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## Electrochemical treatment of highly concentrated wastewater: A review of experimental and modeling approaches from lab- to full-scale

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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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## 18 **Abstract**

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

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

## 40 **Electrochemical wastewater treatment strategies**

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

47 *Electrochemical technologies: an overview*

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

63 *Anode materials*

64 In electrochemical processes, the electrode material is a determining factor, as electrode  
65 properties define the type of treatment and thus the chemical and electrochemical  
66 mechanisms that are involved. Anodes with a high  $\text{O}_2$  evolution overpotential have the  
67 capacity to produce  $\cdot\text{OH}$  on their surfaces ( $\text{M}(\cdot\text{OH})$ ) and are excellent materials for direct  
68 AO, while anodes with the ability to produce chlorine oxidative species from the  
69 oxidation of  $\text{Cl}^-$  are excellent choices for indirect AO of organic pollutants (Martínez-  
70 Huitle et al., 2015). Figure 1 shows the main anode materials typically used for AO. They  
71 have been classified according to their capacity to produce  $\text{M}(\cdot\text{OH})$ , which is linked to

72 their oxidative power. Two main classes can be distinguished: “active” (low O<sub>2</sub> evolution  
73 overpotential) and “non-active” (high O<sub>2</sub> evolution overpotential) anodes. Boron doped  
74 diamond electrodes (BDD) are the most powerful anode materials due to their outstanding  
75 properties, including a high O<sub>2</sub> evolution overpotential (2.8 V vs SHE), high chemical  
76 stability and durability (Brillas et al., 2011). Recently, ceramic sub-stoichiometric anodes  
77 (Ti<sub>4</sub>O<sub>7</sub>) have also demonstrated their remarkable performance in the  
78 degradation/mineralization of organic contaminants, showing similar properties as BDD  
79 (Ganiyu et al., 2016; Olvera-Vargas et al., 2018; Yang et al., 2020); yet, their application  
80 to wastewater treatment is still at the development stage.

81 In contrast, water oxidation on electrodes with a low oxygen evolution  
82 overpotential (“active anodes”) leads to O<sub>2</sub> with negligible formation of M(\*OH) and thus  
83 poor AO performance. Among them, dimensionally-stable anodes (DSA) are good  
84 electrocatalysts for the oxidation of Cl<sup>-</sup> into chlorine oxidants, making them the most  
85 convenient choice for chlorine-mediated oxidation (AO-Cl). The fundamentals and  
86 applications of EAOPs for wastewater treatment have been thoroughly documented in  
87 several review papers of high impact (Brillas & Martínez-Huitle, 2015; Chaplin, 2014;  
88 Feng et al., 2013; Moreira et al., 2017; Sirés et al., 2014).

#### 89 *Cathode material: Fenton-based methods*

90 Cathodes with the electrocatalytic capability of producing H<sub>2</sub>O<sub>2</sub> from the two-electron  
91 reduction of dissolved O<sub>2</sub> represent great options for electrochemical treatment, allowing  
92 the indirect oxidation of organics via the Fenton’s reaction (Brillas et al., 2009). During  
93 EF, the Fe<sup>2+</sup> catalyst for the Fenton’s reaction is continuously regenerated on the cathode  
94 surface through reduction of Fe<sup>3+</sup>. Iron ions are generally supplied as Fe<sup>2+</sup> salts (mainly  
95 FeSO<sub>4</sub>) in catalytic amount. Alternatively, the use of iron anodes can produce the catalyst  
96 in EF-like systems. However, continuous dissolution of a sacrificial anode generates

97 considerable amounts of undesirable Fe sludge. Various types of carbonaceous electrodes  
98 have been used in EF, including carbon felt, carbon cloth, carbon fibers, graphite,  
99 reticulated vitreous carbon (RVC), carbon nanotubes (CNT), carbon sponge, gas  
100 diffusion electrodes (GDE), mesoporous carbon and graphene-based electrodes. Carbon  
101 materials have the advantage of being inexpensive, durable and nontoxic. During the last  
102 two decades a great deal of work has been done on the development of carbon-based  
103 electrodes for EF applications. Excellent papers on the topic and a recent book are  
104 available for review (Babuponnusami & Muthukumar, 2014; Brillas, 2017; Brillas et al.,  
105 2009; Casado, 2019; Huong Le et al., 2017; Nidheesh & Gandhimathi, 2012). Fenton-  
106 based technologies can be substantially improved under UV irradiation, due to the  
107 contribution of different photolytic reactions (photolysis of H<sub>2</sub>O<sub>2</sub> and Fe-carboxylate  
108 complexes, as well as the photoexcitation of semiconductor electrodes). UV-based  
109 EAOPs include photoelectro-Fenton (PEF), solar photoelectro-Fenton (SPEF) and  
110 photoelectrocatalysis (PEC) (Brillas, 2017; Garcia-Segura & Brillas, 2017).

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

121 For the sake of simplicity, EAOPs are generally performed in undivided  
122 electrochemical reactors in which the anode and the cathode are contained in the same  
123 compartment (more details of this subject will be discussed in the next section). In this  
124 kind of reactor configuration, the performance of the process can be optimized when both  
125 the cathodic and the anodic processes contribute to the oxidative degradation of the  
126 organic pollutants. In this scenario, the coupled EF-AO system has shown great  
127 degradation/mineralization efficiencies when treating several organic pollutants and  
128 waste streams (Olvera-Vargas et al., under review; Oturan et al., 2015; Sopaj et al., 2016)  
129 In this “paired” process,  $\cdot\text{OH}$  are generated through two routes: i) by the homogeneous  
130 Fenton’s reaction (EF) promoted by  $\text{H}_2\text{O}_2$  cathodic formation in carbonaceous electrodes  
131 and; ii) by the discharge of water at powerful anodes like BDD and  $\text{Ti}_4\text{O}_7$  (AO). As  
132 mentioned above, greater amounts of  $\cdot\text{OH}$  are produced when the process is carried out  
133 under UV/vis irradiation (EF and PEC).

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

#### 140 ***Defining the treatment strategy***

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

146 the physicochemical characteristics of the effluent: amount of organic matter COD, BOD<sub>5</sub>  
147 and TOC), TSS, TDS, ionic species (inorganic ions and metals) and pH; ii) the goal of  
148 the treatment in terms of water quality targets and; iii) the properties of the treatment  
149 process to be applied. Once these three points have been addressed, the second step  
150 involves the evaluation and optimization of the process. Again, three main actions need  
151 to be taken: i) optimization of the process according to the main variables affecting the  
152 efficiency (current/potential, types of electrodes, pH, catalyst concentration in the case of  
153 EF, supporting electrolyte) (Brillas & Martínez-Huitle, 2015; Chaplin, 2014; Feng et al.,  
154 2013; Moreira et al., 2017; Sirés et al., 2014); ii) cost/efficiency evaluation and; iii) design  
155 of large-scale systems (engineering of the electrochemical reactors and electrodes). The  
156 optimization is generally done in small-scale setups and the data obtained from this  
157 evaluation will serve as the starting point for the development of a pilot. This strategy  
158 selection flow is illustrated in Figure 2. In the following sub-sections a discussion of the  
159 most relevant strategies that have been applied to the treatment of highly polluted waste  
160 streams by electrochemical methods is presented, focusing on EAOPs and EC. A  
161 compilation of representative works that have been considered is displayed in Table 1.

### 162 *Anodic oxidation*

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



171 been proposed by the German and American companies CONDIAS GmbH<sup>1</sup> and  
172 Advanced Diamond Technologies Inc.<sup>2</sup>, respectively. The main limitations of BDD  
173 technology are still the high price of the electrodes and their size (restricted to 1 m<sup>2</sup>), but  
174 as the market grows, larger electrodes are becoming available.

175 As an example of AO applied to high strength wastewater, municipal landfill  
176 leachate previously submitted to biological activated sludge was treated by AO-BDD in  
177 a pilot plant in which 250 L of leachate were treated (Anglada, Urriaga, et al., 2010). The  
178 “DiaCell” treatment unit consisted of three electrochemical reactors, each one containing  
179 five DiaCell sets, where each set comprised ten single cells (anode/cathode pairs). BDD  
180 were used as anodes, while the cathodes consisted of stainless steel (SS). The electrodes  
181 were circular with a surface area of 70 cm<sup>2</sup> and separated by 1 mm. The setup was  
182 operated in discontinuous mode at a flow rate of 300 L min<sup>-1</sup>. More than 70% of COD  
183 removal and 37% of NH<sub>4</sub><sup>+</sup> removal were achieved following 8 h of treatment at 450 mA  
184 m<sup>2</sup> (COD<sub>0</sub> = 3106 - 4057 mg L<sup>-1</sup> and [NH<sub>4</sub><sup>+</sup>]<sub>0</sub> = 1300 - 1355 mg L<sup>-1</sup>). It is important to  
185 mention that the leachate influent was filtered before electrochemical treatment, while no  
186 electrolyte was added because of the high conductivity of leachate (high Cl<sup>-</sup> content).

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

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

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

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

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

217 yielded a maximum of 22% of DOC removal with formation of halogenated intermediates  
218 when using a Ti/SnO<sub>2</sub>-Sb anode (Bagastyo et al., 2013). In contrast, the treatment with  
219 BDD enhanced the DOC removal to 41% with negligible formation of halogenated  
220 substances. Moreover, the mineralization yield (DOC abatement) was improved to 51%  
221 when the Cl<sup>-</sup> ions were previously removed by electrodialysis and substituted with SO<sub>4</sub><sup>2-</sup>  
222 .

### 223 *Electro-Fenton*

224 Because it makes use of inexpensive carbon cathodes, EF has been one of the most widely  
225 used EAOPs. However, one of its main limitations lies in its narrow operational pH range  
226 (optimal pH value of around 3), which restricts its large-scale application because of the  
227 need for pH adjustment before and after treatment. Thus, the number of reports of highly  
228 concentrated real wastewater treatment by AO surpasses that using EF. Research on EF  
229 has been mostly carried out on synthetic solutions containing moderate amounts of COD  
230 and TOC, with concentration ranging between 10 and 300 mg L<sup>-1</sup>. As mentioned above,  
231 BDD anodes have sometimes been combined with EF, significantly enhancing the  
232 mineralization performance (cf. Table 1). However, since EF relies on the cathodic  
233 production of H<sub>2</sub>O<sub>2</sub> and the homogenous Fenton's reaction, inert anodes, such as Pt, have  
234 also been used to a great extent (cf. Table 1). Following the same reasoning, despite their  
235 low mineralization capability, DSA electrodes arise as a very attractive option for EF,  
236 especially when only partial mineralization of refractory effluents is required (EF as part  
237 of a sequential treatment process). In this way, •OH production is ensured by the electro-  
238 Fenton reaction (Olvera-Vargas et al., under review; Sopaj et al., 2016), while the process  
239 benefits from the commercial advantages of DSA. In addition, if Cl<sup>-</sup> ions are present in  
240 the target effluent, the performance can be improved by indirect chlorine oxidation (AO-  
241 Cl).

242 For example, landfill leachates were treated by means of EF using an undivided  
243 electrochemical reactor operated in batch mode (Oturán et al., 2015). Pt or BDD were  
244 used as anodes, while high surface area 3D carbon felt served as the cathode. The initial  
245 total organic carbon concentration of the leachates ranged from 1600 to 3100 mg L<sup>-1</sup>,  
246 containing several recalcitrant organic pollutants, such as polycyclic aromatic  
247 hydrocarbons (PAHs), humic substances, volatile organic compounds (VOCs), organo-  
248 halogenated compounds (OCHs), alkylphenols, pesticides and pharmaceuticals. 93.2% of  
249 TOC removal was achieved following 18 h of treatment by EF-BDD at 1 A of current,  
250 constant air flow at 1 L min<sup>-1</sup> and room temperature. The use of a Pt anode (EF-Pt) yielded  
251 78.2% of TOC removal under the same experimental conditions. In both cases, all the  
252 identified organic contaminants were almost totally degraded during treatment. It is  
253 important to mention that the pH of the samples was adjusted to 3 prior to EF treatment.  
254 Moreover, due to the high amount of TSS, it was recommended to filter the sample before  
255 electrolysis, since suspended solids can clog the channels of electrochemical reactors and  
256 contribute to electrode passivation.

257 EF paired with BDD was recently used to treat real refractory pharmaceutical  
258 wastewater (COD = 1253.3, TOC = 431.5 and BOD<sub>5</sub>/COD = 0.18) (Olvera-Vargas et al.,  
259 under review). EF-BDD achieved 97.1% of TOC removal in 6 h of treatment at 4.17 mA  
260 cm<sup>-2</sup> and 0.2 mM of Fe<sup>2+</sup>, much higher than the 64.3% of TOC removal achieved when  
261 EF was combined with a DSA. However, EF-DSA proved equally efficient at increasing  
262 the biodegradability of the effluent with much lower operational costs (BOD<sub>5</sub>/COD =  
263 0.68 at US\$ 1.46 m<sup>-3</sup> in 4 h vs. 1.68 US\$ m<sup>-3</sup> for BOD<sub>5</sub>/COD = 0.58 in 4 h with EF-BDD).  
264 These findings demonstrate not only the superb potential of EF to treat real  
265 pharmaceutical wastewater with a complex matrix, but also its versatility to reach  
266 different targets of water quality depending upon the anode materials: EF-DSA to

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

274 In another recent work, waste streams from the electronics industry were treated  
275 by EF using a graphene-based GDE (Garcia-Rodriguez et al., 2018). The experiments  
276 were performed in an undivided electrolytic reactor using BDD as anode and 0.4 L of  
277 wastewater sample. Before trial, the effluent was conditioned by adding 0.05 M of  $\text{K}_2\text{SO}_4$   
278 and diluted  $\text{H}_2\text{SO}_4$  to bring the pH to 3. Following 3 h of treatment, the TOC was depleted  
279 by 90% when the initial TOC concentration was  $53.46 \text{ mg L}^{-1}$  and by 60% when the initial  
280 TOC was  $299.31 \text{ mg L}^{-1}$ . The enhanced production of  $\text{H}_2\text{O}_2$  (up to  $495 \text{ mg L}^{-1}$   
281 accumulated in the solution in 3 h of electrolysis) promoted by the graphene-coated GDE  
282 maintained the continuous formation of  $\cdot\text{OH}$  through the Fenton's reaction that took care  
283 of the mineralization of the organic constituents.

### 284 ***Electrocoagulation***

285 For waste streams with high amounts of suspended solids and colloids, including oily  
286 waters, EC emerges as the most feasible option. EC is in fact a well-known  
287 electrochemical technology whose utilization can be traced back to the beginning of the  
288 20<sup>th</sup> century (Mollah et al., 2001). Very comprehensive reviews on EC, its principles and  
289 fields of application are available in the literature (Garcia-Segura et al., 2017; Hakizimana  
290 et al., 2017; Moussa et al., 2017). EC is able to remove organic pollutants, heavy metals  
291 and other inorganic species (sulfate, phosphate, chloride and nitrate, among others) by

292 adsorption onto the electrochemically-produced coagulants followed by settling or  
293 flotation. Moreover, colloidal suspensions are easily destabilized and coagulated  
294 afterwards. Non-toxic and inexpensive iron and aluminum electrodes are generally used.  
295 In fact, EC has become a powerful and effective wastewater treatment technology widely  
296 used even for large-scale applications. For example, F&T water solutions<sup>3</sup> manufactures  
297 EC systems with capacities between 10 to 1000 GPM. Powell Water<sup>4</sup>, WaterTectonics<sup>5</sup>,  
298 Elgressy Engineering Services LTD<sup>6</sup>. and Hydroleap<sup>7</sup> are other companies that  
299 commercialize EC technologies. Their main drawbacks lie in the production of secondary  
300 iron/aluminum sludge that requires further disposal and the important consumption of  
301 electrical energy. Besides, as EC is primarily a separation technology, it does not provide  
302 high efficiencies in the removal of dissolved organic carbon. For this reason, EC has been  
303 coupled to other oxidative technologies when high TOC removal is the goal (Thiam et  
304 al., 2014). In this respect, EC is a perfect candidate for integrated systems either as a  
305 conditioning (reduction of solids and the organic charge) or polishing (elimination of  
306 remaining inorganic species) step (Bocos et al., 2016). Combined processes will be  
307 covered in depth in a dedicated section of this review.

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

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

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

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

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

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

310 suspended solids was conducted by Kobya et al. (Kobya et al., 2016). The experiments  
311 were conducted in a continuous flow reactor, in which the electrochemical cell was  
312 equipped with two pairs of electrodes connected in monopolar parallel configuration with  
313 a space of 20 mm between them. When Fe electrodes were used, 85% and 76% of COD  
314 and TOC removal, respectively, were achieved under optimal conditions: inlet flow rate  
315 of  $0.01 \text{ L min}^{-1}$ , 80 min of treatment and a current density of  $65 \text{ A m}^2$ . With Al electrodes,  
316 COD and TOC removal efficiencies of 77% and 72%, respectively, were attained. The  
317 final COD and TOC values after treatment were lower than the permissible limit of  
318 discharge into the sewage, according to the Turkish legislation.

319 In a different study conducted at pilot scale (working volume of  $53 \text{ m}^3 \text{ day}^{-1}$ ) in a  
320 Korean municipal wastewater treatment plant, a 200-L EC reactor with Al cylindrical  
321 electrodes placed concentrically with a gap of 2.25 cm was operated at 10 V and a  
322 hydraulic retention time of 2 min (Nguyen et al., 2014). The treatment system was capable  
323 of removing total phosphorus (mainly  $\text{PO}_4^{3-}$ ) by 97.21-100% and COD by 95.38%, while  
324  $\text{NH}_4^+$  was totally removed. Moreover, TSS were reduced to less than  $5 \text{ mg L}^{-1}$  and  
325 coliform bacteria to  $30 \text{ MPN mL}^{-1}$ .

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

### 330 **Scale up and system integration: state of the art and perspectives**

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

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

#### 354 ***Modeling of EAOPs***

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



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

### 367 *Anodic oxidation modeling*

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

$$373 \quad j_{lim}(t) = nFk_m COD(t) \quad (1)$$

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

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

$$383 \quad COD(t) = COD^0 \left(1 - \frac{\alpha A k_m}{V_R} t\right) \quad (2)$$

$$384 \quad COD(t) = \alpha COD^0 \exp\left(-\frac{Ak_m}{V_R} t + \frac{I-\alpha}{\alpha}\right) \quad (3)$$

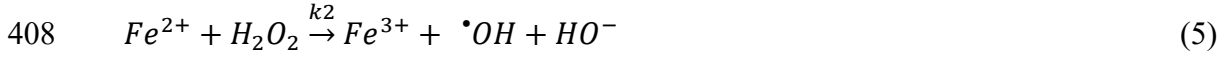
385           Where  $COD^0$  is the initial  $COD$  ( $\text{mol O}_2 \text{ m}^{-3}$ ),  $\alpha$  is the ratio of applied current  
 386 density and the initial limiting current density ( $j_{appl}/j_{lim}^0$ ),  $A$  corresponds to the electrode  
 387 area ( $\text{m}^2$ ) and  $V_R$  the reactor volume ( $\text{m}^3$ ).

388           Although, this model has been applied successfully in many studies (Aquino et  
 389 al., 2014; Barbosa et al., 2016; Steter et al., 2014) and can satisfactorily predict the  
 390 experimental data, its application is limited to direct oxidation processes at the electrode.  
 391 Other approaches of mathematic models for anodic oxidation include that of Cañizares et  
 392 al. (Cañizares et al., 2004), Mascia et al. (Mascia et al., 2010) and Lan et al. (Lan et al.,  
 393 2018), among others. One of the most interesting studies with respect to this review  
 394 consists of the works by Anglada et al (Anglada et al., 2009; Anglada et al., 2011;  
 395 Anglada, Urriaga, et al., 2010), dealing with modeling of real highly concentrated  
 396 wastewater in pilot-scale reactors. They have demonstrated that the rate of COD removal  
 397 is directly proportional to the ratio of the anode area over treated wastewater volume. In  
 398 this way, they used kinetic modeling for COD elimination to calculate the required total  
 399 anode area for scale up (Anglada, Urriaga, et al., 2010).

#### 400 *Electro-Fenton modeling*

401 Unlike AO, which mostly takes place at the anode surface, EF modeling is more complex  
 402 because it involves a series of reactions that take place concomitantly at the cathode  
 403 surface and in the bulk solution. The main reactions include the generation of  $\text{H}_2\text{O}_2$   
 404 through the reduction of oxygen via  $2e^-$  (Eq. 4), the  $\cdot\text{OH}$  radical generation (Eq. 5) and  
 405 the organics degradation (Eq. 6). Furthermore, there are several side reactions that coexist  
 406 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  $-\frac{d(S)}{dt} = k_3[\cdot OH][S]$  (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  $[H_2O_2] = j_{appl} \frac{k_1 K_{ad}[O_2]}{k_2 \lambda [Fe^{2+}]_0 (1 + K_{ad}[O_2])} (1 - e^{-k_2 \lambda [Fe^{2+}]_0 t})$  (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  $\lambda$  is a fixed ratio of  
 426  $[Fe^{2+}]_0/[Fe^{2+}]$ .

427  $Ln \frac{[S_0]}{[S]} + a([S_0] - [S]) = b \left( t - \frac{1 - e^{-ct}}{c} \right)$  (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  $c = k_2 \lambda [Fe^{2+}]_0$ .

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

435 Despite the good first approach of Liu's model for EF, it does not provide detailed  
436 information about byproduct evolution during the degradation process, which is of  
437 paramount importance when those intermediates are more toxic than the original  
438 pollutant. To overcome this drawback, Mousset et al. (Emmanuel Mousset et al., 2016)  
439 developed a more comprehensive model, using MATLAB<sup>®</sup> to perform the simulations.  
440 This model considered five representative steps: (i) in situ electrogeneration of  $\text{H}_2\text{O}_2$ ; (ii)  
441 Fenton's reaction in bulk solution; (iii) ferrous ion electroregeneration; (iv) scavenging  
442 reactions and; (v) degradation and mineralization of phenol. All these parameters had an  
443 impact on the kinetics of the process; however, step four, corresponding to the scavenging  
444 reaction between  $\text{Fe}^{2+}$  and  $\cdot\text{OH}$  (Eq. 7), was the most important, followed by step 1 (Eq.  
445 4), characteristic of this electrochemical process. It is worth highlighting that this model  
446 was able to predict the profile concentrations of ten compounds and was well  
447 corroborated by experimental data.

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

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

$$460 \quad ICE_{global} = \frac{4FV}{I} * \frac{d[COD]}{dt} = ICE_{anode} + ICE_{cathode} + ICE_{MO} \quad (14)$$

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

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

#### 469 *Electrocoagulation modeling*

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

$$477 \quad t = \frac{mnF}{M_{Al}A_0N} \quad (15)$$

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

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

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

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

503 *Computational fluid dynamics in electrochemical reactors*

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

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

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

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

#### 534 *Reactor design and scale up studies*

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

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



553 degradation of alizarin blue black B, a bipolar AO system was 1.2 fold more efficient  
554 than a monopolar AO system, furthermore, the former one didn't accumulate large  
555 amounts of intermediates unlike the monopolar system (Abdessamad et al., 2013), a  
556 higher efficiency with a bipolar configuration was also observed by (Thiam et al., 2014)  
557 during EC process and was attributed to a higher consumption of the electrode which  
558 resulted in a greater coagulation ability and faster pollutant removal. In the other hand,  
559 (Nidheesh et al., 2020) found that a monopolar connection was more efficient for the  
560 removal of COD and color from wastewater, being able to remove up to 55% and 56%,  
561 respectively, whereas a bipolar configuration removed 43% of COD and 48% of color.  
562 Moreover, (Kahraman & Şimşek, 2020) despite finding a slightly more efficient pollutant  
563 removal with a bipolar EC system, the higher cost due to the potential increase (up to 18.3  
564 V) made the monopolar system more feasible where the potential was less than 5V. The  
565 increase in potential with a bipolar configuration, could explain why in EF processes is  
566 preferred a monopolar configuration (Sultana et al., 2018) since high potentials would  
567 lead to parasitic reactions such as H<sub>2</sub> evolution and O<sub>2</sub> reduction via 4 electrons.

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

577 Most EAOP studies have been performed in electrochemical cells with volumes  
578 around 100 mL. Even though the number of studies making use of actual industrial waste  
579 streams, usually with high pollutant concentrations, is increasing (Garcia-Segura et al.,  
580 2018), there is still a lack of information regarding their treatment in larger volumes. To  
581 the best of our knowledge, the applications of EAOPs for the treatment of highly  
582 concentrated waste streams at volumes above 1 L (and up to 1 m<sup>3</sup>) are compiled in Table  
583 2. When the scale up process is carried out, many factors need to be considered for the  
584 design of such reactors, including the final effluent target quality, treatment capacity and  
585 time. However, as mentioned by Anglada et al. (Anglada, Urtiaga, et al., 2010), the A V<sup>-</sup>  
586 <sup>1</sup> ratio also needs to be taken into account and thus this ratio is also presented in Table 2.

587 From Table 2, the average electrode area needed for the treatment of one cubic  
588 meter of wastewater was 1.9 m<sup>2</sup>, 6.6 m<sup>2</sup> and 9.2 m<sup>2</sup> for EF/SPEF (combined or alone),  
589 AO and EC, respectively. It thus appears that processes involving the Fenton's reagent  
590 need a lower electrode area; however, most of the effluents tested were less concentrated,  
591 which has consequences in terms of a higher energy consumption per mass unit of  
592 pollutant. On the other hand, EC requires a higher electrode area and the electrodes  
593 lifespan is shorter than for the other 2 processes. Finally, AO appears to be a versatile  
594 process, with electrode area as low as 0.71 m<sup>2</sup> per m<sup>3</sup> of treated wastewater; however, the  
595 cost of the electrodes is the highest among the three families of electrochemical processes.  
596 Although the number of wastewater treatment electrochemical studies at pilot or  
597 industrial scale is increasing, there is still a critical need for real field and long-term  
598 studies, where the constraints related to their operation and application will be identified  
599 and solved.

600 ***Energy requirements***

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

609 The energy consumption expressed in kWh per kg of pollutant in the above-  
610 mentioned studies (Table 2), was evaluated using Eq. 16 (García-Rodríguez et al.,  
611 2016).

$$612 \text{ Energy Consumption (kWh kg}^{-1} \text{ pollutant)} = \frac{E_{cell}It*1000}{V_s\Delta(\text{pollutant})_{exp}} \quad (16)$$

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

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

624 *Integration of electrochemical methods in multi-stage sequential strategies*

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

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

650 et al., 2011). EAOPs can turn refractory organics into biodegradable compounds over  
651 short treatment times, that can then be more easily handled biologically, while reduced  
652 electrochemical treatment times represent significant savings. The opposite treatment  
653 strategy can also be applied, where the biodegradable fraction of the effluent can first be  
654 treated by biological means, while the remaining refractory pollutants can be degraded  
655 by EAOPs as a subsequent step, as long as the refractory compounds are not toxic to the  
656 biomass. This option can also be advantageous when the effluent displays high initial  
657 organic load (Kishimoto et al., 2017). Recent reviews on this topic with more detailed  
658 information can be found in the literature (Ganzenko et al., 2014; Olvera-Vargas et al.,  
659 2017).

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

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

675 glass cell with either DSA-RuO<sub>2</sub> or BDD anodes and a carbon-PTFE GDE fed with  
676 compressed air at 1 L min<sup>-1</sup>. Filtration units made use of flat ultrafiltration, NF and RO  
677 membranes at 20 °C. AO without previous filtration (BDD anode, 12 V of potential, 800  
678 rpm of stirring and 25 °C) only removed 65% of the initial TOC of the alkaline effluent,  
679 while NF removed TOC up to 72%. Yet, when combined, NF followed by AO resulted  
680 in an overall 96% of TOC removal in the alkaline effluent, following 9 h of AO treatment  
681 of the NF permeate. In this work, the suspended solids were first removed by  
682 microfiltration.

683         Anaerobically-digested food processing wastewater with high concentration of  
684 organic matter and NH<sub>4</sub><sup>+</sup> (COD = 3200-3400 mg L<sup>-1</sup>, NH<sub>4</sub><sup>+</sup> = 3352-3790 mg L<sup>-1</sup> and TSS  
685 = 15580 mg L<sup>-1</sup>) was treated by a sequential system consisting of electrochemical  
686 peroxidation, (ECP, consisting of EC with iron anodes and external addition of H<sub>2</sub>O<sub>2</sub>),  
687 followed by AO (Shin et al., 2017). ECP was conducted in a 1-L undivided reactor with  
688 2 pairs of SS electrodes positioned in parallel. The AO reactor also consisted of an  
689 undivided cell with a capacity of 0.3 L, a DSA and a SS cathode. Under optimal  
690 conditions, the integrated ECP-AO process achieved over 90% of TOC removal and  
691 almost complete NH<sub>4</sub><sup>+</sup> removal, following 150 min of ECP (pH 3, 28.5 mA cm<sup>-2</sup>, H<sub>2</sub>O<sub>2</sub>  
692 feeding at 1.67 mmol min<sup>-1</sup> and 1 M NaCl as supporting electrolyte) and 150 min of AO  
693 (pH 5, and 200 mA cm<sup>-2</sup>). During the ECP step more than 80% of the TOC was removed,  
694 while NH<sub>4</sub><sup>+</sup> was reduced by less than 25%. ECP benefits from a highly oxidative  
695 environment, where the addition of H<sub>2</sub>O<sub>2</sub> and the dissolution of the iron anode promote  
696 the production of •OH through the Fenton's reaction. The remaining TOC and NH<sub>4</sub><sup>+</sup> after  
697 ECP were removed by AO, mainly by intermediacy of chlorine species generated in the  
698 bulk solution. Before treatment, the wastewater sample was centrifuged at 3000 rpm for  
699 20 min to remove the solids.

700 Anaerobically digested sludge generated in a poultry farm (COD = 26,200 mg L<sup>-1</sup>,  
701 TOC = 5,235 mg L<sup>-1</sup> and TSS = 31,300 mg L<sup>-1</sup>) was successfully treated by means of  
702 an integrated process consisting of ECP with Fe electrodes followed by EF with a carbon  
703 brush cathode and a BDD anode (Olvera-Vargas et al., 2019). The synergistic effects of  
704 electrocoagulation and advanced oxidation during ECP (initial pH of 5, [H<sub>2</sub>O<sub>2</sub>]/[Fe<sup>2+</sup>]  
705 ratio of 5, 15.38 mA cm<sup>-2</sup> and 2 h of treatment) achieved 89.3% and 85.6% of COD and  
706 TSS removal, respectively, increasing sludge dewaterability and reducing the amount of  
707 disposable solid sludge in the process. The wastewater treated by ECP was then subjected  
708 to further EF treatment, once the solid sludge was removed from the effluent (such residue  
709 was free of coliforms and rich in nutrients with fertilizing potential). EF achieved almost  
710 total mineralization of the remaining organic matter under optimal conditions (pH 3 and  
711 25 mA cm<sup>-2</sup>), leaving only 24.6 mg L<sup>-1</sup> and 16.3 mg L<sup>-1</sup> of COD and TOC, respectively,  
712 after 8 h of treatment. This two-step electrochemical process proved to be a powerful  
713 strategy to deal with highly polluted and viscous sludge.

714 In another sequential treatment, (diluted) pharmaceutical wastewater was pre-  
715 treated by EF followed by activated sludge (AS) (Mansour et al., 2015). The AS treatment  
716 was conducted in 500-mL reactors containing 200 mL of the effluent and 1 g L<sup>-1</sup> of  
717 biomass from a municipal wastewater treatment plant, at neutral pH and with different  
718 organic nutrients. For EF, 1-L samples were treated in an undivided electrolytic cell  
719 containing a Pt anode and a carbon-felt cathode under constant aeration and stirring. The  
720 optimal conditions determined for EF were 0.69 mM of Fe<sup>2+</sup>, 466 mA of current and pH  
721 3. Following 180 and 300 min of EF treatment, the BOD<sub>5</sub>/COD ratio (biodegradability  
722 index) increased from 0.40 to 0.45 and 0.47, respectively, while the TOC was only  
723 reduced by 14 and 18%, respectively. After 15 days of AS treatment, the overall TOC  
724 removal was 80% and 89% for following 180 min and 300 min of electrolysis,

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

728 A large-scale sequential system including several steps was reported by Moreira  
729 et al. (Moreira, Soler, et al., 2015), in which the remediation of sanitary landfill leachate  
730 was conducted by a sequence of AS, coagulation and PEF/SPED. In this case, the aim of  
731 the electrochemical treatment was to increase the biodegradability of the pre-treated  
732 effluent as a conditioning step prior to biological polishing. The AS process was  
733 performed in a 12-L bioreactor with a conical bottom where 8 L of landfill leachate were  
734 treated using AS from a municipal WWTP. The pH was maintained between 6.5 and 9.  
735 After sedimentation, the supernatant was transferred to a coagulation tank where  $\text{FeCl}_3$   
736 was added at pH 3 and stirred for 15 min. 48 h of sedimentation were required, after  
737 which the effluent was sent to the electrochemical treatment. The 35-L electrochemical  
738 flow-through system comprised an electrochemical filter-press cell with a BDD anode  
739 and a carbon-PTFE GDE cathode, a 20-L recirculation tank and a photoreactor. The AS  
740 treatment achieved 13-33% of DOC removal, as well as total oxidation of  $\text{NH}_4^+$  and  
741 complete alkalinity removal. With the coagulation process, 63% of DOC abatement was  
742 attained. During the SPEF process, 53% of the remaining DOC was removed at a current  
743 density of  $200 \text{ mA cm}^{-2}$ ,  $60 \text{ mg L}^{-1}$  of  $\text{Fe}^{2+}$ , pH of 2.8 and  $20 \text{ }^\circ\text{C}$  (after SPEF, the effluent  
744 was neutralized to pH 7.5 and submitted to 3 h of clarification to remove the remaining  
745 sludge). Overall, 87% of DOC removal was achieved at the end of the multi-stage  
746 treatment. The resulting COD,  $\text{BOD}_5$  and total nitrogen values were still above the  
747 Portuguese and European regulations for discharge. However, Zhan-Wellens  
748 biodegradability tests indicated that the sample was suitable for further biological  
749 treatment to comply with the discharge limits.



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

761

### 762 **Applications to highly concentrated wastewater: nature, characteristics and** 763 **challenges**

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

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

777         Highly concentrated solutions need particular attention and their physical and  
778 chemical characteristics determine the type of treatment to be applied, based on pH,  
779 electrical conductivity, chemical oxygen demand (COD), total organic carbon (TOC),  
780 biodegradability – that can be assessed with the biochemical oxygen demand after 5 days  
781 (BOD<sub>5</sub>) over COD ratio – total inorganic carbon (TIC), total suspended solids (TSS), total  
782 dissolved solids (TDS), main inorganic ions (Cl<sup>-</sup>, SO<sub>4</sub><sup>2-</sup>, NH<sub>3</sub>/NH<sub>4</sub><sup>+</sup>, NO<sub>2</sub><sup>-</sup>, NO<sub>3</sub><sup>-</sup>, PO<sub>4</sub><sup>3-</sup>)  
783 and heavy metals analyzes. In the last two decades, electrochemical processes have  
784 attracted a lot of interest for their high versatility, i.e. their possibility to treat solutions  
785 containing low or high concentrations of organics and/or inorganics. In this section, five  
786 categories of wastewater are investigated, alongside their properties and influence on the  
787 type of electrochemical processes to be applied, with emphasis on electrochemical  
788 advanced oxidation processes (EAOPs), in combination or not with electrocoagulation  
789 (EC). They consist of (i) high COD effluents of industrial origin; (ii) hypersaline  
790 effluents; (iii) solutions contaminated with a mixture of organic and inorganic  
791 contaminants; (iv) effluents with high rheological complexity and; (v) solutions with high  
792 COD load but low pollutant content.

### 793 ***High COD effluents of industrial origin***

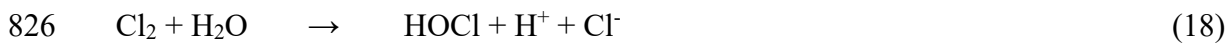
794 Industrial waste streams are often characterized by high COD content and need a specific  
795 treatment. The main industry sectors that have attracted attention due to their recalcitrance  
796 to biological processes comprise textile, petroleum, tannery, chemical, pharmaceutical  
797 and paper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

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

#### 814 *Hypersaline effluents*

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

824 2012):



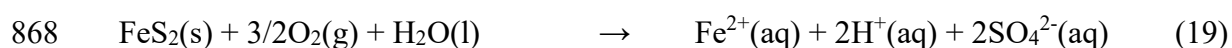
827           It is important to highlight that the formation of halogenated by-products, such as  
828 trihalomethane (THM), could pose a health risk (Hurwitz et al., 2014). Though the initial  
829 COD of RO concentrate is quite low (14-190 mg-O<sub>2</sub> L<sup>-1</sup>, Table 5), organic intermediates  
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-O<sub>2</sub> L<sup>-1</sup>, generally basic pH (8-9)  
845 and high concentrations of inorganic ions, such as Cl<sup>-</sup> (1.5 – 6 g L<sup>-1</sup>), SO<sub>4</sub><sup>2-</sup> (0.1-1.8 g L<sup>-1</sup>)  
846 and NH<sub>3</sub> (0.4-3.2 g L<sup>-1</sup>). Such complex solutions are non-biodegradable (BOD<sub>5</sub>/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

849 chlorine species, such as HClO, as described earlier (Eqs. 1-2) (Cossu et al., 1998). This  
850 oxidizing agent participates in the degradation and mineralization of organic compounds;  
851 however, some organo-halogenated transformation byproducts can also be generated.  
852 These compounds are known to be potentially toxic and therefore deserve particular  
853 attention. The initial presence of ammonia at high level could also constitute a drawback,  
854 since this species can be oxidized to nitrate (NO<sub>3</sub><sup>-</sup>), an unwanted final product at  
855 concentrations higher than the disposal regulations (Anglada, Urriaga, et al., 2010). The  
856 formation of this ion must therefore be carefully monitored. In order to have an overall  
857 approach for a cost-effective complete removal of organic and inorganic species,  
858 combination with separation processes (e.g., nanofiltration, reverse osmosis, etc) (Cui et  
859 al., 2018; El Kateb et al., 2019; Fernandes et al., 2019; Ukundimana et al., 2018) and/or  
860 biological technologies (Anglada, Urriaga, et al., 2010; Baiju et al., 2018; Ding et al.,  
861 2018; El Kateb et al., 2019; Moreira, Soler, et al., 2015; Turro et al., 2012; Zhao et al.,  
862 2010) has been suggested in previous studies. The advantages of such combined  
863 approaches will be reviewed in a dedicated section towards the end of this review.

864 Another challenging source of mixed contamination (organic and inorganic) that  
865 represents a major problem in the mining industry is acid mine drainage (AMD),  
866 generated by the oxidation of sulfides during ore processing, leading to sulfuric acid (Eq.  
867 19).



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

874 oxidation and its water discharge regulation is becoming more stringent (Nariyan et al.,  
875 2018). In parallel, ferric hydroxide (Fe(OH)<sub>3</sub>) precipitation is another environmental  
876 issue. Several studies on the efficiency of electrochemical treatment of AMD as shown  
877 in Table 7. The primary goal of electrochemistry is to increase the pH, as an alternative  
878 to chemical neutralization and precipitation, by reducing H<sup>+</sup> into hydrogen gas (H<sub>2</sub>(g)) at  
879 the cathode, according to Eq. 20:



881 For this purpose, the operation in divided cell is favored in order to avoid the  
882 oxidation of Fe<sup>2+</sup> into Fe<sup>3+</sup> that then precipitates on the electrodes – especially on the  
883 cathode – and subsequently causes electrode fouling (Chartrand & Bunce, 2003). The  
884 assistance of chemical alkalization using CaO as pre-treatment before the  
885 electrochemical step has also demonstrated synergistic effects (Orescanin & Kollar, 2012;  
886 Radić et al., 2014). Additional benefits can be obtained from such electrochemical  
887 treatment by electrodeposition of metals and by producing a dewaterable sludge for  
888 possible valorization (Bunce et al., 2001). Another interesting feature is the elimination  
889 of inorganics such as sulfate by EC (Mamelkina, Tuunila, et al., 2019; Nariyan et al.,  
890 2017). It is finally worthy of note that the initial presence of iron could enhance the  
891 electrochemical process efficiency by involving ·OH radicals through the Fenton’s  
892 reaction in an EF process.

### 893 ***Effluents with high rheological complexity***

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

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

910         The presence in sludge of initial salts such as chloride ions induces several  
911 positive effects on the process efficiency. The electro-oxidation of Cl<sup>-</sup> at Ti/RuO<sub>2</sub> anode  
912 further allows the generation of hypochlorous acid (HClO, Eqs. 1-2) (Bureau et al., 2012;  
913 Song et al., 2010), a powerful oxidizing agent known to inhibit the activity of micro-  
914 organisms. HClO could also oxidize the organic compounds presents in the sludge so that  
915 a decrease of COD is observed. Finally, a decrease of phosphate (PO<sub>4</sub><sup>3-</sup>) initially present  
916 in the sludge was also observed (Bureau et al., 2012), which could be attributed to the  
917 oxidation of ferrous into ferric iron by HClO (Eq. 21) and then Fe<sup>3+</sup> reaction with H<sub>2</sub>PO<sub>4</sub><sup>-</sup>  
918 formed a FePO<sub>4</sub> precipitate (Eq. 22) (Bureau et al., 2012):



921         In addition, calcium (Ca), potassium (K) and magnesium (Mg) concentrations  
922 increased by 20% to 50%, while total nitrogen content remained stable (Bureau et al.,  
923 2012), ensuring that the fertilizing value of the sludge was not compromised. Finally,

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

### 929 ***High COD load with low pollutant content***

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

937 In soil washing/flushing technologies, extracting agents are added most of the  
938 time in a water solution before washing the excavated soil or flushing the non-excavated  
939 soil in order to solubilize the hydrophobic organic pollutants (Mousset et al., 2013). A  
940 good washing agent is a compound with high extraction efficiency (thus requiring as low  
941 an amount of agents as possible), low ability to adsorb onto soil and low toxicity towards  
942 soil microorganisms. They usually have amphiphilic properties that allow dissolving soil  
943 pollutants into the water solution. There are many kinds of agents that have been tested  
944 in the literature including co-solvents, dissolved organic matter, deoxyribonucleic acids,  
945 vegetable oils, fatty acid methyl esters, cyclodextrins and surfactants (Trellu et al., 2016).  
946 Most of them (except for co-solvents) have a high molecular weight ranging from 250 to  
947 1500 g mol<sup>-1</sup>. As soil washing/flushing only transfers the pollution from solid to liquid  
948 matrix, there is a need for a post-treatment to remove the contaminants. In the past few



949 years, advanced electrochemical treatment has appeared as a promising alternative to  
950 biological post-treatment, since those pollutants are barely biodegradable (Mousset et al.,  
951 2017).

952 Table 9 presents the state of the art of electrochemical treatment of soil  
953 washing/flushing solutions. Generally, it is observed that organic pollutants represent  
954 only 1% to 5% of the total organic carbon fraction, the remaining part consisting mostly  
955 of the extracting agent, while the organic matter content extracted from soil is negligible  
956 (Mousset et al., 2017). Interestingly, using different families of extracting agents has  
957 shown to affect the electrochemical treatment efficiency. Surfactants have a long C-C  
958 bond hydrophobic chain and a hydrophilic head, which allows them to form micelles after  
959 reaching a minimal concentration, known as the critical micelle concentration (CMC,  
960 Figure 5a) (Mousset et al., 2014a). These micelles can then trap the hydrophobic organic  
961 contaminants (HOCs), leading to steric hindrance during the oxidation of the pollutant.  
962 Thus, the surfactants are preferentially degraded as compared to the pollutants.  
963 Alternative agents such as cyclodextrins have also been proposed (Mousset & Oturan,  
964 2014), with a toroidal shape that allows trapping the organics into their hydrophobic  
965 cavity (Figure 5b). The functional groups at the surface of cyclodextrins (carboxylic  
966 groups, hydroxyl groups, etc) allow them to bound to ferrous iron under EF treatment,  
967 with hydroxyl radicals being produced close to the contaminants. The pollutant was  
968 therefore preferentially degraded as compared to the cyclodextrin, thanks to the ternary  
969 complex formation ( $\text{Fe}^{2+}:\text{HPCD}:\text{HOC}$ ) according to Eqs. 23 and 24:



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

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

986

## 987 **Conclusion**

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

999 concentrated wastewater, it may prove more beneficial to apply electrochemistry as a pre-  
1000 or post- treatment approach, combined with a more economical conventional biological  
1001 treatment system. Process optimization and cost-efficiency are at the core of a suitable  
1002 treatment strategy; yet, there is a lack of a focused and consistent approach to scale up  
1003 electrochemical reactors. In fact, there are very few pilot studies and therefore, the final  
1004 section of this review emphasized the importance of modeling electrochemical processes,  
1005 as well as the rheological and mass transfer conditions in large scale electrochemical  
1006 reactors. By combining various modeling approaches, electrochemical wastewater  
1007 treatment can be understood in a more systematic manner and adequate reactors can be  
1008 designed. We defined the electrode area over treated volume ratio ( $A/V$ ) as a key  
1009 parameter, which in turn influences energy consumption. We concluded that in most  
1010 cases, the specific energy consumption is lower when the wastewater contains a high  
1011 concentration of contaminants. At a time that electrochemistry for wastewater treatment  
1012 reaches maturity, highly contaminated waste streams may therefore provide the best niche  
1013 to demonstrate the potential of electrochemical wastewater treatment at large scale.

#### 1014 **Declaration of interest statement**

1015 The authors declare that they have no known competing financial interests or personal  
1016 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 <sub>5</sub> = 324, pH = 4.8, SO <sub>4</sub> <sup>2-</sup> = 38, Cl <sup>-</sup> = 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 <sup>-</sup> = 592.02, ClO <sub>3</sub> <sup>-</sup> = 1.17, NH <sub>4</sub> <sup>+</sup> = 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 <sub>4</sub> <sup>2-</sup> = 242, Cl <sup>-</sup> = 1500	AO	10	Ru <sub>0.7</sub> Ir <sub>0.3</sub> O <sub>2</sub>	SS mesh	FR	30% DOC removal	(Radjenovic, Bagastyo, et al., 2011)

Municipal sanitary landfill leachate	COD = 3385, BOD <sub>5</sub> = 500, C = 22.60, pH = 8.4, SO <sub>4</sub> <sup>2-</sup> = 11, Cl <sup>-</sup> = 2574, NH <sub>4</sub> <sup>+</sup> = 1591, NO <sub>3</sub> <sup>-</sup> = 1.9, PO <sub>4</sub> <sup>3-</sup> = 31	AO	10	BDD	BDD	FR	51% COD removal and 32% ammonia removal	(Anglada et al., 2011)
Municipal sanitary landfill leachate	COD = 3782, BOD <sub>5</sub> = 560, pH = 8.4, Cl <sup>-</sup> = 3702, NH <sub>4</sub> <sup>+</sup> = 3143	AO	0.8	RuO <sub>2</sub> -IrO <sub>2</sub>	SS	UR	26.5% COD removal and 81.1% NH <sub>4</sub> <sup>+</sup> removal	(Zhang et al., 2010)
Industrial landfill leachate	COD = 17100-18400, pH = 9, Fe = 20, Cl <sup>-</sup> = 52300, NH <sub>4</sub> <sup>+</sup> = 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 <sub>4</sub> <sup>2-</sup> = 140-199, Cl <sup>-</sup>	AO	250	BDD	SS	Pilot FR	70% of COD removal and 37% of NH <sub>4</sub> <sup>+</sup>	(Anglada, Urtiaga, et al., 2010)

landfill leachate	= 1615-1819, $\text{NH}_4^+$ = 896-980, $\text{NO}_3^-$ = 5-1207							
Municipal sanitary landfill leachate	$\text{NH}_4^+$ = 1934, COD = 4434, TOC = 2782, BOD <sub>5</sub> = 640, TSS = 317, C = 12.77, pH = 8.35	AO	NA	BDD	SS	FR	Total $\text{NH}_4^+$ removal	(Cabeza, Urtiaga, Rivero, et al., 2007)
Municipal sanitary landfill leachate	DOC = 1600-3100, TSS = 15310-17602, pH = 8- 9, $\text{SO}_4^{2-}$ = 60-240, $\text{Cl}^-$ = 5000-6200, $\text{NH}_4^+$ = 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 <sub>5</sub> = 1-10, $\text{NO}_3^-$ = 1035-1192, $\text{SO}_4^{2-}$	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 <sup>-</sup> = 3046- 3822, pH = 2.2-2.9							
Textile	COD = 1224, BOD <sub>5</sub> = 323.8, TOC = 394.6, Cl <sup>-</sup> = 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 <sub>5</sub> = 790, TSS = 3712, SO <sub>4</sub> <sup>2-</sup> = 910	AO / EF / PEF	0.1	BDD	C-PTFE GDE	UR	80% TOC removal	(Flores et al., 2017)
RO concentrate	COD = 120-150, BOD <sub>5</sub> = 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 <sup>-</sup> = 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 <sub>2</sub> -coated carbon membrane	SS	UR	>90% COD removal	(Yang et al., 2012)

Carwash wastewater	COD = 572, BOD <sub>5</sub> = 178, pH = 6.4, C = 1.6, Anionic surfactants = 95.5	AO	0.4	BDD/Pb O <sub>2</sub>	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 <sup>-</sup> = 2000, pH = 6.7 C = 5.3	AO	20	DSA- RuO <sub>2</sub>	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 monopol ar	Fe or Al	FTR (contin ous flow)	85% COD removal (Fe) and 77% (Al)	(Koby et al., 2016)

				connecti on				
Municipal	COD = 238.63, BOD <sub>5</sub> = 205.76, SS = 281.9, pH = 7.7, NH <sub>4</sub> <sup>+</sup> = 29.85,	EC	200	Al	Al	FTR (continu ous flow)	97.27-100% Total-P and NH <sub>4</sub> <sup>+</sup> removal	(Nguyen et al., 2014)
Olive oil mill wastewater	COD = 1393.3, TOC = 581.1, pH = 6.83, TSS = 1890, TN = 16.7, Cl <sup>-</sup> = 41.8, SO <sub>4</sub> <sup>2-</sup> = 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 <sub>4</sub> <sup>3-</sup> =27, SO <sub>4</sub> <sup>2-</sup> = 500	EC	0.5	Fe/Al	Fe/Al	FR	Complete PO <sub>4</sub> <sup>3-</sup> 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	SO <sub>4</sub> <sup>2-</sup> = 13000, TDS = 19790, pH = 2.68, C = 10.66	EC	0.5	Al	Fe	UR	41% of SO <sub>4</sub> <sup>2-</sup> 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 <sub>5</sub> = 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 <sub>0</sub> , respectively)	(Garcia-Rodriguez et al., 2018)

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

Pharmaceutical	COD = 1253.3, TOC = 431.5, BOD <sub>5</sub> /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 <sup>2</sup> )	A V <sup>-1</sup> (m <sup>2</sup> m <sup>-3</sup> )	Current Density (mA cm <sup>-2</sup> )	Energy consumption (kWh kg <sup>-1</sup> pollutant)	Ref.
AO	Tannery	1.2	1202.5 mg L <sup>-1</sup> COD	51.53	90	16	1.3	25	16.1 kWh kg <sup>-1</sup> COD	(Liu et al., 2017)
AO	Washing machine	1	783 mg L <sup>-1</sup> COD	79.7	360	63.5	6.4	16.6	-	(Durán et al., 2018)
AO	landfill leachate	10	3385 mg L <sup>-1</sup> COD	30	180	70	0.7	257	134 kWh kg <sup>-1</sup> COD	(Anglada et al., 2011)
AO	rubber manufacturing process	200	1500 mg L <sup>-1</sup> COD	80	600	2800	1.4	60	77 kWh kg <sup>-1</sup> COD	(Urriaga et al., 2014)



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

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

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

SPEF-AO	Industrial textile wastewater	10	1763 mg L <sup>-1</sup> COD	93	360	50	0.5	25	4.2 kWh kg <sup>-1</sup> <sub>COD</sub>	(Salazar et al., 2019)
SPEF-AO	Mixture of parabens	2.5	100 mg L <sup>-1</sup> TOC	66	240	20	0.8	10	84 kWh kg <sup>-1</sup> <sub>TOC</sub>	(Steter et al., 2018)
SPEF-AO	Mixture of azo dyes	2.5	105 mg L <sup>-1</sup> TOC	98.8	300	20	0.8	100	660 kWh kg <sup>-1</sup> <sub>TOC</sub>	(dos Santos et al., 2018)
SPEF-AO	EC pre-treated cheese whey wastewater	2	23,029 mg L <sup>-1</sup> TOC	27.1	420	20	1	100	7.3 kWh kg <sup>-1</sup> <sub>TOC</sub>	(Tirado et al., 2018)
SPEF-AO	Mixture of pesticides	75	71 mg L <sup>-1</sup> DOC	32	120	400	0.5	74	1201.4 kWh kg <sup>-1</sup> <sub>DOC</sub>	(Salmerón et al., 2019)
SPEF	benzoic acid solution	10	100 mg L <sup>-1</sup> 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 <sub>5</sub> = 180-300, TSS = 525-630, NH <sub>4</sub> <sup>+</sup> = 1300-1355, Cl <sup>-</sup> = 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 <sub>5</sub> = 1196, 175, NA pH = 8.4, 7.7, 7.5,	BIO-Fenton + AO	1	BDD	SS	FR	Overall, > 95% of COD removal and	(Urriaga et al., 2009)

landfill leachate	SO <sub>4</sub> <sup>2-</sup> = 438,500,3100, Cl <sup>-</sup> = 2124, 1876, 1460, NH <sub>4</sub> <sup>+</sup> = 1225, 750, 700, NO <sub>3</sub> <sup>-</sup> = NA, 700, 660						complete NH <sub>4</sub> <sup>+</sup> removal	
Landfill leachate	COD = 7200, BOD <sub>5</sub> /COD = 0.03, Cl <sup>-</sup> = 6000, Fe = 70, pH = 8.2, TDS = 9500	Heterogeneous EF + BIO	1	Ti/TiO <sub>2</sub>	Graphite	UR	82% of COD removal with EF, overall 97%	(Baiju et al., 2018)
Winery wastewater	COD = 10240, C = 1.10, TSS = 2860, pH = 3.7, Fe = 17	AO + EC + Electromagnetization + Clarification	8	SS	SS	UR	77% COD removal, 99% removal of PO <sub>4</sub> <sup>3-</sup> and TSS	(Orescanin et al., 2013)

		on + O <sub>3</sub> / H <sub>2</sub> O <sub>2</sub> / UV						
Winery wastewater	COD = 12000, 380, BOD <sub>5</sub> = 7950, 150, TSS = 81, 72, pH = 3.7, 8.3	BIO + AO / EF / PEF / SPEF	1.25	BDD	C-PTFE GDE	FR	COD, BOD <sub>5</sub> , NH <sub>4</sub> <sup>+</sup> , NO <sub>3</sub> <sup>-</sup> and SO <sub>4</sub> <sup>2-</sup> above regulatory levels	(Moreira, Boaventura, et al., 2015)
Cellulose bleaching	COD = 1250 (acid), 1500 (basic), BOD <sub>5</sub> = 563 (acid), 595 (basic), pH = 2.5, 10.5, Cl <sup>-</sup> =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 <sub>5</sub> = 1517, TSS = >1988, pH = 8.3, Cl <sup>-</sup> = 1131, NH <sub>4</sub> <sup>+</sup> = 177, NO <sub>3</sub> <sup>-</sup> = 1.5	EC + AO	0.1	DSA (IrO <sub>2</sub> -Ta <sub>2</sub> O <sub>5</sub> )	Pt	UR	85%	(Borbón et al., 2014)
Dairy wastewater	COD = 5600, TOC = 2464, BOD <sub>5</sub> /COD = 0.8, TDS = 4530, pH = 5.87, Cl <sup>-</sup> = 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 <sub>5</sub> = 528, TSS = 445-530, pH = 3.7-4.3, Cl <sup>-</sup> = 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 = $2.4 \times 10^3$ CFU mL <sup>-1</sup>	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 <sub>4</sub> <sup>+</sup> = 3352-3790, SS = 6260-6400, TS = 15580	ECP + AO	1 / 0.3	SS/DSA	SS/Ti	UR	>90% TOC and NH <sub>4</sub> <sup>+</sup> removal	(Shin et al., 2017)
Municipal wastewater	COD = 495, BOD <sub>5</sub> = 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 <sub>2</sub> )				
Pulp and paper	COD = 1450, BOD <sub>5</sub> = 350, TSS = 350, TDS = 1050, Cl <sup>-</sup> = 325, pH = 6.72	Permanganate oxidation + ECP + Co <sub>3</sub> /UV/peroxymonosulfate	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/GD E	UR (columns )	51% TOC removal	(Díez et al., 2017)
Slaughterhouse wastewater	COD = 1494, TSS = 19717, BOD <sub>5</sub> = 685, pH = 7.8, NH <sub>4</sub> <sup>+</sup> = 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 <sup>-</sup> = 3547, C = 15.5	EC + UV-assisted AO	3.2 L for EC, 2 L for AO	Fe for EF, BDD for AO	Al/AISI3 04 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 <sub>5</sub> /COD = 0.43, TDS = 8370, pH = 7.2, Cl <sup>-</sup> = 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 <sub>2</sub> L <sup>-1</sup> , pH 5.2, conductivity = 0.5 mS cm <sup>-1</sup> )	EC/Fered-Fenton	Cathode: Fe (22.5 cm <sup>2</sup> ) Anode: Fe (22.5 cm <sup>2</sup> )	TOC removal, energy consumption	(Başaran Dindaş et al., 2020)
Pharmaceutical	Pharmaceutical wastewater (COD = 12 g-O <sub>2</sub> L <sup>-1</sup> , pH 8.5, conductivity = 7 mS cm <sup>-1</sup> )	AO	Cathode: stainless steel (78 cm <sup>2</sup> ) Anode: BDD (78 cm <sup>2</sup> )	Current density, flow rate, TOC removal	(Domínguez et al., 2012)
Chemical	Chemical industry wastewater (COD = 1.3 g-O <sub>2</sub> L <sup>-1</sup> , pH 6.7, conductivity = 5.3 mS cm <sup>-1</sup> , [Cl <sup>-</sup> ] = 0.4 g L <sup>-1</sup> )	AO	Cathode: Pt (63 cm <sup>2</sup> ) Anode: Ti/IrO <sub>2</sub> , Ti/RuO <sub>2</sub> (63 cm <sup>2</sup> )	Anode materials, temperature, current density, COD removal, energy consumption	(Valero et al., 2014)
Chemical	Chemical industry wastewater (COD = 1.0 g-O <sub>2</sub> L <sup>-1</sup> , pH 6.1,	AO	Cathode: graphite (100 cm <sup>2</sup> )	COD and TOC removal, by-products formation	(Rajkumar & Palanivelu, 2004)

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

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



	g L <sup>-1</sup> , conductivity = 1.5 mS cm <sup>-1</sup> , pH 6.9)				
Petroleum	Petroleum wastewater (COD=1.6 g-O <sub>2</sub> L <sup>-1</sup> , conductivity = 4.6 mS cm <sup>-1</sup> , pH 7.5, [Cl <sup>-</sup> ] = 22.4 g L <sup>-1</sup> )	AO	Cathode: Ti (15 cm <sup>2</sup> ) Anode: Ti/Pt, BDD (10 cm <sup>2</sup> )	Anode materials, current density, electrolyte concentration, agitation rate, COD removal, energy consumption	(Rocha et al., 2012)
Petroleum	Petroleum wastewater (COD=0.7 g-O <sub>2</sub> L <sup>-1</sup> , conductivity = 5.1 mS cm <sup>-1</sup> , pH 7.8, [Cl <sup>-</sup> ] = 1.6 g L <sup>-1</sup> )	AO	Cathode: Ti (63.5 cm <sup>2</sup> ) Anode: Ti/IrO <sub>2</sub> -Ta <sub>2</sub> O <sub>5</sub> , Ti/PbO <sub>2</sub> , BDD (63.5 cm <sup>2</sup> )	Anode materials, current density, COD removal, energy consumption	(da Silva et al., 2013)
Paper mill	Paper mill industry wastewater (COD = 5-6 g-O <sub>2</sub> L <sup>-1</sup> , pH 8, conductivity = 7 mS cm <sup>-1</sup> )	AO	Cathode: stainless steel (682 cm <sup>2</sup> ) Anode: PbO <sub>2</sub> (682 cm <sup>2</sup> )	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 <sub>2</sub> L <sup>-1</sup> , pH 12.8-13, conductivity = 0.2 mS cm <sup>-1</sup> )	AO	Cathode: stainless steel (141 cm <sup>2</sup> )  Anode: Ti/RuO <sub>2</sub> (11 cm <sup>2</sup> )	Current density, flow rate, color and COD removal, current efficiency	(Chanworrawoot, 2012)
Paper mill	Paper mill industry wastewater (COD = 0.3 g-O <sub>2</sub> L <sup>-1</sup> , pH 6.8, conductivity = 1.1 mS cm <sup>-1</sup> )	AO	Cathode: stainless steel (50 cm <sup>2</sup> )  Anode: BDD, Ti/Ti <sub>0.5</sub> Ru <sub>0.45</sub> Sn <sub>0.05</sub> O <sub>2</sub> (50 cm <sup>2</sup> )	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 <sup>-1</sup> , COD = 120-150 mg-O <sub>2</sub> L <sup>-1</sup> , pH 7.2-7.6)	EF	Cathode: graphite felt (120 cm <sup>2</sup> ) Anode: Pt wire	[Fe <sup>2+</sup> ], cathodic potential, [H <sub>2</sub> O <sub>2</sub> ], pH	(Zhou et al., 2012)
RO concentrate from a tertiary steel WWTP (Conductivity = 22.3 mS cm <sup>-1</sup> , [Cl <sup>-</sup> ] = 6.4 g L <sup>-1</sup> , COD = 120-190 mg-O <sub>2</sub> L <sup>-1</sup> , pH 8.3)	AO	Cathode: stainless steel (3 cm <sup>2</sup> ) Anode: BDD, Ti/IrO <sub>2</sub> -Ta <sub>2</sub> O <sub>5</sub> , Ti/IrO <sub>2</sub> -RuO <sub>2</sub> ) (3 cm <sup>2</sup> )	Anode materials, pH, COD removal, energy consumption	(Zhou et al., 2011)
RO concentrate from tertiary WWTP (Conductivity = 5 mS cm <sup>-1</sup> , [Cl <sup>-</sup> ] = 1.2 g L <sup>-1</sup> , COD = 170 mg-O <sub>2</sub> L <sup>-1</sup> , pH 8.0)	AO	Cathode: stainless steel (projected surface area: 24 cm <sup>2</sup> ) Anode: Ti/IrO <sub>2</sub> -Ta <sub>2</sub> O <sub>5</sub> , Ti/RuO <sub>2</sub> -IrO <sub>2</sub> , Ti/Pt-IrO <sub>2</sub> , Ti/PbO <sub>2</sub> , Ti/SnO <sub>2</sub> -Sb	Anode materials, COD and inorganic removal, by-products evolution	(Bagastyo et al., 2011)

		(projected surface area: 24 cm <sup>2</sup> )		
RO concentrate from tertiary WWTP spiked with pharmaceuticals (Conductivity = 4.2 mS cm <sup>-1</sup> , [Cl <sup>-</sup> ] = 1.5 g L <sup>-1</sup> , DOC = 57 mg-C L <sup>-1</sup> , pH 7.5)	AO	Cathode: stainless steel (projected surface area: 24 cm <sup>2</sup> )  Anode: Ti/RuO <sub>2</sub> -IrO <sub>2</sub> (projected surface area: 24 cm <sup>2</sup> )	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 <sup>-1</sup> , [Cl <sup>-</sup> ] = 1.6 g L <sup>-1</sup> , DOC = 57 mg-C L <sup>-1</sup> , pH 7.7)	AO	Cathode: stainless steel (projected surface area: 24 cm <sup>2</sup> )  Anode: Ti/Ru <sub>0.7</sub> -Ir <sub>0.3</sub> O <sub>2</sub> , Ti/SnO <sub>2</sub> -Sb (projected surface area: 24 cm <sup>2</sup> )	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 <sup>-1</sup> , [Cl <sup>-</sup> ] = 1.4 g L <sup>-1</sup> , COD = 136 mg-O <sub>2</sub> L <sup>-1</sup> , pH 7.7)	AO	Cathode: stainless steel (41 cm <sup>2</sup> ) Anode: BDD (41 cm <sup>2</sup> )	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 <sup>-1</sup> , [Cl <sup>-</sup> ] = 0.14 g L <sup>-1</sup> , COD = 145 mg-O <sub>2</sub> L <sup>-1</sup> , pH 6.8)	AO	Cathode: stainless steel (projected surface area: 24 cm <sup>2</sup> ) Anode: BDD (41 cm <sup>2</sup> ) and Ti/Pt-IrO <sub>2</sub> , Ti/SnO <sub>2</sub> -Sb (projected surface area: 24 cm <sup>2</sup> )	Anode materials, COD removal, salts evolution, by-products evolution	(Bagastyo et al., 2013)
RO concentrate from tertiary WWTP (Conductivity = 6 mS cm <sup>-1</sup> , [Cl <sup>-</sup> ] = 1.4 g L <sup>-1</sup> , COD = 140 mg-O <sub>2</sub> L <sup>-1</sup> , pH 7.7)	AO	Cathode: BDD (41 cm <sup>2</sup> ) Anode: BDD (41 cm <sup>2</sup> )	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 <sup>-1</sup> , [Cl <sup>-</sup> ] = 0.3-0.6 g L <sup>-1</sup> , DOC = 14-30 mg-C L <sup>-1</sup> , pH 7.6-7.9)	AO	Cathode: stainless steel (70 cm <sup>2</sup> ) Anode: BDD (70 cm <sup>2</sup> )	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 <sup>-1</sup> , [Cl <sup>-</sup> ] = 0.6 g L <sup>-1</sup> , COD = 5 mg-O <sub>2</sub> L <sup>-1</sup> , pH 8.2)	AO	Cathode: Ti/RuO <sub>2</sub> (60 cm <sup>2</sup> ) Anode: BDD (60 cm <sup>2</sup> )	COD and inorganic species evolution	(Van Hege et al., 2002)
RO concentrate pharmaceuticals WWTP (Conductivity = 2.7 mS cm <sup>-1</sup> , [Cl <sup>-</sup> ] = 0.5-0.8 g L <sup>-1</sup> , DOC = 17 mg-C L <sup>-1</sup> , pH 7.5)	AO	Cathode: stainless steel (70 cm <sup>2</sup> ) Anode: BDD (70 cm <sup>2</sup> )	Pollutant removal	(Urriaga et al., 2013)
RO concentrate from WWTP spiked with pharmaceuticals (Conductivity = 2.6 mS cm <sup>-1</sup> , [Cl <sup>-</sup> ] = 0.15 g L <sup>-1</sup> , TOC = 17 mg-C L <sup>-1</sup> , pH 8.4)	AO	Cathode: Pt Anode: BDD (12 cm <sup>2</sup> )	Kinetics of degradation and mineralization, by-products and pathway, ecotoxicity	(Lütke Eversloh et al., 2014)

RO concentrate from WWTP (Conductivity = 17 mS cm <sup>-1</sup> , [Cl <sup>-</sup> ] = 1.5 g L <sup>-1</sup> , DOC = 272 mg-C L <sup>-1</sup> , pH 8.3)	AO	Cathode: Ti (150 cm <sup>2</sup> ) Anode: Ti/PbO <sub>2</sub> (300 cm <sup>2</sup> )	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 <sup>-1</sup> , COD = 7.2 g-O <sub>2</sub> L <sup>-1</sup> , BOD <sub>5</sub> /COD = 0.03, [Cl <sup>-</sup> ] = 6 g L <sup>-1</sup> , Fe = 0.07 g L <sup>-1</sup> )	EF	Cathode: graphite (25 cm <sup>2</sup> ) Anode: Ti/TiO <sub>2</sub> (25 cm <sup>2</sup> )	Aeration, catalyst reusability, COD removal	(Baiju et al., 2018)
Landfill leachate (pH = 8.2, TDS = 9.5 g L <sup>-1</sup> , COD = 6.2 g-O <sub>2</sub> L <sup>-1</sup> , BOD <sub>5</sub> /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 <sup>-1</sup> , COD = 1.2 g-O <sub>2</sub> L <sup>-1</sup> , [Cl <sup>-</sup> ] = 1.6 g L <sup>-1</sup> , [NH <sub>3</sub> ] = 0.38 g L <sup>-1</sup> )	AO	Cathode: steel Anode: Ti/SnO <sub>2</sub> , Ti/PbO <sub>2</sub> (24 cm <sup>2</sup> )	pH, COD and ammonium evolution	(Cossu et al., 1998)
Landfill leachate (pH = 5.8-6.8, COD = 53.3 g-O <sub>2</sub> L <sup>-1</sup> , [Cl <sup>-</sup> ] = 2.6-4.5 g L <sup>-1</sup> , [SO <sub>4</sub> <sup>2-</sup> ] = 0.4-0.74 g L <sup>-1</sup> , [NH <sub>3</sub> ] = 0.9-1.3 g L <sup>-1</sup> )	AO	Cathode: stainless steel (1.5 m <sup>2</sup> ) Anode: Ti/Pt (0.85 m <sup>2</sup> )	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 <sup>-1</sup> , COD = 0.8 g-O <sub>2</sub> L <sup>-1</sup> , BOD <sub>5</sub> /COD < 0.1, [Cl <sup>-</sup> ] = 1.8 g L <sup>-1</sup> )	AO	Cathode: stainless steel Anode: Ti/PbO <sub>2</sub> (48 cm <sup>2</sup> )	pH, current density, COD removal, energy consumption	(Panizza et al., 2010)
Landfill leachate (pH = 8.3, conductivity = 12.8 mS cm <sup>-1</sup> , COD = 4.4 g-O <sub>2</sub> L <sup>-1</sup> , BOD <sub>5</sub> /COD = 0.14, [Cl <sup>-</sup> ] = 3.2 g L <sup>-1</sup> , [SO <sub>4</sub> <sup>2-</sup> ] = 0.04 g L <sup>-1</sup> , [NH <sub>3</sub> ] = 1.9 g L <sup>-1</sup> )	AO	Cathode: stainless steel (70 cm <sup>2</sup> ) Anode: BDD (70 cm <sup>2</sup> )	COD and ammonium removal	(Cabeza, Urtiaga, & Ortiz, 2007)
Landfill leachate (pH = 7.8, conductivity = 27.6 mS cm <sup>-1</sup> , COD = 4.2 g-O <sub>2</sub> L <sup>-1</sup> , BOD <sub>5</sub> /COD = 0.09, [Cl <sup>-</sup> ] = 3.2 g L <sup>-1</sup> , [SO <sub>4</sub> <sup>2-</sup> ] = 1.9 g L <sup>-1</sup> , [NH <sub>3</sub> ] = 2.3 g L <sup>-1</sup> )	AO	Cathode: Ta (10 cm <sup>2</sup> ) Anode: BDD (10 cm <sup>2</sup> )	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 <sub>2</sub> L <sup>-1</sup> , [Cl <sup>-</sup> ] = 1.2 g L <sup>-1</sup> , [SO <sub>4</sub> <sup>2-</sup> ] = 0.25 g L <sup>-1</sup> , [NH <sub>3</sub> ] = 0.22 g L <sup>-1</sup> )	AO	Cathode: graphite (30 cm <sup>2</sup> ) Anode: graphite/PbO <sub>2</sub> (30 cm <sup>2</sup> )	COD and DOC removal, inorganic evolution (sulfate, chloride, nitrate, bicarbonate)	(Mandal et al., 2020)

Landfill leachate (pH = 8.0, COD = 2.7 g-O <sub>2</sub> L <sup>-1</sup> , [Cl <sup>-</sup> ] = 2.9 g L <sup>-1</sup> , [NH <sub>3</sub> ] = 2.8 g L <sup>-1</sup> , Fe = 0.005 g L <sup>-1</sup> )	EF/AO	Cathode: Ti (projected surface area: 15 × 10 cm <sup>2</sup> ) Anode: Ti/RuO <sub>2</sub> -IrO <sub>2</sub> -SnO <sub>2</sub> -TiO <sub>2</sub> (projected surface area: 15 × 10 cm <sup>2</sup> )	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 <sup>-1</sup> , [Cl <sup>-</sup> ] = 5-6.2 g L <sup>-1</sup> , [SO <sub>4</sub> <sup>2-</sup> ] = 0.06-0.24 g L <sup>-1</sup> , [NH <sub>3</sub> ] = 1.9-3.2 g L <sup>-1</sup> )	EF/AO	Cathode: carbon felt (140 cm <sup>2</sup> ) Anode: Pt, BDD (24 cm <sup>2</sup> )	Current density, TOC removal efficiency	(Oturán et al., 2015)
Landfill leachate (pH = 7.8, COD = 1.0 g-O <sub>2</sub> L <sup>-1</sup> , [Cl <sup>-</sup> ] = 21 g L <sup>-1</sup> , [NH <sub>3</sub> ] = 0.44 g L <sup>-1</sup> )	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)

<p>Landfill leachate (pH = 8.5, conductivity = 17.8 mS cm<sup>-1</sup>, COD = 1.8 g-O<sub>2</sub> L<sup>-1</sup>, [Cl<sup>-</sup>] = 2.4 g L<sup>-1</sup>, [NH<sub>3</sub>] = 1.7 g L<sup>-1</sup>)</p>	<p>EC/AO</p>	<p>Cathode: graphite  Anode: Ti/RuO<sub>2</sub>-IrO<sub>2</sub>,  Ti/RuO<sub>2</sub>-IrO<sub>2</sub>-mZVI  (macroscopic zero-valent  iron) (15 cm<sup>2</sup>)</p>	<p>Anode material, voltage, TOC and COD removal, ammonia evolution,</p>	<p>(Sun et al., 2020)</p>
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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, [SO <sub>4</sub> <sup>2-</sup> ] = 0.45 g L <sup>-1</sup> )	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, [SO <sub>4</sub> <sup>2-</sup> ] = 12 g L <sup>-1</sup> , [Fe] = 0.61 g L <sup>-1</sup> )	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, [SO <sub>4</sub> <sup>2-</sup> ] = 11 g L <sup>-1</sup> , [Fe] = 0.92 g L <sup>-1</sup> )	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-/bipolar), 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 ( $168 \text{ cm}^2$ ) Anode: Fe ( $168 \text{ cm}^2$ )	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 <sup>2</sup> ) Anode: Fe (168 cm <sup>2</sup> )	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 <sup>2</sup> ) Anode: Fe, Al (168 cm <sup>2</sup> )	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 <sup>2</sup> ) Anode: Fe, Al (26 cm <sup>2</sup> )	Current density, electrode materials, iron removal, energy consumption	(Foudhaili et al., 2019)
Real acid mine drainage solutions (pH = 6.9-7.5, $[\text{SO}_4^{2-}] = 1.3\text{-}2.9 \text{ g L}^{-1}$ )	EC	Cathode: Fe (26 cm <sup>2</sup> ) Anode: Fe (26 cm <sup>2</sup> )	Toxicity assays	(Foudhaili, Jaidi, et al., 2020)
Iron in acid mine drainage model solution (pH = 2.4-2.6, $[\text{Fe}] = 0.47\text{-}0.52 \text{ g L}^{-1}$ , $[\text{SO}_4^{2-}] = 1.3\text{-}4.9 \text{ g}$ $\text{L}^{-1}$ ) and real acid mine drainage solution (pH = 2.4-	EC	Cathode: Fe (11 × 11 cm <sup>2</sup> ) Anode: Fe (11 × 11 cm <sup>2</sup> )	Current density, Sulfate concentration	(Foudhaili, Lefebvre, et al., 2020)

2.6, [Fe] = 0.04-0.07 mg L <sup>-1</sup> , [SO <sub>4</sub> <sup>2-</sup> ] = 1.3-2.9 g L <sup>-1</sup> )				
Acid mine drainage solution (pH = 3.0, [SO <sub>4</sub> <sup>2-</sup> ] = 1.9 g L <sup>-1</sup> , [Fe] = 0.45 g L <sup>-1</sup> )	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 <sup>-1</sup> )	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 <sup>2</sup> , respectively Anode: Ti/IrO <sub>2</sub> (7.2 cm <sup>2</sup> )	pH, current	(Bunce et al., 2001)
Iron/Sulfate in acid mine drainage model solution (pH = 1.9, [FeSO <sub>4</sub> ] = 0.56 g L <sup>-1</sup> )	AO	Cathode: reticulated vitreous carbon Anode: Ti/IrO <sub>2</sub> (7.2 cm <sup>2</sup> )	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 <sup>-1</sup> , Fe(II) = 0.30 g L <sup>-1</sup> )	AO	Cathode: stainless steel (10 cm <sup>2</sup> ) Anode: Ti/IrO <sub>2</sub> (7 cm <sup>2</sup> )	Reactor design	(Wang et al., 2003)
Acid mine drainage solution (pH = 2.4, [SO <sub>4</sub> <sup>2-</sup> ] = 1.3 g L <sup>-1</sup> , [Fe] = 0.23 g L <sup>-1</sup> )	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 <sub>4</sub> <sup>2-</sup> ] = 1 g L <sup>-1</sup> )	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 <sub>2</sub> L <sup>-1</sup> , TS = 15.9 g L <sup>-1</sup> )	EC	Cathode: Fe (11.5 × 6 cm <sup>2</sup> ) Anode: Fe (11.5 × 6 cm <sup>2</sup> )	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 <sub>2</sub> L <sup>-1</sup> , DS = 23.3 g L <sup>-1</sup> , Total coliforms = 7.9 × 10 <sup>6</sup> MPN)	Fered-Fenton	Cathode: graphite (10 × 6 cm <sup>2</sup> ) Anode: graphite (10 × 6 cm <sup>2</sup> )	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 <sub>2</sub> L <sup>-1</sup> , TSS	Electrochemical peroxidation/EF	Cathode: steel (5 × 6.5 cm <sup>2</sup> )	Current density, pH, TSS & COD & TOC removal, inorganic evolution, cost evaluation	(Olvera-Vargas et al., 2019)

= 31.3 g L <sup>-1</sup> , TDS = 9.4 g L <sup>-1</sup> , Total coliforms = 2.4 × 10 <sup>3</sup> CFU mL <sup>-1</sup> )		Anode: steel (5 × 6.5 cm <sup>2</sup> )		
Activated sludge from municipal WWTP (pH 6.7-7.0, COD = 17.4-19.0 g-O <sub>2</sub> L <sup>-1</sup> , TS = 8-18.2 g L <sup>-1</sup> )	AO	Cathode: Ti/RuO <sub>2</sub> (70 cm <sup>2</sup> ) Anode: Ti/RuO <sub>2</sub> (70 cm <sup>2</sup> )	Current density, pH, type of salt, cost evaluation	(Song et al., 2010)
Municipal wastewater sludge	AO	Cathode: Ti (908 cm <sup>2</sup> ) Anode: Ti/RuO <sub>2</sub> (504 cm <sup>2</sup> )	Current density, electrolyte concentration	(Bureau et al., 2012)
Activated sludge from paper mill WWTP (pH 7.8-8.2, COD = 0.55 g-O <sub>2</sub> L <sup>-1</sup> , TS = 16.3 mg L <sup>-1</sup> , Total coliforms = 2.8 × 10 <sup>5</sup> CFU g-DS <sup>-1</sup> )	AO	Cathode: Ti (908 cm <sup>2</sup> ) Anode: Ti/RuO <sub>2</sub> (504 cm <sup>2</sup> )	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 <sup>-1</sup> )	AO	Cathode: Ti/PbO <sub>2</sub> (10 × 10 cm <sup>2</sup> ) Anode: Ti/PbO <sub>2</sub> (10 × 10 cm <sup>2</sup> )	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 <sub>2</sub> L <sup>-1</sup> , TS = 9.1-13.7 g L <sup>-1</sup> )	AO	Cathode: Ti/RuO <sub>2</sub> (44 cm <sup>2</sup> ) Anode: Ti/RuO <sub>2</sub> (44 cm <sup>2</sup> )	pH, current density	(Heng & Isa, 2014)

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

		cm <sup>2</sup> ) and Fe (16 cm <sup>2</sup> )		
Activated sludge from municipal WWTP (pH 7.0, soluble COD = 0.14 g-O <sub>2</sub> L <sup>-1</sup> , MLSS = 2.5 g L <sup>-1</sup> , VSS/TSS = 0.68)	AO/Fered-Fenton	Cathode: Ti (10 × 10 cm <sup>2</sup> ) Anode: Ti/RuO <sub>2</sub> -IrO <sub>2</sub> (10 × 10 cm <sup>2</sup> )	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 <sup>-1</sup> , VSS/TSS = 0.46-0.52)	AO, AO/EF	Cathode: Ti (8 × 7 cm <sup>2</sup> ) Anode: Ti/RuO <sub>2</sub> -IrO <sub>2</sub> (8 × 7 cm <sup>2</sup> )	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 <sub>2</sub> L <sup>-1</sup> , TSS = 24 g L <sup>-1</sup> )	AO, AO/EF	Cathode: Ti, active carbon fiber (10 × 7 cm <sup>2</sup> ) Anode: Ti/RuO <sub>2</sub> (10 × 7 cm <sup>2</sup> )	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 <sup>2</sup> ) Anode: Pt grid (3 cm diameter, 4.5 cm height)	Current density	(Murati et al., 2009)
Phenanthrene (17 mg L <sup>-1</sup> )	Tween 80 (0.75 g L <sup>-1</sup> ) and HPCD (10 g L <sup>-1</sup> )	Soil washing/EF	Cathode: carbon felt (150 cm <sup>2</sup> ) Anode: Pt grid (3 cm diameter, 5 cm height)	[Fe <sup>2+</sup> ], current density, biodegradability and toxicity of solution	(Mousset et al., 2014a)

Phenanthrene (16 mg L <sup>-1</sup> )	HPCD (9 g L <sup>-1</sup> )	Soil washing/EF	Cathode: carbon felt (10 cm <sup>2</sup> ) Anode: Pt grid (3 cm diameter, 5 cm height), Ti/RuO <sub>2</sub> -IrO <sub>2</sub> (40 cm <sup>2</sup> ), BDD (40 cm <sup>2</sup> )	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 <sup>2</sup> ) Anode: Pt sheet (1 cm <sup>2</sup> )	Current density, toxicity of solution	(Hanna et al., 2005)
16 polycyclic aromatic hydrocabons (PAHs) (1090 mg kg <sup>-1</sup> )	Tween 80 (7.5 g L <sup>-1</sup> ) or HPCD (7.5 g L <sup>-1</sup> )	Soil washing/EF	Cathode: carbon felt (150 cm <sup>2</sup> )	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 <sup>-1</sup> )	Tween 80 (11 g L <sup>-1</sup> )	Soil flushing/EF	Cathode: carbon felt (150 cm <sup>2</sup> ) Anode: BDD (40 cm <sup>2</sup> )	pH, biodegradability and toxicity of solution	(Huguenot et al., 2015)
Phenanthrene (500 mg kg <sup>-1</sup> )	SDS or Tween 80 (1 g L <sup>-1</sup> )	Soil washing/AO	Cathode: stainless steel (78 cm <sup>2</sup> ) Anode: DSA (78 cm <sup>2</sup> ), BDD (78 cm <sup>2</sup> )	anode materials, pH, conductivity	(Sáez et al., 2010)
Oxyfluorfen (100 mg kg <sup>-1</sup> )	SDS (0.1-5 g L <sup>-1</sup> )	Soil washing/AO	Cathode: stainless steel (78 cm <sup>2</sup> )	Concentration of extracting agent, turbidity, zeta	(dos Santos et al., 2016)



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

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

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

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

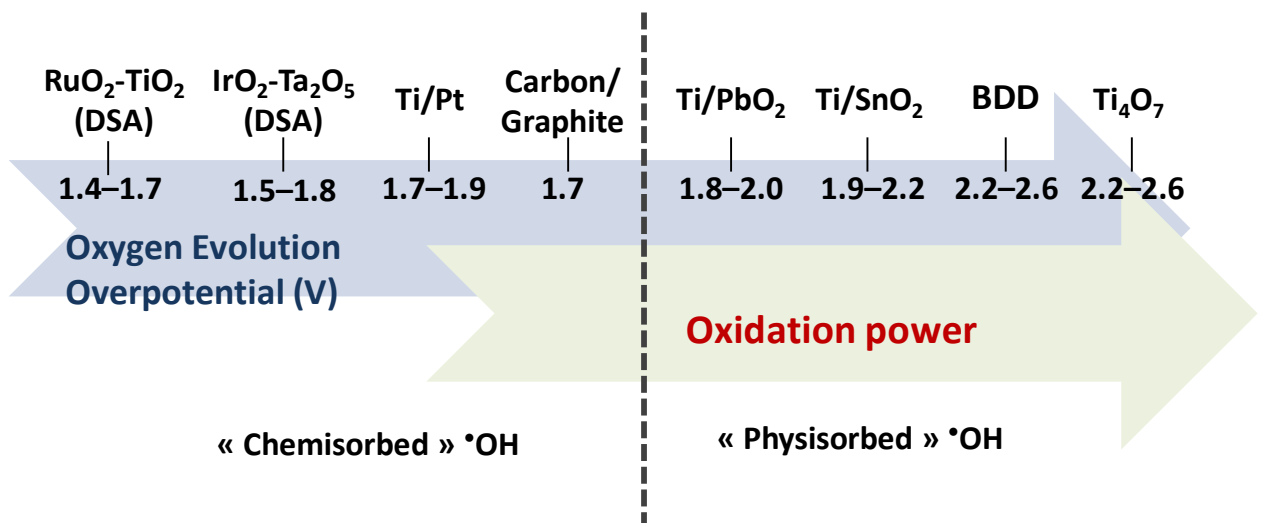


Figure 1. Classification of the electrode materials used in AO.

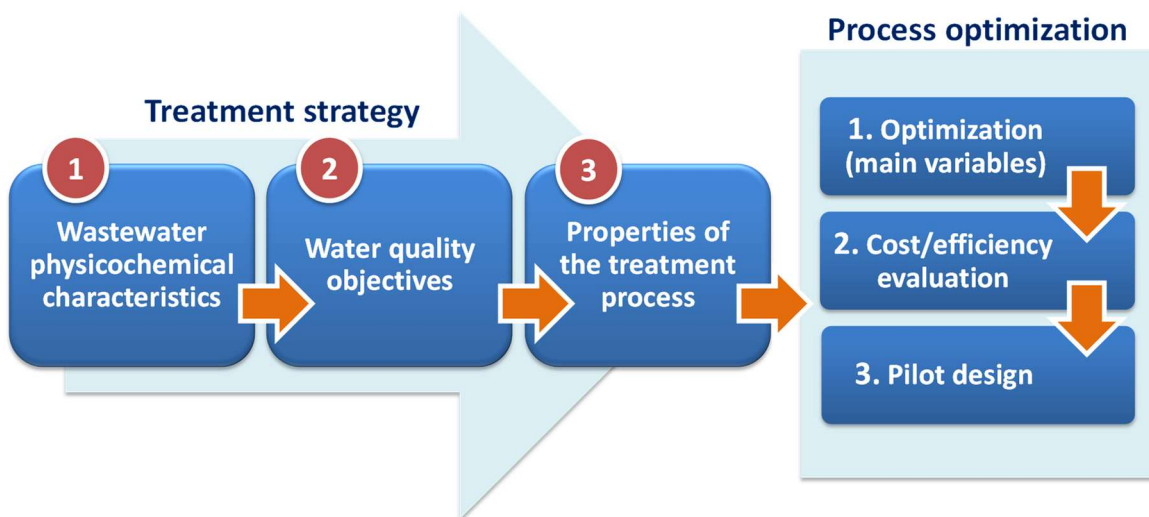


Figure 2. Main steps involved in the selection of the most suitable treatment strategy.

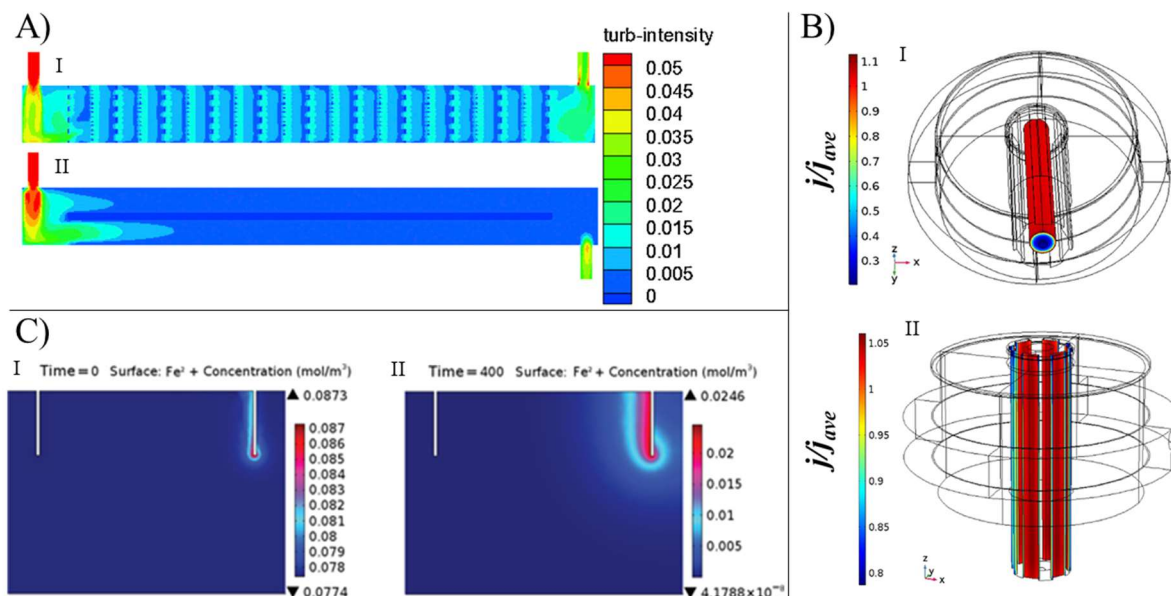


Figure 3. CFD simulation for a) the turbulent intensity distribution in a vertical-flow tubular electrochemical reactor (I) and a concentric tubular electrochemical reactor (II) for AO of phenol wastewater, b) normalized current density distribution in a rotating cylinder electrode (RCE) reactor for 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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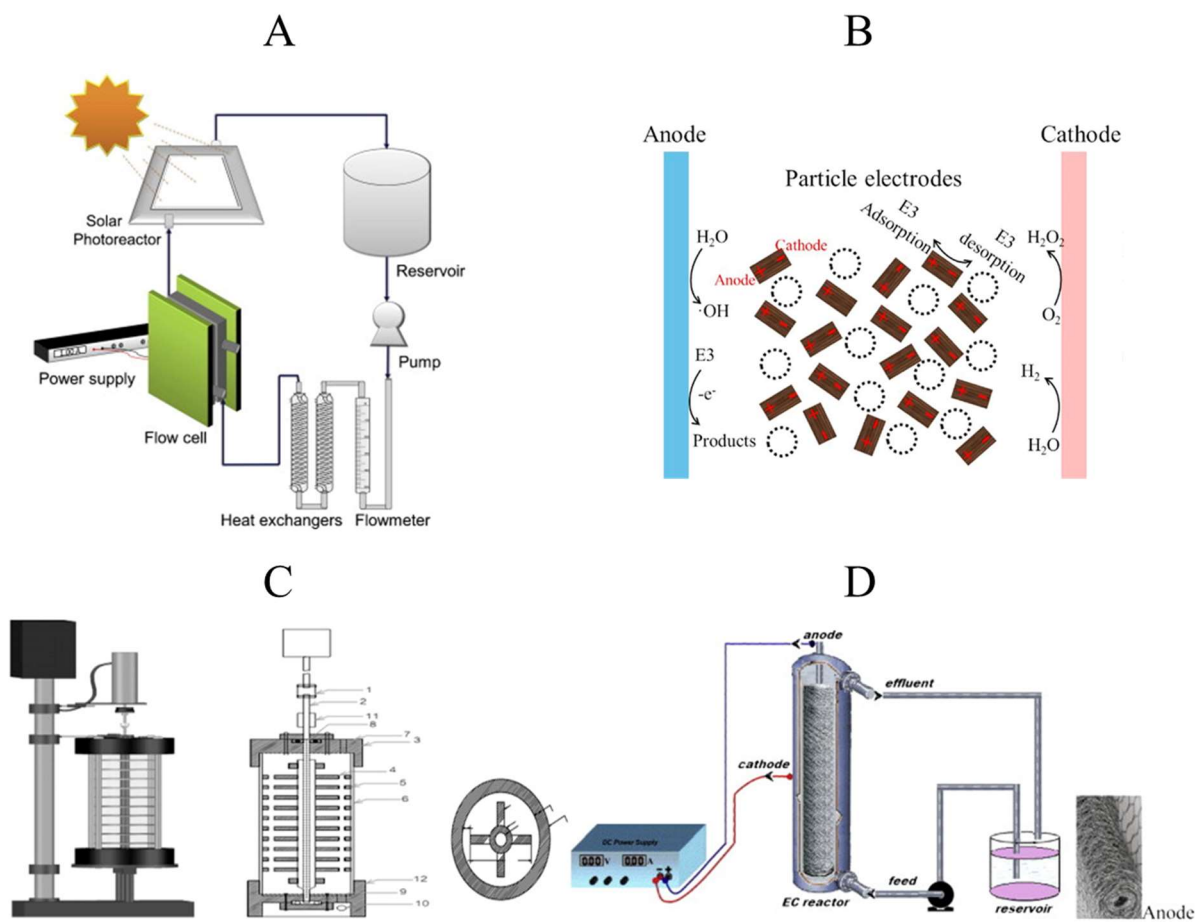


Figure 4. Reactor designs for EAOPs, a) Filter press reactor, b) fluidized bed reactor, c) Rotating electrode reactor and d) fixed bed reactor (Adapted with permission from: (Naje et al., 2016; Olvera-Vargas et al., 2015; Shen et al., 2017; Tezcan Un & Aytac, 2013)). Copyright Elsevier.

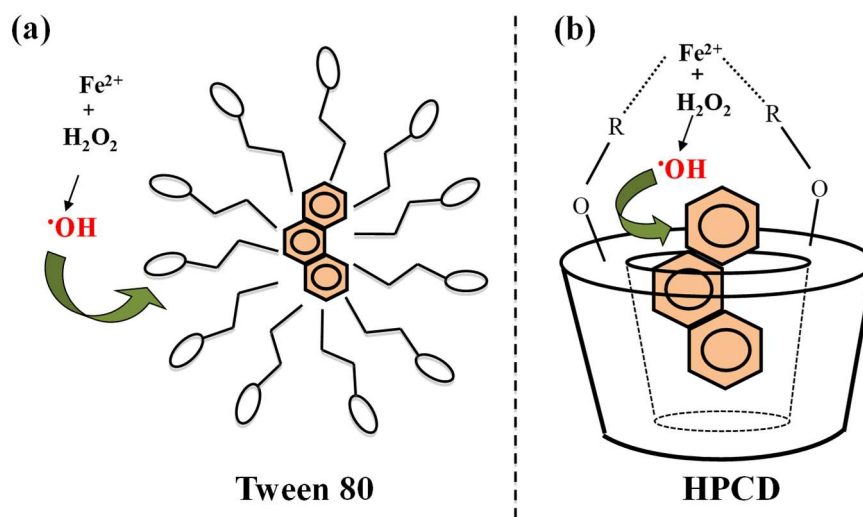


Figure 5. Schematic representation of two different ways of •OH oxidative degradation of hydrophobic organic pollutant in the presence of (a) surfactant (Tween 80) or (b) cyclodextrin (HPCD) in aqueous solution. Adapted with permission from (Mousset et al., 2014a). Copyright Elsevier.

### Figure captions

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