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Removal of pharmaceuticals and personal care products from wastewater via anodic oxidation and electro-Fenton processes: current status and needs regarding their application

Iván Lozano[a], Pabel Cervantes-Aviles[a,b,*], Arturo Keller[b,c] and Claudia López Aguilar[d]

[a] Escuela de Ingeniería y Ciencias, Tecnológico de Monterrey, Campus Puebla, Vía Atlixcáyotl 5718, Reserva Territorial Atlixcáyotl, Puebla, Puebla 72453, México
[b] University of California, Center for Environmental Implications of Nanotechnology, Santa Barbara, CA, USA 93106
[c] Bren School of Environmental Science and Management, University of California at Santa Barbara, CA, USA 93106
[d] Facultad de Ciencias Químicas, Benemérita Universidad Autónoma de Puebla, Av. San Claudio, Ciudad Universitaria, Puebla 72570, México
*Corresponding author. E-mail: pabel.cervantes@tec.mx; pcervantes@bren.ucsb.edu

PC-A, 0000-0001-9665-2959

ABSTRACT

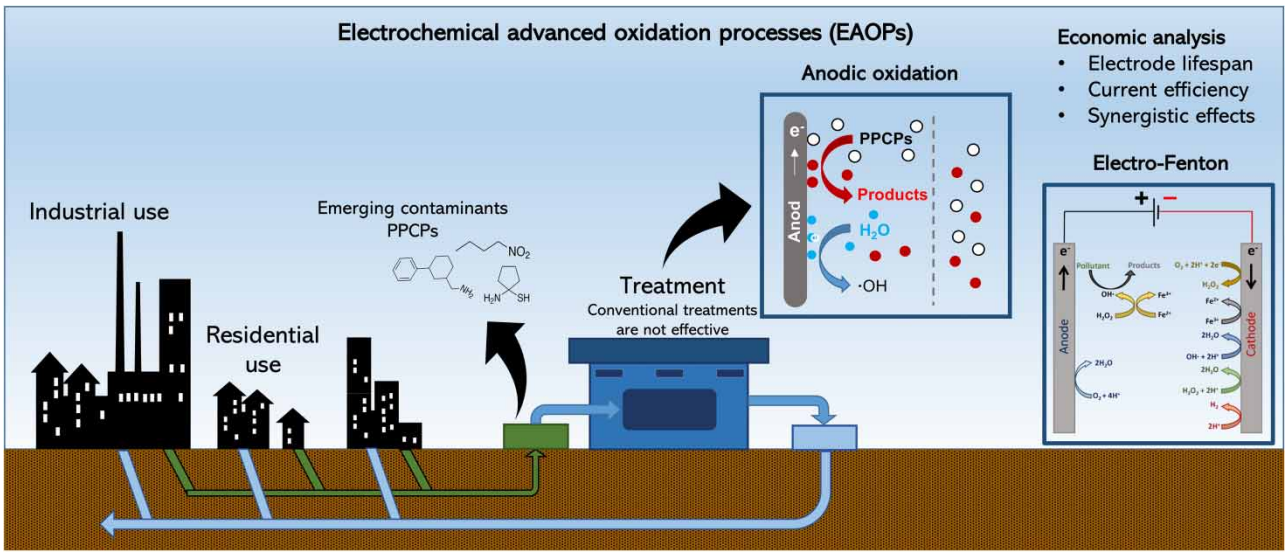
This review provides a current opinion on the most recent works that have been published toward the application of electrochemical advance oxidation processes (EAOPs) for the degradation of pharmaceutical and personal care products (PPCPs) in water streams. Advances in the application of anodic oxidation (AO)- and electro-Fenton (EF)-based processes are reported, including operational conditions, electrode performance, and removal. Although AO- and EF-based processes can easily reach 100% removal of PPCPs, mineralization is desirable to avoid the generation of potential toxic byproducts. The following section exploring some techno-economic aspects of the application of EAOPs is based on electrode selection, operational costs as well as their use as cotreatments, and their synergistic effects. Finally, this short review ends with perspectives about the emerging topics that are faced by these technologies applied for the degradation of PPCPs in research and practice.

Key words: anodic oxidation, electro-Fenton, PPCPs, removal, techno-economic aspects

HIGHLIGHTS

- EAOPs are an attractive alternative for PPCPs such as antibiotics and personal care products.
- Anodic oxidation and electro-Fenton can fully mineralize PPCPs in water at short terms with some trade-offs.
- Techno-economical aspects such as the lifetime of electrodes and operating costs are the main constrain for EAOPs application.
- Cotreatments and synergistic processes could be considered for the successful application of EAOPs.

GRAPHICAL ABSTRACT



1. INTRODUCTION

Pharmaceuticals and personal care products (PPCPs) are constantly diversifying and increasing in production (Khan *et al.* 2022). Due to their potential human and ecological effects, they are considered emerging contaminants (ECs). Pharmaceuticals mainly consist of antibiotics, analgesics, hormones, anti-inflammatory drugs, beta-blockers, among others, while personal care products include chemicals used for cosmetics, surfactants, food additives, hygiene products, and fragrances (Pai & Wang 2022). Moreover, metabolites and transformation products of PPCPs generated in the environment or during their removal are also considered ECs (Zhang *et al.* 2022). Many of these contaminants are also considered recalcitrant to conventional water treatment (Patel *et al.* 2019).

The presence of PPCPs in aquatic and land ecosystems has been found to be linked to their existence in the influents and effluents of wastewater treatment plants (WWTPs). In wastewater streams, the concentration of PPCPs reaches up to 10,000 ng/L (for analgesics in Africa) and its concentration decreases between 10 and 90% (Adeleye *et al.* 2022). Several factors influence the removal of PPCPs during wastewater treatment, such as the pollutant's chemical structure, concentration, and solubility, as well as the technology utilized for treatment. Activated sludge and anaerobic digestion are the prevailing technologies employed in wastewater treatment. Nevertheless, these methods typically exhibit limited efficiency in degrading PPCPs (Hidrovo *et al.* 2022).

In some cases, conventional treatments such as activated sludge have shown partial success in removing pharmaceuticals like diclofenac, ibuprofen, and triclosan, particularly in acclimated cultures (Kim *et al.* 2017). However, this process is time-consuming, resulting in relatively low degradation rates (Patel *et al.* 2019). Consequently, using anaerobic processes for the biodegradation of other PPCPs may require larger treatment units, leading to increased costs. Overall, both aerobic and anaerobic conditions present several limitations for the complete removal of PPCPs through biodegradation. On the other hand, emerging treatments such as electrochemical advanced oxidation processes (EAOPs) have shown great potential for PPCP degradation under certain operational conditions (Moreira *et al.* 2017; Patel *et al.* 2019; Pai & Wang 2022).

EAOPs are a promising up-and-coming type of water treatment that has been extensively studied in recent decades (Klavarioti *et al.* 2009; Barrera-Díaz *et al.* 2014; Sirés *et al.* 2014; Moreira *et al.* 2017; Lozano *et al.* 2022). Many efforts have been put toward studying the degradation capabilities of EAOPs in synthetic and, to a lesser extent, real water matrices. There are some features that make EAOPs an attractive alternative for organic degradation, including PPCPs: the experimental setup is relatively simple, especially for non-radiation systems such as anodic oxidation (AO) and electro-Fenton (EF) (Feng *et al.* 2013). In these processes, operational costs are mostly comprised of electric energy consumption (Muddemann *et al.* 2021). Moreover, high degradation and mineralization efficiencies can be achieved by a combination of direct oxidation (AO) and oxidants (mainly $^{\bullet}OH$)-mediated oxidation (Hu *et al.* 2021), abating of persistent organic pollutants, and a lack of

additional reactants. Despite the many aspects that make EAOPs a very promising technology (Moradi *et al.* 2020), there is a clear lack of real-life application, and commercially available water treatment units in the field are scarcely reported. In this review, we highlight some of the most recent studies on the removal of PPCPs via AO and EF-based processes, their operating conditions, trade-offs, as well as some of the techno-economic aspects facing their realistic applications.

2. ELECTROCHEMICAL ADVANCED OXIDATION PROCESSES

The mechanism of action in EAOPs involves the simultaneous application of electrochemical oxidation and advanced oxidation techniques, leading to the generation of powerful oxidants that facilitate the degradation of organic pollutants in water and wastewater. During EAOPs, electrochemical reactions occur at the surface of specially designed electrodes, resulting in the formation of highly reactive species, such as hydroxyl radicals ($^{\bullet}OH$). These radicals possess strong oxidizing potential and exhibit a non-selective attack on organic compounds, breaking down complex molecules into simpler, less harmful byproducts. Additionally, EAOPs can promote the regeneration of oxidants through the *in situ* electrogeneration of H_2O_2 and other active oxygen species. The synergistic effects of the generated oxidants, coupled with the enhanced mass transfer at the electrode–electrolyte interface, ensure efficient pollutant degradation even at low concentrations. Moreover, the use of various electrode materials and configurations in EAOPs enables fine-tuning of the process to suit specific applications, making it a versatile and promising technology for sustainable water treatment. In the following subsections, we address the main characteristics of some of the most representative types of EAOPs, namely AO, EF, and photoelectrocatalysis.

2.1. Anodic oxidation

AO is one of the most direct methods to remove several types of PPCPs, mainly antibiotics such as amoxicillin (Sopaj *et al.* 2015), cefalor (Kurt 2020), cefazolin (Sordello *et al.* 2021), ciprofloxacin (Lima *et al.* 2020), sulfonamides (Fabiańska *et al.* 2014), sulfamethoxazole (Loos *et al.* 2018), tetracycline (Oturan *et al.* 2013), and trimethoprim (González *et al.* 2011), among others. AO can be described as an electrochemical oxidation process that occurs on the anode's surface, or in a mediated oxidative process, or both (Klavarioti *et al.* 2009). The oxidative process of PPCPs in water can be heterogeneous and homogeneous. The heterogeneous process occurs when the PPCPs are directly oxidized in the electrode surface involving the following steps: transport from the solution to the surface of the electrode, adsorption of the molecule, and electron transfer to the pollutant combined with direct electrochemical reaction, desorption of the products, and transport to the water matrix. Homogeneous oxidation of the PPCPs in water is mediated by the oxidants produced on the anode surface from components present in the matrix, or other ions added to the media to promote oxidation (Panizza & Cerisola 2009; Feng *et al.* 2013). However, byproducts formation during degradation of PPCPs when they are not fully mineralized may have similar or more adverse effects to the aquatic organisms than the original compound.

Hydroxyl radicals ($^{\bullet}OH$), generated either from water or hydroxide ions oxidation, are the strongest oxidants and are readily produced in AO (Sirés *et al.* 2007). However, weaker oxidants such as H_2O_2 from $^{\bullet}OH$ dimerization and O_3 are also produced from water (Kapałka *et al.* 2009), complementing both anodic processes in a highly efficient way: the direct oxidation of pollutants on the anode surface, and the formation of $^{\bullet}OH$ and subsequent pollutant oxidation. Since hydroxyl radicals have a short lifespan (Yang *et al.* 2017), the oxidation of PPCPs mainly occurs in the anode surface and its vicinity, representative of the heterogeneous nature of the AO treatment. Therefore, anode selection is crucial to successfully perform the degradation processes until the mineralization of PPCPs in wastewater treatment.

Boron-doped diamond (BDD) electrodes can be found in the literature as the most successfully applied anode material for PPCPs degradation (García-Montoya *et al.* 2015; Moreira *et al.* 2017; Loos *et al.* 2018; Lozano *et al.* 2022). This trend can also be seen in the compilation of the last 5 years studies presented in Table 1; BDD electrodes are chosen mainly because they possess the largest known oxygen overpotential range about 2.2–2.6 V vs. standard hydrogen electrode (SHE) which allows them to be operated in the potential range of water decomposition to form hydroxyl radicals, some of the strongest oxidizing agents while minimizing oxygen evolution (Enache *et al.* 2009; Moreira *et al.* 2017). High contaminant (PPCPs) concentrations, ranging from 0.5 to 100 mg L^{-1} with 50 mg L^{-1} can be treated, which is greater than the concentration detected in most water bodies and wastewaters, which ranges from ng L^{-1} to μg L^{-1} (Ebele *et al.* 2017; Hena *et al.* 2021).

The comparisons of many electrodes in the market tend to favor BDD electrodes as the best choice for AO, which is four times better than dimensional stable anodes (DSA), such as steel/IrO_2–Sb_2O_3 and Ti/IrO_2–Sb_2O_3, for degradation of amoxicillin (Sopaj *et al.* 2015), and is more efficient in removing tetracycline over time than Pt and Ti/RuO_2 electrodes (Oturan

Table 1 | Removal efficiencies of some PPCPs and experimental conditions of anodic oxidation process

Compounds	Electrode	Concentration	Medium	Experimental conditions	Efficiency (%)	Oxidation products	References
Cefaclor	Sn/Sb/Ni-Ti	50 mg L^{-1}	750 mg L^{-1} KCl	50 mA cm^{-2} for 30 min, pH 7	100 (TOC)	None reported, mineralization	Kurt (2020)
Cefazolin	Pt sheet	100 μM	Synthetic urine	150 mA cm^{-2} for 20 min pH 6	99.5% (HPLC-UV)	Transformation intermediates that arise from the oxidation of the different CFZ sulfur atoms.	Sordello *et al.* (2021)
Ciprofloxacin	Gas diffusion (GDE)	30 mg L^{-1}	0.1 mol L^{-1} K_2SO_4	50 mA cm^{-2} for 90 min at 20 °C and stirring pH 2.5	99.3 (HPLC) 25.3 (TOC)	Aliphatic acids	Lima *et al.* (2020)
Sulfamethoxazole	BDD	0.5 mg L^{-1}	Simulated wastewater	0.9 A for 180 min, $Q =$ 250 L h^{-1}, T = RT	D: 72.9 (HPLC)	Several transformation products	Loos *et al.* (2018)
Diclofenac	BDD	0.5 mg L^{-1}	Simulated wastewater	0.9 A for 180 min, $Q =$ 250 L h^{-1}, $T =$ RT	D: 73.7 (HPLC)	Up to 6 transformation products	Loos *et al.* (2018)
Iopromide	BDD	0.5 mg L^{-1}	Simulated wastewater	0.9 A for 180 min, $Q =$ 250 L h^{-1}, $T =$ RT	D: 32.3 (HPLC)	Several transformation products	Loos *et al.* (2018)
17-alpha-ethinylestradiol	BDD	0.5 mg L^{-1}	Simulated wastewater	0.9 A for 180 min, $Q =$ 250 L h^{-1}, $T =$ RT	D: 69.7 (HPLC)	Several transformation products	Loos *et al.* (2018)
Caffeine	BDD, PbO_2, Pt	50 mg L^{-1}	0.1 M Na_2SO_4	200 mA/cm^2 for 150 min, $T = 25$ °C,	BDD: 100 (TOC) PbO_2: 40 (TOC) Pt: 15 (TOC)	None reported	Chen *et al.* (2015)

et al. 2013). BDD has also been reported as the best option for complete mineralization of caffeine when compared to PbO_2 and Pt electrodes with 40 and 15% of mineralization, respectively (Chen *et al.* 2015). Numerous studies indicate that BDD electrodes are the best all-around choice for organic pollutant degradation (Kapałka *et al.* 2009; Garcia-Segura *et al.* 2012; Ambuludi *et al.* 2013; Chen *et al.* 2014; Sopaj *et al.* 2015; Kouskouki *et al.* 2019); however, full mineralization is highly recommended to avoid undesirable byproducts formation. The most commonly reported oxidation byproducts are carboxylic acids (Ganiyu *et al.* 2018; Cai *et al.* 2020), although oxidation byproducts are not always reported due to the complexity of detection and quantification, and they can vary substantially based on experimental conditions such as current density (50–200 mA cm^{-2}), water media, pH (2.5–7), time (20–180 min), among other factors. Most studies have used Na_2SO_4 (5–100 mM) as electrolytic media based on its inert and neutral nature to avoid pH changes and secondary chemical reactions (Table 1). In addition, sulfate and some anions (such as phosphate and carbonate), promote the formation of stable oxidants such as $(SO_4^-)^\bullet$ and $(PO_4^{2-})^\bullet$ for BDD and some PbO_2-coated electrodes (Sirés *et al.* 2014). Further research using real water and wastewater matrices is needed to scale up AO processes and to better understand the mass transport of the pollutants from the water matrix to the anode surface and its vicinity under more realistic conditions.

The type of pollutant also influences the operating conditions for PPCPs degradation via AO. For example, the removal of diclofenac was higher than iopromide under the same conditions (Loos *et al.* 2018), suggesting that increasing the current density or time of exposition removal could enhance. This is because iopromide has more peptide (amide) bonds than diclofenac, requiring more energy to be degraded. Another polar covalent bond that can be challenging to break down through AO is the C–F which corresponds to per- and polyfluorinated substances, which are present in cosmetics and may potentially reach wastewater. The mineralization of these and other highly stable chemical substances is needed to promote the application of the AO process.

2.2. Electro-Fenton

Electrochemical oxidation processes based on the Fenton reaction, also known as EF processes, are some of the most sought-after EAOPs to this day, given their high mineralization potential of organic compounds (Panizza *et al.* 2014). The basis of EF is the Fenton's reaction in which Fe^{2+} is oxidized to Fe^{3+} by electrogenerated H_2O_2, forming hydroxyl radicals ($^\bullet OH$) and a hydroxide ion (OH^-).

EF processes can be assisted with solid catalyst which is known as a heterogeneous EF (H-EF) process, and by UV radiation such as photoelectron-Fenton (PEF) and solar photoelectron-Fenton (SPEF). In the case of heterogeneous processes, the aim is to promote $^\bullet OH$ formation from H_2O_2 and Fe^{2+}, while in the UV-assisted processes, Fe^{3+} complexes formed are photoexcited to produce both Fe^{2+} and $^\bullet OH$ increasing efficacy overall and avoiding unnecessary reactions (Lozano *et al.* 2022).

To achieve the best performance for EF processes, a careful selection of both electrodes (anode and cathode) (da Silva *et al.* 2021; Olvera-Vargas *et al.* 2021a) and the experimental conditions between them must be conducted (He & Zhou 2017). The most common cathodes are carbon-based, while the materials for anodes that can degrade PPCPs have been BDD, Pt-, Ti-, RuO_2- and IrO_2-based, either as mesh or sheet (Table 2). The production of $^\bullet OH$ is the main difference for all anodes. In general, BDD, PbO_2 and IrO_2-based anodes result in a higher removal of PPCPs (Droguett *et al.* 2020). Other factors influencing the removal of PPCPs are the experimental medium, usually Na_2SO_4, ranging from 5 to 100 mM, pH between 3 and 4, current density ranges between 3.8 and 150 mA·cm^{-2}, as well as a certain amount of catalyst in the case of heterogeneous process and irradiance (1.1–55 W·m^{-2}) for PEF and SPEF processes. Full removal of PPCPs in a short time (from 20 to 360 min) can be achieved at concentration ranging from 0.1 to 0.6 mM with some trade-offs such as pH, catalyst, and current density.

More research is needed that focuses on removal mechanisms, optimization of catalysts and influence of interferences in real wastewater such as radical scavengers and ionic strength, among other factors. The main expected outcomes are the optimization of the operating conditions such as pH, current density, Fe^{2+} and H_2O_2 concentration, O_2 flow rate, and electrode gap distance. However, little is known about the application of new working electrodes made of unconventional carbon sources such as, organic fraction of urban waste or agricultural waste, which will decrease the net environmental impact of this EF process. Moreover, moving from assisted to natural radiation supply in the case of PEF and SPEF, respectively, is a highly relevant factor that influences the sustainability of EF-based technologies applied for PPCP removal.

3. TECHNO-ECONOMIC ASPECTS TO CONSIDER

There are many aspects to consider when deciding the type of technology to use for water treatment, such as the type of contaminants to treat and regulatory requirements. However, for any new technology to either compete or find a niche

Table 2 | Operating conditions for PPCPs removal via electro-Fenton (EF) based technologies: heterogeneous electro-Fenton (H-EF), photo-electro-Fenton (PEF), heterogeneous photoelectro-Fenton (H-PEF), and solar photoelectro-Fenton (SPEF)

Process	Compound	Electrode	Concentration	Medium	Experimental conditions	Reference
EF	Mixture of: methyl paraben (MeP) ethyl paraben (EtP) propyl paraben (PrP)	BDD (Anode)	MeP = 0.3 mM EtP = 0.3 mM PrP = 0.3 mM	5 mM Na_2SO_4	$j = 10$ mA cm^{-2} removal time = 360 min pH = 3	Steter *et al.* (2018)
H-EF	Cephalexin	IrO_2-based (anode), DSA®-O_2 plate	50 mg L^{-1}	50 mM Na_2SO_4	$j = 50$ mA cm^{-2} removal time = 30 min pH = 3 $T = 25$ °C Catalyst: chalcopyrite	Droguett *et al.* (2020)
EF	Thiamphenicol	IrO_2-based (anode), DSA®-O_2 plate	50 mg L^{-1}	20 mM Na_2SO_4	$j = 30$ mA cm^{-2} removal time = 90 min pH = 3 $T = 35$ °C	Thiam *et al.* (2020)
H-EF	Thiamphenicol	IrO_2-based (anode), DSA®-O_2 plate	50 mg L^{-1}	20 mM Na_2SO_4	$j = 30$ mA cm^{-2} removal time = 45–90 min pH initial = 4 $T = 35$ °C Catalyst: pyrite	Thiam *et al.* (2020)
EF	Cephalexin	RuO_2/Ti mesh electrode	200 mg L^{-1}	50 mM Na_2SO_4	$j = 18$ mA cm^{-2} removal time = 60 min pH = 3 T = 25 °C	Zhang *et al.* (2019b)
EF	Ciprofloxacin	RuO_2/Ti mesh electrode	200 mg L^{-1}	50 mM Na_2SO_4	$j = 18$ mA cm^{-2} removal time = 60 min pH = 3 $T = 25$ °C	Wang *et al.* (2018)
H-PEF	Cephalexin	IrO_2-based (anode), DSA®-O_2 plate	50 mg L^{-1}	50 mM Na_2SO_4	$j = 50$ mA cm^{-2} removal time = 15 min pH = 3 Irradiance = 5 W m^{-2} $T = 25$ °C Catalyst: chalcopyrite	Droguett *et al.* (2020)
PEF	Thiamphenicol	IrO_2-based (anode), DSA®-O_2 plate	50 mg L^{-1}	20 mM Na_2SO_4	$j = 30$ mA cm^{-2} removal time = 75 min pH = 3 Irradiance = 5 W m^{-2} $T = 35$ °C	Thiam *et al.* (2020)
H-PEF	Thiamphenicol	IrO_2-based (anode), DSA®-O_2 plate BDD (anode)	50 mg L^{-1}	20 mM Na_2SO_4	$j = 30$ mA cm^{-2} removal time = 60 min pH initial = 4 Irradiance = 5 W m^{-2} $T = 35$ °C Catalyst: pyrite	Thiam *et al.* (2020)
PEF	Cephalexin	RuO_2/Ti mesh electrode	200 mg L^{-1}	50 mM Na_2SO_4	$j = 18$ mA cm^{-2} removal time = 60 min pH = 3 Irradiance = 1.5 mW cm^{-2} $T = 25$ °C	Zhang *et al.* (2019b)
PEF	Ciprofloxacin	RuO_2/Ti mesh electrode	200 mg L^{-1}	50 mM Na_2SO_4	$j = 18$ mA cm^{-2} removal time = 40 min pH = 3 Irradiance = 0.11 mW cm^{-2}	Wang *et al.* (2018)

(*Continued.*)

Table 2 | Continued

Process	Compound	Electrode	Concentration	Medium	Experimental conditions	Reference
					(UVA) $T = 25$ °C	
SPEF	Mixture of: methyl paraben (MeP) ethyl paraben (EtP) propyl paraben (PrP)	BDD (Anode)	MeP = 0.3 mM EtP = 0.3 mM PrP = 0.3 mM	5 mM Na_2SO_4 Na^+, NH_4^+, SO_4^{2-}, Cl^-, NO_2^- and NO_3^- Urban wastewater	$j = 10$ mA cm^{-2} removal time = 180 min mineralization time = 240 min pH = 3	Steter *et al.* (2018)
SPEF	Levofloxacin	Platinized Ti plate	30–60 mg L^{-1} (as DOC)	50 mM Na_2SO_4	Ecath = −0.30 V l SHE removal time = 180–360 min Irradiance = 55 W m^{-2} pH = 3	Coria *et al.* (2018)

Note: All studied presented 100% of removal efficiency.

application, one of the most important aspects to consider is economic viability. When considering the economic viability of EAOPs, one can study aspects such as: cost of electrodes, energy consumption, cotreatments, and synergistic effects.

3.1. Electrodes

BDD is one of the most sought-after electrode materials for many types of EAOP. Recently, Lozano *et al.* (2022) reviewed the application of EAOPs for the removal of PPCPs and found that BDD was the most commonly used material in the AO studies reviewed, given its exceptional capacity for $^{\bullet}OH$ production and chemical stability (Oturan 2021). More recently, BDD has also been increasingly studied in EF–AO combination processes for PPCP degradation due to the high synergy found between techniques (Cai *et al.* 2020; Zhu *et al.* 2020; Olvera-Vargas *et al.* 2021b; Oturan 2021; Mbaye *et al.* 2022). Oturan (2021) recently reviewed the impact that BDD anodes have on the performance of EF processes and found a clear and strong enhancement in both degradation and removal efficiency.

Although BDD shows a very marked advantage in degradation and removal efficiency over other types of anode materials (He *et al.* 2019), using BDD can significantly increase treatment cost. Recently, Stirling *et al.* (2020) used a techno-economic analysis to identify what they describe as key innovations to increase electrochemical oxidation competitiveness in point-of-use treatment systems. Their results identified electrode material as one of the main elements that are driving capital costs and concluded that a tenfold decrease in BDD cost and exploring alternative emergent materials such as Ti_4O_7 can significantly improve affordability of electrochemical devices in the market. Salazar-Banda *et al.* (2021) reviewed the most recent advances in the development of novel materials for both anodes and cathodes and found significant advancements in novel electrode preparation involve new heating methods, different solvents, and modifications in their compositions. Even when these modifications to electrode preparation clearly show an improvement in efficiency, stability and electrocatalytically activity, Salazar *et al.* concluded that additional efforts in large-scale trials are necessary to accurately test the applicability of these materials (Salazar-Banda *et al.* 2021).

One of the most important parameters to consider when determining the viability of an electrode material is its estimated lifetime, given that it directly impacts initial investment and operational costs when considered as a consumable. In their techno-economic analysis, Stirling *et al.* (2020) considered electrode lifetime as a key assumption in their calculations for an EAOP point-of-use device. Although electrode lifetime is clearly a very important factor when determining an EAOP economic viability, not much information is available regarding this subject. In this regard, the review published by Moradi *et al.* (2020) focuses on electrode lifetime as a key parameter for further EAOP development. Moradi *et al.* describe several aspects that affect electrode lifetime such as: applied current density, soluble compounds, method of preparation, electrode shape, pH, temperature, and in the case of composites bonded to a substrate, they identified oxygen bubbles and the creation of high-pressure zones at the electrode surface, since they are the main sources of corrosion during EAOP operation. Given the time consuming and highly difficult task of obtaining real service lifetime values for novel electrodes, Moradi *et al.*

point to accelerated lifetime tests under strong acidic conditions (0.5 M H_2SO_4) with a high current density (200 mA cm^{-2}) as way to estimate the approximate service lifetime of electrodes (Lim *et al.* 2018; Yao *et al.* 2019; Zhang *et al.* 2019a; Xia *et al.* 2020). Data analysis demonstrated that BDD, TiN, and IrO_2 doping significantly increased the lifetime of PbO_2 and DSA electrodes, reaching between 3.9 and 10.4 years of service (Moradi *et al.* 2020).

3.2. Operational costs

Although electrode costs tend to be the bulk of the economic burden in EAOPs (especially for BDD-based processes), operational costs can also diminish commercial interest in EAOP application. Current efficiency is the main parameter to consider for assessing operational costs, as energy consumption can more than quadruple for processes that operate at high current densities where the process is controlled by mass transfer (Radjenovic & Sedlak 2015). Many efforts have been made to address this problem, from new enhancements in electrode preparation (Hu *et al.* 2021; Salazar-Banda *et al.* 2021) to new reactor designs like the one recently reported by Muddemann *et al.* (2021) in which a process involving a novel BDD anode deposited on a tantalum substrate and a gas diffusion electrode (GDE) as cathode are used to treat an artificial wastewater sample containing phenol and showed that the novel BDD–GDE system can achieve complete mineralization with a reduction in energy consumption of 25, 86, and 92% when compared with BDD-stainless steel, ozonation, and peroxonation systems, respectively. The increase in energy efficiency can be attributed to an improvement in the mass transfer conditions given the 3D structure of GDE electrodes and the absence of the hydrogen evolution reaction which doesn't contribute to contaminant degradation. Muddemann *et al.* also estimated a lifetime of approximately 18 years for the novel BDD electrode based on surface corrosion analysis, and through an operational expenses (OPEX) analysis of a case study comparing with classic niobium-based BDD electrodes, they found that replacement costs can be diminished by a factor of 5, effectively reducing operational costs.

3.3. Cotreatments and synergistic processes

The coupling of EAOPs with synergistic processes, such as photo and ultrasonic irradiation, often leads to an increase in degradation and/or removal efficiency, and it is a very promising and researched subject that merits its own classification in EAOP technologies. EF processes can be enhanced by the irradiation of UV light (PEF) by increasing Fe^{2+} generation due to $Fe(OH)_2^+$ photoreduction and photodecarboxylation of Fe(III)-carboxylate species (Brillas 2020). However, the high cost of artificial UV lamps, both initial and operational, represents an important obstacle for applying PEF processes at a larger scale. Synergistic analysis considering both degradation efficiency and energy consumption can help determine to what extent photo-irradiation is worth the additional electric consumption as the additional costs can be several times greater than non-irradiated EAOPs (Khataee *et al.* 2012). Photo-irradiation operational costs can be reduced using solar driven EAOPs during some seasons, and in some locations, the great potential of free, renewable, and environmentally friendly solar radiation makes them very attractive for degradation of any organic pollutant, including PPCPs (Brillas 2020; Lozano *et al.* 2022). One of the main drawbacks of solar driven EAOPs is the relative lower degradation efficiencies obtained when compared to UV irradiation since most semiconductors used, such as TiO_2, work better under UV irradiation (approximately 420 nm). In recent years, several efforts have been made to increase the effectiveness of solar radiation in EAOPs. In this regard, various novel semiconductor electrodes considering heteroatom doping and composites with enhanced absorption ability to visible light have increased the viability of solar driven EAOPs (Brillas 2020; Ye *et al.* 2021; Lozano *et al.* 2022). Recently, Karthikeyan *et al.* (2020) did in-depth review of the recent advances in some of the most popular photocatalysts such as TiO_2, ZnO, WO_3, CuO, and SnO_2, reporting that minimizing the band gap energy and electron–hole recombination rates are key elements in further development of photocatalysts. They also noted that large-scale application requires the exploration of novel preparation methods that are facile, efficient, and cost-effective.

Ultrasound (US) radiation can also have a synergistic effect with EAOPs and significantly improve degradation efficiency (Lozano *et al.* 2022). However, US technology is quite costly, and one must keep in mind that the benefits may not be worth the additional investment, both in the substantial initial cost of acquiring US equipment and the additional operational costs related to energy consumption. In this regard, careful optimization of operating parameters must be conducted to determine the conditions in which US radiation is justified, such as the recently published study by Kacem *et al.* (2022) in which they determined that US radiation could be used for the degradation of a model compound (Mordant orange-1) when in the presence of sulfate $\geq$20 mM for the entire process or just for the initial part when sulfate is not present. Nevertheless, they found that synergistic effects tend to be higher for low current values.

Some of the most promising applications of EAOPs lies within their highly versatile nature, which is mainly explored when they are used as a cotreatment to achieve a certain objective, such as meeting a discharge criteria. There are many examples of EAOPs employed as an additional treatment. Recently, Feng *et al.* (2021) published a study in which they treated landfill leachate using a combination of EAOPs (BDD anodes), biological treatment (anoxic–aerobic), and ultrafiltration. Feng *et al.* found that the highest current efficiency occurred at low current values (1 mA/cm^2). Most interestingly, they analyzed the impact of different operational conditions (mainly current density) on the overall operational cost and found that combining EAOPs and biological treatment had the lowest operational cost (0.76 USD/m^3) of all the technologies they analyzed. Additionally, they found that the highest cost savings (up to 54%) were achieved for the lowest current density value studied, which corresponds to the highest current efficiency, in accordance with some of the aspects discussed in the previous subsection.

4. CONCLUSIONS

EAOPs are undoubtedly a very promising set of technologies that could drastically change the way water treatment is done in the next decade. There is no clear unique research path to make EAOPs more competitive, and current research seems to be correctly focusing on all the factors that could increase EAOP's market share, including their application to specific PPCPs such as antibiotics, anti-inflammatory drugs, and others. Given the potential problems generated by transformation products during EAOPs, complete mineralization is an important matter for EAOP further development; however, more efforts such as the one recently proposed by Stirling *et al.* (2020) to identify key innovations through techno-economic analysis or case studies like the one proposed in the recent publication of Muddemann *et al.* (2021) using OPEX analysis to study the economic viability of novel reactor configurations are needed to help complement the many studies already available on EAOP novel materials and their application on specific pollutants (He *et al.* 2019; Karthikeyan *et al.* 2020; Hu *et al.* 2021; Salazar-Banda *et al.* 2021; Lozano *et al.* 2022). Finally, accurate data on key elements of EAOPs such as electrode lifetime (Moradi *et al.* 2020) are vital for further economic analysis.

DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

CONFLICT OF INTEREST

The authors declare there is no conflict.

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