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Analysis of the Effective Factors on the Electro Remediation of Polluted Soils, an Action Towards Green Technology and Sustainable Environmental Improvement

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تحلیلی بر شناخت عوامل تأثیرگذار در فرایند الکتروشیمی به منظور پاکسازی عناصر آلاینده از خاک، گامی در راستای فناوری سبز و توسعه پایدار محیط زیستی

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Abstract:
Today, the issue of sustainable development has become one of the most important global issues and has become the focus of attention and human attention in the economic, social, and environmental perspectives. Various human activities, as well as the development of industries and the expansion of urbanization with waste and wastewater, have contaminated soil. Different methods are available to clean up the soil about economic issues. One of them is the electro remediation method, which is often applied in low-permeability fine-grained soils and is successful as a greenhouse effect in reducing the toxicity of heavy metals and is a new and developing approach for organic pollutants. This paper will be presented the effective parameters, the strong points as well as limitations, to clarify the basic concepts in the electro remediation process. Conduct experiments with the three-dimensional arrangement of electrodes instead of the linear arrangement of electrodes could be more practical. Using geosynthetic electrodes to prevent corrosion of metallic electrodes, combining the electro remediation process with other designs and different operational modes, and overcome to the problem of reducing the electro-osmosis flow and reverse electro-osmosis flow with pH control are methods to enhance the rate of removal of pollutants from soils.

Keywords: Remediation of Contaminated Soil; Sustainable Development; Effective Parameters; Enhanced Electro Remediation.

چکیده:
امروزه مسئله توسعه پایدار یکی از موضوعات مهم در سطح جهانی به شمار رفته و به نقطه تمرکز و توجه بشر در دیدگاه‌های اقتصادی، اجتماعی و زیست‌محیطی تبدیل شده است. فعالیت‌های مختلف انسانی و نیز توسعه صنایع و گسترش شهرنشینی با تولید پسماند و پساب، آلودگی خاک را به همراه داشته است. روش‌های متفاوتی برای پاکسازی خاک با توجه به مسائل اقتصادی در دسترس است. یکی از آنها، روش الکتروشیمی می‌باشد که اغلب در خاک‌های ریزدانه با نفوذپذیری پایین کاربرد دارد و به‌عنوان روشی کارآمد در راستای فناوری سبز و کاهش سمیت فلزات سنگین از خاک موفقیت‌آمیز بوده و برای آلاینده‌های آلی نیز یک رویکرد جدید و درحال‌توسعه است. مقاله حاضر از نظر هدف کاربردی است و ضمن ارائه مفاهیم تحلیلی- توصیفی از نوع اسنادی، به مفاهیم بنیادین در عملیات پاکسازی به روش الکتروشیمی و بررسی پارامترهای مؤثر بر میزان راندمان حذف آلاینده از خاک می‌پردازد و به‌عنوان نتیجه‌گیری می‌توان با بهره‌گیری از تحلیل در مقیاس سه بعدی در عملیات الکتروشیمی نتایج را به‌صورت عملی‌تر بررسی کرد و با استفاده از الکترودهای ژئوسنتتیک در جهت جلوگیری از خوردگی الکترودهای فلزی، کنترل pH و نیز با بهره‌گیری طراحی‌ها و حالت‌های مختلف عملیاتی ترکیب‌شده با روش الکتروشیمی بر مشکل کاهش جریان الکترواسمز و وقوع جریان الکترواسمز معکوس غلبه کرد و در نهایت راندمان حذف را افزایش داد.

واژه‌های کلیدی: پاکسازی خاک، توسعه پایدار، پارامترهای مؤثر، الکتروشیمی بهبودیافته.

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Introduction

Thousands of heavy metal-contaminated sites, organic compounds, and other hazardous wastes have been identified that have adverse effects on groundwater, soil, and ecosystem quality (van Cauwenberghe 1997). In the United States in 1990, nearly 217,000 contaminated sites were identified, and the US government has raised $ 187 billion to treat those areas within 30 years (Bhandari et al. 2007). In Europe in 1998, more than 600,000 contaminated sites were identified; it raised concerns and challenges about remediation contaminated soil in these areas (Bhandari et al. 2007). Soil pollution has become a critical environmental issue due to human activities and inefficient waste management methods (Habibul et al. 2016). Sustainable development obtains the needs of the current generation without compromising the ability of future generations to obtain their needs. It seems that the goal of sustainable development is to preserve human societies through a kind of development that does not destroy the fundamental systems that support environmental life (Zare et al. 2018). Many researchers and international organizations related to the subject have considered the environmental dimension one of the essential dimensions of sustainable development (Zare et al. 2018). The move towards green technologies, which means expanding clean technologies to minimize threats to human and environmental health, which will help reduce the environmental impact of technology assemblies and will have a positive impact on the development of products that are environmentally friendly and economically viable as a workaround for sustainable environmental development (Sarmadi et al. 2018). Appropriate and desirable interaction between the concepts of curriculum and sustainable development plays an essential role in the sustainability of the world. Proper attention to creating and organizing appropriate education and curriculum is one of the prerequisites for sustainable development (Zare et al. 2018). Soil destruction is a significant threat to development in most economies around the world (Keesstra et al. 2016). Soil protection is a set of strategies to prevent soil erosion and is part of the environmental knowledge of the soil (Farhadi et al. 2017). Education can be considered as a suitable solution to deal with environmental problems and achieve a healthier and cleaner environment for future generations, creating a biological balance and helping to reduce environmental problems (Meiboudi et al. 2017). Also, many studies have been conducted to emphasize the importance of extension training in accepting and applying soil protection proceedings (Farhadi et al. 2017). There are several remediation technologies for contaminated soils, which can be categorized into biological, physicochemical, thermal degradation, and soil stabilization treatments or in situ and ex situ treatments (Pham and Sillanpää 2015a). Each of them has its advantage and disadvantages, and in reality, the selection of proper soil remediation strategies is site-specific with various aspects for consideration. Among many different soil remediation technologies, electro remediation has emerged as a potential measure, especially for in situ soil treatment. In contrast, EK has been known mostly for its application in metal removal. The use of EK in organic decontamination is still in the development stage (Pham and Sillanpää 2015a). In this paper, we attempted to briefly introduce the EK method to move toward sustainable environmental development to achieve green technology, focusing on the elimination of organic pollutants using the EK method, citing analytical-descriptive concepts of documentary type to identify influential factors of EK, and EK Enhanced by Design & Operation Modes.

1- Electro remediation

In 1808, Reus first observed an electric field with a direct current flowing into a mixture of clay and water, followed by water moving in capillary tubes under the electric field from the anode to the cathode, and when the electric potential was cut off, the water flow stopped immediately (Acar and Alshawabkeh 1996). In the electro remediation process, two electrodes (anode and a cathode) are inserted into the sample, which creates an electric field with the passage of direct current with low intensity (Alshawabkeh and Bricka 2000). This method is widely used in fine-grained soils with low permeability (Shin et al. 2016; Gomes et al. 2012) because they are electrically and chemically active due to their unique mineralogical properties (Shang et al. 2004).

Electro remediation process in fine-grained, saturated, and semi-saturated soils and two laboratory and field scales is widely practiced (Zou et al. 2016). Shariatmadari et al. showed constant potential conditions consume lower energy consumption (Shariatmadari 1997). The electro remediation process of pollutant transfer from soil solution is based on five main mechanisms. Electrolysis reaction, ion migration, electrophoresis, and diffusion are fundamental electro remediation mechanisms. Diffusion plays a minimal performance in the remediation process because of the slowness of this process in removing organic pollutants (Virkutyte et al. 2002). Electrosmosis, induced by electric gradient, is a phenomenon of pore fluid, involving non-ionic species of soluble viruses, which moved toward electrodes. Ionic migration is the movement of ions toward the opposite charge electrodes. Electrophoresis is the transfer of colloidal particles with an electric charge and pollutants by the application of voltage gradient (Reddy et al. 2009). Figure 1 shows the main mechanisms of electro remediation reactions schematically.

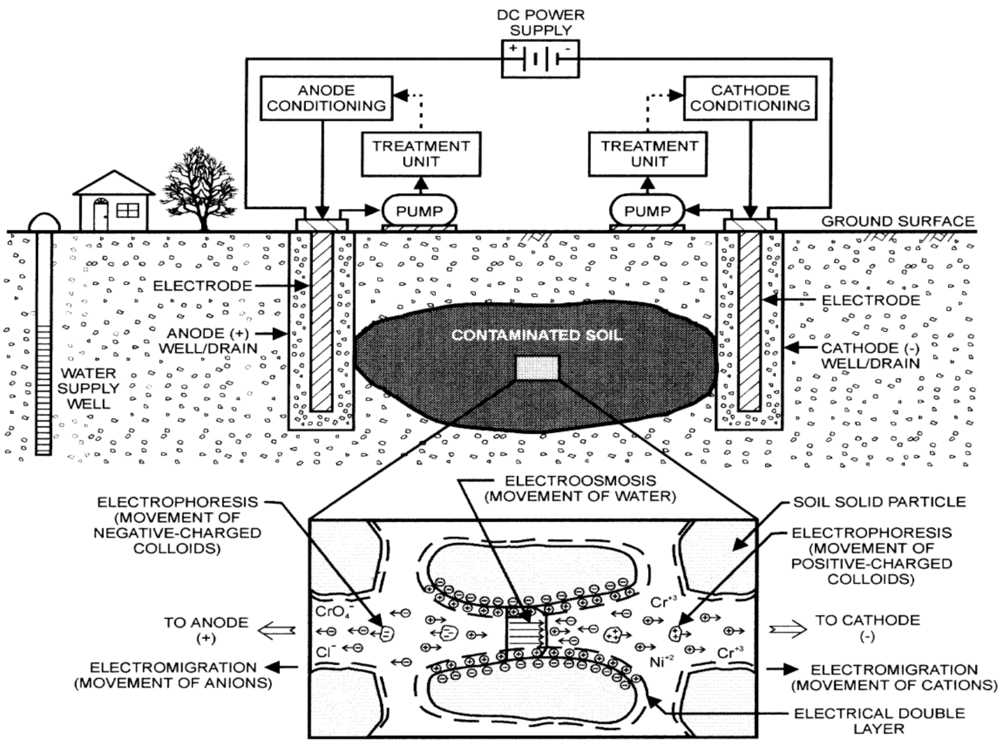

Figure 1. In-situ electro remediation (Reddy & Shirani, 1997)

In general, the surface of soil particles often has a negative charge as a result of which the electroosmotic flow moves along the surface layer towards the cathode (Lynch et al. 2007; Suzuki et al. 2014). Ionic migration is more commonly used to remove heavy metals and electroosmotic is often used to remove organic pollutants. The electroosmotic flow maintains positive (from anode to cathode) in case of the zeta potential remains negative. The zeta potential is of the double layer region around a colloidal particle indicates the degree of stability of the colloidal particles in the solution (Moayedi et al. 2014). In acidic conditions where the zeta potential is positive, the reverse electroosmotic flow occurs to the anode (Ammami et al. 2015). After the application of electric current in the soil, electrolysis reactions of water in the electrodes occur. Electrolysis in the cathode produces H_2 and hydroxide (reduction reaction as shown in Eq.1), and in the anode, produce hydronium and o_2 (oxidation reaction as shown in Eq.2) (Ramírez et al. 2015). Based on the results of these reactions, an acidic front is formed near the anode (decreasing in pH) and a basic front (increasing the pH) near the cathode that migrates to each other.

Eq.1: $4H_2O + 4e^- \rightarrow 4OH^- + 2H_2\uparrow$

Eq.2: $2H_2O + 4e^- \rightarrow 4H^+ + O_2\uparrow$

2-1-Absorption-desorption reaction:

The adsorption process refers to the separation of contamination from the solution and its sorption to the solid phase or soil surface. The adsorption process involves surface adsorption or ion exchange. The desorption process will be the reverse process and is responsible for releasing contaminants from the soil surface (Acar and Alshawabkeh 1993). Sedimentation-Dissolution Reaction: Sedimentation and dissolution reactions are essential in the electro remediation process. Contaminants are dissolved by the migration of hydrogen ions and acidification of the soil environment, and sedimentation of pollutants, such as heavy metals, takes place in areas with high pH (Saberi 2015).

Oxidation-reduction reaction: changes in soil electrochemistry during electro remediation processes cause various reactions, including oxidation and reduction of elements that are strongly affected by soil pH. Soil buffering capacity (resistance to pH change) is a crucial characteristic of the soil and can be significantly affected by geochemical reactions that are highly dependent on soil pH changes (Mosavat et al. 2012)..

Table 1. Advantages and limitations of electronic method (Virkutyte et al., 2002; Yeung et al., 1997)

Advantages	Limitations
• Can be applied in soils with low and heterogeneous permeability as well as a wide range of pollutants • Could be in saturated and semi-saturated environments • High flexibility in combination with other methods of decontamination to increase efficiency • High speed in launching and commissioning • No need to drill too much on the infected Site • Performance flexibility in and out of place	• Requires acidic environment to remove heavy metals • Low concentration of the desired ions and high concentration of non-target ions • Existence of large amounts of iron, iron oxide or sand and aggregates that cause heterogeneous in the soil • When high voltage is applied to the soil, the efficiency decreases due to the increase in temperature

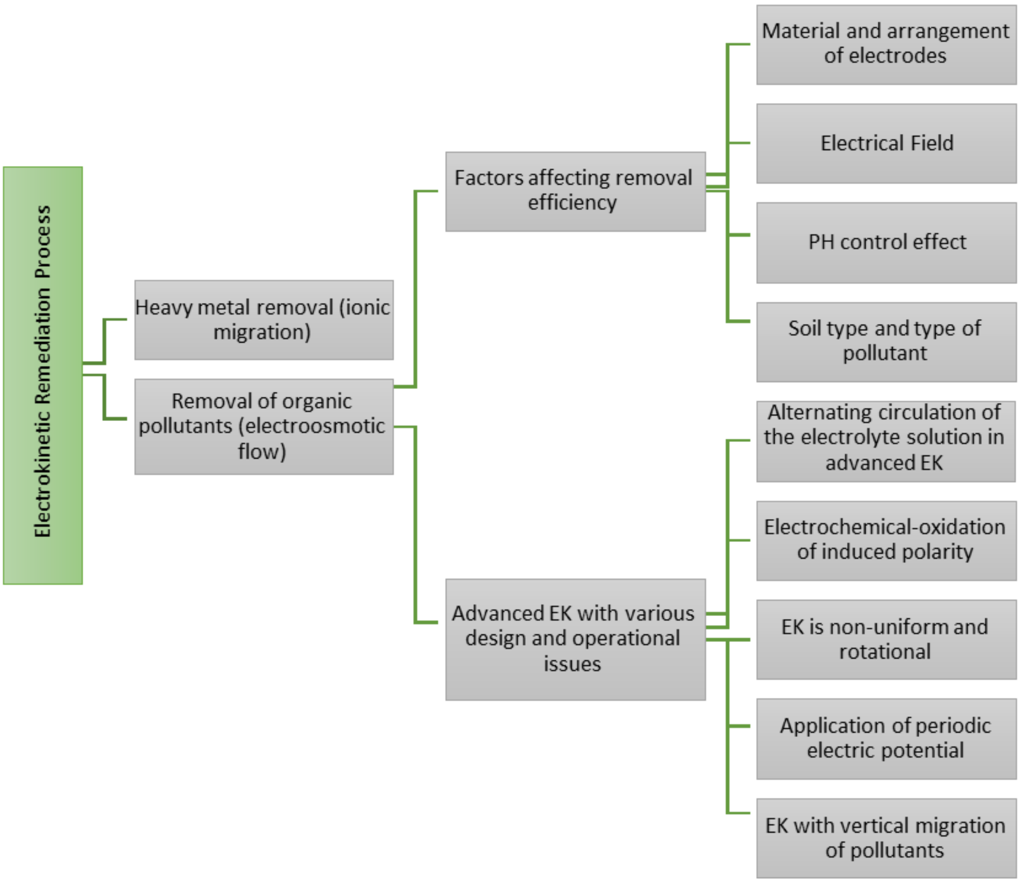

Figure 2. electro remediation process in organic pollutants

2-2- Advantages and limitations of the electro remediation method

The electro remediation method has attracted many researchers and environmental experts' attention in recent decades due to its many unique advantages(Reddy et al. 2009). Table 1 presents the advantages and limitations of the electro remediation method.

Figure 2 indicates a summary of studies on organic pollutants

3- Factors affecting the efficiency of the electro remediation process

Factors such as the type of electrodes, the type and texture of the soil, the position of the electrodes, the electric current, the type and concentration of pollutants, the cost and optimal time to perform the process affect the removal efficiency (López-Vizcaíno et al. 2014; Reddy and Chinthamreddy 1999). Some of the main influential factors are listed below.

3-1-Effect of soil type and pollutant type

In general, the soil is heterogeneous and consists of clay layers, sand, and clay layers among the sand layers. Soil nature has a significant effect on the removal of organic and inorganic pollutants (Alcántara et al. 2012). examined Pb and phenanthrene removal in kaoline and sandy soils. The results showed that phenanthrene removal was lower in sandy soil, attributed to the high organic matter content of sandy soils (Alcántara et al. 2012). Suang Shu et al. (2014) studied the elimination of pyrene by the electro remediation method in two types of soil (sandy soil and loam soil). After remediation, the efficiency of pyrene removal in sandy soil was measured to be 56.8% and in loamy soil 20.1% for 7 days. As a result, the removal efficiency in sandy soils is higher due to a higher flow rate and lower pH (Xu et al. 2014). Gajis et al. (2014) in a feasibility study of the electro remediation process to remove several organic pollutants in sandy loam soils and sandy soils, at the end of all experiments the percentage of contaminants remaining in the soil was between (17% to 50%) for sandy loam and between (27% to 48%) for sandy (Yeung et al. 1997).

3-2-Effect of pH Control

The electroosmotic flow is the fundamental mechanism of electro remediation transmission of inorganic and non-ionic pollutants. To achieve the desired efficiency, the electroosmotic flow should be from cathode to anode. Higher pH increases the cation exchange capacity and increases the absorption of heavy metals in the soil; therefore, to achieve proper removal of contaminants from the soil, it should keeping the pH low so that the contaminants remain in the solution phase. On the other hand, pH should be kept high enough to maintain the negative potential of the zeta potential (Yeung et al. 1997).

Alcantara et al. (2010) conducted a study on the remediation of a mixture of several polyaromatic hydrocarbons (PAHs) from kaolin soil. They observed that anode pH control causes a high electroosmotic flow. At the end of the experiment, about 40% of the initial hydrocarbons were removed (Alcántara et al. 2010). In standard electro remediation - Fenton remediation, Bokus et al. (2015) removed the maximum 67% of PAHs and pesticide pyrimethanil. As well as Bokus et al. (2015), to overcome the precipitation of the metals near the cathode chamber, used pH complexion and pH control agents in the cathode reservoir (Bocos et al. 2015).

3-3-Electrode Materials

Appropriate electrodes are often selected based on their performance in terms of current density, chemical stability, and resistance to corrosion (Pham and Sillanpää 2015b). Tsai et al. (2010) evaluated electro remediation efficiency (EK) using electrodes of different materials (graphite and iron) .Their results of the experiments indicated that the most suitable efficiency was achieved when the iron electrode was used (Tsai et al. 2010).

Mendes et al. (2012) examined the possibility of removing hydrocarbons from contaminated soil by analyzing a variety of electrodes. For this purpose, several types of electrodes were analyzed for use as an anode. They also used two types of carbon fabric electrodes and titanium as cathodes. The results described that the carbon fabric absorbs organic compounds; consequently, its use as a cathode in the remediation of pollution was discarded (Méndez et al. 2012). Jalali et al. (2014) used geosynthetic electrodes to solve problems such as corrosion of metal electrodes and their short life. Discolored clay substrate and crack formation in the clay bed, due to corrosion and separation of metal electrodes in the acidic environment near the anode that was resolved using geosynthetic as electrodes (Jalali 2014).

3-4-Effect of electrode arrangement and direct and periodic electric field (one-dimensional-two-dimensional)

Wu et al. (2013) used numerical simulations to identify and evaluate the electrode configuration to electro remediation at chemical oxidation sites (EK-ISCO) in low permeability soils (Wu et al. 2013). Zhi Wu et al. (2013) found that closing electrode spacing results in faster EK-ISCO remediation (Wu et al. 2013). Assessed electro remediation soil flushing (EKSF) technologies for the removal of 2,4-dichlorophenoxyacetic acid from spiked soils using an electrode arrangement consisting of one cathode encircled by six anodes (1c6a) and one anode encircled by six cathodes (1a6c) (Risco et al. 2016). The volume of water extricated from cathodes with 1a6c is seven times higher than that of the 1c6a approach. The arrangement 1c6a was the most effective and accomplished a transfer of 70% of the herbicide included in the soil to flushing water (Risco et al. 2016). Lee et al. (2016) conducted a small-scale 3D electro remediation process laboratory study to increase the remediation efficiency of oil-contaminated soil and heavy metals. Lee et al. (2016) used 0.10 M KH_2PO_4 as an anodic solution and removed more than 95%,50%, and approximately 20% of copper metal in 21 days (Lee et al. 2016).

4-Advanced electro remediation with various design and operating conditions

Using EK, coupled with other enhancement technologies, not only can the remediation time be effectively saved, but also organic contaminants decompose in the soil and therefore do not require additional remediation (Ren et al. 2014).

4-1-Circulation electrolytes in enhanced electro remediation (CEEK)

One of the main disadvantages of EK remediation is the increase in acidity during EK remediation. The increasing acidity of the soil environment dramatically destroys soil components and have a complementary effect on soil zeta potential, which reduces electroosmotic flow. Therefore, by establishing a flow circulation system, by neutralizing the pH, the acidity of the soil can be avoided (Lin et al. 2016). Zhou et al. (2016) investigated the effect of ammonia continuous circulation enhanced electro remediation of fluorine contaminated soil and to examine its impact on soil pH after remediation. The results showed that with the continuous circulation of ammonia as an electrolyte solution, it increases the migration of fluoride and reduces energy consumption (Zhu et al. 2016).

4-2-Electro-Chemical-Geo-Oxidation

In Stuttgart, Germany, ECRTs has been used as an innovative, cost, and time-effective method for organic pollution remediation (Chang and Cheng 2007). ECRTs can remove organic contaminants in situ and aquifers and groundwater using electro remediation oxidation (Pham et al. 2009). ECGO is an in-site technology available at ManTech International to remove organic contaminants in water and soil (van Cauwenberghe 1997). Sang et al. (2018) used the electro remediation oxidation method in-site (EK-ISCO) as an innovative and effective method to remove oil contaminants from the soil to solve the problem of high external oxidation costs (Song et al. 2018).

4-3-Non-uniform and rotary electro remediation

Lou et al. (2006) used non-uniform electro remediation transportation processes using an electrode matrix and a rotational operation mode to combine organic pollutants (phenols) and damaged bacteria in the soil (Luo et al. 2006). The results showed that non-uniform electro remediation processes could increase the biological decomposition of phenol in the soil, the efficiency of which depends on the operating state of the electric field (Luo et al. 2006). Fan et al. (2007), using the non-uniform electro remediation process in a hexagonal matrix resulting from the arrangement of electrodes during two standard operating modes (two-sided and rotary), achieved the removal of organic pollutants more efficiently and cheaper (Fan et al. 2007).

4-4-Periodic voltage

Hahladakis et al. (2016) concentrated on assessing the electro remediation of real contaminated sediments with heavy metals and polycyclic aromatic hydrocarbons (PAHs), periodic voltage, and recently experimented non-ionic surfactants. The results showed that the “day on-night off” utilization mode of voltage, in conjunction with the chosen solubilizing agents, favored the overall EK removal process (Hahladakis et al. 2016; Asadollahfardi and Rezaee 2018).

4-5-Upward electro remediation soil remediation

An upward electro remediation soil remediation (UESR) method applying vertical non-uniform electric field generated by point-shaped electrodes was presented for the removal of heavy metals from contaminated soils. A non-uniform upward electric field was generated between an anode set in soil and a cathode placed on the soil surface. Unlike conventional electro remediation that uses boreholes or trenches for horizontal migration of heavy metals, the UESR process used vertical non-uniform electric field, causing upward transportation of heavy metals to the top surface of the remediated soil (Cheng et al. 2007).

Giannis et al. 2012, conducted the vertical electro remediation -flushing technology for the simultaneous removal of organic pollutants (phenanthrene and pyrene) and heavy metals (Cd, Pb, Zn) from contaminated soils. The highest removal efficiency was obtained for Cd, Zn and Pb,82%,73%, and 37% respectively from natural soil after 8 days of remediation. In terms of the organic pollutants, under the conditions accompanied the electro remediation-flushing remediation, low removal efficiency for phenanthrene (29%) and pyrene (19%) was obtained. However, it is remarkable that without the use of any solubilizing agent, the organic pollutants could be removed following the pore fluid's movement (Giannis et al. 2012).

Conclusions

Researchers' investigation over the past 22 years has led to many models that need to be taken to design future projects to improve contaminated sites as a step towards sustainable development Therefore, the recovery of contaminated soils with organic contaminants is very specific. The results obtained in modifying and improving a site cannot be used for other infected sites. This is due to the high impact of the physical and chemical properties of soil and possible interactions with organic pollutants. This is due to the high complexity of the electro remediation process and the presence of many factors and changing the effective parameters in the results of electro remediation soil improvement and improve efficiency. Besides, chemicals used to improve electro remediation may complicate system behavior, and the results of contamination removal may vary significantly from site to site; Recommendations that can be considered in future studies and to achieve success in soil conservation programs and its optimal use using training to identify the electroconvulsive method and the factors influencing the removal of organic pollutants are:

1) Extensive and comprehensive studies should be conducted on the simultaneous remediation of organic pollutants and heavy metals using an electro remediation process.
2) Compared to other methods of physical and chemical remediation methods, electro remediation requires more time to process, lasting from a few days to a few months. On the other hand, due to the low solubility of organic pollutants and the low efficiency of removal, it is possible to improve it by (1) combining with other processes (2) Designing and operating various operational issues. (3) Investigating the factors influencing the efficiency of this technology
3) Combining the electro remediation process with other designs and different operational modes, in addition to increasing the efficiency of remediation of organic pollutants in the soil, saves the time and costs of the reaction.
4) In addition to focusing on the removal efficiency of pollutants, it is important to achievement to lower and more efficient energy consumption; it is recommended to reduce the test time and the potential difference between the electrodes and the type of electrode used. Another effective parameter of this system is the use of other types of electrodes with polymeric (geosynthetic) conductive materials.

Conduct experiments with the three-dimensional arrangement of electrodes instead of the linear arrangement of electrodes and change the number and distance of electrodes in the three-dimensional arrangement and finally compare with the one-dimensional arrangement that has not been done in previous studies.

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