

Review



# Key Points of Advanced Oxidation Processes (AOPs) for Wastewater, Organic Pollutants and Pharmaceutical Waste Treatment: A Mini Review

Pavlos K. Pandis <sup>1</sup><sup>(D)</sup>, Charalampia Kalogirou <sup>1,2</sup>, Eirini Kanellou <sup>1,3</sup>, Christos Vaitsis <sup>1</sup>, Maria G. Savvidou <sup>1</sup><sup>(D)</sup>, Georgia Sourkouni <sup>2</sup><sup>(D)</sup>, Antonis A. Zorpas <sup>3,\*</sup><sup>(D)</sup> and Christos Argirusis <sup>1,\*</sup>

- <sup>1</sup> Laboratory of Inorganic Materials Technology, School of Chemical Engineering, National Technical University of Athens, Greece, 9 Iroon Polytechniou Str., Zografou Campus, 15780 Athens, Greece; ppandis@chemeng.ntua.gr (P.K.P.); charalampia-stamatia.kalogirou@tu-clausthal.de (C.K.); eirini.kanellou@st.ouc.ac.cy (E.K.); vaitsis@central.ntua.gr (C.V.); msavvid@central.ntua.gr (M.G.S.)
- <sup>2</sup> Clausthal Centre for Materials Technology (CZM), Clausthal University of Technology, Leibnizstr. 9, 38678 Clausthal-Zellerfeld, Germany; cogsa@tu-clausthal.de
- <sup>3</sup> Laboratory of Chemical Engineering and Engineering Sustainability, Faculty of Pure and Applied Sciences, Open University of Cyprus, Giannou Kranitiodi 33, Latsia, 2220 Nicosia, Cyprus
- Correspondence: antonis.zorpas@ouc.ac.cy or antoniszorpas@yaoo.com (A.A.Z.); amca@chemeng.ntua.gr (C.A.)

**Abstract:** Advanced oxidation procedures (AOPs) refer to a variety of technical procedures that produce OH radicals to sufficiently oxidize wastewater, organic pollutant streams, and toxic effluents from industrial, hospital, pharmaceutical and municipal wastes. Through the implementation of such procedures, the (post) treatment of such waste effluents leads to products that are more susceptible to bioremediation, are less toxic and possess less pollutant load. The basic mechanism produces free OH radicals and other reactive species such as superoxide anions, hydrogen peroxide, etc. A basic classification of AOPs is presented in this short review, analyzing the processes of  $UV/H_2O_2$ , Fenton and photo-Fenton, ozone-based (O<sub>3</sub>) processes, photocatalysis and sonolysis from chemical and equipment points of view to clarify the nature of the reactive species in each AOP and their advantages. Finally, combined AOP implementations are favored through the literature as an efficient solution in addressing the issue of global environmental waste management.

Keywords: advanced oxidation process; wastewater; green chemistry; waste management; sonochemistry

## 1. Introduction

Advanced oxidation processes (AOPs) comprise environmentally friendly and innovative techniques in order to treat wastewater and enrich anti-pollutant technology procedures [1–3]. The environmental impacts of the above processes are considered to be of high importance due to the high reaction rates and efficiencies, as well as the lack of highly dangerous byproducts promoting green chemistry aspects [2–8]. Common to all advanced oxidation processes is the formation of hydroxyl radicals (•OH), which are capable of acting as the main group of chemicals towards the decontamination and/or biodegradation of highly toxic and heavy pollutants in many fields of the global circular economy [6,9-16]. These processes enhance the oxidation kinetics for the mineralization of contaminants to carbon dioxide or to other harmless or beneficial byproducts, especially in water purification systems [17,18]. Classifications are reported in the literature considering the source of the production of hydroxyl radicals. Amongst numerous oxidizing chemicals described in the literature, the dominant classification is mainly categorizing AOPs [1,4,5,7,12,19–24] into (a) UV–hydrogen peroxide processes, (b) Fenton and photo-Fenton, (c) ozone-based processes, (d) photocatalysis and (e) sonolysis. Although reviews on the use of AOPs for purification systems have been reported in the literature [2,7,25], the



Citation: Pandis, P.K.; Kalogirou, C.; Kanellou, E.; Vaitsis, C.; Savvidou, M.G.; Sourkouni, G.; Zorpas, A.A.; Argirusis, C. Key Points of Advanced Oxidation Processes (AOPs) for Wastewater, Organic Pollutants and Pharmaceutical Waste Treatment: A Mini Review. *ChemEngineering* **2022**, *6*, 8. https://doi.org/10.3390/ chemengineering6010008

Academic Editor: Josep Font

Received: 30 November 2021 Accepted: 5 January 2022 Published: 18 January 2022

**Publisher's Note:** MDPI stays neutral with regard to jurisdictional claims in published maps and institutional affiliations.



**Copyright:** © 2022 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https:// creativecommons.org/licenses/by/ 4.0/). scope of this review is to coherently present and summarize the key points in each process, to classify the aspects of the chemical reactions and fully comprehend the role of each AOP. Schematic illustrations and classifications are presented in order to properly comprehend the basic concepts and uses of each AOP. This review offers the ability to easily encourage the proper choice of AOP for specific wastewater. Finally, the benefits of the combined use of AOP techniques are reported and typical reactor paradigms are referred to in each AOP presented from an engineering point of view.

#### 2. UV/H<sub>2</sub>O<sub>2</sub> (Ultraviolet/Hydrogen Peroxide Processes)

Generally, the presence of UV light radiation, if properly combined with hydrogen peroxide, may effectively produce OH radicals; simultaneously, UV light acts as a natural disinfectant agent for many organic pollutants and wastewater systems. The applications of such systems have been reported in many fields, varying from the pharmaceutical sector [26–28] to the textile industry [9,29], organic pollutant removal [3,10,23,30–33], etc. [34–36]. The chemistry of this procedure has been investigated by modeling the mechanisms and processes [29,37], concluding that the substitution of expensive H<sub>2</sub>O<sub>2</sub> with other chemicals, such as Cl<sub>2</sub>, may be a useful alternative and excellent candidate at low pH values and low UV doses [38]. The main chemical reaction occurring in such systems is described below [2,29,39]:

$$H_2O_2 + hv \rightarrow 2^{\bullet}OH \tag{1}$$

Through the effect of radiation, the hydrogen peroxide molecules form OH radicals. This decomposition is attributed to a Haber–Weiss mechanism, which is initiated by cleavage of the O–O bond. Further chemical OH production is initiated by the following reactions [40]:

$$H_2O_2 + {}^{\bullet}OH \rightarrow H_2O + HO_2^{\bullet}$$
<sup>(2)</sup>

$$H_2O_2 + HO_2^{\bullet} \rightarrow H_2O + O_2 + {}^{\bullet}OH$$
(3)

However, in some cases, radical recombination occurs, as follows [41]:

$$^{\bullet}OH + HO^{\bullet} \rightarrow H_2O_2$$
 (4)

$$^{\bullet}OH + HO_2^{\bullet} \rightarrow H_2O + O_2 \tag{5}$$

$$\mathrm{HO}_{2}^{\bullet} + \mathrm{HO}_{2}^{\bullet} \rightarrow \mathrm{H}_{2}\mathrm{O}_{2} + \mathrm{O}_{2} \tag{6}$$

Many factors affect the efficiency of the above reactions, such as the concentration of hydrogen peroxide, the source of UV radiation and the physicochemical characteristics of fluid involved in the oxidation process. In Figure 1, the effect of the  $H_2O_2$  concentration is indicative of the degradation of carbamazepine.

Equations (1)–(6) are used by different groups to describe the synergistic utilization of UV radiation and H<sub>2</sub>O<sub>2</sub>. For example, Verma et al. [28] described the combination of UV-C radiation and H<sub>2</sub>O<sub>2</sub> for the successful degradation of the toxic anatoxin-a, suggesting the alternative effect of the harsher UV-C radiation instead of UV-A. Anatoxin-A is extremely toxic; therefore, the experiments were conducted in Lamil Plus Class II microbiological safety cabinets (Karstulan Metalli Tek-Fin Oy, Finland). The UV radiation was produced by DUV LEDs (TO39FW), developed by Sensor Electronic Technology, Columbia. Bagheri et al. [32] tried to overcome the excess recombination using only vacuum-UV processes, emphasizing the effect of fluid concentrations. A prototype annular photoreactor made of Plexiglas was photo-assisted by mercury 42 W amalgam lamps; all these experiments were conducted under continuous flow. These parameters were also investigated by Crapulli et al. [37], for mechanistic modelling, combining different concentrations and UV radiation wavelengths in order to estimate the higher penetration depths of hydroxyl radicals (•OH). Their concentration profiles were gathered under air- and oxygen-saturated conditions corresponding to Re = 1587 over a period of 60 min in a vacuum-UV reactor with cooling capabilities maintaining constant power consumption. Zoschke et al. [39]

concluded that UV radiation with an alternative medium, such as O<sub>3</sub>, is more effective than H<sub>2</sub>O<sub>2</sub>. All experiments were performed in custom-made laboratory-scale reactors, under a UV lamp with nominal power ranging from 11 to 60 W and volume reactor capacities from 0.1 to 3.1 L. Their reactor setup included a glass tank, a pump and a stainless-steal irradiation reactor tube holding the UV lamp. Nevertheless, the main asset from this AOP is the strong synergy of the photolytic dissociation of  $H_2O_2$ , along with photolytic activity [3]. Especially in the pharmaceutical industry, decomposition of the products requires the energy of this AOP in order to achieve maximum efficiency of the reactions [2,26,38,39]. The UV/H<sub>2</sub>O<sub>2</sub> process has the major advantage of utilizing UV light as a disinfectant in order to deactivate microorganisms; the radiation also assists the photolysis of hydrogen peroxide into the reactive hydroxyl radicals (•OH) needed for the process. In addition, this process is an advantageous option for treating natural and wastewater with turbid flows, utilizing the lower scattering of light. Even though some of its applications are more viable than others (such as the mineralization of some compounds found in water, including some residual pharmaceuticals), the overall conclusion is that  $UV/H_2O_2$  systems have found their place at the top of the list of the most promising technologies for water and wastewater treatment [10,21,28,35].



**Figure 1.** Carbamazepine degradation under UV light upon different H<sub>2</sub>O<sub>2</sub> concentrations (adapted from [42]).

## 3. Fenton and Photo-Fenton

Radical and heavy pollutants necessitate the use of UV radiation with more effective species, which will ensure higher catalyst effectivity at different working pH conditions [7]. In Fenton processes, reactions are favored due to the use of cheaper and less hazardous materials, simpler and less sophisticated equipment, along with ecologically meaningful cyclic reuse of reactants. The Fenton process was introduced by Fenton [43], who tried to enhance the dissociation of  $H_2O_2$  under highly acidic conditions. The reactions are described by the simplified equations as follows [44]:

$$Fe^{2+} + H_2O_2 \rightarrow Fe^{3+} + {}^{\bullet}OH + OH^-$$
 (7)

$$H_2O_2 + {}^{\bullet}OH \rightarrow H_2O + HO_2^{\bullet}$$
(8)

$$Fe^{2+} + {}^{\bullet}OH \rightarrow Fe^{3+} + OH^-$$
 (9)

$$Fe^{3+} + HO_2^{\bullet} \rightarrow Fe^{2+} + O_2 + H^+$$
 (10)

$$^{\bullet}OH + ^{\bullet}OH \rightarrow H_2O_2$$
 (11)

Organic pollutant + 
$${}^{\bullet}OH \rightarrow degraded products$$
 (12)

This synergistic combination of such radicals has been proven more effective in heavy and large molecules [7,12,35,45,46], without diminishing the ability to use AOPs in wastewater [45–47], landfill pretreatment [2,48] and drinking water purification [2,3]. In 1894, H.J.H. Fenton published the first research on the capabilities of Fe<sup>2+</sup> ions to drastically oxidize chemicals under specific conditions. The reaction solution was named after him, and later recognized as "Fenton reagent". Although this reagent is currently known as a mixture of H<sub>2</sub>O<sub>2</sub> and Fe<sup>2+</sup> ions, and the oxidation procedure is capable of successfully treating a wide range of waste streams, the moieties that are produced and are responsible for the oxidation is still under scientific research and debate [49–51].

Although Fenton's reagent was discovered over 100 years ago, its use as an oxidizing process for destroying high-molecular organic toxic substances was not applied until the 1960s [52]. The main benefit from applications of the Fenton process is the comprehensive damage of pollutants to harmless compounds, e.g.,  $CO_2$ ,  $H_2O$  and inorganic salts. Fenton's method is considered as an "on–off" process compared with other biological methods; moreover, it is environmentally friendly (decomposes into  $O_2$  and  $H_2O$ ), easy to apply, cost-effective, and can easily be applied for the treatment of wastewater [50], olive oil mill waste [49], the de-colorization of textile wastewater [53], etc.

The Fenton method involves four phases. Initially, the pH is modified to low acidity. Then, the main oxidation reaction takes place at pH values between 3 and 5. The wastewater is then neutralized at pH values of 7 to 8, usually using NaOH or CaOH to stop the reaction; at the end, precipitation occurs [49,54–56].

During the process, the OH radicals are produced through the decomposition of  $H_2O$  at pH < 6, and within a time window of  $1 \times 10^{-3}$  s, it reacts with the organic substances (RH) in the pollutant. The assumed reactions are as follows:

$$Fe^{2+} + H_2O_2 \rightarrow Fe^{3+} + OH^- + {}^{\bullet}OH$$

$$RH + {}^{\bullet}OH \rightarrow R^{\bullet} + H_2O$$

$$R^{\bullet} + Fe^{3+} \rightarrow product + Fe^{2+}$$

$$Fe^{2+} + {}^{\bullet}OH \rightarrow Fe^{3+} + OH^-$$

$$Fe^{3+} + H_2O_2 \rightarrow Fe^{2+} + H^+ + HO_2^{\bullet}$$
(13)

The combination of UV radiation with  $Fe^{2+}$  or  $Fe^{3+}$  enhances the rate of degradation of the pollutant. This photo-Fenton AOP is hindered only by the accumulation of  $Fe^{3+}$ ions, which are inhibitors to the process. During this AOP, however, more OH radicals are reproduced through photochemical regeneration by UV/solar radiation, according to the following reactions:

$$Fe^{3+} + H_2O + hv \rightarrow Fe^{2+} + {}^{\bullet}OH + H^+$$
 (14)

$$Fe^{3+} + H_2O_2 + hv \rightarrow Fe^{2+} + H^+ + HO_2^{\bullet}$$
 (15)

In low pH environments (ca. 2–3), Fenton and photo-Fenton reactions are more favored and stable. All reactions described in Equation (13) are favored at such pH values, producing more soluble and active  $Fe^{2+}$  and  $Fe^{3+}$  ions [57,58]. The typical effects of pH value on methyl orange degradation is shown in Figure 2.



**Figure 2.** pH effect on methyl orange degradation in the Fenton process (adapted and edited from [57,58]).

Fenton and photo-Fenton processes have been described in the literature by Mahdad et al. [59]. The use of such processes for the post-treatment of leachate in municipal solid waste significantly lowered the COD content, simultaneously increasing the BOD/COD ratio. The photo-Fenton process resulted in prominent results in color and COD reduction, in comparison with the Fenton process. Parameters including pH, H<sub>2</sub>O<sub>2</sub> concentration, UV radiation time and iron sulfate dosage contribute to estimations of the efficient parameters in the above processes.

Punzi et al. [60] validated the photo-Fenton process over the degradation of a textile azo-dye. The results were monitored by custom-made anaerobic 0.6 L glass reactors, totally covered with a biofilm. The results seemed efficient, because COD reduction >90% was reported during the post-treatment, while simultaneously achieving reductions in toxicity. Fiorentino et al. [61] used a solar-driven Fenton process for the treatment of real urban wastewater purification. They managed to surpass the pH barrier for sufficient bacterial inactivation and antibiotic degradation, although were not able to fully control the environmental impact of the byproducts. Their research was equipped with two identical polyvinylchloride batch mode reactors of 15 L capacity under constant 21 W/m<sup>2</sup> solar radiation. Further research is reported [12,23] on the aspects and conditions through which Fenton-like processes are affecting pharmaceutical treatments in natural waters and organic pollutants. The conclusion that conventional wastewater treatments are not as prominent as Fenton-like AOPs is encouraging the researchers to use more environmentally friendly and "green" chemical procedures. Finally, in the field of mining, Wang et al. [62] verified the effectiveness of Fenton processes in catalyzing lactate in pyrite minerals. The process produced in situ OH radicals and  $H_2O_2$  and the lactate oxidation lasted 10 d, demonstrating the potential of the process in wastewater treatment. Batch reactors made of 30 mL Pyrex vials were used with Teflon septa and rubber gaskets. The irradiation light source was a 500 W tungsten iodine lamp under a 340–3500 nm wavelength.

Fenton and photo-Fenton processes exploit the advantages of both the Fe reagent and UV radiation to the highest degree. Organic pollutants are favored by the capability of Fe ions to react with several active pollutants, especially in water purification procedures.

The rates depend on the pH value of the aqueous solution; however, wastes with values of acidic pH are mostly favored [21,41,42,52]. In general, homogeneous Fenton processes, the major disadvantage is outlined as the rates of the reactions, which are particularly slow, especially at high pH values. In addition, the products possess very high concentrations of metal-containing sludge. The method, by itself, lacks efficiency, and the heterogeneous Fenton process is often utilized due to the low requirements of the sludge preparation step, leading to lower operational costs [7,12,42].

## 4. Electro-Fenton

There has been growing attention to the use of electrochemical methods for treating hazardous wastes. The organic and toxic pollutants contained in such type of wastes, such as phenols, which exist in the structure of liquid olive oil waste, pesticides, etc., are usually destroyed by anodic oxidation as a result of the creation of oxidants such as hydroxyl radicals, ozone, etc. [63]. Electrochemical oxidation has been effectively applied in domestic sewage, the purification of olive oil wastewater [64], leachates from landfill sites [64], tannery and textile wastes [65,66], methyl parathion pesticide [63], etc. These methods are considered as environmentally friendly, because they do not use any other type of toxic substances (Figure 3). Electrochemical oxidation depends on the concentration of the prime solution such as NaCl, the pH, the range of temperature, as well as the recirculation time and the size and type of the anode and voltage applied. The electrochemical reactions are very complex and well known. However, considering that the anode is a Ti/Pt electrode, the following reactions characterized the electrochemical oxidation of aqueous solutions, which contain organic matter [63].



Figure 3. Electro-Fenton schematic (adapted from [63]).

All reactions were conducted in the electrochemical reactor, which contained reaction solutions, electrolytes, an anode and a cathode. Initially, the anode discharged the water, producing hydroxyl radicals which were adsorbed onto the active sites of the electrode surface (M[]), according to the following reaction:

$$H_2O + M[] \rightarrow M[OH^-] + H^+ + e^-$$
 (16)

Subsequently, the absorbed hydroxyl radical oxidized the organic matter as follows:

$$R + M[OH^{-}] \rightarrow M[] + RO + H^{+} + e^{-}$$
 (17)

where RO denotes the oxidized organic matter, which can be continuously formed by the hydroxyl radicals. Due to their high oxidation potential, the radicals •OH, O• and ClOH• have a very rapid lifetime compared with other oxidants (i.e., Cl<sub>2</sub>, ClO<sub>2</sub>, O<sub>2</sub>, O<sub>3</sub> and  $H_2O_2$ ). Principal (Cl<sub>2</sub> and  $O_2$ ) as well as secondary ( $H_2O_2$ ,  $O_3$  and ClO<sub>2</sub>) oxidants which are produced from the degradation process exhibit a relatively long life and are diffused into the areas away from the electrodes continuing the oxidation process (indirect oxidation). Due to catalytic activity of the anode, direct electro-chemical oxidation is affected, as are the diffusion rates of organic compounds at the active points of the anode and the applied current density. On the other hand, indirect electrochemical oxidation processes are influenced by the circulation rates of secondary oxidants within the solution, the pH and the temperature range. Secondary oxidants do not convert all pollutants to  $H_2O$  and  $CO_2$ ; therefore, effective contaminant deprivation is constructed through the electrochemical process. In solutions with pH < 6, the oxidation process creates secondary ions and free radicals (ClO<sup>-</sup>, O<sub>3</sub>, etc.) which, in turn, act as oxidation agents. In moderate alkaline pH conditions, these radicals are produced with the addition of some  $H_2O_2$  through a cycle of chloride and chlorine products, whereas in strong alkaline values, ClO<sup>3-</sup> is produced, which is a more stable form of radical, thus limiting the procedure. Consequently, chlorides are decreased in acidic conditions throughout the electrolysis progression, creating ClO<sub>2</sub>, whereas the formation of chlorates is favored in highly alkaline conditions by the chlorides. Pharmaceuticals, as well as antibiotics (such as ciprofloxacin amoxicillin, sulfamethoxazole, tetracycline and erythromycin), betablockers (such as nadolol, bisoprolol and atenolol), non-steroidal anti-inflammatory medicines (such as ibuprofen, naproxen and aspirin), neuroleptics (such as fluphenazine, clozapine and chlorpromazine), estrogens (such as nilestriol, progynova and diethylstilbestrol), and lipid regulators (such as olbemox, lovastatin and simvastatin) have commonly been identified in various water samples due to their decomposition in human and animals bodies [67,68]. For example, many antibiotics have been identified in different water bodies, including surface water, urban sewage, hospital sewage, etc., with concentrations ranging from several nanograms to several micrograms per liter. Zhang et al. considered electrochemical anoxic oxidation to be one of the most suitable processes to remove pharmaceuticals through the mechanism of electron transfer and reactive species oxidation [69]. For example, ibuprofen was entirely removed through electrochemical anoxic oxidation with a Ti/Pt/PbO<sub>2</sub> anode in 60 min, with COD and TOC values from 60% to 95% and 48% to 92%, respectively [70].

#### 5. Ozone-Based (O<sub>3</sub>) Processes

Involving  $O_3$  in aqueous reactions may produce various unwanted byproducts due to its high redox potential. The two pathways that may occur are direct and indirect reactions, with the former being preferable. The two different half-reactions that may occur in water are:

$$O_{3} + 2H^{+} + 2e^{-} \rightarrow O_{2} + H_{2}O$$

$$O_{3} + H_{2}O + e^{-} \rightarrow O_{2} + 2HO^{-}$$
(18)

Usually, ozone is utilized to break C=C bonds in organic compounds. In its indirect reactions, it creates different radicals depending on the pH values.

$$O_3 + OH^- \rightarrow HO_2^{\bullet} + O_2^{\bullet^-} (1 < pH < 7)$$
 (19)

$$O_{3} + OH^{-} \rightarrow HO_{2}^{\bullet} + O_{2} (8 < pH < 14) O_{3} + HO_{2}^{\bullet} \rightarrow HO_{2}^{\bullet} + O_{3}^{\bullet^{-}}$$
(20)

Under alkaline conditions,  $HO_2^{\bullet}$  is highly unstable, propagating reactions such as those detailed below [71]:

$$HO_{2}^{\bullet} \rightarrow O_{2}^{\bullet-} + H^{+}$$

$$O_{3} + O_{2}^{\bullet-} \rightarrow O_{3}^{\bullet-} + O_{2}$$

$$O_{3}^{\bullet-} + H^{+} \rightarrow HO_{3}^{\bullet}$$

$$HO_{3}^{\bullet} \rightarrow {}^{\bullet}OH + O_{2}$$

$$O_{3} + {}^{\bullet}OH \rightarrow HO_{4}^{\bullet}$$

$$HO_{4}^{\bullet} + HO_{2}^{\bullet} + O_{2}$$
(21)

HO<sub>4</sub> radicals ultimately produce more HO<sub>2</sub> radicals, creating OH radicals for the AOP [18]. The most significant problem with such systems is the control of ozone byproducts such as bromate and N-nitrosodimethylamine, which are both carcinogenic. These also raise issues in the biological stability of the outcomes of the AOP, because acids, aldehydes and degradable organic compounds are produced [72]. Two major categorizations are included in these O<sub>3</sub>-based processes: ozone/hydrogen peroxide and ozone/UV. The former is favored in general wastewater treatments because the production of hydroxyl radicals is enhanced through the dissociation of  $H_2O_2$  [71,73], whereas in the latter, UV radiation photolyzes O<sub>3</sub> to the production of  $H_2O_2$ ; in turn, this product produces the necessary OH radicals [40], as expressed below:

$$O_3 + H_2O + hv \rightarrow H_2O_2$$
  
$$H_2O_2 + hv \rightarrow 2 \bullet OH$$
(22)

Ozone/hydrogen peroxide has been already implicated in the pharmaceutical sector, as stated by Almomani et al. [74]. The oxidation of various antibiotics, estrogens, etc., has been monitored, and the parameters for higher yields are mainly the concentrations, directly increasing the rates of the oxidation reactions. A fully controlled double-sided glass reactor was operated in semi batch modes, with proper mass flow controllers, washing traps, a diffuser, and pH and temperature controls. An effluent-rich ozone/hydrogen peroxide catalyst was also tested for anticancer, antipsychotic and painkiller drugs. A COD reduction was achieved, reaching 88% in a pH range of  $4\sim6$ . A custom-made assembly with a glass reactor supported by ozone and oxygen generator systems with controllable flows was used for the experiments, with 200 mL of wastewater in each batch run [75]. Finally, lake water and wastewater samples were treated with ozone/hydrogen peroxide AOPs, in a batch reactor of 500 mL at pH = 8, resulting in the increased transformation of atrazine (ATR), sulfamethoxazole (SMX) and N-nitroso-dimethylamine (NDMA), whilst keeping the bromate formation at a percentage of 70% [31].

Ozone-based processes produce the strongest oxidants in large quantities due to the  $O_3$  splitting reaction. Byproducts are more biologically stable but lead to the formation of aldehydes such as formaldehyde, which is a genotoxic compound. Although it has a powerful oxidation capability, there are some drawbacks to its use in aqueous reactions, due to its low solubility in water. The mass transfer of gas–liquid interfaces and self-decomposition are the limiting steps for water purification systems. Generally, this AOP is used as a pretreatment step in wastewater treatment, with no suggestions in major water purification procedures [28,68,69,71].

#### 6. Photocatalysis

Semiconductor materials, upon exposure to ultraviolet or visible radiation, can provide surface-activated regions on which redox reaction occur, exploiting the formation of electrons and holes on their lattice. This procedure is called photocatalysis, and usually, products of the redox cycle produce HO<sup>•</sup> and  $O_2^{\bullet-}$  radicals [76,77]. The dual nature of this process involves the ability to excite the semiconductor materials and alter the physical state of the reactants at a significant rate. Band theory explains the ways of quantifying this ability in terms of band-gap energy (E<sub>g</sub>). The moment that semiconductor materials are exposed to light or UV irradiation, the photonic energy emitted from the radiation is

absorbed from an electron in the valence band, creating an electron hole and leading to its excitation to the conduction band. Figure 4 depicts a typically illustrated mechanism of pollutants via photocatalysis on TiO<sub>2</sub>, according to valence bond theory (VBT).



Figure 4. Indicative mechanism of photocatalysis on TiO<sub>2</sub> (adapted from [78]).

In general, metal oxides are the widest ranging applications for these AOPs, with  $TiO_2$ , zinc oxide and other binary metal oxides being the most abundantly used [79–81]. The categorization of different semiconductors according to their bandgap is reported in Table 1.

In lowering the bandgap of photocatalysts, less energy is demanded to initiate the photo-oxidation process; thus, lower irradiation energy is needed. All photocatalysts operate in the UV region, although considerable effort has been made to extend the photoinitiation to under visible light [82–84]. Doping with metals or non-metals in order to modify the existing energy bandgap has abundantly been investigated in the literature [84–87]. Due to the stability, low cost and non-toxicity of photocatalytic TiO<sub>2</sub>, its use is abundant in pollutant removal [88]. Hunge Y.M. et al. compared pharmaceutical drug removal from waste with Fenton [89], photo-Fenton [90], TiO<sub>2</sub> photocatalysis [91] and ZnO photocatalysis [92] oxidation processes. Of these, the ZnO UV-assisted process proved inefficient in terms of pollutant degradation. Daneshvar et al. reported the latter method to be a plausible substitute for TiO<sub>2</sub> photocatalysis due to similar photocatalytic mechanisms [93].

Xiang Li et al. used 50 mL glass beakers under 500 W of UV radiation at a 420 nm wavelength with TiO<sub>2</sub>, trying to resolve the major arsenic contamination problem for global public health [95,96]. This issue had been addressed through different approaches such as UV/Fe(III)-complexes [97], Fe(II)/H<sub>2</sub>O<sub>2</sub> and TiO<sub>2</sub> photocatalysis [98]. TiO<sub>2</sub> photocatalysis had proven to be a promising method for arsenic-contaminated water through oxidizing As(III) to the less toxic and less mobile form, As(V) [95]. Another issue that arises is that TiO<sub>2</sub> is not able to directly absorb visible light. The technique which may be used for the decomposition of environmental pollutants under solar radiation is through dye-sensitized photocatalytic oxidation (SPCO) over a TiO<sub>2</sub> catalyst [99]. All the above experiments require inexpensive setups: mainly in glass and/or Duran beakers, under stirring, and radiated under UV lamps emitting in UVA ranges at a maximum power of 500 W.

Semiconductor	Bandgap (eV @300K)
ZnS (Wurtzite)	3.91
ZnS (Zinc blende)	3.54
SnO <sub>2</sub>	3.60
TiO <sub>2</sub>	3.20
ZnO	3.03
WO <sub>3</sub>	2.60
CdS	2.42
Fe <sub>2</sub> O <sub>3</sub>	2.20
CdO	2.10
Cu <sub>2</sub> O	2.10
CdSe	1.70
AlSb	1.58
CdTe	1.56
GaAs	1.42

Table 1. Bandgaps of semiconductors (adapted from [94]).

Stepnowski P. et al. [100] reported the degradation of imidazolium ionic liquids using the UV photolysis of ionic liquids. In this investigation, 100 mL of liquid was used in a closed reaction cell, with a Degussa P25 load as the catalyst with a 1000 W xenon arc lamp. The  $H_2O_2/UV$  system was found to be the most effective regarding the degradation of all the compounds. After 360 min of the experiment, approximately 23% of both ionic liquids was mineralized. A rapid degradation rate was observed for methyl imidazole, for which degradation was complete after 180 min [100,101].

Generally, photocatalysis is used in wastewater treatment, utilizing the surface of the catalyst in order to most efficiently decontaminate the pollutants. It is the most used AOP for water pollutant degradation because it has low operational costs. Despite its vast uses in many fields, photocatalysis is the most efficient and environmentally friendly technique compared with the other AOPs. The remediation of wastewater through this AOP is especially prominent, and yields byproducts with low pollutant loads in the environment. The efficiency of the procedures is mainly attributed to the catalysts used and in the power of the radiation. As in all UV processes, light emission clearly has a dual role: as a disinfectant and power source of catalysis initiation. Limitations are due to: (i) the improper selection of catalysts, which leads to higher bandgap energies, not suitable for maximum catalytic efficiency because the production of hydroxyl radicals (•OH) is suppressed; and (ii) the radiation wavelength [74,76,78,88,92].

#### 7. Sonolysis (Ultrasound (US) Radiation)

Sonochemistry is "green" branch chemistry that uses ultrasounds either to promote or modify chemicals reactions. Compared with different sources of energy input into the reaction, ultrasonic treatment can induce a wide range of chemical and physical consequences in a non-equilibrium state, applicable for the synthesis and modification of materials, leading to non-equilibrated physicochemical characteristics, catalytic activity, surface morphology and shape. Ultrasound has frequencies ranging from 20 kHz to 1 GHz. Low-frequency treatment (power ultrasound, up to 100–200 kHz) is used to perform sonochemical processes and ultrasonic cleaning, whereas high-frequency ultrasound is most commonly used for medical diagnostics, detection, and the monitoring of processes, e.g., fluid transport. Generally, ultrasound exceeds the vibrational wavelengths on molecular and atomic scales; thus, it does not react with molecules. It creates cavitation phenomena in which bubbles are quickly formed, explode, and collapse inside a liquid medium, producing high energy

conversions to chemical energy [102,103]. The liquid medium undergoes an expansion step (rarefaction), a compression step during the compression phase of the sound field, and finally, the collapse of the formed bubble. The first two steps occur in cycles, and during cavitation phenomena, thermal transport and local hotspots are negligible. This explains the minimal temperature increase in the liquid medium. The critical diameter of the collapsing bubble is dependent on the frequency of the ultrasound and the liquid medium. The localized high temperatures and pressures provide conditions for alternative chemical reaction conditions with high product yields [102,104,105].

More specifically, three phases/zones have been recognized in sonocatalytic reactions. The internal conditions of bubbles have temperatures and pressures of approximately 5000 °C and 1000 atm, respectively, comprising an initial zone with high heating and cooling rates. The second zone consists of a gas/liquid transition region surrounding the bubble. In this zone, temperature reaches a value of ~1900 °C, and this phase has a width of 200 nm and usually consists of nonvolatile compounds. The third phase is the bulk liquid phase, where shockwaves, jets, shear forces, as well as temperature and pressure, can be kept at normal values, producing free radicals. Hydrophilic chemicals are driven in bulk, while hydrophobic chemicals remain in the boundary/interface zone. Volatile chemicals are incorporated into the bubble via evaporation [106,107]. According to the nature of the chemical/pollutant, hydrophilic compounds are treated with OH radicals in the bulk region; hydrophobic non-volatiles are degraded in the interface, whereas volatile pollutants are thermally treated inside the bubble [104]. A simple illustrated aspect of this is depicted in Figure 5 for the butyric acid (BA), before (Figure 5a) and after bubble collapse during sonolysis (Figure 5b).

The degradation of molecules via the formation, growth and collapse of a bubble in a liquid based on acoustic cavitation is called sonolysis. A bubble's collapse can occur within a few nanoseconds, reaching high temperature and pressure: up to 5000 K and 500 atm, respectively. Under these conditions, water molecules in the form of gas from the bubbles can produce hydroxyl radicals ( $H_2O \rightarrow \bullet OH + H \bullet$ ). Sonolysis is an advanced oxidation process commonly used for wastewater treatments [108]. It is a successful and highly efficient method; therefore, ultrasound reactors are finding more and more applications on an industrial scale [22,109].



**Figure 5.** (a) Gas–liquid interface before bubble collapse, and (b) sonolysis of butyric acid after bubble collapse. P, products or intermediates (adapted from [110]).

The chemistry behind this AOP involves the dissociation of the water into oxygen and nitrogen, and the further atomization of these molecules. Intermediate radicals are produced, followed occasionally by light emissions [103,111,112], as seen below:

$$\begin{array}{l} H_2O+))) \rightarrow H^{\bullet} + {}^{\bullet}OH \\ O_2+))) \rightarrow 2O^{\bullet} \\ N_2+))) \rightarrow 2N^{\bullet} \\ H^{\bullet} + O_2 \rightarrow 2HOO^{\bullet} \\ O^{\bullet} + H_2O \rightarrow 2 {}^{\bullet}OH \\ H^{\bullet} + H^{\bullet} \rightarrow H_2 \\ {}^{\bullet}OH + {}^{\bullet}OH \rightarrow H_2O_2 \\ N^{\bullet} + {}^{\bullet}OH \rightarrow HOO + H^{\bullet} \\ NO + {}^{\bullet}OH \rightarrow HNO_2 \\ 2HOO^{\bullet} \rightarrow H_2O_2 + O_2 \\ H^{\bullet} + HOO^{\bullet} \rightarrow H_2O_2 \end{array}$$

As mentioned above, US irradiation has gained considerable interest from the scientific community; therefore, commercial US reactors have been used in various cases of waste treatment, such as the purification of water from various pharmaceuticals or dyes, and some researchers have even developed cavitation devices. Saharan et al. [113] optimized three different cavitation devices, with single-hole orifice plates, circular and a slit venturi, for the degradation of Orange-D dye. They found that the geometry of the cavitation devices affects the cavitation event. The dimensions and parameters of the ultrasound devices were inspired by Bashir et al. [114]. Madhavan et al. [115] have demonstrated the favorable sonolytic degradation of Orange-D in a relatively acidic environment, close to pH 6 under 213 kHz at a 35% power amplitude from an ELAC LVG-60 RF generator. Many factors, such as the intensity and frequency of the ultrasound, the geometry of reactors, bulk temperature and the water matrix may affect the process efficiency. The most important, however, seems to be the presence of dissolved gases [5].

Pharmaceuticals, which are frequently detected in water, have drawn interest because they cannot biodegrade and persist and remain toxic [116]. They are considered emerging contaminants with potential environmental and health risks. Researchers have used different AOPs for the treatment of pharmaceutical wastewater in everyday life [117]. De Bel et al. [118] degraded ciprofloxacin from deionized water at 544 kHz. Naddeo et al. [119] achieved the degradation of diclofenac and carbamazepine from urban wastewater following first-order kinetics. Lianou et al. [120] degraded piroxicam from ultrapure water using different power densities. Ziylan-Yavas et al. [121] succeeded in the degradation of ibuprofen from ultrapure water with the application of various single ultrasonic frequencies. Serna et al. [122] degraded oxacillin from distilled water at 275 kHz. Guyer et al. [123] achieved the degradation of diclofenac from Milli-Q water at 861 kHz. The organology of the above experiments involved horn-type digital sonifiers at fixed frequencies of 20 kHz to 600 MHz at maximum power outputs of 500 W. Ultrasonic horns with titanium tips 1 cm in diameter were positioned with their ends 3 cm from the bottom of the rounded glass flasks. All the flasks were cooled in order to avoid temperature-affected phenomena.

Organic pollutants can be degraded via sonolysis. According to the work of Wang et al., there are two pathways to do it. The first method is using a cavitation bubble produced by ultrasound with high pressure and high temperature, and the second method employs reactive species which are produced due to molecular water decomposition from ultrasound [124]. The application of ultrasound in order to activate persulphate has drawn the interest of researchers because the cavitation bubbles can activate the efficiency of the persulphate [125]. Wei et al. [126] reported that ultrasonic irradiation activated persulphate for the degradation of carbamazepine, dioxines, trichloroethane and bisphenol A, both with and without thermal activation. The elimination of ibuprofen in water has been well addressed in the sequential use of ultrasonic frequencies, as reported by Ziylian-Yavas et al. [121].

In sonolysis, the production of hydroxyl radicals (•OH) is mostly favored due to the extreme conditions of the energy input for the procedure. These extreme conditions,

(23)

although short-lasting, are sufficient for producing large quantities of •OH, which promote the degradation of pollutants in water. This AOP is gaining favor, in comparison with the others mentioned above, due to the lack of reagents, ease of use and selectiveness of parameter setting according to the pollutant nature. The power consumption of the process is significantly lower than all the others, and the major disadvantages are the insufficient research on parameter selection on the different kinds of pollutants and their concentrations, and the elevated economical costs during operation [100,102,107,112].

### 8. Single and Combined AOPs

In real water matrices, the energy requirements are strongly associated with the nature of the contaminant. Regarding the removal of a pollutant, a comparison table (Table 2) is presented depicting the possible concentrations of water purification for the abovementioned AOPs and the total degradation percentages of heavy wastewater, such as leachates, and low-concentration pollutants such as antibiotics.

Experimental Parameters								
AOP	Waste/ Contaminant	pН	Time [min]	Other (Volume, Concentration, Power, Frequencies, etc.)	COD Reduction [%]	Waste Degradation [%] *	Ref.	
O <sub>3</sub> /Fenton	Leachate	7	90	1700 mg/L	65	n/a	[127]	
O <sub>3</sub> /Fenton	Leachate	7	60	400 m - I	72	n/a	[40]	
Fenton	Leachate	3	40	400 mg L -	46	n/a	[48]	
Fenton	Leachate	4.5	80	<b>7</b> 40 /I	56	n/a	[50]	
UV/Fenton	Leachate	4.5	80	740 mg/L -	84	n/a	נאכן	
UV/Fenton	Amoxicillin	2	50	6 W	81	n/a	[92]	
Fenton	Amoxicillin	3	50	104 mg/L,	80	n/a		
UV/Fenton	Amoxicillin	3	50	104 mg/L,	81	n/a	[76]	
UV/TiO <sub>2</sub> /H <sub>2</sub> O <sub>2</sub>	Amoxicillin	5	300	600 mL, 6 W,100 mg/L,	26	n/a	- [/0]	
UV/TiO <sub>2</sub>	Amoxicillin	3	300	600 mL, 6 W, 50 mg/L,	12	n/a	•	
UV/TiO <sub>2</sub>	Amoxicillin	3	30	600 mL, 6 W, 50 mg/L	12	n/a	[01]	
UV/TiO <sub>2</sub> /H <sub>2</sub> O <sub>2</sub>	Amoxicillin	5	30	600 mL, 6 W,100 mg/L	26	n/a	. [91]	
$O_3/UV/H_2O_2$	Enrofloxacin	11	20	1.4 L, 15 W	88	n/a		
O <sub>3</sub> /UV	Enrofloxacin	11	30	1.4 L, 15 W	75	n/a	[26]	
O <sub>3</sub> /UV/H <sub>2</sub> O <sub>2</sub> Fenton	Enrofloxacin	11	15	1.4 L, 15 W	100	n/a		
Sonolysis	ibuprofen	4.3	180	20 mg/L, 12 kHz, 1 L	n/a	10		
Sonolysis	Ibuprofen	4.3	180	20 mg/L, 20 kHz, 1 L	n/a	40	-	
Sonolysis	Ibuprofen	4.3	180	20 mg/L, 862 kHz	n/a	80	-	
Fenton	Ibuprofen	2.6	180	20 mg/L	n/a	80	[128]	
Sonolysis/Fenton	Ibuprofen	2.6	120	20 mg/L, 12 kHz, 1 L	n/a	95		
Sonolysis/Fenton	Ibuprofen	2.6	120	20 mg/L, 20 kHz, 1 L	n/a	97	•	
Sonolysis/Fenton	ibuprofen	2.6	120	20 mg/L, 862 kHz	n/a	100	•	
Sonolysis/Fenton	Diclofenac	n/a	40	20 mg/L, 20 kHz, 200 mL	n/a	73	[129–131]	
Sonolysis	Ibuprofen	n/a	180	14.6μg/L, 45 kHz	n/a	60	[110]	
Sonolysis	Diclofenac	5.3	180	16.6 μg/L, 200 mL, 45 kHz	n/a	60	— [119]	
Sonolysis	Ibuprofen	3	30	21 mg/L–300 mL, 300 kHz	n/a	98	[132]	
UV/H <sub>2</sub> O <sub>2</sub>	Ibuprofen	7	<30 min	11 W	n/a	60	[133]	

Table 2. Comparison of AOP degradation efficiencies in leachates and antibiotic effluents.

\* in terms of concentration degradation ratio.

In order to depict the use of AOP towards most significant polluted wastewaters, Table 2 is representative regarding the efficiency of the degradation in similar cases. For leachate treatment, ozone-based and photo-Fenton processes are more effective than Fenton processes. The use of such AOPs is induced due to the major advantages reported in each case. In the case of the leachate, due to the mixed nature of the waste (aqueous, oilbased, etc.), photo-Fenton performed more efficiently than ozone-based Fenton processes, although it exceeded the time parameters. Nevertheless, for heavily contaminated leachates, combined photo- and ozone-based Fenton techniques should be more favored over single Fenton processes.

In the case of antibiotics, their low concentrations in water are not favorably degraded by photocatalytic AOPs. In addition, although it seems that degradation only using low-frequency ultrasound irradiation is insufficient, sometimes, the combination of ultrasound with other AOPs has better effects in terms of COD reduction and waste degradation. Combined energy inputs overcome the obstacles of the byproducts of each AOP separately [128]. It is proven that only ultrasound did not affect the degradation of a pharmaceutical pollutant. Naddeo et al. [131] coupled ultrasound and Fenton reactions to enhance the degradation of diclofenac, sulfamethoxazole and carbamazepine from distilled water in single and mixed solutions. Additionally, Adityosulindro et al. [128] combined Fenton reactions and high-frequency (862 kHz) ultrasound, and succeeded in degrading ibuprofen from distilled water and wastewater, even achieving rates of 100%.

The feasibility of combined Fenton techniques was also suggested by Elmola et al. [47], in cases of the treatment of antibiotics. The combination suggests that AOP may be used in tandem with sequencing batch reactor (SBR) methods. In addition, Tokomura et al. [134] combined a wide range of AOPs for the removal of certain pharmaceuticals. In the textile industry, the synergistic effect of sonolysis and photocatalysis throughout Fenton processes surpasses the energy barriers of the decolorization of complex azodyes systems [135]. Even in the cases of azoic dyes, the use of different AOPs suggests a more synergistic combination of the techniques [136]. In cases of heavy pollution and municipal wastewater treatment [137–140], Amr et al. [127] stabilized municipal leachate with ozonation combined with Fenton processes, reducing the treatment period and yielding a sufficient value of COD removal (0.63 kg/kg COD). Organic pollutants such as organosulfur, nitro derivatives of benzene, BTEX and phenol were subjected to 100% oxidation in the combined use of sonolysis with ozone/hydrogen peroxide AOP [141]. Additionally, the viability of a multi-barrier treatment through combined  $H_2O_2/UVC$  techniques allowed the reuse of wastewater after eliminating selective contaminants [36]. The use of AOP versus physical or chemical processes is favored mainly because most contaminants present in wastewater are found to be recalcitrant to the physical/chemical treatment process.

Figure 6 presents an overview of the mechanisms of the aforementioned AOPs.

UV/H <sub>2</sub> O <sub>2</sub>	Fenton and Photo Fenton	Electro Fenton
$H_2O_2 \xrightarrow{hv} 2HO^{\bullet}$	$Fe^{2+} + H_2O_2 \rightarrow Fe^{3+} + HO^{\bullet} + OH^{-}$ $Fe^{3} + H_2O_2 \xrightarrow{hv} Fe^{2+} + HO^{\bullet} + H^{+}$	$H_2O + M[] \rightarrow M[OH-] + H^+ + e^-$ R + M[OH-] → M[] + RO + H <sup>+</sup> + e
O <sub>3</sub> Processes	Photocatalysis	Sonolysis
$O_3 + H_2O + hv \rightarrow H_2O$ $H_2O_2 + hv \rightarrow 2^{\bullet}OH$	$MOx \xrightarrow{hv} h_{VB}^{+} + e_{CB}^{-}$ $O_{2} + e_{CB}^{-} \rightarrow O_{2}^{\bullet}$ $H_{2}O + h_{VB}^{+} \rightarrow HO^{\bullet} + H^{+}$	$H_2O \xrightarrow{)))} H^{\bullet} + {}^{\bullet}OH$

Figure 6. Overview of the mechanisms in AOP.

In every AOP, the production of free and strong OH radicals is achieved through different sources of power and materials. Firstly, in UV peroxide AOPs, the power from UV radiation is capable of producing the necessary OH radicals. In contrast to Fenton processes, the addition of  $Fe^{3+}/Fe^{2+}$  ions with  $H_2O_2$  (Fenton reagent) produces highly active  $Fe^{2+}/Fe^{3+}$  ions, respectively, with OH radicals providing the necessary catalytic system to properly treat specific wastewater. In electro-Fenton reactions, the process utilizes electrical energy to split the water molecule into OH free radicals with the simultaneous presence of Fe ions. Ozone-based processes use photons (hv) to initiate the production of hydrogen peroxide which, in turn, produces OH radicals. In photocatalysis, a metal-oxide catalyst is used, which utilizes the UV/Vis radiation to create electron hole pairs for the generation of OH free radicals. Ultimately, energy from the ultrasound (sonolysis) is efficient for the rapid production of H and OH radicals with high yield.

In Table 3, the main advantages and disadvantages of each AOP are presented.

Table 3. Advantages and disadvantages of AOPs.

АОР	Advantages	Disadvantages	Ref.
UV/H <sub>2</sub> O <sub>2</sub>	<ul> <li>Simultaneous use of UV as a disinfectant;</li> <li>Treats wastewater utilizing low light scattering;</li> <li>Low cost.</li> </ul>	<ul> <li>Inefficient for heavily polluted streams;</li> <li>High operating costs;</li> <li>Large quantities.</li> </ul>	[142–148]
Fenton-based	<ul> <li>Exploitation of Fe ions and UV/voltage;</li> <li>The versatility of Fe ions to react with pollutants;</li> <li>Highly active in acidic pH conditions.</li> </ul>	<ul> <li>Slow reaction rate;</li> <li>Metal-containing sludge byproducts;</li> <li>Fenton reagent preparation cost;</li> <li>Chlorate formation in alkaline solution.</li> </ul>	[149–155]
Ozone-based	<ul> <li>High reaction rate (high production of OH due to O<sub>3</sub> splitting);</li> <li>More stable byproducts;</li> <li>Effective in pretreatment for water purification;</li> </ul>	<ul> <li>Formation of genotoxic compounds;</li> <li>Low solubility in water/drawbacks in aqueous reactions;</li> <li>Self-decomposition.</li> </ul>	[156–159]
Photocatalysis	<ul> <li>Stability, low-cost, non-toxicity;</li> <li>Effective due to catalyst surface in water pollutant degradation;</li> <li>Environmentally friendly/low pollutant load of byproducts;</li> </ul>	<ul> <li>Improper catalyst selection;</li> <li>Radiation wavelengths require high operational costs.</li> </ul>	[160–168]
Sonolysis	<ul> <li>Higher OH production;</li> <li>Lack of reagents;</li> <li>Parameter versatility.</li> </ul>	<ul> <li>Parameter versatility;</li> <li>Uncontrolled byproducts;</li> <li>Targets low-concentration wastewater.</li> </ul>	[169–176]

#### 9. Conclusions

AOPs have been fully implemented in wastewater treatment, the pharmaceutical industry, textiles, and at the general circular economy scale globally. Categorization of the processes and applications mentioned in the research can be used to evaluate the effect of each AOP separately, as well as in combination. Single AOPs may be used to purify and treat waste and wastewater at very efficient rates; however, combinations of AOPs have been implemented in many cases, with prominent results. This review represents a guide useful for rapidly assessing the drawbacks and limitations of each AOP in the prospect of potential uses in wastewater treatments. Major advantages and disadvantages of each process are presented, focusing on the operation costs, reaction rates and formation

of byproducts. Major limitations are the concentrations of wastewater effluents and the up-scalability of the aforementioned AOPs. The equipment required is inexpensive because it consists of custom-made vessels and glass apparatus. Especially for the treatment of wastewater contaminated by antibiotics, Fenton-based and UV processes seem to favor degradation of the effluent streams. Nevertheless, the combined efficiency of AOPs holds the key to a more rational, cost-effective, "green" process of handling global pollution and contaminant problems. The efficient removal of vital parameters in each case will create the necessary global database and shed light on global pollution solutions.

**Author Contributions:** Writing—original draft preparation, P.K.P., C.K., E.K., C.V., M.G.S.; writing—review and editing, P.K.P.; visualization, P.K.P.; supervision, G.S., C.A. and A.A.Z. All authors have read and agreed to the published version of the manuscript.

**Funding:** This study has been partially funded by the European Union's Horizon 2020 research and innovation programme, under grant agreement number 870292 (BioICEP).

Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Not applicable.

Data Availability Statement: Not applicable.

Acknowledgments: The authors would like to acknowledge National Technical University of Athens (Greece), Clausthal University of Technology (Germany) and Open University of Cyprus for their support and research facilities. They also declare that this study has been partially funded by the European Union's Horizon 2020 research and innovation programme, under grant agreement number 870292 (BioICEP).

Conflicts of Interest: The authors declare no conflict of interest.

#### References

- 1. Covinich, L.G.; Bengoechea, D.I.; Fenoglio, R.J.; Area, M.C. Advanced Oxidation Processes for Wastewater Treatment in the Pulp and Paper Industry: A Review. *Am. J. Environ. Eng.* **2014**, *4*, 56–70. [CrossRef]
- Miklos, D.B.; Remy, C.; Jekel, M.; Linden, K.G.; Drewes, J.E.; Hübner, U. Evaluation of advanced oxidation processes for water and wastewater treatment—A critical review. *Water Res.* 2018, 139, 118–131. [CrossRef]
- 3. Sillanpää, M.; Ncibi, M.C.; Matilainen, A. Advanced oxidation processes for the removal of natural organic matter from drinking water sources: A comprehensive review. *J. Environ. Manag.* 2018, 208, 56–76. [CrossRef]
- 4. Mota, A.L.N.; Albuquerque, L.F.; Beltrame, L.T.C.; Chiavone-Filho, O.; Machulek, A., Jr.; Nascimento, C.A.O. Advanced oxidation Processes and Their Application in The Petroleum Industry: A Review. *Braz. J. Pet. Gas* **2008**, *2*, 122–142.
- Klavarioti, M.; Mantzavinos, D.; Kassinos, D. Removal of residual pharmaceuticals from aqueous systems by advanced oxidation processes. *Environ. Int.* 2009, 35, 402–417. [CrossRef]
- Michael, I.; Rizzo, L.; McArdell, C.S.; Manaia, C.; Merlin, C.; Schwartz, T.; Dagot, C.; Fatta-Kassinos, D. Urban wastewater treatment plants as hotspots for the release of antibiotics in the environment: A review. *Water Res.* 2013, 47, 957–995. [CrossRef]
- Bokare, A.D.; Choi, W. Review of iron-free Fenton-like systems for activating H2O2 in advanced oxidation processes. J. Hazard. Mater. 2014, 275, 121–135. [CrossRef]
- Katheresan, V.; Kansedo, J.; Lau, S.Y. Efficiency of various recent wastewater dye removal methods: A review. J. Environ. Chem. Eng. 2018, 6, 4676–4697. [CrossRef]
- 9. Phugare, S.S.; Kalyani, D.C.; Surwase, S.N.; Jadhav, J.P. Ecofriendly degradation, decolorization and detoxification of textile effluent by a developed bacterial consortium. *Ecotoxicol. Environ. Saf.* **2011**, *74*, 1288–1296. [CrossRef]
- 10. Keen, O.S.; McKay, G.; Mezyk, S.P.; Linden, K.G.; Rosario-Ortiz, F.L. Identifying the factors that influence the reactivity of effluent organic matter with hydroxyl radicals. *Water Res.* **2014**, *50*, 408–419. [CrossRef]
- Krishnan, S.; Rawindran, H.; Sinnathambi, C.M.; Lim, J.W. Comparison of various advanced oxidation processes used in remediation of industrial wastewater laden with recalcitrant pollutants. In *IOP Conference Series: Materials Science and Engineering*; IOP Publishing: Bristol, UK, 2017; Volume 206, p. 012089.
- 12. Mirzaei, A.; Chen, Z.; Haghighat, F.; Yerushalmi, L. Removal of pharmaceuticals from water by homo/heterogonous Fenton-type processes—A review. *Chemosphere* **2017**, *174*, 665–688. [CrossRef]
- Tian, J.; Wu, C.; Yu, H.; Gao, S.; Li, G.; Cui, F.; Qu, F. Applying ultraviolet/persulfate (UV/PS) pre-oxidation for controlling ultra-filtration membrane fouling by natural organic matter (NOM) in surface water. *Water Res.* 2018, 132, 190–199. [CrossRef]
   Zorpas, A.A.; Lasaridi, K. Measuring waste prevention. *Waste Manag.* 2013, 33, 1047–1056. [CrossRef]
- 15. Lasaridi, K.; Chroni, C.; Antonis, A.; Zorpas, K.; Abeliotis, K. Chapter 4: Waste Prevention. In *Sustainable Solid Waste Management*; Harokopio University: Athens, Greece, 2016; pp. 53–92.

- Perez-Gimeno, A.; Navarro-Pedreno, J.; Almendro-Candel, M.B.; Gomez, I.; Zorpas, A.A. The use of wastes (organic and inorganic) in land restoration in relation to their characteristics and cost. *Waste Manag. Res. J.-Ternational Solid Wastes Public Clean.* Assoc. ISWA 2019, 37, 502–507. [CrossRef]
- 17. Prousek, J. Advanced Oxidation Processes for water treatment, Chemical Processes. Chem. Listy 1996, 90, 229–237.
- Andreozzi, R.; Caprio, V.; Insola, A.; Marotta, R. Advanced oxidation by ozone to remove recalcitrance from waste waters—A review. *Environ. Technol.* 1999, 22, 51–59.
- Gil, L.A.G.A.; Vicente, M.Á. The Handbook of Environmental Chemistry—Applications of Advanced Oxidation Processes (AOPs) in Drinking Water; Springer International Publishing: Berlin/Heidelberg, Germany, 2019.
- Oturan, M.A.; Aaron, J.-J. Advanced Oxidation Processes in Water/Wastewater Treatment: Principles and Applications. A Review. *Crit. Rev. Environ. Sci. Technol.* 2014, 44, 2577–2641. [CrossRef]
- Asghar, A.; Raman, A.A.A.; Daud, W.M.A.W. Advanced oxidation processes for in-situ production of hydrogen peroxide/hydroxyl radical for textile wastewater treatment: A review. J. Clean. Prod. 2015, 87, 826–838. [CrossRef]
- 22. Boczkaj, G.; Fernandes, A. Wastewater treatment by means of advanced oxidation processes at basic pH conditions: A review. *Chem. Eng. J.* **2017**, *320*, 608–633. [CrossRef]
- Matafonova, G.; Batoev, V. Recent advances in application of UV light-emitting diodes for degrading organic pollutants in water through advanced oxidation processes: A review. *Water Res.* 2018, 132, 177–189. [CrossRef]
- Macías-Quiroga, I.F.; Henao-Aguirre, P.A.; Marín-Flórez, A.; Arredondo-López, S.M.; Sanabria-González, N.R. Bibliometric analysis of advanced oxidation processes (AOPs) in wastewater treatment: Global and Ibero-American research trends. *Environ. Sci. Pollut. Res. Int.* 2021, 28, 23791–23811. [CrossRef]
- 25. Garcia-Costa, A.L.; Alves, A.; Madeira, L.M.; Santos, M.S. Oxidation processes for cytostatic drugs elimination in aqueous phase: A critical review. *J. Environ. Chem. Eng.* **2021**, *9*, 104709. [CrossRef]
- Bobu, M.; Yediler, A.; Siminiceanu, I.; Zhang, F.; Schulte-Hostede, S. Comparison of different advanced oxidation processes for the degradation of two fluoroquinolone antibiotics in aqueous solutions. *J. Environ. Sci. Heal Part A Toxic Hazard. Subst. Environ. Eng.* 2013, 48, 251–262. [CrossRef]
- 27. Jamil, T.S.; Roland, H.; Michael, H.; Jens-Uwe, R. Homogeneous photocatalytic processes for degradation of some endocrine disturbing chemicals under UV irradiation. *J. Water Process. Eng.* **2017**, *18*, 159–168. [CrossRef]
- Verma, S.; Sillanpää, M. Degradation of anatoxin-a by UV-C LED and UV-C LED/H<sub>2</sub>O<sub>2</sub> advanced oxidation processes. *Chem. Eng. J.* 2015, 274, 274–281. [CrossRef]
- Imoberdorf, G.; Mohseni, M. Kinetic study and modeling of the vacuum-UV photoinduced degradation of 2,4-D. *Chem. Eng. J.* 2012, 187, 114–122. [CrossRef]
- Saritha, P.; Aparna, C.; Himabindu, V.; Anjaneyulu, Y. Comparison of various advanced oxidation processes for the degradation of 4-chloro-2 nitrophenol. J. Hazard. Mater. 2007, 149, 609–614. [CrossRef]
- Katsoyiannis, I.A.; Canonica, S.; von Gunten, U. Efficiency and energy requirements for the transformation of organic micropollutants by ozone, O<sub>3</sub>/H<sub>2</sub>O<sub>2</sub> and UV/H<sub>2</sub>O<sub>2</sub>. Water Res. 2011, 45, 3811–3822. [CrossRef]
- Bagheri, M.; Mohseni, M. Impact of hydrodynamics on pollutant degradation and energy efficiency of VUV/UV and H<sub>2</sub>O<sub>2</sub>/UV oxidation processes. *J. Environ. Manag.* 2015, 164, 114–120. [CrossRef]
- 33. Ike, I.A.; Karanfil, T.; Cho, J.; Hur, J. Oxidation byproducts from the degradation of dissolved organic matter by advanced oxidation processes—A critical review. *Water Res.* **2019**, *164*, 114929. [CrossRef]
- Azbar, N.; Yonar, T.; Kestioglu, K. Comparison of various advanced oxidation processes and chemical treatment methods for COD and color removal from a polyester and acetate fiber dyeing effluent. *Chemosphere* 2004, 55, 35–43. [CrossRef]
- Kusvuran, E.; Gulnaz, O.; Irmak, S.; Atanur, O.M.; Yavuz, H.I.; Erbatur, O. Comparison of several advanced oxidation processes for the decolorization of Reactive Red 120 azo dye in aqueous solution. *J. Hazard. Mater.* 2004, 109, 85–93. [CrossRef]
- Rueda-Márquez, J.J.; Pintado-Herrera, M.G.; Martín-Díaz, M.L.; Acevedo-Merino, A.; Manzano, M.A. Combined AOPs for potential wastewater reuse or safe discharge based on multi-barrier treatment (microfiltration-H2O2/UV-catalytic wet per-oxide oxidation). *Chem. Eng. J.* 2015, 270, 80–90. [CrossRef]
- Crapulli, F.; Santoro, D.; Sasges, M.R.; Ray, A.K. Mechanistic modeling of vacuum UV advanced oxidation process in an annular photoreactor. *Water Res.* 2014, 64, 209–225. [CrossRef]
- 38. Rosenfeldt, E.; Boal, A.K.; Springer, J.; Stanford, B.; Rivera, S.; Kashinkunti, R.D.; Metz, D.H. Comparison of UV-mediated Advanced Oxidation. *J.-Am. Water Work. Assoc.* **2013**, *105*, 29–33. [CrossRef]
- 39. Zoschke, K.; Dietrich, N.; Bornick, H.; Worch, E. UV-based advanced oxidation processes for the treatment of odour compounds: Efficiency and by-product formation. *Water Res.* **2012**, *46*, 5365–5373. [CrossRef]
- 40. Oppenlaender, T. Photochemical Purification of Water and Air: Advanced Oxidation Processes (AOPs): Principles, Reaction Mechanisms, Reactor Concepts; John Wiley & Sons: Hoboken, NJ, USA, 2007.
- Edalatmanesh, M.; Dhib, R.; Mehrvar, M. Kinetic modeling of aqueous phenol degradation by UV/H<sub>2</sub>O<sub>2</sub> process. *Int. J. Chem. Kinet.* 2008, 40, 34–43. [CrossRef]
- 42. Ali, F.; Khan, J.A.; Shah, N.S.; Sayed, M.; Khan, H.M. Carbamazepine degradation by UV and UV-assisted AOPs: Kinetics, mechanism and toxicity investigations. *Process. Saf. Environ. Prot.* 2018, 117, 307–314. [CrossRef]
- 43. Fenton, H.J.H. Oxidation of tartaric acid in presence of iron. J. Chem. Soc. Trans. 1894, 65, 899–910. [CrossRef]

- 44. Haber, R.W.F. Unpaarigkeit und Radikalketten im Reaktionsmechanismus organischer und enzymatischer Vorgänge. *Ber. Dtsch. Chem. Ges.* **1931**, *64*, 2844–2856. [CrossRef]
- 45. Cañizares, P.; Lobato, J.; Paz, R.; Rodrigo, M.; Sáez, C. Advanced oxidation processes for the treatment of olive-oil mills wastewater. *Chemosphere* **2007**, *67*, 832–838. [CrossRef]
- Blanco, J.; Torrades, F.; de la Varga, M.; García-Montaño, J. Fenton and biological-Fenton coupled processes for textile wastewater treatment and reuse. *Desalination* 2012, 286, 394–399. [CrossRef]
- 47. Elmolla, E.S.; Chaudhuri, M. The feasibility of using combined Fenton-SBR for antibiotic wastewater treatment. *Desalination* **2012**, 285, 14–21. [CrossRef]
- 48. Cortez, S.; Teixeira, P.; Oliveira, R.; Mota, M. Evaluation of Fenton and ozone-based advanced oxidation processes as mature landfill leachate pre-treatments. *J. Environ. Manag.* 2011, *92*, 749–755. [CrossRef] [PubMed]
- Zorpas, A.A.; Costa, C.N. Combination of Fenton oxidation and composting for the treatment of the olive solid residue and the olive mile wastewater from the olive oil industry in Cyprus. *Bioresour. Technol.* 2010, 101, 7984–7987. [CrossRef] [PubMed]
- 50. Zorpas, A.A. Alternative treatment of urban wastewater using electrochemical oxidation. *Desalination Water Treat.* 2011, 27, 268–276. [CrossRef]
- 51. Zorpas, A.A. Chemical oxidation and Membrane Bioreactor for the treatment of Household heating wastewater. *Desalination Water Treat.* **2013**, *51*, 6952–6960. [CrossRef]
- 52. Huang, C.P.; Dong, C.; Tang, Z. Advanced Chemical Oxidation: Its Present role and Potential Future in Hazardous waste treatment. *Waste Manag.* **1993**, *13*, 361–377. [CrossRef]
- 53. Meric, S.; Selcuk, H.; Belgiorno, V. Acute toxicity removal in textile finishing wastewater by Fenton's oxidation, ozone and coagulation-flocculation processes. *Water Res.* 2005, *39*, 1147–1153. [CrossRef] [PubMed]
- 54. Kang, Y.W.; Hwang, K.Y. Effects on Reaction Conditions on the Oxidation Efficiency in the Fenton Process. *Wat. Res.* **2000**, *34*, 2786–2790. [CrossRef]
- 55. Xu, X.R.; Zhao, Z.Y.; Li, X.Y.; Gu, J.D. Chemical oxidative degradation of methyl tert-butyl ether in aqueous solution by Fenton's reagent. *Chemosphere* **2004**, *55*, 73–79. [CrossRef] [PubMed]
- 56. Zorpas, A.A.; Inglezakis, V.J. Intergraded Applied Methodology for the Treatment of Heavy Polluted Waste Waters from Olive Oil Industries. *Appl. Environ. Soil Sci.* 2011, 2011, 1–14. [CrossRef]
- 57. Hsueh, C.L.; Huang, Y.H.; Wang, C.C.; Chen, C.Y. Degradation of azo dyes using low iron concentration of Fenton and Fenton-like system. *Chemosphere* 2005, *58*, 1409–1414. [CrossRef] [PubMed]
- 58. Yang, C.; Wang, D.; Tang, Q. The synthesis of NdFeB magnetic activated carbon and its application in degradation of azo dye methyl orange by Fenton-like process. *J. Taiwan Inst. Chem. Eng.* **2014**, *45*, 2584–2589. [CrossRef]
- Mahdad, F.; Younesi, H.; Bahramifar, N.; Hadavifar, M. Optimization of Fenton and photo-Fenton-based advanced oxidation processes for post-treatment of composting leachate of municipal solid waste by an activated sludge process. *KSCE J. Civ. Eng.* 2016, 20, 2177–2188. [CrossRef]
- Punzi, M.; Anbalagan, A.; Börner, R.A.; Svensson, B.-M.; Jonstrup, M.; Mattiasson, B. Degradation of a textile azo dye using biological treatment followed by photo-Fenton oxidation: Evaluation of toxicity and microbial community structure. *Chem. Eng. J.* 2015, 270, 290–299. [CrossRef]
- 61. Fiorentino, A.; Esteban, B.; Garrido-Cardenas, J.A.; Kowalska, K.; Rizzo, L.; Aguera, A.; Pérez, J.A.S. Effect of solar photo-Fenton process in raceway pond reactors at neutral pH on antibiotic resistance determinants in secondary treated urban wastewater. *J. Hazard. Mater.* **2019**, *378*, 120737. [CrossRef]
- 62. Wang, W.; Qu, Y.; Yang, B.; Liu, X.; Su, W. Lactate oxidation in pyrite suspension: A Fenton-like process in situ generating H<sub>2</sub>O. *Chemosphere* **2012**, *86*, 376–382. [CrossRef] [PubMed]
- 63. Arapoglou, D.; Vlyssides, A.; Israilides, C.; Zorpas, A.; Karlis, P. Detoxification of methyl-parathion pesticide in aqueous solutions by electrochemical oxidation. *J. Hazard. Mater.* **2003**, *98*, 191–199. [CrossRef]
- 64. Lyssides, A.G.V.; Israelides, C.J.; Mourafeti, V.N.; Karvouni, G. Olive Oil Wastewater treatment with the use of An Electrolysis System. *Bioresour. Technol.* **1997**, *61*, 163–170.
- Vlyssides, A.G.; Loizidou, M.; Karlis, P.K.; Zorpas, A.A.; Papaioannou, D. Electrochemical Oxidation of a Textile Dye Wastewater using a Ptr/Ti electrode. J. Hazard. Mater. 1999, 70, 41–52. [CrossRef]
- 66. Vlyssides, A.; Papaioannou, D.; Loizidoy, M.; Karlis, P.; Zorpas, A. Testing an electrochemical method for treatment of textile dye wastewater. *Waste Manag.* 2000, 20, 569–574. [CrossRef]
- 67. Zhang, J.; Zhou, Y.; Yao, B.; Yang, J.; Zhi, D. Current progress in electrochemical anodic-oxidation of pharmaceuticals: Mechanisms, influencing factors, and new technique. *J. Hazard. Mater.* **2021**, *418*, 126313. [CrossRef]
- Chang, X.; Meyer, M.; Liu, X.; Zhao, Q.; Chen, H.; Chen, J.-A.; Qiu, Z.; Yang, L.; Cao, J.; Shu, W. Determination of antibiotics in sewage from hospitals, nursery and slaughter house, wastewater treatment plant and source water in Chongqing region of Three Gorge Reservoir in China. *Environ. Pollut.* 2010, *158*, 1444–1450. [CrossRef] [PubMed]
- 69. Wang, Y.; Zhou, C.; Wu, J.; Niu, J. Insights into the electrochemical degradation of sulfamethoxazole and its metabolite by Ti/SnO<sub>2</sub>-Sb/Er-PbO<sub>2</sub> anode. *Chin. Chem. Lett.* **2020**, *31*, 2673–2677. [CrossRef]
- Ciríaco, L.; Anjo, C.; Correia, J.; Pacheco, M.; Lopes, A. Electrochemical degradation of Ibuprofen on Ti/Pt/PbO<sub>2</sub> and Si/BDD electrodes. *Electrochim. Acta* 2009, 54, 1464–1472. [CrossRef]
- 71. Beltran, F.J. Ozone Reaction Kinetics for Water and Waste water Systems; Lewis Publishers: Boca Raton, FL, USA, 2003.

- Krasner, S.W.; Mitch, W.; McCurry, D.L.; Hanigan, D.; Westerhoff, P. Formation, precursors, control, and occurrence of nitrosamines in drinking water: A review. *Water Res.* 2013, 47, 4433–4450. [CrossRef]
- 73. Rakness, K.L. *Process Design, Operation, and Optimization, Ozone in Drinking Water Treatment;* American Water Works Association: Denver, CO, USA, 2005.
- 74. Almomani, F.A.; Shawaqfah, M.; Bhosale, R.R.; Kumar, A. Removal of emerging pharmaceuticals from wastewater by ozone-based advanced oxidation processes. *Environ. Prog. Sustain. Energy* **2016**, *35*, 982–995. [CrossRef]
- Patel, S.; Mondal, S.; Majumder, S.K.; Das, P.; Ghosh, P. Treatment of a Pharmaceutical Industrial Effluent by a Hybrid Process of Advanced Oxidation and Adsorption. ACS Omega 2020, 5, 32305–32317. [CrossRef]
- 76. Elmolla, E.S.; Chaudhuri, M. Comparison of different advanced oxidation processes for treatment of antibiotic aqueous solution. *Desalination* **2010**, 256, 43–47. [CrossRef]
- 77. Vaitsis, C.; Mechili, M.; Argirusis, N.; Kanellou, E.; Pandis, P.K.; Sourkouni, G.; Zorpas, A.; Argirusis, C. Ultrasound-Assisted Preparation Methods of Nanoparticles for Energy-Related Applications. In *Nanotechnology and the Environment*; IntechOpen: London, UK, 2020.
- Etacheri, V.; Di Valentin, C.; Schneider, J.; Bahnemann, D.B.D.; Pillai, S.C. Visible-light activation of TiO2 photocatalysts: Advances in theory and experiments. J. Photochem. Photobiol. C Photochem. Rev. 2015, 25, 1–29. [CrossRef]
- Karunakaran, C.; Senthilvelan, S. Photocatalysis with ZrO<sub>2</sub>: Oxidation of aniline. *J. Mol. Catal. A Chem.* 2005, 233, 1–8. [CrossRef]
   Chan, S.H.S.; Wu, T.Y.; Juan, J.C.; Teh, C.Y. Recent developments of metal oxide semiconductors as photocatalysts in advanced
- oxidation processes (AOPs) for treatment of dye waste-water. *J. Chem. Technol. Biotechnol.* 2011, *86*, 1130–1158. [CrossRef]
  81. Wang, H.; Zhang, L.; Chen, Z.; Hu, J.; Li, S.; Wang, Z.; Liu, J.; Wang, X. Semiconductor heterojunction photocatalysts: Design, construction, and photocatalytic performances. *Chem. Soc. Rev.* 2014, *43*, 5234–5244. [CrossRef]
- 82. Nyamukamba, P.; Tichagwa, L.; Mamphweli, S.; Petrik, L. Silver/Carbon Codoped Titanium Dioxide Photocatalyst for Improved Dye Degradation under Visible Light. *Int. J. Photoenergy* **2017**, 2017, 3079276. [CrossRef]
- Stucchi, M.; Bianchi, C.; Pirola, C.; Vitali, S.; Cerrato, G.; Morandi, S.; Argirusis, C.; Sourkouni, G.; Sakkas, P.; Capucci, V. Surface decoration of commercial micro-sized TiO<sub>2</sub> by means of high energy ultrasound: A way to enhance its photocatalytic activity under visible light. *Appl. Catal. B Environ.* 2015, *178*, 124–132. [CrossRef]
- 84. Stucchi, M.; Bianchi, C.; Argirusis, C.; Pifferi, V.; Neppolian, B.; Cerrato, G.; Boffito, D. Ultrasound assisted synthesis of Agdecorated TiO<sub>2</sub> active in visible light. *Ultrason. Sonochem.* **2018**, *40*, 282–288. [CrossRef] [PubMed]
- 85. Singh, R.R.N. To Study the Effect of the Concentration of Carbon on Ultraviolet and Visible Light Photo Catalytic Activity and Characterization of Carbon Doped TiO<sub>2</sub>. J. Nanomed. Nanotechnol. **2015**, *6*, 1.
- Poulia, A.; Sakkas, P.; Kanellopoulou, D.; Sourkouni, G.; Legros, C.; Argirusis, C. Preparation of metal–ceramic composites by sonochemical synthesis of metallic nano-particles and in-situ decoration on ceramic powders. *Ultrason. Sonochem.* 2016, 31, 417–422. [CrossRef]
- Stucchi, M.; Bianchi, C.L.; Pirola, C.; Cerrato, G.; Morandi, S.; Argirusis, C.; Sourkouni, G.; Naldoni, A.; Capucci, V. Copper NPs decorated titania: A novel synthesis by high energy US with a study of the photocatalytic activity under visible light. *Ultrason. Sonochem.* 2016, *31*, 295–301. [CrossRef]
- Pekakis, P.A.; Xekoukoulotakis, N.; Mantzavinos, D. Treatment of textile dyehouse wastewater by TiO<sub>2</sub> photocatalysis. *Water Res.* 2006, 40, 1276–1286. [CrossRef]
- 89. Elmolla, E.; Chaudhuri, M. Optimization of Fenton process for treatment of amoxicillin, ampicillin and cloxacillin antibiotics in aqueous solution. *J. Hazard. Mater.* 2009, 170, 666–672. [CrossRef]
- 90. Elmolla, E.S.; Chaudhuri, M. Degradation of the antibiotics amoxicillin, ampicillin and cloxacillin in aqueous solution by the photo-Fenton process. *J. Hazard. Mater.* **2009**, *172*, 1476–1481. [CrossRef]
- Elmolla, E.S.; Chaudhuri, M. Photocatalytic degradation of amoxicillin, ampicillin and cloxacillin antibiotics in aqueous solution using UV/TiO<sub>2</sub> and UV/H<sub>2</sub>O<sub>2</sub>/TiO<sub>2</sub> photocatalysis. *Desalination* 2010, 252, 46–52. [CrossRef]
- Elmolla, E.S.; Chaudhuri, M. Degradation of amoxicillin, ampicillin and cloxacillin antibiotics in aqueous solution by the UV/ZnO photocatalytic process. J. Hazard. Mater. 2010, 173, 445–449. [CrossRef] [PubMed]
- Daneshvar, N.; Salari, D.; Khataee, A. Photocatalytic degradation of azo dye acid red 14 in water on ZnO as an alternative catalyst to TiO<sub>2</sub>. J. Photochem. Photobiol. A Chem. 2004, 162, 317–322. [CrossRef]
- 94. Ameta, S.C.; Ameta, R. Advance Oxidation Processes for Wastewater Treatment; Elsevier: Amsterdam, The Netherlands, 2018.
- 95. Lee, W.C.H. Photocatalytic Oxidation of Arsenite in TiO2 Suspension: Kinetics and Mechanisms. *Environ. Sci. Technol* 2002, *36*, 3872–3878. [CrossRef]
- Buschmann, J.; Canonica, S.; Lindauer, U.; Hug, S.J.; Sigg, L. Photoirradiation of Dissolved Humic Acid Induces Arsenic(III) Oxidation. *Environ. Sci. Technol.* 2005, 39, 9541–9546. [CrossRef] [PubMed]
- 97. Benjamin, W.P.I.; Kocar, D. Photochemical Oxidation of As(III) in Ferrioxalate Solutions. Environ. Sci. Technol. 2003, 37, 1581–1588.
- Fei, H.; Leng, W.; Li, X.; Cheng, X.; Xu, Y.; Zhang, J.; Cao, C. Photocatalytic Oxidation of Arsenite over TiO<sub>2</sub>: Is Superoxide the Main Oxidant in Normal Air-Saturated Aqueous Solutions? *Environ. Sci. Technol.* 2011, 45, 4532–4539. [CrossRef]
- Wei, Z.; Sun, Y.; Felix, N. Visible-Light Induced Water Detoxification Catalyzed by PtII Dye Sensitized Titania. J. Am. Chem. Soc. 2008, 130, 12566–12567.
- Stepnowski, P.; Zaleska, A. Comparison of different advanced oxidation processes for the degradation of room temperature ionic liquids. J. Photochem. Photobiol. A Chem. 2005, 170, 45–50. [CrossRef]

- 101. Hunge, Y.M. Basics and advanced developments in photocatalysis—A review (Mini review). *Int. J. Hydrol.* **2018**, *2*, 539–540. [CrossRef]
- 102. Pestman, J.M.; Engberts, J.B.F.N.; De Jong, F. Sonochemistry: Theory and applications. *Recl. Des Trav. Chim. Des Pays-Bas* 2010, 113, 533–542. [CrossRef]
- 103. Adewuyi, Y.G. Sonochemistry: Environmental Science and Engineering Applications. *Ind. Eng. Chem. Res.* 2001, 40, 4681–4715. [CrossRef]
- 104. Pdtrier, C.; Francony, A. Ultrasonic waste-water treatment: Incidence of ultrasonic frequency on the rate of phenol and carbon tetrachloride degradation. *Ultrason. Sonochem.* **1997**, *4*, 295–300. [CrossRef]
- 105. Sakkas, P.M.; Schneider, O.; Sourkouni, G.; Argirusis, C. Sonochemistry in the service of SOFC research. *Ultrason. Sonochem.* **2014**, 21, 1939–1947. [CrossRef] [PubMed]
- Gogate, P.R. Cavitational reactors for process intensification of chemical processing applications: A critical review. *Chem. Eng. Process. Process Intensif.* 2008, 47, 515–527. [CrossRef]
- Vaitsis, C.; Sourkouni, G.; Argirusis, C. Metal Organic Frameworks (MOFs) and ultrasound: A review. *Ultrason. Sonochem.* 2018, 52, 106–119. [CrossRef]
- 108. Deng, Y.; Zhao, R. Advanced Oxidation Processes (AOPs) in Wastewater Treatment. Curr. Pollut. Rep. 2015, 1, 167–176. [CrossRef]
- Gagol, M.; Soltani, R.D.C.; Przyjazny, A.; Boczkaj, G. Effective degradation of sulfide ions and organic sulfides in cavitation-based advanced oxidation processes (AOPs). Ultrason. Sonochem. 2019, 58, 104610. [CrossRef] [PubMed]
- Okitsu, K.; Nanzai, B.; Kawasaki, K.; Takenaka, N.; Bandow, H. Sonochemical decomposition of organic acids in aqueous solution: Understanding of molecular behavior during cavitation by the analysis of a heterogeneous reaction kinetics model. *Ul-Trasonics Sonochemistry* 2009, *16*, 155–162. [CrossRef] [PubMed]
- 111. Suslick, K.S. The Chemical Effects of Ultrasound. Sci. Am. 1989, 260, 80-86. [CrossRef]
- 112. Suslick, K.; Didenko, Y.; Fang, M.M.; Hyeon, T.; Kolbeck, K.J.; McNamara, W.B.; Mdleleni, M.M.; Wong, M. Acoustic cavitation and its chemical consequences. *Philos. Trans. R. Soc. London. Ser. A Math. Phys. Eng. Sci.* **1999**, 357, 335–353. [CrossRef]
- Saharan, V.K.; Rizwani, M.A.; Malani, A.A.; Pandit, A.B. Effect of geometry of hydrodynamically cavitating device on degradation of orange-G. Ultrason. Sonochem. 2013, 20, 345–353. [CrossRef]
- 114. Bashir, T.A.; Soni, A.G.; Mahulkar, A.V.; Pandit, A.B. The CFD driven optimisation of a modified venturi for cavitational activity. *Can. J. Chem. Eng.* **2011**, *89*, 1366–1375. [CrossRef]
- 115. Madhavan, J.; Grieser, F.; Ashokkumar, M. Degradation of orange-G by advanced oxidation processes. *Ultrason. Sonochem.* **2010**, 17, 338–343. [CrossRef] [PubMed]
- Kanakaraju, D.; Glass, B.D.; Oelgemoeller, M. Advanced oxidation process-mediated removal of pharmaceuticals from water: A review. J. Environ. Manag. 2018, 219, 189–207. [CrossRef]
- 117. Jose, J.; Philip, L. Continuous flow pulsed power plasma reactor for the treatment of aqueous solution containing volatile organic compounds and real pharmaceutical wastewater. *J. Environ. Manag.* **2021**, *286*, 112202. [CrossRef] [PubMed]
- De Bel, E.; Janssen, C.; De Smet, S.; Van Langenhove, H.; Dewulf, J. Sonolysis of ciprofloxacin in aqueous solution: Influence of operational parameters. *Ultrason. Sonochem.* 2011, 18, 184–189. [CrossRef]
- Naddeo, V.; Landi, M.; Scannapieco, D.; Belgiorno, V. Sonochemical degradation of twenty-three emerging contaminants in urban wastewater. *Desalination Water Treat.* 2013, 51, 6601–6608. [CrossRef]
- Lianou, A.; Frontistis, Z.; Chatzisymeon, E.; Antonopoulou, M.; Konstantinou, I.; Mantzavinos, D. Sonochemical oxidation of piroxicam drug: Effect of key operating parameters and degradation pathways. J. Chem. Technol. Biotechnol. 2017, 93, 28–34. [CrossRef]
- 121. Ziylan-Yavas, A.; Ince, N.H. Single, simultaneous and sequential applications of ultrasonic frequencies for the elimination of ibuprofen in water. *Ultrason. Sonochem.* 2018, 40, 17–23. [CrossRef] [PubMed]
- 122. Serna-Galvis, E.A.; Silva-Agredo, J.; Giraldo-Aguirre, A.L.; Florez-Acosta, O.A.; Torres-Palma, R.A. High frequency ultrasound as a selective advanced oxidation process to remove penicillinic antibiotics and eliminate its antimicrobial activity from water. *Ultrason. Sonochem.* **2016**, *31*, 276–283. [CrossRef]
- Güyer, G.T.; Ince, N.H. Degradation of diclofenac in water by homogeneous and heterogeneous sonolysis. *Ultrason. Sonochem.* 2011, 18, 114–119. [CrossRef]
- Wang, J.; Wang, S. Reactive species in advanced oxidation processes: Formation, identification and reaction mechanism. *Chem. Eng. J.* 2020, 401, 126158. [CrossRef]
- 125. Ike, I.A.; Linden, K.G.; Orbell, J.D.; Duke, M. Critical review of the science and sustainability of persulphate advanced oxidation processes. *Chem. Eng. J.* 2018, 338, 651–669. [CrossRef]
- 126. Wei, Z.; Villamena, F.A.; Weavers, L.K. Kinetics and Mechanism of Ultrasonic Activation of Persulfate: An in Situ EPR Spin Trapping Study. *Environ. Sci. Technol.* 2017, *51*, 3410–3417. [CrossRef]
- Abu Amr, S.S.; Aziz, H.A. New treatment of stabilized leachate by ozone/Fenton in the advanced oxidation process. *Waste Manag.* 2012, 32, 1693–1698. [CrossRef]
- Adityosulindro, S.; Barthe, L.; González-Labrada, K.; Haza, U.J.J.; Delmas, H.; Julcour, C. Sonolysis and sono-Fenton oxidation for removal of ibuprofen in (waste)water. Ultrason. Sonochem. 2017, 39, 889–896. [CrossRef]
- 129. Hou, L.; Zhang, H.; Xue, X. Ultrasound enhanced heterogeneous activation of peroxydisulfate by magnetite catalyst for the degradation of tetracycline in water. *Sep. Purif. Technol.* **2012**, *84*, 147–152. [CrossRef]

- 130. Chandak, S.; Ghosh, P.K.; Gogate, P.R. Treatment of real pharmaceutical wastewater using different processes based on ultra-sound in combination with oxidants. *Process Saf. Environ. Prot.* 2020, 137, 149–157. [CrossRef]
- 131. Naddeo, V.; Uyguner-Demirel, C.S.; Prado, M.; Cesaro, A.; Belgiorno, V.; Ballesteros, F. Enhanced ozonation of selected pharmaceutical compounds by sonolysis. *Environ. Technol.* **2015**, *36*, 1876–1883. [CrossRef]
- Mendez-Arriaga, F.; Torres-Palma, R.A.; Petrier, C.; Esplugas, S.; Gimenez, J.; Pulgarin, C. Ultrasonic treatment of water contaminated with ibuprofen. *Water Res.* 2008, 42, 4243–4248. [CrossRef] [PubMed]
- 133. Kwon, M.; Kim, S.; Yoon, Y.; Jung, Y.; Hwang, T.-M.; Lee, J.; Kang, J.-W. Comparative evaluation of ibuprofen removal by UV/H<sub>2</sub>O<sub>2</sub> and UV/S<sub>2</sub>O<sub>8</sub><sup>2-</sup> processes for wastewater treatment. *Chem. Eng. J.* **2015**, 269, 379–390. [CrossRef]
- Tokumura, M.; Sugawara, A.; Raknuzzaman, M.; Mamun, H.-A.-; Masunaga, S. Comprehensive study on effects of water matrices on removal of pharmaceuticals by three different kinds of advanced oxidation processes. *Chemosphere* 2016, 159, 317–325. [CrossRef]
- 135. Maroudas, A.; Pandis, P.K.; Chatzopoulou, A.; Davellas, L.-R.; Sourkouni, G.; Argirusis, C. Synergetic decolorization of azo dyes using ultrasounds, photocatalysis and photo-fenton reaction. *Ultrason. Sonochem.* **2021**, *71*, 105367. [CrossRef] [PubMed]
- 136. Faouzi, M.; Cañizares, P.; Gadri, A.; Lobato, J.; Nasr, B.; Paz, R.; Rodrigo, M.; Saez, C. Advanced oxidation processes for the treatment of wastes polluted with azoic dyes. *Electrochim. Acta* **2006**, *52*, 325–331. [CrossRef]
- Voukkali, I.; Loizia, P.; Pedreño, J.N.; Zorpas, A.A. Urban strategies evaluation for waste management in coastal areas in the framework of area metabolism. *Waste Manag. Res. J. Int. Solid Wastes Public Clean. Assoc. ISWA* 2021, 39, 448–465. [CrossRef] [PubMed]
- 138. Zorpas, A.A.; Coumi, C.; Drtil, M.; Voukalli, I. Municipal sewage sludge characteristics and waste water treatment plant effectiveness under warm climate conditions. *Desalination Water Treat.* **2011**, *36*, 319–333. [CrossRef]
- 139. Zorpas, A.A.; Vlyssides, A.G.; Loizidou, M. Dewatered anaerobically-stabilized primary sewage sludge composting: Metal leachability and uptake by natural Clinoptilolite. *Commun. Soil Sci. Plant Anal.* **1999**, *30*, 1603–1613. [CrossRef]
- Zorpas, A.A.; Coumi, C.; Drtil, M.; Voukalli, I.; Samaras, P. Operation description and physicochemical characteristics of influent, effluent and the tertiary treatment from a sewage treatment plant of the Eastern Region of Cyprus under warm climates. *Desalination Water Treat.* 2010, 22, 244–257. [CrossRef]
- 141. Gagol, M.; Przyjazny, A.; Boczkaj, G. Highly effective degradation of selected groups of organic compounds by cavitation based AOPs under basic pH conditions. *Ultrason. Sonochem.* **2018**, *45*, 257–266. [CrossRef]
- Agarkoti, C.; Gogate, P.R.; Pandit, A.B. Coupling of acoustic/hydrodynamic cavitation with ozone (O<sub>3</sub>), hydrogen peroxide (H<sub>2</sub>O<sub>2</sub>), magnesium oxide (MgO) and manganese dioxide (MnO<sub>2</sub>) for the effective treatment of CETP effluent. *Sep. Purif. Technol.* 2021, 284, 120281. [CrossRef]
- Chen, L.; Li, H.; Qian, J. Degradation of roxarsone in UV-based advanced oxidation processes: A comparative study. J. Hazard. Mater. 2021, 410, 124558. [CrossRef] [PubMed]
- Cotton, C.; Dotson, A.; Jousset, S.; Linden, K.; Collins, J. Applying UVAOP at an existing WTP: Effects on disinfection strategy and DBP formation. In Proceedings of the Water Quality Technology Conference and Exposition, Savannah, Georgia, 14–18 November 2010; pp. 3008–3013.
- Li, W.; Zhang, M.; Wang, H.; Lian, J.; Qiang, Z. Removal of recalcitrant organics in reverse osmosis concentrate from coal chemical industry by UV/H2O2 and UV/PDS: Efficiency and kinetic modeling. *Chemosphere* 2021, 287, 131999. [CrossRef]
- 146. Meng, T.; Sun, W.; Su, X.; Sun, P. The optimal dose of oxidants in UV-based advanced oxidation processes with respect to primary radical concentrations. *Water Res.* 2021, 206, 117738. [CrossRef]
- 147. Tang, Y.; Lee, C.S.; Walker, H.; Gobler, C.; Apul, O.; Venkatesan, A.K.; Mao, X. Effect of residual H<sub>2</sub>O<sub>2</sub> on the removal of advanced oxidation byproducts by two types of granular activated carbon. *J. Environ. Chem. Eng.* **2021**, *9*, 106838. [CrossRef]
- Wang, H.; Hasani, M.; Wu, F.; Warriner, K. Pre-oxidation of spent lettuce wash water by continuous Advanced Oxidation Process to reduce chlorine demand and cross-contamination of pathogens during post-harvest washing. *Food Microbiol.* 2021, 103, 103937. [CrossRef]
- Ahmed, N.; Vione, D.; Rivoira, L.; Carena, L.; Castiglioni, M.; Bruzzoniti, M. A Review on the Degradation of Pollutants by Fenton-Like Systems Based on Zero-Valent Iron and Persulfate: Effects of Reduction Potentials, pH, and Anions Occurring in Waste Waters. *Molecule* 2021, 26, 4584. [CrossRef]
- 150. Ahmed, Y.; Zhong, J.; Yuan, Z.; Guo, J. Simultaneous removal of antibiotic resistant bacteria, antibiotic resistance genes, and micropollutants by a modified photo-Fenton process. *Water Res.* **2021**, *197*, 117075. [CrossRef]
- 151. Butt, A.L.; Mpinga, J.K.; Tichapondwa, S.M. Photo-Fenton Oxidation of Methyl Orange Dye Using South African Ilmenite Sands as a Catalyst. *Catalysts* **2021**, *11*, 1452. [CrossRef]
- 152. Gorozabel-Mendoza, M.L.; Filho, O.A.E.; Zambrano-Intriago, L.A.; Baquerizo-Crespo, R.J.; Giler-Molina, J.M.; Rodríguez-Díaz, J.M. Degradation of Blue 1 and Yellow 6 Dyes in Binary Mixture Using Photo-Fenton/Sunlight System: Optimization by Factorial Designs. *Water Air Soil Pollut.* 2021, 232, 500. [CrossRef]
- 153. Rodrigues-Silva, F.; Lemos, C.R.; Naico, A.A.; Fachi, M.M.; Amaral, B.D.; de Paula, V.C.; Rampon, D.S.; Beraldi-Magalhães, F.; Prola, L.D.; Pontarolo, R.; et al. Study of isoniazid degradation by Fenton and photo-Fenton processes, by-products analysis and toxicity evaluation. *J. Photochem. Photobiol. A Chem.* 2021, 425, 113671. [CrossRef]

- 154. Vieira, M.M.; Dornelas, A.S.P.; Carlos, T.D.; Pallini, A.; Gravato, C.; Pereira, D.H.; Sarmento, R.A.; Cavallini, G.S. When treatment increases the contaminant's ecotoxicity: A study of the Fenton process in the degradation of methylene blue. *Chemosphere* **2021**, 283, 131177. [CrossRef]
- 155. Widyastuti, N.; Hidayat, M.; Purnomo, C.W. Enhanced Biogas Production from Sugarcane Vinasse using Electro-Fenton as Pre-treatment Method. In *IOP Conference Series: Earth and Environmental Science;* IOP Publishing: Bristol, UK, 2021; Volume 830.
- 156. Brillas, E. A critical review on ibuprofen removal from synthetic waters, natural waters, and real wastewaters by advanced oxidation processes. *Chemosphere* **2021**, *286*, 131849. [CrossRef] [PubMed]
- 157. Dutschke, M.; Schnabel, T.; Schütz, F.; Springer, C. Degradation of chlorinated volatile organic compounds from contaminated ground water using a carrier-bound TiO<sub>2</sub>/UV/O<sub>3</sub>-system. *J. Environ. Manag.* **2021**, *304*, 114236. [CrossRef]
- 158. Gautam, P.; Popat, A.; Lokhandwala, S. Advances & Trends in Advance Oxidation Processes and Their Applications. In *Advanced Industrial Wastewater Treatment and Reclamation of Water*; Springer: Berlin/Heidelberg, Germany, 2022; pp. 45–69.
- 159. Vieira, W.T.; de Farias, M.B.; Spaolonzi, M.P.; da Silva, M.G.C.; Vieira, M.G.A. Latest advanced oxidative processes applied for the removal of endocrine disruptors from aqueous media—A critical report. *J. Environ. Chem. Eng.* **2021**, *9*, 105748. [CrossRef]
- Bolujoko, N.B.; Unuabonah, E.I.; Alfred, M.O.; Ogunlaja, A.; Ogunlaja, O.O.; Omorogie, M.O.; Olukanni, O.D. Toxicity and removal of parabens from water: A critical review. *Sci. Total Environ.* 2021, 792, 148092. [CrossRef]
- Cardoso, I.M.F.; Cardoso, R.M.F.; da Silva, J.C.G.E. Advanced Oxidation Processes Coupled with Nanomaterials for Water Treatment. *Nanomaterial* 2021, 11, 2045. [CrossRef]
- 162. Fosso-Kankeu, E.; Pandey, S.; Ray, S.S. *Photocatalysts in Advanced Oxidation Processes for Wastewater Treatment;* Springer: Berlin/Heidelberg, Germany, 2020.
- 163. Evgenidou, E.; Ofrydopoulou, A.; Malesic-Eleftheriadou, N.; Nannou, C.; Ainali, N.M.; Christodoulou, E.; Bikiaris, D.N.; Kyzas, G.Z.; Lambropoulou, D.A. New insights into transformation pathways of a mixture of cytostatic drugs using Polyester-TiO<sub>2</sub> films: Identification of intermediates and toxicity assessment. *Sci. Total Environ.* 2020, 741, 140394. [CrossRef]
- Kusuma, T.D.; Jyothi, M.S.V.N.; Rao, C.P.; Maliyekkal, S.M. Advanced Oxidation Processes: A Promising Route for Abatement of Emerging Contaminants in Water. In *Nanomaterials and Nanocomposites for Environmental Remediation*; Springer: Berlin/Heidelberg, Germany, 2021; pp. 275–305.
- 165. Liu, H.; Li, C.; Zhang, T.; Xu, Z.; Li, Y.; Li, B.; Tian, S. UV facilitated synergistic effects of polymetals in ore catalyst on peroxy-monosulfate activation: Implication for the degradation of bisphenol S. *Chem. Eng. J.* **2022**, 431, 133989. [CrossRef]
- 166. Kaus, N.H.M.; Imam, S.S.; Aziz, A.W.; Lee, H.L.; Adnan, R.; Ibrahim, M.L. Controlled growth of BiFeO<sub>3</sub> nano-particles in the presence of alginate template for adsorptive removal of different dyes. *Colloids Surf. A Physicochem. Eng. Asp.* 2021, 615, 126294. [CrossRef]
- 167. Kaus, N.H.M.; Rithwan, A.F.; Adnan, R.; Ibrahim, M.L.; Thongmee, S.; Yusoff, S.F.M. Effective Strategies, Mechanisms, and Photocatalytic Efficiency of Semiconductor Nanomaterials Incorporating rGO for Environmental Contaminant Degradation. *Catalysts* 2021, 11, 302. [CrossRef]
- Zare, E.N.; Iftekhar, S.; Park, Y.; Joseph, J.; Srivastava, V.; Khan, M.A.; Makvandi, P.; Sillanpaa, M.; Varma, R.S. An overview on non-spherical semiconductors for heterogeneous photocatalytic degradation of organic water contaminants. *Chemosphere* 2021, 280, 130907. [CrossRef]
- Liu, P.; Wu, Z.; Abramova, A.V.; Cravotto, G. Sonochemical processes for the degradation of antibiotics in aqueous solutions: A review. Ultrason. Sonochem. 2021, 74, 105566. [CrossRef] [PubMed]
- 170. Luo, T.; Xu, J.; Li, J.; Wu, F.; Zhou, D. Strengthening arsenite oxidation in water using metal-free ultrasonic activation of sulfite. *Chemosphere* **2021**, *281*, 130860. [CrossRef] [PubMed]
- 171. Poblete, R.; Cortes, E.; Salihoglu, G.; Salihoglu, N.K. Ultrasound and heterogeneous photocatalysis for the treatment of vinasse from pisco production. *Ultrason. Sonochem.* **2020**, *61*, 104825. [CrossRef] [PubMed]
- 172. Rayaroth, M.P.; Aravindakumar, C.T.; Shah, N.S.; Boczkaj, G. Advanced oxidation processes (AOPs) based wastewater treatment— Unexpected nitration side reactions—A serious environmental issue: A review. *Chem. Eng. J.* **2022**, 430, 133002. [CrossRef]
- 173. Rossi, G.; Mainardis, M.; Aneggi, E.; Weavers, L.K.; Goi, D. Combined ultrasound-ozone treatment for reutilization of primary effluent—A preliminary study. *Environ. Sci. Pollut. Res.* **2021**, *28*, 700–710. [CrossRef]
- 174. Sourkouni, G.; Kalogirou, C.; Moritz, P.; Gödde, A.; Pandis, P.K.; Höfft, O.; Vouyiouka, S.; Zorpas, A.A.; Argirusis, C. Study on the influence of advanced treatment processes on the surface properties of polylactic acid for a bio-based circular economy for plastics. *Ultrason. Sonochem.* 2021, 76, 105627. [CrossRef] [PubMed]
- 175. Sunasee, S.; Leong, K.H.; Wong, K.T.; Lee, G.; Pichiah, S.; Nah, I.W.; Jeon, B.H.; Yoon, Y.; Jang, M. Sonophotocatalytic degradation of bisphenol A and its intermediates with graphitic carbon nitride. *Environ. Sci. Pollut. Res.* 2019, 26, 1082–1093. [CrossRef] [PubMed]
- Wang, J.; Wang, S. Toxicity changes of wastewater during various advanced oxidation processes treatment: An overview. J. Clean. Prod. 2021, 315, 128202. [CrossRef]