1 **Electrochemical treatment of highly concentrated wastewater: a**
2 **review of experimental and modeling approaches from lab- to full-**
3 **scale**

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Abstract

This review proposes an insight into the prospects of electrochemistry for the treatment of highly concentrated effluents in three sections. The first section focuses on the challenges inherent to such kind of wastewater, divided into five categories: industrial wastewater (e.g., pharmaceutical, electronics, chemical, food-processing), hypersaline effluents (e.g., RO concentrates), solutions contaminated with a mixture of organic and inorganic contaminants (e.g., leachate, mining), highly viscous solution (or non-Newtonian liquid) (e.g., sludge) and solutions of high COD load but with low pollutant content (e.g., from soil washing). The second section of this review then focuses on the treatment strategies to ensure that the electrochemical treatment is adapted to these very specific waste streams, including the influence of operating conditions, electrode materials and processes (with special emphasis on anodic oxidation, electro-Fenton and electrocoagulation). The final part focuses on the perspectives of electrochemical treatment of challenging wastewater, by giving the engineering parameters to ensure successful upscaling of electrochemical processes in terms of modeling mass transport, charge transfer and hydrodynamics, reactor designs and energy requirements. The review concludes on process combinations, where electrochemistry could complement traditional methods of treatment, in order to improve the overall efficiency of the integrated system.

Keywords: electrochemical advanced oxidation processes, high organic load wastewater, industrial wastewater treatment

Electrochemical wastewater treatment strategies

This section of the review presents the most relevant treatment strategies involving electrochemical technologies that have been applied to handle waste streams with high levels of contamination as defined in the previous section, with a focus on organic pollutants and real effluents. Waste streams polluted with high concentrations of inorganic and ionic species will only be briefly mentioned. Moreover, emphasis has been placed on EAOPs and EC.

Electrochemical technologies: an overview

Electrochemical technologies have proven to be powerful methods for the treatment of numerous kinds of waste streams (Martínez-Huitle et al., 2015; Moreira et al., 2017). The success of the treatment relies first of all on the physicochemical properties of the wastewater and second, on the electrode materials and thus the type of process that is applied. EAOPs, based on the production of $\cdot\text{OH}$, are highly efficient in degrading dissolved organic pollutants, especially aromatic/cyclic and unsaturated compounds that possess the highest reaction constants with $\cdot\text{OH}$ (Buxton et al., 1988). Hence, effluents containing important amounts of such organic contaminants are excellent candidates for EAOP treatment, including pharmaceutical, food processing, textile, reverse osmosis (RO) concentrates and landfill leachate effluents, as well as other waste streams from various chemical sectors. Highly loaded effluents are also ideal candidates for $\cdot\text{OH}$ -based EAOPs because high concentrations of organics avoid mass transport limitations that are generally associated with electrochemical methods. Nevertheless, very high contents of organic matter require longer treatment times and extensive energy consumption, having a negative impact on the feasibility of the treatment.

Anode materials

In electrochemical processes, the electrode material is a determining factor, as electrode properties define the type of treatment and thus the chemical and electrochemical mechanisms that are involved. Anodes with a high O_2 evolution overpotential have the capacity to produce $\cdot\text{OH}$ on their surfaces ($\text{M}(\cdot\text{OH})$) and are excellent materials for direct AO, while anodes with the ability to produce chlorine oxidative species from the oxidation of Cl^- are excellent choices for indirect AO of organic pollutants (Martínez-Huitle et al., 2015). Figure 1 shows the main anode materials typically used for AO. They have been classified according to their capacity to produce $\text{M}(\cdot\text{OH})$, which is linked to

their oxidative power. Two main classes can be distinguished: “active” (low O₂ evolution overpotential) and “non-active” (high O₂ evolution overpotential) anodes. Boron doped diamond electrodes (BDD) are the most powerful anode materials due to their outstanding properties, including a high O₂ evolution overpotential (2.8 V vs SHE), high chemical stability and durability (Brillas et al., 2011). Recently, ceramic sub-stoichiometric anodes (Ti₄O₇) have also demonstrated their remarkable performance in the degradation/mineralization of organic contaminants, showing similar properties as BDD (Ganiyu et al., 2016; Olvera-Vargas et al., 2018; Yang et al., 2020); yet, their application to wastewater treatment is still at the development stage.

In contrast, water oxidation on electrodes with a low oxygen evolution overpotential (“active anodes”) leads to O₂ with negligible formation of M(•OH) and thus poor AO performance. Among them, dimensionally-stable anodes (DSA) are good electrocatalysts for the oxidation of Cl⁻ into chlorine oxidants, making them the most convenient choice for chlorine-mediated oxidation (AO-Cl). The fundamentals and applications of EAOPs for wastewater treatment have been thoroughly documented in several review papers of high impact (Brillas & Martínez-Huitle, 2015; Chaplin, 2014; Feng et al., 2013; Moreira et al., 2017; Sirés et al., 2014).

Cathode material: Fenton-based methods

Cathodes with the electrocatalytic capability of producing H₂O₂ from the two-electron reduction of dissolved O₂ represent great options for electrochemical treatment, allowing the indirect oxidation of organics via the Fenton’s reaction (Brillas et al., 2009). During EF, the Fe²⁺ catalyst for the Fenton’s reaction is continuously regenerated on the cathode surface through reduction of Fe³⁺. Iron ions are generally supplied as Fe²⁺ salts (mainly FeSO₄) in catalytic amount. Alternatively, the use of iron anodes can produce the catalyst in EF-like systems. However, continuous dissolution of a sacrificial anode generates

considerable amounts of undesirable Fe sludge. Various types of carbonaceous electrodes have been used in EF, including carbon felt, carbon cloth, carbon fibers, graphite, reticulated vitreous carbon (RVC), carbon nanotubes (CNT), carbon sponge, gas diffusion electrodes (GDE), mesoporous carbon and graphene-based electrodes. Carbon materials have the advantage of being inexpensive, durable and nontoxic. During the last two decades a great deal of work has been done on the development of carbon-based electrodes for EF applications. Excellent papers on the topic and a recent book are available for review (Babuponnusami & Muthukumar, 2014; Brillas, 2017; Brillas et al., 2009; Casado, 2019; Huong Le et al., 2017; Nidheesh & Gandhimathi, 2012). Fenton-based technologies can be substantially improved under UV irradiation, due to the contribution of different photolytic reactions (photolysis of H_2O_2 and Fe-carboxylate complexes, as well as the photoexcitation of semiconductor electrodes). UV-based EAOPs include photoelectro-Fenton (PEF), solar photoelectro-Fenton (SPEF) and photoelectrocatalysis (PEC) (Brillas, 2017; Garcia-Segura & Brillas, 2017).

Enormous progress has been made in the development of advanced electrode materials for EAOPs, including composite electrodes coated with nanomaterials, metallic catalysts and semiconductor photocatalysts (Garcia-Segura & Brillas, 2017; Huong Le et al., 2017; Moreira et al., 2017). Nonetheless, such electrodes are still at the development stage and further work is needed before large-scale applications are envisaged. In fact, the performances of novel electrodes are generally investigated in synthetic diluted solutions of model organic pollutants, but practical applications require robust, durable and mechanically reliable electrodes. Commercially available electrodes commonly used in electrochemical wastewater treatment include BDD, DSA, Pt, Ti, carbon felt and carbon-PTFE-GDE.

For the sake of simplicity, EAOPs are generally performed in undivided electrochemical reactors in which the anode and the cathode are contained in the same compartment (more details of this subject will be discussed in the next section). In this kind of reactor configuration, the performance of the process can be optimized when both the cathodic and the anodic processes contribute to the oxidative degradation of the organic pollutants. In this scenario, the coupled EF-AO system has shown great degradation/mineralization efficiencies when treating several organic pollutants and waste streams (Olvera-Vargas et al., under review; Oturan et al., 2015; Sopaj et al., 2016). In this “paired” process, $\cdot\text{OH}$ are generated through two routes: i) by the homogeneous Fenton’s reaction (EF) promoted by H_2O_2 cathodic formation in carbonaceous electrodes and; ii) by the discharge of water at powerful anodes like BDD and Ti_4O_7 (AO). As mentioned above, greater amounts of $\cdot\text{OH}$ are produced when the process is carried out under UV/vis irradiation (EF and PEC).

Electrochemical technologies are considered green processes because they utilize electrons (electric current) to generate the oxidizing agents, thus avoiding the handling of hazardous chemicals. Furthermore, they do not virtually generate secondary wastes. The main constraint limiting their large-scale application is related to technical aspects and economic considerations (total capital investment, total product cost, energy consumption).

Defining the treatment strategy

Most of the investigations on electrochemical processes have been carried out in bench-scale reactors making use of synthetic solutions of target pollutants or simulated waste streams. However, the use of synthetic solutions avoids real conditions, which are crucial for the development of large-scale treatment systems. When defining the most appropriate treatment strategy, there are three key points that need to be taken into consideration: i)

the physicochemical characteristics of the effluent: amount of organic matter COD, BOD₅ and TOC), TSS, TDS, ionic species (inorganic ions and metals) and pH; ii) the goal of the treatment in terms of water quality targets and; iii) the properties of the treatment process to be applied. Once these three points have been addressed, the second step involves the evaluation and optimization of the process. Again, three main actions need to be taken: i) optimization of the process according to the main variables affecting the efficiency (current/potential, types of electrodes, pH, catalyst concentration in the case of EF, supporting electrolyte) (Brillas & Martínez-Huitle, 2015; Chaplin, 2014; Feng et al., 2013; Moreira et al., 2017; Sirés et al., 2014); ii) cost/efficiency evaluation and; iii) design of large-scale systems (engineering of the electrochemical reactors and electrodes). The optimization is generally done in small-scale setups and the data obtained from this evaluation will serve as the starting point for the development of a pilot. This strategy selection flow is illustrated in Figure 2. In the following sub-sections a discussion of the most relevant strategies that have been applied to the treatment of highly polluted waste streams by electrochemical methods is presented, focusing on EAOPs and EC. A compilation of representative works that have been considered is displayed in Table 1.

Anodic oxidation

For the treatment of highly polluted effluents, the first approach consists of the evaluation of individual electrochemical processes to assess their feasibility. AO has been the most commonly used technology because of its simplicity and versatility with respect to the composition of the effluents. Indeed, AO has shown a greater range of applicability than EF because its efficiency relies almost entirely on the anode material and is not pH restricted. BDD has been the most widely utilized electrode, followed by DSA. A great number of studies have demonstrated the outstanding oxidation capabilities of BDD during both AO and EF (cf. Table 1). Commercial applications of EAOPs with BDD have

been proposed by the German and American companies CONDIAS GmbH¹ and Advanced Diamond Technologies Inc.², respectively. The main limitations of BDD technology are still the high price of the electrodes and their size (restricted to 1 m²), but as the market grows, larger electrodes are becoming available.

As an example of AO applied to high strength wastewater, municipal landfill leachate previously submitted to biological activated sludge was treated by AO-BDD in a pilot plant in which 250 L of leachate were treated (Anglada, Urtiaga, et al., 2010). The “DiaCell” treatment unit consisted of three electrochemical reactors, each one containing five DiaCell sets, where each set comprised ten single cells (anode/cathode pairs). BDD were used as anodes, while the cathodes consisted of stainless steel (SS). The electrodes were circular with a surface area of 70 cm² and separated by 1 mm. The setup was operated in discontinuous mode at a flow rate of 300 L min⁻¹. More than 70% of COD removal and 37% of NH₄⁺ removal were achieved following 8 h of treatment at 450 mA m² (COD₀ = 3106 - 4057 mg L⁻¹ and [NH₄⁺]₀ = 1300 - 1355 mg L⁻¹). It is important to mention that the leachate influent was filtered before electrochemical treatment, while no electrolyte was added because of the high conductivity of leachate (high Cl⁻ content).

DSA are stable and durable materials used in many industrial processes such as chlorine production, electrowining of metal ores and water electrolysis, among others (Trasatti, 2000). Since they are commercially available for large-scale applications, they are convenient materials for practical use. DSA are used especially for effluents containing important amounts of Cl⁻ ions by promoting the production of chlorine

¹ Condias - Ihr Partner für elektrochemische Verfahren mit Diamantelektroden | Ihr Partner für elektrochemische Verfahren mit Diamantelektroden. <http://condias.de/> (accessed April 27, 2018).

² Advanced Diamond Technologies. <http://www.thindiamond.com/> (accessed April 27, 2018).

oxidizing species by electrochemical oxidation of Cl^- . In this way, chlorine species are the prime oxidizing agent in the bulk solution (AO-Cl). Yet, because of their restricted capacity to generate $\text{M}(\bullet\text{OH})$, DSA anodes are known to achieve only moderate degradation/mineralization yields: indeed, the standard reduction potentials of Cl_2 ($E^\circ(\text{Cl}_2/\text{Cl}^-) = 1.36 \text{ V vs SHE}$) and HClO ($E^\circ(\text{HClO}/\text{Cl}_2) = 1.49 \text{ V vs SHE}$) are lower than that of $\bullet\text{OH}$ ($E^\circ(\bullet\text{OH}/\text{H}_2\text{O}) = 2.8 \text{ V vs SHE}$) (Deborde & von Gunten, 2008). For example, an RO concentrate contaminated with 28 pharmaceuticals and pesticides, and high amounts of Cl^- (1500 mg L^{-1}) was treated by AO using a $\text{Ti/Ru}_{0.7}\text{Ir}_{0.3}\text{O}_2$ mesh anode (Radjenovic, Bagastyo, et al., 2011). 10 L of sample were treated in a batch setup operated in recirculation mode with a flow rate of 162 mL min^{-1} . The electrochemical reactor consisted of a divided filter-press cell (114 mL-capacity) containing a DSA and a SS cathode both with 24 cm^2 of projected surface area. 25% of the dissolved organic carbon (DOC) was removed at 250 A m^{-2} and 437 A h m^{-3} , while 20 of the 28 organic pollutants were totally degraded. Spectrophotometric analyses revealed the formation of chloro-, bromo- and hydroxylated byproducts which were suspected to induce the high toxicity levels found after treatment, according to bioluminescence toxicity tests.

As illustrated in the previous example, electrochemical technologies have been applied to the treatment of RO/nanofiltration (NF) concentrates with significant advantages, since these effluents contain high concentrations of electrolytes, beneficial for electrochemical methods. Furthermore, the presence of salts, such as Cl^- , contributes to the oxidation capacity through the formation of oxidizing chlorine derivatives (mainly in the case of AO-DSA). However, DSA may entail the formation of chlorinated compounds, which is an important limitation, owing to their toxicity. This can be avoided when BDD is utilized due to its superior mineralization performance. For example, it was reported that the AO treatment of Cl^- -rich RO concentrates from municipal WWTPs

yielded a maximum of 22% of DOC removal with formation of halogenated intermediates when using a Ti/SnO₂-Sb anode (Bagastyo et al., 2013). In contrast, the treatment with BDD enhanced the DOC removal to 41% with negligible formation of halogenated substances. Moreover, the mineralization yield (DOC abatement) was improved to 51% when the Cl⁻ ions were previously removed by electrodialysis and substituted with SO₄²⁻.

Electro-Fenton

Because it makes use of inexpensive carbon cathodes, EF has been one of the most widely used EAOPs. However, one of its main limitations lies in its narrow operational pH range (optimal pH value of around 3), which restricts its large-scale application because of the need for pH adjustment before and after treatment. Thus, the number of reports of highly concentrated real wastewater treatment by AO surpasses that using EF. Research on EF has been mostly carried out on synthetic solutions containing moderate amounts of COD and TOC, with concentration ranging between 10 and 300 mg L⁻¹. As mentioned above, BDD anodes have sometimes been combined with EF, significantly enhancing the mineralization performance (cf. Table 1). However, since EF relies on the cathodic production of H₂O₂ and the homogenous Fenton's reaction, inert anodes, such as Pt, have also been used to a great extent (cf. Table 1). Following the same reasoning, despite their low mineralization capability, DSA electrodes arise as a very attractive option for EF, especially when only partial mineralization of refractory effluents is required (EF as part of a sequential treatment process). In this way, •OH production is ensured by the electro-Fenton reaction (Olvera-Vargas et al., under review; Sopaj et al., 2016), while the process benefits from the commercial advantages of DSA. In addition, if Cl⁻ ions are present in the target effluent, the performance can be improved by indirect chlorine oxidation (AO-Cl).

For example, landfill leachates were treated by means of EF using an undivided electrochemical reactor operated in batch mode (Oturan et al., 2015). Pt or BDD were used as anodes, while high surface area 3D carbon felt served as the cathode. The initial total organic carbon concentration of the leachates ranged from 1600 to 3100 mg L⁻¹, containing several recalcitrant organic pollutants, such as polycyclic aromatic hydrocarbons (PAHs), humic substances, volatile organic compounds (VOCs), organohalogenated compounds (OCHs), alkylphenols, pesticides and pharmaceuticals. 93.2% of TOC removal was achieved following 18 h of treatment by EF-BDD at 1 A of current, constant air flow at 1 L min⁻¹ and room temperature. The use of a Pt anode (EF-Pt) yielded 78.2% of TOC removal under the same experimental conditions. In both cases, all the identified organic contaminants were almost totally degraded during treatment. It is important to mention that the pH of the samples was adjusted to 3 prior to EF treatment. Moreover, due to the high amount of TSS, it was recommended to filter the sample before electrolysis, since suspended solids can clog the channels of electrochemical reactors and contribute to electrode passivation.

EF paired with BDD was recently used to treat real refractory pharmaceutical wastewater (COD = 1253.3, TOC = 431.5 and BOD₅/COD = 0.18) (Olvera-Vargas et al., under review). EF-BDD achieved 97.1% of TOC removal in 6 h of treatment at 4.17 mA cm⁻² and 0.2 mM of Fe²⁺, much higher than the 64.3% of TOC removal achieved when EF was combined with a DSA. However, EF-DSA proved equally efficient at increasing the biodegradability of the effluent with much lower operational costs (BOD₅/COD = 0.68 at US\$ 1.46 m⁻³ in 4 h vs. 1.68 US\$ m⁻³ for BOD₅/COD = 0.58 in 4 h with EF-BDD). These findings demonstrate not only the superb potential of EF to treat real pharmaceutical wastewater with a complex matrix, but also its versatility to reach different targets of water quality depending upon the anode materials: EF-DSA to

increase the biodegradability of refractory wastewater vs. more powerful (though costlier) EF-BDD for *quasi*-complete mineralization. Furthermore, this work presented an interesting account on the respective contribution of homogeneous $\bullet\text{OH}$ and heterogeneous BDD($\bullet\text{OH}$) generated during EF-BDD, whose synergistic effects accelerated the mineralization of organics, with homogeneous $\bullet\text{OH}$ dominating during the first treatment stages, while BDD prevailed in the last stages through electron transfer reactions.

In another recent work, waste streams from the electronics industry were treated by EF using a graphene-based GDE (Garcia-Rodriguez et al., 2018). The experiments were performed in an undivided electrolytic reactor using BDD as anode and 0.4 L of wastewater sample. Before trial, the effluent was conditioned by adding 0.05 M of K_2SO_4 and diluted H_2SO_4 to bring the pH to 3. Following 3 h of treatment, the TOC was depleted by 90% when the initial TOC concentration was 53.46 mg L^{-1} and by 60% when the initial TOC was 299.31 mg L^{-1} . The enhanced production of H_2O_2 (up to 495 mg L^{-1} accumulated in the solution in 3 h of electrolysis) promoted by the graphene-coated GDE maintained the continuous formation of $\bullet\text{OH}$ through the Fenton's reaction that took care of the mineralization of the organic constituents.

Electrocoagulation

For waste streams with high amounts of suspended solids and colloids, including oily waters, EC emerges as the most feasible option. EC is in fact a well-known electrochemical technology whose utilization can be traced back to the beginning of the 20th century (Mollah et al., 2001). Very comprehensive reviews on EC, its principles and fields of application are available in the literature (Garcia-Segura et al., 2017; Hakizimana et al., 2017; Moussa et al., 2017). EC is able to remove organic pollutants, heavy metals and other inorganic species (sulfate, phosphate, chloride and nitrate, among others) by

adsorption onto the electrochemically-produced coagulants followed by settling or flotation. Moreover, colloidal suspensions are easily destabilized and coagulated afterwards. Non-toxic and inexpensive iron and aluminum electrodes are generally used. In fact, EC has become a powerful and effective wastewater treatment technology widely used even for large-scale applications. For example, F&T water solutions³ manufactures EC systems with capacities between 10 to 1000 GPM. Powell Water⁴, WaterTectonics⁵, Elgressy Engineering Services LTD⁶. and Hydroleap⁷ are other companies that commercialize EC technologies. Their main drawbacks lie in the production of secondary iron/aluminum sludge that requires further disposal and the important consumption of electrical energy. Besides, as EC is primarily a separation technology, it does not provide high efficiencies in the removal of dissolved organic carbon. For this reason, EC has been coupled to other oxidative technologies when high TOC removal is the goal (Thiam et al., 2014). In this respect, EC is a perfect candidate for integrated systems either as a conditioning (reduction of solids and the organic charge) or polishing (elimination of remaining inorganic species) step (Bocos et al., 2016). Combined processes will be covered in depth in a dedicated section of this review.

As an example of EC as stand-alone technology, the treatment of 3.5 L of real textile wastewater with a high COD concentration of 2000 mg L⁻¹ and 230 mg L⁻¹ of

³ F& T Water Solutions | Electrocoagulation for Industrial Wastewater Treatment. <http://www.ftwatersolutions.com/> (accessed April 27, 2018).

⁴ Powell Water - Powell Electrocoagulation Waste Water Treatment. <http://powellwater.com/> (accessed April 27, 2018).

⁵ WaterTectonics - Advanced Water Treatment Solutions. <http://www.watertectonics.com/> (accessed April 27, 2018).

⁶ Elgressy Engineering Services Ltd. <http://www.elgressy.com/> (accessed April 27, 2018).

⁷ hydroleap. <https://www.hydroleap.com/> (accessed April 27, 2018).

suspended solids was conducted by Kobya et al. (Kobya et al., 2016). The experiments were conducted in a continuous flow reactor, in which the electrochemical cell was equipped with two pairs of electrodes connected in monopolar parallel configuration with a space of 20 mm between them. When Fe electrodes were used, 85% and 76% of COD and TOC removal, respectively, were achieved under optimal conditions: inlet flow rate of 0.01 L min⁻¹, 80 min of treatment and a current density of 65 A m². With Al electrodes, COD and TOC removal efficiencies of 77% and 72%, respectively, were attained. The final COD and TOC values after treatment were lower than the permissible limit of discharge into the sewage, according to the Turkish legislation.

In a different study conducted at pilot scale (working volume of 53 m³ day⁻¹) in a Korean municipal wastewater treatment plant, a 200-L EC reactor with Al cylindrical electrodes placed concentrically with a gap of 2.25 cm was operated at 10 V and a hydraulic retention time of 2 min (Nguyen et al., 2014). The treatment system was capable of removing total phosphorus (mainly PO₄³⁻) by 97.21-100% and COD by 95.38%, while NH₄⁺ was totally removed. Moreover, TSS were reduced to less than 5 mg L⁻¹ and coliform bacteria to 30 MPN mL⁻¹.

In conclusion of this section, we would like to highlight that works dealing with electrochemical processes as individual systems are mostly focused on the removal of organic contaminants and they do not usually address the fate of inorganic species, including supporting electrolytes and other ionic compounds formed during treatment.

Scale up and system integration: state of the art and perspectives

Much of the vast experimental work reported with respect to EAOPs has been conducted at laboratory scale with volumes lower than 1 L. Great progress has been made in the optimization of such processes in these conditions, improving the electrode materials as well as the operational parameters (applied current density, flow rates, electrolysis time,

etc.). In this final section, we wish to bring attention to the recent efforts in bringing these technologies to practical wastewater treatment applications, an indication of the new state of maturity that they have reached in the recent years. The approach is two folds: (i) need for scale-up through a combined modeling and experimental approach and; (ii) system integration with traditional treatments, as part of an overall wastewater treatment system. In the previous section, we have reported a methodology to select an overall strategy for electrochemical treatment, in relation with the characteristics of the wastewater to be treated (Figure 2). Therefore, the engineering parameters of the reactor must be tailored according to the type of EAOP and the effluent. Additionally, there are certain general considerations to take into account when scaling up an electrochemical process. In this section, the parameters of utmost importance for scaling up an electrochemical reactor are addressed in a general form. In order to assess the feasibility of the wastewater treatment process, the first step always consists of a laboratory test, followed by pilot scale runs and ending with an industrial or commercial application. Usually, modular reactors composed of several electrode pairs are the most common approaches in electrochemical industrial processes. Therefore, the most critical stage is the transition from laboratory to pilot scale, where one or two pairs of electrodes, can be used as cell stack in the pilot plant. This allows estimating the efficiency as well as the number of cell stack modules needed for commercial applications.

Modeling of EAOPs

Modeling is a powerful tool for scaling up electrochemical processes. Modeling relies on mathematic equations, in order to predict parameters, such as the chemical oxygen demand (COD) evolution, mass-transport coefficients, intermediate profile concentrations, etc. under an extensive number of operating conditions. Modeling is useful to reduce test trials and operating costs; moreover, the use of computational fluid

dynamics (CFD) has been very helpful to study the hydrodynamic behavior of electrochemical reactors, which allows improving their design. In spite of the wide range of modeling classification, most of the reviewed models presented next, are semi-empirical models, meaning they are located in between the phenomenological and mechanistic models (Zhen et al., 2014). Since every electrochemical treatment process is unique, different approaches have been made in an effort to have a better understanding of each specific process.

Anodic oxidation modeling

Comninellis and collaborators were the pioneers in the development of mathematical models for AO (Gherardini et al., 2001; Panizza et al., 2001; Rodrigo et al., 2001; Simond et al., 1997). Their model, based on the relation between the limiting current (j_{lim}) and COD (Eq. 1), considers that the degradation of the pollutant is carried out only at the anode surface.

$$j_{lim}(t) = nFk_m COD(t) \quad (1)$$

where $j_{lim}(t)$ refers to the limiting current density ($A\ m^{-2}$) at time t , F is the Faraday's constant ($C\ mol^{-1}$), n is the number of exchanged electrons, k_m states for the mass transfer coefficient ($m\ s^{-1}$) in the electrochemical cell and COD is the chemical oxygen demand of the electrolyte at time t .

This model considers two possible working regimens: i) when the applied current density (j_{appl}) is below the limiting current density, the system is under current control and; ii) on the contrary, when the applied current density exceeds the limiting current density, the system becomes controlled by mass transfers. COD evolution can then be described by Eqs 2 and 3, respectively.

$$COD(t) = COD^0 \left(1 - \frac{\alpha A k_m}{V_R} t \right) \quad (2)$$

$$COD(t) = \alpha COD^0 \exp\left(-\frac{Ak_m}{V_R} t + \frac{I-\alpha}{\alpha}\right) \quad (3)$$

Where COD^0 is the initial COD (mol O_2 m^{-3}), α is the ratio of applied current density and the initial limiting current density (j_{appl}/j_{lim}^0), A corresponds to the electrode area (m^2) and V_R the reactor volume (m^3).

Although, this model has been applied successfully in many studies (Aquino et al., 2014; Barbosa et al., 2016; Steter et al., 2014) and can satisfactorily predict the experimental data, its application is limited to direct oxidation processes at the electrode. Other approaches of mathematic models for anodic oxidation include that of Cañizares et al. (Cañizares et al., 2004), Mascia et al. (Mascia et al., 2010) and Lan et al. (Lan et al., 2018), among others. One of the most interesting studies with respect to this review consists of the works by Anglada et al (Anglada et al., 2009; Anglada et al., 2011; Anglada, Urtiaga, et al., 2010), dealing with modeling of real highly concentrated wastewater in pilot-scale reactors. They have demonstrated that the rate of COD removal is directly proportional to the ratio of the anode area over treated wastewater volume. In this way, they used kinetic modeling for COD elimination to calculate the required total anode area for scale up (Anglada, Urtiaga, et al., 2010).

Electro-Fenton modeling

Unlike AO, which mostly takes place at the anode surface, EF modeling is more complex because it involves a series of reactions that take place concomitantly at the cathode surface and in the bulk solution. The main reactions include the generation of H_2O_2 through the reduction of oxygen via $2e^-$ (Eq. 4), the $\cdot OH$ radical generation (Eq. 5) and the organics degradation (Eq. 6). Furthermore, there are several side reactions that coexist with the main process, e.g. Eqs 7-10, among others.

414 In spite of the complexity to predict organic pollutant abatement by EF, Liu et
 415 al. (Liu et al., 2007) presented the first kinetic model using phenol as model organic
 416 pollutant. Their model used the rate constants presented in Eqs 4 to 10 and is based on
 417 the assumption that the organic substance (S) is mainly degraded by $\cdot OH$ radicals, with
 418 a reaction rate described by Eq. 11.

$$419 \quad -\frac{d(S)}{dt} = k_3[\cdot OH][S] \quad (11)$$

420 Using Eq. 11 as starting point, they presented a pair of equations to describe
 421 H_2O_2 accumulation (Eq. 12) and the organic substance degradation (Eq. 13), deduction
 422 described elsewhere (Liu et al., 2007)).

$$423 \quad [H_2O_2] = j_{appl} \frac{k_1 K_{ad}[O_2]}{k_2 \lambda [Fe^{2+}]_0 (1 + K_{ad}[O_2])} \left(1 - e^{-k_2 \lambda [Fe^{2+}]_0 t}\right) \quad (12)$$

424 Where j_{appl} is the applied current density, K_{ad} corresponds to the adsorption
 425 equilibrium constant of dissolved oxygen on the cathode surface and λ is a fixed ratio of
 426 $[Fe^{2+}]_0/[Fe^{2+}]$.

$$427 \quad Ln \frac{[S_0]}{[S]} + a([S_0] - [S]) = b \left(t - \frac{1 - e^{-ct}}{c}\right) \quad (13)$$

428 Where $a = k_3/k_4 \lambda [Fe^{2+}]_0$, $b = j_{appl} k_1 k_3 K_{ad}[O_2]/k_4 \lambda [Fe^{2+}]_0 (1 +$
 429 $K_{ad}[O_2])$ and $c = k_2 \lambda [Fe^{2+}]_0$.

The validation of this model was carried out through the variation of three parameters: initial concentration of iron $[\text{Fe}^{2+}]_0$, dissolved oxygen concentration $[\text{O}_2]$ and applied current density (j_{appl}). The model demonstrated a good fitting with experimental data, which indicates its suitability to describe the kinetics of EF reaction, not only mathematically but chemically too.

Despite the good first approach of Liu's model for EF, it does not provide detailed information about byproduct evolution during the degradation process, which is of paramount importance when those intermediates are more toxic than the original pollutant. To overcome this drawback, Mousset et al. (Emmanuel Mousset et al., 2016) developed a more comprehensive model, using MATLAB[®] to perform the simulations. This model considered five representative steps: (i) in situ electrogeneration of H_2O_2 ; (ii) Fenton's reaction in bulk solution; (iii) ferrous ion electroregeneration; (iv) scavenging reactions and; (v) degradation and mineralization of phenol. All these parameters had an impact on the kinetics of the process; however, step four, corresponding to the scavenging reaction between Fe^{2+} and $\cdot\text{OH}$ (Eq. 7), was the most important, followed by step 1 (Eq. 4), characteristic of this electrochemical process. It is worth highlighting that this model was able to predict the profile concentrations of ten compounds and was well corroborated by experimental data.

Recently, Mousset et al. (Mousset et al., 2019) proved for the first time through a mathematical model, that COD concentration has a direct impact on the predominant oxidation process in an electrochemical reactor combining EF and AO, showing that low COD concentrations lead to a process controlled by mass transfer where EF has the highest contribution, while AO shows reduced efficiency under these conditions. In contrast, the treatment of an effluent with a high initial COD load will initially be dominated by AO before reaching a point where EF then controls the overall process.

Mousset et al. (Mousset et al., 2019) proposed a mathematical equation for a global instantaneous current efficiency (ICE_{global}) in a reactor combining EF and AO-BDD, taking into consideration the electro-oxidation at the surface of the anode, the EF process at the cathode and the indirect mediated oxidation (MO) caused by reactions at the anode (Eq. 14).

$$ICE_{global} = \frac{4FV}{I} * \frac{d[COD]}{dt} = ICE_{anode} + ICE_{cathode} + ICE_{MO} \quad (14)$$

Where F is the Faraday's constant, V the volume of the treated solution and I corresponds to the applied current.

In this way, their research highlighted the importance of the reactor design to favor either one or the other electrochemical process depending on the initial organic load of the wastewater. Besides the presented approaches, other research groups have also tried to optimize the EF process through the use of modeling (Alvarez-Gallegos & Silva-Martínez, 2018; Khataee et al., 2014; Qiu et al., 2015). However, to our best knowledge, none of these models is yet validated at large scale.

Electrocoagulation modeling

One of the first proposed models for EC was done by Mameri et al. (Mameri et al., 1998), who carried out the defluorination of water using bipolar Al electrodes and found a relation between the electrode area divided by the working volume ($A \text{ V}^{-1}$) and the treatment time. Furthermore, they evidenced the existence of an optimal current density for a fixed $A \text{ V}^{-1}$ relation. Through the use of an equation based on the Faraday's relation (Eq. 15), they predicted the necessary treatment time (t), well corroborated by experimental data.

$$t = \frac{mnF}{M_{Al}A_0N} \quad (15)$$

Where m is the theoretical Al mass needed for the elimination of the fluoride ions (kg), n the Al valence, F corresponds to the Faraday's constant (C mol^{-1}), M_{Al} corresponds to the mass of Al (kg), A_0 is the electrode area (m^2) and N the cells number.

Another approach of EC modeling is through the adsorption phenomenon, where the electrogenerated metallic hydroxides are considered to be the adsorbent. In this sense, Carmona et al. (Carmona et al., 2006) showed that the experimental data for the abatement of oil from water using Al electrodes fitted well a Langmuir-type empirical equation, describing the efficiency of the treatment based on the applied current density and pollutant concentration.

Another interesting model that takes into account mass transfers during electrolysis was proposed by Lu et al. (Lu et al., 2017). They found that the electrochemical cell experienced different compound concentrations along the electrode gap, forming three regions: i) an acid front (dominated by H^+); ii) a base front (dominated by HO^-) and; iii) a buffering area where amorphous $\text{Al}(\text{OH})_{3(\text{s})}$ is formed.

There is a considerable amount of proposed EC models in the literature (Cañizares et al., 2008; Graça et al., 2019; Safonyk & Prysiazniuk, 2019; Xiang et al., 2018; Yehya et al., 2014), with different approaches depending upon the several mechanisms that can take place in complex processes, including electrochemical phenomena (e.g. charge transport, electrodes interface, electrochemical kinetics, etc.), adsorption, flocculation, flotation, settling and complexation (Hakizimana et al., 2017). These mechanisms depend mainly upon the electrodes material and effluent characteristics. Although Fe and Al represent the gold-standard electrodes for EC, the vast variety of industrial effluents has prevented a general systematic approach so far. This emphasizes the need to find out the most accurate mechanisms, according to the type of wastewater in order to achieve a model with good predictive value.

Computational fluid dynamics in electrochemical reactors

In the recent years, the use of CFD to study electrochemical reactors has gained popularity. The development of softwares such as ANSYS Fluent[®], COMSOL Multiphysics[®], OpenFOAM[®], among others, has allowed a better understanding of the hydrodynamics of the system, allowing the prediction of flow patterns, velocity profiles, concentration profiles, reaction rate distribution at the electrodes, etc. Besides the acquisition of numerical data, the visual representation of a wide variety of parameters facilitates the understanding of the system. In the last decade, different research groups have made use of this type of software to improve and understand systems such as AO, EC and EF, for example, Figure 3a shows the mixing comparison of a vertical-flow tubular and a concentric tubular electrochemical reactors, as can be seen the turbulent intensity of wastewater was higher around the electrodes in the former reactor, which translates as a better mass transfer of the active substances towards the electrodes surface (Wang et al., 2015), Figure 3b illustrates the current distribution in the electrodes of an EC reactor (Villalobos-Lara et al., 2020) and Figure 3c shows the generation of Fe^{2+} that takes place in the cathode due to the reduction of Fe^{3+} after different time intervals during the EF process (Reza et al., 2015).

CFD is helpful in improving the reactor design by predicting the efficiency of the process. For example, Martínez-Delgadillo et al. (Martínez-Delgadillo et al., 2010) showed the importance of the electrochemical reactor inlet and how it affects the performance of Cr(VI) removal. In this way, the estimation of the fluid behavior can be done beforehand to correct and evaluate the reactor operation. Another remarkable study was conducted by Choudhary et al. (Choudhary & Mathur, 2017), using a rotating 3D anode in an EC reactor. They showed the effect of rotation speed on mass transfers and also evidenced the influence of the hydrodynamic regime on the efficiency of the process.

In this way, they were able to calculate the optimal flow rate and rotation speed using CFD simulation.

As seen above, modeling through mathematical equations and CFD are important tools that can improve our understanding of electrochemical wastewater treatment processes. The integration of both, which is rarely seen, would considerably facilitate the scale-up process of electrochemical reactors for wastewater treatment.

Reactor design and scale up studies

Typical electrochemical cell designs include 2D and/or 3D electrodes. Within 2D electrodes, the most common geometry consists of planar electrodes usually in the form of parallel plates or concentric cylinders. On the other hand, 3D electrodes comprise porous materials and fluidized bed systems, having the advantage of a high electrode area x volume⁻¹ ratio. Meanwhile, 2D materials have the benefit of a more homogeneous electrode potential, which is reflected in a good reaction rate distribution over the electrode surface, making them the most popular choice for large-scale reactor design.

The design of the reactor usually includes two types of electrode arrangement, monopolar and bipolar. Monopolar refers to a situation where the electrodes are polarized either negatively or positively, usually accomplished by connecting all of them to the negative or positive terminal of a power supply. On the other hand, a bipolar arrangement comprises electrodes without any electrical connection placed in between monopolar electrodes, where each face of these bipolar electrodes will have the opposite charge of the parallel electrode besides it. In this way they may adopt a negative and positive charge at the same time. Despite being a common studied parameter especially in EC processes, there is always the need of evaluating the electrode arrangement for each particular system, since there is not a unique criterion that can be applied in general. In one hand, there are studies concluding that a bipolar system has a better efficiency, e.g. during the

degradation of alizarin blue black B, a bipolar AO system was 1.2 fold more efficient than a monopolar AO system, furthermore, the former one didn't accumulate large amounts of intermediates unlike the monopolar system (Abdessamad et al., 2013), a higher efficiency with a bipolar configuration was also observed by (Thiam et al., 2014) during EC process and was attributed to a higher consumption of the electrode which resulted in a greater coagulation ability and faster pollutant removal. In the other hand, (Nidheesh et al., 2020) found that a monopolar connection was more efficient for the removal of COD and color from wastewater, being able to remove up to 55% and 56%, respectively, whereas a bipolar configuration removed 43% of COD and 48% of color. Moreover, (Kahraman & Şimşek, 2020) despite finding a slightly more efficient pollutant removal with a bipolar EC system, the higher cost due to the potential increase (up to 18.3 V) made the monopolar system more feasible where the potential was less than 5V. The increase in potential with a bipolar configuration, could explain why in EF processes is preferred a monopolar configuration (Sultana et al., 2018) since high potentials would lead to parasitic reactions such as H₂ evolution and O₂ reduction via 4 electrons.

Despite the great advances in the understanding of EAOPs, there is a lack of systematic approach to carry out the reactor design, which is reflected in their great variety; however, many studies agree that the mass transfer and the area of the electrode per unit of volume should be as high as possible, resulting in a compact reactor. Over the past decades a wide range of reactor designs have thus been developed, including filter presses (Figure 4a), fluidized beds (Figure 4b), rotating electrodes (Figure 4c) and fixed beds (Figure 4d). Such designs can be used for any of the processes mentioned earlier. The main differences between reactors for AO, EF or EC lie in the electrode materials and in the operating conditions, with minor modifications to the reactor design.

Most EAOP studies have been performed in electrochemical cells with volumes around 100 mL. Even though the number of studies making use of actual industrial waste streams, usually with high pollutant concentrations, is increasing (Garcia-Segura et al., 2018), there is still a lack of information regarding their treatment in larger volumes. To the best of our knowledge, the applications of EAOPs for the treatment of highly concentrated waste streams at volumes above 1 L (and up to 1 m³) are compiled in Table 2. When the scale up process is carried out, many factors need to be considered for the design of such reactors, including the final effluent target quality, treatment capacity and time. However, as mentioned by Anglada et al. (Anglada, Urtiaga, et al., 2010), the A V⁻¹ ratio also needs to be taken into account and thus this ratio is also presented in Table 2.

From Table 2, the average electrode area needed for the treatment of one cubic meter of wastewater was 1.9 m², 6.6 m² and 9.2 m² for EF/SPEF (combined or alone), AO and EC, respectively. It thus appears that processes involving the Fenton's reagent need a lower electrode area; however, most of the effluents tested were less concentrated, which has consequences in terms of a higher energy consumption per mass unit of pollutant. On the other hand, EC requires a higher electrode area and the electrodes lifespan is shorter than for the other 2 processes. Finally, AO appears to be a versatile process, with electrode area as low as 0.71 m² per m³ of treated wastewater; however, the cost of the electrodes is the highest among the three families of electrochemical processes. Although the number of wastewater treatment electrochemical studies at pilot or industrial scale is increasing, there is still a critical need for real field and long-term studies, where the constraints related to their operation and application will be identified and solved.

Energy requirements

AO, EC and EF, are mainly driven by electricity, which makes energy consumption the prime expense for these electrochemical processes. Energy consumption mostly arises from pumping, aeration and applied current to carry out the electrolysis, all of these parameters being directly related to the operational costs. Other operational costs, including electrode replacement, sludge management, chemicals cost, maintenance, among others, are not covered in this review but can be found elsewhere (Cañizares et al., 2009; Garcia-Segura et al., 2017; Ibarra-Taquez et al., 2017; Liu et al., 2017; E. Mousset et al., 2016).

The energy consumption expressed in kWh per kg of pollutant in the above-mentioned studies (Table 2), was evaluated using Eq. 16 (García-Rodríguez et al., 2016).

$$\text{Energy Consumption (kWh kg}^{-1} \text{ pollutant)} = \frac{E_{cell} I t * 1000}{V_s \Delta(\text{pollutant})_{exp}} \quad (16)$$

Where E_{cell} is the average potential difference (V) of the electrochemical reactor, I corresponds to the applied current (A), t is the time of electrolysis (h), V_s is the treated volume (L), $\Delta(\text{pollutant})_{exp}$ corresponds to the pollutant decay (mg L^{-1}), and a conversion factor of $1000 \text{ Wh mg}^{-1} = 1 \text{ kWh kg}^{-1}$ is applied.

From Table 2, it can be seen that in most cases, the energy consumption is inversely proportional to the concentration of contaminants. One of the main explanations could be the better mass transfer towards the electrodes, as evidenced in other studies (Garcia-Rodriguez et al., 2018). This is important, because the volumes generated for highly polluted wastewater are typically smaller than for wastewater with low concentrations, where dilution creates larger amounts of wastewater. This would render the treatment of highly concentrated wastewater typically more cost-effective.

Integration of electrochemical methods in multi-stage sequential strategies

Due to the complex composition of real and highly contaminated waste streams, the application of a single universal treatment method is generally not feasible. In most cases, proper treatment can only be achieved through a series of sequential steps, depending upon the treatment strategy, the type of contaminants and the final water quality objective (Figure 2). When combining different treatment methods in a sequential integrated process, the main advantages of the individual processes can be refined under optimal conditions, while the other processes forming part of the overall treatment train can overcome the individual drawbacks inherent to each method. In this scenario, the integration of electrochemical technologies with other methods shows potential to become an excellent solution for the treatment of numerous waste streams with different characteristics. Coupled systems are designed not only to attain the treatment objectives, but also to reduce the treatment costs, through the conception of a technologically and economically feasible strategy. From this perspective, it is clear that when conceptualizing coupled systems, conventional methods will always be the first option to combine with advanced technologies. The outstanding oxidizing capabilities of EAOPs can then be exploited for example to degrade refractory and toxic organics that resist biological degradation or to prevent membrane fouling (Ganiyu et al., 2015; Ganzenko et al., 2014). Table 3 compiles a number of studies dealing with the coupling of electrochemical technologies with biological processes, membrane technology, other AOPs and flocculation/coagulation, among others.

The combination of electrochemical technologies with biological methods is especially promising because biological methods are by far the most utilized form of wastewater treatment at the industrial scale, due to their economic and technological assets. However, when refractory effluents cannot be treated by microbiological attack, the use of highly oxidative technologies becomes one the most convenient options (Oller

et al., 2011). EAOPs can turn refractory organics into biodegradable compounds over short treatment times, that can then be more easily handled biologically, while reduced electrochemical treatment times represent significant savings. The opposite treatment strategy can also be applied, where the biodegradable fraction of the effluent can first be treated by biological means, while the remaining refractory pollutants can be degraded by EAOPs as a subsequent step, as long as the refractory compounds are not toxic to the biomass. This option can also be advantageous when the effluent displays high initial organic load (Kishimoto et al., 2017). Recent reviews on this topic with more detailed information can be found in the literature (Ganzenko et al., 2014; Olvera-Vargas et al., 2017).

Membrane technology has also become a prime actor of the wastewater industry. However, membrane fouling caused by chemical agents is a major drawback of membranes, alongside the need to dispose the concentrated brine rejects. In this respect, the integration of membranes with electrochemical processes is very promising, where chemicals responsible for fouling can be first degraded by EAOPs prior to membrane filtration or EAOPs can be applied as a post-treatment for the concentrates. A third alternative that has been explored is the use of electrocatalytic membranes that function as both separation and oxidation units with self-fouling control properties; yet, these systems are still in the early stages of development (Yang et al., 2012). More extensive information with regard to membrane/EAOP systems can be found in a review paper by Ganiyu et al. (Ganiyu et al., 2015). Some relevant sequential strategies that have been used in the treatment of highly loaded hard-to-treat waste streams are described below.

The acidic and alkaline effluents from a cellulose bleaching industry (details of the sample composition can be found in Table 3) were treated by a sequential filtration-AO process (Salazar et al., 2015). The AO reactor consisted of an undivided cylindrical

glass cell with either DSA-RuO₂ or BDD anodes and a carbon-PTFE GDE fed with compressed air at 1 L min⁻¹. Filtration units made use of flat ultrafiltration, NF and RO membranes at 20 °C. AO without previous filtration (BDD anode, 12 V of potential, 800 rpm of stirring and 25 °C) only removed 65% of the initial TOC of the alkaline effluent, while NF removed TOC up to 72%. Yet, when combined, NF followed by AO resulted in an overall 96% of TOC removal in the alkaline effluent, following 9 h of AO treatment of the NF permeate. In this work, the suspended solids were first removed by microfiltration.

Anaerobically-digested food processing wastewater with high concentration of organic matter and NH₄⁺ (COD = 3200-3400 mg L⁻¹, NH₄⁺ = 3352-3790 mg L⁻¹ and TSS = 15580 mg L⁻¹) was treated by a sequential system consisting of electrochemical peroxidation, (ECP, consisting of EC with iron anodes and external addition of H₂O₂), followed by AO (Shin et al., 2017). ECP was conducted in a 1-L undivided reactor with 2 pairs of SS electrodes positioned in parallel. The AO reactor also consisted of an undivided cell with a capacity of 0.3 L, a DSA and a SS cathode. Under optimal conditions, the integrated ECP-AO process achieved over 90% of TOC removal and almost complete NH₄⁺ removal, following 150 min of ECP (pH 3, 28.5 mA cm⁻², H₂O₂ feeding at 1.67 mmol min⁻¹ and 1 M NaCl as supporting electrolyte) and 150 min of AO (pH 5, and 200 mA cm⁻²). During the ECP step more than 80% of the TOC was removed, while NH₄⁺ was reduced by less than 25%. ECP benefits from a highly oxidative environment, where the addition of H₂O₂ and the dissolution of the iron anode promote the production of [•]OH through the Fenton's reaction. The remaining TOC and NH₄⁺ after ECP were removed by AO, mainly by intermediacy of chlorine species generated in the bulk solution. Before treatment, the wastewater sample was centrifuged at 3000 rpm for 20 min to remove the solids.

Anaerobically digested sludge generated in a poultry farm ($\text{COD} = 26,200 \text{ mg L}^{-1}$, $\text{TOC} = 5,235 \text{ mg L}^{-1}$ and $\text{TSS} = 31,300 \text{ mg L}^{-1}$) was successfully treated by means of an integrated process consisting of ECP with Fe electrodes followed by EF with a carbon brush cathode and a BDD anode (Olvera-Vargas et al., 2019). The synergistic effects of electrocoagulation and advanced oxidation during ECP (initial pH of 5, $[\text{H}_2\text{O}_2]/[\text{Fe}^{2+}]$ ratio of 5, 15.38 mA cm^{-2} and 2 h of treatment) achieved 89.3% and 85.6% of COD and TSS removal, respectively, increasing sludge dewaterability and reducing the amount of disposable solid sludge in the process. The wastewater treated by ECP was then subjected to further EF treatment, once the solid sludge was removed from the effluent (such residue was free of coliforms and rich in nutrients with fertilizing potential). EF achieved almost total mineralization of the remaining organic matter under optimal conditions (pH 3 and 25 mA cm^{-2}), leaving only 24.6 mg L^{-1} and 16.3 mg L^{-1} of COD and TOC, respectively, after 8 h of treatment. This two-step electrochemical process proved to be a powerful strategy to deal with highly polluted and viscous sludge.

In another sequential treatment, (diluted) pharmaceutical wastewater was pre-treated by EF followed by activated sludge (AS) (Mansour et al., 2015). The AS treatment was conducted in 500-mL reactors containing 200 mL of the effluent and 1 g L^{-1} of biomass from a municipal wastewater treatment plant, at neutral pH and with different organic nutrients. For EF, 1-L samples were treated in an undivided electrolytic cell containing a Pt anode and a carbon-felt cathode under constant aeration and stirring. The optimal conditions determined for EF were 0.69 mM of Fe^{2+} , 466 mA of current and pH 3. Following 180 and 300 min of EF treatment, the BOD_5/COD ratio (biodegradability index) increased from 0.40 to 0.45 and 0.47, respectively, while the TOC was only reduced by 14 and 18%, respectively. After 15 days of AS treatment, the overall TOC removal was 80% and 89% for following 180 min and 300 min of electrolysis,

respectively. These results demonstrate the potential of EF as pre-treatment to increase the biodegradability of refractory waste streams, a strategy that deserves more exploration (Olvera-Vargas et al., 2017).

A large-scale sequential system including several steps was reported by Moreira et al. (Moreira, Soler, et al., 2015), in which the remediation of sanitary landfill leachate was conducted by a sequence of AS, coagulation and PEF/SPED. In this case, the aim of the electrochemical treatment was to increase the biodegradability of the pre-treated effluent as a conditioning step prior to biological polishing. The AS process was performed in a 12-L bioreactor with a conical bottom where 8 L of landfill leachate were treated using AS from a municipal WWTP. The pH was maintained between 6.5 and 9. After sedimentation, the supernatant was transferred to a coagulation tank where FeCl_3 was added at pH 3 and stirred for 15 min. 48 h of sedimentation were required, after which the effluent was sent to the electrochemical treatment. The 35-L electrochemical flow-through system comprised an electrochemical filter-press cell with a BDD anode and a carbon-PTFE GDE cathode, a 20-L recirculation tank and a photoreactor. The AS treatment achieved 13-33% of DOC removal, as well as total oxidation of NH_4^+ and complete alkalinity removal. With the coagulation process, 63% of DOC abatement was attained. During the SPEF process, 53% of the remaining DOC was removed at a current density of 200 mA cm^{-2} , 60 mg L^{-1} of Fe^{2+} , pH of 2.8 and 20°C (after SPEF, the effluent was neutralized to pH 7.5 and submitted to 3 h of clarification to remove the remaining sludge). Overall, 87% of DOC removal was achieved at the end of the multi-stage treatment. The resulting COD, BOD_5 and total nitrogen values were still above the Portuguese and European regulations for discharge. However, Zhan-Wellens biodegradability tests indicated that the sample was suitable for further biological treatment to comply with the discharge limits.

In summary, electrochemical technologies have demonstrated their extraordinary capacity to treat a great diversity of waste streams containing high loads of organic pollutants and other types of chemicals. Their outstanding oxidative power has made them top choices to integrate as part of sequential treatment systems. Such systems are undoubtedly the most feasible way to clean wastewater either for discharge or for reuse. Electrochemical technologies are currently reaching maturity for industrial-scale applications. The design and construction of electrochemical reactors and pilot plants (tightly linked to the development of electrode materials) is crucial for the success of electrochemical processes in the years to come. Besides, fundamental research remains essential to further position electrochemical methods as a valid technology for treating highly contaminated waste streams.

Applications to highly concentrated wastewater: nature, characteristics and challenges

Many industrial sectors and other human activities result in the production of high quantities of very highly loaded effluents and waste streams. The type of contamination may vary but the challenge always lies in the lack of a gold standard for the effective treatment of such kinds of wastewater. Electrochemistry may occupy a niche there because a lot of the characteristics of highly concentrated waste streams make them ideal candidates for electrochemical treatment. For instance, the volumes generated are usually low, giving an edge to electrochemical approaches, that are generally well adapted to small decentralized treatment systems. Furthermore, the high concentration of salts and organics may act like a natural electrolyte and may provide catalysts and reactive species for electrochemical reactions, thus lowering the treatment costs as compared to the treatment of diluted effluents. Yet, specific strategies need to be implemented and there

are in fact very few instances of pilot or full-scale demonstration of electrochemical treatment for such kind of wastewater.

Highly concentrated solutions need particular attention and their physical and chemical characteristics determine the type of treatment to be applied, based on pH, electrical conductivity, chemical oxygen demand (COD), total organic carbon (TOC), biodegradability – that can be assessed with the biochemical oxygen demand after 5 days (BOD₅) over COD ratio – total inorganic carbon (TIC), total suspended solids (TSS), total dissolved solids (TDS), main inorganic ions (Cl⁻, SO₄²⁻, NH₃/NH₄⁺, NO₂⁻, NO₃⁻, PO₄³⁻) and heavy metals analyzes. In the last two decades, electrochemical processes have attracted a lot of interest for their high versatility, i.e. their possibility to treat solutions containing low or high concentrations of organics and/or inorganics. In this section, five categories of wastewater are investigated, alongside their properties and influence on the type of electrochemical processes to be applied, with emphasis on electrochemical advanced oxidation processes (EAOPs), in combination or not with electrocoagulation (EC). They consist of (i) high COD effluents of industrial origin; (ii) hypersaline effluents; (iii) solutions contaminated with a mixture of organic and inorganic contaminants; (iv) effluents with high rheological complexity and; (v) solutions with high COD load but low pollutant content.

High COD effluents of industrial origin

Industrial waste streams are often characterized by high COD content and need a specific treatment. The main industry sectors that have attracted attention due to their recalcitrance to biological processes comprise textile, petroleum, tannery, chemical, pharmaceutical and paper mill activities. Electrochemical processes and in particular EAOPs have attracted a lot of interest due to their strong ability to degrade and mineralize heavily loaded effluents containing xenobiotic contaminants, such as synthetic dyes, phenolic

compounds, aliphatic hydrocarbons, aromatic hydrocarbons, pesticides, lignosulfonate compounds, antibiotics, anti-inflammatory drugs, stimulants, analgesics, etc. (Garcia-Segura et al., 2018). Selected studies are listed in Table 4. All these effluents have in common a high COD content varying from $\sim 1.0 \text{ g-O}_2 \text{ L}^{-1}$ to more than hundreds $\text{g-O}_2 \text{ L}^{-1}$ (Chanworrawoot, 2012; Valero et al., 2014). Electrochemical processes have the capability to mineralize those effluents until quasi-complete mineralization, especially with EAOPs (anodic oxidation, AO, with boron-doped diamond anode (BDD) and electro-Fenton, EF) that make use of very strong oxidizing agents like hydroxyl radicals ($\cdot\text{OH}$). The limiting factor is the time required to reach high levels of organic removal that can considerably increase the electrical energy consumption. The amount of organic by-products generated is also substantial due to the high initial quantity of carbon content. Some of these degradation intermediates can potentially be more toxic than the parent compounds, so that ecotoxicity studies have to be performed to verify the evolution of toxicity with electrolysis time (Gargouri et al., 2014).

Hypersaline effluents

Besides industrial wastewater, waste streams containing high amount of salts are typically found in reverse osmosis (RO) concentrates. These hypersaline brines constitute a major drawback of such membranes and often cannot be discharged into surface water or by deep well injection, due to increasingly stringent environmental restrictions (Van Hege et al., 2002). The advantage of using electrochemical technology is that the high salinity – with conductivity ranging from 2 to 22 mS cm^{-1} – ensures great conduction of current, which reduces the energy requirements (Table 5). In addition, the high chloride ion (Cl^-) content ($0.3\text{-}1.4 \text{ g L}^{-1}$; Table 5) can generate hypochlorous acid (HClO), which is an oxidizing agent able to degrade organic compounds (Eqs. 17-18) (Perez-Gonzalez et al.,

824 2012):

827 It is important to highlight that the formation of halogenated by-products, such as
828 trihalomethane (THM), could pose a health risk (Hurwitz et al., 2014). Though the initial
829 COD of RO concentrate is quite low (14-190 mg-O₂ L⁻¹, Table 5), organic intermediates
830 are still formed (Lütke Eversloh et al., 2014). Because maintaining a short electrolysis
831 time is usually preferred for minimizing electrical energy consumption, close monitoring
832 of those potentially toxic by-products and their level of ecotoxicity is advised.

833 *Solutions contaminated with a mixture of organic and inorganic contaminants*

834 The treatment of mixtures of inorganic and organic compounds represents another
835 technical issue. Landfilling is the most important solid waste management solution
836 performed worldwide owing to its low-cost and simplicity (Fernandes et al., 2015;
837 Mandal et al., 2017). Leachate collection systems are conventionally implemented and
838 the characteristics of these leachates depend upon the hydrogeology, the age of the
839 landfill, the composition and moisture content of the waste, as well as seasonal variations
840 (Mandal et al., 2017). The great challenge of landfill leachates lies in their varying
841 composition, high concentration and their toxicity. Several studies proposed to deal with
842 raw landfill leachate using electrochemical technologies, as reviewed by Mandal et al.
843 (2017), an up-to-date selection of which is given in Table 6. The amount of carbon content
844 is usually very high, with COD varying from 1 to 50 g-O₂ L⁻¹, generally basic pH (8-9)
845 and high concentrations of inorganic ions, such as Cl⁻ (1.5 – 6 g L⁻¹), SO₄²⁻ (0.1-1.8 g L⁻¹)
846 and NH₃ (0.4-3.2 g L⁻¹). Such complex solutions are non-biodegradable (BOD₅/COD
847 < 0.2), which restricts the secondary treatment possibilities. The advantage of electro-
848 oxidation is that the initial presence of chloride ions leads to the formation of active

chlorine species, such as HClO, as described earlier (Eqs. 1-2) (Cossu et al., 1998). This oxidizing agent participates in the degradation and mineralization of organic compounds; however, some organo-halogenated transformation byproducts can also be generated. These compounds are known to be potentially toxic and therefore deserve particular attention. The initial presence of ammonia at high level could also constitute a drawback, since this species can be oxidized to nitrate (NO_3^-), an unwanted final product at concentrations higher than the disposal regulations (Anglada, Urtiaga, et al., 2010). The formation of this ion must therefore be carefully monitored. In order to have an overall approach for a cost-effective complete removal of organic and inorganic species, combination with separation processes (e.g., nanofiltration, reverse osmosis, etc) (Cui et al., 2018; El Kateb et al., 2019; Fernandes et al., 2019; Ukundimana et al., 2018) and/or biological technologies (Anglada, Urtiaga, et al., 2010; Baiju et al., 2018; Ding et al., 2018; El Kateb et al., 2019; Moreira, Soler, et al., 2015; Turro et al., 2012; Zhao et al., 2010) has been suggested in previous studies. The advantages of such combined approaches will be reviewed in a dedicated section towards the end of this review.

Another challenging source of mixed contamination (organic and inorganic) that represents a major problem in the mining industry is acid mine drainage (AMD), generated by the oxidation of sulfides during ore processing, leading to sulfuric acid (Eq. 19).

The chemical composition of AMD depends on the mined rock mass and hydrogeology; they are typically characterized by acidic pH as low as 2, abundance of dissolved species, including sulfate, iron and other toxic metals (mainly lead, copper, cadmium, and arsenic) (Bejan & Bunce, 2015). Sulfate is usually found at high concentration (in the range of hundreds to thousands of mg L^{-1}) from (di-)sulfide air/water

oxidation and its water discharge regulation is becoming more stringent (Nariyan et al., 2018). In parallel, ferric hydroxide ($\text{Fe}(\text{OH})_3$) precipitation is another environmental issue. Several studies on the efficiency of electrochemical treatment of AMD as shown in Table 7. The primary goal of electrochemistry is to increase the pH, as an alternative to chemical neutralization and precipitation, by reducing H^+ into hydrogen gas ($\text{H}_2(\text{g})$) at the cathode, according to Eq. 20:

For this purpose, the operation in divided cell is favored in order to avoid the oxidation of Fe^{2+} into Fe^{3+} that then precipitates on the electrodes – especially on the cathode – and subsequently causes electrode fouling (Chartrand & Bunce, 2003). The assistance of chemical alkalization using CaO as pre-treatment before the electrochemical step has also demonstrated synergistic effects (Orescanin & Kollar, 2012; Radić et al., 2014). Additional benefits can be obtained from such electrochemical treatment by electrodeposition of metals and by producing a dewaterable sludge for possible valorization (Bunce et al., 2001). Another interesting feature is the elimination of inorganics such as sulfate by EC (Mamelkina, Tuunila, et al., 2019; Nariyan et al., 2017). It is finally worthy of note that the initial presence of iron could enhance the electrochemical process efficiency by involving $\cdot\text{OH}$ radicals through the Fenton's reaction in an EF process.

Effluents with high rheological complexity

Most of the electrochemically treated solutions have a simple rheological nature with Newtonian behavior, following the common Newton's law of viscosity. In Newtonian fluids, the contaminant concentration is easily homogenized in the bulk solution upon reaching stationary stage with sufficient convection. However, in non-Newtonian liquids, the pollutants concentration is not homogeneous even with good stirring. Then, the

challenge is to increase the contact between the electro-generated oxidants and the contaminants in the electrochemical cell. Sludge from wastewater treatment plant (WWTP) is an example of non-Newtonian liquid (Ratkovich et al., 2013). The sludge from WWTP is characterized by a high moisture content, usually higher than 97% alongside high total solids (TS) content, in the range of 1 g L⁻¹ to several tens of g L⁻¹, resulting in a solid-liquid mixture (Cai et al., 2019; Chen et al., 2019). WWTPs generate a lot of sludge from primary treatment and biological processes that then need to be disposed safely. Land application is one of the most appropriate solution to benefit from the fertilizers present in the sludge after appropriate pre-treatment. Several publications proposed to pre-treat the sludge mainly for dewatering purpose by electrochemical process, a selection of which is presented in Table 8.

The presence in sludge of initial salts such as chloride ions induces several positive effects on the process efficiency. The electro-oxidation of Cl⁻ at Ti/RuO₂ anode further allows the generation of hypochlorous acid (HClO, Eqs. 1-2) (Bureau et al., 2012; Song et al., 2010), a powerful oxidizing agent known to inhibit the activity of micro-organisms. HClO could also oxidize the organic compounds presents in the sludge so that a decrease of COD is observed. Finally, a decrease of phosphate (PO₄³⁻) initially present in the sludge was also observed (Bureau et al., 2012), which could be attributed to the oxidation of ferrous into ferric iron by HClO (Eq. 21) and then Fe³⁺ reaction with H₂PO₄⁻ formed a FePO₄ precipitate (Eq. 22) (Bureau et al., 2012):

In addition, calcium (Ca), potassium (K) and magnesium (Mg) concentrations increased by 20% to 50%, while total nitrogen content remained stable (Bureau et al., 2012), ensuring that the fertilizing value of the sludge was not compromised. Finally,

electro-oxidation pre-treatments allowed sludge disinfection, a decrease in organic carbon content, metal deposition and a raise in nutrient content (Drogui et al., 2013; Masihi & Gholikandi, 2018; Tuan & Sillanpaa, 2020). The potential problems of electrode fouling and corrosion as well as biofilm development in long-term use have not yet been discussed in detail and should be tested in further studies.

High COD load with low pollutant content

The advanced electrochemical treatment of solutions highly loaded with COD but containing low pollutant content mostly alludes to soil remediation. Organic pollutants that remain in the soil have different properties regarding their volatility, hydrophobicity, water solubility and therefore different technologies have been tested, including thermal treatment, biological processes and physicochemical techniques. Among the latter, soil washing and soil flushing have emerged as cost-efficient alternatives to thermal processes that denature the soil and to bioremediation when pollutants are biorefractory.

In soil washing/flushing technologies, extracting agents are added most of the time in a water solution before washing the excavated soil or flushing the non-excavated soil in order to solubilize the hydrophobic organic pollutants (Mousset et al., 2013). A good washing agent is a compound with high extraction efficiency (thus requiring as low an amount of agents as possible), low ability to adsorb onto soil and low toxicity towards soil microorganisms. They usually have amphiphilic properties that allow dissolving soil pollutants into the water solution. There are many kinds of agents that have been tested in the literature including co-solvents, dissolved organic matter, deoxyribonucleic acids, vegetable oils, fatty acid methyl esters, cyclodextrins and surfactants (Trellu et al., 2016). Most of them (except for co-solvents) have a high molecular weight ranging from 250 to 1500 g mol⁻¹. As soil washing/flushing only transfers the pollution from solid to liquid matrix, there is a need for a post-treatment to remove the contaminants. In the past few

years, advanced electrochemical treatment has appeared as a promising alternative to biological post-treatment, since those pollutants are barely biodegradable (Mousset et al., 2017).

Table 9 presents the state of the art of electrochemical treatment of soil washing/flushing solutions. Generally, it is observed that organic pollutants represent only 1% to 5% of the total organic carbon fraction, the remaining part consisting mostly of the extracting agent, while the organic matter content extracted from soil is negligible (Mousset et al., 2017). Interestingly, using different families of extracting agents has shown to affect the electrochemical treatment efficiency. Surfactants have a long C-C bond hydrophobic chain and a hydrophilic head, which allows them to form micelles after reaching a minimal concentration, known as the critical micelle concentration (CMC, Figure 5a) (Mousset et al., 2014a). These micelles can then trap the hydrophobic organic contaminants (HOCs), leading to steric hindrance during the oxidation of the pollutant. Thus, the surfactants are preferentially degraded as compared to the pollutants. Alternative agents such as cyclodextrins have also been proposed (Mousset & Oturan, 2014), with a toroidal shape that allows trapping the organics into their hydrophobic cavity (Figure 5b). The functional groups at the surface of cyclodextrins (carboxylic groups, hydroxyl groups, etc) allow them to bound to ferrous iron under EF treatment, with hydroxyl radicals being produced close to the contaminants. The pollutant was therefore preferentially degraded as compared to the cyclodextrin, thanks to the ternary complex formation ($\text{Fe}^{2+}:\text{HPCD}:\text{HOC}$) according to Eqs. 23 and 24:

Interestingly, iron can be present initially in the soil solution at concentrations high enough to avoid external addition (Mousset et al., 2016). Such treatment even allows

reusing the cyclodextrins for further soil washing/flushing cycles in order to decrease the treatment cost. Still, Tween 80 surfactant could extract ten times more pollutants from soil than cyclodextrin, which counterbalanced the ability of cyclodextrin to be more easily recycled (Mousset et al., 2017). Even if surfactants are the most important family of agents employed in soil washing/flushing owing to their cost-effectiveness, it is important to emphasize that an overview of the complete soil remediation treatment needs to be obtained before the selection of a suitable extracting agent can be made. Additionally, treating soil washing/flushing solutions electrochemically leads to high energy consumption (100-500 kWh m⁻³) (Mousset et al., 2017) and the faradaic yield, i.e. the ratio between the electric energy devoted to the target compound and the total electric energy applied, is in this case an important parameter to optimize when treating such soil solutions.

Conclusion

In conclusion, this review has provided an insight into the specificities of highly concentrated waste streams, why and how electrochemical treatments constitute a promising option for such effluents. The first section defined the types of wastewater that display the best potential for electrochemical treatment. Several advantages make them particularly suitable for electrochemistry and in particular their high salinity, providing a source of natural electrolyte and favoring the formation of active species with great decontamination and disinfection potential. Challenges exist in terms of variability, need for high faradaic yields, and special care to be given to degradation byproducts before discharge (e.g., organo-halogenated compounds). To face these challenges, we examined a number of strategies for electrochemical wastewater treatment, with special focus on AO, EF and EC, alone or in combination with other processes. In particular, for highly

concentrated wastewater, it may prove more beneficial to apply electrochemistry as a pre- or post- treatment approach, combined with a more economical conventional biological treatment system. Process optimization and cost-efficiency are at the core of a suitable treatment strategy; yet, there is a lack of a focused and consistent approach to scale up electrochemical reactors. In fact, there are very few pilot studies and therefore, the final section of this review emphasized the importance of modeling electrochemical processes, as well as the rheological and mass transfer conditions in large scale electrochemical reactors. By combining various modeling approaches, electrochemical wastewater treatment can be understood in a more systematic manner and adequate reactors can be designed. We defined the electrode area over treated volume ratio (A/V) as a key parameter, which in turn influences energy consumption. We concluded that in most cases, the specific energy consumption is lower when the wastewater contains a high concentration of contaminants. At a time that electrochemistry for wastewater treatment reaches maturity, highly contaminated waste streams may therefore provide the best niche to demonstrate the potential of electrochemical wastewater treatment at large scale.

Declaration of interest statement

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 paper.

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Table 1. Compilation of representative publications on stand-alone electrochemical processes applied to the treatment of highly polluted wastewater.

Wastewater	Composition*	Process	Reactor capacity (L)	Anode	Cathode	Reactor type**	Efficiency	Reference
Textile	COD = 470, TSS = 68, pH = 8.8,	AO	0.5	BDD	Zr	UR	85% TOC removal	(Tsantaki et al., 2012)
Textile	COD = 5957, pH = 7.3	AO	Bench-scale	DSA	SS	FTR	94-99% of color removal, COD no reported	(Vaghela et al., 2005)
Textile	COD = 1224, BOD ₅ = 324, pH = 4.8, SO ₄ ²⁻ = 38, Cl ⁻ = 234	EF	0.5	Pt	Carbon fiber	UR	75.2% COD removal	(Wang et al., 2010)

Pharmaceutical	COD = 12000, TOC = 1600, C = 7.00, pH = 7, TS = 5000	AO	0.6	BDD	SS	FR	Almost complete TOC removal	(Domínguez et al., 2012)
Municipal and textile after RO	COD = 158.08, pH = 8.21, Cl ⁻ = 592.02, ClO ₃ ⁻ = 1.17, NH ₄ ⁺ = 48.24, C = 3.99	AO	1	BDD	Ti	FR	> 80% COD removal	(Van Hege et al., 2002)
Industrial after UF-RO (spiked with 28 micropollutants)	DOC = 57, C = 4.250, pH=7.5, Fe=0.22, SO ₄ ²⁻ = 242, Cl ⁻ = 1500	AO	10	Ru _{0.7} Ir _{0.3} O ₂	SS mesh	FR	30% DOC removal	(Radjenovic, Bagastyo, et al., 2011)

Municipal sanitary landfill leachate	COD = 3385, BOD ₅ = 500, C = 22.60, pH = 8.4, SO ₄ ²⁻ = 11, Cl ⁻ = 2574, NH ₄ ⁺ = 1591, NO ₃ ⁻ = 1.9, PO ₄ ³⁻ = 31	AO	10	BDD	BDD	FR	51% COD removal and 32% ammonia removal	(Anglada et al., 2011)
Municipal sanitary landfill leachate	COD = 3782, BOD ₅ = 560, pH = 8.4, Cl ⁻ = 3702, NH ₄ ⁺ = 3143	AO	0.8	RuO ₂ - IrO ₂	SS	UR	26.5% COD removal and 81.1% NH ₄ ⁺ removal	(Zhang et al., 2010)
Industrial landfill leachate	COD = 17100-18400, pH = 9, Fe = 20, Cl ⁻ = 52300, NH ₄ ⁺ = 1200	AO	1.1	carbon plate	SS	UR	83% DOC removal	(Nageswara Rao et al., 2009)
Municipal sanitary	COD = 920-1448, C = 10.00-10.90, pH = 8.1-9.4, SO ₄ ²⁻ = 140-199, Cl ⁻	AO	250	BDD	SS	Pilot FR	70% of COD removal and 37% of NH ₄ ⁺	(Anglada, Urtiaga, et al., 2010)

landfill leachate	= 1615-1819, NH_4^+ = 896-980, NO_3^- = 5-1207							
Municipal sanitary landfill leachate	NH_4^+ = 1934, COD = 4434, TOC = 2782, BOD_5 = 640, TSS = 317, C = 12.77, pH = 8.35	AO	NA	BDD	SS	FR	Total NH_4^+ removal	(Cabeza, Urtiaga, Rivero, et al., 2007)
Municipal sanitary landfill leachate	DOC = 1600-3100, TSS = 15310-17602, pH = 8- 9, SO_4^{2-} = 60-240, Cl^- = 5000-6200, NH_4^+ = 1900- 3200, with and without filtration	AO, EF	0.25	BDD	CF	UR	93% DOC removal (EF)	(Oturán et al., 2015)
Municipal sanitary landfill	COD = 1030-1505, DOC = 337-430, BOD_5 = 1-10, NO_3^- = 1035-1192, SO_4^{2-}	SPEF	1.18	BDD	Pt, C- PTFE GDE	FR	80% DOC removal	(Moreira et al., 2016)

leachate (previously subjected to biological and coagulation)	= 1749-1917, Cl ⁻ = 3046- 3822, pH = 2.2-2.9							
Textile	COD = 1224, BOD ₅ = 323.8, TOC = 394.6, Cl ⁻ = 233.9, C = 2.91	EF	0.5	Pt	Carbon fiber cloth	UR	75.2% COD removal	(Wang et al., 2010)
Olive oil mill wastewater	COD = 2368, BOD ₅ = 790, TSS = 3712, SO ₄ ²⁻ = 910	AO / EF / PEF	0.1	BDD	C-PTFE GDE	UR	80% TOC removal	(Flores et al., 2017)
RO concentrate	COD = 120-150, BOD ₅ = 15-18, TOC = 25-35, pH = 7.2-7.6, C = 22.30	EF	0.2	Pt	Carbon felt	UR	62% COD removal	(Zhou et al., 2012)

Slaughterhouse wastewater	COD = 405.6, TSS = 150, Cl ⁻ = 364, pH = 8.5	EF	1	DSA/BD D	Carbon felt	UR	83.50% COD removal	(Paramo-Vargas et al., 2015)
Soils contaminated with petroleum (washing solution)	COD = 22000, TOC = 5100	EF	0.4	BDD	Carbon felt	UR	>97% TOC removal	(Huguenot et al., 2015)
Oily water (synthetic)	COD = 1000 , pH = 7	AO	Bench-scale	TiO ₂ -coated carbon membrane	SS	UR	>90% COD removal	(Yang et al., 2012)

Carwash wastewater	COD = 572, BOD ₅ = 178, pH = 6.4, C = 1.6, Anionic surfactants = 95.5	AO	0.4	BDD/Pb O ₂	SS	UR-R	Almost total COD removal	(Panizza & Cerisola, 2010)
Melodonin wastewater (glucose and glycine)	COD = 2500, TOC = 1000, pH = 7	AO	0.6	BDD	SS	FR	Complete COD removal	(Cañizares et al., 2009)
Almond industry wastewater	COD = 1300, TOC = 410, TSS = 33, Cl ⁻ = 2000, pH = 6.7 C = 5.3	AO	20	DSA-RuO ₂	SS	FR	80% COD removal	(Valero et al., 2014)
Textile	COD = 2000 DOC = 485, pH = 6.8, TSS = 230	EC	3.5	Fe or Al in monopolar	Fe or Al	FTR (continuous flow)	85% COD removal (Fe) and 77% (Al)	(Kobyas et al., 2016)

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Municipal	COD = 238.63, BOD ₅ = 205.76, SS = 281.9, pH = 7.7, NH ₄ ⁺ = 29.85,	EC	200	Al	Al	FTR (continuous flow)	97.27-100% Total-P and NH ₄ ⁺ removal	(Nguyen et al., 2014)
Olive oil mill wastewater	COD = 1393.3, TOC = 581.1, pH = 6.83, TSS = 1890, TN = 16.7, Cl ⁻ = 41.8, SO ₄ ²⁻ = 1558.3	EC-PEF	0.15 / 0.1	Fe/BDD	Fe/ C-PTFE GDE	UR	40% TOC and COD (EC), 97% (coupled EC-PEF)	(Flores et al., 2018)
Phosphates (synthetic)	PO ₄ ³⁻ =27, SO ₄ ²⁻ = 500	EC	0.5	Fe/Al	Fe/Al	FR	Complete PO ₄ ³⁻ removal	(Lacasa et al., 2011)
Copper mineral processing	As(V) = 2200, pH = 2.1	EC	1	Fe	Fe	UR	Complete removal of As(V)	(Nuñez et al., 2011)

Acid mine drainage	$\text{SO}_4^{2-} = 13000$, TDS = 19790, pH = 2.68, C = 10.66	EC	0.5	Al	Fe	UR	41% of SO_4^{2-} removal	(Nariyan et al., 2017)
Washing machine wastewater	COD = 783, TOC = 680, C = 1.29, pH = 15	AO	1	BDD/Pt	Ti	FR	88% COD removal (BDD), 71% (Pt)	(Durán et al., 2018)
Soils contaminated with PAH (washing solutions)	Total PAH = 2.2 and 31.1, TOC (from PAH) = 37 and 272 *	EF	0.4	Pt	Carbon felt	UR	>99% PAH removal	(Mousset et al., 2016)
Electronics wastewater	TOC = 53.46 and 299.31, COD = 190, BOD ₅ = 49, acetone = 73.03, isopropanol = 10.01,	EF	0.4	BDD	Graphen e-PTFE-GDE	UR	90% and 60% TOC removal (for low and high TOC ₀ , respectively)	(Garcia-Rodriguez et al., 2018)

	NH ₄ ⁺ = 100, PO ₄ ³⁻ = 636, pH = 8.50, C = 2.1							
RO concentrate	COD = 53, DOC = 53, pH = 7.8, Cl ⁻ = 1526, NO ₃ ⁻ = 14.1, SO ₄ ²⁻ = 253	AO	1	BDD/DS A (Pt- IrO ₂ and SnO ₂ - Sb ₂ O ₅)	SS	FR (undivided)	74% COD removal and 51% DOC (BDD)	(Bagastyo et al., 2013)
RO concentrate from Municipal WWTP	COD = 17, pH = 7.5, TSS = 5.7, NH ₄ ⁺ = 120, C = 2665, Cl ⁻ = 501-808, SO ₄ ²⁻ = 514-560, NO ₃ ⁻ = 95-145	AO	2	BDD	SS	FR	95% removal of 12 selected micropollutants	(Urtiaga et al., 2013)
RO concentrate from WWTP	COD = 272, pH = 8.3, Cl ⁻ = 1500, C = 2.6, TN = 40.8	AO	0.82	Ti/PbO ₂	Ti	UR	72% COD and 18% of TN removal	(Wang et al., 2018)

Pharmaceutical	COD = 1253.3, TOC = 431.5, BOD ₅ /COD = 0.18, pH = 8.1, C = 2.2	EF	0.5	BDD and DSA	Carbon brush	UR	Total COD removal and 97.1% of TOC removal	(Olvera-Vargas et al., under review)
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Table 2. AO, EC and EF studies carried out at volumes above one liter.

Process	Wastewater	Treated volume (L)	Pollutant concentration	Removal (%)	Treatment time (min)	Electrode area (cm ²)	A V ⁻¹ (m ² m ⁻³)	Current Density (mA cm ⁻²)	Energy consumption (kWh kg ⁻¹ pollutant)	Ref.
AO	Tannery	1.2	1202.5 mg L ⁻¹ COD	51.53	90	16	1.3	25	16.1 kWh kg ⁻¹ COD	(Liu et al., 2017)
AO	Washing machine	1	783 mg L ⁻¹ COD	79.7	360	63.5	6.4	16.6	-	(Durán et al., 2018)
AO	landfill leachate	10	3385 mg L ⁻¹ COD	30	180	70	0.7	257	134 kWh kg ⁻¹ COD	(Anglada et al., 2011)
AO	rubber manufacturing process	200	1500 mg L ⁻¹ COD	80	600	2800	1.4	60	77 kWh kg ⁻¹ COD	(Urtiaga et al., 2014)

AO	landfill leachate	250	860 mg L ⁻¹ COD	80	420	10500	4.2	30	53 kWh kg ⁻¹ _{COD}	(Anglada, Ortiz, et al., 2010)
AO	Industrial landfill leachate	1.1	17750 mg L ⁻¹ COD	80	360	140	12.7	21.4	6.8 kWh kg ⁻¹ _{COD}	(Nageswara Rao et al., 2009)
AO	Almond industry	20	1300 mg L ⁻¹ COD	70	330	3300	16.5	32	319.1 kWh kg ⁻¹ _{COD}	(Valero et al., 2014)
AO	1,4-Dioxane	66	1300 mg L ⁻¹ 1,4-DX	54	206 h	8400	12.7	0.3	345.5 kWh kg ⁻¹ _{1,4-DX}	(Blotevogel et al., 2019)
AO	cooking wastewater	500	3160 mg L ⁻¹ COD	59.5	212	16320	3.3	0.5	0.2 kWh kg ⁻¹ _{COD}	(Liu et al., 2020)
EC	oily wastewater	333	870 mg L ⁻¹ COD	77	20	80000	6.3	5	3 kWh kg ⁻¹ _{COD}	(Bilgili et al., 2016)

EC	petroleum refinery	1000	4700 mg L ⁻¹ COD	35	60	36500	3.6	1.8	-	(El-Naas et al., 2016)
EC	Tannery	2	6000 mg L ⁻¹ COD	95	360	105	5.3	40	5.5 kWh kg ⁻¹ _{COD}	(Elabbas et al., 2016)
EC	Winery	8	10240 mg L ⁻¹ COD	55	310	937.5	11.7	70	75.2 kWh kg ⁻¹ _{COD}	(Orescanin et al., 2013)
EC	Textile	3.5	2000 mg L ⁻¹ COD	85	80	660	18.9	6.5	45 kWh kg ⁻¹ _{COD}	(Kobyas et al., 2016)
EC	Table olive processing wastewaters	200	1000 mg L ⁻¹ COD	42.5	50	9600	4.8	5.7	1.3 kWh kg ⁻¹ _{COD}	(Benekos et al., 2019)
EC	Raw cheese whey wastewater	3	29,563 mg L ⁻¹ _{TOC}	22.1	60	140	4.7	30	3.3 kWh kg ⁻¹ _{TOC}	(Tirado et al., 2018)

EC-Fenton	Olive mill wastewaters	36	95000 mg L ⁻¹ COD	48	240	6451	17.9	3.1	0.8 kWh kg ⁻¹ _{COD}	(Khoufi et al., 2009)
EF	Aged landfill leachate	1.5	490 mg L ⁻¹ COD	83.5	80	24	1.6	30	-	(Lu, 2019)
EF	Aniline solution	30	780 mg L ⁻¹ TOC	61	120	100	0.3	200	81.4 kWh kg ⁻¹ _{TOC}	(Brillas & Casado, 2002)
EF	Bisphenol A	1.5	379 mg L ⁻¹ COD	18	90	67	4.5	120	-	(Chmayssem et al., 2017)
EF-Adsorption	Phenol	1.2	940 mg L ⁻¹ Phenol	99	120	120	10	2.5	0.6 kWh kg ⁻¹ _{phenol}	(Garcia-Rodriguez et al., 2020)
SPEF-AO	Winery wastewater	1.3	380 mg L ⁻¹ COD	68.4	240	10	0.8	25	19.6 kWh kg ⁻¹ _{COD}	(Moreira, Boaventura, et al., 2015)

SPEF-AO	Industrial textile wastewater	10	1763 mg L ⁻¹ COD	93	360	50	0.5	25	4.2 kWh kg ⁻¹ _{COD}	(Salazar et al., 2019)
SPEF-AO	Mixture of parabens	2.5	100 mg L ⁻¹ TOC	66	240	20	0.8	10	84 kWh kg ⁻¹ _{TOC}	(Steter et al., 2018)
SPEF-AO	Mixture of azo dyes	2.5	105 mg L ⁻¹ TOC	98.8	300	20	0.8	100	660 kWh kg ⁻¹ _{TOC}	(dos Santos et al., 2018)
SPEF-AO	EC pre-treated cheese whey wastewater	2	23,029 mg L ⁻¹ TOC	27.1	420	20	1	100	7.3 kWh kg ⁻¹ _{TOC}	(Tirado et al., 2018)
SPEF-AO	Mixture of pesticides	75	71 mg L ⁻¹ DOC	32	120	400	0.5	74	1201.4 kWh kg ⁻¹ _{DOC}	(Salmerón et al., 2019)
SPEF	benzoic acid solution	10	100 mg L ⁻¹ TOC	95	60	100	1	200	-	(Casado et al., 2006)

Table 3. Compilation of representative publications on the treatment of highly polluted wastewater by electrochemical processes in combination with other methods.

Wastewater	Composition*	Process	Reactor capacity (L)	Anode	Cathode	Reactor type**	Efficiency	Reference
Municipal sanitary landfill leachate	COD = 3106-4057, BOD ₅ = 180-300, TSS = 525-630, NH ₄ ⁺ = 1300-1355, Cl ⁻ = 2220-2780	BIO + Coagulation / aeration + EF, SPEF-UV, SPEF	25	BDD	C-PTFE GDE	FR	78% DOC removal (SPEF)	(Moreira, Soler, et al., 2015)
Municipal sanitary	COD = 4430, 1300 and 200, BOD ₅ = 1196, 175, NA pH = 8.4, 7.7, 7.5,	BIO-Fenton + AO	1	BDD	SS	FR	Overall, > 95% of COD removal and	(Urtiaga et al., 2009)

landfill leachate	$\text{SO}_4^{2-} = 438,500,3100$, Cl^- $= 2124, 1876, 1460$, $\text{NH}_4^+ = 1225, 750, 700$, $\text{NO}_3^- = \text{NA}, 700, 660$						complete NH_4^+ removal	
Landfill leachate	$\text{COD} = 7200$, $\text{BOD}_5/\text{COD} = 0.03$, $\text{Cl}^- =$ 6000 , $\text{Fe} = 70$, $\text{pH} = 8.2$, $\text{TDS} = 9500$	Heterogen eous EF + BIO	1	Ti/TiO ₂	Graphite	UR	82% of COD removal with EF, overall 97%	(Baiju et al., 2018)
Winery wastewater	$\text{COD} = 10240$, $\text{C} = 1.10$, $\text{TSS} = 2860$, $\text{pH} = 3.7$, Fe $= 17$	AO + EC + Electroma gnetizatio n + Clarificati	8	SS	SS	UR	77% COD removal, 99% removal of PO_4^{3-} and TSS	(Orescanin et al., 2013)

		on + O ₃ / H ₂ O ₂ / UV						
Winery wastewater	COD = 12000, 380, BOD ₅ = 7950, 150, TSS = 81, 72, pH = 3.7, 8.3	BIO + AO / EF / PEF / SPEF	1.25	BDD	C-PTFE GDE	FR	COD, BOD ₅ , NH ₄ ⁺ , NO ₃ ⁻ and SO ₄ ²⁻ above regulatory levels	(Moreira, Boaventura, et al., 2015)
Cellulose bleaching	COD = 1250 (acid), 1500 (basic), BOD ₅ = 563 (acid), 595 (basi), pH = 2.5, 10.5, Cl ⁻ =488.2 (acid), 349.9 (basic), TOC = 499 (acid), 594 (basic)	Filtration + AO	0.2	DSA or BDD	C-PTFE GDE	UR	96% NF+AO	(Salazar et al., 2015)

Dairy wastewater	COD = 3859, 395(after EC), BOD ₅ = 1517, TSS = >1988, pH = 8.3, Cl ⁻ = 1131, NH ₄ ⁺ = 177, NO ₃ ⁻ = 1.5	EC + AO	0.1	DSA (IrO ₂ -Ta ₂ O ₅)	Pt	UR	85%	(Borbón et al., 2014)
Dairy wastewater	COD = 5600, TOC = 2464, BOD ₅ /COD = 0.8, TDS = 4530, pH = 5.87, Cl ⁻ = 654, C = 3.2	EC + Phytoremediation	1	Al	Fe	UR	86.4% of COD removal with EC, Overall 97% of COD removal	(Akansha et al., 2020)
Tannery	COD = 9922-10180, BOD ₅ = 528, TSS = 445-530, pH = 3.7-4.3, Cl ⁻ = 1239, Fe = 2-2.8	EC / AO / EF / PEF + PEF-UVA	0.25	BDD	Fe/BDD	UR	95% EC+PEF	(Isarain-Chavez et al., 2014)

Anaerobic sludge from poultry farm	COD = 26200, TOC = 5236, TSS = 31300, TDS = 9400, pH = 7.8-8.2, total coliforms = 2.4×10^3 CFU mL ⁻¹	ECP + EF	0.5	Fe for ECP BDD for EF	Fe for ECP Carbon brush for EF	UR	89.3% of COD removal by ECP, 91.6% of COD and 87.2% of TOC removal by EF, Overall, 99.1% of COD and 93.2% of TOC removal	(Olvera-Vargas et al., 2019)
Anaerobically-digested food wastewater	COD = 3210-3420, TN = 4832-4728, NH ₄ ⁺ = 3352-3790, SS = 6260-6400, TS = 15580	ECP + AO	1 / 0.3	SS/DSA	SS/Ti	UR	>90% TOC and NH ₄ ⁺ removal	(Shin et al., 2017)
Municipal wastewater	COD = 495, BOD ₅ = 359	BIO + EC/AO and	0.3	For EC, Fe	SS	FR	80% COD combined process	(Kishimoto et al., 2017)

(spiked with 1,4-dioxane)		EC/AO + BIO		For AO, DSA (Ti/RuO ₂)				
Pulp and paper	COD = 1450, BOD ₅ = 350, TSS = 350, TDS = 1050, Cl ⁻ = 325, pH = 6.72	Permanga nate oxidation + ECP + Co ₃ / UV / peroxymo nosulfate	2 / 0.35 / 0.15	Fe	Fe	UR	overall 95% COD removal	(Jaafarzadeh et al., 2017)
Olive oil mill wastewater	COD = 100000, polyphenols = 12000	ECP + Anaerobic digestion + EC	3	cast Fe	Cast Fe	UR	65.8% of polyphenols by ECP and reduction of	(Khoufi et al., 2006)

							toxicity; overall 97% COD removal	
Winery wastewater	COD = 86100, TOC = 60100, pH=3.4	PEF + LED radiation (light emitting diode)	0.04	Graphite	Graphite/ Ni-foam/GDE	UR (columns)	51% TOC removal	(Díez et al., 2017)
Slaughterhouse wastewater	COD = 1494, TSS = 19717, BOD ₅ = 685, pH = 7.8, NH ₄ ⁺ = 181	Anaerobic digestion + SPEF	0.5 / 0.1	BDD	C-PTFE GDE	UR	90% COD removal with BIO and 97% overall	(Vidal et al., 2016)
Cheese whey wastewater	TOC = 29563, pH = 5.64, TSS = 5880, TN = 1106, Cl ⁻ = 3547, C = 15.5	EC + UV-assisted AO	3.2 L for EC, 2 L for AO	Fe for EF, BDD for AO	Al/AISI304 for EC,	UR for EC, FR for AO	22-27% of TOC removal with EC, Overall 49.1% TOC removal	(Tirado et al., 2018)

					Pt for AO			
Pharmaceutical	COD = 72153.34, TOC = 2045.68, Trimethoprom = 0.2 mM	Aerobic sludge + EF	1	Pt	Carbon felt	UR-R	Overall 89% of TOC removal	(Mansour et al., 2015)
Mixed wastewater from WWTP	COD = 1152, TOC = 274, BOD ₅ /COD = 0.43, TDS = 8370, pH = 7.2, Cl ⁻ = 3691	EF with persulfate + BIO	1	Ti/Pt	Graphite felt	UR	60% of COD removal with EF persulfate, Overall 94% COD removql	(Popat et al., 2019)

Table 4. Literature review of industrial wastewater treatment by electrochemical processes.

Kind of industry	Matrix	Process	Electrode materials	Main parameters tested	Reference
Pharmaceutical	Pharmaceutical wastewater (COD = 4 g-O ₂ L ⁻¹ , pH 5.2, conductivity = 0.5 mS cm ⁻¹)	EC/Fered-Fenton	Cathode: Fe (22.5 cm ²) Anode: Fe (22.5 cm ²)	TOC removal, energy consumption	(Başaran Dindaş et al., 2020)
Pharmaceutical	Pharmaceutical wastewater (COD = 12 g-O ₂ L ⁻¹ , pH 8.5, conductivity = 7 mS cm ⁻¹)	AO	Cathode: stainless steel (78 cm ²) Anode: BDD (78 cm ²)	Current density, flow rate, TOC removal	(Domínguez et al., 2012)
Chemical	Chemical industry wastewater (COD = 1.3 g-O ₂ L ⁻¹ , pH 6.7, conductivity = 5.3 mS cm ⁻¹ , [Cl ⁻] = 0.4 g L ⁻¹)	AO	Cathode: Pt (63 cm ²) Anode: Ti/IrO ₂ , Ti/RuO ₂ (63 cm ²)	Anode materials, temperature, current density, COD removal, energy consumption	(Valero et al., 2014)
Chemical	Chemical industry wastewater (COD = 1.0 g-O ₂ L ⁻¹ , pH 6.1,	AO	Cathode: graphite (100 cm ²)	COD and TOC removal, by-products formation	(Rajkumar & Palanivelu, 2004)

	conductivity = 3.3 mS cm ⁻¹ , [Cl ⁻] = 0.5 g L ⁻¹)		Anode: Ti/IrO ₂ -RuO ₂ (28 cm ²)		
Textile	Tannery wastewater after biological treatment (COD = 1.6-1.7 g-O ₂ L ⁻¹ , 6.2 mS cm ⁻¹ , pH 7.3-7.9, [Cl ⁻] = 720 mg L ⁻¹)	AO	Cathode: stainless steel Anode: Ti/PbO ₂ (272 cm ²)	pH, color removal	(Ling et al., 2016)
Textile	Tannery wastewater after biological treatment (COD = 1.3 g-O ₂ L ⁻¹ , 1.9 mS cm ⁻¹ , pH 8.8, [Cl ⁻] = 180 mg L ⁻¹)	AO	Cathode: graphite Anode: Ti/Pt	Electrolyte concentration, current density, temperature, Color removal, COD removal, current efficiency	(Wang et al., 2009)
Tannery	Tannery wastewater (COD=2.4 g-O ₂ L ⁻¹ , conductivity = 18.5 mS cm ⁻¹ , pH 7.8, [Cl ⁻] = 5.5 g L ⁻¹)	AO	Cathode: stainless steel (100 cm ²)	Anode materials, current density, energy consumption	(Szpyrkowicz et al., 2005)

			Anode: Ti/Pt–Ir, Ti/RhO _x –TiO ₂ , Ti/PdO– Co ₃ O ₄ (100 cm ²)		
Tannery	Synthetic tannery wastewater (COD=2-20 g-O ₂ L ⁻¹ , tannic acid = 1-3 g/L, conductivity = 10-30 mS cm ⁻¹ , pH 3-13, [Cl ⁻] = 1.5-28 g L ⁻¹)	AO	Cathode: stainless steel (25 cm ²) Anode: Ti/IrO ₂ , Ti/PbO ₂ (25 cm ²)	Anode materials, temperature, pH, current density, current efficiency	(Panizza & Cerisola, 2004)
Tannery	Tannery wastewater (COD = 10 g-O ₂ L ⁻¹ , conductivity = 6.3- 9.1 mS cm ⁻¹ , pH 3.7-4.3, [Cl ⁻] = 1.3 g L ⁻¹)	AO, EF	Cathode: BDD, Fe (3 cm ²) Anode: BDD, Fe (3 cm ²)	Electrode materials, current density, mineralization evolution, current efficiency	(Isarain-Chavez et al., 2014)
Petroleum	Petroleum wastewater (COD=19.8 g-O ₂ L ⁻¹ , total petroleum hydrocarbons = 11.2	AO	Cathode: Pt grid Anode: Ta/PbO ₂ and BDD (7 cm ²)	Anode materials, COD and TPH removal, by-products, ecotoxicity	(Gargouri et al., 2014)

	g L ⁻¹ , conductivity = 1.5 mS cm ⁻¹ , pH 6.9)				
Petroleum	Petroleum wastewater (COD=1.6 g-O ₂ L ⁻¹ , conductivity = 4.6 mS cm ⁻¹ , pH 7.5, [Cl ⁻] = 22.4 g L ⁻¹)	AO	Cathode: Ti (15 cm ²) Anode: Ti/Pt, BDD (10 cm ²)	Anode materials, current density, electrolyte concentration, agitation rate, COD removal, energy consumption	(Rocha et al., 2012)
Petroleum	Petroleum wastewater (COD=0.7 g-O ₂ L ⁻¹ , conductivity = 5.1 mS cm ⁻¹ , pH 7.8, [Cl ⁻] = 1.6 g L ⁻¹)	AO	Cathode: Ti (63.5 cm ²) Anode: Ti/IrO ₂ -Ta ₂ O ₅ , Ti/PbO ₂ , BDD (63.5 cm ²)	Anode materials, current density, COD removal, energy consumption	(da Silva et al., 2013)
Paper mill	Paper mill industry wastewater (COD = 5-6 g-O ₂ L ⁻¹ , pH 8, conductivity = 7 mS cm ⁻¹)	AO	Cathode: stainless steel (682 cm ²) Anode: PbO ₂ (682 cm ²)	Current density, pH, electrolyte concentration, color and COD removal, energy consumption	(El-ashtoukhy et al., 2009)

Paper mill	Paper mill industry wastewater (COD = 130 g-O ₂ L ⁻¹ , pH 12.8-13, conductivity = 0.2 mS cm ⁻¹)	AO	Cathode: stainless steel (141 cm ²) Anode: Ti/RuO ₂ (11 cm ²)	Current density, flow rate, color and COD removal, current efficiency	(Chanworrawoot, 2012)
Paper mill	Paper mill industry wastewater (COD = 0.3 g-O ₂ L ⁻¹ , pH 6.8, conductivity = 1.1 mS cm ⁻¹)	AO	Cathode: stainless steel (50 cm ²) Anode: BDD, Ti/Ti _{0.5} Ru _{0.45} Sn _{0.05} O ₂ (50 cm ²)	Current density, flow rate, temperature, pH, NaCl concentration, energy consumption	(Klidi et al., 2018)

Table 5. Literature review of RO concentrate treatment by electrochemical processes.

Matrix	Process	Electrode materials	Main parameters tested	Reference
RO concentrate (Conductivity = 22.3 mS cm ⁻¹ , COD = 120-150 mg-O ₂ L ⁻¹ , pH 7.2-7.6)	EF	Cathode: graphite felt (120 cm ²) Anode: Pt wire	[Fe ²⁺], cathodic potential, [H ₂ O ₂], pH	(Zhou et al., 2012)
RO concentrate from a tertiary steel WWTP (Conductivity = 22.3 mS cm ⁻¹ , [Cl ⁻] = 6.4 g L ⁻¹ , COD = 120-190 mg-O ₂ L ⁻¹ , pH 8.3)	AO	Cathode: stainless steel (3 cm ²) Anode: BDD, Ti/IrO ₂ -Ta ₂ O ₅ , Ti/IrO ₂ -RuO ₂) (3 cm ²)	Anode materials, pH, COD removal, energy consumption	(Zhou et al., 2011)
RO concentrate from tertiary WWTP (Conductivity = 5 mS cm ⁻¹ , [Cl ⁻] = 1.2 g L ⁻¹ , COD = 170 mg-O ₂ L ⁻¹ , pH 8.0)	AO	Cathode: stainless steel (projected surface area: 24 cm ²) Anode: Ti/IrO ₂ -Ta ₂ O ₅ , Ti/RuO ₂ -IrO ₂ , Ti/Pt-IrO ₂ , Ti/PbO ₂ , Ti/SnO ₂ -Sb	Anode materials, COD and inorganic removal, by-products evolution	(Bagastyo et al., 2011)

		(projected surface area: 24 cm ²)		
RO concentrate from tertiary WWTP spiked with pharmaceuticals (Conductivity = 4.2 mS cm ⁻¹ , [Cl ⁻] = 1.5 g L ⁻¹ , DOC = 57 mg-C L ⁻¹ , pH 7.5)	AO	Cathode: stainless steel (projected surface area: 24 cm ²) Anode: Ti/RuO ₂ -IrO ₂ (projected surface area: 24 cm ²)	DOC, pharmaceuticals and inorganic evolution, by-products and toxicity evolution	(Radjenovic, Bagastyo, et al., 2011)
RO concentrate from tertiary WWTP spiked with pharmaceuticals (Conductivity = 4.3 mS cm ⁻¹ , [Cl ⁻] = 1.6 g L ⁻¹ , DOC = 57 mg-C L ⁻¹ , pH 7.7)	AO	Cathode: stainless steel (projected surface area: 24 cm ²) Anode: Ti/Ru _{0.7} -Ir _{0.3} O ₂ , Ti/SnO ₂ -Sb (projected surface area: 24 cm ²)	Anode materials, current density, pharmaceutical removal, by-products and toxicity evolution	(Radjenovic, Escher, et al., 2011)

Electrodialysed RO concentrate from tertiary WWTP (Conductivity = 6.0 mS cm ⁻¹ , [Cl ⁻] = 1.4 g L ⁻¹ , COD = 136 mg-O ₂ L ⁻¹ , pH 7.7)	AO	Cathode: stainless steel (41 cm ²) Anode: BDD (41 cm ²)	pH, COD and DOC removal, by-products evolution, energy consumption	(Bagastyo et al., 2012)
Electrodialysed RO concentrate from tertiary WWTP (Conductivity = 1.4 mS cm ⁻¹ , [Cl ⁻] = 0.14 g L ⁻¹ , COD = 145 mg-O ₂ L ⁻¹ , pH 6.8)	AO	Cathode: stainless steel (projected surface area: 24 cm ²) Anode: BDD (41 cm ²) and Ti/Pt-IrO ₂ , Ti/SnO ₂ -Sb (projected surface area: 24 cm ²)	Anode materials, COD removal, salts evolution, by-products evolution	(Bagastyo et al., 2013)
RO concentrate from tertiary WWTP (Conductivity = 6 mS cm ⁻¹ , [Cl ⁻] = 1.4 g L ⁻¹ , COD = 140 mg-O ₂ L ⁻¹ , pH 7.7)	AO	Cathode: BDD (41 cm ²) Anode: BDD (41 cm ²)	Reactor configuration, COD removal, energy consumption, by-products, ecotoxicity	(Bagastyo et al., 2014)

RO concentrate from tertiary WWTP (Conductivity = 2.3-5.1 mS cm ⁻¹ , [Cl ⁻] = 0.3-0.6 g L ⁻¹ , DOC = 14-30 mg-C L ⁻¹ , pH 7.6-7.9)	AO	Cathode: stainless steel (70 cm ²) Anode: BDD (70 cm ²)	Initial RO concentrate matrix, Pollutant removal, inorganic and organic by-products evolution	(Pérez et al., 2010)
RO concentrate mixed domestic and textile WWTP (Conductivity = 4.0 mS cm ⁻¹ , [Cl ⁻] = 0.6 g L ⁻¹ , COD = 5 mg-O ₂ L ⁻¹ , pH 8.2)	AO	Cathode: Ti/RuO ₂ (60 cm ²) Anode: BDD (60 cm ²)	COD and inorganic species evolution	(Van Hege et al., 2002)
RO concentrate pharmaceuticals WWTP (Conductivity = 2.7 mS cm ⁻¹ , [Cl ⁻] = 0.5-0.8 g L ⁻¹ , DOC = 17 mg-C L ⁻¹ , pH 7.5)	AO	Cathode: stainless steel (70 cm ²) Anode: BDD (70 cm ²)	Pollutant removal	(Urriaga et al., 2013)
RO concentrate from WWTP spiked with pharmaceuticals (Conductivity = 2.6 mS cm ⁻¹ , [Cl ⁻] = 0.15 g L ⁻¹ , TOC = 17 mg-C L ⁻¹ , pH 8.4)	AO	Cathode: Pt Anode: BDD (12 cm ²)	Kinetics of degradation and mineralization, by-products and pathway, ecotoxicity	(Lütke Eversloh et al., 2014)

RO concentrate from WWTP (Conductivity = 17 mS cm ⁻¹ , [Cl ⁻] = 1.5 g L ⁻¹ , DOC = 272 mg-C L ⁻¹ , pH 8.3)	AO	Cathode: Ti (150 cm ²) Anode: Ti/PbO ₂ (300 cm ²)	COD and inorganic (chloride, total nitrogen) removal, e energy consumption	(Wang et al., 2018)
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Table 6. Literature review of raw landfill leachate treatment by electrochemical processes.

Matrix	Process	Electrode materials	Main parameters tested	Reference
Landfill leachate (pH = 8.3, conductivity = 41.3 mS cm ⁻¹ , COD = 7.2 g-O ₂ L ⁻¹ , BOD ₅ /COD = 0.03, [Cl ⁻] = 6 g L ⁻¹ , Fe = 0.07 g L ⁻¹)	EF	Cathode: graphite (25 cm ²) Anode: Ti/TiO ₂ (25 cm ²)	Aeration, catalyst reusability, COD removal	(Baiju et al., 2018)
Landfill leachate (pH = 8.2, TDS = 9.5 g L ⁻¹ , COD = 6.2 g-O ₂ L ⁻¹ , BOD ₅ /COD = 0.03)	EF	Cathode: graphite Anode: graphite	Voltage, pH, catalyst reusability, COD removal	(Sruthi et al., 2018)
Landfill leachate (pH = 8.3, conductivity = 12 mS cm ⁻¹ , COD = 1.2 g-O ₂ L ⁻¹ , [Cl ⁻] = 1.6 g L ⁻¹ , [NH ₃] = 0.38 g L ⁻¹)	AO	Cathode: steel Anode: Ti/SnO ₂ , Ti/PbO ₂ (24 cm ²)	pH, COD and ammonium evolution	(Cossu et al., 1998)
Landfill leachate (pH = 5.8-6.8, COD = 53.3 g-O ₂ L ⁻¹ , [Cl ⁻] = 2.6-4.5 g L ⁻¹ , [SO ₄ ²⁻] = 0.4-0.74 g L ⁻¹ , [NH ₃] = 0.9-1.3 g L ⁻¹)	AO	Cathode: stainless steel (1.5 m ²) Anode: Ti/Pt (0.85 m ²)	pH, COD and ions (ammonia, nitrate, phosphorus, calcium) evolution, by products formation, energy consumption	(Vlyssides et al., 2001)

Landfill leachate (pH = 8.2, conductivity = 9.8 mS cm ⁻¹ , COD = 0.8 g-O ₂ L ⁻¹ , BOD ₅ /COD < 0.1, [Cl ⁻] = 1.8 g L ⁻¹)	AO	Cathode: stainless steel Anode: Ti/PbO ₂ (48 cm ²)	pH, current density, COD removal, energy consumption	(Panizza et al., 2010)
Landfill leachate (pH = 8.3, conductivity = 12.8 mS cm ⁻¹ , COD = 4.4 g-O ₂ L ⁻¹ , BOD ₅ /COD = 0.14, [Cl ⁻] = 3.2 g L ⁻¹ , [SO ₄ ²⁻] = 0.04 g L ⁻¹ , [NH ₃] = 1.9 g L ⁻¹)	AO	Cathode: stainless steel (70 cm ²) Anode: BDD (70 cm ²)	COD and ammonium removal	(Cabeza, Urtiaga, & Ortiz, 2007)
Landfill leachate (pH = 7.8, conductivity = 27.6 mS cm ⁻¹ , COD = 4.2 g-O ₂ L ⁻¹ , BOD ₅ /COD = 0.09, [Cl ⁻] = 3.2 g L ⁻¹ , [SO ₄ ²⁻] = 1.9 g L ⁻¹ , [NH ₃] = 2.3 g L ⁻¹)	AO	Cathode: Ta (10 cm ²) Anode: BDD (10 cm ²)	Current density, pH, COD, biodegradability, ammonia and nitrate evolution, energy consumption	(Fudala-Ksiazek et al., 2018)
Landfill leachate (pH = 7.9, COD = 0.75 g-O ₂ L ⁻¹ , [Cl ⁻] = 1.2 g L ⁻¹ , [SO ₄ ²⁻] = 0.25 g L ⁻¹ , [NH ₃] = 0.22 g L ⁻¹)	AO	Cathode: graphite (30 cm ²) Anode: graphite/PbO ₂ (30 cm ²)	COD and DOC removal, inorganic evolution (sulfate, chloride, nitrate, bicarbonate)	(Mandal et al., 2020)

Landfill leachate (pH = 8.0, COD = 2.7 g-O ₂ L ⁻¹ , [Cl ⁻] = 2.9 g L ⁻¹ , [NH ₃] = 2.8 g L ⁻¹ , Fe = 0.005 g L ⁻¹)	EF/AO	Cathode: Ti (projected surface area: 15 × 10 cm ²) Anode: Ti/RuO ₂ -IrO ₂ -SnO ₂ -TiO ₂ (projected surface area: 15 × 10 cm ²)	pH, Hydraulic retention time, inter-electrode gap, COD removal, by-products evolution	(Zhang et al., 2012)
Landfill leachate (pH = 8.0-9.0, TOC = 1.6-3.1 g-C L ⁻¹ , [Cl ⁻] = 5-6.2 g L ⁻¹ , [SO ₄ ²⁻] = 0.06-0.24 g L ⁻¹ , [NH ₃] = 1.9-3.2 g L ⁻¹)	EF/AO	Cathode: carbon felt (140 cm ²) Anode: Pt, BDD (24 cm ²)	Current density, TOC removal efficiency	(Oturán et al., 2015)
Landfill leachate (pH = 7.8, COD = 1.0 g-O ₂ L ⁻¹ , [Cl ⁻] = 21 g L ⁻¹ , [NH ₃] = 0.44 g L ⁻¹)	EF/AO	Cathode: hydrothermal carbonization biochar Anode: iron filings in the 3D Fe/C granules electrode system	Voltage, COD and TOC removal, ammonia evolution, reusability of electrode	(Yu et al., 2020)

Landfill leachate (pH = 8.5, conductivity = 17.8 mS cm ⁻¹ , COD = 1.8 g-O ₂ L ⁻¹ , [Cl ⁻] = 2.4 g L ⁻¹ , [NH ₃] = 1.7 g L ⁻¹)	EC/AO	Cathode: graphite Anode: Ti/RuO ₂ -IrO ₂ , Ti/RuO ₂ -IrO ₂ -mZVI (macroscopic zero-valent iron) (15 cm ²)	Anode material, voltage, TOC and COD removal, ammonia evolution,	(Sun et al., 2020)
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Table 7. Literature review of acid mine drainage treatment by electrochemical processes.

Matrix	Process	Electrode materials	Main parameters tested	Reference
Acid mine drainage solution	EC	Cathode: sulphide/graphide block Anode: Fe	pH, redox potential, metal elements concentrations	(Shelp et al., 1996a)
Acid mine drainage solution (pH = 3.2, $[\text{SO}_4^{2-}] = 0.45 \text{ g L}^{-1}$)	EC	Cathode: sulphide/graphide block Anode: Al, Zn	Anode materials, pH, redox potential, metal elements concentrations	(Shelp et al., 1996b)
Acid mine drainage solution (pH = 2.6, $[\text{SO}_4^{2-}] = 12 \text{ g L}^{-1}$, $[\text{Fe}] = 0.61 \text{ g L}^{-1}$)	EC	Cathode: Fe followed by Al Anode: sacrificial Fe followed by Al	Color, turbidity and inorganic removal	(Orescanin & Kollar, 2012)
Acid mine drainage solution (pH = 2.7, $[\text{SO}_4^{2-}] = 11 \text{ g L}^{-1}$, $[\text{Fe}] = 0.92 \text{ g L}^{-1}$)	EC	Cathode: Fe followed by Al Anode: sacrificial Fe followed by Al	Color, turbidity and inorganic removal, toxicity evolution	(Radić et al., 2014)

Acid mine drainage solution (pH = 2.9)	EC	Cathode: stainless steel ($7 \times 5 \text{ cm}^2$) Anode: Fe, Al ($7 \times 5 \text{ cm}^2$)	Anode materials, current density, Cd removal	(Nariyan et al., 2016)
Acid mine drainage solution (pH = 2.7, $[\text{SO}_4^{2-}] = 13 \text{ g L}^{-1}$, $[\text{Fe}] = 0.77 \text{ g L}^{-1}$)	EC	Cathode: stainless steel ($7 \times 5 \text{ cm}^2$) Anode: Fe, Al ($7 \times 5 \text{ cm}^2$)	Anode materials, current density, inorganic (metals and sulfate) removal	(Nariyan et al., 2017)
Acid mine drainage solution (pH = 2.7, $[\text{SO}_4^{2-}] = 13 \text{ g L}^{-1}$, $[\text{Fe}] = 0.77 \text{ g L}^{-1}$)	EC	Cathode: stainless steel ($7 \times 5 \text{ cm}^2$) Anode: Fe, Al ($7 \times 5 \text{ cm}^2$)	Anode materials, current density, electrode connection configuration (mono-/bi-polar), energy consumption	(Nariyan et al., 2018)
Sulfate/nitrate/chloride/copper/nickel/zinc/ammonia in acid mine drainage model solution ($[\text{SO}_4^{2-}] = 3 \text{ g L}^{-1}$)	EC	Cathode: Fe (168 cm^2) Anode: Fe (168 cm^2)	pH, current density, sulfate removal, flow mode (batch/continuous)	(Mamelkina et al., 2017)

Sulfate/nitrate/chloride/copper/ nickel/zinc/ammonia in acid mine drainage model solution ($[\text{SO}_4^{2-}] = 3 \text{ g L}^{-1}$)	EC	Cathode: Fe (168 cm^2) Anode: Fe (168 cm^2)	pH, flow mode (batch/continuous), inorganic (ammonia, nitrate, copper, nickel, zinc, sulfate) removal	(Mamelkina et al., 2018)
Sulfate/nitrate/chloride/copper/ nickel/zinc/ammonia in acid mine drainage model solution ($[\text{SO}_4^{2-}] = 1\text{-}3 \text{ g L}^{-1}$)	EC	Cathode: Fe (168 cm^2) Anode: Fe, Al (168 cm^2)	current density, pH, electrode materials, sulfate removal,	(Mamelkina, Tuunila, et al., 2019)
Iron in acid mine drainage model solution ($[\text{Fe}] = 0.5 \text{ g L}^{-1}$)	EC	Cathode: Fe, Al (26 cm^2) Anode: Fe, Al (26 cm^2)	Current density, electrode materials, iron removal, energy consumption	(Foudhaili et al., 2019)
Real acid mine drainage solutions ($\text{pH} = 6.9\text{-}7.5$, $[\text{SO}_4^{2-}] = 1.3\text{-}2.9 \text{ g L}^{-1}$)	EC	Cathode: Fe (26 cm^2) Anode: Fe (26 cm^2)	Toxicity assays	(Foudhaili, Jaidi, et al., 2020)
Iron in acid mine drainage model solution ($\text{pH} = 2.4\text{-}2.6$, $[\text{Fe}] = 0.47\text{-}0.52 \text{ g L}^{-1}$, $[\text{SO}_4^{2-}] = 1.3\text{-}4.9 \text{ g L}^{-1}$) and real acid mine drainage solution ($\text{pH} = 2.4\text{-}$	EC	Cathode: Fe ($11 \times 11 \text{ cm}^2$) Anode: Fe ($11 \times 11 \text{ cm}^2$)	Current density, Sulfate concentration	(Foudhaili, Lefebvre, et al., 2020)

2.6, [Fe] = 0.04-0.07 mg L ⁻¹ , [SO ₄ ²⁻] = 1.3-2.9 g L ⁻¹)				
Acid mine drainage solution (pH = 3.0, [SO ₄ ²⁻] = 1.9 g L ⁻¹ , [Fe] = 0.45 g L ⁻¹)	Sono-EC	Cathode: stainless steel Anode: Mg	Iron and copper removal	(Morgan et al., 2017)
Acid mine drainage solution (pH = 3.0-3.9, [Fe] = 1.7-2.3 t yr ⁻¹)	Sono-EC	Cathode: stainless steel Anode: Mg	Reactor design, flow rate, inorganic removal, energy consumption	(Rose et al., 2019)
Acid mine drainage solution	AO	Cathode: platinum, copper, graphite and carbon felt, with areas of 4.5, 6.4, 6.8 and 6.9 cm ² , respectively Anode: Ti/IrO ₂ (7.2 cm ²)	pH, current	(Bunce et al., 2001)
Iron/Sulfate in acid mine drainage model solution (pH = 1.9, [FeSO ₄] = 0.56 g L ⁻¹)	AO	Cathode: reticulated vitreous carbon Anode: Ti/IrO ₂ (7.2 cm ²)	Current density	(Chartrand & Bunce, 2003)

Iron/Arsenite spiked in acid mine drainage model solution (pH = 2.0-2.1, Fe(III) = 0.26 g L ⁻¹ , Fe(II) = 0.30 g L ⁻¹)	AO	Cathode: stainless steel (10 cm ²) Anode: Ti/IrO ₂ (7 cm ²)	Reactor design	(Wang et al., 2003)
Acid mine drainage solution (pH = 2.4, [SO ₄ ²⁻] = 1.3 g L ⁻¹ , [Fe] = 0.23 g L ⁻¹)	AO	Cathode: Ti Anode: Ti, graphite, BDD	Anode materials, current density	(Park et al., 2015)
Sulfate/nitrate/chloride/cyanide in acid mine drainage model solution ([SO ₄ ²⁻] = 1 g L ⁻¹)	EC, AO, EC/AO	Cathode: Fe, Al, C, Ti Anode: Fe, Al, C, Ti	Electrode materials, pH, current density, inorganic (cyanide, sulfate, nitrate) removal	(Mamelkina, Vehmaanpera, et al., 2019)

Table 8. Literature review of WWTP sludge treatment by electrochemical processes (TS: total solids; DS: dry sludge; MLSS: mixed liquid suspension solids; MPN: most probable number).

Matrix	Process	Electrode materials	Main parameters tested	Reference
Activated sludge from municipal WWTP (pH 7.0, soluble COD = 0.09 g-O ₂ L ⁻¹ , TS = 15.9 g L ⁻¹)	EC	Cathode: Fe (11.5 × 6 cm ²) Anode: Fe (11.5 × 6 cm ²)	pH, current density, electrolyte concentration, COD and TOC removal, sludge disintegration	(Sari Erkan & Onkal Engin, 2020)
Anaerobic sludge from conventional WWTP (pH 6.8, COD = 22.5 g-O ₂ L ⁻¹ , DS = 23.3 g L ⁻¹ , Total coliforms = 7.9 × 10 ⁶ MPN)	Fered-Fenton	Cathode: graphite (10 × 6 cm ²) Anode: graphite (10 × 6 cm ²)	Voltage, COD and total coliforms removal, zeta potential, dewaterability/filterability	(Masihi & Gholikandi, 2018)
Anaerobic sludge from poultry farm (pH 7.8-8.2, COD = 26.2 g-O ₂ L ⁻¹ , TSS	Electrochemical peroxidation/EF	Cathode: steel (5 × 6.5 cm ²)	Current density, pH, TSS & COD & TOC removal, inorganic evolution, cost evaluation	(Olvera-Vargas et al., 2019)

= 31.3 g L ⁻¹ , TDS = 9.4 g L ⁻¹ , Total coliforms = 2.4×10^3 CFU mL ⁻¹)		Anode: steel (5×6.5 cm ²)		
Activated sludge from municipal WWTP (pH 6.7-7.0, COD = 17.4-19.0 g-O ₂ L ⁻¹ , TS = 8-18.2 g L ⁻¹)	AO	Cathode: Ti/RuO ₂ (70 cm ²) Anode: Ti/RuO ₂ (70 cm ²)	Current density, pH, type of salt, cost evaluation	(Song et al., 2010)
Municipal wastewater sludge	AO	Cathode: Ti (908 cm ²) Anode: Ti/RuO ₂ (504 cm ²)	Current density, electrolyte concentration	(Bureau et al., 2012)
Activated sludge from paper mill WWTP (pH 7.8-8.2, COD = 0.55 g-O ₂ L ⁻¹ , TS = 16.3 mg L ⁻¹ , Total coliforms = 2.8×10^5 CFU g-DS ⁻¹)	AO	Cathode: Ti (908 cm ²) Anode: Ti/RuO ₂ (504 cm ²)	Dewaterability, coliforms removal, evolution of organics and inorganics, cost evaluation	(Drogui et al., 2013)

Anaerobic sludge from municipal WWTP (pH 7.2-7.4, TS = 2.9-3.2%)	AO	Cathode: stainless steel Anode: Ti	Electro-migration of organic matter and metals, dewaterability	(Tuan & Sillanpaa, 2010)
Anaerobic sludge from municipal WWTP (pH 7.2-7.4, TS = 2.9-3.2%)	AO	Cathode: stainless steel Anode: Ti	Pressure, voltage, electro-migration of organic matter and metals, dewaterability	(Tuan & Sillanpaa, 2020)
Sludge from municipal WWTP (pH 6.4-6.6, TS = 32.5-34.3 g L ⁻¹)	AO	Cathode: Ti/PbO ₂ (10 × 10 cm ²) Anode: Ti/PbO ₂ (10 × 10 cm ²)	Supporting electrolyte, NaCl dosage, dewaterability, cost evaluation	(Xiao et al., 2019)
Activated sludge from municipal WWTP (pH 6.8-6.9, soluble COD = 0.15-0.33 g-O ₂ L ⁻¹ , TS = 9.1-13.7 g L ⁻¹)	AO	Cathode: Ti/RuO ₂ (44 cm ²) Anode: Ti/RuO ₂ (44 cm ²)	pH, current density	(Heng & Isa, 2014)

Activated sludge from municipal WWTP (pH 6.7, soluble COD = 0.10 g-O ₂ L ⁻¹ , TSS = 6.3 g L ⁻¹)	AO	Cathode: Ti/RuO ₂ (10 × 7 cm ²) Anode: Ti/RuO ₂ (10 × 7 cm ²)	Voltage, dewaterability	(Zhen et al., 2013)
Activated sludge from municipal WWTP	AO/EC	Cathode: Ti/RuO ₂ and Al and Fe (180 cm ²) Anode: Ti/RuO ₂ and Al and Fe (180 cm ²)	Voltage, COD removal, dewaterability	(Gharibi et al., 2013)
Activated sludge from municipal WWTP (pH 6.7-6.8, TSS = 16-37 g L ⁻¹)	AO/EC	Cathode: Ti (12 × 6 cm ²) and Fe (16 cm ²) Anode: Ti/RuO ₂ IrO ₂ (12 × 6	Voltage, dewaterability	(Li et al., 2016)

		cm ²) and Fe (16 cm ²)		
Activated sludge from municipal WWTP (pH 7.0, soluble COD = 0.14 g-O ₂ L ⁻¹ , MLSS = 2.5 g L ⁻¹ , VSS/TSS = 0.68)	AO/Fered-Fenton	Cathode: Ti (10 × 10 cm ²) Anode: Ti/RuO ₂ -IrO ₂ (10 × 10 cm ²)	pH, current density, dewaterability/filterability, cost evaluation	(Cai et al., 2019)
Sludge from thickening tank in municipal WWTP (pH 6.7-6.9, TOC = 2.7 g g-DS ⁻¹ , VSS/TSS = 0.46-0.52)	AO, AO/EF	Cathode: Ti (8 × 7 cm ²) Anode: Ti/RuO ₂ -IrO ₂ (8 × 7 cm ²)	Voltage, TOC removal, dewaterability/filterability, cost evaluation	(Hu et al., 2018)
Activated sludge from municipal WWTP (pH 6.8, soluble COD = 0.14 g-O ₂ L ⁻¹ , TSS = 24 g L ⁻¹)	AO, AO/EF	Cathode: Ti, active carbon fiber (10 × 7 cm ²) Anode: Ti/RuO ₂ (10 × 7 cm ²)	Current density, pH, dewaterability	(Chen et al., 2019)

Table 9. Literature review of soil washing/flushing solutions treatment by electrochemical processes (BCD: beta-cyclodextrin; DSA: dimensionally stable anode; HPCD: hydroxypropyl-beta-cyclodextrin; SDS: sodium dodecyl sulfate; Tween 80: polyoxyethylene (20) sorbitan monooleate; MMO: metal mixed-oxide).

Pollutant	Extracting agent	Process	Electrode materials	Main parameters tested	Reference
Trinitrotoluene (TNT) (0.2 mM)	BCD (1 mM)	Soil washing/EF	Cathode: carbon felt (60 cm ²) Anode: Pt grid (3 cm diameter, 4.5 cm height)	Current density	(Murati et al., 2009)
Phenanthrene (17 mg L ⁻¹)	Tween 80 (0.75 g L ⁻¹) and HPCD (10 g L ⁻¹)	Soil washing/EF	Cathode: carbon felt (150 cm ²) Anode: Pt grid (3 cm diameter, 5 cm height)	[Fe ²⁺], current density, biodegradability and toxicity of solution	(Mousset et al., 2014a)

Phenanthrene (16 mg L ⁻¹)	HPCD (9 g L ⁻¹)	Soil washing/EF	Cathode: carbon felt (10 cm ²) Anode: Pt grid (3 cm diameter, 5 cm height), Ti/RuO ₂ -IrO ₂ (40 cm ²), BDD (40 cm ²)	Anode materials, current density, biodegradability and toxicity of solution	(Mousset et al., 2014b)
Pentachlorophenol (PCP) (0.77 mM)	HPCD (5 mM)	Soil washing/EF	Cathode: carbon felt (10 cm ²) Anode: Pt sheet (1 cm ²)	Current density, toxicity of solution	(Hanna et al., 2005)
16 polycyclic aromatic hydrocarbons (PAHs) (1090 mg kg ⁻¹)	Tween 80 (7.5 g L ⁻¹) or HPCD (7.5 g L ⁻¹)	Soil washing/EF	Cathode: carbon felt (150 cm ²)	Number of SW cycles, pH, Soil respirometry	(Mousset et al., 2016)

			Anode: Pt grid (3 cm diameter, 5 cm height)		
Total petroleum hydrocarbons (TPH) (3900-6100 mg kg ⁻¹)	Tween 80 (11 g L ⁻¹)	Soil flushing/EF	Cathode: carbon felt (150 cm ²) Anode: BDD (40 cm ²)	pH, biodegradability and toxicity of solution	(Huguenot et al., 2015)
Phenanthrene (500 mg kg ⁻¹)	SDS or Tween 80 (1 g L ⁻¹)	Soil washing/AO	Cathode: stainless steel (78 cm ²) Anode: DSA (78 cm ²), BDD (78 cm ²)	anode materials, pH, conductivity	(Sáez et al., 2010)
Oxyfluorfen (100 mg kg ⁻¹)	SDS (0.1-5 g L ⁻¹)	Soil washing/AO	Cathode: stainless steel (78 cm ²)	Concentration of extracting agent, turbidity, zeta	(dos Santos et al., 2016)

			Anode: DSA (78 cm ²), BDD (78 cm ²)	potential, mean particle size, ions evolution	
Oxyfluorfen (100 mg kg ⁻¹)	SDS (0.1-5 g L ⁻¹)	Soil washing/Ultrasound-AO	Cathode: stainless steel (70 cm ²) Anode: BDD (70 cm ²)	Concentration of extracting agent, turbidity, zeta potential, mean particle size, ions evolution	(Vieira Dos Santos et al., 2017)
Petroleum (COD: 487 and 2943 mg-O ₂ L ⁻¹ ; TOC 359 and 1298 mg-C L ⁻¹)	SDS (0.1-5 g L ⁻¹)	Soil washing/(Ultrasound/UV)-AO	Cathode: stainless steel (78 cm ²) Anode: BDD (78 cm ²)	initial organic load, concentration of extracting agent, turbidity, zeta potential, mean particle size, ions evolution	(dos Santos et al., 2017)
Phenanthrene (25 mg L ⁻¹) and humic acid (10 mg L ⁻¹)	Tween 80 (1.31 g L ⁻¹)	Soil washing/AO	Cathode: stainless steel (24 cm ²)	current density, supporting electrolyte	(Trellu et al., 2017)

			Anode: BDD (24 cm ²)		
Phenanthrene (25 mg L ⁻¹) and humic acid (10 mg L ⁻¹)	Tween 80 (1.31 g L ⁻¹)	Soil washing/AO	Cathode: stainless steel (28 cm ²) Anode: BDD (28 cm ²)	Ecotoxicity, biodegradability, carboxylic acids	(Trellu et al., 2016)
Phenanthrene (456 mg kg ⁻¹)	HPCD (10-40 g L ⁻¹)	Soil washing/AO	Cathode: graphite (52 cm ²) Anode: graphite (52 cm ²)	concentration of extracting agent	(Gómez et al., 2010)
Atrazine (100 mg kg ⁻¹)	SDS (0.1 g L ⁻¹)	Soil washing/AO	Cathode: stainless steel (78 cm ²) Anode: BDD (78 cm ²)	COD and TOC removal, intermediates evolution, mean particle size, zeta potential	(dos Santos et al. 2015a) (elec comm)

Atrazine (100 mg kg ⁻¹)	SDS (0.1-5 g L ⁻¹)	Soil washing/AO	Cathode: stainless steel (78 cm ²) Anode: BDD (78 cm ²)	COD and TOC removal, sulfate and intermediates evolution, mean particle size, zeta potential	(dos Santos et al. 2015b) (JHM)
Lindane (100 mg kg ⁻¹)	SDS (0.1-20 g L ⁻¹)	Soil washing/AO	Cathode: stainless steel (78 cm ²) Anode: BDD (78 cm ²)	COD and TOC removal, sulfate evolution, mean particle size	(Munoz-Morales et al. 2017)
Clopyralid (100 mg kg ⁻¹)	Deionized water	Soil washing/AO	Cathode: stainless steel (78 cm ²) Anode: BDD (78 cm ²)	Clopyralid removal, intermediates evolution	(Rodriguez et al. 2018)
Pendimethalin (100 mg kg ⁻¹)	SDS (5-50 g L ⁻¹)	Soil washing/AO	Cathode: stainless steel (78 cm ²)	TOC removal, sulfate evolution, mean particle size	(Almazan-Sanchez et al. 2017)

			Anode: BDD (78 cm ²)		
Clopyralid (100 mg kg ⁻¹)	Synthetic wastewater	Soil washing/AO	Cathode: BDD (78 cm ²) Anode: BDD (78 cm ²)	TOC removal, intermediate evolution, energy consumption	(Cotillas et al. 2017)
Lontrel [®] (commercial herbicide based on 72% w/w of clopyralid) (1000 mg kg ⁻¹)	Deionized water	Soil washing/AO	Cathode: stainless steel (78 cm ²) Anode: BDD, Ru-MMO, Ir-MMO, carbon felt (78 cm ²)	TOC removal, intermediates evolution, toxicity and biodegradability evolution	(Carboneras et al. 2018)

Figure 1. Classification of the electrode materials used in AO.

Figure 2. Main steps involved in the selection of the most suitable treatment strategy.

Figure 3. CFD simulation for a) the turbulent intensity distribution in a vertical-flow tubular electrochemical reactor (I) and a concentric tubular electrochemical reactor (II) for AO of phenol wastewater, b) normalized current density distribution in a rotating cylinder electrode (RCE) reactor for EC, I) RCE surface and II) working electrode plates and c) concentration profile of Fe^{2+} around the cathode during EF process at (I) $t = 0$ s and (II) $t = 400$ s.

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Figure 4. Reactor designs for EAOPs, a) Filter press reactor, b) fluidized bed reactor, c) Rotating electrode reactor and d) fixed bed reactor (Adapted with permission from: (Naje et al., 2016; Olvera-Vargas et al., 2015; Shen et al., 2017; Tezcan Un & Aytac, 2013)). Copyright Elsevier.

Figure 5. Schematic representation of two different ways of $\bullet\text{OH}$ oxidative degradation of hydrophobic organic pollutant in the presence of (a) surfactant (Tween 80) or (b) cyclodextrin (HPCD) in aqueous solution. Adapted with permission from (Mousset et al., 2014a). Copyright Elsevier.

Figure captions

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