence of electrokinetic treatment on the dissolved oxygen (DO) concentrations in the groundwater of polluted soils (Ramírez et al. 2014).

Due to the low diffusion rate of oxygen, it is a challenge to develop an appropriate alternative to supply a high enough DO concentration to meet the demand for in situ soil aerobic remediation processes (Ramírez et al. 2014). Different alternatives have been used to increase the concentration of DO in the media, such as air sparging or biosparging, liquid delivery systems, and bioventing (Balcke et al. 2004; Vogt et al. 2004).

Additionally, several products, such as oxygen micro-bubbles and oxygen-releasing compounds (ORCs), have been extended to oxygenate soil and groundwater (Kunukcu 2007; Jechalke et al. 2010; Zawierucha and Malina 2011; Chun et al. 2013).

Mena et al. (Ramírez et al. 2014) have suggested that the oxygen demand for aerobic in situ soil bioremediation could also be supplied by transport of the oxygen generated by the water oxidation reaction at the surface of the anode in an electro-bioremediation process.

They concluded that, with regard to the effect of the voltage, it was also observed that applying high electric current did not increase the values of the DO concentrations in the sampling points distributed across the soil section. It is likely that, due to the low permeability of the clay soils, the oxygen generated at the anode was not transported through the soil. Therefore, in aerobic biological treatment of low-permeability soils, the oxygen generated at the anode electrode surface by the water oxidation reaction would not spread adequately to meet the necessary oxygen requirements.

Some organic pollutants such as polycyclic aromatic hydrocarbons (PAHs) (Wick et al. 2004), alkanes (Kim et al. 2005), halogenated hydrocarbons (Ho et al. 1999aa, b; Jackman et al. 2001; DeFlaun and Condee 1997), and phenols (Luo et al. 2005; Yee et al. 1998; Ho et al. 1995) have been removed successfully by a combination of electrokinetic with bioremediation.

Combination of geosynthetics and electrokinetic

Geosynthetics have been widely used in environmental industries and civil engineering for a long time and are well established as providing reinforcement, separation, filtration, and drainage and also acting as impermeable members, barriers, and passive materials in these applications (Hamir et al. 2001; Jones et al. 2011).

However, a new application in which they are coupled with electrokinetic can be recognized, where the geosynthetic plays an active role, initiating physical or chemical changes to the soil in which it is installed, in addition to providing the expected functions (Glendinning et al. 2005).

The idea of Electro Kinetic Geosynthetic (EKG) (or electrically conductive geosynthetic) materials was suggested for the first time by Jones et al. (1996). In fact, EKGs, besides providing reinforcement, drainage, and filtration of soils, can also be improved by electrokinetic techniques for transportation of chemical species and water across fine-grained low-permeability soils like clays. Table 2 shows the functions of electrically conductive geosynthetics, which are used in practical applications (Jones et al. 2011):

EKG has been used as an anode electrode for the reinforcement of soil, with needle-punched EKG as the cathode. Some pullout tests showed an improvement in the reinforcement bond of up to 211 % and enhancement in shear strength of up to 200 % in comparison with the values obtained when the geosynthetics were not electrically conductive (Hamir et al. 2001).

Usually there are three fundamental applications for electrically conductive geosynthetics or active geosynthetics (Glendinning et al. 2005, 2008; Jones et al. 2011):

1. Electrophoretic action, which increases the speed of solid settlement from liquids.
2. Electro-osmotic action, which involves dewatering and a decrease in volume.

Table 2 Functions of electrically conductive geosynthetics in practical applications (Jones et al. 2011)

	Function	Effects
Electrokinetic	Electro-osmosis	Water flow
		Pore pressure change
		Volume change
	Electrophoresis	Particle movement
		Particle orientation
	Ion migration	Solute movement
	Electrolysis of water	Oxygen evolution
	Heating	Joule heating (electrode)
		Resistive heating (soil)
	Oxidation reactions	Soil cementation
		Reduction in soil plasticity
	Reducing reactions	Electrowinning of metal ions
		Evolution of ammonia
Geosynthetics	Drainage	Water flow
		Gas flow
	Reinforcement	Tensile strength
In-plane stiffness	Filtration	Barrier to solids entrained in flow
	Separation	Strengthening and prevent mixing
	Containment	Physical containment of solids
	Membrane action	Barrier to flow (containment of fluids)
	Sorption	Capture of liquids or dissolved species

3. Improvement of strength by consolidation with electro-osmotic action.

The main purpose of electrokinetic is the remediation of polluted soils, and conductive geosynthetics can be used to effect the movement of pollutants across soil to the electrodes and afterwards to adsorb them. Since hydraulic permeability is a function of the grain size, electro-osmotic permeability is effectively independent of grain size. In other words, electro-osmosis can result in flow rates 100–10,000 times greater than hydraulic flow in fine-grained materials (Jones et al. 2008).

State-of-the-art uses of EKGs include the following:

1. Decreasing the cost of disposal through the use of EKG for soil consolidation or volume reduction in industrial wastes (Alshawabkeh et al. 2004).
2. Increasing shear strength by the use of conductive reinforcement, which enables the use of cohesive fines and very wet material as fill for reinforced structures (Glendinning et al. 2005).
3. Preventing liquefaction of susceptible soils (like saturated loose sands) with electrically conductive band drains.
4. Attaining rapid drawdown of the phreatic surface in comparison with currently possible dewatering with conventional well-pointing technology in fine-grained low-permeability soils (McLoughlin 2005; Glendinning et al. 2006).
5. EKG technology could help to enhance mining methods and to improve the soil conditions in the vicinity of the tunnel or to reduce post-construction settlements associated with the tunnel.
6. The stability of slopes could be increased by applying direct current between appropriately positioned electrodes. In other words, negative pore pressure that is generated at the anode will increase the soil strength and cohesion between the EKG electrode (nail) and perimeter soil, and therefore the nails remain in the soil permanently (Jones et al. 2011).
7. Shallow foundations that are constructed on problematic soils with the capability of swelling

and shrinkage can be treated by EKG technology. Therefore, this method controls the moisture of prone strata with adjustment of water as necessary to stop changes in volume.

8. Shear strength improvement of low-permeability soils, especially clays, with about ten times faster improvement and consolidation in comparison with prefabricated vertical drains (PVD) treatment (Chew et al. 2004).

Recommendations for future research

- To improve the shear strength of low-strength soils, the use of electrical pile or sheet pile is recommended.
- Electrokinetic could be used in embankment dams in order to reduce pore pressure and prevent hydraulic fracture.
- In marine usage, for rapid dewatering of bed sludge and fine soils, electrokinetic is a very efficient alternative, and more research is needed.
- Problematic soils which show shrinkage and swelling behaviour could be remediated by electrokinetic technology.
- Electrokinetic sheet pile could be used as a barrier to stop leakage of pollutant in the vicinity of emission industries.
- Investigation of the bearing capacity of shallow foundations on fine content soils or enhanced sludge by electrokinetic could lead to new perspectives for geotechnical engineering.

Conclusion

Although polluted soils occupy a small part of land areas, their risk to plants, animals, humans, and groundwater is too high. Remediation technologies have been used for many years in order to mitigate or remove pollutants from soils. Selection of the best method for remediation depends on many factors such as soil and sediment characteristics, amount of pollutant (concentrations), future use of contaminated land, purpose of remediation, allowable amount of contaminants in the medium, type of pollutant, available methods, economic conditions, and time to remediate. However, remediation in complex site conditions, such as low permeability and complex composition of some clays or heterogeneous subsurface conditions, has some deficiencies. Therefore, there is no technology that can achieve the best results, but mixing electrokinetic with other remediation methods like bioremediation and geosynthetics promises to be the most effective method so far. A new emerging *in situ* hybrid technology has been proposed to increase the mobility and the possibilities of interaction among micro-organisms, pollutants, and nutrients in the soil. This technique is called electrokinetic-enhanced bioremediation or electro-bioremediation and uses synergistic effects of bioremediation and electrokinetic in the remediation of organic contaminants. Some organic pollutants such as polycyclic aromatic hydrocarbons, alkanes, halogenated hydrocarbons, and phenols have been removed by a combination of electrokinetic with bioremediation. Geosynthetics have been widely used for a long time to provide filtration, separation, reinforcement, drainage, and to act as impermeable members, barriers, and passive materials. Electrically conductive geosynthetics or Electro Kinetic Geosynthetics (EKGs), besides providing reinforcement, drainage, and filtration of soils, can be improved by electrokinetic techniques for transporting chemical species and water across fine-grained low-permeability soils like clays. EKG was used as an anode electrode for the reinforcement of soils, with needle-punched EKG as the cathode. Pullout tests showed an improvement in the reinforcement bond of up to 211 % and enhancement in shear strength of up to 200 % in comparison with the values obtained when the geosynthetics were not electrically conductive.

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