A review of wastewater treatment technologies for the degradation of pharmaceutically active compounds: carbamazepine as a case study

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Abstract

The potential toxicological effects of pharmaceutically active compounds (PhACs) in natural water bodies have received considerable critical attention over the past years, encouraging numerous researchers to develop novel technologies to address the limitations of current wastewater treatment methods. Among the wide range of reported PhACs, carbamazepine is a well-known anticonvulsant that has often been used as a target compound in various studies because of its prevalence and resistance against degradation. The aim of this article is to summarize the research progress attained to date regarding the removal of this pharmaceutical as well as to evaluate and compare the performance of biological, physical, chemical and hybrid treatments. The review conducted on the available literature revealed that novel advanced oxidation processes (AOPs) and advanced reduction processes (ARPs) are a comprehensive set of promising techniques with an outstanding effectiveness, as most treatments allow for degradation rate constants in the order of 4 to 12 h^{-1}, meaning that 99% carbamazepine removal can be attained in less than 70 min under optimal conditions. In particular, electrochemical and photocatalytic AOPs have consistently reported successful degradation results.

Abbreviations: *AC:* Activated carbon, *AOP(s):* Advanced oxidation process(es), *API:* Active pharmaceutical ingredient, *ARP(s):* Advanced reduction process(es), *BDD:* Boron-doped diamond, *CAS:* Conventional activated sludge, *CBZ:* Carbamazepine, *CEC(s):* Contaminant(s) of emerging concern, *COD:* Chemical oxygen demand, *DOC:* Dissolved organic carbon, *DOM:* Dissolved organic matter, *EO:* Electrochemical oxidation, *GAC:* Granular activated carbon, *IW:* Industrial wastewater, *MBR*: Membrane bioreactor, *MOF(s)*: Metal-organic framework(s), *MWCO:* Molecular weight cut-off, *MWWTP:* Municipal wastewater treatment plant, *NF:* Nanofiltration, *nZVI:* Nano zero valent iron, *PAC:* Powdered activated carbon, *PFAS:* Per- and polyfluoroalkyl substances, *PhACs:* Pharmaceutically active compounds, *PMS:* Peroxymonosulfate, *PPCPs*: Pharmaceuticals and personal care products, *PS:* Persulfate, *PNECs:* Predicted no-effect-concentrations, *RO:* Reverse osmosis, *TOC:* Total organic carbon, *US:* Ultrasound, *UV:* Ultraviolet, *WW:* Wastewater, *WWTP:* Wastewater treatment plant.

1   Introduction

The latest advances in high-accuracy analytical techniques have shed light on the presence of numerous contaminants of emerging concern (CECs) in wastewater streams at concentrations ranging from ng L^{-1} to μg L^{-1} [1–3]. The CEC category refers not only to compounds recently detected in water bodies and whose effects are unknown but also to contaminants that have already been identified and whose persistence is becoming a problematic threat [4–6]. Compounds such as pharmaceutically active compounds (PhACs), pharmaceuticals and personal care products (PPCPs), endocrine disruptors, pesticides, flame retardants, perfluorinated compounds and household chemicals are common CECs found in wastewater that originate from diverse sources (e.g., industries, households, hospitals, landfills, agriculture, irrigation activities, etc.) [1, 7–10]. Numerous studies have highlighted the adverse ecotoxicological and human health effects that CECs may cause both in the short and long term [1, 8, 11–15]. Therefore, considerable research effort has been devoted to the development of novel technologies to solve this ever-rising issue in an effective, cost-efficient and sustainable manner [16–23].

Carbamazepine (CBZ) is a common anticonvulsant primarily used to treat epilepsy and nerve pain by reducing the spread of seizure activity in the brain [24]. It is currently consumed in large quantities worldwide, with an estimated consumption growth from 742 tons in 1995 to 1214 tons in 2015 [25]. After its oral administration, only 72% is absorbed by the human body and metabolized up to a 99% level, while the rest is discharged into the sewage system [26]. Its likelihood for biodegradation, photodegradation and volatilization is negligible, making it a suitable candidate for CEC degradation studies, especially regarding PhACs [26, 27]. In addition, it has been used as a contamination indicator in water and wastewater effluents [28]. In fact, it has been reported as the most frequently detected active pharmaceutical ingredient (API) in river catchments, being found in 62% of the 1,052 sampling sites analyzed along 258 rivers worldwide [29]. The concentration of carbamazepine in water streams has been reported to reach 23 ng L^{-1} in drinking water, 83 ng L^{-1} in groundwater, 23 μg L^{-1} in surface water, 200 μg kg^{-1} dry weight in sewage sludge and 259 μg L^{-1} in municipal and urban wastewater [11, 30]. These values exceed predicted no-effect concentrations (PNECs) for ecological toxicity (i.e., 25 ng L^{-1}), becoming a potential threat to aquatic ecosystems [1, 31, 32]. Specifics on the potential toxicological effects of carbamazepine exposure on aquatic species and human health have been reviewed by Hai et al. (2018), where the limited number of studies in the field as well as their diverse results call for additional risk assessments and thorough monitoring [33]. Due to its persistence and increasing presence, carbamazepine has therefore been internationally recognized as a concerning pollutant, included in the NORMAN List of Emerging Substances [30] and classified as an Endocrine Disrupting Chemical by the US Environmental Protection Agency [34].

The issue with the uncontrolled release of PhACs such as carbamazepine lies in the fact that they can eventually terminate into natural systems such as surface water, groundwater and soils, given that conventional (physico)chemical and biological treatments are not designed to achieve their complete elimination [2–4, 10]. Nevertheless, these conventional methods can be used as complementary steps in wastewater treatment since they offer unique advantages, such as being simple and efficient when removing biodegradable compounds as well as economic, environmentally friendly and safe [35, 36]. Among novel treatment methods, advanced oxidation processes (AOPs) have received particular attention over the past years. They are a wide set of technologies that rely on the generation of powerful species, mainly oxidative radicals, to degrade CECs [2, 6, 37, 38]. Similarly, advanced reduction processes (ARPs) operate based on the generation of reductive radicals [39, 40]. In both cases, radicals are species characterized by having an unpaired electron,

and thus showing a strong tendency to interact with other compounds to attain an even number of electrons, either by donating the unpaired one (i.e., acting as a reducing agent) or by accepting another electron (i.e., acting as an oxidizing agent) [41]. Radical generation methods can be categorized based on their dependence on energy sources (i.e., electro, sono, thermo or photochemical processes), an opposed lack of energy requirements (i.e., chemical and catalytic systems) as well as in case they show a hybrid design. There is a substantial body of literature regarding AOPs, as they have been shown to be suitable for both the pre- and posttreatment of contaminated wastewater. The pretreatment operation is preferred for highly toxic water influents to increase their biodegradability, while posttreatment applications are advised as a polishing step to increase degradation and mineralization efficiencies [42–44]. In particular, AOPs based on the generation of hydroxyl radicals ($^{\bullet}$OH) show the largest contribution, as reflected by previous scientometric analyses [45, 46]. In parallel, attention has also been dedicated to sulfate radical ($SO_4^{\bullet-}$)-based AOPs, which despite appearing later, have quickly gained importance in the field (Fig. 1). As further discussed in this review, carbamazepine degradation has accordingly been more extensively investigated through both types of AOPs than with other treatments.

The present article draws on the literature on the removal of carbamazepine from wastewater streams, including both real and synthetic effluents, to illustrate and compare the performances of multiple physical, biological and chemical technologies, with a special emphasis on the latter in the form of AOPs. Further experimental details of the different literature studies under review can be found in Tables 1 and 2.

Top 50 Keywords in Advanced Oxidation Processes
Evolution of recurrence for the most common keywords found in AOPs literature

Keyword	N times
advanced oxidation processes	4.774
wastewater	1.832
treatment	1.810
water	1.343
ozone	1.313
ultraviolet	1.277
radical	1.109
photocatalysis	1.022
oxidation	980
degradation	954
hydrogen peroxide	860
fenton	785
process	698
hydroxyl	626
organic	607
reaction	566
acid	527
dye	503
kinetics	438
photofenton	388
pollutants	376
pharmaceutical	354
sulfate	352
carbon	348
removal	345
toxicity	345
persulfate	337
catalysis	325
titanium oxide	325
iron	324
disinfection	312
membrane	300
effluent	294
reactor	281
catalyst	258
solar	256
heterogeneous	255
mineralization	255
compound	242
biodegradation	240
activated	239
chemical	229
contaminant	218
electrofenton	216
oxygen	206
electrochemical	205
oxides	203
emerging	202
dioxide	199
industrial	199

Figure 1: Evolution of the Top 50 keywords in the field of AOPs. This ranking is based on the number of times that a specific keyword has appeared in either the title, abstract or author keywords of a certain scientific publication related to the field of AOPs. To this end, the keyword combination *[wastewater OR waste water OR effluent OR water OR aqueous solution]* with *["advanced oxidation"]* has been applied to all documents indexed in Scopus. Accordingly, 8712 documents were screened, and their corresponding keywords were identified and ranked across time periods of 5 years. The total frequency of each keyword measured until the end of 2020 is shown in the last column. In particular, the keywords *radical*, *hydroxyl* and *sulfate* have been highlighted in the visualization to emphasize their relevance and recurrence.

2 Advanced oxidation processes

A vast literature exists on the application of advanced oxidation processes (AOPs) for carbamazepine degradation, comprising multiple types of radicals and activation methods. In this review, they have been grouped as (i) chemical and catalytic oxidation, (ii) electrochemical oxidation, and (iii) photo(catalytic)oxidation, depending on the primary activation method for oxidative radicals. For additional information on the underlying mechanisms and operating conditions of each AOP treatment, the reader is referred to the reviews by Cuerda-Correa et al. (2020) [6] and Wang and Zhuan (2020) [47] for $^\bullet$OH-based AOPs, and to the reviews by Guerra-Rodríguez et al. (2018) [48] and Giannakis et al. (2021) [49] for $SO_4^{\bullet-}$-based AOPs.

2.1 Chemical and catalytic oxidation

Among the chemical-based activation methods, the most relevant AOPs are those degradation systems based on ozonation, Fenton reaction and $SO_4^{\bullet-}$ radicals. Ozonation applied to different effluents containing less than 1 mg L^{-1} carbamazepine has shown consistent degradation results with efficiencies close to 100% among different studies, with an estimated ozone dosage in the order of 0.6 g O_3/g dissolved organic carbon (DOC) [3, 50, 51]. This is because carbamazepine is characterized by electron-rich moieties that easily react with ozone as well as by the cogeneration of powerful $^\bullet$OH and secondary radicals (Eqs. 1-8) [3]. On the downside, ozonation is heavily influenced by the presence of suspended solids, nitrite ions (NO_2^-), and effluent organic matter, which is also likely to present numerous ozone reactive moieties, leading to competition reactions that may hamper carbamazepine removal [3, 52].

$$O_3 + CBZ \rightarrow CBZ_{oxid} \tag{1}$$

$$O_3 + OH^- \rightarrow O_2 + HO_2^- \leftrightarrow H_2O_2 \tag{2}$$

$$O_3 + HO_2^- \rightarrow HO_2^\bullet + O_3^{\bullet-} \tag{3}$$

$$HO_2^\bullet \leftrightarrow H^+ + O_2^{\bullet-} \tag{4}$$

$$O_3 + O_2^{\bullet-} \rightarrow O_3^{\bullet-} + O_2 \tag{5}$$

$$O_3^{\bullet-} + H^+ \rightarrow HO_3^\bullet \tag{6}$$

$$HO_3^\bullet \rightarrow {}^\bullet OH + O_2 \tag{7}$$

$$\bullet OH + O_3 \rightarrow HO_2^\bullet + O_2 \qquad (8)$$
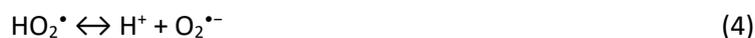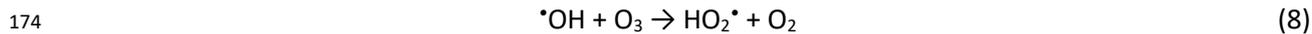
Regarding other $^\bullet OH$-based chemical activation methods, the classical Fenton reaction (Eq. 9) has been extensively investigated, which is based on the use of hydrogen peroxide (H_2O_2) and an iron catalyst to release $^\bullet OH$ radicals that yield pollutant degradation [53]. For carbamazepine removal, there is quite some variability in the results for Fenton-related treatments, ranging from ca. 7% up to 100% degradation [54–56]. The key factors affecting the treatment performance are the initial iron concentration since higher values yield higher degradation efficiencies, and the complexity of the water matrix since chloride (Cl^-), carbonate (CO_3^{2-}) and bicarbonate (HCO_3^-) ions are the main cause for $^\bullet OH$ scavenging, and thus decrease degradation rates [54, 56]. In terms of reported degradation rate constants, Matta et al. (2011) attained a value of 11.34 h^{-1}, with both Fe(II) and H_2O_2 concentrations up to 0.25 mM [54]. In recent years, Fenton processes have also been combined with $SO_4^{\bullet-}$-based systems, such as in the two-staged nanoscale zero valent iron (nZVI)-activated persulfate (PS) and Fenton process investigated by Wu et al. (2020), which yielded 77% carbamazepine degradation [57]. The main advantage of this system is the partial substitution of PS by H_2O_2, which reduces both operational costs and the release of sulfate (SO_4^{2-}) ions in the effluent [57].

$$Fe^{2+} + H_2O_2 \rightarrow Fe^{3+} + {}^\bullet OH + OH^- \qquad (9)$$
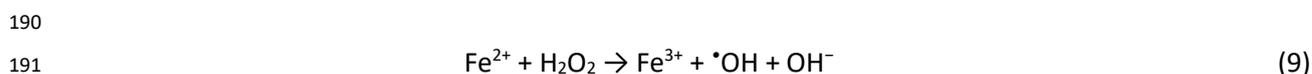
Finally, peroxymonosulfate (HSO_5^-, PMS) and persulfate ($S_2O_8^{2-}$, PS) are two common precursors for the generation of $SO_4^{\bullet-}$ radicals in conventional $SO_4^{\bullet-}$-based AOPs [58]. Matta et al. (2011) tested the activation of PMS for carbamazepine degradation through several cobalt-based catalysts (Eq. 10). Their results showed that the presence of Cl-substitutes in the catalyst could reduce the reaction rate constant by almost 40%, meaning that after 35 min of treatment, the degradation percentage could oscillate between 88% and 100% depending on the catalyst used. Additionally, the complexity of the water matrix had an inhibitory effect on the degradation, as the final carbamazepine removal was 50% after 1 h of treatment in a real wastewater effluent. The maximum degradation rate constant obtained in such a system was approximately 14.87 h^{-1} [54]. In the case of Fe(II)-activated PS (Eq. 11), opposite results were attained by Rao et al. (2014), as the presence of Cl^- ions accelerated the carbamazepine degradation rate, in contrast to the scavenging caused by nitrate (NO_3^-), sulfate (SO_4^{2-}) and dihydrogen phosphate ($H_2PO_4^-$) ions. In fact, 10 mM Cl^- enabled 99% degradation in 1 min, whereas a chloride-free system eliminated 60% of carbamazepine in 10 min, arguing that this could be caused by the cogeneration of additional chlorine-derived radicals [59]. Recently, researchers have also explored the use of novel catalysts based on both iron and cobalt for the activation of PMS. Wu et al. (2019) developed cobalt ferrite nanoparticles ($nCoFe_2O_4$) under several supports, of which organo-montmorillonite was found to be particularly promising as an inexpensive and effective support material [60]. It exhibited high carbamazepine removal efficiencies during three successive cycles (i.e., 73 − 93% degradation and up to 2.34 h^{-1} rate constant) due to the superior performance of Co^{2+} to generate $SO_4^{\bullet-}$ radicals, while ensuring less than 0.03% toxic cobalt leaching into the effluent [60]. A stable layered double oxide (LDO)-supported Co-Fe bimetal catalyst (Co_2FeAl-LDO) was investigated by Sun et al. (2020), leading to complete carbamazepine removal and faster degradation kinetics (i.e., up to 13.22 h^{-1}) due to not only the specific catalyst structure but also the synergistic effect of cobalt, iron and aluminium [61]. Their catalyst was effective up to the fifth cycle (i.e., 88% degradation) with an optimal catalyst dose of 60 mg L^{-1} and an initial PMS concentration of 0.2 mM, leading to 0.4 mg L^{-1} cobalt leaching [61].
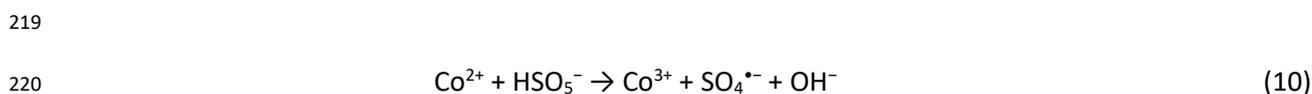
$$Co^{2+} + HSO_5^- \rightarrow Co^{3+} + SO_4^{\bullet-} + OH^- \qquad (10)$$

$$Fe^{2+} + S_2O_8^{2-} \rightarrow Fe^{3+} + SO_4^{\bullet-} + SO_4^{2-} \tag{11}$$

2.2 Electrochemical oxidation

Electrochemical oxidation is a popular approach for carbamazepine degradation, although comparison among experimental conditions is a complex matter since they are characterized by multivariate designs, where the material, size and shape of the electrodes, current density, pH, temperature and anolyte composition have all been reported to play a relevant role in the final removal efficiency. Across most studies under review, higher current densities, anolyte and precursor concentrations generally led to higher degradation rates, except for the electro-Fenton treatment because of the iron-related competition reactions [62]. First, the type of electrodes used is a distinguishable feature of electrochemical treatments since their nature directly influences their selectivity and surface reactivity [63].

Boron-doped diamond (BDD) electrodes have been reported to be powerful anode materials to generate multiple types of oxidative radicals on their surface, including $^{\bullet}OH$ and $SO_4^{\bullet-}$. One of their main advantages is that radicals can be formed without the need for chemical precursors since electron transfer mechanisms directly generate $^{\bullet}OH$ from water electrolysis and $SO_4^{\bullet-}$ from SO_4^{2-} ions (Eqs. 12 and 13). Farhat et al. (2015) compared the effects of sulfate- and nitrate-containing anolytes with a BDD anode, showing that the sulfate-based medium yielded reaction rate constants up to 10-15 times higher for multiple contaminants, even at low initial SO_4^{2-} concentrations (i.e., 1.6 mM). In the case of carbamazepine, the reaction rate constant obtained was approximately 29.40 h^{-1}, whereas under the same experimental conditions, the rate decreased to approximately 2.40 h^{-1} when $NaNO_3$ replaced Na_2SO_4 as the anolyte [64]. Song et al. (2018) studied the contribution of BDD electrochemical treatment on PS- and PMS-based AOPs, and in both cases, it was observed that carbamazepine degradation could significantly increase from 0.16 h^{-1} and 0.21 h^{-1} with PS and PMS alone, up to 6.81 h^{-1} and 18.77 h^{-1} with the added electrochemical oxidation, respectively [65]. Fast carbamazepine degradation using BDD (i.e., 11.34 h^{-1}) was also observed by García-Espinoza et al. (2018) in a spiked wastewater effluent when NaCl anolyte was used [27]. García-Espinoza and Nacheva (2019) further investigated the role of the anolyte species by testing BDD performance in the presence of Na_2SO_4, NaCl and NaBr as well as if no additional anolyte is added. Assays revealed that electrochemical oxidation was considerably faster in NaBr and NaCl (i.e., up to 22.81 h^{-1} and 7.08 h^{-1}, respectively) than in Na_2SO_4 synthetic solutions (i.e., up to 1.23 h^{-1}), both with and without oxygen addition [66]. Nonetheless, when applied to a spiked secondary effluent, carbamazepine degradation showed faster kinetics than in the synthetic solution with Na_2SO_4 anolyte (i.e., up to 2.75 h^{-1}). The rationale behind these results is that the increased matrix complexity enabled the generation of additional oxidants at the anode surface, such as HClO and $H_2S_2O_8$ [66].

$$BDD + H_2O \rightarrow BDD(^\bullet OH) + H^+ + e^- \tag{12}$$

$$BDD + SO_4^{2-} \rightarrow BDD(SO_4^{\bullet-}) + e^- \tag{13}$$
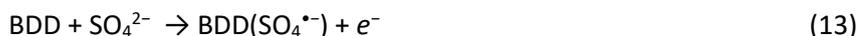
Another commonly used electrode material is Ti/PbO_2, although it has shown slower degradation kinetics than BDD. García-Espinoza et al. (2016) investigated the performance of Ti/PbO_2 anodes in a cylindrical mesh shape to promote the production of reactive oxygen species, such as O_3, H_2O_2 and $^\bullet OH$ radicals, obtaining a rate of approximately 0.77 h^{-1} [67]. Tran et al. (2017) tested rectangular Ti/PbO_2 anodes and attained a maximum degradation rate constant of 0.81 h^{-1} [68]. Circular Ti/PbO_2 and BDD anodes were compared by García-Gómez et al. (2014), and in this case, Ti/PbO_2 led to a slightly higher degradation (i.e., approximately 8% higher), although the attained kinetics were still similar to other Ti/PbO_2 studies (i.e., 1.26 h^{-1}) [63]. Other researchers have also investigated the performance of alternative electrode materials, such as Ti/Ta_2O_5-SnO_2 by Gurung et al. (2018). In comparison to conventional Ti/PbO_2, their newly developed anode showed a slightly lower carbamazepine degradation efficiency (i.e., 71.7% in contrast to 77.9% after 8 h). However, this material turned out to be more energy-efficient and effective during the treatment of a spiked membrane bioreactor effluent, while neither leaching heavy metals nor being affected by the pH [69]. A novel blue-colored TiO_2 nanotube array membrane filter anode was studied by Xu et al. (2021), which, due to the generation of multiple radical species (i.e., $SO_4^{\bullet-}$, $^\bullet OH$, $O_2^{\bullet-}$), showed fast degradation kinetics up to 24.18 h^{-1}. Their study also showed that the presence of Cl^- enhanced carbamazepine degradation since additional $Cl^\bullet$ radicals were formed, whereas NO_3^- inhibited the degradation rate because of a set of proposed competition reactions with $^\bullet OH$ radicals [70]. Finally, a mediated electrochemical oxidation process based on Co(III) was reported by Liu et al. (2019), where no radical species were detected and the main oxidizing substance was Co(III) ions, allowing for complete carbamazepine degradation in 50 min [4, 71].

Komtchou et al. (2015) compared electrochemical oxidation with the electro-Fenton process and electroperoxidation process. In their electro-Fenton study, different types of anode materials were investigated, where BDD allowed for up to 75% higher carbamazepine degradation and 4-fold higher TOC removal than Ti/Pt and Ti/SnO_2 [62]. However, it was revealed that the anode selection significantly affected the underlying degradation mechanisms, as BDD showed a predominant contribution of direct oxidation of carbamazepine (i.e., corresponding to 36% degradation) in contrast to electro-Fenton and electroperoxidation (i.e., attributed to 23% and 14% degradation, respectively). On the other hand, Ti/Pt promoted the electro-Fenton reaction (i.e., 36% carbamazepine degradation) over electroperoxidation (i.e., 21%) and direct oxidation (i.e., 8.6%) [62]. In comparison to other studies, the lower degradation percentages attained in the direct oxidation scenario may be attributed to the considerably lower current density applied.

2.3 Photo(catalytic)oxidation

Carbamazepine degradation by direct photolysis, both in natural and simulated sunlight, has not been found to be a successful treatment since considerably low degradation rate constants have been reported (i.e., lower than 0.01 h^{-1}) [72–74]. Photolysis in real wastewater effluents has shown slightly higher degradation rates (i.e., up to 0.022 h^{-1}) when additional matrix constituents enabled indirect photolysis pathways. For instance, the presence of NO_3^- ions promoted photodegradation given that it is a photosensitizer, and in contrast, humic acid hindered it since it absorbed part of the

available irradiation energy and reformed the parent compound [72]. As a result, photodegradation treatment has often been studied in combination with oxidant species, such as chlorine, ozone or hydrogen peroxide, as well as with the Fenton treatment, $SO_4^{\bullet-}$ precursors and metal catalysts.

Deng et al. (2013) performed a comparative study on the performance of different common oxidants with UV irradiation, namely, UV/H_2O_2, UV/PMS and UV/PS. Based on energy requirements, oxidant costs and operating conditions, the UV/PS system was proposed as the most efficient and economic method with a degradation rate constant up to 10.83 h^{-1}. In contrast, UV/H_2O_2 and UV/PMS yielded maximum degradation rate constants of approximately 7.45 h^{-1} and 1.55 h^{-1}, respectively. Experimental results also revealed that increasing the oxidant dose had a positive influence on the kinetics and that optimal pH conditions were acidic for UV/H_2O_2 and UV/PS (i.e., pH 3 and 5, respectively) and basic for UV/PMS (i.e., pH 11). Both UV/H_2O_2 and UV/PS were inhibited in the presence of Cl^- and CO_3^{2-}, whereas the UV/PMS system showed a degradation increase under certain conditions [75]. Other studies have also reported high carbamazepine degradation efficiencies (i.e., over 79% removal) within short reaction times (i.e., from 9 s to 3 min) under the combined treatment of UV with these oxidants [3, 51, 76]. Additionally, it was found that the presence of Fe(II) as a catalyst in the UV/PMS and UV/PS systems enhanced the removal efficiency, reaching almost complete degradation [76]. In another study, the carbamazepine degradation rate constant under UV/PMS/Fe(II) treatment was reported to reach 11.21 h^{-1} [74]. Few studies have also reported UV/O_3 as an effective treatment for the removal of carbamazepine [51, 77].

Regarding chlorine-based UV methods, Wang et al. (2016) observed that neither UV irradiation nor chlorination could effectively degrade carbamazepine when applied individually. However, the combination of both under acidic conditions showed a synergistic effect resulting from the combined generation of $^{\bullet}OH$ and $Cl^{\bullet}$ radicals, which allowed for a degradation rate constant of up to 85.2 h^{-1}. In simulated wastewater, the same treatment showed an inhibition of approximately 30% since dissolved organic matter (DOM) acted as an oxidation consumer and light filter as well as bicarbonate ions (HCO_3^-) reacted with both $^{\bullet}OH$ and $Cl^{\bullet}$ to generate carbonate radicals ($CO_3^{\bullet-}$), which are milder oxidant species towards organic compounds [78]. Sichel et al. (2011) also previously reported the potential of UV/chlorine systems from an energy point of view. Their research showed that 90% carbamazepine degradation could be attained in simulated wastewater at the technical scale (i.e., 250 L h^{-1} continuous flow reactor), while showing approximately 75% energy reduction compared to UV/H_2O_2 [79].

Improving the degradation efficiency in a UV system is possible via the addition of a photocatalyst such as a semiconductor oxide. In particular, TiO_2 has been widely investigated because of its low cost, high stability and activity, abundance, and feasibility for immobilization on different supports [80]. Franz et al. (2020) compared a heterogeneous TiO_2 mesh and conventional TiO_2 powders (i.e., Degussa P25) and reported that carbamazepine degradation rate constants were 1.08 h^{-1} and 10.44 h^{-1}, respectively. This difference originates from the various active surface areas of the catalysts, which also led to different UV dosage efficiencies. Nonetheless, even if the dispersed TiO_2 powders are more effective, the supported mesh is more easily recovered and regenerated [80]. Similar degradation kinetics have been observed on other heterogeneous [3, 74, 81, 82] and homogeneous TiO_2 systems [83], as summarized in Table 1. Other photocatalysts have also been explored for carbamazepine degradation, such as bismuth-, zinc- and tungsten-derived oxides. Given that the morphology and optical abilities of these catalysts directly influence adsorption and reaction mechanisms taking place, the results on carbamazepine removal are diverse. Under specific conditions, monoclinic-$BiPO_4$ and triclinic-WO_3 photocatalysts have reported degradation rate constants up to 7.86 h^{-1} and 7.64 h^{-1}, respectively [84, 85]. The highest degradation rate constant has been reported in acidic synthetic wastewater when using $BiOCl_{0.875}Br_{0.125}$, achieving

a value of 25.80 h^{-1} [83]. In the case of ZnO nanoparticles, photodegradation of carbamazepine has shown slower kinetics (i.e., 0.84 h^{-1}), coupled with reduced photocatalytic activity due to the photocorrosion of ZnO in aqueous solutions [81]. Finally, photo-Fenton treatment has also been explored for the degradation of carbamazepine in wastewater treatment [3, 56]. In the work by Klamerth et al. (2010), the solar photo-Fenton treatment displayed a degradation rate constant up to 0.84 h^{-1} even at low iron and H_2O_2 concentrations, although its performance was affected by the presence of $^\bullet$OH radical scavengers such as CO_3^{2-}. A main limiting factor for the application of this photo-Fenton system in real wastewater was that tests with *Vibrio fischeri* showed that the generated degradation products led to a toxicity increase [56].

3   Advanced reduction processes

Advanced reduction processes (ARPs) operate in an opposite manner to AOPs, as they rely on the formation of highly reactive reducing radicals, such as hydrated electrons (e_{aq}^-), hydrogen atoms ($H^\bullet$), sulfite radicals ($SO_3^{\bullet-}$) and sulfur dioxide radicals ($SO_2^{\bullet-}$), to degrade oxidized compounds [41, 86]. In particular, ARPs have been applied to the degradation of persistent per- and polyfluoroalkyl substances (PFAS), as already shown in the state-of-the-art review by Cui et al. (2020), where the chemical mechanisms behind several types of ARPs are discussed [86]. Regarding the degradation of carbamazepine, Yu et al. (2021) investigated for the first time its removal in a UV/sulfite ARP. Experimental results showed synergistic effects, where 99.6% carbamazepine degradation was attained after 30 min (i.e., with a degradation rate constant of 20.4 h^{-1}), in contrast to the nonsignificant degradation efficiencies observed when sulfite and UV were used independently [40]. The degradation rate constant was found to increase with increasing sulfite concentration, UV irradiance and pH, attaining optimal values under alkaline conditions since e_{aq}^- was the dominant reductive species [40].

4   Biological treatments

Biological wastewater treatments rely on the degradation of contaminants by the metabolism of microbial cultures. For the degradation of carbamazepine, conventional activated sludge (CAS) processes alone or in combination with membrane bioreactors (MBR) have been investigated, where the former entail a settling tank to separate the formed sludge and the latter are based on sludge separation via microfiltration or ultrafiltration membranes [33]. In both treatments, almost negligible carbamazepine degradation efficiencies have been observed (i.e., typically below 10%) [26]. These poor results are attributed to carbamazepine's moderate hydrophilicity (i.e., logP = 2.45), low water-sludge distribution coefficient (i.e., 1.2 L kg_{ss}^{-1}), and the presence of an electron withdrawing functional group (i.e., amide group) in its structure, which increases the biodegradation resistance [33]. Higher degradation efficiencies have been attained by Tahir et al. (2021) by selectively enriching wastewater with sulfate-reducing and fermentative acidogenic bacteria. After 144 h, their anaerobic bioreactor yielded 46% carbamazepine degradation, coupled with 36% total organic carbon (TOC) removal, 98% sulfate (SO_4^{2-}) reduction and the production of several volatile fatty acids (i.e., acetic acid, propionic acid and butyric acid) [87]. In addition, under these conditions, the degradation of carbamazepine was higher than that in a mixed microbial community (i.e., 36%), and the enriched microbial community was more resistant to the toxicity resulting from increased carbamazepine concentrations [87]. In a different study, carbamazepine degradation was enhanced

by the combination of activated sludge and gamma irradiation in a two-step treatment, leading up to 99.8% carbamazepine removal and 79.3% mineralization when 800 Gy irradiation was applied [88].

As an alternative to bacterial-dominated activated sludge processes, white-rot fungi have been found to be more effective in degrading recalcitrant compounds due to the generation of extracellular and intracellular enzymes, the latter being more suitable to target the amide group in the carbamazepine structure [33]. Nonetheless, attained efficiencies have been observed to be dependent on the fungal species and subsequent enzymes as well as on the need for operation under sterile conditions to avoid bacterial contamination [33]. In the treatment investigated by Hata et al. (2010), carbamazepine was treated with ligninolytic enzymes (i.e., laccase and manganese peroxidase). In their work, they showed that there was no significant carbamazepine degradation if laccase alone was used, whereas manganese peroxidase allowed for 14% elimination after one cycle of 24 h. In the presence of laccase with a redox mediator (i.e., 1-hydroxybenzotriazole), a single treatment cycle led to 22% removal in 24 h, and repeated 8-h cycles for a period of 48 h showed 60% elimination [89]. In the presence of crude lignin peroxidase, other studies reported less than 15% elimination after 2 h [90]. The highest removal efficiencies have been observed through specific whole-cell white-rot fungi under sterile conditions, where multiple enzymes are generated [33]. For instance, species such as *Bjerkandera* sp. R1, *Bjerkandera adusta* and *Pleurotus ostreatus* (PC9) allowed for 99% removal in a span of 14 to 32 incubation days [91, 92]. Nevertheless, lower efficiencies have also been observed (i.e., less than 60% elimination) for several white-rot fungal cultures under different operating conditions [33, 93].

5   Physical treatments

Activated carbon (AC) is a well-established material used in adsorption processes because of its high porosity, surface area and affinity to interact with a wide range of compounds, eventually retaining them on its surface [3, 94]. It is typically used in either powered or granular form, referred to as PAC and GAC, respectively [95, 96]. Given that carbamazepine is considered an adsorbable compound that stays in neutral form regardless of the operating pH, it has been found to be suitable for AC separation [33, 97]. Under PAC, over 90% removal effectiveness has been reported, even if low carbon doses were used [3]. In the same study, a GAC bed reactor enabled the removal of 72% carbamazepine in 23,400 bed volumes [3]. The difference between both methods lies in the fact that PAC has a larger specific surface area for removal [33]. More recently, metal-organic framework (MOF)-derived nanoporous carbons have attracted growing interest as adsorbents because of their high porosity, specific surface area and chemical stability [98, 99]. In the study by Yu et al. (2022), MOF-5 showed a higher adsorption capacity and faster uptake rate than ZIF-8-derived nanoporous carbon and commercial PAC for the removal of carbamazepine, with the graphitic defects being the key feature to improve the adsorption [99]. Their MOF-5 could be reused for 5 cycles, attaining removal efficiencies higher than 87% [99]. Other recently developed adsorbents with a maximum capacity for carbamazepine adsorption ranging from approximately 208 to 286 mg g^{-1} are activated palm kernel shell [100], zirconia/porous carbon nanocomposites [101], hierarchically structured carbon composites [102] and biomass activated carbon [103], which are comparable to the adsorption capacity of commercial PAC (i.e., 225 – 274 mg g^{-1}) [100, 104].

Regarding membrane filtration in wastewater treatment, pollutant removal relies on the effectiveness of a pressure-driven physical barrier to impede the passage of target compounds and hence separating contaminants from the influent. In wastewater treatment facilities at a large scale,

nanofiltration (NF) and reverse osmosis (RO) are the most commonly used types [3]. In terms of carbamazepine separation, Radjenovic et al. (2008) reported carbamazepine rejection percentages higher than 97% and 99% when NF and RO membranes were used, respectively [105]. Hai et al. (2018) also reviewed available studies on carbamazepine removal via NF and RO, showing that the final efficiency was strongly dependent on the molecular weight cut-off (MWCO) of the membranes and their fouling [33].

Both adsorption and filtration systems are essentially separation methods that do not entail degradation mechanisms, and consequently, they need to be coupled with an additional unit to carry out pollutant elimination, typically through advanced oxidation processes. Nonetheless, both are useful techniques to reduce the wastewater volume to be further treated and hence minimize the need for large-scale operations.

6   Hybrid treatments

The purpose of hybrid processes is to combine multiple wastewater treatment methods to promote a synergistic effect that can enhance pollutant degradation. To this end, carbamazepine degradation has been explored under sonoelectrooxidation [68], sono-Fenton [106], radiolysis/H_2O_2 [31], photoelectrocatalysis [80, 107] and UV/PS/Fe(II)/US/H_2O_2 treatments [108].

Tran et al. (2017) investigated the performance of an electrochemical cell in combination with ultrasound, where Na_2SO_4 was used as the anolyte and Ti/PbO_2 and Ti were used as the anode and cathode, respectively. Their results revealed that the degree of synergy increased with decreasing current intensity and increasing ultrasound power. After 30 days of treatment, the observed carbamazepine degradation percentage (i.e., 99.5%) was higher than that with electrolysis alone (i.e., 91%). In this setup, the maximum reported degradation rate constant corresponded to ca. 0.76 h^{-1} [68]. Ultrasound technology has also been investigated in combination with the Fenton process by Ghauch et al. (2011), where low frequency ultrasound irradiation was dedicated to cleaning the iron catalyst due to corrosion. Recycling experiments showed that reused iron catalysts could promote the satisfactory degradation of carbamazepine (i.e., up to 85% per cycle) after 4 successive cycles [106]. Similar to the conventional Fenton reaction, the degradation efficiency of the US/Fe(0)/H_2O_2 system is strongly dependent on the acidic nature of the solution [106]. Liu et al. (2016) demonstrated that electron beam radiolytic treatment combined with H_2O_2 addition could further promote carbamazepine degradation and mineralization since both oxidative and reductive species could be generated simultaneously. It was also reported that the addition of H_2O_2 contributed to the decomposition of some mutagenic and carcinogenic transformation products [31].

Regarding photoelectrocatalytic treatments, Daghrir et al. (2013) carried out a comparative study on carbamazepine degradation utilizing Ti/TiO_2 nanostructured electrodes in combination with cathodes of different natures. Optimal degradation was observed with a vitreous carbon cathode, reaching up to 73.5% degradation (i.e., 0.24 h^{-1} reaction rate constant) and 21.2% mineralization after 120 min [107]. Faster degradation kinetics were reported by Franz et al. (2020) when using TiO_2 meshes in photoelectrocatalysis (i.e., 4.56 h^{-1}). In fact, when combining photocatalysis with electrochemical treatment, an increase by a factor of 4.2 was observed with respect to heterogeneous photocatalysis on a supported TiO_2 mesh [80]. As previously discussed in section 2.3, conventional photocatalysis with commercial Degussa P25 TiO_2 powders was in this study more

effective (i.e., 10.44 h^{-1}) than the photoelectrocatalytic treatment, although powdered catalyst presents the drawback of a more complex recovery at the industrial scale [80].

Finally, a more extensive hybrid system was proposed by Monteagudo et al. (2015), which was dedicated to the simultaneous activation of PS by Fe^{2+} ions, thermal energy, ultrasound and UV irradiation, in addition to the catalytic activity of Fe^{2+} on H$_2$O$_2$ decomposition under acidic conditions. In contrast to the mineralization efficiencies observed in isolated or binary combinations of these methods (i.e., less than 26.4% TOC removal), the hybrid setup could reach 99% mineralization [108]. This study revealed that the presence of sulfate electrolytes inhibited the coalescence of cavitation bubbles and increased the concentration of SO$_4^{\bullet-}$ radicals. In addition, the iron catalyst could be regenerated through UV irradiation [108].

7   Opportunities and limitations

Conventional physical and biological treatments are not considered stand-alone solutions to degrade carbamazepine. The former is a separation process that requires further treatment, and the latter typically entails considerably slow kinetics (i.e., less than 0.004 h^{-1}) to eventually attain minor or negligible carbamazepine degradation efficiencies (Table 2). Nonetheless, both can help increase the efficiency of other treatments, such as AOPs. Physical treatments preconcentrate wastewater and hence reduce the reactor volume needed and its derived operational costs, while biological treatments enhance mineralization efficiencies by targeting biodegradable compounds in the effluent. Likewise, and despite the successful kinetics observed in the UV/sulfite system (i.e., between 5.2 and 20.4 h^{-1}), ARPs for carbamazepine degradation have not been extensively investigated, as shown by the limited number of available research articles (Table 2). Consequently, additional studies are needed to evaluate their strengths and pitfalls as well as their suitability for industrial applications.

As reflected in Table 1 and Figure 2, the possibilities for further implementation of AOPs are more extensive. Most treatments can be found within a window of 4 to 12 h^{-1} degradation rate constant (Fig. 3), meaning that 99% carbamazepine removal can be attained in a time span between 20 to 70 min. The largest body of literature is dedicated to UV-based treatments, which range from the low kinetics attained via photolysis alone (i.e., less than 0.03 h^{-1}) up to the notably higher degradation rates observed in UV/chlorine systems (i.e., up to 85.2 h^{-1}). However, before taking the leap towards the industrial implementation of UV-based treatments, not only the degradation effectiveness of a single study is the key parameter to take into consideration. In the case of UV/chlorine treatment, it is essential to optimize chlorine dosages and mitigate scavenging effects in complex wastewater matrices, such as those caused by DOM and HCO$_3^-$ ions. In addition, it is imperative to quantify the risks of increased effluent toxicity since the presence of Cl$^-$ ions in wastewater can further lead to the production of species such as chlorate (ClO$_3^-$) and perchlorate (ClO$_4^-$). Other combinations of UV and chemical precursors, such as PS, PMS and the Fenton reaction, have reported either similar or lower rates in comparison to their chemical-based counterparts. This evidence indicates that the development of hybrid treatments should also be closely evaluated. It is commonly assumed that synergistic effects are intrinsic to hybrid setups, although as seen in the case of carbamazepine degradation (Fig. 2), considerably slower degradation kinetics can also be triggered.

13
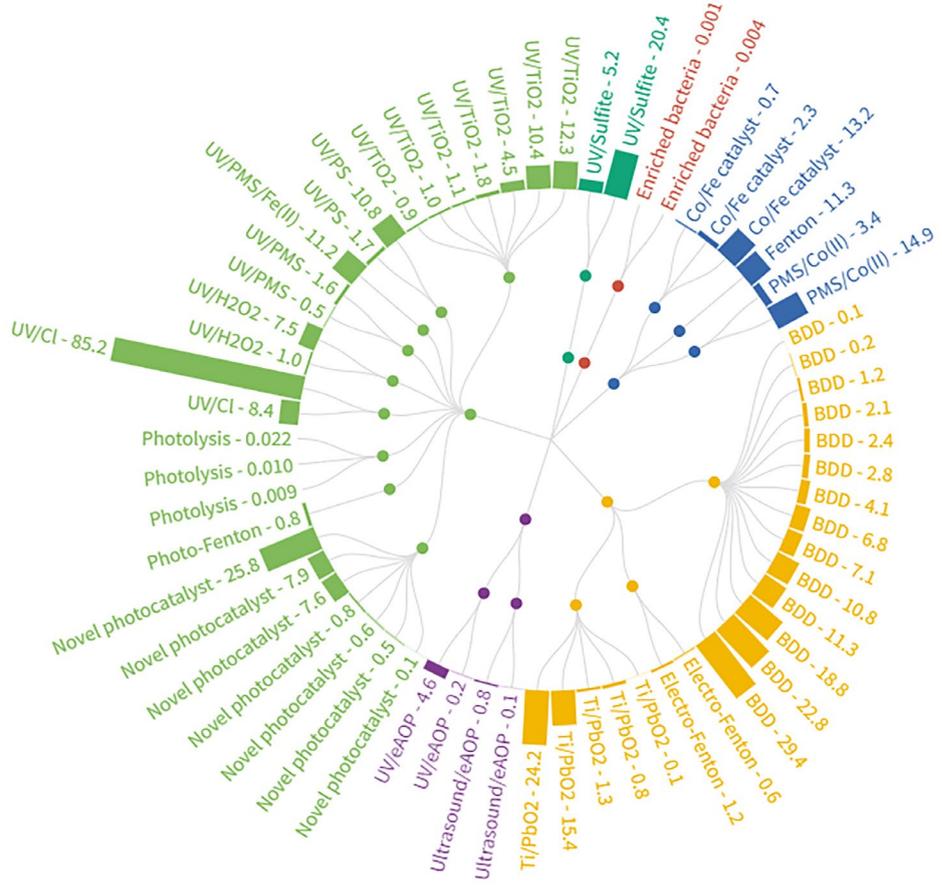
Figure 2: Carbamazepine degradation rates for several wastewater treatments. The height of the bars corresponds to the value of the pseudo-first order kinetic constant (h^{-1}) reported in a specific literature reference.
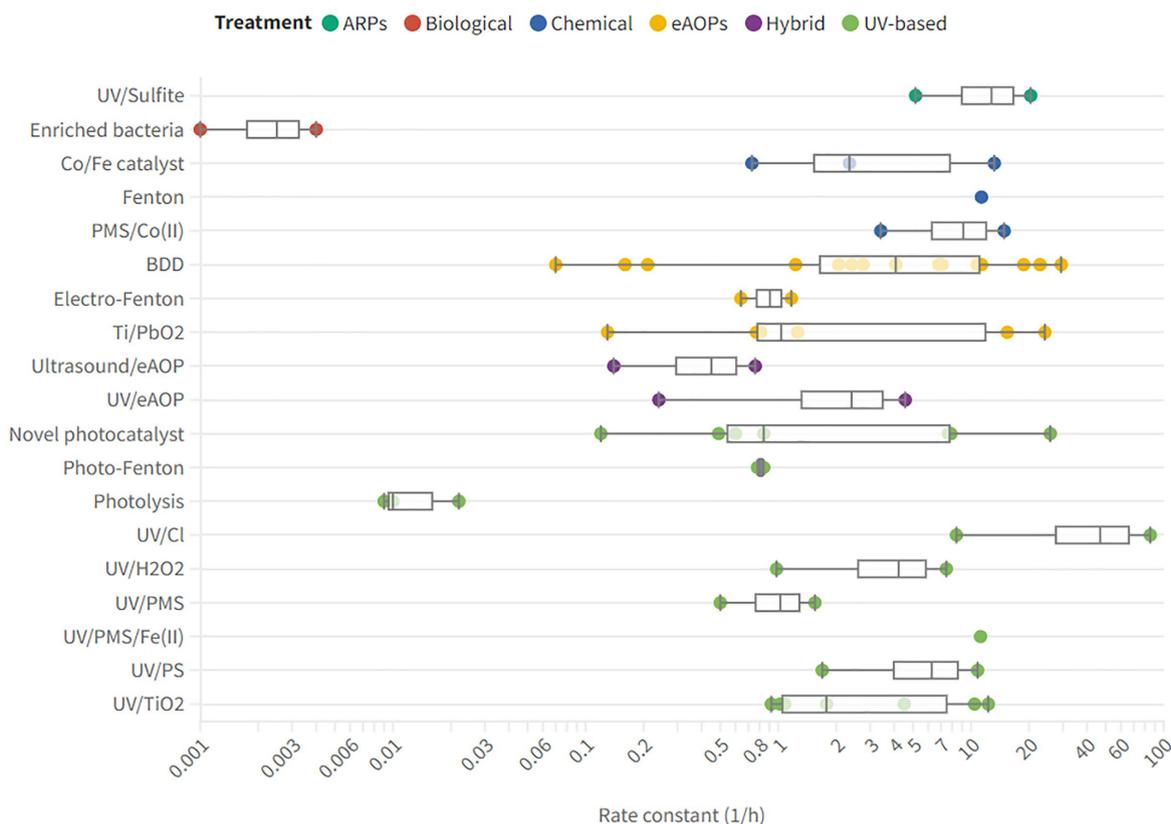
Figure 3: Average carbamazepine degradation rates (h^{-1}) for several wastewater treatments.

Chemically activated AOPs, such as ozonation, Fenton process and sulfate radical precursor-based oxidation, have been reported to be effective for carbamazepine degradation (Table 1), although they also pose several advantages and limitations for future industrial applications. Ozonation presents the advantages of operating under mild conditions while leading to high removal percentages (i.e., close to 100%). However, the use of ozone is costly, yields low mass transfer efficiencies and utilization rates during the AOP treatment, and is strongly influenced by suspended solids and organic matter. The formation of bromate through ozonation also adds as a constraint that the use of wastewater containing Br^- ions should be avoided or pretreated. Fenton treatments are easy to operate and provide high degradation kinetics (i.e., 11.3 h^{-1}); although a low pH is needed, iron catalyst consumption and the co-production of large amounts of iron sludge hinder its application in continuous industrial setups. Sulfate radical-based AOPs that rely on PMS and PS present high degradation efficiencies (i.e., up to 14.9 h^{-1}) and can be applied to a wide range of wastewater since the advantages of both $^\bullet OH$ and $SO_4^{\bullet-}$ radicals can be exploited depending on the application at hand. The drawbacks of these treatments are not only the need for considerable amounts of chemical precursors but also the associated potential increase in sulfate residuals and acidity of the treated effluents. To overcome these limitations, subsequent posttreatment should be implemented, which also increases the overall operational costs. These chemical-based AOPs also have in common that they typically rely on the catalytic generation of oxidative radicals, where the implications of applying homogeneous or heterogeneous catalysts are quite diverse. Homogeneous catalysts are burdensome to recover, and hence it is complicated to prevent the leaching of metals and catalyst components into effluents. On the other hand, heterogeneous catalysts mitigate these leaching effects, but catalyst reuse remains a challenging matter. As a result, heterogeneous catalysts are preferred and typically implemented in the form of magnetic nanoparticles, although it

15

579   is necessary to further improve their reusability while decreasing their production costs.
580

581   Based on the reviewed literature (Table 1), electrochemical and photocatalytic AOPs have
582 consistently reported effective carbamazepine degradation results, more specifically,
583 electrochemical treatments driven by the use of boron-doped diamond (BDD) or Ti/PbO$_2$ anodes and
584 photocatalytic treatments involving TiO$_2$ or novel photocatalysts. The maximum degradation
585 efficiencies attained by BDD electrodes, Ti/PbO$_2$ electrodes and novel photocatalysts oscillate
586 between 24.2 and 29.4 h^{-1} under optimal conditions, with the TiO$_2$ photocatalysts being slightly less
587 efficient (i.e., up to 12.3 h^{-1}). Nonetheless, degradation rate constants as low as 0.1 h^{-1} have also
588 been observed for these treatments when the presence of radical scavengers has played a major
589 hindering role. The added value of these technologies is that effective carbamazepine degradation
590 can be attained under mild operating conditions and with minimal or no addition of chemicals to
591 generate oxidative radicals. In addition, the use of renewable energies contributes to a reduction in
592 operational costs and environmental impacts. On the other hand, both electrochemical and
593 photocatalytic treatments showcase limited energy use efficiencies, either in the form of electricity
594 or light, respectively, which need to be further optimized. In light of a possible future
595 implementation, other main limitations to be overcome for these AOPs are the high costs that
596 advanced electrodes and photocatalysts entail, the need for reducing their performance variability
597 due to wastewater composition effects and the potential formation of toxic byproducts.

Table 1: Summary of reported carbamazepine (CBZ) degradation in AOPs for wastewater treatment.

Treatment		Matrix	Initial CBZ (mg L^{-1})	Removal	Rate constant	Comments	Ref.
Chemical oxidation	Ozonation	Synthetic and Spiked WWTP effluent	1	60 – 99%	-	• O$_3$ flow: 2.6 g h^{-1}	[51]
		Spiked river effluent	5·10^{-4}	100%	-	• pH: 7.3 • [O$_3$]: 0.006 mM	[50]
		WWTP effluent	-	97 – 100%	-	• [O$_3$]: 0.61 ± 0.04 g/g DOC	[3]
	Fenton	Synthetic and WWTP effluent	11.81	7 – 83%	11.34 h^{-1}	• pH: 3 • [H$_2$O$_2$]: 0.15 – 0.25 mM • [Fe(II)]: 0.15 – 0.25 mM	[54]
		WWTP effluent	1·10^{-3}	100%	-	• pH: 3 • [H$_2$O$_2$]: 0.895 mM • [Fe(II)]: 0.358 mM	[55]
		WWTP effluent	0.1	6.6 – 18%	-	• pH: 3 • [H$_2$O$_2$]: 0.147 – 1.470 mM • [Fe]: 0.09 mM	[56]
	Fenton/PS	Synthetic	50	77%	-	• pH: 6.8 • [H$_2$O$_2$]: 0.5 mM • [PS]: 1 mM • [nZVI]: 2 mM	[57]
	PMS/Co(II)	Synthetic and WWTP effluent	11.81	50 – 100%	3.40 – 14.87 h^{-1}	• pH: 3 • [PMS]: 0.15 – 0.25 mM • [Co(II)]: 0.15 – 0.25 mM	[54]
	PS/Fe(II)	Synthetic	5.91	3 – 99%	-	• pH: 2 – 7.87 • [Fe(II)]: 0.05 – 0.75 mM • [PS]: 0.125 – 1.5 mM • [Cl$^-$]: 0 – 10 mM	[59]
	Other catalysts	Synthetic	5	93%	2.34 h^{-1}	• pH: 6.8 • Organo-montmorillonite supported nCoFe$_2$O$_4$ catalyst compared with other supports • [nCoFe$_2$O$_4$/OMt]: 0.4 g L^{-1} • [PMS]: 0.5 mM	[60]

		Matrix	Concentration	Removal	Rate constant	Conditions	Ref.
		Synthetic	5 – 20	29 – 100%	0.73 – 13.22 h^{-1}	• pH: 4 – 10 • Layered double oxide-supported Co-Fe bimetal catalysts • [Co$_2$FeAl-LDO]: 20 – 80 mg L^{-1} • [PMS]: 0.05 – 0.4 mM	[61]
Electrochemical oxidation	BDD anodes	Synthetic	0.47	100%	29.40 ± 2.10 h^{-1}	• pH: 2 • Current density: 200 A m^{-2} • Electrolyte: Na$_2$SO$_4$ (40 mM)	[64]
		Synthetic	0.47	100%	2.40 ± 0.37 h^{-1}	• pH: 2 • Current density: 200 A m^{-2} • Electrolyte: NaNO$_3$ (60 mM)	[64]
		Synthetic	12	8.6 – 36%	-	• pH: 3 • Current density: 18 A m^{-2} • Electrolyte: Na$_2$SO$_4$ (7 mM)	[62]
		Synthetic	10	82%	0.07 – 1.23 h^{-1}	• pH: 4 • Current density: 403.23 A m^{-2} • Electrolyte: Na$_2$SO$_4$ (7 mM) • With and without O$_2$ addition	[4, 66]
		Synthetic	10	-	4.07 – 7.08 h^{-1}	• pH: 4 • Current density: 403.23 A m^{-2} • Electrolyte: NaCl (7 mM) • With and without O$_2$ addition	[4, 66]
		Synthetic	10	-	10.78 – 22.81 h^{-1}	• pH: 4 • Current density: 403.23 A m^{-2} • Electrolyte: NaBr (7 mM) • With and without O$_2$ addition	[4, 66]
		Synthetic	1.18	-	0.16 – 6.81 h^{-1}	• Current density: 0 – 100 A m^{-2} • Electrolyte: NaClO$_4$ (50 mM) • [PS]: 0 – 5 mM	[65]
		Synthetic	1.18	-	0.21 – 18.77 h^{-1}	• Current density: 0 – 100 A m^{-2} • Electrolyte: NaClO$_4$ (50 mM) • [PMS]: 0 – 5 mM	[65]
		Spiked IW effluent	10	100%	2.06 – 2.75 h^{-1}	• Current density: 403.23 A m^{-2} • No electrolyte addition • With and without O$_2$ addition	[66]
		Spiked WWTP effluent	10	88.7%	11.34 h^{-1}	• pH: 2 • Current density: 161.29 A m^{-2} • Electrolyte: NaCl (14 mM)	[27]

Ti-based anodes	Synthetic	2 – 30	53 – 80.3%	0.47 – 1.57 $M^{-1}s^{-1}$	• Ti/Ta_2O_5-SnO_2 and Ti/PbO_2 anodes • pH: 2 – 10 • Current density: 10 – 170 A m^{-2} • Electrolyte: Na_2SO_4 (100 mM)	[69]
	Synthetic	10	45.4 – 83.9%	0.77 h^{-1}	• Ti/PbO_2 cylindrical mesh anode • pH: 7 • Current density: 24.1 – 241.5 A m^{-2} • Electrolyte: Na_2SO_4 (7 mM) • Pure O_2 addition at 0 – 3 L min^{-1}	[67]
	Synthetic	10	-	0.13 – 0.81 h^{-1}	• Ti/PbO_2 anode • pH: 6.9 – 7.4 • Current density: 909 – 13,636 A m^{-2} • Electrolyte: Na_2SO_4 (10 mM)	[68]
	Synthetic	10	53.5 – 89.9%	1.26 h^{-1}	• Ti/PbO_2 and Ti/BDD anodes • pH: 7 • Current intensity: 1 – 2 A • Electrolyte: Na_2SO_4 (2.82 mM) • Energy: 11.300 – 80.379 kWh m^{-3}	[63]
	Synthetic	10	99%	15.42 – 24.18 h^{-1}	• Blue-colored TiO_2 nanotube arrays membrane filter anode • Current density: 10 – 100 A m^{-2} • Electrolyte: Na_2SO_4 (10 mM) • Energy: 0.0246 – 0.6489 kWh m^{-3}	[70]
	Synthetic	5	98%	-	• Ti/Pt anode • pH: 5.5 • Current density: 140 A m^{-2} • Electrolyte: Na_2SO_4 (25 mM)	[4, 71]
	Spiked MBR effluent	0.01	100%	-	• Ti/Ta_2O_5-SnO_2 anode • pH: 6 • Current density: 90 A m^{-2} • Electrolyte: Na_2SO_4 (0 – 100 mM)	[69]
Electro-Fenton	Synthetic	12 ± 2	29 – 73%	0.64 – 1.17 h^{-1}	• Nb/BDD, Ti/Pt and $TiSn_2$ anodes • pH: 3 • Current density: 18 – 177 A m^{-2} • [Fe^+]: 0 – 0.75 mM • Electrolyte: Na_2SO_4 (7 mM)	[62]

	Electro-peroxidation	Synthetic	12	30 – 50%	-	• Nb/BDD and Ti/Pt anodes • pH: 3 • Current density: 18 A m^{-2} • Electrolyte: Na$_2$SO$_4$ (7 mM)	[62]
Hybrid processes	Activated sludge/Gamma irradiation	WWTP effluent	17	91.8 – 99.8%	-	• 300 – 800 Gy irradiation	[88]
	Sonoelectro-oxidation	Synthetic	10	24.9 – 81.3%	0.14 – 0.76 h^{-1}	• Ti/PbO$_2$ anode • pH: 6.9 – 7.4 • Current density: 909 – 1,3636 A m^{-2} • US Power: 20 – 40 W • Electrolyte: Na$_2$SO$_4$ (10 mM)	[68]
	Sono-Fenton	Synthetic	9.92	12 – 95%	-	• pH: 3 – 9 • US frequency: 40 kHz • [Fe(0)]: 0.44 – 3.57 mM • [H$_2$O$_2$]: 12.5 – 100 mM	[106]
	UV/PS/Fe(II)/US/H$_2$O$_2$	Synthetic	15	87.7 – 99%	-	• pH: 2.8 • UV irradiation: 7.81·10^{-6} E s^{-1} • US amplitude: 100% • [PS]: 300 – 900 mg L^{-1} • [Fe(II)]: 5 – 15 mg L^{-1} • [H$_2$O$_2$]: 100 mg L^{-1}	[108]
	Radiolysis/H$_2$O$_2$	Synthetic	75	100%	0.77 – 2.63 kGy^{-1}	• 1.8 MeV and 0 – 10 mA electron beam • Irradiation doses: 0.5 – 20 kGy • [H$_2$O$_2$]: 0 – 200 mM	[31]
	Photoelectro-catalysis	Synthetic	10 – 20	21.6 – 73.5%	0.24 h^{-1} 2.36 M^{-1} s^{-1}	• Ti/TiO$_2$ anode, and SS, Gr, VC, a-C1 and a-C2 cathodes • pH: 5.5 – 7.3 • Current density: 2.7 – 33.6 A m^{-2} • UV irradiation: 69 W m^{-2} • Electrolyte: Na$_2$SO$_4$ (50 mM)	[107]
		Synthetic	0.1	99%	4.56 ± 0.12 h^{-1}	• TiO$_2$ mesh anode • Potential: 122.1 V m^{-2} • UV irradiation: 916 W m^{-2} • Energy: 7.58 kWh m^{-3}	[80]
Photodegradation	Photolysis	Synthetic	0.15	50%	0.009 ± 0.0007 h^{-1}	• Natural sunlight • pH: 8.0 ± 0.1 • Solar irradiance: 7.7 – 9.2 kW m^{-2}	[72]

	Water	Value	%	Rate	Conditions	Ref.
	Synthetic	9.5	15 – 100%	0.01 – 1.30 h⁻¹	• Simulated sunlight • pH: 2.9 – 9.0 • Not sparged, sparged with O_2 and sparged with N_2 • Solar irradiance: 55 W m⁻²	[73]
	WWTP effluent	11.81	8%	-	• Simulated sunlight • pH: 7.8 • Photonic flux: 7.9·10⁶ E min⁻¹	[74]
	WWTP effluent	0.15	50%	0.022 ± 0.014 h⁻¹	• Natural sunlight • pH: 7.85 ± 0.21 • Solar irradiance: 7.7 – 9.2 kW m⁻²	[72]
UV/Chlorine	Synthetic	2	98%	8.4 – 85.2 h⁻¹	• pH: 4.9 – 10 • UV irradiation: 2.1 – 14.8 W m⁻² • [NaOCl]: 0.03 – 0.63 mM	[78]
	Simulated WW	1·10⁻³	50 – 90%	-	• pH: 7 • UV: 40 – 200 W • [HOCl]: 0.008 – 0.116 mM	[79]
	Simulated WW	1·10⁻³	17 – 20%	-	• pH: 7 • UV: 40 – 200 W • [ClO_2]: 0.003 – 0.092 mM	[79]
UV/H_2O_2	Synthetic	2.5 – 10	63 – 100%	0.98 – 7.45 h⁻¹	• pH: 3 – 11 • UV irradiation: 1.53 W m⁻² • [H_2O_2]: 0.5 – 5 mM	[44, 75]
	Synthetic and Spiked WWTP effluent	1	73 – 96%	-	• [H_2O_2]: 0.5 mL L⁻¹	[51]
	WWTP effluent	3.33·10⁻⁴	82 – 99%	-	• UV irradiation: 70 W m⁻² • [H_2O_2]: 0.59 – 1.47 mM	[3]
	WWTP effluent	0.043 ± 0.037	50 – 83%	-	• Continuous operation • UV contact time: 9 – 28 s • pH: 7.17 ± 0.08 • UV irradiation: 95.5 W m⁻² • [H_2O_2]: 0.5 – 5 mM	[76]
UV/O_3	Synthetic and Spiked WWTP effluent	1	81 – 99%	-	• O_3 flow: 2.6 g h⁻¹ • UV lamp: 15 W	[51]

	Spiked WWTP effluent	$1 \cdot 10^{-4}$	100%	-	• Steady state time: 180 min • Hydraulic retention time: 41 min • pH: 8.5 • O_3: 50 g Nm^{-3} (gas phase) • UV: 10 W (UVA-LEDs)	[77]
UV/PMS	Synthetic	2.5 – 10	39 – 80%	0.50 – 1.55 h^{-1}	• pH: 3 – 11 • UV irradiation: 1.53 W m^{-2} • [PMS]: 0.5 – 5 mM	[44, 75]
	WWTP effluent	0.043 ± 0.037	73 – 100%	-	• Continuous operation • UV contact time: 9 – 28 s • pH: 7.17 ± 0.08 • UV irradiation: 95.5 W m^{-2} • [PMS]: 0.5 – 5 mM	[76]
UV/PMS/Fe(II)	WWTP effluent	11.81	100%	11.21 ± 0.02 h^{-1}	• pH: 3 • Photonic flux: $7.9 \cdot 10^6$ E min^{-1} • [PMS]: 0.2 mM • [Fe(II)]: 0.1 mM	[74]
	WWTP effluent	0.043 ± 0.037	88 – 100%	-	• Continuous operation • UV contact time: 9 – 28 s • pH: 7.17 ± 0.08 • UV irradiation: 95.5 W m^{-2} • [PMS]: 0.5 – 5 mM • [Fe(II)]: 0.5 – 5 mM	[76]
UV/PS	Synthetic	2.5 – 10	80 – 99%	1.69 – 10.83 h^{-1}	• pH: 3 – 11 • UV irradiation: 1.53 W m^{-2} • [PS]: 0.5 – 5 mM	[44, 75]
	WWTP effluent	0.043 ± 0.037	47 – 79%	-	• Continuous operation • UV contact time: 9 – 28 s • pH: 7.17 ± 0.08 • UV irradiation: 95.5 W m^{-2} • [PS]: 0.5 – 5 mM	[76]
UV/PS/Fe(II)	WWTP effluent	0.043 ± 0.037	96 – 100%	-	• Continuous operation • UV contact time: 9 – 28 s • pH: 7.17 ± 0.08 • UV irradiation: 95.5 W m^{-2} • [PS]: 0.5 – 5 mM • [Fe(II)]: 0.5 – 5 mM	[76]

	Matrix	Concentration	Removal	Kinetic constant	Remarks	Ref.
UV/TiO$_2$	Synthetic	0.1	100%	10.44 h^{-1}	• Degussa P25 powders • Half-life time: 4.5 min • UV irradiation: 916 W m^{-2} • Energy: 3.30 kWh m^{-3}	[80]
	Synthetic	0.1	65%	1.08 ± 0.42 h^{-1}	• TiO$_2$ mesh • Half-life time: 36 min • UV irradiation: 916 W m^{-2} • Energy: 31.98 kWh m^{-3}	[80]
	Synthetic	1	60%	12.30 ± 0.66 h^{-1}	• Degussa P25 • pH: 5 • UV irradiance: 710 W m^{-2} • [P25]: 0.5 g L^{-1}	[83]
	Spiked WWTP effluent	0.1	50 − 80%	-	• TiO$_2$ immobilized on glass spheres • Natural sunlight	[3]
	WWTP effluent	11.81	-	1.78 ± 0.01 h^{-1}	• pH: 7.8 • Photonic flux: 7.9·10^6 E min^{-1} • [TiO$_2$]: 0.5 g L^{-1}	[74]
	WWTP effluent	2.95·10^{-4}	46 − 100%	0.92 h^{-1}	• TiO$_2$ nanoparticles • UV irradiance: 69 W m^{-2}	[81]
	WWTP effluent	5 − 20	70 − 99%	1.02 − 4.50 h^{-1}	• Commercial TiO$_2$ • pH: 3.4 − 10 • UV irradiance: 15.08 ± 0.41 W m^{-2} • [TiO$_2$]: 0.5 − 2 g L^{-1}	[82]
Photo-Fenton	WWTP effluent	3.33·10^{-4}	66 − 94%	-	• pH: 6 − 7.5 • UV irradiation: 70 W m^{-2} • [H$_2$O$_2$]: 0.59 − 1.47 mM • [Fe]: 0.036 − 0.072 mM	[3]
	WWTP effluent	0.1	100%	0.78 − 0.84 h^{-1}	• pH: 3 • UV irradiation: 30 W m^{-2} • [H$_2$O$_2$]: 0.147 − 1.470 mM • [Fe]: 0.09 mM	[56]
Other photocatalytic systems	Synthetic	1	99%	0.60 − 25.80 h^{-1}	• BiOCl$_{0.875}$Br$_{0.125}$ • pH: 4 − 9 • UV irradiance: 710 W m^{-2} • [BiOCl$_{0.875}$Br$_{0.125}$]: 0.5 g L^{-1}	[83]

Synthetic	5.91	80 – 100%	0.49 – 7.64 h^{-1}	• Triclinic-WO$_3$ • pH: 4.6 – 6 • UV lamp: 420 nm • UV intensity: $1.5 \cdot 10^{-6}$ E L^{-1} s^{-1} • [WO$_3$]: 0.3 g L^{-1} • [PS]: 2 mM • [CH$_3$OH]: 0 – 100 mM	[85]
Synthetic and Simulated WW	5	20 – 100%	0.12 – 7.86 h^{-1}	• BiPO$_4$ (hexagonal and monoclinic) • UV lamp: 100 W • [BiPO$_4$]: 1 g L^{-1}	[84]
WWTP effluent	$2.95 \cdot 10^{-4}$	41 – 92%	0.84 h^{-1}	• ZnO nanoparticles • UV irradiance: 69 W m^{-2}	[81]

609 **Table 2:** Summary of reported carbamazepine (CBZ) degradation in other methods for wastewater treatment.

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Treatment		Matrix	Initial CBZ (mg L^{-1})	Removal	Rate constant	Comments	Ref.
Advanced reduction processes	UV/Sulfite	Synthetic	5	5 – 99.6%	5.16 – 20.40 h^{-1}	• pH: 3 – 11 • UV irradiation: 9 – 32 W m^{-2} • [Sulfite]: 0 – 1.6 mM	[40]
Biological treatment	Laccase	Synthetic	4.73	22 – 60%	-	• pH: 4.5 • [Laccase]: 10 nkat mL^{-1} • [1-hydroxybenzotriazole]: 0 – 0.2 mM • Culture: *T. versicolor*	[89]
	Lignin peroxidase	Synthetic	5	7 – 15%	-	• pH: 2.5 – 6 • [H$_2$O$_2$]: 0.7 mM • Culture: *P. chrysosporium*	[90]
	Manganese peroxidase	Synthetic	4.73	14%	-	• pH: 4.5 • [MnP]: 10 nkat mL^{-1} • [MnSO$_4$]: 0.1 mM • [Glucose]: 25 mM • [Glucose oxidase]: 3.33 nkat mL^{-1} • Culture: *P. chrysosporium*	[89]
	Multiple enzymes	Synthetic	10	47 – 58%	-	• Cultures: *T. versicolor, G. lucidum, I. lacteus, P. chrysosporium*	[93]
		Synthetic	1	99%	-	• Cultures: *Bjerkandera sp. R1, B. adusta, P. chrysosporium*	[91]
		Synthetic	1·10^{-3}	48 – 99%	-	• Cultures: *P. ostreatus Florida N001, P. ostreatus PC9, P. ostreatus Florida F6.*	[87]
	Enriched bacteria	WWTP effluent	10 – 40	10 – 46%	0.001 – 0.004 h^{-1}	• Enriched sulfate-reducing and fermentative acidogenic bacteria compared to mixed microbial communities • Influence of COD:SO$_{42-}$ and CBZ dose	[92]
Physical treatment	PAC	WWTP effluent	2.21 – 4.62·10^{-4}	90 – 92%	-	• [PAC]: 10 – 20 mg m^{-1} • 20 – 60 min contact time	[3]
	GAC	MWWTP effluent	1.10·10^{-4}	72%	-	• 23,400 bed volumes treated • 14 min empty bed contact time	[3]

MOFs	Synthetic	10	87 – 100%	Q_m: 663.70 mg g^{-1}	• Comparison between MOF-5-derived nanoporous carbon, ZIF-8-derived nanoporous carbon and commercial PAC	[99]
	Synthetic	$1 \cdot 10^{-3}$	-	K_F: 57.56 – 73.79 (ng/mg)(L/ng)$^{1/n}$	• Two GAC adsorbents • Carbon dosage: 10 – 800 ng L^{-1} • Adsorption equilibria after 12 days • (1/n) = 0.42 – 0.43	[97]
NF	Groundwater	20	97 – 100%	-	• pH: 5.6 – 6.1 • Feed flow rate: 360 m^3 h^{-1} • Permeate flow rate: 234 m^3 h^{-1} • Filtration in two stages, comprising 31 and 15 membrane modules with 6 NF membranes each	[105]
RO	Groundwater	20	99 – 100%	-	• pH: 5.6 – 6.1 • Feed flow rate: 486 m^3 h^{-1} • Permeate flow rate: 356.4 m^3 h^{-1} • Filtration in two stages, comprising 40 and 20 membrane modules with 6 RO membranes each	[105]

8 Conclusions

There is an urgent need to tackle the ubiquitous presence of recalcitrant contaminants in natural water bodies, which pose a potential threat to aquatic ecosystems and human health. As discussed in this review, advanced oxidation processes (AOPs) have been effective in the degradation of pharmaceutically active compounds such as carbamazepine, as opposed to biological and physical treatments. In particular, high degradation efficiencies have been attained through electrochemical and photocatalytic AOPs under multiple experimental conditions. However, future research should be dedicated to enhancing the adaptability and practicability of AOPs, such as by reducing chemical, energy and catalyst needs. In addition, their resilience against radical scavengers and wastewater components needs to be improved to promote their implementation at a larger scale. Finally, along with reporting degradation efficiencies of the targeted PhACs, studies should focus on elucidating the underlying degradation mechanisms to ensure that toxic byproducts are not discharged.

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Competing interests

The authors declare no competing interests.

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