Electrochemical treatment of wastewater to remove contaminants from the production and disposal of plastics: a review

Borislav N. Malinović1 · Jernej Markelj2 · Andreja Žgajnar Gotvajn2 · Irena Kralj Cigić2 · Helena Prosen2

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Abstract
Wastewater is a major source of contaminants originating from the production, usage, and disposal of plastic materials. Due to their poor biodegradability, these contaminants in municipal wastewater treatment plants, additional advanced oxidation processes such as electrochemical treatments have been developed to improve the standard biological treatment. Here we review the applications of electrochemical treatments of wastewater for the removal of the following plastic contaminants: bisphenol A, phthalic acid esters, and benzotriazoles. We present the effectiveness of treatment in terms of contaminant removal and mineralization; the identification of transformation products; toxicity assessment; and process energy requirements. In the present review, we have focused on the applications of electrochemical treatments of wastewater for the removal of three important groups of contaminants originating mainly from plastics: bisphenol A, phthalic acid esters, and benzotriazoles. The review focuses on the research of electrochemical treatments for these contaminants in the last five years. The papers are assessed from the point of i) effectiveness of treatment in terms of contaminant removal and mineralization; ii) identification of transformation products; iii) toxicity assessment; iv) processes’ energy requirements. Electrochemical treatments were confirmed to be a viable option for the removal of selected contaminants from wastewater.

Keywords Bisphenol A · Benzotriazoles · Electrochemical treatment · Emerging contaminants · Plastic materials · Phthalic acid esters · Wastewater

Introduction
Human activities introduce synthetic chemicals into the environment, influencing environmental processes and affecting living organisms. Therefore, these chemicals are regarded as contaminants. Most problematic older contaminants have been restricted, so the focus recently shifted to emerging contaminants (ECHA 2020; NORMAN 2020; Richardson and Kimura 2020; Yusuf et al. 2021). The generally applied definition is that these are contaminants that exhibit some adverse effect on humans or other living organisms and are not included in monitoring programs or not commonly regulated (Geissen et al. 2015; Richardson and Kimura 2017). Evaluation of their adverse effects on living organisms is problematic, so effect-based monitoring or prioritization by weighted average risk quotient is recommended (Yang et al. 2022b; Yusuf et al. 2021).

NORMAN is a network of organizations and laboratories for the monitoring of emerging contaminants. Currently, NORMAN database has 21 categories, consisting mainly of lower molecular weight compounds (NORMAN 2020). More complex materials are also considered as emerging contaminants, e.g. nanomaterials and microplastics (Geissen et al. 2015; Richardson and Kimura 2020; Richardson and Ternes 2018). The most harmful compounds are included in the watchlists in the European Union, USA, and elsewhere, but most remain unregulated.
Emerging contaminants are principally released into the environment via wastewater treatment plants using aerobic digestion processes (Richardson and Kimura 2017; Taheran et al. 2018). Several novel treatments have been proposed to enhance removal of contaminants from wastewater and can be roughly divided based on the prevalent removal mechanism (Akporie and Conradie 2020; Kaloudas et al. 2021; Mecha and Chollom 2020; Mousset and Dionysiou 2020; Mudhoo and Sillanpää 2021; Wu et al. 2021; Zhong et al. 2021). These treatments are shown in Fig. S1 and are frequently combined. Here we review electrochemical treatments of selected emerging contaminants in wastewater, originating mainly from the production and disposal of plastic materials (Fig. 1). Three groups were considered: bisphenol A, o-phthalic acid esters, and benzotriazoles. This review is an abridged version of the chapter published by Malinović et al. (2021) in the series Environmental Chemistry for a Sustainable World, volume 66.

Properties, toxicity, and occurrence of bisphenol A, phthalic acid esters and benzotriazoles

Emerging contaminants share some common features:

(i) Concentrations in the environment are very low (Taheran et al. 2018). Adverse effects on living organisms are the consequence of long-term exposure, causing carcinogenicity, endocrine disruption, reproductive health risks, or epigenetic changes (Richardson and Kimura 2017; Sharavanian et al. 2020).

(ii) Most are (bio)degradable, but are considered “pseudopersistent” due to continuous release into the environment. They are transformed into new compounds that are not necessarily less problematic (Richardson and Kimura 2017; Taheran et al. 2018).

(iii) The main point of entry of emerging contaminants in the environment is wastewater treatment plants using aerobic digestion processes (Richardson and Kimura 2017; Taheran et al. 2018), but agriculture and industry also contribute a part of emissions (Geissen et al. 2015).

(iv) Their environmental fate, spatial, and temporal distribution are poorly understood and are likely to change due to climate changes (Richardson and Kimura 2017).

(v) Emerging contaminants are mostly studied in wastewaters, surface waters, and groundwaters (Richardson and Kimura 2020; Richardson and Ternes 2018), although occurrences in other environmental samples, plants, animals, and food were also confirmed.

(vi) They are present worldwide, but are generally well documented only in European Union, USA, Canada, and Australia (Richardson and Kimura 2020; Richardson and Ternes 2018), while much less information is available for the rest of the world.

We are especially interested in anticorrosives, such as benzotriazoles, and substances present in plastics, such as phthalates, bisphenol A, and benzotriazolic ultraviolet filters. These emerging contaminants are listed also in NORMAN network (NORMAN 2020). Their properties and occurrence are detailed below.
Bisphenol A

Bisphenol A or 4,4′-(propane-2,2-diyl)diphenol is a precursor in the production of widely used types of plastic, i.e. polycarbonates and epoxy resins (Aravind et al. 2019). As a high production chemical, bisphenol A is used in containers for beverages and food, resin coatings, infant feeding bottles—banned in European Union since 2011, electronic equipment, construction glazing, sports safety equipment, thermal paper, medical devices, dental sealants, and similar (Liu et al. 2021b).

The most important emission sources are wastewater treatment plants, runoff, landfill leachate, and industrial discharges. Typical concentrations found in environmental samples (Corrales et al. 2015) are 5–370 µg/L in wastewater treatment plants effluents; up to 56 µg/L in surface water; 0.014–1.3 µg/L in tap water; around 10 µg/kg in sewage sludge and biosolids; more than 100,000 µg/kg in industrial effluents; 0.01–1,000 µg/kg in soil and sediments with values above 100 µg/kg in electronic waste recycling and disposal sites. Persistence in the environment is variable (Liu et al. 2021b), with average half-life time of 3–4 days in natural waters and up to 340 days in soils and sediments. Consequently, bisphenol A is classified as a priority pollutant in European Union Water Framework Directive.

Bisphenol A is moderately toxic to terrestrial and aquatic plants (MubarakAli et al. 2019; Xiao et al. 2020) and has a low potential for bioconcentration in aquatic organisms (Liu et al. 2021b). In human biomonitoring studies, bisphenol A was found in urine, blood, milk, and other biological samples (Gramec Skledar and Peterlin Mašič 2016) and was shown to have genotoxic, reproductive toxic, cytotoxic, neurotoxic, and endocrine-disrupting effects (Liu et al. 2021b). Human exposure has been linked to adverse health effects, such as reduced basal testosterone secretion, low birth weight of newborns, diabetes, obesity, and cardiovascular diseases. European Food Safety Authority set the limit of tolerable daily intake of bisphenol A to 4 µg/kg body weight per day (Gramec Skledar and Peterlin Mašič 2016).

Phthalic acid esters

Phthalic acid esters or phthalates comprise of dialkyl or aryl–alkyl diesters of o-phthalic acid that are commonly added as plasticizers up to 70% to improve the durability and flexibility of plastics. Phthalates are found in different consumer and industrial products, including personal care products and food contact materials (Radke et al. 2019).

Phthalates accounted for over 55% of global plasticizer consumption in 2020, and their use is expected to increase (IHS Markit 2022). Phthalates are not chemically bonded to the polymer, which means relatively easy release due to exposure to sunlight, changes in temperature or pH, and contact with solvents. For example, the release of some phthalates from baby bottles is 50–150 µg/kg of food content (Padervand et al. 2020). Consequently, phthalates are ubiquitous in the environment, food, beverages and were also detected in most microplastics (Padervand et al. 2020).

Ubiquitous occurrence of phthalates leads to human exposure. As some can adversely impact the endocrine system (Tijani et al. 2016), Environmental Protection Agency set an exposure limit of up to 6 mg di-(2-ethylhexyl) phthalate per day and 263 mg dimethyl phthalate per day for a healthy male (Benjamin et al. 2017), although it is estimated that human exposure to phthalates may exceed 1.0 g per day.

Some phthalates are classified as priority pollutants by environmental protection agencies, e.g. in European Union, USA, Canada, and China. The European Commission in 2020 banned certain phthalates that are classified as toxic to reproduction in all articles in concentration equal or above 0.1% and classified some as very toxic to aquatic life. A guideline value for maximum concentration in drinking water is set to 8 µg/L by World Health Organization (WHO 2013). The International Agency for Research on Cancer has classified di-(2-ethylhexyl) phthalate as possibly carcinogenic to humans.

Benzotriazoles

Polar benzotriazoles comprise of 1H-benzotriazole and its polar derivatives, e.g. tolyltriazole. They are used as anticorrosive agents and added to industrial fluids, de-icing liquids for aircraft operation, domestic-use products, and similar (Kraševč and Prosen 2018, 2021). Non-polar derivatives are used as ultraviolet stabilizers in plastics, food packaging, cosmetics, textiles, etc. (Montesdeoca-Esponda et al. 2020). More recently, substituted benzotriazoles were applied as organic photovoltaic materials in solar cells (Cui and Li 2019; Li et al. 2022a).

Polar benzotriazoles are ubiquitously present in environmental waters, river sediments, soils, house dust, indoor air, and atmospheric aerosols, while their chlorinated transformation products are detected in disinfected water (Carighini et al. 2015; Richardson and Ternes 2018; Shi et al. 2019; Yang et al. 2022a). Emissions into the environment are mainly by airport drainwater and wastewater treatment plants. In European Union, over 97% of effluent wastewater contained benzotriazoles with concentrations 2–3 µg/L and up to 44 µg/L in untreated wastewater (Cantwell et al. 2015). 5,6-dimethyl-1H-benzotriazole and 5-chloro-1H-benzotriazole were detected in human urine and adipose tissue (Li and Ding 2021; Wang et al. 2015). The biodegradability of 1H-benzotriazole is low with half-life 114 days at aerobic and 315 days at sulphate-reducing conditions (Liu et al. 2011).
Non-polar benzotriazoles enter the environment by sludge disposal, wash-out from clothes, leaching from microplastics, and atmospheric deposition (Yang et al. 2022a). They adsorb on sediments or sludge in wastewater treatment plants (Liu et al. 2012). Non-polar benzotriazoles were found in marine organisms, e.g. up to 66.1 ng/g in dolphins (Nakata et al. 2010) or up to 46 ng/g in coastal fish (Montesdeoca-Esponda et al. 2020). Bioconcentration factors are up to 105, while half-lives are estimated to be 60 days in waters and 542 days in sediments (Lu et al. 2016).

Acute toxicity of benzotriazoles to animals is rather low (Shi et al. 2019) and the most toxic for fish cell cultures was 5-chloro-1H-benzotriazole (Zeng et al. 2016). Gati-dou et al. (2017) found slight inhibition of *Lemna minor* by five polar benzotriazoles only at quite high concentration. As for chronic effects, 1H-benzotriazole shows endocrine-disrupting activity (Speltini et al. 2016) and is a suspect human carcinogen (Wang et al. 2013). Polar benzotriazoles may interfere with foetal mitochondrial function (Chen et al. 2020).

Some benzotriazoles are classified as emerging contaminants by Water Environment Federation, Environmental Protection Agency, and European Union Candidate List of Substances of Very High Concern for Authorization (ECHA 2020). However, no environmental limit concentrations are as yet set for them, except for some isolated cases for drinking water (Pervova et al. 2010; Richardson and Ternes 2018).

**Electrochemical technologies for wastewater treatment**

Although electrochemical treatments have long been known, they have globally regained their importance over the last decades due to tightened environmental regulations on wastewater and increased efficiency. Available options are presented here. Treatments are usually developed and tested on laboratory scale in either batch, recirculation, or continuous mode (Fig. 2).

**Classical electrochemical treatments**

Classical electrochemical treatments include electrodeposition or electroreduction, electrocoagulation, electroflotation, electrodialysis, and electrodissolution. Electrooxidation is discussed in the next section because of its recent revival due to application of new electrode materials, classifying it among advanced electrochemical processes.

Cathodic electrodeposition or electroreduction is mainly used for the removal of heavy metals or regeneration of precious metals from electrolytes or wastewater. Many inorganic and organic pollutants can be removed from wastewater with lower probability of by-product formation, e.g. dechlorination of chlorinated organic compounds (Lei et al. 2019) or reduction of nitrite and nitrate (Kim et al. 2017; Malinovic et al. 2015).

Electrocoagulation is the formation of metal hydroxides as coagulants by electrolytic dissolution of sacrificial anode. Coagulation occurs primarily at cathode and coagulated pollutants are removed by sedimentation or electroflotation by evolved small hydrogen bubbles. Typical application is heavy metal removal from industrial wastewater (Ebba et al. 2021; Moussa et al. 2017). Only a small amount of sludge is generated (Garcia-Segura et al. 2017). The main disadvantages are the replacement of sacrificial anodes and sometimes electrode passivation.

In electroflotation, electrolysis of water produces very small bubbles of hydrogen and oxygen that make pollutants float on the water surface (Santiago Santos et al. 2018). Electroflotation is desirable for removing oil from emulsions (Genc and Goc 2018), dyes, surfactants, biological pollutants (Kolesnikov et al. 2019), and printing wastewater (Safwat et al. 2019). Combined electrocoagulation and

![Fig. 2 Laboratory electrochemical reactors for the development of electrochemical treatments: a—batch mode; b—recirculation mode; c—continuous mode; + = anode, – = cathode](image-url)
Electroflotation was applied for removal of microplastics from wastewater (Akarsu et al. 2021).

Electrodialysis is used to separate ionic substances from water, which can be applied to wastewater. The effluent flows through the middle compartment between two ion-exchange membranes, while electrodes are placed on the walls of the electrolyser. In electric field, ions move towards the electrodes and a concentrated solution remains between the membranes without any reaction or chemicals. Electrodialysis is applied to treat effluents from industrial processes and wastewater (Arola et al. 2019; Gurreri et al. 2020; Mohammadi et al. 2021).

Similarly, electrofiltration is a membrane process aided by electrical potential gradient. Electrified membranes were developed in recent years, leading to reduced energy consumption, reduced required potentials, elimination of electrode corrosion, and formation of by-products (Sun et al. 2021; Zhu and Jassby 2019).

Although successful for certain purposes, classical electrochemical treatments are rarely used to remove emerging contaminants from wastewater. The only exception is electrocoagulation.

**Advanced and combined electrochemical treatments**

Over the last few decades, several new and more effective technologies were developed for wastewater treatment, such as electrochemical advanced oxidation processes or new combinations of existing methods. These processes include electrooxidation, electro-Fenton, photoelectrochemical, and sonoelectrochemical technologies. Combination of these often leads to greater process efficiency, reduced reaction time, and increased process economy.

Electrocoagulation has recently been combined with ozonation by introducing ozone into the electrocoagulation reactor and was found to be more efficient than electrocoagulation alone (Wagh et al. 2020). Electrocoagulation can also be paired with electro-Fenton to allow iron coagulants to precipitate organic carbon that unnecessarily consumes hydroxyl radicals. Coupling microbial fuel cells with electrocoagulation was reported for treatment of municipal wastewater (Mei et al. 2019; Safwat 2019) with significantly reduced operational cost.

Today, electrooxidation is the most popular electrochemical technique for wastewater treatment. In direct electrooxidation, pollutants are degraded on anode surface by direct electron transfer from the adsorbed pollutant. Process is theoretically possible at lower potentials than for oxygen evolution and has a poor efficiency (Feng et al. 2016). However, electrodes are likely to inhibit further oxidation due to adsorbed pollutants on the surface. In indirect electrooxidation, pollutants are oxidized in the bulk solution by anodically generated redox reagents, such as chlorine and hypochlorite, hydrogen peroxide, ozone, persulphate, and metal mediators.

Electrooxidation can also occur indirectly at the anode by physisorbed reactive oxygen species causing predominantly complete degradation and by chemisorbed reactive oxygen species that selectively form oxidation products (Comninellis 1994). Generally, the former are much more efficient for oxidation of organic pollutants than chemisorbed species due to their high standard potential ($E^0 = 2.80 \text{ V vs. standard hydrogen electrode}$) that provides complete mineralization.

Many advanced oxidation processes are eco-friendly and based on the in situ production of the hydroxyl radical. Among them, electrochemical processes have significant advantages. The electrocatalytic properties of the anodic material determine the mechanism of electrooxidation. Hydroxyl radicals are the strongest oxidants feasible for wastewater treatment, but anodes with high overvoltage for oxygen evolution should be used to avoid consumption of radicals by competitive reactions. Boron-doped diamond anode is such a novel electrode material that can completely mineralize highly polluted wastewater with high efficiency up to 100% (Martínez-Huitle and Panizza 2018).

Besides hydroxyl radical, other strong oxidation agents can be formed at the anode, such as hydrogen peroxide and ozone. Oxidants electrogenerated from dissolved salts are active chlorine and peroxy-compounds, i.e. peroxodisulphate and sulphate radicals (Groenen Serrano 2018).

Ozone is a strong oxidant with high standard potential ($E^0 = 2.10 \text{ V vs. standard hydrogen electrode}$). If produced electrochemically, less acid intermediates are produced than with non-electrochemical ozonizer (Lara-Ramos et al. 2020). Hydrogen peroxide can be produced both on anode and cathode during the electrolysis, but cathodic production requires an oxygen gas supply. For regeneration of $\text{H}_2\text{O}_2$, good contact between the cathode, oxygen, and wastewater is required, making porous cathodes such as gas-diffusion electrodes and three-dimensional electrodes the preferred choice (Moreira et al. 2017).

The efficiency of electrooxidation treatment of wastewater can be enhanced by the addition of anions, which serve both to yield better conductivity and to electrogenerate strong oxidizing agents, such as active chlorine species. Presence of chlorine in treated wastewater could lead to the formation of organochlorinated species, which exhibit increased toxicity and are usually more recalcitrant than the parent molecules (Moreira et al. 2017). Sulphate addition causes formation of reactive sulphate species which function as secondary oxidants. Depending on the process parameters, electrode material, and type of pollutants, sulphate ions can achieve the same or higher efficiency than chlorides (Prosen et al. 2020). Unlike chlorides, sulphates are not considered to be pollutants. Peroxodisulphate can also be electrogenerated.
by sulphate oxidation at very high potentials (above 2 V vs. standard hydrogen electrode), strongly depending on the electrode material (Groenen Serrano 2018).

Electro-Fenton processes are also very popular as an efficient and economical alternative to degrade pollutants in wastewater. They are based on the synthesis of \( \text{H}_2\text{O}_2 \) and regeneration of ferrous ions by cathodic reduction of ferric ions on the cathode. \( \text{H}_2\text{O}_2 \) is usually produced by reduction of \( \text{O}_2 \) from compressed air in an acidic medium. Fenton reaction produces ferric iron that can be reduced at the same potential as for the electrogeneration of \( \text{H}_2\text{O}_2 \) (Oturan and Oturan 2018).

In the electro-Fenton process, the Fenton reagent and hydroxyl radicals are constantly formed, thus avoiding waste generation. The most important parameters are the type of anode, current intensity, oxygen solubility, generation rate of hydrogen peroxide, and pH (Jose Rueda-Marquez et al. 2020). Cathode materials are important for the generation of the Fenton reagent and are most often carbon felt or gas-diffusion electrodes, while glassy carbon and graphite are only sometimes used (Jain et al. 2018; Nair et al. 2021). Significant advances in efficiency and application have been made in recent years, leading to some new electro-Fenton-based processes: heterogeneous electro-Fenton, bioelectro-Fenton, sonoelectro-Fenton, photoelectro-Fenton, and solar photoelectro-Fenton.

Photoelectrocatalysis represents the synergy between photocatalytic and electrocatalytic processes. The process uses a semiconductor—photocatalyst—under ultraviolet or solar light illumination to generate electron–hole pairs with the possibility to degrade organic compounds by hydroxyl radical at the surface of semiconductor. Photoelectrocatalytic systems use different anode materials to degrade various organic pollutants (Antolini 2019).

Ultraviolet or solar irradiation of the photoactive electrode is the basis of photoassisted electrochemical treatments. Ultraviolet radiation sources may be UVA with wavelengths 315–400 nm, UVB at 285–315 nm, and UVC below 285 nm. In photoelectro-Fenton, electro-Fenton is combined with ultraviolet irradiation (Su et al. 2013; Zhu et al. 2019) or sunlight (GilPavas et al. 2018; Murillo-Sierra et al. 2018). The regeneration of ferrous ions is possible with a combination of \( \text{H}_2\text{O}_2 \) and a radiation source whereby a hydroxyl radical is formed. Faster pollutant removal can be achieved with photoelectro-Fenton compared to electro-Fenton (Casado 2019).

Electrochemical processes can lead to passivation of the electrode surface due to adsorption of pollutants, intermediates, or products and are often limited by mass transport in the system, significantly reducing the efficiency (Feng et al. 2016). Ultrasound can prevent both and can have a synergistic effect with electrochemical treatment by increasing mass transport. Sonoelectrochemical treatment causes a homolytic fracture of \( \text{H}_2\text{O} \) leading to the formation of the reactive oxygen species (Radha and Sirisha 2018). Cavitation microbubbles are created, causing intensive mixing and cleaning of the electrode surfaces. Ultrasound paired with electro-Fenton is known as sonoelectro-Fenton.

In addition to the technologies we have briefly described, there are many other less used and many other combinations that have proven effective in laboratory research for the treatment of a wide range of pollutants. However, wastewater treatment under realistic conditions requires scaling up, which presents new challenges, outlined in “Effectiveness of electrochemical treatments and comparison with other advanced oxidation processes” section.

**Electrochemical treatment of bisphenol A in wastewater**

Bisphenol A has a significant health impact and is present in a wide range of environmental systems, such as water, sediments, soil, and air (Corrales et al. 2015). This is probably the reason that bisphenol A is frequently used as a target contaminant (Jia et al. 2019; Oh et al. 2019) to investigate the removal from wastewater using various approaches, including electrochemical treatments.

Table 1 lists the recent applications of electrochemical treatments for the removal of bisphenol A from model aqueous systems and wastewater. Number of applications is quite large compared to other contaminants reviewed in this paper. Detailed parameters of treatments are provided in Table S1 in Supplementary Materials, offering opportunity for data comparison. Applications are further reviewed below with an emphasis on the outcome of the treatment, especially transformation products.

A great variety of applied electrodes can be observed. Typical electrodes are discussed in Sect. Electrochemical technologies for wastewater treatment, and only a few novelties or modifications were presented such as dimensionally stable anodes (Chen et al. 2017; Chmayssem et al. 2017; Wang et al. 2021; Yu et al. 2021; Zhang et al. 2020), composite electrodes (Sun et al. 2020; Zhai et al. 2021), membrane electrodes (Li et al. 2020b), or modified boron-doped diamond (Huang et al. 2017). There are special anodes for the photoprocesses, such as WO\(_3\) electrodes (Goulart et al. 2019; Xiao et al. 2019), while plain or boron-doped multiwalled carbon nanotube electrodes (Bakr and Rahaman 2017, 2019) or tubular coal-based carbon membrane (Pan et al. 2019) are used for electrochemical filtration.

For the cathodes, there are more options such as carbon or modified carbon (Cao et al. 2018; Chmayssem et al. 2019). Signifi-
| Process                          | Biophenol A                                                                 | Phthalic acid esters                                                                 | Benzotriazoles                                                                 |
|---------------------------------|-----------------------------------------------------------------------------|-------------------------------------------------------------------------------------|--------------------------------------------------------------------------------|
| Electrooxidation                | Aliakbari et al. (2019), Aravind et al. (2019), Deng et al. (2018), Huang et al. (2017), Wu et al. (2019), Yu et al. (2021) | Chen et al. (2022), Deng et al. (2020), Souza et al. (2014a), Souza et al. (2014c), Turan et al. (2020), Xu et al. (2021) | Ahmed et al. (2016), Ahmad and Ghambari (2018), Ding et al. (2010), Wu et al. (2013) |
| Electrocatalytic oxidation      | Chen et al. (2017), Li et al. (2020b), Sun et al. (2019b), Sun et al. (2020), Zhai et al. (2021) | Deng et al. (2010), Prosen et al. (2020), Saha et al. (2020), Wu et al. (2013), Xing et al. (2012) | Ahmed et al. (2016) |
| Photoelectrocatalysis           | Burgos-Castillo et al. (2018), Chen et al. (2021), Cui et al. (2019), Li et al. (2019), Zhang et al. (2018), Zhang et al. (2019), Zhu et al. (2020) | Feng et al. (2021), Wang et al. (2019a), Wang et al. (2019b), Wang et al. (2021), Xiong et al. (2021) | Ahmed et al. (2016), Ahmad and Ghambari (2018), Wang et al. (2019a) |
| Electro-Fenton                  | Burgos-Castillo et al. (2018), Cao et al. (2018), Chmayssem et al. (2017), Chu et al. (2021), Li et al. (2020a), Liu et al. (2021c), dos Santos et al. (2021a), Li et al. (2021a), Xiong et al. (2021) | Ren et al. (2018), Wang et al. (2020) | Ahmed et al. (2016), Ahmad and Ghambari (2018) |
| Combination of electrolysis, granular activated carbon, and $O_3$ | Fang et al. (2022a) | | Wang et al. (2019a) |

Table 1: Electrochemical treatment of biophenol A, phthalic acid esters, and benzotriazoles. More detailed review of the electrochemical treatments separately for each group of contaminants is available in Tables S1, S2, and S3 in Supplementary Materials.
One of the most popular electrochemical treatments for bisphenol A removal is electro-Fenton, which can be performed as a separate process or in combination with oxidation or photoprocesses. Chmayssem et al. (2017) identified some competing oxidation reactions, but showed that these had a negligible effect on bisphenol A removal. No transformation products were identified, although some were obviously present even at over 99% removal, as seen from the residual total organic carbon measurement. These products were speculated to be low-chain carboxylic acids. Zhang et al. (2020) reported a three-dimensional heterogeneous electro-Fenton system with good degradation efficiency and confirming pseudo-first-order kinetics. Bisphenol A degradation was explained by hydroxyl addition, bridge cleavage, and ring-opening reactions. The degradation products were identified using liquid chromatography-mass spectrometry (LC–MS) (Fig. 3).

Jhones dos Santos et al. (2021a, b) studied the degradation of bisphenol A in acidic solutions, observing large production of sulphate radical and confirming pseudo-first-order kinetics. Based on the gas chromatography-mass spectrometry (GC–MS) analyses, the initial stages of degradation were suggested and the structure of transformation products was described (Fig. 3). Similar work by dos Santos et al. (2021b) showed that combination of graphene sheets and nanoparticles provided excellent catalytic capacity in electro-Fenton mineralization. Two degradation intermediates were identified.

Divyapriya et al. (2017) studied the formation of $\text{H}_2\text{O}_2$ and hydroxyl radicals at different working electrode materials and different $\text{pH}$ at electro-Fenton conditions, again with model contaminant bisphenol A. $\text{H}_2\text{O}_2$ was formed at all electrode materials. The formation was enhanced in strongly alkaline medium compared to acidic due to competitive reactions, as well as at more negative potentials, but was diminished at composite materials containing $\text{Fe}_3\text{O}_4$ by increasing $\text{Fe}_3\text{O}_4$ ratio. One degradation product of bisphenol A was identified: 4-isopropenyl phenol (Fig. 3).
Some novel electrode materials for electro-Fenton were applied. Zhao et al. (2018) prepared an iron-copper-embedded carbon aerogel and comparison with carbon aerogels embedded with Fe or Cu only was made. The main focus was to investigate the mechanism of oxygen reduction reaction leading to the formation of $\text{H}_2\text{O}_2$ or $\text{H}_2\text{O}$. Finally, the process was applied to real samples of sanitary wastewater and was shown to effectively lower total organic carbon and total nitrogen content. Wang et al. (2021) investigated the Cu-based electro-Fenton process and the mineralization pathway for degradation of bisphenol A with Cu/N–C catalysts. Several degradation products were detected (Fig. 3), formed by bridge cleavage, formation of polymeric products, and aromatic ring cleavage. Cao et al. (2018) applied spent graphite from the lithium-ion batteries as cathode. Powder was used raw or leached either with acid or with acid and alkali. The best performance was achieved with the acid-leached spent graphite powder.

Several electrochemical advanced oxidation processes were compared for the degradation of bisphenol A in the study by Burgos-Castillo et al. (2018), namely oxidation with electrochemically generated $\text{H}_2\text{O}_2$, electro-Fenton, photoelectro-Fenton, and solar photoelectro-Fenton. Bisphenol A degradation and transformation products were monitored (Fig. 3). The treatment efficiency was in the following decreasing order: electrochemically generated $\text{H}_2\text{O}_2$, electro-Fenton, photoelectro-Fenton, solar photoelectro-Fenton. Authors explained this order by higher power of sunlight in comparison with lamp. Mineralization was the most efficient at lower current density. In chloride-containing medium, chlorinated aromatic transformation products were formed, while sulphate-containing medium resulted in aromatic intermediates. These products were then degraded to short-chain carboxylic acids.

Another important group of electrochemical treatments for bisphenol A removal are electrooxidations, either alone or in combination with chemical and biochemical treatments. Sun et al. (2019b), Sun et al. (2020), and Zhai et al. (2021) proposed the degradation pathway of bisphenol A in the electrocatalytic wet air oxidation process, which is a highly efficient technology for rapid mineralization under room conditions. Intermediate degradation products $p$-isopropenyl phenol, benzoquinone, and propanedioic acid were identified using GC–MS (Fig. 3). In addition, oxidation kinetics were described as first order. Sun et al. (2019b) and Sun et al. (2020) additionally assessed the toxicity of treated samples by measuring inhibition of *Escherichia coli* growth. While initial solution with bisphenol A showed severe inhibition of bacterial growth, the latter was obviously accelerated by the treated solution. Thus, conversion to compounds that can be used as bacterial nutrients could be suggested.

Zhang and co-authors (Zhang et al. 2021a, 2021b) investigated graphene oxide electrodes or the reduced version modified with S, N, or F. The effect of doping agents was investigated and improved catalytic activity was observed. These modified materials were relatively stable. The degradation products were characterized with GC–MS, and linear chain compounds, such as undecane and nonane, were found (Fig. 3).

Chen et al. (2017) used particle electrodes made of N-doped graphene aerogel for the degradation of bisphenol A in simulated wastewater. The new material provided high specific area and was more effective. Several aromatic intermediates were identified by liquid chromatography-tandem mass spectrometry (LC–MS/MS): hydroxylated bisphenol A, isopropylphenol, hydroquinone, and phenol (Fig. 3). Possible polymeric products were speculated, but none were identified. In the next phase, ring opening occurred, and the identified products were butanetraol, 2-methylpentyl alcohol, and short-chain organic acids such as maleic and oxalic.

The same research group (Zhang et al. 2018) synthetized a polypyrrole/reduced graphene oxide aerogel and used it as particle electrode for electrocatalytic oxidation of bisphenol A by catalytic approach with reduction of Cr(VI). Authors monitored transformation products by LC–MS/MS and identified different hydroxylated derivatives: 4-isopropylphenol, 4-isopropenylphenol, benzoquinone, 2,5,5-trimethyl-2-hexene, and divinylxythane (Fig. 3).

In the study by Wu et al. (2019), bisphenol A was degraded at newly prepared boron-doped graphene aerogel-modified gas diffusion electrode. Complete degradation was achieved in 1 h, with intermediate transformation products $p$-benzoquinone, phenol, 4-isopropylphenol, and 4-hydroxyacetone, which were further degraded to short-chain organic acids.

Electrooxidation at boron-doped diamond anode was systematically studied by Li et al. (2017). Besides the degradation rate of bisphenol A, transformation products, current efficacy, and consumption of chloride ions were also monitored. Some identified products were different chlorinated compounds, which were more persistent than bisphenol A.

Aravind et al. (2019) studied electrooxidation of bisphenol A that was eluted from thermal cash receipts, which is a significant source of bisphenol A in wastepaper, cellulose, and recycled paper products. Ti–$\text{TiO}_2$/$\text{IrO}_2$/RuO$_2$ anode was used. Several transformation products were identified by GC–MS and nuclear magnetic resonance (Fig. 3). Moreover, biodegradability, toxicity, and estrogenicity of the samples were determined by naphthalene-degraded consortia of microorganisms. Bisphenol A was toxic to bacteria, while the treatment increased biodegradability as the result of reduced toxicity. By modelling of acute eco-toxicity of identified aromatic transformation products, three of these were found to be mutagenic and carcinogenic. Increased
biodegradability led to the biodegradation of toxic products to more simple and non-toxic compounds, such as maleyl acetate, malate, fumarate, acetopyruvate, and oxoacidic acid.

Huang et al. (2017) studied samples of real wastewater, river water, and groundwater. Bisphenol A was degraded by electrooxidation with boron-doped diamond anode, and this treatment was compared to the combined treatment with the addition of Ce(IV) ions or to the treatment with Ce(IV) alone. The addition of Ce(IV) as a pure chemical or recycled from real spent thin-film transistor liquid–crystal display chromium-etching solutions was also compared. Although the removal was most efficient by treatment with Ce(IV) ions only, complete total organic carbon removal could be achieved only by electrooxidation. Similarly, main transformation product p-benzoquinone could not be degraded by Ce(IV) alone. The processes were then tested on real wastewater and environmental water samples spiked with bisphenol A. The degradation rate of contaminant was similar as in synthetic solution, but complete total organic carbon removal was not achieved even after 2 h of treatment. This study clearly shows that the results for synthetic solutions should not simply be extrapolated to real environmental samples.

Another group of electrochemical processes that are used for bisphenol A degradation are photoelectrochemical treatments. WO₃ photoanode was used in photoelectrocatalytic degradation by Xiao et al. (2019) and Goulart et al. (2019). In the first case bisphenol A was degraded using WO₃ photoanode and Xe lamp to simulate solar light and to irradiate the anode. In the second case modified Cu-doped WO₃ photoanode with different percentages of Cu addition was used. The electrode was irradiated by visible light and H₂O₂ was used as an additional oxidant. Bisphenol A was more effectively removed in the first case.

Li et al. (2019) prepared Co₃O₄/BiVO₄ nanostructured films by electrospinning and used the material as photoanode illuminated with Xe lamp at wavelengths above 420 nm. Photoanode with BiVO₄ only was also tested and found to be much less effective. Different modes of operation were tested: electrooxidation only, photochemical only, combination, and resulted in 21%, 55%, and 96% of bisphenol A removal, respectively.

Yan et al. (2019) designed a photocatalytic sulphate radical-based advanced oxidation process, which was catalysed by a visible light-responsive catalyst g-C₃N₄/Fe₂O₃ and enhanced by electro-assisted persulphate activation using a low-voltage bioelectricity supplied by a microbial fuel cell. High removal of bisphenol A was achieved, while two other tested catalysts g-C₃N₄ and Fe₂O₃ showed significantly lower performance. The g-C₃N₄/Fe₂O₃ catalyst could be recycled 5 times. This study is especially interesting because of the use of renewable sources visible light and microbial fuel cells. To compare, Chen et al. (2021) applied combination of electrolysis and ultraviolet irradiation to activate sulphite and obtain sulphate radicals. Approximately 30% contaminant removal was achieved after 30 min.

Another useful material for photoelectrodes is three-dimensional structured reduced graphene oxide-polyaniline/TiO₂ composite synthesized by Cui et al. (2018) and irradiated by a Hg lamp at λ 320 nm. Working electrodes made of TiO₂ only or polyaniline/TiO₂ were also tested. Different pollutants were degraded: phenol, bisphenol A, 2,4-dichlorophenol, and coking wastewater. Removal was the most effective with the composite material.

Although electrofiltration is considered to be a classical electrochemical treatment and not an electrochemical advanced oxidation process, it is nevertheless still under development and finds useful applications. Bakr and Rahaman (2017) applied pristine and boron-doped multiwalled carbon nanotubes for the removal of bisphenol A. LC–MS analysis was performed to identify the transformation products (Fig. 3). Authors postulated superoxide species as the dominant factor in the degradation and identified several transformation products, such as hydroxylated phenol derivatives, i.e. hydroquinone, which further decomposed to short-chain aliphatic compounds. In the presence of NaCl, some chlorinated phenolic intermediates and aliphatic compounds were found.

Bakr and Rahaman (2019) also tested bisphenol A and ibuprofen degradation with an improved cross-flow electrochemical filtration system using a superconductive blended membrane made of multiwalled carbon nanotubes and buckypaper. Identical membranes were used as anodes and cathodes, separated by a porous Teflon rubber separator. Transformation products were identified by LC–MS: both pollutants degraded to aliphatic organic acids with the common intermediates hydroquinone and p-benzoquinone (Fig. 3). Similarly, Pan et al. (2019) designed an electrochemical microfiltration process based on the coal-based carbon tubular membrane as anode. Transformation products were identified by LC–MS: p-benzoquinone and 4-isopropyphenol (Fig. 3); oxalic, maleic, and acetic acid that were subsequently mineralized.

Finally, use of an electrochemical system may not be applied per se to degrade pollutants, but as an aid to another advanced oxidation process. Jia et al. (2019) enhanced the Mn(II)-catalysed advanced oxidation process for sulphate radical production from sulphite by maintaining Mn(II) catalytic activity via constant oxygen production using the electrolysis of water with two mixed-metal oxide electrodes. The combined process was tested on bisphenol A and compared to the process without electrolytic production of O₂, where significantly lower removal of pollutant was observed. The developed process was subsequently tested on several water contaminants with equally good removal efficiencies.
Similarly, Deng et al. (2018) designed a combined electro-advanced oxidation process. A Mn$_{0.6}$Zn$_{0.4}$Fe$_2$O$_4$ catalyst was prepared from spent alkaline Zn-Mn batteries and applied for heterogeneous electro-activation of peroxysulphate in a cell with Ti/IrO$_2$-RuO$_2$ anode. Experiments were performed with or without catalyst, peroxysulphate, or electric current. The most effective bisphenol A removal was achieved with the combination of all three.

Alikarami et al. (2019) combined electrochemical and photocatalytic processes. The degradation of bisphenol A occurred because of photocatalytic generation of hydroxyl radicals on ZnO nanoparticles that were electrochemically generated from the sacrificial Zn anode. Separate photolysis and electrochemical processes were much less effective. The removal efficiency was further enhanced by the addition of sodium percarbonate or peroxymonosulphate. Degradation could be further increased by the application of ultrasound. In biotoxicity assay, the initial inhibition in bisphenol A-containing solution was decreased after 2 h of treatment. A peak in toxicity was seen after 30 min of treatment. In conclusion, two-hour treatment converted bisphenol A to less toxic products.

To summarize, some notable novelties in the field of electrochemical treatments for the removal of bisphenol A from aqueous systems have been identified in the reviewed papers. There are also a fairly large number of studies investigating kinetics (dos Santos et al. 2021a; Jhones dos Santos et al. 2021a, b; Sun et al. 2020; Wang et al. 2021; Xiong et al. 2021; Zhai et al. 2021). Promising technologies are used for the electrochemical treatment, such as electrochemical filtration in multiwalled carbon nanotubes (Bakr and Rahaman 2017, 2019). Also, new materials are employed, such as carbon aerogels as novel mesoporous carbon materials with a three-dimensional net structure of the only aerogel with electrical conductivity (Chen et al. 2017) and graphene oxide-polyaniline/titanium dioxide composite (Cui et al. 2018). Very important, the recycling of used materials from batteries was reported in the case of spent graphite from lithium-ion batteries (Cao et al. 2018) and in the case of spent raw material from alkaline Zn-Mn batteries (Deng et al. 2018).

The main drawback of many reviewed papers is the lacking assessment of the treatment outcomes. Most often, only target pollutant removal is monitored together with chemical oxygen demand or total organic carbon reduction. Transformation products’ identification is neglected in many papers, and toxicity studies are even more scarce. There are some notable exceptions (Alikarami et al. 2019; Aravind et al. 2019; Li et al. 2017; Sun et al. 2019b, 2020), where partial or more comprehensive toxicity assessment was done, making these papers especially valuable.

Electrochemical treatments of phthalic acid esters in wastewater

In recent years, the electrochemical treatment of $o$-phthalic acid esters or phthalates is gaining increasing attention. Most studies shown in Table 1 used dimethyl phthalate, diethyl phthalate, or dibutyl phthalate as model compounds (Chen et al. 2022; Hou et al. 2016; Souza et al. 2014b).

Some researchers also prepared synthetic wastewater (Deng et al. 2020) or used real samples (Feng et al. 2021). Electrochemical treatment was used in a variety of experimental setups, with a selection of anode materials used in direct electrochemical oxidation, including Ti (Turan et al. 2020), boron-doped diamond (Souza et al. 2014a), and modified PbO (Chen et al. 2022; Deng et al. 2020; Souza et al. 2014c). Coupled processes have also been applied, such as electro-Fenton system with CeO$_2$ nanoparticles (Ren et al. 2018), electro-peroxone (Hou et al. 2016), and photoelectrocatalysis (Feng et al. 2021; Wang et al. 2019a). A more detailed overview of the electrochemical treatments can be found in Table S2 in Supplementary Materials.

In a study by Souza et al. (2014a), direct electrooxidation of dimethyl phthalate using a boron-doped diamond electrode proved to be very effective. Dimethyl phthalate was completely removed after about 4.5 h, although low concentrations of oxalic acid were still present. However, boron-doped diamond electrode has two important disadvantages, namely high cost and cumbersome electrode preparation. Direct electrooxidation was also attempted in simulated synthetic wastewater using oxidized Ti and graphite electrodes (Turan et al. 2020). At optimal conditions, di-n-octyl phthalate was not detected in the solution after only 20 min. A composite electrode IrO$_2$-Ta$_2$O$_5$/Ti was tested by Xu et al. (2021) and showed promising electrocatalytic properties with the removal efficiency of 90% and 56% for dibutyl phthalate and total organic carbon, respectively, after one hour. Stability study showed no decrease in removal efficiency after three runs and negligible loss of Ta and Ir after 100 h of use.

Recently, a popular alternative is the modified lead dioxide electrode (PbO$_2$), which is relatively easy to prepare, reliable, and has promising electrocatalytic activity. Souza et al. (2014c) tested a fluoride-doped PbO$_2$ film electrodeposited on a Ti substrate (Ti/PbO$_2$F) and reported a 57% removal rate for dimethyl phthalate and a 35% total organic carbon removal rate after 4.5 h. Similar results were reported by Deng et al. (2020) who prepared a Ce-doped Ti/PbO$_2$ electrode. The removal efficiencies of dimethyl phthalate, diethyl phthalate, and dibutyl phthalate in synthetic wastewater after 4 h were 42%, 26%, and 63%, respectively. After 10 h, the removal efficiencies increased to over 95%, with the exception of diethyl phthalate at 81%. Chen
et al. (2022) demonstrated the use of polytetrafluoroethylene (PTFE)-doped PbO2 electrodeposited on a layer of vertically aligned TiO2 nanotubes (TiO2NTs). The comparison between Ti/PbO2, Ti/TiO2NTs/PbO2, and Ti/TiO2NTs/PbO2-PTFE showed that the conventional Ti/PbO2 had poorer degradation efficiency towards dibutyl phthalate, although the difference between electrodes with and without PTFE was not significant. However, the Ti/TiO2NTs/PbO2-PTFE electrode exhibited the lowest energy consumption, albeit at the expense of stability.

Ren et al. (2018) compared a traditional and modified electro-Fenton system using CeO2 nanoparticles as catalysts for H2O2 generation using four different electrolytes at initial pH 3.0: NaH2PO4, NaNO3, NaCl, and Na2SO4. An improvement in removal efficiency was observed in Na2SO4, while no significant change was observed in NaNO3 and NaCl. CeO2 nanoparticles showed a negative effect in NaH2PO4. Conventional ozonation is not an effective method for removing phthalic acid esters because they generally react very slowly with ozone. Therefore, Hou et al. (2016) tested electro-peroxone process in which H2O2 is generated in situ by cathodic reduction of O2 with a mixture of O2 and O3 sparged into the reactor. Further reactions produce hydroxyl radical. The study showed that the electro-peroxone process was indeed much more effective compared to ozonation alone, which removed only 14% total organic carbon and 99% of diethyl phthalate in one hour. In contrast, total organic carbon removal with the electro-peroxone process ranged 76–92% after one hour, and diethyl phthalate was completely degraded after only 5 min.

Photoelectrocatalytic degradation of dimethyl phthalate was demonstrated by Feng et al. (2021) using a copper-doped graphite carbon nitride composite on a carbon paper. The process was carried out under constant oxygen flow and imitated sunlight, removing the ultraviolet part of the spectrum. Complete degradation of dimethyl phthalate was achieved after one hour in a wide pH range of 3–11. Additionally, dimethyl phthalate removal process was applied to coal water slurry gasification wastewater with promising results. Chemical oxygen demand decreased by 95% within 8 h, while total organic carbon decreased by 98.5% within 3 h. In another photocatalytic degradation study,

![Fig. 4](image-url) Currently known electrochemical degradation pathways and transformation products of o-phthalic acid esters, namely dimethyl phthalate, diethyl phthalate, and dibutyl phthalate (Chen et al. 2022; Deng et al. 2020; Feng et al. 2021; Hou et al. 2016; Souza et al. 2014a; Souza et al. 2014c; Wang et al. 2019a). Main degradation pathways include dealkylation, hydroxyl addition, decarboxylation, and cleavage of the benzene ring. Degradation produces variety of transformation products, including monomethyl, monoethyl, or monobutyl phthalate, depending on the phthalic acid ester, phthalic acid, hydroxyphthalic acids, phthalic anhydride, hydroxybenzoic acids, mono-, di-, and tri-hydroxybenzene and several aliphatic carboxylic acids. Additional chlorinated phenolic derivatives were observed in chloro-containing matrix. Masses of individual compounds in Da account for stable isotopes with the lowest masses. Means of detection are specified in parenthesis for each product (GC–MS—gas chromatography-mass spectrometry; HPLC–UV—liquid chromatography-ultraviolet detection; HPLC–MS/(MS)—liquid chromatography-(tandem) mass spectrometry)
graphene-loaded TiO₂ nanotube array electrodes were used to degrade dibutyl phthalate (Wang et al. 2019a). Presence of anions negatively affected the efficiency in the following order: NO₃⁻, HCO₃⁻, SO₄²⁻, Cl⁻. Addition of electron acceptors improved the photocatalytic activity with the greatest improvement for H₂O₂.

The degradation processes have been studied for dimethyl phthalate, diethyl phthalate, and dibutyl phthalate. There are many possible pathways depending on a variety of factors, such as the predominant reactive species, ions present in the solution, and others. However, the major degradation pathways are dealkylation, hydroxyl addition, decarboxylation, and benzene ring cleavage, as shown in Fig. 4.

Dealkylation produces the following transformation products: monomethyl phthalate, monoethyl phthalate, and monobutyl phthalate, depending on the original phthalate (Deng et al. 2020). In the next step, phthalic acid is formed, followed by hydroxyl addition and formation of hydroxyphthalic acids (Hou et al. 2016) or removal of water and formation of phthalic anhydride (Feng et al. 2021; Wang et al. 2019a). Meanwhile, the hydroxyl radical can also initiate the decarboxylation and hydroxylation of phthalic acid to form hydroxybenzoic acids and mono-, di-, and tri-hydroxybenzene (Souza et al. 2014a, c). Additionally, formation of undesirable organochlorine compounds is possible in the presence of NaCl. In a study by Souza et al. (2014a), degradation of dimethyl phthalate was compared using a boron-doped diamond electrode with Na₂SO₄ or NaCl as the supporting electrolyte and found that the concentration of transformation products was higher when NaCl was used. In addition to the higher concentration of monomethyl phthalate and phthalic acid, other chlorinated phenol derivatives were observed in NaCl that were not detected in Na₂SO₄: 3-chlorophenol, 2,4-dichlorophenol, 2,4,6-trichlorophenol, and 4-chlororesorcinol. From here, the aromatic ring can open and further oxidize to produce a variety of low molecular weight organic acids, including succinic, maleic, oxalic, and formic acid. These aliphatic carboxylic acids can be completely mineralized to carbon dioxide and water. Another proposed pathway (Chen et al. 2022) is the hydroxyl radical attack on two carbon atoms relative to the carboxyl position, leading to ring opening and the formation of transformation products with long-chain structures.

Thus, it can be concluded that degradation mechanisms and products of electrochemical treatments are quite well known, but a thorough toxicological assessment of the treated wastewater is again missing.

## Electrochemical treatments of benzotriazoles in wastewater

Removal of 1H-benzotriazole and its analogues in wastewater treatment plants range from insignificant to 69% (Liu et al. 2012). These contaminants are also refractory to degradation by ozonation (Wang et al. 2020), which calls for more effective treatments. Studies on electrochemical removal are scarce and are presented in Table 1, with more details in Table S3 in Supplementary Materials.

Direct electrooxidation is rarely encountered. Xing et al. (2012) used boron-doped diamond electrode in Na₂SO₄ electrolyte and confirmed hydroxyl radicals as the dominant oxidants, removing 72% of 1H-benzotriazole in 5 h. In a more recent study by Prosen et al. (2020), two different anode materials were tested: boron-doped diamond and mixed-metal oxide IrO₂/RuO₂ on titanium. The former performed better with removal efficiencies after 2.5 h 94.4% in NaCl and 97.9% in H₂SO₄, while electrolytes Na₂CO₃ and Na₂SO₄ provided lower removal. Several chlorinated transformation products were identified in NaCl by liquid chromatography-tandem mass spectrometry (LC–MS/MS), while less problematic hydroxylated products were detected with H₂SO₄, which was selected as the optimal electrolyte.

Saha et al. (2020) investigated combination of vertical flow constructed wetland and direct electrooxidation on boron-doped diamond or mixed-metal oxide anode. The aim was to remove organic chemicals from saline cooling tower blowdown water, containing 0.8 ± 0.1 mg/L of 1H-benzotriazole. Electrooxidation was applied after or before wetland. In the first configuration, 97% of 1H-benzotriazole was removed by wetland and the remaining 3% by electrooxidation. In the second configuration, electrooxidation on boron-doped diamond removed 100% and on mixed-metal oxide 75% of contaminant in 3 h, but toxic transformation products in both cases negatively affected the wetland. Toxicity was evaluated by Vibrio fischeri assay and was abated by wetland; unidentified products were presumably chlorinated due to Cl⁻ presence in the sample.

Combined electrochemical treatment with light irradiation was used by Ding et al. (2010). Photoelectrocatalytic treatment was performed with a TiO₂-modified glassy carbon electrode and compared to direct electrooxidation, direct photolysis, and photocatalysis. Best results, 90% removal of pollutant in 180 min, were observed with a combination of photocatalysis and electrochemical oxidation. Wu et al. (2013) also investigated a heterogeneous photoelectro-Fenton process with ZnFe₂O₄ nanoparticles, graphite electrodes, and ultraviolet lamp. The best removal of 67% was seen with photoelectrocatalysis, followed by 78% by heterogeneous electro-Fenton process with ZnFe₂O₄ nanoparticles.
and additional light source. At optimized parameters, final removal efficiency of 1H-benzotriazole was 91%.

Ahmadi et al. (2016) tested photoperoxid-coagulation to remove 1H-benzotriazole with activated carbon fibre cathode and Fe anode in Na2SO4. At the optimal conditions, the removal was 96% in 60 min, the main mechanisms being coagulation and oxidation. Ahmadi and Ghanbari (2018) developed also a photoelectro-peroxone-Fe0 process to degrade several contaminants, including 1H-benzotriazole. Hydrogen peroxide was electrogenerated in situ, and ultraviolet C lamps were used for photo-formation of hydroxyl radical. Fe0 was a source of ferrous ions which formed hydroxyl radical with H2O2. At optimal conditions, complete removal of 1H-benzotriazole was achieved in 30 min.

Electro-peroxone process was applied by Wang et al. (2019b) to electrogenerate H2O2 from mixture of O3 and O2. Several antimicrobials were degraded in municipal wastewater, including 1H-benzotriazole and 5-methyl-1H-benzotriazole. Ozonation required up to 3 min to completely degrade both contaminants in synthetic sample, while the same was achieved in 1 min with electro-peroxone. In real effluent wastewater, 100% degradation required up to 5 min for both processes. 1H-benzotriazole was slower to degrade than 5-methyl-1H-benzotriazole.

Wang et al. (2020) tested various treatments for nitrobenzene removal: electrolysis by granular activated carbon as 3D electrode; ozonation; and the combination of both. The most effective combination of both processes was applied also to 1H-benzotriazole. The removal rate constant was 0.10 min−1, which can be translated into 98% removal in 40 min.

Several authors observed that even after complete removal of 1H-benzotriazole, transformation products were still present. Yet, very little is known about electrochemical degradation pathways, which are presented in Fig. 5. Ding et al. (2010) identified products from ultraviolet–visible spectra and by liquid chromatography. Authors proposed degradation mechanism with opening of the triazole ring and 2-aminobenzenediazonium formation. Loss of amino group yields benzenediazonium and further aniline. Ding et al. (2012) applied gas chromatography–mass spectrometry and theoretical calculations to identify 2-nitroaniline, 1,2-dinitrobenzene, and 2-nitrophenol. Prosen et al. (2020) identified several products by LC–MS/MS: monochloro-benzotriazole, methyl-benzotriazole, and dichloro-benzotriazole in electrolyte NaCl; aminophenol, mono-, di- and trihydroxy-benzotriazole in electrolyte H2SO4.

To conclude, electrochemical treatments of benzotriazole derivatives are still in their nascent phase and there are a

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**Fig. 5** Currently known electrochemical degradation pathways of 1H-benzotriazole from Ding et al. (2010), Prosen et al. (2020), Xing et al. (2012), and Wu et al. (2013). Main degradation pathways are the opening of the triazole ring and 2-aminobenzenediazonium formation, followed by the loss of amino group to yield benzenediazonium and aniline. Identified degradation products are 2-nitroaniline, 1,2-dinitrobenzene, 2-nitrophenol, aminophenol, mono-, di-, and trihydroxy-benzotriazole. Chlorinated products are formed in the presence of chloride: monochloro-benzotriazole, methyl-benzotriazole, and dichloro-benzotriazole. Means of detection are specified in parenthesis for each product (GC–MS—gas chromatography–mass spectrometry; HPLC–UV—liquid chromatography–ultraviolet detection; LC–MS/MS—liquid chromatography–(tandem) mass spectrometry; UV—ultraviolet detection). BTA—1H-benzotriazole
lot of grey areas, especially regarding the transformation products, degradation mechanisms, and toxicity assessment. Another fact worth mentioning is that no papers on electrochemical treatment of non-polar derivatives were found.

**Effectiveness of electrochemical treatments and comparison with other advanced oxidation processes**

**Cost-effectiveness analysis**

Energy consumption as the largest part of the total costs of electrochemical treatment is easy to compare and identify areas for improvement. Operating costs are usually expressed as energy required to remove mass of pollutant in raw influent, kWh/kg$_{\text{pollutant}}$; by chemical oxygen demand (COD), kWh/kg$_{\text{COD}}$; by total organic carbon (TOC), kWh/kg$_{\text{TOC}}$; or as energy required to treat specific volume of wastewater, kWh/m$^3$. There is no uniform approach of assessing energy efficiency for all types of wastewater treatment plants; therefore, the final decision has to be done by the user.

In selected eleven studies on electrooxidation of the real wastewater from the textile industry, consumption of electricity was 0.26–100 kWh/h (Garcia-Segura et al. 2018), while a comparison of nine other studies of different wastewaters shows energy consumption in the range of 0.2–345.5 kWh/kg$_{\text{COD}}$ (Garcia-Rodriguez et al. 2022). In three studies on electrodialysis of different wastewaters, electricity consumption ranged 0.16–21.3 kWh/m$^3$, while four different studies show 1.7–17.5 kWh/m$^3$ of energy consumption for electrocoagulation (Simon et al. 2018). The above data show a huge range of electricity requirements for the same processes, depending on the type of wastewater and process parameters.

Papers comparing the costs of conventional and electrochemical treatments are rare and due to their scarcity very valuable. For example, industrial plants are developed for electrocoagulation where costs for chemicals are reduced tenfold in comparison with chemical precipitation technique, 3.2-fold relative to chemical coagulation of textile wastewater or nearly twofold compared to chemical coagulation of metallurgical wastewater (Moussa et al. 2017). In a rare study at real-scale electrocoagulation plant for domestic wastewater treatment for a settlement with 17,500 inhabitants, which was monitored for 12 months, energy consumption was 0.49–0.54 kWh/m$^3$ or 0.24–0.28 EUR/m$^3$ (Koyuncu and Ariman 2020).

There are many suggestions for reducing operational costs such as electrodes with a large active surface (Moussa et al. 2017; Rondinini et al. 2018), as well as modification of the distance between electrodes, which has a linear influence on the electrolyte resistance and thus on the electricity consumption (Santiago Santos et al. 2018). The most interesting studies are those that show the cost-effectiveness of a certain treatment approach in terms of reactor design development and the possibility of a scale-up process. The treatment of pharmaceutical pollutants in water by electro-Fenton process confirmed that reactor configuration is the key point for efficiency (Monteil et al. 2019). Another interesting study was performed by Martínez-Huitle et al. (2015), who point out that cell design is a key factor for successful electrolytic treatment of wastewater in terms of efficiency and economy, where mass and heat transport, reactions kinetics, and current efficiency should be considered.

The principles of dimensional analysis applicable to scale-up of chemical reactors cannot be applied in the electrochemical reactor design because not all the similarity criteria match. The scale-up principle of chemical processes requires two scaling parameters used to define dimensionless variables. Commonly used criteria are geometric, kinematic, and thermal similarities between reactors. In electrochemical reactor, an additional criterion for scale-up is the current/potential similarity (Goodridge and Scott 1995). The geometric similarity criterion cannot be fulfilled for electrochemical cells, as increasing distances and electrodes would result in a large voltage drop and an increase in energy consumption. Thermal similarity is difficult to apply because of the effect of Joule heating inside the reactor. Additionally, an important parameter is the mass transfer at the electrode surface where the reactions take place. Most often, mass transfer is the bottleneck of all electrochemical processes, including wastewater treatments.

In case of increased energy consumption or complex wastewater, electrochemical treatments can be successfully combined with other treatments which lead to greater process efficiency and increased process economy. Optimizing the process in terms of treatment efficacy and acceptable costs can be as simple as maintaining optimal values of technological parameters, while in combined techniques more effort is needed to identify the potential for such optimizations.

**Toxicity evaluation after electrochemical treatments**

Any treatment efficiency evaluation should include toxicity and biodegradability testing. Fate of the treated wastewater determines the selection of toxicity test. If biological treatment will be applied, biotests with a mixed culture of microorganisms from activated sludge are the most appropriate (ISO 8192:2007 2020). In the case of further anaerobic biological treatment, the test measuring biogas production is a good choice (ISO 13641–I:2003 2020). For effluents in natural waters a battery of biotests with decomposers, i.e. bacteria; producers, i.e. algea Scenedesmus subspicatus or duckweed Lemna minor; and consumers, i.e. brine shrimp.
Artemia salina, water flea Daphnia magna, fish Brachidionio rerio, should be applied.

In recent studies only one or two biotests were applied (Aravind et al. 2019; Borowska et al. 2016; Han et al. 2018; Xiao et al. 2020). Some bioassays recently used for the contaminants in this review are presented in Table S4 in Supplementary Materials. Even more often, no assessment of toxicity after the treatment was included (Murrell and Dorman 2020; Pan et al. 2019). In several studies, toxic transformation products and increased toxicity were determined during advanced oxidation processes and electrochemical treatments even in the case of good removal rates accompanied by substantial mineralization (Aravind et al. 2019; MubarakAli et al. 2019).

Another approach to evaluate the toxicity is the application of calculations by the Environmental Protection Agency Toxicity Estimation Software Tool, which are based on Quantitative Structure–Activity Relationship methodology (Wu et al. 2020). This approach is more feasible for mixture of pollutants; method is faster, cheaper and requires practically no chemicals, making it more environmentally friendly (Sigurnjak Bureš et al. 2021).

The hazard of treated effluents in a natural environment is related to the bioavailability and (bio)accumulation of transformation products. Comprehensive biodegradability study can holistically identify those products that are environmentally relevant due to their recalcitrant and toxic properties. In common environmental conditions, assessment of ready biodegradability is usually enough. Readily biodegradable substances mineralize rapidly, i.e. in less than 28 days in common environmental conditions after a short lag phase and with non-hazardous transformation products. However, the available reviewed papers mostly lack such an assessment.

Comparison with other advanced oxidation processes

A variety of different technologies are applied to upgrade or replace conventional biological treatment approaches that are not efficient enough when persistent micropollutants are considered (Tiwari et al. 2017; van Gijn et al. 2021). A variety of conventional systems are well established and could be effective enough (Tang et al. 2019), but wastewater treatment plants are hot spots for emerging contaminants and their upgrade for removal of those pollutants should be a priority (Éramo et al. 2019; Sabri et al. 2018).

Chemical oxidation is an effective option for the treatment of wastewaters containing persistent micropollutants. One disadvantage of such systems is a high demand on electrical energy for devices such as ozonizers, ultraviolet lamps, and ultrasound generators, resulting in higher treatment costs and limited knowledge on the behaviour of the oxidation products in the biological post-treatment (Gulde et al. 2021).

Advanced oxidation processes are technologies involving in situ generation of hydroxyl radicals (Ribeiro et al. 2015). Various oxidation reactions and highly efficient generation of non-selective hydroxyl radicals with a high oxidation potential are employed in the processes often enhanced by catalytic, i.e. by TiO₂, ZnO, WO₃, Bi₂O₃, MoO₃, etc.; and photocatalytic processes by ultraviolet or visible light. Hydroxyl radicals are generated through chemical or photochemical reactions. Chemically, hydroxyl radicals can be generated using strong oxidants such as hydrogen peroxide and ozone or by combination of these (Sharma et al. 2018). Technologically feasible approaches are a combination of (i) ozone and ultraviolet, (ii) hydrogen peroxide and ultraviolet, (iii) photo-Fenton and Fenton-like reactions, and (iv) photocatalytic oxidation by ultraviolet and TiO₂. H₂O₂ combination with ultraviolet also enhances production of hydroxyl radicals which propagate a chain reaction and finally lead to the formation of O₂ and H₂O (Liu et al. 2021a). In photocatalytic oxidation, TiO₂ as a metal oxide semiconductor is excited after absorption of ultraviolet light and is able to oxidize a variety of organic compounds. Ultraviolet light can also be combined with chlorine to generate OH, Cl, Cl₂⁻, and ClO⁻ radicals (Gulde et al. 2021; Joseph et al. 2021).

Hydrodynamic cavitation was found to have a synergistic effect on advanced oxidation processes (Braeutigam et al. 2012; Patil et al. 2014; Zupanc et al. 2013). Many researchers used such hybrid processes (Choi et al. 2019; Gogate et al. 2014; Raut-Jadhav et al. 2013). Cavitation phenomena combine formation, cyclic growth, and rarefaction leading to the terminal implosive collapse of vapour bubbles in the liquid phase. These phenomena can be generated by turbulent flows, fast moving particles, boiling, electrical discharge, and laser or ultrasonic irradiation. Hydrodynamic cavitation can be efficiently coupled with other advanced oxidation processes, which led to the degradation of pollutants due to both mechanical, i.e. extreme shear forces in the bulk media, and chemical effects in the fluid (Čehovin et al. 2017; Raut-Jadhav et al. 2013).

Conclusion

Wastewater treatment plants are recognized as one of the most important emission sources of emerging contaminants in the environment and advanced oxidation processes are constantly improved to better remove contaminants from effluents. In electrochemical treatments, electrons are the main reactive species and additional chemicals are not required as wastewaters mostly have a satisfactory conductivity due to salts content. Transformation products cause secondary pollution, but can be avoided by correct choice.
of technique and electrode material. The equipment is relatively simple, easily amenable to automation and modular adaptation. However, electrical energy consumption and replacement of electrodes cause high operating costs. The former can be solved by using renewable energy sources and the latter by the use of recycled materials, with some examples given in the present review. Additional improvements are the development of new materials, e.g. nanomaterials, and new electrode shapes.

Reviewed electrochemical treatments are mostly small-scale applications. Wider industrial use could be achieved by better electrocatalytic activity of the electrodes; improved reactor design and hydrodynamics; electrode arrangement; and current efficiency. Process kinetics are frequently studied and very important in scaling-up. Another vein of research is the coupling of electrochemical treatments with conventional or advanced oxidation processes. Combination treatment is generally more effective due to synergistic effect.

Despite these advances and advantages, complete mineralization of contaminants is usually not achieved due to substantial energy requirements. Therefore, transformation products remain in the treated wastewater, but the reviewed studies generally lack the characterization and toxicity assessment of products. A major advance towards wider acceptance of electrochemical treatments would be a better elucidation of the outcome: What remains in the treated wastewater and how toxic is it? Therefore, additional studies need to be conducted in this field, at least for the contaminants covered in the present review.

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Declarations

Conflict of interest The authors have no relevant financial or non-financial interests to disclose.

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