



## Current perspective of advanced electrochemical oxidation processes in wastewater treatment and life cycle analysis

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### ARTICLE INFO

#### Keywords:

Advanced oxidation process  
Wastewater treatment  
Real application  
Life cycle analysis

### ABSTRACT

The water crisis is one of the most alarming problems of this century. Thus, strategies are being sought not only to manage water use, but also to reuse it through efficient treatments that make it possible to achieve a circular water economy. In this sense, Electrochemical Advanced Oxidation Processes (EAOPs) have been shown to produce water with the quality necessary for its reuse. The application of EAOPs in water treatment plants is one of the current needs, so research efforts are focused on scaling them up to an industrial level. The application of these processes is limited, mainly due to energy consumption; however, by means of optimal conditions and designs, different studies have shown that the application of these processes can be successful and compete with conventional treatments. This work compiles studies of applications of EAOPs to treat real wastewater, where the removal efficiencies, optimal operating conditions and their energy consumption or operational cost are indicated. Advantages, disadvantages, and future challenges or needs of the different EAOPs are also presented. An analysis of studies on Life Cycle Analysis (LCA) of EAOPs is also presented and the need to standardize the functional unit to conduct such studies was concluded.

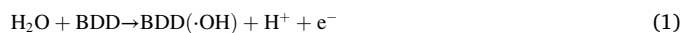
### 1. Introduction

The correct treatment of wastewater is a relevant topic within environmental conservation, pursuing sustainable development. To address this issue, there have been developed different techniques (physical, chemical or biological)

or a combination of them. In this sense, within water treatment plants, biological treatment is the most widely used. Biological treatments, however, currently do not respond correctly to the needs of final water quality since they have not demonstrated to be efficient in the treatment of landfill leachates and removal of recalcitrant compounds present at high concentration in industrial effluents and with extreme pH. In addition, their operation tends to be in very long periods of time and large operational areas. Faced with these inefficiencies and disadvantages, there is a growing need to find environmentally friendly technologies that provide adequate wastewater treatment and above all a viable application [1–3].

Advanced oxidation processes (AOPs) have been demonstrated to be a possible option for the efficient treatment of wastewater and their

plausible application within water treatment plants. AOPs are physico-chemical processes that have the main advantage of operating at pressure and temperature close to ambient conditions, degrading the contaminants present in the water to other more environmentally friendly molecules or achieving total mineralization [4,5]. AOPs can act in a non-selective but very active way, since they involve the generation of highly oxidizing species, i.e hydroxyl radicals ( $\cdot\text{OH}$ ) via Eq. 1, which have a high oxidation potential (2.8 V) and are responsible for the organic pollutants removal via oxidation [6,7]. In addition to  $\cdot\text{OH}$  (Eq. 1), other reactive oxygen species (ROS) generated through AOPs, for example by anodic oxidation using Boron-Doped Diamond (BDD) electrodes can be produced, i.e.  $\text{O}_3$  (Eq. 2),  $\text{S}_2\text{O}_8^{2-}$  (Eq. 3)  $\text{O}_2^{\cdot-}$ , and  $\text{HO}_2^{\cdot}$  are also capable of degrading pollutants. And recently, it has been shown that ions, like  $\text{Cl}^-$  can produce  $\text{Cl}^{\cdot}$  (Eq. 4),



In the presence of  $\text{SO}_4^{2-}$ ,

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In the presence of  $\text{Cl}^-$ ,



However, research on the application of AOPs in plants to treat wastewater, is a topic of current interest as studies on the feasibility of these processes are required. Table 1 shows an analysis of different advanced oxidation processes, their advantages, limitations and needs, in order to establish the current situation in the application of these processes [8–14].

It is important to mention that electrocoagulation is not an advanced oxidation process; however, since it is an electrochemical treatment with high efficiency and application in treatment plants, it has been included in this table and will be below described in more detail.

This work aims to concentrate and analyze the literature related with electrochemical advanced oxidation processes (EAOPs) applied to real

wastewater recovery. In addition, studies regarding life cycle assessment of these EAOPs are also revised.

## 2. Electrochemical cell design

The main requirements on the application of electrochemical advanced oxidation processes such as energy consumption have been studied to optimize the processes. In this context, other aspects that deserve special attention are the design of the electrochemical cell and the electrodes design (area and material). The former must be oriented to obtain a uniform current and a distribution of potential aiming to optimize power consumption in relation to selectivity and rate of oxidation.

A mechanism for electrochemical reactions comprises four main stages: a) transfer of mass to the electrode, b) adsorption and desorption at the electrode surface, c) direct transfer of electrons at the electrode surface, and d) chemical reactions linked with electron transfer. The

**Table 1**  
Insight into advanced oxidation processes for real application.

Technology	Advantages	Limitations	Needs
Electro-oxidation	<ul style="list-style-type: none"> <li>Highly efficient in the removal of pollutants by degradation and mineralization.</li> <li>No need to add chemicals (environmentally friendly).</li> <li>Potential of oxidation is stable in a wide pH range.</li> <li>Wide variety of electrode materials</li> <li>Operation is easy and the equipment to conduct it is simple.</li> <li>Little or no sludge production</li> <li>Renewable energy like solar can be used.</li> </ul>	<ul style="list-style-type: none"> <li>High cost of electrodes</li> <li>High energy consumption</li> <li>Anode instability and deposition of substances on its surface (e.g. due to foaming caused by surfactants and corrosion inhibitors).</li> <li>Removal efficiencies highly dependent on influent flow rate and initial COD concentration.</li> </ul>	<ul style="list-style-type: none"> <li>Electrodes to obtain high efficiencies and reduce treatment time.</li> <li>New sources of electrical energy to increase sustainability of the process</li> </ul>
Ozone ( $\text{O}_3$ )	<ul style="list-style-type: none"> <li>Easy operation since ozone can be electrogenerated on site, thus storage is not required.</li> <li>Low maintenance and operating costs</li> <li>Efficient over a wide pH range, acting through different mechanisms.</li> <li>High efficiency as an oxidant and disinfectant.</li> <li>It is currently the most widely used advanced oxidation process in water treatment plants.</li> </ul>	<ul style="list-style-type: none"> <li>Need for an ozone generator (however, there is the possibility of using electrogenerated ozone).</li> <li>High voltages and air or oxygen supply required when generated by Corona Discharge method.</li> <li>Efficiency of removal is slightly lower than other AOPs.</li> </ul>	<ul style="list-style-type: none"> <li>To improve the efficiency of organic compounds removal.</li> <li>Research on ozone electrogeneration, electrode materials and cell design.</li> <li>Combine oxidation processes and other stages of wastewater treatment to increase efficiency.</li> </ul>
Electrocoagulation	<ul style="list-style-type: none"> <li>The use of external chemicals is not required, which reduces the cost of acquisition, transportation and storage and there is no need to neutralize them at the end of the treatment.</li> <li>The resulting water is clear and odor is also removed.</li> <li>The produced solids can be easily separated by settling.</li> <li>It generates effluent with lower total dissolved solids content, which allows for lower water recycling costs if the water is reused.</li> <li>The operation costs are minimal compared to the conventional process using polymers.</li> </ul>	<ul style="list-style-type: none"> <li>The "sacrificial electrodes" dissolve because of oxidation and must be replaced regularly.</li> <li>The use of electricity can be expensive in many places.</li> <li>Over time it can lose efficiency, due to the occurrence of electrode passivation.</li> <li>The treatment of produced sludge</li> <li>High conductivity of the wastewater is required.</li> </ul>	<ul style="list-style-type: none"> <li>Use of solar systems as a response to energy consumption,</li> <li>Coupling with other technologies.</li> <li>Develop a sustainable approach to the utilization of the <math>\text{H}_2</math> gas generated.</li> </ul>
Electrofenton	<ul style="list-style-type: none"> <li>Higher removal efficiency than traditional Fenton process.</li> <li>Lower reagent cost because no addition of reagents is required, as <math>\text{Fe}^{2+}</math> and <math>\text{H}_2\text{O}_2</math> can be generated electrically.</li> <li>Lower iron concentration required (regenerates constantly and rapidly as <math>\text{Fe}^{2+}</math> at the cathode)</li> </ul>	<ul style="list-style-type: none"> <li>Acidic pH conditions are required (the need for acidification and final requalification of the aqueous matrix)</li> <li>Special designs are required to separate the formed sludge. This sludge is a precipitate of iron compounds formed at pH higher than 3.</li> <li>High cost of electrode materials and energy consumption</li> </ul>	<ul style="list-style-type: none"> <li>Appropriate tank, cells and system designs.</li> <li>Research on low-cost electrode materials.</li> <li>Energy supply by solar radiation (photoelectrofenton).</li> </ul>
Electro-photocatalysis	<ul style="list-style-type: none"> <li>Better efficiency than classical catalysis and photocatalysis</li> <li>High efficiency in short times</li> <li>Reduced recombination rate of photogenerated electrons and holes</li> <li>Improved removal efficiency also due to generation of active chlorine from chloride ions.</li> </ul>	<ul style="list-style-type: none"> <li>Energy consumption is higher than traditional photocatalysis</li> <li>Higher costs than the traditional electrocatalytic process, except when photocatalysis is driven by sun-light.</li> <li>The cost of separating the catalyst once it has been used.</li> <li>Need for more extensive pretreatment to improve uniform radiation distribution over the entire surface of the catalyst on a larger scale.</li> </ul>	<ul style="list-style-type: none"> <li>Maximization of efficiency towards special reactor configuration and flow pattern</li> <li>Economic feasibility still under evaluation</li> </ul>
Ultrasound-assisted electrochemical treatment	<ul style="list-style-type: none"> <li>Higher <math>\bullet\text{OH}</math> production.</li> <li>Lack of reagents.</li> <li>Parameter versatility.</li> <li>Not require any extra chemicals</li> </ul>	<ul style="list-style-type: none"> <li>Parameter versatility.</li> <li>Uncontrolled by products.</li> <li>Targets low-concentration wastewater.</li> </ul>	<ul style="list-style-type: none"> <li>Appropriate system designs.</li> <li>System for large volumes of viable wastewater</li> </ul>

design of electrochemical cells for oxidation processes holds significant importance as it enables the attainment of elevated degradation and energy efficiencies by ensuring proper mass transport, appropriate materials, electrodes, and cell geometry. It is desirable that cells meet certain characteristics: i) low cost of electrode materials, maintenance, and operation, ii) easy operation and simplicity during scale-up, iii) convenience and reliability, proper design, installation, operation and maintenance and monitoring procedures, iv) low cell potential difference, v) large surface area electrodes working at uniform current density and v) high potential and conversion rates that could be achieved with high mass transport rates [15].

In the process of scaling up electrochemical AOPs, the configuration of an electrochemical cell plays a fundamental role for its viable application. There are key components to consider: reactor configuration, flow mode, operation mode and electrode construction. The correct selection of these components allows maximizing pollutant degradation and energy cost reduction [16–18]. Batch electrochemical cells are usually used at laboratory scale, due to the simplicity of the system configuration and the relatively low initial cost. Also, this configuration is the preferred one when starting a treatment because it allows to have a better control of the operating conditions and their influence on degradation; however, these conditions are constantly changing and are often far from the real conditions of a treatment plant. For this reason, the design of electrochemical cells seeking scale-up is oriented towards a continuous flow regime more suitable for treating large volumes of wastewater, which is characterized by decreasing mass transfer problems due to turbulence promoters, as well as avoiding electrode passivation, resulting in a higher production of oxidizing agents. In addition, continuous flow improves faradaic efficiency, resulting in lower energy consumption, higher selectivity, and lower electrolyte loads than with batch mode. Lower electrolyte loadings reduce waste streams and simplify purification systems. Table 2 shows the differences in operating parameters depending on the operating mode that are important to consider for scale-up [19].

Continuous flow reactors can operate by flow-by or flow-through. In flow-by the fluid flow direction is parallel to the electrodes, so that the aqueous solution cannot pass through the electrode, so the geometry is usually a plate. This geometry limits the active surface of the electrode, which is where the degradation occurs either directly or indirectly of the contaminants, so there is a limited mass transfer that results in low treatment efficiency and high energy consumption. However, these mass transfer problems can be overcome by modifying the fluid flow rate, switching between laminar and turbulent flow, determining the residence time of the reactants in the reactor and on the electrodes surface. Studies have been carried out comparing the degradation rates depending on the type of flow within the system [18]. On the other hand, the continuous flow-through mode refers to when the fluid direction is perpendicular to the electrode; this type of flow requires porous or mesh electrodes between 0.1 and 1.0 mm wide [61]. By using this type of electrodes, a larger active area is obtained, which could

**Table 2**  
Operating mode influence on electrochemical reactor parameters. Taken with permission from Elsevier [19].

Parameter	Operating mode	
	Batch	Flow-through
Mass transfer	Complex	Clearly defined
Electrode passivation	High	Low
Electrode distance	Large	Small (zero-gap)
Potential/current distribution	Less uniform	Uniform
Ohmic drop	High	Low
Temperature regulation	Poor	Good
Area/Volume ratio	Small	High
Treated volume	Large	Small
Mineralization rate	Slow	Fats
Energy use	High	Low

increase the mass transfer of the reagent, thus improving the degradation efficiency of different pollutants. In addition, the porous structure could improve the current efficiency by providing more reaction sites and reducing the applied current. Therefore, it is beneficial to improve the electron utilization efficiency and increase the electrode lifetime [20]. A study by Perez et al., to obtain high degradation efficiency and low energy consumption for wastewater treatment, compared the development of a microfluidic flow-through cell with a narrow internal electrode spacing with a commercial continuous flow cell, and they obtained that the developed microfluidic cell required 4–10 times less current and 6–15 times less energy consumption than the commercial continuous flow cell [21].

Fig. 1 shows two different commercial cells that have been tested for electro-oxidation (DiaClean®) and ozone generation (CabECO®). These cells have a continuous flow configuration and are equipped with boron-doped diamond electrodes. The CabECO® cell is a PEM electrolyser, it has a MEA (Membrane Electrode Assembly) with BDD electrode and Nafion membrane. DiaClean® and CabECO® have been tested by different research groups for wastewater treatment and disinfection and have been inserted in real applications [16,22–26].

As mentioned above, a correct cell design allows us to minimize energy consumption, which is one of the most debated aspects of AOPs applications Vs. conventional treatments. The energy consumption,  $EC$ , is defined by Eq. (5) [29], where  $U$  is voltage (V),  $i$ , is current (A),  $t$  is time in h,  $v$  is the sample volume (L),

$$EC \left( \frac{kWh}{L} \right) = \frac{U * i * t}{v} \quad (5)$$

The cost per liter of wastewater treated can be calculated with Eq. (6) [30], where the cost is expressed in US dollars per liter (US\$/L).

$$Cost \left( \frac{US\$}{L} \right) = EC \left( \frac{kWh}{L} \right) (\$kW) \quad (6)$$

### 3. Application of electrochemical advanced oxidation processes

The application of electrochemical advanced oxidation processes (EAOPs) in water treatment plants or in real water is a topic of current interest, because once their efficiency is known in the laboratory, their industrial scaling-up is necessary. In order to do so, all the needs and disadvantages shown in Table 1 must be overcome. To overcome these disadvantages, the study of electrode materials, the correct design of the reactor and the knowledge of the optimum operating conditions are essential. Some of the advanced oxidation processes nowadays have been successfully applied, reaching good efficiencies, and demonstrating their feasibility.

#### 3.1. Electro-oxidation

Electrochemical oxidation is known to generate high amounts of hydroxyl radicals, which are highly oxidant and capable of achieving complete mineralization of contaminants, i.e. transforming them into carbon dioxide, water, and inorganic molecules. Electrochemical oxidation can occur by two different mechanisms; directly the pollutants are oxidized at the electrode-solution interface and indirectly, the hydroxyl radicals generated from the oxidation of water are responsible for the degradation [31,32]. In addition, if the wastewater contains sulfate and chloride salts (which is a common situation in real waste), these species can be oxidized by direct electron transfer at the electrode surface or by the electrogenerated  $\bullet OH$ , forming more stable oxidants, which can act in the bulk solution during treatment. This production of oxidants helps to minimize the problems associated with mass transport limitations, which are often encountered during electrochemical treatment of poorly concentrated wastewater. As is known, the main mediator species for chloride and sulfate containing solutions are hypochlorite and peroxosulfate, respectively [33].

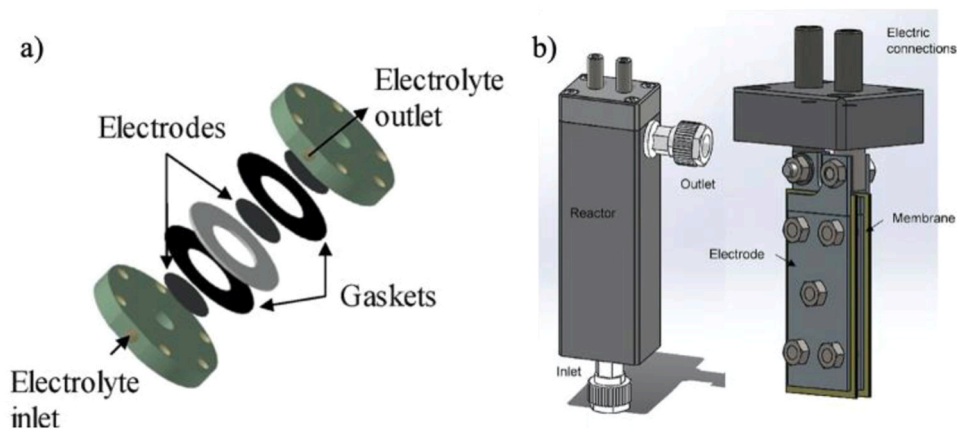


Fig. 1. Commercial electrochemical cells a) DiaClean® Taken with permission from Elsevier [27], b) CabECO® Taken with permission from Elsevier [28].

Within this technology the electrode material is one of the key parameters. One of the most studied and used materials in the last two decades are Boron-Doped Diamond (BDD) anode electrodes due to their properties such as high stability even at acidic pH, wide potential window, and low absorption [34]. In wastewater treatment, electrodes with a high oxygen evolution overpotential are preferred because a complete mineralization of the pollutants to carbon dioxide is achieved, which is a very important feature of BDD. At the same time these electrodes not only achieve the electrogeneration of hydroxyl radicals, but also the

production of other oxidizing species is possible: ozone ( $O_3$ ), hydrogen peroxide ( $H_2O_2$ ), active chlorinated species (hypochlorous acid ( $HClO$ ), chlorine gas ( $Cl_2$ )), peroxodisulfate ( $S_2O_8^{2-}$ ), ferrate ( $Fe_2O_4^{2-}$ ), peroxydicarbonate ( $C_2O_6^{2-}$ ), and peroxodiphosphate ( $P_2O_4^{4-}$ ) [19]. All these characteristics make BDD electrodes attractive for wastewater treatment and have been applied with great efficiency to many different types of complex organic molecules. However, these characteristics can at the same time play against the use of BDDs in some wastewater qualities or in cases where selective degradation is sought, due to their high removal

Table 3

Real applications of electro-oxidation processes.

Wastewater	Cell design	Efficiency	Energy consumption or cost	Ref.
Pharmaceutical/hospital wastewater	CPC reactor Anode: (Nb-BDD) Cathode: carbon-poly- tetrafluoroethylene (PTFE) GDE 73.6 mA/cm <sup>2</sup>	100% removal of Pentaclorofenol 100% removal of Diclofenaco 84.1% removal of Terbutryn	5.0 kWh/m <sup>3</sup>	[41]
Wastewater from a treatment plant	Microreactor flow by Anode: BDD Cathode: Nickel IE gap (μm): 50 Current density (A/ m <sup>2</sup> ): 160 Flow rate (mL/min): 0.1–0.5	60–80% TOC removal	0.4 \$/m <sup>3</sup>	[42]
Wastewater of petroleum industry (Ferry Terminal Almirante Barroso)	Plug-flow reactor (PFR) Anode and cathode: Titanium and Ti/ RuO <sub>2</sub> Time: 60 min Current density (mA/cm <sup>2</sup> ): 30 Flow rate (mL/ min): 0.54	100% COD removal	40.2 kWh/ kg COD US\$ 38/kg COD	[43]
Distillery wastewater	Batch reactor Anode: Ti-RuO <sub>2</sub> Cathode: Ti Current (A): 2 Current density (mA/cm <sup>2</sup> ): 20	56.86% COD removal	3.36 mg COD/Wh	[44]
Industrial cattle slaughterhouse wastewater	Monopolar batch bench reactor Anode: MMO-O <sub>2</sub> (Ti/IrO <sub>2</sub> -Ta <sub>2</sub> O <sub>5</sub> ) Cathode: Stainless steel Time: 5 h Current density (mA/cm <sup>2</sup> ): 20	81.66% TOC removal	0.034 kWh/m <sup>3</sup>	[45]
Textile wastewater	Batch reactor Anode: DSA-Cl <sub>2</sub> (Ti/Ru <sub>0.3</sub> Ti <sub>0.7</sub> O <sub>2</sub> ) Cathode: 304 Stainless steel Time:200 min Current density (mA/cm <sup>2</sup> ): 30	100% COD removal	0.24–0.41 kWh/m <sup>3</sup>	[46]
Real high salinity wastewater	Continuous membrane flow cell Anode: β-PbO <sub>2</sub> /Ti Cathode: Carbon/polytetrafluoroethylene Proton exchange membrane (Fumasep®FKD-PK-75) Anolyte: (NH <sub>4</sub> ) <sub>2</sub> SO <sub>4</sub> Catholyte: H <sub>2</sub> SO <sub>4</sub> Time: 240 Cell Voltage:2.5 V	100% COD removal	146.94 kWh/ m <sup>3</sup>	[47]

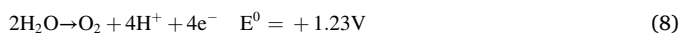
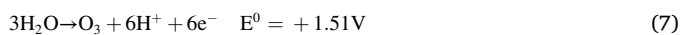
power. For example, Hao et al. evaluated the mineralization of phenol using BDD anodes. Their findings revealed that the mineralization started to be significant after phenol concentration decrease was appreciable. reduction in TOC commenced subsequent to a substantial decrease in phenol concentration [35]. In this sense, BDD electrodes resulted in better results than those attained with DSA electrodes. Nevertheless, perchlorate, along with other unwanted chlorinated byproducts, was detected at the conclusion of the electrolysis process, posing challenges for treated water or water.

On the other hand, one of the main limitations of BDD electrodes has been their high cost [36]. In response to this problem, the study of electrode materials that share the same characteristics as BDD but at a lower cost has been one of the research trends within this technology. Thus, electrodes such as Mixed metal oxide (MMO) electrodes have achieved better performances and lower energy consumption compared to BDD [37–39]. A study on concentrated reverse osmosis treatment showed that the Instantaneous Current Efficiency (ICE) at 25 mA and one hour of treatment was 0.295, 0.234 and 0.288 with BDD, Ti/Ir-O<sub>2</sub>-RuO<sub>2</sub> and Ti/IrO<sub>2</sub>-Ta<sub>2</sub>O<sub>5</sub> respectively, and the energy consumption of 0.158, 0.048 and 0.050 kWh/g Chemical Oxygen Demand (COD) respectively, which shows that the CE (Current Efficiency) is slightly higher in BDD electrodes, but the energy consumption is three times higher with BDD electrodes than with DSA [40]. Likewise, the design of cells in continuous systems has been shown to reduce energy consumption. Table 3 shows real applications of electrooxidation, the cell design, electrodes, operating conditions and efficiencies achieved.

In the application of EAOPs, specifically electro-oxidation, it is important to carry out toxicity studies, since the process can reduce organic matter, but at the same time it can produce by-products that increase the toxicity of the treated water. A study of the degradation of tetracycline by electro-oxidation shows that even with 85% TOC removal after 30 min of treatment at least 3 intermediates considered toxic are present, and these compounds degrade to carboxylic acid after 40 min of treatment with 95% of TOC removal [48]. Ganiyu S. et al., studied the degradation of Amoxicillin (AMX) using Ti<sub>4</sub>O<sub>7</sub> electrodes and observed that during the first minutes of oxidation the toxicity of water increased due to the presence of intermediates that were more toxic than AMX up to 240 minutes, subsequently bioluminescence inhibition reached its minimum value after 360 minutes of electrolysis, indicating mineralization/degradation of both AMX and its oxidation reaction intermediates into less toxic and biodegradable short-chain carboxylic acid [49]. Accordingly, toxicity analysis is indispensable in EAOPs because a high percentage of removal can be achieved but water might have a high or important toxicity, and this water cannot pass to a second biological treatment or be reused.

### 3.2. Electrogenated ozone

Ozone can be electrogenerated by oxidation of water. For this to happen, it is necessary to work with electrodes that present a high anodic overpotential for the oxygen evolution reaction or to apply quite large current densities, since the electrogeneration of ozone might compete with the production of oxygen that occurs at a lower potential (Eqs. 7 and 8),



The main advantage of ozone electrogeneration over conventional technologies such as Corona Discharge is that the production of ozone through water and directly in it is feasible, which improves its solubility and efficiency in organic degradation. Also, that air or oxygen is not needed since ozone is generated by water through reaction 7. As it is known, ozone shows low solubility and stability in water, so by its electrochemical generation, the selection of the electrolyte and temperature control is important [50–52]. At this point the electrochemical

reactor design is of paramount importance since it will dictate the proper and optimum use of the electrochemically produced ozone. An example of this, is the Downflow Bubble Column Electrochemical Reactor (DBCER) reported by [53] where ozone was in-situ produced at pilot scale with BDD electrodes. An advantage of this technology is that its design allows 100% consumption of the produced ozone.

Ozone electrogeneration has been studied for many years. However, this technology has been evolving, which allows it to be today one of the most promising technologies in water treatment, due to the advantages mentioned above in Table 1, but also to the efficiencies that electro-ozonation has shown. During the history of ozone research, various electrode materials and cell designs have been studied. As a first alternative, platinum electrodes were evaluated, which achieved high ozone production efficiencies, but at very extreme temperature and pH conditions. Subsequently, PbO<sub>2</sub> electrodes were shown to achieve similar efficiencies to platinum electrodes under milder operating conditions; however, traces of lead were found in the treated water. Finally, BDD electrodes have proven to be an excellent alternative due to their very high oxygen overpotential, their ability to work at mild pH conditions and without the presence of by-products due to their high stability that prevent corrosion [11,51]. Several studies on electrochemical ozone production have shown that current efficiencies of 10%, 19% and 42% can be achieved with OFM-PbO<sub>2</sub>, Pt(10)–Ta(90) and BDD electrodes, respectively [54–56].

Ozone electrogeneration for wastewater treatment is at the stage of scaling up from laboratory to real applications. Table 4 summarizes the current applications of ozone electrogeneration in wastewater treatment. The cell design is critical for a process where one of the constraints is the solubility of ozone and the cost of efficient electrode materials such as BDD. In this sense, a correct cell design would allow to reduce energy consumption and, in this sense, the operational cost. PEM electrolyzers are a very used technology, because they allow to work efficiently with liquids of very low conductivity and low voltages [25,57]. For example, a study by Lara-Ramos J. et al., 2020 shows that ozone generation by PEM electrolyzer compared to the corona discharge method, achieves higher dissolved ozone concentrations 0.55 vs.

**Table 4**  
Summary of applications of ozone electrogeneration [11].

Application	Treatment conditions	Efficiency	Ref.
Landfill leachate	Ozone electrogeneration by Ti/RuO <sub>2</sub> -IrO <sub>2</sub> anode Batch reactor Voltage and current applied: 9 V and 4 A Temperature: <15 °C pH: 7.4	64.8% COD removal in 97 min	[61]
Removal of Total coliforms and Pseudomonas aeruginosa	Ozone electrogeneration by CabECO® cell (4 DIACHEM® electrodes with two Nafion separation membranes) Continuous reactor Current intensity: 4 A Room temperature	3 Log unit removal (total coliforms) and 1 Log unit removal (Pseudomonas aeruginosa) in 60 min	[62]
Fecal-polluted water	Ozone electrogeneration by CabECO® cell (4 DIACHEM® electrodes with two Nafion separation membranes) Continuous reactor Current density: 41.7–1666.6 Am <sup>-2</sup> Room temperature	99% Pseudomonas Aeruginosa and Total Coliforms removal for 0.02 Ah L <sup>-1</sup>	[63]
Leachate concentrate	Ozone electrogeneration by Ti/ATO anode Continuous reactor Current density: 5–40 mA cm <sup>-2</sup> pH: 7.3	60% Removal TOC efficiency in 240 min	[64]



0.39 mg/dm<sup>3</sup>, a degradation efficiency of 40% Clopyralid removal with PEM electrolyzer and only 5% with corona discharger, and 6.5 W compared to 45 W of energy consumption [58].

However, the PEM electrolyzer requires the study of membranes that can withstand the conditions of real water, since Nafion membranes show a rapid degradation and clogging. In this sense, the future of the use of PEM electrolyzers for water treatment is focused on the study of stable membranes [59]. Attention is also being paid to taking advantage of the high H<sub>2</sub> generation that this type of electrochemical cells is capable of producing. In this way, water treatment would not only generate O<sub>3</sub> and improve water quality, but it could also produce green hydrogen [60].

Ozone electrogeneration is one of the most useful AOPs in real industrial or municipal applications. There are examples of mobile water disinfection installations, such as the solar-assisted "SafeWater Africa" project. The aim of the project was to study and develop an autonomous and decentralized 'Made in Africa' water treatment system for rural and periurban areas which is highly efficient in the degradation of harmful pollutants and which is accepted by the rural communities to access to safe water. The purification system includes an electrochemical oxidation technology based on the electrochemical reactor used for ozone production (CabECO® cell), which is based on the electrochemical oxidation of critical low-power substances and applies diamond-coated metal electrodes. The entire water purification system contains the following additional technologies Electrocoagulation and electrodialysis for inorganic substances (Spain), remote monitoring and quality control of the system (South Africa), solar energy supply (South Africa), and the use of a solar power supply (South Africa) [65].

In Mexico, meanwhile, in 2016, the project "Wastewater treatment plant for the Costa Rica syndicate" was launched. The Junta de Agua Potable y Alcantarillado de Culiacán (JAPAC) was a pioneer in the country in using the ozonation technique for cleaning wastewater, which allows reusing the vital liquid in agriculture at a low cost and without environmental impact. The main proposal is to eliminate the chlorination technique and incorporate ozone. The technique consists of concentrating oxygen and applying an electric discharge. The ozone produced is applied to the bottom of the water in the treatment plant to bring the gas bubbles into contact with the liquid to be treated [66].

### 3.3. Electrocoagulation

As mentioned above, electrocoagulation is not an AOP, however it is an electrochemical process that has a high efficiency in removing suspended solids and colloidal material in short times which helps to purify water. It has an advantage over conventional coagulation because there is no need to add coagulants to the water, but they are generated in situ through the use of electrodes, which translates into operational savings that are attractive to the industry and this is the reason for being an electrochemical technology with one of the largest real applications in wastewater treatment.

Electrocoagulation (EC) is a wastewater treatment based on the destabilization reaction of colloidal particles using coagulants generated *in-situ*, through the electrolytic oxidation of an appropriate soluble anode material. The most commonly used anodes to date are Fe and Al, due to their low cost and high valence (+III). For example, in the case of the iron anode, Fe<sup>2+</sup> is generated at the anode by the oxidation of iron, while OH<sup>-</sup> is produced by the reduction of H<sub>2</sub>O. The Fe<sup>2+</sup> ion is an active coagulant precursor that forms insoluble hydroxides (Fe(OH)<sub>2(s)</sub>), and Fe(OH)<sub>3(s)</sub> and then these iron hydroxides act as a coagulant/flocculant for suspended solids and give rise to high-density flocs that subsequently settle down. In turn, the production of H<sub>2</sub> at the cathode leads to flotation of the suspended particles on the surface (electroflotation process) [67]. Thus, the electrocoagulation process essentially consists of three stages: 1) electrochemical reactions at the electrode surface and formation of coagulant species (electrochemistry); 2) destabilization of contaminants (e.g., colloids, emulsions, suspensions); and 3) removal of

flocs by flotation and sedimentation [67].

Studies have demonstrated the feasibility of electrocoagulation versus conventional coagulation. During the treatment of cadmium-containing wastewater, Khaled et al., 2019 studied the operational cost of electrocoagulation (price of electric power, electrode materials and pH correction) obtaining a cost for electrocoagulation of 0.06 USD/m<sup>3</sup>, versus 2.11 USD/m<sup>3</sup> for coagulation [68]. On the other hand, it has been shown that the energy consumption of electrocoagulation can be higher than that of conventional coagulation, for 100 m<sup>3</sup> y, 246.4 USD vs. 118.7 USD, respectively. However, the material cost of the chemical coagulation process (1232 USD) was higher than that of electrocoagulation (492.8 USD) [69].

On the other hand, the electrocoagulation process presents other disadvantages that could increase its operational cost such as: rapid consumption of sacrificial anodes, resulting in periodic replacement, reduced process efficiency due to passivation of electrodes, and the floc generated as a result of the treatment requires additional treatment because it contains a high concentration of metal ions that cannot be discharged directly into the environment [13]. In response to these disadvantages, the combination of EC with other water treatment techniques such as membrane processes (microfiltration, ultrafiltration, reverse osmosis), oxidation processes (electrooxidation and ozonation) have been studied [67]. According to Table 5, the coupling of EC with other technologies not only improves the quality of treated water, but also reduces treatment costs.

### 3.4. Electro-Fenton

The Electro-Fenton (EF) process is an advanced electrochemical oxidation process. This process is based on the well-known Fenton reaction for the *in-situ* generation of •OH hydroxyl radicals. The difference of the conventional Fenton process, the continuous *in situ* production of H<sub>2</sub>O<sub>2</sub> occurs from the reduction of dissolved oxygen in a suitable cathode material and ferrous ions (Fe<sup>2+</sup>) are added to the solution and are continuously regenerated or are released by an iron source (sacrificial anode). The reaction between H<sub>2</sub>O<sub>2</sub> and Fe<sup>2+</sup> generates hydroxyl radicals, which will be the species responsible for the degradation of organic compounds [76,77].

The EF process has shown efficiencies in the degradation of organic compounds such as dyes [64], pharmaceuticals, pesticides, among others [68]. It also achieves high efficiencies in the treatment of real wastewater (Table 6). However, one of its main limitations for its application has been the optimal operating conditions, such as an acid pH and the non-recyclability of the catalyst (Fe<sup>2+</sup>), in addition to the generation of sludge [78]. To address these disadvantages, the use of heterogeneous iron-containing catalytic sources has been studied [79]. One study evaluated the use of metallurgical slag as a photocatalyst for the degradation of diclofenac, which allowed working at pH 7, achieving complete degradation of diclofenac at 90 min and 87% mineralization at 300 min [80].

The insertion of EF into real treatment plants has been studied in China. One study proposed by Zhou X. et al., about coupling EF with peroxy-coagulation as a feasible pre-treatment for high-strength refractory coke plant wastewater, proposed the use of a batch reactor, using a graphite electrode as anode and cathode (Fig. 2). It demonstrates that the EF process is efficient in the pretreatment of wastewater, since it achieves the elimination or transformation of dissolved organic matter, showing efficiency to degrade complex organic compounds of refractory wastewater due to ring opening and chain breaking. The B/C ratio also increased from 0.11 initially to 0.43 after the EF process, greatly improving the biodegradability of the wastewater, and reducing toxicity, making it suitable for further biological treatment [84].

An important improvement has been observed when combining electro-Fenton with other AOPs. Thus, this is a strategy that is advantageous because improves the whole process by generating not only hydroxyl radicals but also other strong oxidants like SO<sub>4</sub><sup>•-</sup> and Cl. On the

Table 5

Real applications of electrocoagulation single or coupled to other treatments.

Wastewater/ Technology	Cell design	Efficiency	Energy consumption or cost	Ref.
Poultry slaughterhouse wastewater	Batch reactor Iron and copper electrodes 30 V Volume: 2 L 60 min	100% removal total coliforms	0.29 kW/h	[70]
Textile wastewater/ Electrocoagulation-Flotation and Pulsed Power Plasma	Batch reactor Aluminum Electrodes Volume: 600 mL	81.25% TOC removal	0.581 kWh/Kg TOC	[71]
Real textile industry wastewater (Mehr nasaj textile factory)/ Electrocoagulation-Nanofiltration	Electrocoagulation: Batch reactor Aluminum electrodes Volume: 2.5 L Nanofiltration: Dead-end stirred cell filtration Polyethersulfone (PES) NF membrane Volume: 400 mL 0.9 Mpa	74% COD removal 95% color removal	—	[72]
Chocolate industry wastewater	Parallel flow column reactor Recirculation system Aluminum electrodes 2 pairs Solar energy Sodium sulfate (1 M) as electrolyte pH: 6.34 60 min	63% COD removal 97% color removal	4.01 USD/m <sup>3</sup>	[73]
Swine slaughterhouse effluents	Prepilot scale Filter-press FM01-LC-type electrochemical reactor Semicontinuous process Plate aluminum electrodes 25 mA/cm <sup>2</sup>	72.7% TOC removal	19.80 kW h/m <sup>3</sup>	[74]
Produced water (Majnoon oil field)	EC-EO Batch reactor Anodes: Al and graphite plates Cathodes: 3 plates of stainless steel Time: 150 min Density current. 26.77 mA/cm <sup>2</sup>	93.43% COD removal 97.82% turbidity removal	128.6 kWh/kg COD.	[75]

other hand, it is also possible to increase the Fe (II) regeneration rate from Fe (III) photoreduction in solar electro-Fenton (SPEF) and this increases the OH generation and reduces the amount of generated sludge and therefore the inherent cost of its management [85].

Another alternative that has emerged to be able to work at not so extreme pH is the so-called Electro-Fenton Like, which is based on using other materials as catalyst. In this context, Co, Cu, Mn, Ce as well as other elements, exhibit various oxidation states and can work as H<sub>2</sub>O<sub>2</sub> dissociation catalysts [86]. A study by Yelong Zou et al., assessed the efficiency of using copper and cobalt as catalyst for sulfamethoxazole (SMX) degradation in a cylindrical batch reactor, using Pt as anode and CuCo-O@CNTs/NF as cathode which allowed working at pH 5.6 and achieved 100% SMX removal and 39% TOC removal, the cathode was able to achieve 94% SMX removal even after ten times of reuse in the Electro-Fenton type system, which proved to be a good alternative for real application [87]. Another study focused on the treatment of textile dyeing wastewater by Heterogeneous Electro-Fenton evaluated the reuse of nickel converter slags, obtaining a favorable response because the NLPC had a high catalytic activity and could degrade efficiently in a shorter time (30 minutes), in addition to excellent stability and recyclability [88].

### 3.5. Electrophotocatalysis

Electrophotocatalysis (EPC) is a process that combines heterogeneous photocatalysis with electrochemistry. In EPC, a semiconductor photocatalyst, SC, is used as an electrode to produce oxidizing species (mainly •OH radicals) through the excitation of the photocatalyst by the action of UV/Vis radiation and under an external potential [89]. One of the most widely used photocatalysts is TiO<sub>2</sub>, since it has demonstrated high efficiency in generating hydroxyl radicals when irradiated with UV

light, it is environmentally friendly and has a low cost. Due to these properties, its efficiency in various wastewater matrices and strategies to increase this efficiency have been studied numerous times, among them are; 1) a high surface area through the fabrication of nanostructures (more photocatalytic active sites) and higher efficiency in charge transfer, 2) doping with other atoms such as Fe, Cu, Cr, B, N (second generation photocatalysts) and 3) composite materials with metals (Pd, Pt, Au) [90].

The main drawback of this technology has been the recovery of the photocatalyst from the treated water before its discharge or reuse, to address this limitation, the deposition of the photocatalyst on support materials has been proposed. However, this is one of the main research gaps as this reduces the available surface area causing loss of photocatalytic activity and limitations in mass transfer, requiring the study of different support materials.

Regarding the design of the reactor or cell, most of the reported studies are based on continuous or discontinuous flow undivided cells, equipped with two or three electrodes, the photoanode, the cathode and the reference electrode, in addition to the irradiation source, either internal or external, such as UV lamps or solar radiation. One of the key components in this type of reactor is the electrode material and it must be characterized by being chemically and physically stable, exhibiting a high photoactivity in both, the UV and visible range and high electrical conductivity [91]. This is one of the advanced oxidation processes that has been developed recently; however, its application to real waters or industrial effluents might be limited by the complexity of real waters which can exert a negative impact on the treatment efficiency since there are chemical and optical interferences with light, due to dissolved organic matter, inorganic ions and pollutants [92].

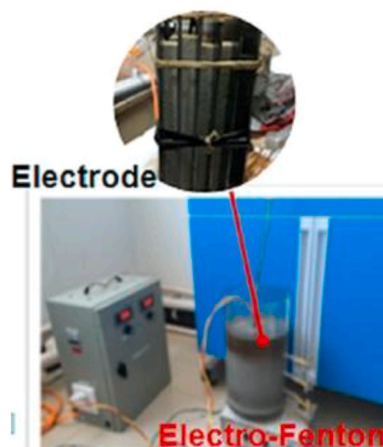
Table 7 shows a summary of the works reported with real water. It can be observed that most of the works couple this technology to another

**Table 6**

Real applications of the Homogeneous and Heterogeneous Electro-Fenton process.

Wastewater	Cell design	Efficiency	Energy consumption or cost	Ref.
Textile industrial wastewater	Cylindrical glass reactor Anode: graphite electrode Cathode: graphite electrode SZVI: 0.6 g/L Current density (mA/cm <sup>2</sup> ): = 20 Time: 30 min pH: 6.5 AC/ CFO	Maximum reductions of approximately 100%, 67%, and 59% in color, COD, and TOC, respectively. The BOD5/COD ratio from 0.15 to 0.54	—	[78]
Textile industry wastewater (Mink Blanket industry, Ludhiana, Punjab)	Continuous reactor Anode: two Ti/Ru <sub>2</sub> electrodes Cathode: two aluminum electrodes Continuous and steady aeration Current (A): 1.10 C <sub>Fe</sub> : 0.55 mM V: 1.5 L Time: 137 min	85% COD removal 94% Color removal	15 kWh/ kg COD \$9.75/ kg COD	[81]
Kerman Hospital Wastewater	Cylindrical glass reactor Anode: iron electrode Cathode: iron electrode Current density (mA/cm <sup>2</sup> ): 8 H <sub>2</sub> O <sub>2</sub> : 122.5 μL/L pH: 2.75 V: 250 mL Time: 137 min Room temperature	Degradation yield of 99.5%	15 kWh/ m <sup>3</sup>	[82]
Effluent treatment plant, India	Batch reactor Anode: 2 Pt/Ti electrodes Cathode: 2 graphite felt V: 1 L Catalyst: modified laterite soil Voltage: 3 V pH: 3 and natural pH (7.8) Time: 60 min Room temperature	55.4% COD removal at pH 3 41.30% COD removal at the natural pH of wastewater (pH 7.8)	0.179 kWh/ k COD	[83]

treatment in order to improve its performance and be a more viable option for its application. It was demonstrated that the combination of EPC with O<sub>3</sub> improves the performance of EPC for the treatment of produced waters which are highly complex waters. Ozone contributes to decrease the color of the effluent, facilitating photons to reach the



**Fig. 2.** Pre-treatment proposal for high-strength refractory coke plant wastewater by Electro-Fenton with peroxi-coagulation. Used with permission from Elsevier [84].

**Table 7**

Actual water treatment studies using electrophotocatalysis.

Wastewater	Cell design	Efficiency	Ref
Oilfields wastewater.	Microreactor Anode: Immobilizing boron carbon nitride (BCN) nanosheets on the copper electrode Cathode: coil type copper electrodes Irradiation: two 8 W UV lamps pH: 3.8	81% COD removal	[94]
Azo dye wastewater	Continuous cross-flow reactor Conductive membrane: Polyvinylidene fluoride (PVDF)-polyaniline (PANI)-TiO <sub>2</sub> Cathode: Pt Light intensity of 100 mW/cm <sup>2</sup>	73% decolorization rates in 60 min	[95]

cylindrical TiO<sub>2</sub> nanotube photoanode to enhance the generation of electron-hole pairs and thus increase the concentration of hydroxyl radicals in solution achieving total removal of color and turbidity and a COD removal of 73% [93]. Therefore, although this technology is still under development to be efficiently applied in water treatment plants, its coupling to advanced processes that have been widely studied and tested could be a good alternative.

Within the advanced oxidation processes, especially electro-photocatalysis, a great advantage is the use of solar radiation, since this eliminates the need for an artificial irradiation source, which results in cost reduction and greater viability. However, as we know, solar radiation is very inconsistent, so the design of facilities and reactors that can reduce the problems related to this is of utmost importance. A reactor design has been reported by McMichael et al. [96] (Fig. 3), which uses solar radiation as an energy source to achieve inactivation of *E. coli* and *P. Aeruginosa* equipped with a TiNT-mesh photoanode. At the same time the results showed that the photocurrent correlates linearly with the change in UV intensity, which could potentially be developed into a quality assurance method when using actual solar irradiation.

In summary, the typical documented disadvantages of the electro-Fenton process can be overcome by producing H<sub>2</sub>O<sub>2</sub> in-situ, adding a photons source and applying material design to prepare a cathode allowing the recovery of the active species through a redox cycle. An example of this, albeit applied to synthetic water only, is the gas diffusion electrode (GDE) prepared by [97], consisting of CuFeO<sub>2</sub>-NO/PBC (PBC=porous biochar) on nickel foam. This GDE was proven to generate and activate H<sub>2</sub>O<sub>2</sub>. The use of this electrode also eliminates the need of



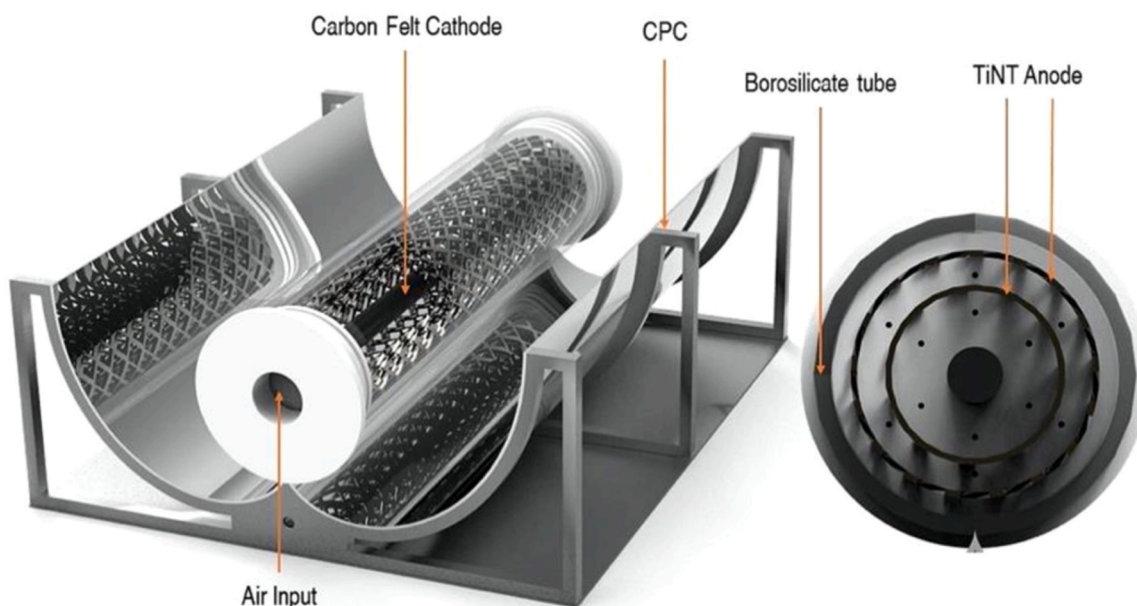


Fig. 3. Solar PEC reactor configuration from [96]. Used with permission from Elsevier.

powder recovery demanded in other photo-electrocatalytic processes. This work also presents a topic that can be considered trendy and is the use of biochar, from alfalfa in this case, to prepare GDEs and other carbon-based materials.

### 3.6. Ultrasound

In recent years, the use of ultraviolet and ultrasound technologies coupled with electrochemical treatments has been studied with the aim of improving the production of oxidants and/or increasing the degradation of organic compounds. Ultrasound (US) has frequencies ranging from 20 kHz to 1 GHz. Generally, ultrasound exceeds vibrational wavelengths at the molecular and atomic scale and therefore does not react with molecules. It creates cavitation phenomena in which bubbles are rapidly formed, bursting and collapsing within a liquid medium, producing high energy conversions into chemical energy. In this sense, it has been shown that ultrasound enhances electrochemical degradation

through physical and chemical mechanisms [98].

Physically, sonication forms microbubbles and the collapse of these bubbles generates extremely high temperature, pressure and these conditions give rise to rupture forces, thus organic compounds tend to be directly pyrolyzed. On the other hand, chemically it acts by homolytic fragmentation of dissolved  $\text{H}_2\text{O}$  and  $\text{O}_2$  by different oxidizing species such as  $\cdot\text{OH}$ ,  $\text{HO}_2^\cdot$  and  $\text{O}^\cdot$  and  $\text{SO}_4^{\cdot-}$  [99]. In addition to this, the ultrasound wave also causes a change in the chemical composition of the electrolyte, which generates new radicals and their derivatives based on the effect of the cavitation phenomenon.

One of the great advantages of coupling ultrasound to electrochemical treatments is the cleaning of the electrodes used, due to the shock waves produced by the cavitation microbubbles. Thus, promoting an increased ion transfer rate within the electrode area and mass transfer rate with the bulk, at the same time the instantaneous high temperature and pressure generated by the ultrasonic cavitation can activate the electrode surface, providing activation energy for the electrode reaction

Table 8

Studies of the coupling of ultrasound to coupled advanced oxidation processes for wastewater treatment.

Technology/ Wastewater	Cell Design	Efficiency	Energy consumption or cost	Ref.
Electrocoagulation-Flotation (ECF) vs. Electrocoagulation-Flotation / Ultrasound (ECF/US) Swine slaughterhouse wastewater	Cylindrical glass reactor Anode: Al Cathode: Al/ Fe 3500 mL 5–20 V Ultrasound frequency: 40 kHz	ECF: 60 min ECF/ US: 35 min 95.5% color, 96.2% turbidity, 93.4% nitrogen, 90.7% BOD, 90% COD.	ECF 5.310 kWh/m <sup>3</sup> 1.031 \$/m <sup>3</sup> ECF/US 5.146 kWh/m <sup>3</sup> 0.745 \$/m <sup>3</sup>	[103]
Electrodisinfection (ED) vs. Sono-electrodisinfection ED/US	Continuous reactor recirculation Anode: BDD Cathode: Stainless- steel 2.0 L 5–20 V Ultrasound frequency: 24 kHz	ED: 200 min disinfection rate of 0.0242 min <sup>-1</sup> ED/ US: 120 min disinfection rate of 0.1428 min <sup>-1</sup>	—	[104]
Ti/RuO <sub>2</sub> vs. Ti/RuO <sub>2</sub> /US /cosmetic industry wastewater	Circular glass reactor 500 mL Ti/RuO <sub>2</sub> anode and cathode Ultrasound frequency: 33 kHz	Ti/RuO <sub>2</sub> : 65% COD removal Ti/RuO <sub>2</sub> /US: 80% COD removal	Ti/RuO <sub>2</sub> /US 217.36 \$/ m <sup>3</sup> 824.07 kWh/ m <sup>3</sup>	[101]

[100]. In this sense, the coupling of electrochemical technologies and ultrasound increases the degradation speed of contaminants, the useful life and in-situ cleaning of the electrode, eliminates electrochemical polarization, as well as reduces treatment times and energy consumption [101,102].

Several studies have demonstrated the increased efficiency of electrochemical processes when assisted by ultrasound. In Table 8, it can be seen that US not only improves the degradation or process efficiency, but it can also reduce treatment times, energy consumption and in some cases total treatment costs. Showing that the cost of treatment can be reduced by about 30%, in addition to improving water quality [103], as well as increasing disinfection rates by about 6 times [104].

#### 4. Feasibility of the insertion of advanced electrochemical oxidation processes in industrial water treatment plants

As demonstrated throughout this review, electrochemical cells are at a stage of technological maturity that allows their insertion in industrial water treatment trains with high POP loads. Although one of its main disadvantages may be the energy consumption or the first investment, these processes seem to be a good alloy to achieve the circular economy of water, where these consumptions could be recovered. The circular water economy seeks to take wastewater and through appropriate treatments achieve its reuse either for irrigation or other purposes (which have been gaining interest in the last decade), as wastewater is rich in valuable substances that could have a secondary use [105]. Such is the case of the electrogeneration of ozone described above. This process has proven to be completely sustainable, since the wastewater is the raw material for the generation of ozone, which will be subsequently used for the treatment of such water, where the final product will be treated water, which can meet the quality of irrigation water. In addition, the ozone generated in gaseous phase can have some other industrial application. In this way, advanced electrochemical oxidation processes have demonstrated not only to achieve a good quality of treated water, but also to take advantage of wastewater to generate value-added products and mainly energy.

It is known that the main products in the electrolysis of water are oxygen and hydrogen. Hydrogen is an excellent source of fuel and energy storage, so its production has gained worldwide interest. Currently, most hydrogen is produced by natural gas reforming and coal gasification, which results in high CO<sub>2</sub> emissions and efficiencies of 70–75% and 45–65%, respectively [106]. Due to the generation of CO<sub>2</sub> and its contribution to greenhouse gases and global warming, it has been necessary to search technologies that can compete in efficiency with the conventional ones and turn out to be a green process. Table 9

Hydrogen is not the only product that can be generated by wastewater, different studies have shown that it is possible to recover heavy metal ions from wastewater. Electrochemical recovery is an environmentally friendly option, with operational versatility and can be automated compared to other methods (adsorption, chemical precipitation, etc.). In this way, Ag<sup>+</sup> ions can be recovered by electrodeposition with efficiencies of 80–100%. [112]. Copper has been recovered from an industrial wastewater with nanocarbon film and the platinum electrode [113], as well as leaching solution with graphite and copper electrodes [114,115]. Other products such as bromine can also be recovered by electro-oxidation, with an efficiency of 77.84% (101.040 kg/h) and an energy consumption of 5.71 kJ/h. [116]. Another study shows the recovery of formic acid from wastewater containing a high concentration of Cl<sup>-</sup> using an ion-capture electrochemical system integrated with a liquid-membrane chamber with efficiencies of 586–1827 mg/L, high purity (80.6%–84.1%), acceptable extraction efficiency (61.5%–68.0%) and low energy consumption (2.43 kWh/kg of FA) in semi-continuous operation (14.5 h) [117]. Lehui Ren, et al., 2022, recovered phosphorus from wastewater using a cathodic membrane filtration reactor [118].

According to Fig. 4, the AOPs (Advanced Oxidation Processes)

**Table 9**

Studies focused on the production of hydrogen from wastewater by electrochemical processes.

Technology/ wastewater	Removal efficiency	Hydrogen efficiency
Pico-hydro power (PHP)-Microbial electrolysis cell (MEC) coupled system/ Palm Oil Mill Effluent (POME) wastewater [107]	57 ± 2.1% CE 73 ± 0.8% COD	75 ± 1.9 r(H <sub>2</sub> ) 1.16 ± 0.08 Q (m <sup>3</sup> H <sub>2</sub> /m <sup>3</sup> d) 894.42 ± 0.3 YH <sub>2</sub> mmol H <sub>2</sub> /g COD
Photoelectrochemical treatment/ wastewater: ammonia, urea and formamide [108]	80% COD	30 mol (ammonia) 140 mol (urea) 240 mol (formamide)
Au/TiO <sub>2</sub> / Industrial and municipal wastewater [109]	-	11 μmol/ L (industrial wastewater) 23 μmol/ L (municipal wastewater) 44 mol/ L
Cu/TiO <sub>2</sub> / municipal wastewater produced before the biological treatment [110]	-	
TiO <sub>2</sub> /SS/ oilfield-produced wastewater [111]	80% degradation	12.36 μmol/hour (synthetic produced wastewater) 9.11 μmol/hour (real produced wastewater)

implemented would help to improve the biodegradability of the wastewater, and then move on to conventional biological treatments that would now be able to degrade the organic matter contained in the water and reach the maximum permissible values to be able to discharge or reuse it. This coupling would allow relatively fast kinetics, no solids generation, and removal of excess NH<sub>3</sub> above the requirements of biological oxidation.

An example of the application of EAOPs prior to biological treatment (Fig. 4A) would be the integration of advanced ozonation as a pretreatment to enhance the efficiency of conventional biological processes by pre-breaking down recalcitrant organic compounds in municipal wastewater. The presence of recalcitrant organic compounds has proven to be an obstacle to the overall efficiency of biological processes, as some of these compounds are difficult to biodegrade by conventional microorganisms. Given this resistance, it is proposed to implement an advanced ozonation process as pretreatment. Ozone (O<sub>3</sub>) is used to generate highly reactive hydroxyl radicals, which attack and decompose recalcitrant compounds. Catalytic ozonation may be one of the most promising technologies with the highest net energy gain potential for this application, because the reaction between ozone and catalyst can generate heat, which can be captured and used to supply heat to biological treatments or produce energy that would be supplied to the treatment [119]. Using a continuous monitoring system, the effectiveness of ozonation is evaluated in real time by measuring the concentration of recalcitrant compounds before and after pretreatment. The ozone-pretreated wastewater enters the conventional biological process. The organic load is now more easily biodegradable by microorganisms, improving the efficiency of the biological treatment. This integration would allow to reach adequate removal levels for recalcitrant compounds, complying with environmental regulations. This reduces the burden on biological processes, prolongs the life of the systems and ensures compliance with environmental standards.

There are also other studies on the application of EO coupled with secondary treatments, using commercial electrochemical cells, such as the CONDIACELL® cell. Xing Du et al. [120] propose a hybrid Nanofiltration-Electrooxidation) NF-EO system for the treatment and reuse of secondary wastewater using a simple "waste-treatment-waste" green strategy. Electrochemical oxidation of NF concentrate with BDD anodes was found to remove organic contaminants while generating controllable chloramines that could mitigate biofouling in the downstream NF process. The NF-EO process was able to completely remove antibiotics and bacteria and maintained good stability for 10

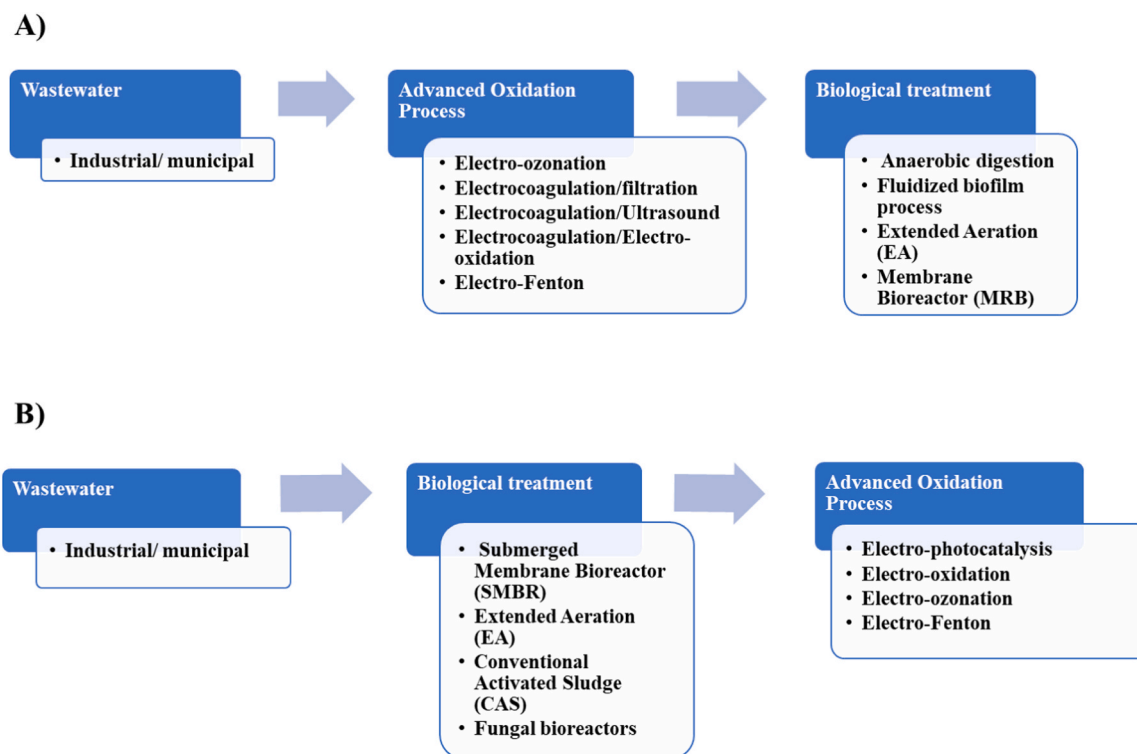


Fig. 4. Proposals of advanced oxidation processes at wastewater treatment plant.

consecutive cycles. It should be noted that chloramines electrogenerated by EO to alleviate NF membrane biofouling could avoid redundant dosing of environmentally damaging chemicals. This study provides a promising "waste-treatment-waste" technology for sustainable wastewater reuse in engineered, agricultural, and natural systems affected by chloramine.

The proposal of a treatment train including an EAOP at the end (Fig. 4B), can be an effective strategy to improve the quality of the final effluent and comply with environmental standards. For example, in a treatment train that includes conventional stages such as screening, sedimentation, and an aerobic biological process for the removal of the industrial organic load. Due to the nature of industrial water, persistent industrial chemicals are encountered after conventional treatment, which require further attention. Given the persistent nature of the identified contaminants, it was decided to integrate a photocatalysis process at the end of the treatment train. Photocatalysis uses catalysts and ultraviolet light to generate highly reactive hydroxyl radicals. A continuous monitoring system evaluates the effectiveness of photocatalysis in real time and the results of the combined treatment, verifying that the treated effluent complies with applicable environmental regulations before discharge to the receiving body. In this way the integration of photocatalysis at the end of the treatment train improves the overall efficiency in the removal of persistent pollutants, ensures that the final effluent meets environmental standards prior to discharge to the environment and contributes to sustainability by specifically addressing persistent pollutants that could have a negative impact on the receiving environment. The performance of a solar photocatalysis reactor coupled to an ultrafiltration system on wastewater from a wastewater treatment plant in Baghdad, Iraq, was studied by Nisreen S. Ali et al. [121]. The process consisted of ultraviolet (UV), UV/H<sub>2</sub>O<sub>2</sub>, and UV/TiO<sub>2</sub> nanocatalysts as pretreatment steps to prevent fouling of the ultrafiltration (UF) membrane, thus decreasing TOC and turbidity by photocatalysis.

## 5. Life cycle analysis of EAOPs

Life Cycle Assessment (LCA) is a methodology applied to evaluate the potential environmental burdens and human health impacts implied by processes, products, activities or services, due to the implicit resource consumption or to the generation of by-products or emissions. This methodology is based on the standards ISO 14040 and 14044 [122,123] and is generally discretized in four general stages: Goal and scope definition, life cycle inventory, life cycle impact assessment and interpretation of the obtained impacts [122]. The indicators of potential environmental impacts can be classified into mid-point and end-point. To calculate these indicators, as well as their contribution, and to conduct sensibility and uncertainty analyses, there is a widely used software by the EAOPs related works presented in Table 10, SimaPro.

Table 10 summarizes the existing literature on LCA of electrochemical advanced oxidation processes. It can be observed that the related works are rather scarce. In this section, however, the goal, inventory, impact categories and main results of such works will be analyzed and discussed to make a recommendation for future similar analyses.

A detailed and precise life cycle inventory (LCI) is fundamental for a proper LCA. To do so, the first step is to select a functional unit and to establish a system boundary, alternative scenarios, qualitative and quantitative description of unit processes, including categories of chemicals to the air, water and soil emissions, consumed resources (water and energy, for instance), land use, material recovery or alternative use. Then a table summarizing all inputs and outputs of any resource must be generated, with the corresponding units and data quality. These data are the required to achieve the defined study scope or goal. The data quality refers to the source of data, i.e. experimental or theoretical, includes specific laboratory scale operations and extraction processes; allocation principles and procedures including documentation and justification and uniform application. All entries on LCI, are associated with impacts from the Ecoinvent database with the help of Simapro software (Pre-Consultants).

According to Table 10, there is not a standard functional unit (FU) for

**Table 10**  
Summary of the literature related to Life Cycle Assessment (LCA) of Electrochemical Advanced Oxidation Processes (EAOP).

Reference	Goal	Software	FU	Method	Inventory analysis	Electrode material	Midpoint indicators	Endpoint indicators	Some Relevant Results
[124]	LCA of AOP to treat the effluent of an olive mill	SimaPro 7.3.3	1 L of olive mill wastewater	IPCC 2007 ReCiPe version	Reactor type Reactor material Reactor inputs Operating parameters Organics Energy requirements	Boron-doped diamond	GWP 0.16 kgCO <sub>2</sub> eq	Human Health, damage Ecosystems diversity Resource availability	The environmental impacts of all three AOPs show that <b>human health</b> is primarily affected followed by impacts onto <b>resources depletion</b> . Compared to photocatalysis and WAO, EO is a more sustainable technology
[128]	To estimate the environmental impact of sludge recovery processes	eBalance	1 g dried sludge material powder	Ecoinvent v3.1 and CLCD database	Added chemicals Required energy Electrode materials Remained compounds	Ref. electrode: Ag/ AgCl Counter electrode: Pt wire	EP 2.1 × 10 <sup>-4</sup> <b>kg Peq</b> HT 1.60 × 10 <sup>-10</sup> <b>kg 1,4DCB-eq</b> GWP 0.48 <b>kgCO<sub>2</sub>eq</b>	NR	The use of generated sludge for electro-Fenton, reduces FRC by 73.7% and GWP by more than 97.1%, HTPc and E by 87–95% and 54.2–90.2%, compared to the usual sludge disposal method.
[137]	Analyze the environmental impacts of coupled ion exchange (IX) and Electrochemical oxidation (EO) process trains to treat polyfluoroalkyl substances (PFASs)-contaminated groundwater.	SimaPro 7.0	1000 m <sup>3</sup> groundwater.	Tool for Reduction and Assessment of Chemicals and Other Environmental Impacts (TRACI)	Sodium chloride deionized water electricity heat	Ti <sub>4</sub> O <sub>7</sub> anode and stainless-steel cathode	ODP 0.002 kg CFC-11 eq GWP 72.2 kgCO <sub>2</sub> eq C-CTUh 1.03 × 10 <sup>-6</sup> kg 1,4DCB-eq N-CTUh 4.92 × 10 <sup>-6</sup> kg 1,4DCB-eq	NR	The major contributors to the GWP were electricity and resin manufacturing.
[125]	LCA and comparison of five scenarios to remove phenolic compounds: (i) adsorption by AC, (ii) <b>electro-Fenton process by sacrificial anode</b> , (iii) solar photo-Fenton, (iv) solar photocatalysis with TiO <sub>2</sub> , and (v) solar photocatalysis with TiO <sub>2</sub> /AC	SimaPro®	1 m <sup>3</sup> of phenol	CML 2000 baseline	Reactor, added chemicals, energy consumption, Remaining compounds Sludge Emitted CO <sub>2</sub>	Stainless steel electrodes	GWP 26.77 kgCO <sub>2</sub> eq ODP 1.61 × 10 <sup>-6</sup> kg CFC-11 eq HT 5.53 kg 1,4DCB-eq MAE 4812.32 kg 1,4DCB-eq E 0.007 kg Peq	NR	Electro-Fenton implies the highest contribution to most of the assessed categories and this is due to the energy consumed by the process. PO and AP are affected using <b>stainless-steel</b> electrodes. AP is also affected using Na <sub>2</sub> SO <sub>4</sub> , which also poses some impact (from 3% to 10%) on human toxicity.
[126]	LCA and comparison of bioelectrochemical and integrated microbial fuel cell systems for sustainable wastewater	SimaPro 9.2	1 L of wastewater	ReCiPe 2016	Construction material, Energy consumption, direct GHG emission, by-products generation, and effluent	System coupled with graphite-based anode and Pt.	GWP 1.66 × 10 <sup>-2</sup> kgCO <sub>2</sub> eq HTPc 2.85 kg 1,4DCB-eq MEP 9.03E-6	Human health, ecosystems, and resources.	It was demonstrated that a double chamber air-cathode cell is the most environmentally friendly option under all

(continued on next page)

Table 10 (continued)

Reference	Goal	Software	FU	Method	Inventory analysis	Electrode material	Midpoint indicators	Endpoint indicators	Some Relevant Results
	treatment and resource recovery						kg Peq FFP $-3 \times 10^{-2}$ kg oil eq MET 1.10 kg 1,4DCB-eq		endpoint damage categories. The hotspot is electricity consumed during operation and this has the largest contribution to GWP (90%).
[127]	Evaluate the environmental impact and environmental hotspots of an integrated membrane system in treating anaerobic palm oil mill effluent (POME) with several scenarios involving adsorption and electro-oxidation as pretreatment processes.	SimaPro v9	1 m <sup>3</sup> of treated wastewater	ReCiPe 2016	Materials and fuels Pretreated anaerobic POME Water storage Steel, low-alloyed Chromium steel pipe Electricity, voltage Pumps Cleaning system (water heating) Prefilter Output to technosphere Products and co-products Treated POME effluent	Fe electrode	GWP 582.9 <b>kgCO<sub>2</sub>eq</b> ODP 9.87 <b>kg CFC-11 eq</b> METP 0.067 kg Peq HTPc 7.8 kg 1,4DCB-eq HTPnc 198.5 kg 1,4DCB-eq FFP 58.09 kg oil eq MET 11.80 kg 1,4DCB-eq	Three areas of protection (i.e., human health, ecosystems, and resources).	It can be concluded from the in-depth 'cradle-to-gate' LCA that both the adsorption and the electro-oxidation integrated <b>membrane treatment systems</b> are mainly impacted by the production of a hollow fiber membrane module contributing at <b>42–99% at the midpoint level</b> , <b><math>8.61 \times 10^{-4}</math> (adsorption integrated membrane) and <math>8.45 \times 10^{-4}</math> (electro-oxidation integrated membrane) DALY</b> are the overall characterization factors of the endpoint categories that indicate the impact it has on <b>human health</b> .
[129]	LCA of the pilot-scale electrochemical oxidation of carbamazepine (CBZ)	SimaPro 9.3.0.2	1 mg of CBZ removed per m <sup>3</sup> of wastewater treated during one day of operation	ReCiPe	Chemical and energy consumption in the different scenarios under study, for the vertical plate stirred tank reactor. Sodium nitrate Sodium sulfate Sodium hydroxide Electricity electrodes Electricity recirculation pump Electricity in-out pumps Electricity dosing pump	BDD anodes and one stainless steel cathode	GWP 7.6 kgCO <sub>2</sub> eq MET 0.26 kg 1,4DCB-eq ODP $1.25 \times 10^{-5}$ kg CFC-11 eq FFP 2.03 kg oil eq E $6.78 \times 10^{-3}$ kg Peq HTPc 0.38 kg 1,4DCB-eq HTPnc 8.43 kg 1,4DCB-eq	Human Health, damage Ecosystems diversity Resource availability	Pumping accounts for 51–74% in the categories of (GWP), (TA), (FE) and (FRS). Sodium nitrate accounts for 59–62% contribution to the was the ME category. Recirculation, because of pumping, increases the environmental burdens.



the LCA of Electrochemical advanced oxidation (EAOP's). This FU can be a volume of treated wastewater [124–127], or a certain removed mass of a specific pollutant. Other FU that could be used are removed toxicity, units of removed TOC or removed COD. Due to the physicochemical inherent complexity of wastewater, however, the authors consider that a certain volume of treated wastewater, should be used as FU to assess EAOP's, this would be helpful for the final aim of conducting LCA that is the capability of making decisions regarding the use of one technology or another in terms of efficiency, environmental and human impacts.

For the inventory analysis, there is also a variety of considered inputs and outputs. Nevertheless, all works listed in Table 10, focus on the reaction system to establish the inventory. In all cases, reactor inputs are considered, i.e. energy, water added chemicals; however, only few consider the construction material [113–115] and the type of electrode [128] which becomes important not only because dictates the oxidation process efficiency, ergo the consumed energy, but also the sludge generated that will affect environmental and human impact categories [30]. In most of the cases, the remained compounds in the effluent are also considered in the output of the cycle inventory analysis.

Regarding the assessed mid-point and end-point indicators, these also vary according to the study. Most of the works, 6 out of 7, summarized in Table 10, determine the GWP of the assessed process. This was found to vary in the range 7.6 kg CO<sub>2</sub>-eq per g of removed carbamazepine [129] to 583.87 kg CO<sub>2</sub>-eq per m<sup>3</sup> of treated palm oil mill effluent [127]. GWP is an important environmental impact indicator and is directly related to the sustainability of the assessed process since corresponds to its carbon footprint. Thus, it is mostly affected by the energy requirements of the assessed technology and therefore by all operating parameters dictating the pollutants removal efficiency like reactor, reactor material, physicochemical characteristics of electrodes, type of effluent and added chemicals. It has been demonstrated, albeit with electrocoagulation [30] and electrochemical technologies for partial oxidation [130], that this indicator can be significantly reduced (ca. 92%) using photovoltaic solar panels. It is worth clarifying, that the electrode material is not only important because of the oxidation efficiency but also because its elaboration or extraction might represent environmental burdens. Take as an example, activated carbon electrodes that are made from fossil-based coals that imply the green-house gases emission during their production [131]. Recently, [129], demonstrated the importance of type of reactor by conducting the LCA of carbamazepine removal by EAOP. For this purpose, the environmental burdens of a DiaCell 1001 electrochemical cell and a vertical plate stirred tank reactor were compared, and it was concluded that from an environmental point of view is better to use the former than the latter. Another important conclusion is that an EAOP is environmentally less harmful than other AOPs like ozonation and photo-Fenton and that pumping was the operation contributing the most to GWP because of the fossil energy consumption. In the same context, Chatzisyseon et al., 2013, demonstrated that the environmental sustainability of AOPs increases in the following order EO>WAO>UV/TiO<sub>2</sub>.

Other widely reported mid-point environmental impact is the potential marine ecotoxicity (ME), which is related to the addition of non-ferrous metals (cobalt, copper, manganese, molybdenum, nickel and zinc) to oceans and to the generated toxic effects [132]. The units of this indicator are kilograms of 1,4-Dichlorobenzene (kg 1,4DB-eq). The use of fossil energy contributes importantly to this indicator and also does the electrode material. In this sense, it is worth pointing out that some metals exhibit more aquatic burdens than benzene [133]. On this regard, for instance, Ni and Cu, usually employed in the composition of some electrodes, pose high marine ecotoxicity values, 3.47 E10 and 5.15 E09, respectively [133]. Despite this, ME is not an indicator widely reported in the LCA of EAOP. Razman et al., 2022, report a value of 11.8 kg 1,4-DCB while the value reported by Magdy et al., 2021, is two orders of magnitude higher (4812.32 kg 1,4-DCB eq). It is worth noticing, that the former study was a coupled treatment EO-Membrane

of a palm oil mill effluent while the latter was on the treatment of phenolic solutions by electro-Fenton, thus the relatively high ME reported value was ascribed to the remained organic compounds in solution and to the energy consumption.

Ozone depletion potential (ODP) is related to the damage that a chemical can cause to the ozone layer. Such chemicals are characterized for being persistent and also for having in their structure some specific elements able to interact with ozone, like chlorine and bromine. Thus, the UVB radiation reaching earth increases due to the atmospheric ozone concentration decreasing because of the interaction between ozone and halogens. As widely reported, skin cancer and cataracts are a negative consequence of the increase in the UVB radiation [132]. The potential environmental and human impacts of chemicals to ozone layer are converted to their equivalent of kilograms of trichlorofluoromethane, freon-11 or chlorofluorocarbon equivalents (CFC-11 eq) [134]. Although a zero ODP is desirable, an acceptable value in US has been established to be 0.2 [135] (Finlayson-Pitts & Pitts, 2000). If a software is not used, the equations reported in [135] can be used to calculate this value. For the coupled EO-M treatment, this indicator has been reported to be  $9.89 \times 10^{-5}$  Kg CFC11-eq [116] while it was reported  $1.61 \times 10^{-6}$  Kg CFC11-eq as for the electro-Fenton process [125].

Because a significantly consumed resource in EAOPs is electricity, it is important to establish the ratio between the energy content of fossil resource and the energy content of crude oil. This ratio is an environmental impact indicator and is known fossil depletion or fossil resource scarcity (FFP), and is provided for crude oil, natural gas, hard coal, brown coal and peat [132]. The units of this indicator are kg oil eq. This indicator is reported only by few works presented in Table 10. It is worth highlighting the negative value reported by Chin et al., 2022, which means that is an avoided impact. This is plausible because the assessed system is a microbial cell with Pt. The highest reported value is 58.09 kg oil eq by Razman et al., 2022 and this might be due to the energy consumed by the 67 pairs of electrodes assessed in such a study.

Regarding eutrophication (E), this indicates the potential burdens of macronutrients (nitrogen, phosphorous and carbon) in both aquatic and terrestrial ecosystems [132]. The eutrophication potential is expressed in kilograms of phosphate (kg PO<sub>4</sub><sup>3-</sup> eq.) and based on the authors experience, should be included in all LCA of wastewater treatment technologies when there are nitrogen and phosphorous compounds in the treated effluent composition. It varies in a range of  $9.03 \times 10^{-6}$  – 6.78 kg PO<sub>4</sub><sup>3-</sup> eq and it depends on the FU, the type of matrix and type of reactor.

Another reported indicator is the human toxicity potential with carcinogenic or non-carcinogenic effects, HTPc and HTPnc, respectively. It is also expressed in kg 1,4-dichlorobenzene- equivalents (1,4DCB-eq) and as can be seen in Table 10, it varies between  $1.6 \times 10^{-10}$  [128] and 8.43 [129]. The International Agency for Research on Cancer (IARC), part of the World Health Organization (WHO), evaluated the carcinogenic risk of 844 substances (mixtures) to humans by assigning a carcinogenicity class to each substance [136].

## 6. Conclusions

The insertion of EAOPs in water treatment plants is a strategy for wastewater treatment that is still in the study phase for scaling up. Most of the articles published and presented in this review report are on laboratory conditions, often very controlled, so that efforts should focus on starting to implement these processes in pilot plants or on a larger scale to evaluate the applicability of these systems in real case scenarios. However, as discussed, these technologies show favorable responses for the removal of contaminants in real matrices, where the selection of electrodes, reactor design and operating parameters are key points to achieve not only high removal efficiencies, but also low energy consumption that make these treatments completely viable and sustainable options that can compete with conventional processes currently applied in wastewater treatment plants.

One of the main advantages of these electrochemical processes is that they do not only lead to the pollutants removal, but also allow obtaining value-added products from them or energy, thus being a dual option for water remediation and energy solution. Therefore, it is necessary to exploit these technologies from this perspective, increasing research in the application of EAOPs as treatment, but also as recovery or production technologies. This will make their implementation more attractive at the industrial level, as they would be highly sustainable processes, capable of producing the energy necessary for their operation or recovering products to give them a second life, which would allow recovering investment and operation costs.

Finally, and regarding the life cycle assessment (LCA) of EAOP applied to real wastewater treatment, the literature regarding this topic is currently scarce and it represents an important area of research to achieve Sustainable Development Goal No. 6 (SDG 6). The functional unit (FU) and the assessed environmental impact categories must be carefully elected to obtain meaningful results.

### Declaration of Competing Interest

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

### Acknowledgments

Authors are grateful to CONACYT (Project 320965) for financial support. Technical support of Citlali Martínez Soto is also acknowledged.

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