sustainability

Review

Sustainability in ElectroKinetic Remediation Processes: A Critical Analysis

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Abstract: In recent years, the development of suitable technologies for the remediation of environmental contaminations has attracted considerable attention. Among these, electrochemical approaches have gained prominence thanks to the many possible applications and their proven effectiveness. This is particularly evident in the case of inorganic/ionic contaminants, which are not subject to natural attenuation (biological degradation) and are difficult to treat adequately with conventional methods. The purpose of this contribution is to present a critical overview of electrokinetic remediation with particular attention on the sustainability of the various applications. The basis of technology will be briefly mentioned, together with the phenomena that occur in the soil and how that will allow its effectiveness. The main critical issues related to this approach will then be presented, highlighting the problems in terms of sustainability, and discussing some possible solutions to reduce the environmental impact and increase the cost-effectiveness and sustainability of this promising technology.

Keywords: electrokinetic approach; sub-surface contamination; heavy metals; EKRT; renewable energies; electrochemical technologies; wastewater treatment; soil remediation; photovoltaic; wind

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1. Introduction

The unstoppable industrial progress that began at the end of the 19th century has generated countless cases of environmental contamination, with the release of large quantities of inorganic and organic pollutants.

Several different physicochemical and biological approaches have been suggested for the remediation of contaminated water [1–5] and soils [6–10]; however, selecting a suitable technology is often a difficult yet crucial step for the successful reclamation of a contaminated site [11,12]. Among the various methods investigated, the ElectroKinetic Remediation Technology (EKRT) has proved particularly interesting and efficient, owing to its possibility of being applied in situ [13,14] and to its more environmentally friendly character compared to other approaches [15]. Indeed, one of the main advantages of the electrokinetic process is that it theoretically operates exploiting electrons as the only reagent [16]. However, the use of electrolytes or complexing agents is often mandatory to obtain measurable results in a reasonable time and with sustainable energy consumption.

Unlike other recent reviews on electrochemical applications [17–20], the purpose of this contribution is to present an overview on applications and technical solutions related to the electrokinetic remediation of soils, shifting the focus more on their sustainability compared to previous revisions [21]. The fundamentals of the technology will be briefly mentioned first, together with the phenomena that occur in the soil, which are the basis of its effectiveness.

2. Fundamentals of Electrokinetic Remediation

The EKRT approach consists in applying an electric potential gradient to induce a low electric current across a portion of the contaminated soil to be treated, using electrodes suitably positioned in the subsurface [22]. It can be applied both in situ and ex situ and allows high efficiency even if performed in soils with low permeability [23,24], as the applied electric field can easily reach contaminants embedded deep in the subsurface, which other technologies are unable to reach [25,26].

Depending on the intensity of the resulting electric current and the characteristics of the system (salt content, moisture, soil composition, etc.), different physical, chemical and electrochemical processes are induced. These may allow a substantial migration of species through the soil and towards the wells (electrode housing and electrolyte solution) from where they can eventually be removed [27] by electrodeposition, adsorption, precipitation, or co-precipitation on the electrodes [28], or simply by removing the contaminated electrolyte solution (which can then be treated and reused [29]).

The most relevant phenomena induced in a soil by the applied electric field are listed below [30] and graphically illustrated in Figure 1:

- electroosmosis, i.e., the displacement of the solution naturally present in the soil;
- (electro)migration of electrically charged species;
- electrolysis, a process that occurs on the surfaces of electrodes, generally at the expense of water (decomposition reactions);
- electrophoresis, that is the transport of charged particles of colloidal size present within a stationary fluid, due to the application of an electric gradient.

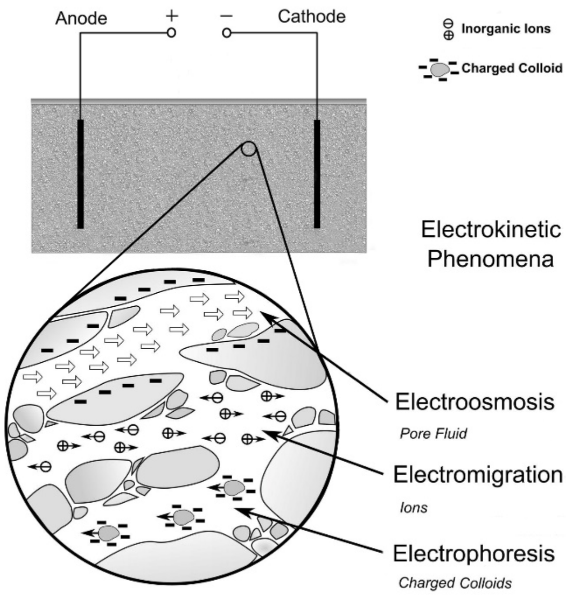

Figure 1. Detail of the main mechanisms occurring during an ElectroKinetic Remediation Technology (EKRT) remediation. Adapted from [31].

Moreover, since soils are usually characterized by a high ionic resistance, an increase in temperature is normally observed (Joule–Thompson effect) [17]; the dissipated energy is proportional to the square of the current flowing through the soil. It is worth noting that, in the presence of volatile pollutants, this can cause significant environmental problems.

Under the action of the electric field generated between electrodes, anions and cations move toward the anodes and the cathodes, respectively (electromigration). In addition, the anodic oxidation of water generates an acidic front, while the reduction of water at the cathodes produces an alkaline front. The H^+ and OH^- species thus formed not only contribute to electromigration but can also allow changes in soil pH with possible repercussions on soil chemistry. Among the resulting chemical reactions, the dissolution or precipitation of salts and minerals can either facilitate or hinder the release of pollutants fixed in the soil [32].

3. Evolution of the Technology and Its Applications

Although the first investigations on electroosmotic flow date back to the beginning of the 19th century [22,33], the electrokinetic approach was applied only several decades later, initially as a consolidation process for fine soils [34] and subsequently for the recovery of heavy metals [35,36].

In the first investigations [23,30], the removal of contaminants was attempted simply by inducing an electric current through electrodes inserted directly into the contaminated soil, but the efficiency of the remediation was low. Numerous improvements have therefore been proposed, including:

- the optimization of the pH of electrolytic solutions [37];
- the use of an ion-exchange membrane to prevent the migration of protons (acidity) and hydroxyl ions (alkalinity) from the electrodes in the soil [37–39];
- the increase in the mobility of pollutants by adding complexing agents [40] and surfactants [41];
- the optimization of the effective volume, by varying the disposition of the electrodes according to the nature of the site and the target contaminants [42,43].

Since electromigration generally provides a greater impact than electroosmosis, for many years EK remediation has mainly focused on charged species such as heavy metals [44–46]. More recently, research has focused on the use of EKR technology for the removal of dangerous organic substances from soil [47] or from marine and river sediments [48]. A plethora of technologies have been reported, ranging from simple electrokinetic soil flushing for soils with low hydraulic conductivity, to the use of permeable reactive barriers loaded with granular activated carbon (GAC) [49], zero-valent iron (ZVI) [50–52], or even microorganisms [53] used to retain or transform the organic species mobilized by the applied electric fields.

The main challenge of EK technology is the conversion of low solubility pollutants into mobile forms in order to extract them. Therefore, enhancing agents, added to the process fluids, are necessary to obtain an effective removal of all type of contaminants. During an electrokinetic soil flushing, surfactants are normally introduced into the process fluid to allow the formation of micelles (charged particles) with the species target of the remediation. These micelles are then transported across the soil under the effect of the electric field applied to promote the removal of organic and inorganic compounds [23,28], causing electrophoresis to significantly contribute to the remediation process of relatively permeable soils.

The ever-increasing attention to environmental sustainability is now catalyzing the interest in new (bio)remediation technologies, which involves the minimum use of chemicals (and external energy) and consequently implies a lower environmental impact [54]. In this context, electro-bioremediation technologies have recently been investigated, especially following the discovery that many microorganisms are capable of degrading environmental contaminants, including polycyclic aromatic hydrocarbons (PAHs), using electrodes as electron acceptors [55–57].

Furthermore, bioelectrochemical systems allow manipulating the redox potential of the contaminated matrix, thus establishing in situ conditions that favor the biodegradation of contaminants [58,59].

On the other hand, the coupling of the electrokinetic approach with bio-remediation and phyto-remediation could represent a more sustainable approach. Their energy requirement is low, the addition of chemicals is often not necessary, and the physicochemical and 'biological' (e.g., the fertility) characteristics of the soil at the end of the treatment are improved compared to the initial situation. EK treatment can increase the bioavailability of organic pollutants by facilitating contact between microbes and nutrients and/or pollutants, and the weak electric current may also directly stimulate microbial activity [60,61] or degrade some of the pollutants through an electrolytic reaction [62]. In addition, the application of an EK treatment can improve the growth and respiration of plants (which in turn can facilitate the removal of metals) and facilitate the spread of microorganisms of the rhizosphere (with possible enhancement of the biodegradation of organic contaminants) [63]. Other authors instead argue that the benefits are linked to the influence of the electric field on enzymatic reactions, water activity, and membrane transport [64].

It has been reported that the elimination of organic and/or inorganic contaminants through the combined use of plants and an electric field applied through the soil to be treated is an effective approach [65], able to control the transport of contaminants in the rhizosphere, as well as to prevent the establishment of strong acid or alkaline fronts in the soil [66]. However, suitable operating conditions must be selected to ensure the survival and development of microorganisms and/or plants [67]. Extreme pH values and high temperatures can be produced during the process, which are two of the most critical parameters for keeping microorganisms active [18,31]. Since most processes induced by the electric field have a negative effect on the viability of microorganisms, the simultaneous optimization of both electrokinetic and biological processes can be very challenging [59]. The application of an electric field can also cause negative effects to plants: for example, O'Connor et al. [68] reported growth inhibition and death of plants located near the electrodes. The negative effects of electricity would be linked to changes in soil pH associated with water electrolysis and phyto-toxicity due to the increasing bioavailability of metals.

Another recently explored approach is the coupling of EKRT and nanotechnology. Nanomaterials and nanoparticles have peculiar properties which, if properly exploited, can allow first-rate performance while maintaining high sustainability [69,70].

Hosseini et al. [71] suggested employing the EK technique to distribute additives such as nano-silica or lime through the pores of a collapsible soil (e.g., loess) to stabilize or improve its physical properties. On the other hand, the approach can also be applied to achieve remediation goals; for example, Czinnerova et al. [72] recently explored the potential of a combined nZVI-EK bioremediation approach to clean up highly polluted aquifers from chlorinated ethenes (nZVI stands for zero-valent nanoscale iron). They demonstrated that EK improves the long-term reactivity of nZVI by also stimulating microbial degradation activity by increasing the groundwater temperature.

4. Side Effects during EKR

The EK approach has several advantages over other remediation technologies: it is easy to operate, relatively cost-effective and can be applied both in-situ and ex-situ [73]. In the case of an in-situ application, the moisture content and the nature of the contaminated soil are the main limiting factors (remediation is theoretically faster in sands and gravels than in silts and clays, but the different hydraulic properties can play against the former, since the interstitial solution content can be much lower). Further problems are related to the possible presence of buried metal conductors and contaminants whose presence was not known.

Figure 2 shows more specifically the main criticalities connected to the use of the EKR, which will be discussed in more detail below.

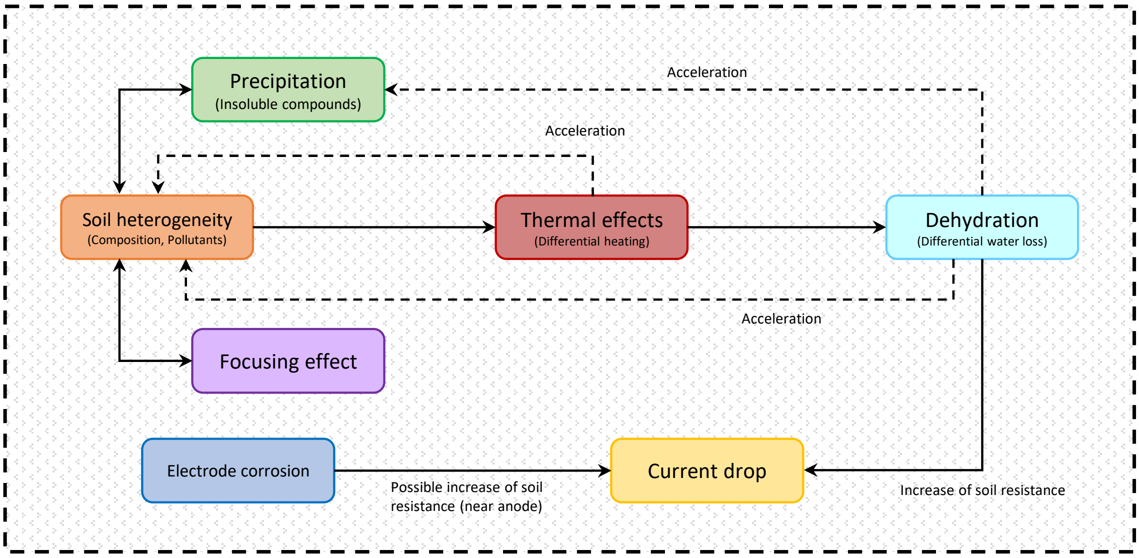

Figure 2. The interconnections between the main negative effects in the electrokinetic approach.

4.1. Thermal Effects

Due to the resistivity of soils, much of the energy supplied when an electric field is applied is dissipated as heat. The greater the resistivity of the soil, the greater the potential required to induce a given amount of current (flow of charged species) and, consequently, the greater the associated thermal effect. Unfortunately, most electrokinetic tests are conducted in the laboratory on small systems (often less than 1 kg of soil), which means that also the currents and potentials involved are proportionally reduced. Moreover, the studied systems are in contact with, and exchange heat with, the surrounding environment, thus any thermal effects related to these tests is reduced. As a result, only a few researchers have commented about thermal effects in their studies [74,75]. In particular, differences were found in the spatial distribution of temperature in the soil, with a more marked increase at/near the cathode than at/near the anode. It has been proposed that the difference is due to the alkaline front originating from the cathode [75,76]. On the one hand, alkalinity reduces the mobility of metal species (decrease in electrical conductivity) through the formation of the respective hydroxides; the clogging caused by the latter also decreases the hydraulic conductivity.

In general, soil thermal conductivity is a complex function of the water content, porosity and temperature of the soil and it is not easy to predict the repercussions this may have on the remediation process. In a recent article, Wen et al. [77] commented "*heating helps reducing the fluid viscosity, which results in enhanced electroosmotic flow*"; this is questionable, because a higher temperature increases the solubility of salts in the pore fluid, and a higher ionic concentration plays against electroosmotic phenomena. Correctly, Wen and collaborators also commented that heating can influence transport of inorganic pollutants in soil by diffusion/evaporation of soil pore water. Moreover, thermal effects can accelerate many chemical reactions and improve the desorption of many organic contaminants from soil [18].

4.2. Precipitation of Insoluble Compounds

Some salts present in the soil solution, especially those of calcium and magnesium, can be involved in precipitation or adsorption reactions at the electrodes, thus reducing their conductivity and/or influencing the electrochemical reactions that allow the passage of electric current. Besides, the precipitation of poorly soluble compounds reduces the porosity and increases the tortuosity and resistivity of the soil, possibly preventing the transport of pollutants.

At relatively high temperatures, the strongly alkaline environment and the dehydration caused by evaporation create favorable conditions for the precipitation of salts in the cathode region. Wen et al. [78] reported that the cumulative adsorption capacity of species at the electrode increases over time, even though the rate of this process decreases over time.

To manage this problem, it is essential to monitor and possibly adjust the pH at the cathodes, by adding acids. Vocciante et al. [29] proposed a particular water management system, in which the solutions from the anode and cathode wells are extracted and sent to a common tank to allow mutual neutralization (if necessary, acid can also be added to bring the pH to values close to neutrality), before being reintroduced into the soil. Other proposed solutions, such as the use of a solution of citric acid and polyaspartic acid as electrolytes, are plausibly capable of destroying the $CaCO_3$ deposits formed on the cathodes [78].

4.3. Electrode Corrosion

For a practical application of EKRT, the choice of the positive electrode (anode) may represent a problem: under the effects of the electrical polarization, an unsuitable electrode can be quickly corroded, with formation of unwanted corrosion byproducts. Furthermore, if the latter are characterized by poor electrical conductivity, corrosion can cause an undesirable increase in the voltage required to generate the current, thus increasing energy consumption [79].

Ideally, the electrodes should have mechanical strength and toughness that allow for easy transportation, and be able to resist deposition of insoluble species as well as corrosion in order to increase service life and reduce operating costs. Stainless steel is a conductive material with high strength, good toughness, and low cost [78]. Xu et al. [80] found substantially similar effects in the use of stainless-steel electrodes and graphite electrodes when treating a soil contaminated with Cd. Although both materials are subject to corrosion during the EKR process, stainless steel releases metal ions, including chromium, causing a secondary pollution problem.

Some electrically conductive polymers, such as polypyrrole (PPy), can be used as a coating to protect stainless steel electrodes from corrosion; unfortunately, PPy cannot provide long-term protection in chloride-containing environments [81]. Kim et al. [82] proposed to use a pulsed polarization system, as this allowed reducing the corrosion rate of the electrode by about 38%.

Overcoming the initial criticisms of the potentially higher costs, and considering that stable electrodes can be reused, conductors based on titanium coated with conductive and catalytically active ceramic films are probably the best solution in terms of both stability and energy optimization.

4.4. Focusing Effect

What is commonly referred to as the “focusing effect” occurs where the acid and alkaline fronts meet, during an EK remediation process, acting as a barrier to the migration of those species (especially heavy metals) that are able to form poorly soluble hydroxide [83,84].

According to Cheng et al. [85], the loss of mobile ions is one of the main reasons for the termination of an EK remediation process. If the process can initially be considered as mainly supported by electromigration, during the course of the process the transport of species due to the electroosmotic flow becomes more and more decisive; however, the latter can be hindered by aggregation of colloidal particles of the soil.

The most common method to counteract the focusing effect is to control the pH at the cathode by keeping it at a low level [18,86]. The desorption of the heavy metals accumulated due to the focusing effect can also be facilitated through an exchange of polarity between the electrodes (with consequent inversion between the acid and alkaline regions). This approach is effective in weakening the focusing effect too [87]. Alternatively, it is possible to prevent the alkalinity produced by the cathodes from entering the soil by using ion exchange membranes [88].

More recently, other approaches have been proposed: Sun et al. [89] suggested to use a super-imposed electric field (SE-EKR) to overcome the problem, while Li et al. [90] recommended using "approaching anodes" (AAs-EKR). Both AAs-EKR and SE-EKR address the focusing effect in a similar way, i.e., by pushing the focusing region towards the cathode. Electrolytes containing chelating agents and/or surfactants can also help overcoming the focusing effect, as they help improve the solubility of metals by forming coordination compounds over a wide range of pH [87,91].

4.5. Dehydration

Due to the action of electroosmotic flow (EOF), water in soil can be driven to move from anode to cathode under the application of a direct current electric field. Although EOF is beneficial for the removal of pollutants, it is necessary to prevent excessive dehydration as this cannot only cause the interruption of electrical contact between the soil and anodes (the first to be affected by the effects of dehydration) but also cause the precipitation of salts originally dissolved in pore water. In addition, the rate of electromigration is also directly influenced by the electrical conductivity and path length (tortuosity) of soil [21].

One of the reasons why electrolyte solutions are used extensively is to avoid these negative effects. Indeed, increasing soil moisture content improves the soil conductivity (thus reducing ohmic effects), and promotes the electromigration [92]. In previous studies, adding an electrolyte chamber, rather than inserting electrodes directly into the soil, greatly increased the electrical conductivity of the system, while at the same time continuously supplementing water to the soil. However, dehydration was still noticeable in the middle of the reactor. In [29], saturation of the volume of soil to be treated with water/electrolyte was proposed, together with a lateral confinement using sheet piles or pumping wells placed just outside the EKRT field and which recirculate the liquid to the inside the field itself, to prevent the dispersion of water and/or contaminants.

The effect of soil water content on the removal of pollutants has rarely been reported in the past scientific literature. Shin et al. [92] conducted the research and found the optimal condition was 50% soil moisture for EKR of soil contaminated by As; compared with a 35% soil moisture, the removal rate of arsenic under optimal conditions was nearly doubled. In addition, there are close correlations between dehydration, precipitation of insoluble compounds, the focusing effect, and the thermal effect in the EKR process. As shown in Figure 2, precipitation of insoluble compounds and focusing effect occurring at/near the cathode may result in high resistance in the region, which can induce local high temperature and lead to a lot of soil moisture to evaporate. Water loss in soil near the cathode will in turn indirectly result in a lot of salt in the soil solution being separated out and enhance the local thermal effect because of the low electrical conductivity of the bulk soil. Local high temperature induces and strengthens the asymmetric distribution of soil conductivity and increases the speed of precipitation phenomena. In general, free water and interstitial water in soil can be displaced by EOF, but EOF assisted by thermal effect can also remove bound water [93], and even completely dry the soil so that it loses the ability to conduct electricity. This vicious circle continues until the current disappears.

5. Energy-Related Aspects

From an application point of view, the engineering implementation of electrokinetic remediation technology is often criticized for its high operating costs, of which electricity consumption is one of the predominant items [21]. To make technology more accessible (sustainable), it is advisable to look for ways to improve the efficiency of use of this energy, as well as to develop self-powered technologies.

5.1. Energy Consumption

The energy consumption of an EKR process is mainly linked to the electric field necessary to induce the movement of the target species of the reclamation. The following main processes can be distinguished: electrokinetic migration (electromigration, electroosmosis, electrophoresis, in order of importance of the contribution to the process), ohmic effects (consumption of energy in the form of heating), and electrolysis reactions. Through appropriate engineering choices [15], it is possible to minimize the energy consumption due to ohmic effects; however, since these are related to the occurrence of the electrical phenomena necessary to implement the technology, they cannot be completely avoided.

In addition to the above, the energy consumption linked to the operation of any pumps (often necessary to manage the process fluids, or the dosage of reactants) as well as the energy required for any treatment of the process fluids must be considered. Although the latter contribution could be considered not strictly linked to the remediation process, an in-situ treatment of process fluids can allow for the reuse of part of these fluids, limiting the consumption of reagents and/or water.

5.2. Energy Saving

A significant amount of the cost of implementing an EKR technology is related to electricity consumption. According to the estimates presented by Alshawabkeh et al. [42], the cost of electricity represents 10–15% of the total cost and 25% of the operating costs. However, as anticipated, the weight of the electrical contribution can be modified following specific engineering choices. For example, Vocciante et al. [15] reported how a different arrangement and a reduced number of electrodes—made possible by using an electrolytic solution and a sophisticated recirculation system—can allow to contain the costs of the plant (in terms of both environmental and economic impact) at the expense of higher energy/operational consumption. Indeed, based on the example application discussed in [15], and considering the improvements and the process water management detailed in [29], the cost of electricity amounts to 30–35% of operating costs (further information will be provided in a future publication).

This apparently counterproductive choice allows one to achieve higher levels of overall sustainability when the technology is combined with the use of renewable energy sources.

The use of pulsed electric fields, widely exploited in other fields such as electrodeposition, can represent another good compromise: Jo et al. [94] reported an electrical energy consumption equal to 42% of that of a conventional process. The energy saving is due to the “switch-off time” between one pulse and the following [95,96]. Such a solution is also useful for reducing the effects of any polarization phenomena. From an operational point of view, the application of a pulsed electric field does not substantially affect the speed of remediation, as the generally high impedance of the system means that the response is not so rapid as to stop the processes in progress, despite the momentary absence of the electric field.

According to Fu et al. [75], another strategy to save on electricity consumption is to increase the conductivity of the system through the addition of appropriate electrolytes. In their studies, the addition of 0.1 M citric acid improved the efficiency of energy use in the treatment of a soil contaminated with Cr.

5.3. Use of Renewable Energies

Renewable energy represents an alternative to conventional electricity and can make these processes not only more sustainable and eco-compatible but also autonomous, while reducing the environmental problems associated with the use of energy from fossil sources [97].

There are several ways of coupling energy from renewable sources to an electrokinetic remediation process: storage and subsequent utilization, random direct coupling, targeted direct coupling, and targeted storage with direct coupling [20].

As for the first approach, the typical energy storage devices are traditional lead-acid batteries. The preliminary energy storage avoids subjecting the remediation system to the risk of potential fluctuations in the production of electricity and also allows to work during periods in which the production of renewable energy is low or zero (i.e., when solar radiation or wind flow are limited by time or by specific climatic conditions [19]). However, this operating mode foresees inevitable energy losses linked to inefficiencies in the conversion of chemical energy into electrical energy. Additionally, traditional batteries have limited capacity and limited power, which may not be high enough to supply the required energy to the remediation plant. In this context, attention has been focused on the use of green energies and on the development of innovative energy storage systems [20,98].

The use of photovoltaic panels and fuel cells based on bio-electrochemical processes (microbial fuel cells) represent two potentially interesting approaches for the development of self-powered technologies. Example studies of electrokinetic remediation powered by solar photovoltaic modules can be found in the literature, relating to soils contaminated by Cr^{VI} [99], As [100], and Cu [101]. Wind turbines, photo-electrocatalytic fuel cells, and triboelectric nanogenerators are additional renewable energy sources that have been tested on a laboratory scale for the reclamation of wastewater and soils, coupled with various electrochemical technologies (electrodialysis, electrooxidation, electro-Fenton, electrokinetic soil remediation) [20,98].

5.3.1. Photovoltaic Solar Cells and Wind Turbines

The intermittency of energy from some renewable sources increases treatment times and reduces process control, due to the variability of the applied power. Some of the obtained results showed that the direct use of such energy sources is questionable, as the remediation trend obtained during the functioning of the process can be affected during the periods in which the process stops, due to the retro-diffusion of pollutants in the absence of electric field [102]. Other authors have reported excellent results in removing metals from the soil using solar energy as a power source [100,103,104]. Hansen and Rojo [105] commented that an intermittent process allows for the system to be depolarized, which improves the results thanks to the synergistic effect of different mechanisms, including dissolution and electrokinetic transport. Hassan et al. [101] obtained a 75% average removal of Cu from a soil originally polluted with 355 mg/kg of metal, achieving a removal of up to 92% near the anode. In the case of As removal, the solar-powered process consumed 50% less energy than an analogous treatment powered by traditional sources [100]. In addition, better results have been obtained by reusing acid effluents as washing fluids [106].

Millán et al. [16] compared three different ways to power an electrokinetic remediation process: direct powering with photovoltaic energy, application of a constant potential gradient, and application of a constant power gradient. Direct powering (without preliminary energy storage) is perhaps the most sustainable strategy, but the results have not been exciting. The authors highlighted a low transport of contaminants per kWh (due to the inversion in the transport of pollutants during the night) and massive evaporation effects (due to the high electric field applied at noon). Regarding efficiency, they found that it is better to work in mild conditions of constant potential gradient. Conditions that are more drastic can negatively affect the soil in terms of pH (more extreme values) and lower water content, leading to less removal of pollutants. As previously commented, evaporation mechanisms are enhanced by operating at high electric fields (due to ohmic heating).

According to Ganiyu and Martínez-Huitle [107], direct powering with photovoltaic energy reduces both investment costs (no need for batteries, inverters, or power supplies) and maintenance costs (absence of used batteries to manage). On the other hand, there are economic and technical disadvantages, such as the additional cost of cables and electricity transport, and the energy loss associated with transport [16]. In terms of process efficiency, there are problems related to mass transfer when working at excessively high or low values of applied current density, as well as problems related to the presence of reversible reactions [108].

Wind turbine-powered electrochemical technologies have been less investigated and implemented, possibly because their installation requires solid and robust foundations that are not suitable for residential areas [20]. Unlike photovoltaic energy, wind power can also be available at night; however, the electricity generated depends on the wind conditions [109,110] and the energy storage system.

The use of wind energy to power the electrokinetic remediation of a soil contaminated by the herbicide 2,4 dichlorophenoxyacetic acid has recently been reported: lower removal efficiencies (53.9% after 15 days) were achieved compared to using electricity from conventional sources (90.2%) [111]. The lower efficiency was attributed to fluctuations in the power supply caused by variations in wind conditions. The use of batteries could have minimized the problem while increasing installation costs.

The typical intermittency of renewable energy sources makes it difficult to use them continuously and constantly. For this reason, energy storage is a key issue to be urgently addressed [112]. Energy can be stored using mechanical, electromagnetic, thermal or electrochemical technologies [108,113]. Lead-acid batteries are the most common storage systems, while other rechargeable batteries, supercapacitors and redox flow batteries (RFBs) represent important alternatives. Among these, lithium-ion batteries and RFBs are undoubtedly the most promising systems [114,115]. The main advantages of these technologies are the decoupling between capacity and power [108,112,115], which allow better management and use of the renewable energy.

5.3.2. Costs and Social Impacts of Solar and Wind Energy

Solar and wind energy are not exempt from negative environmental impacts [116,117]. In the case of the use of photovoltaic panels, land use, modification of the natural habitat, as well as the use of water and hazardous materials in their production must be considered among the negative effects [117].

Wind turbines can shrink, fragment, or degrade the habitat of wildlife, fish, and plants. The rotating blades pose a threat to birds and bats, as well as causing "noise pollution" [116]. Nonetheless, solar and wind energy play an important role by contributing to the containment of global warming, as they allow reducing the consumption of energy from conventional sources and the related CO_2 emissions. In addition, they also have an impact on local development, by stimulating employment in the industrial, agricultural, and construction sectors [116,117].

5.3.3. Self-Powered Technologies

An alternative approach is the conversion of the chemical energy present in organic matter into electrical energy [118]. This can be achieved in a microbial fuel cell (MFC), where microorganisms that oxidize organic matter at the cell anode produce electrons that are transferred to the cathode by external wires; at the cathode, oxygen acts as an electron acceptor, being reduced to water. It has been suggested that the weak electric field generated by the MFC can drive the electrokinetic process. Habibul et al. [119] studied the process on soil contaminated with Cd and Pb, reporting that 31% of Cd and 44% of Pb were removed from the region near the anode after 143 and 108 days, respectively. Song et al. [120] tried to improve the electric current provided by the MFC by supplying a 3% straw to the fuel cell and reported that the removal of Pb and Zn increased from 15% to 37% and from 10% to 25%, respectively. Although MFCs are economical and environmentally friendly, an EKR process guided in this way has significant limitations, mainly low removal efficiency and very long remediation times.

6. Conclusions

Although in-depth studies (both laboratory and pilot scale) are still required to achieve an adequate level of technological readiness, the electrokinetic remediation technology looks very promising in terms of perspective, adaptation, and applications.

Given the critical issues highlighted, it is necessary to rely on all possible measures aimed at reducing energy consumption to increase the sustainability of the approach. First, the process would benefit from a reduction in ohmic effects, as these rarely contribute favorably to the technology. Where possible, the use of electrolytic solutions allows the conductivity of the soil to increase, avoiding the adverse phenomena of dehydration. In case of contamination by ionic species, the increase in the level of humidity (even up to complete saturation) favors electromigration compared to electroosmosis and electrophoresis, thus allowing to reduce treatment times and therefore overall energy consumption. To prevent the introduced solution from becoming a vehicle for the transfer of contamination, the use of suitable geometries capable of "confining" the volume to be treated (for example, using internal cathodes and peripheral anodes, or extraction wells arranged around the area to be treated) can be decisive.

However, the last obstacle to its large-scale implementation is perhaps represented by the optimized recourse to energy from renewable sources. Among these, solar photovoltaic and wind turbines are the most consolidated technologies to complete an eco-sustainable commercialization. Self-powered technologies are the cheapest, but the current and potential values obtainable from these sources do not seem adequate for the purpose (although they appear to be suitable for other electrochemical applications). The coupling between energy from renewable sources and EKR applications also requires the development of efficient energy storage systems, to reduce costs and improve the management of process, minimizing the side effects associated with the production and disposal of these systems.

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