Review Article

Environmental applications of electrochemical technology. What is needed to enable full-scale applications?

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Abstract

In recent years, thousands of scientific articles have considered the application of electrochemical technologies to remediate environmental problems ranging from the treatment of polluted soils to the removal of hazardous species from industrial liquid wastes. New research topics continue to emerge. Despite such research efforts, the technology readiness level (TRL) for many of those technologies remains very low; although most are considered promising, many are far from being introduced as efficient processes into the market. Important barriers need to be overcome to reach high TRLs. Some of these are scientific or technological and generate the opportunity for critical, applied research. Others are related to the lack of components in the value chain of the technology and generate opportunities for entrepreneurs to benefit from an improvement in the TRL. In this short review, a brief description of the current state of the most relevant technologies which are still in low TRL is carried out, highlighting barriers that must be removed to achieve full-scale applications in industry.

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Introduction

Applications of electrochemical technology in environmental remediation have been the focus of many research activities over the last three decades [1]. Thousands of publications have sought to develop new technologies or improve existing processes. After this long period, very few technologies are being applied at full scale and most technologies evaluated remain at the status of ‘promising’ technologies. Most have identifiable benefits, but important technological and cost handicaps can be identified, which are related to missing components in the value chain of the technology.

Electrolytic processes include the cathodic deposition of metals found in the widely used electrowinning and electrorefining processes and the oxidation of organic compounds, either direct on the anode surface or mediated by oxidants produced on the anode or on the cathode surface. This second case is of a great significance because hydrogen peroxide can be efficiently produced from the reduction of oxygen using gas diffusion electrode (GDE) electrodes [2] or more recently using cross-flow electrodes even with the application of high pressures [3], leading to more efficient processes. Electrocoagulation technologies are also initiated by the electrolytic release of coagulants from a sacrifice anode and can be used to break up emulsions in industrial wastes or to remove colloid pollutants in those wastes and during the treatment of surface water [4].

Electrochemically assisted separation processes include not only electrodialysis [5] and capacitive deionisation [6] (which allow concentration of ions in liquids) but also electrokinetic processes (which allow transport of species in solid–liquid mixtures such as soil or sludge) [7,8].

Not all electrochemically assisted processes are at the same technology readiness level (TRL). Large differences are found between electrowinning or electrodialysis and the electrolysis of wastewater polluted with organics. Sometimes, the key to understand these

differences is a necessity for companies aiming to develop those processes, especially when there is a lack of alternatives. When competitive solutions are fully marketed, it is difficult to displace them, unless the advantages of the new technology would help to obtain a very fast payback. Lessons must be learned about the successful implementation of electrodialysis and electrodeposition of metals, to achieve full applicability of other electrochemical technologies and to allow returns on invested money to companies and society.

Many electrochemical technologies, including the electrochemical oxidation of wastewater, electrodisinfection, electrocoagulation and soil electroremediation, are now in a position to improve TRLs. Sometimes, the challenge to be faced is technical; in other cases, it is economic. The following is a brief critique of each of these.

Challenges in water treatment: electrodisinfection

While the use of mixed metal oxide anodes has been demonstrated for the disinfection of water, particularly the production of chlorinated disinfectants, a long path remains to push the technology from traditional disinfection of saline swimming pools or spas, in which many small and medium enterprises (SMEs) are focussing their markets [9].

Two important challenges must be faced to meet this challenge. One is to determine how to avoid the formation of hazardous by-products such as chlorates or trihalomethanes. Chlorates are formed by oxidation of hypochlorite or by its disproportionation, which is a natural process that also occurs during ageing of the disinfected water. This species is related to serious health problems because it is known to affect the nervous system. The second type of hazardous species is even more controversial. Chlorinated organics are formed from the combination of organic matter with active species of chlorine, and these species are related to cancer and other very serious illnesses. These species are not characteristic of the electrochemical process, because they are also formed during the application of conventional chlorination technology. The other challenge is to apply replacements for mixed metal oxide anodes, such as diamond-like coatings, capable of not only oxidising chloride ions but also producing more efficient disinfectants, including hydroxyl radicals. When using these electrodes, problems associated with the production of hazardous species can be aggravated, because of the well-known production of perchlorates via oxidation of chlorates. These new electrode materials open the possibility of promoting the action of other oxidising reagents, for example, ozone and peroxosalts, [10] to help remove resistant pathogens. An adequate contact time between water

and the anodes in the electrolyser, a sufficiently large specific current and the cathodic formation of hydrogen peroxide to prevent further oxidation of chlorine to chlorates and perchlorates are among strategies considered to overcome problems [11]. In this way, R & D departments in many companies are making efforts not only to develop new electrodes but also to design electrochemical cells that avoid the formation of undesirable products and offer improved volumetric electrode area, mass transfer and current efficiency. Important projects include the SafeWaterAfrica European Union (EU) programme, in which two full-scale demonstrators based on this technology are being commissioned to treat water in rural communities in South Africa and Mozambique, and they are expecting to be in operation during 2019 [12].

Challenges enabling electrocoagulation to compete with coagulation

Electrocoagulation is an alternative to conventional coagulation, in which the coagulant is provided by the dissolution of sacrificial electrodes [13]. The simplicity of operation and the side processes involving the production of bubbles are major advantages. In the first case, the dose of coagulant reagent can be adjusted simply by regulating the intensity exerted, and the handling of reagents is avoided. In the second case, a proper mechanical cell design can help to utilise oxygen and hydrogen microbubbles to promote turbulence and enhance the flocculation of particles (so-called electroflocculation). Sometimes, separation can be promoted by flotation, once microbubbles have adhered to the surface of the flocs, diminishing their overall density (in so-called electroflotation). However, authors disagree in this assessment: the main advantage of electrocoagulation, compared with coagulation, is related to the pH regulation. In conventional coagulation, the reagents consist of salts of iron or aluminium; the main reagent in electrocoagulation is hydroxyl ions, and the counter ion does not increase the salinity of the treated waste. Salts used in coagulation perform as Lewis acids and require neutralisation by alkali additions, to obtain an acceptable pH. This increases the salt load of the treated waste and the volume of sludge, resulting in an effluent with a much higher conductivity. Conversely, the electrocoagulation can self-regulate the pH, and there is no necessity of addition of pH-neutralising reagents [14,15].

Although the laboratory design of an electrocoagulation cell is very simple, its scale-up is not as easy [13,16]. It is not always possible to use tank cells with sheets of iron and aluminium, and there is a need to use cheap materials as sacrificial electrodes. Use of low-quality iron or aluminium can enable bipolar electrode configurations to be used. Achieving a low cell voltage is a major challenge, and components of cell voltage are rarely

quantified. The combination of electrocoagulation with free radical–assisted processes is a promising approach to encourage its implantation at full scale [17,18].

Challenges in the treatment of industrial wastes: removal of refractory pollutants

One of the most exciting research topics in recent years has been the development of technologies for the treatment of industrial wastes in cases where the application of biological technologies and other cheap processes are inefficient. In this field, many technologies have been developed, most of them being classified as advanced oxidation processes (AOPs). The key oxidant involved is the hydroxyl radical. This radical has two great advantages as compared with other traditional oxidants, as in the case of chlorine used in the 70s and 80s: the hydroxyl radical minimises formation of hazardous by-products and can be faster and more efficient, because of the harsh oxidation conditions. Despite outstanding developments in this field, reflected in a large bibliography, many challenges remain [19].

Regarding the relationship between these AOPs and electrochemical technology, several features may be noted: (1) the confirmation in 2003 of the active role of hydroxyl radicals in the electrochemical oxidation with diamond electrodes [20] and (2) the introduction of the combination of Fenton and electrochemical technologies [21,22] led to the recognition of a new category of AOP processes, namely electrochemical AOPs [2,23–25].

Regarding the current status of this technology, the application of electrochemical AOPs for the removal of many types of organic pollutants (both, from synthetic and real wastewater) has been widely studied. Many authors have tried to elucidate the role of electrochemical cell design and types of electrode on the electrochemical process. It has been stressed that a key requirement in the potential applicability of the electrochemical technology is the efficient use of all the components of the electrochemical cell [26–28]. This implies not only the implementation of a suitable anode but also the synergistic use of the cathode reaction and the promotion of mediated oxidation processes in the bulk during the treatment. In addition, the increase in the mass transfer rate inside the cell is also important. Thus, an efficient mechanical design of the cell and a careful choice of the operation conditions are critical to achieve good performance. Despite these efforts, the technology is still at an intermediate TRL and major developments are needed to reach higher readiness levels which recognise adequate mechanical design of the electrochemical cell to obtain good patterns for flow and current distribution [29], scale-up either by increasing electrode size or by stacking [30], the

management of hydrogen generated [31,32] and DC electrical power requirements [33]. Many of these points necessitate collaboration between companies and academia; any slowness at this interface must be considered a major handicap to the implementation of electrochemical technology.

Challenges in soil remediation: not only metals but also organics?

Application of electric fields to transport water and pollutants in soil has been known for many decades. The possibilities of using this technology to efficiently remove metals from soil or those derived from the transport of volatile and semivolatile pollutants with the heat produced [34] have been described. Both applications are currently at a high TRL, with many companies applying commercial solutions in the first and over TRL 6 in the case of the second.

The focus of research in recent years, however, has been the use of this technology to remove other pollutants, particularly hazardous organics contained in soil [35] or in marine and river sediments [36]. A plethora of technologies has been reported, ranging from simple electrokinetic soil flushing for soils with low hydraulic conductivity [37] to the more complicated use of permeable reactive barriers loaded with granular activated carbon (GAC), zero-valent iron [38] or even microorganisms [39] have been used to retain or transform organics mobilised by applied electric fields. Scale-up is critical; it affects not only the speed of treatment [40–42] but also the efficiency and selectivity reported in research studies: there is often poor reproducibility of results obtained on the laboratory scale (where electrokinetic processes are rate controlling) to larger scales (where ohmic and heating effects can dominate). Multidisciplinary aspects range from the compaction of soil to the physical position of electrodes. Further research is needed because technical and environmental concerns will demand careful evaluation before full-scale applications. These include controlling side effects during treatment (such as anodic precipitation, heating of the soil, gas formation), as well as evaluating the potential ecotoxicological effects of the surfactants, cosolvents, oxidants or reductants used [43].

Challenges in the treatment of gaseous streams: a new frontier in environmental applications?

Treatment of gaseous streams has not been extensively studied using electrochemical technologies, except for studies focused on the valorisation of gaseous streams in high-temperature fuel cells. However, there is a very interesting market niche, not only related to industrial environments but also complementing other treatment technologies in which gaseous effluents are produced [44]. The application of electrochemically assisted

technologies to remediate soil can lead to significant increases in soil temperature and to release of volatile and semivolatile pollutants that must be treated. The integration of adsorption units with electrochemical cells is the goal and promising results have recently been obtained [45], although further research is needed to scale-up this treatment technology.

Once the pollutant is in the liquid phase, the process becomes easier and similar to wastewater treatment. This means that expertise developed over previous decades can be used. However, some recent studies have pointed out the possibility of using methanol instead of water for collecting more efficiently the organics contained in the gas [46]. This is especially important when electrolysis is combined with adsorption, because methanol is more efficient to regenerate GAC, and results show that combined GAC adsorption–desorption in methanol and electrolysis of the resulting solution allows to remove completely target pollutants in a more efficient way, requiring much less electrical charge than the electrolysis in aqueous phase. However, the efficiency of the overall removal process not only depends on the performance of the electrochemical process but also depends on the adsorption equilibria of the pollutant in methanol media. In any case, preliminary works allow the confirmation of this combined technology as a real challenge in the treatment of gaseous streams polluted with organics.

Challenges in sanitary effluents: is there slot for electrochemical technology?

Medical facilities, such as hospitals, produce sanitary effluents with a high load of a great variety of chemicals (e.g., pharmaceuticals, detergents, disinfectants, heavy metals, radionuclides) and pathogens. No legal regulations have been established to define how to manage and treat sanitary effluents, and thus, they are directly discharged in public sewage for treatment at conventional municipal wastewater treatment plants (WWTPs). The inefficiency of biological treatments from WWTPs to treat sanitary effluents leads the spread of chemicals and pathogens in the environmental water sources, negatively affecting aquatic organisms and human health. This has led to proliferation of numerous studies searching for technological solutions to decrease the impact of sanitary effluents in the environment. Two main strategies have been developed: (1) boosting efficient treatments at WWTPs by the application of additional treatment and (2) pretreatment before their discharge into WWTPs [47]. The second option is emerging as the key alternative, as it addresses the problem in more concentrated effluents and the volume of wastewater treated is further much lower than in the other case. Some authors have focussed on the treatment of hospital urines, where both pharmaceuticals and pathogens are more concentrated and an

improvement in the process efficiency is expected, as well as lower investment and operational costs. Among the available technologies, a single electrochemical technique such as electrochemical oxidation or electro-Fenton and the combination of free radical–assisted electrochemical technologies have demonstrated complete disinfection and partial oxidation of chemicals towards the decrease of toxicity and increase of biodegradability of urines and sanitary effluents [48,49].

Challenges in the sustainable powering of electrochemical technology

Sustainability is currently an overused word. We are far exceeding the capacity of our planet to provide raw materials and energy, and solutions for problems generated should be as much sustainable as possible. Here, it is important to realise that the main requirement of electrochemical processes is electricity and green supply by photovoltaic solar panels or wind turbines has been proven to be efficient to power these devices, especially when the power demand is not so high. This supply can be direct or by means of energy storage devices. In the first cases, there are important differences between the application to water treatment [31] or soil because of the reversibility of the transport processes in soil remediation [50,51] which leads to lose efficiency over nights when photovoltaic (PV) technology is used or when no wind is available to produce electricity. In the second case, the use of redox flow batteries is seeming to be a good alternative because they can store important amounts of energy and help to regulate renewable

Figure 1

Number of publications per year related to different environmental electrochemical technologies according to Scopus.

energy in this electrochemical application [52–54]. Despite this topic is being considered useful in the scientific and technical literature, it requires further development in the next few years to reach high TRLs enabling full-scale application.

Concluding remarks: the big challenge — scaling up and commercialisation

Figure 1 shows a rapid survey carried out with the number of items published per year according to the Scopus since 2000, in the different topics covered in this short review (it is only informative because the keywords are not unique for each technology, and it is not aimed to be an exhaustive evaluation but just a quick outlook that helps the reader to have in mind the bibliometric significance of the research topic), where it can be seen that publications per year related to electrochemical oxidation (keywords used: electrochemical oxidation, electro-fenton, electrofenton, anodic oxidation and electrooxidation) have increased significantly, although they are stabilised in the last five years, suggesting some sort of exhaustion of the new ideas in the topic. In comparing the different technologies, it can be drawn that this high number of publications from electrochemical AOPs is more important than that of the publications related to electrokinetic soil remediation. In turn, these publications remain more important than the number related to electrocoagulation and electrodialysis. It is also interesting to see that the rate of publication is almost constant in all technologies

except for electrodialysis in which a slight increase is observed over recent years. All these data indicate the relevance of the research in environmental electrochemical technologies.

As explained in this work, bibliometric comparison is not the focus here. Rather, real world applicability of the technologies is critical. Although the original idea dates back to the early 70s, it is at the end of the 80s when NASA defined the nine-level TRL scale, which later was adopted by the EU in the mid-2000s to be used in its research framework program H2020. From the initial steps in the research (TRL1: basic principles observed) to the final (TRL9: actual system proven in operational environment), different goals and actors must perform to reach the market from fundamental research. Figure 2 shows what can be considered as the most advanced applications of the environmental electrochemical technology, the approximate TRL reached currently and barriers that are limiting the improvement in the TRL. In some cases, nonenvironmental electrochemical technologies have been also included for the sake of comparison (electrowinning and electrorefining data are provided to point out the differences between the industrial and the environmental processes and purification of acids and alkalis with electrodialysis, for the same reason).

This figure summarises graphically the main highlights introduced in this short review. At this point, it is

Figure 2

	TRL 1	TRL 2	TRL 3	TRL 4	TRL 5	TRL 6	TRL 7	TRL 8	TRL 9	Challenges	
Removal of metal pollutants										0	Electrochemically assisted soil remediation
Removal of volatile & semivolatile organics										1	
Removal of other organics										1	
Desalination										0	Electrodialysis
Purification of acids & bases										0	
Treatment of metal ions polluted wastes										2	
Oxidative treatment of industrial wastes										2,3,4	Electrolysis
Oxidative Treatment of hospitalary wastewater										1	
Treatment of gaseous streams										1,2	
Disinfection not for drinking applications (pools)										0	
Drinking water disinfection										1,4	
Electrowinning or electrorefining										0	
Electrowinning for metal recovery from wastes										2,3	
Dehalogenation of chlorinated wastes										2,3,4	
Treatment of wastes polluted with colloids										4	Electrocoagulation
Treatment of emulsions										4	
Treatment of dye polluted wastewater										4	
Removal of turbidity from water & wastewater										3,4	

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TRL of the main environmental electrochemical technologies, including the key challenges that have to be overpassed to increase this level: no significant challenges (0); further fundamental work (1); components development (membranes, electrodes, cells ...) (2); scale up (3); competition with efficient technologies (4). TRL, technology readiness level.

important to note that increasing the TRL of a technology is a collaborative work not only including the work of base or technological scientists from academy but also the cooperation with small medium-sized enterprises (SME) and large enterprises (LE). Thus, all components of the value chain must be considered to get a successful implementation. The interest in each of these factors is complementary, and their appropriate combination must be considered as the major handicap to environmental electrochemical technology over the next few years.

Conflict of interests

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

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This work elucidates how to select the appropriate electrochemical flow reactor and types of electrode, depending on the laboratory or pilot scale application.