

Review

Photoelectrochemical Degradation of Contaminants of Emerging Concern with Special Attention on the Removal of Acetaminophen in Water-Based Solutions

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Abstract: Despite being an indispensable medium for life and the environment, the deterioration of water quality continues to be a matter of great concern for the scientific community since this problem has generated the need to develop processes for water treatment. Among the wide variety of types of contaminants, the presence of contaminants of emerging concern (CECs) has become an issue of global concern, since they are present in water in low concentrations and can persist in the environment, bioaccumulate and be toxic to human health and aquatic biota. Among CECs, this review focuses on pharmaceutical removal; what favorable effects they have on human and animal health are widely recognized, but the consequences or risks associated with the constant disposal of these products to the environment, through the manufacturing process, consumption, biological excretion or inappropriate disposal, are an area under constant study. The presence of pharmaceuticals in water represents a high risk to the environment because they contain active ingredients that were designed to induce specific pharmacological effects but, when dissolved in water, reach non-target populations and cause undesirable toxicological effects. This review pays attention to the photoelectrochemical removal of paracetamol (PTM), as a model molecule, from water, employing different photoactive materials (TiO₂, BiVO₄, ZnO and tungsten oxides) and the main parameters affecting their performance. The main goal of the present review is to facilitate future researchers to design their experiments concerning the PEC processes for the degradation of pharmaceuticals, specially PTM, and the existing limitations of each system.

Keywords: photoelectrochemistry; photoelectrocatalyst; photocatalysts; photoelectrocatalysis; acetaminophen; paracetamol; pharmaceutical removal

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1. Introduction

The availability of clean water remains a major concern owing to the rise in the water pollution rate. While improper disposal of household effluents and some natural phenomena contributes to water pollution, rapid industrialization is mostly responsible for the recent increase in water pollution, as many effluents from industries are not often properly treated before being discharged into water bodies, or through indiscriminate use of fertilizers during agricultural activity [1]. In particular, the presence of micropollutants, also known as contaminants of emerging concern (CECs), has become an issue of global concern [2]. Indeed, CECs can persist in the environment, bioaccumulate and be toxic to human health and aquatic biota [3,4]. In the last decade, more than 1000 substances (pharmaceuticals, personal care products, steroid hormones and agrochemicals) have been identified as CECs at low concentrations in water [5,6]. In this context, their detection, analysis and, principally, their removal from drinking water and the improvement in

wastewater treatment plants are of great interest. Therefore, the main output of the current review is to facilitate future researchers to design their experiments concerning the photoelectrochemical processes for the degradation of pharmaceutical products.

1.1. Pharmaceutical Pollution

Among CECs, this review focuses on pharmaceuticals that are used therapeutically for human and animal diseases and whose number and variety have increased in the last decades due to advances in medical science and pharmacology [7]. The favorable effects they have on human and animal health, food production and economic welfare are widely recognized. However, the consequences or risks associated with the constant disposal of these products to the environment, through the manufacturing process, consumption, biological excretion or inappropriate disposal, are an area under constant study [8,9]. The presence of pharmaceuticals in water represents a high risk to the environment because they contain active ingredients that were designed to induce specific pharmacological effects but, when dissolved in water, reach non-target populations and cause undesirable toxicological effects [10]. On the other hand, the persistence of these CECs is one of the main concerns for the scientific community since this implies that, even if their use and indiscriminate elimination cease, the concentrations disseminated in the environment will not decrease in the short term, facilitating their interaction with the ecosystem and the flora and fauna that form part of it [11].

The most frequently detected drugs in groundwater are anti-inflammatory drugs, such as diclofenac, paracetamol (PTM) and ibuprofen, due to their high consumption [12]. Chronic exposure to these compounds has been reported to produce toxic effects on biota, such as gill alterations in fish and kidney failure in birds [13]. Another group of widely disseminated CECs is antibiotics [14]. The worldwide consumption of antibiotics is estimated to be between 100,000 and 200,000 tons per year. Therefore, the presence of antibiotics in municipal wastewater, sewage treatment plant effluents and ground and surface water in $\mu\text{g/L}$ amounts (below therapeutic levels), which could favor the emergence of resistant bacteria, is not surprising [15]. In this context, pharmaceutical compounds have been identified as an important group of water pollutants, and, thus, their removal is necessary to avoid the risk of exposure to them. Unfortunately, as these compounds are not completely removed by wastewater treatment plants, extensive contamination of water bodies occurs. Consequently, pharmaceutical pollution supposes a global hazard to environmental and human health, in addition to the delivery of the United Nations Sustainable Development Goals (SDGs). A recent study [16], which examined different rivers around the world, found that the sites most contaminated with active pharmaceutical ingredients (APIs) were those in low- and middle-income countries and were associated with areas of poor wastewater and waste management infrastructure and pharmaceutical manufacturing. The most frequently detected APIs were carbamazepine, metformin and caffeine, which were detected in more than half of the sites monitored. Regarding pharmaceutical frequencies and concentration, four APIs were detected across all continents: caffeine, nicotine, acetaminophen/paracetamol and cotinine, a nicotine metabolite [16].

1.2. Pharmaceutical Removal Techniques

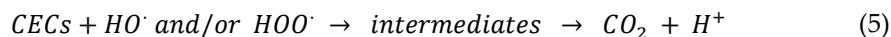
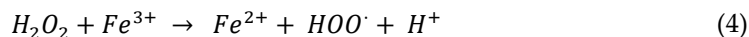
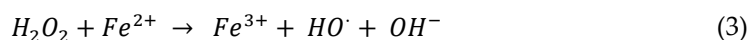
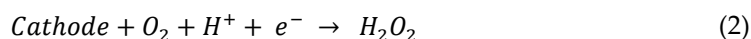
In general terms, the methods for removing pharmaceuticals from water are classified into conventional and advanced oxidation processes (AOPs) [17]. Conventional wastewater treatment includes biological and physicochemical mechanisms [18,19]. However, their potential to remove pharmaceuticals is restricted since these pollutants cannot be digested as a carbon source by microorganisms [20], and the use of chemical treatment, although it improved the efficiency of drug elimination, on a pilot scale, it is necessary to apply upstream and downstream processes that make treatment more expensive [21]. Moreover, conventional methods present certain drawbacks that limit their applicability levels: physical methods could not remove CECs at low concentrations, some chemical techniques have low capacity rates and biological methods usually have long treatment

periods [22]. Although the regulation of CECs in municipal wastewater effluents is still under debate worldwide, some countries, such as Switzerland and Germany, are implementing advanced treatments to remove CECs from wastewater treatment plant effluents (WWTPs) [23]. In fact, secondary biological treatments applied in municipal WWTPs have been shown to provide poor removal for most CECs [23].

Among the newer degradation methods are AOPs, both homogeneous and heterogeneous photocatalytic and catalytic AOPs [24]. The latter have not yet been applied on a large scale, probably because a cost-effective and efficient technology has not yet been achieved. AOPs are processes in which radicals, highly oxidizing species, are generated, which, in sufficient quantities, cause the mineralization of organic matter to carbon dioxide, water and inorganic ions [25]. Their high efficiency is related to the high production of radicals, resulting from the use of a combination of oxidizing agents, radiation (ultraviolet or ultrasonic) and catalysts [26]. AOPs are relatively efficient, promising and suitable for pharmaceutical degradation [27,28]. Additionally, AOPs are included in many operating wastewater treatment trains or are suggested for different reuse alternatives [29]. While different process technologies have already been established at full scale for use as AOPs, several researchers are constantly conducting new studies on numerous emerging AOPs for water treatment (e.g., electrochemical AOPs, plasma, ultrasound, electron beam and microwave). Thus, there are many diverse technologies, involving very different methods of oxidant generation and potentially using a number of different mechanisms for CEC removal. Most of these processes are based on electrical power and pose a huge challenge for the critical assessment of AOPs in terms of their operational costs, sustainability and overall feasibility.

Among the AOPs, electrochemical advanced oxidation processes (EAOPs), including electrocatalytic (EC) and photoelectrocatalytic (PEC) oxidation, are the most important wastewater treatments [22]. EAOPs use electro-catalytically produced hydroxyl radicals for the mineralization of CECs, linking the advantage of hydroxyl radical production and the efficiency of EC.

Electrocatalytic (EC) methods involve the electrooxidation of CECs at the anode (Equation (1)) and the degradation of pollutants by peroxide that is produced by oxygen reduction at the cathode (Equation (2)), which, in the presence of certain metals (e.g., Fe), may produce oxygen-radical species (Equations (3) and (4)) that are powerful oxidants (Equation (5)) [17]:

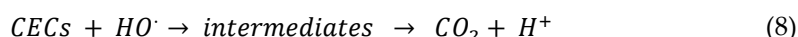
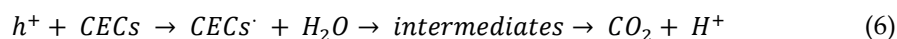


It is important to note that the above reactions are not balanced.

The degradation capability depends on the nature of the anode and the cathode along with operational variables, such as operating current density/voltage, temperature, substrate concentration, stirring and pH, among others [30]. Electrode materials should provide high electrical conductivity, good mechanical properties, strong chemical resistance and elevated electrocatalytic activity towards the water oxidation reaction at the anode and peroxide formation at the cathode, among others [31,32].

The photocatalytic (PC) approach offers the possibility of using naturally available and renewable solar energy as a potential energy source in the presence of a suitable photocatalyst prepared from semiconductors (e.g., transition metal oxides) as a catalyst [33]. When illuminated with a photon of energy higher than the band gap energy, these semiconductors form an electron–hole pair (e^-/h^+). These electron–hole pairs are potent redox species that utilize many organic photodegradation reactions, either directly or indirectly,

through the formation of hydroxyl radicals in solution [30]. In the context of the current manuscript, holes may react with CECs and/or water, creating a radical intermediate that undergoes further reaction to carbon dioxide (Equations (6)–(8)). In addition, if dioxygen is dissolved in solution, it may react with the photogenerated electrons to produce superoxide anion (O_2^-). The last species may produce peroxide or react with CECs. Furthermore, peroxide may oxidize CECs or scavenge other photogenerated electrons to generate $HO\cdot$ and OH^- species, and consequently degrade the pollutants according to Equation (5).

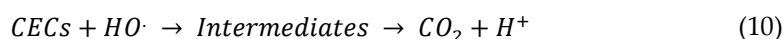


It is important to note that the above reactions are not balanced.

Unfortunately, the photocatalytic activity of most semiconductors is still low due to the lack of enough active sites, large band gap, fast recombination of the generated electron/hole pairs and, of course, low electrical conductivity. Most common heterogeneous catalysts are transition metal oxides, TiO_2 being the most researched due to its excellent chemical and physical stability, low cost, nontoxicity to humans and high photocatalytic activity, but its large band gap makes it operational under high energy input, i.e., UV radiation [34].

Photoelectrochemical (PEC) removal of organic matter present in water is mainly based on the use of semiconductors, as photoactive materials, immobilized on different electrical conductive supports [35]. Particularly, photoelectrocatalysis, consisting of a combination of both photochemical and electrochemical processes aiming to retard and/or avoid the fast recombination of the generated electron/hole pairs during the process, has become a powerful AOP for CEC removal [36]. Furthermore, the anodic overpotential needed in a typical PEC cell is lower compared to what is required for the electrochemical degradation of organics.

In typical PEC processes, the conductive interface is used to immobilize a photocatalyst, and this assembly is called a photoelectrode (PE). When irradiated with UV or a visible light source with a range equal to or greater than the bandgap of the semiconductor material used, the electrons in the valence band (e_{vb}) promoted to the conduction band (e_{cb}) generate electron/holes (e^-/h^+), and from the application of a positive external bias, the photoexcited electrons are transferred to the cathode (in this way, the use of electron scavenger may be avoided and the recombination of the e^-/h^+ pair may be diminished) and the holes become available on the PE surface for the generation of radicals (Equations (6) and (7)) with high oxidative power or directly participating in the degradation of the pollutant due to the formation of radicals in the presence of light at the cathode (Equations (1)–(5) and Equations (9) and (10)) [37,38].



It is important to note that the above reactions are not balanced.

Semiconductor materials (SCMs), such as titania (TiO_2), zinc oxide (ZnO), copper (I and II) oxide [39], bismuth vanadate ($BiVO_4$) [40] and tungsten oxide (WO_3) [41], have been applied as electrodes in PEC processes in water treatment. However, they reveal certain drawbacks, such as instability during the PEC reaction, photo-corrosion, high recombination of photogenerated charges, high electrical resistance and the absorption of light in a small range of the spectrum, such is the case of ZnO and TiO_2 that only absorb in the UV light region. In addition, it should be taken into account that the immobilization of the semiconductor on the surface of the conductive material would produce a decrease in the EC efficiency due to an increase in the high electrical resistance provided by the photoactive material. Therefore, the development of strategies to solve these problems is

necessary to improve the PEC properties of the materials to be applied as photoanodes. In this sense, it was reported that the addition of co-catalysts, doping and the heterojunction formation between semiconductors may decrease the recombination of photogenerated charges and could improve the formation of oxidizing species [42].

Figure 1 shows a simplified PEC cell used for organic pollutant degradation [38]. Any semiconductor (SMC) to be used as a photoanode (or photoelectrode) should be expected to have an adequate band gap and exhibit photochemical stability and acceptable electrical conductivity in aqueous media. In addition, this material should provide a high quantum yield and a considerable adsorption capacity for the organic contaminant.

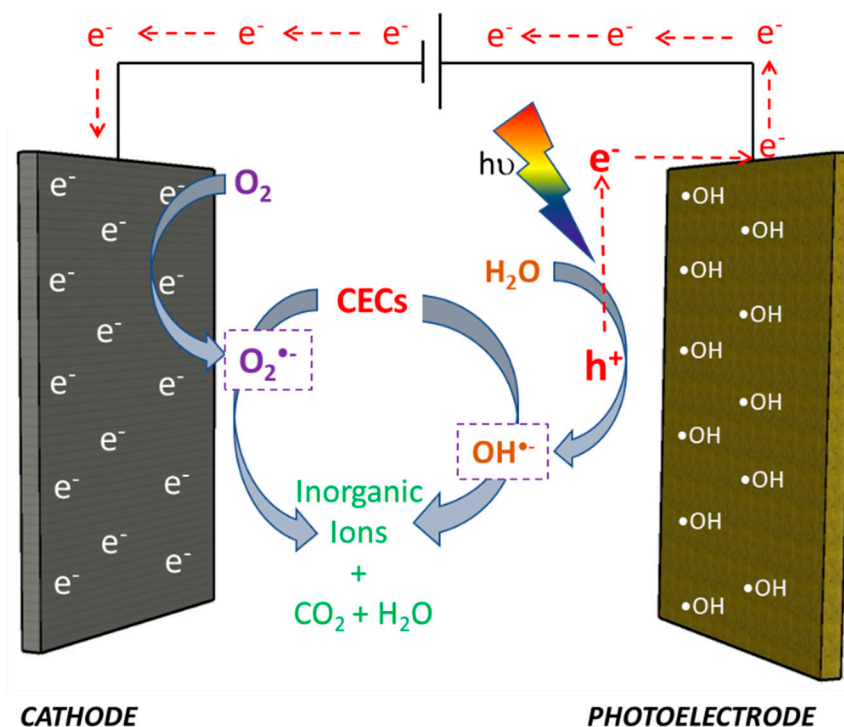


Figure 1. Schematic representation of the PEC process.

1.3. Photoelectrodes for PEC Treatment of Pharmaceuticals

As mentioned above, in a study conducted in different lakes around the world, it was seen that some of the APIs found with higher frequency and concentration were caffeine (stimulant and lifestyle compound), acetaminophen/paracetamol (analgesic), atenolol (β -blocker) and carbamazepine (antiepileptic), among others [16]. In this sense, and taking into account the lack of studies on the subject, it is of the utmost importance to develop technologies that allow for the elimination of these compounds until their mineralization (pharmaceuticals \rightarrow CO_2), using techniques such as photoelectrocatalysis, to avoid the deterioration of aquatic environments. In this section, we describe and discuss the main results reported for the removal of PTM by PEC, PC and EC oxidation and the main parameters that affect the performance of the technique, classifying them according to the photoanode used during the oxidation process.

Acetaminophen/paracetamol is a molecule composed of an aromatic, an amide and a hydroxyl group, as described in Figure 2. Among thousands of pharmaceuticals found in water, wastewater and drinking water, paracetamol is one of the most reported. Different treatments were reported in acetaminophen removal from water, including biological [43], $\text{H}_2\text{O}_2/\text{UV}$ oxidation [44], sonolysis [45], TiO_2 photocatalysis [46], LED/ TiO_2 , Fenton [47], photo-Fenton [48], reverse osmosis [49], activated carbon [50] and ozonation [51].

However, in most of these treatments, only partial mineralization was obtained and reaction times were long. PTM was selected as a model molecule since its structure is the main component or metabolite of several anesthetic and analgesic compounds.

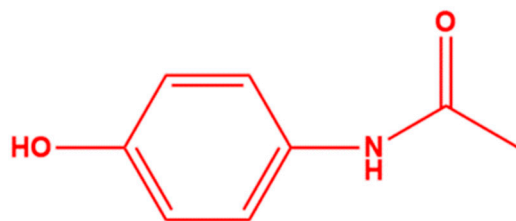


Figure 2. Paracetamol (PTM) molecule.

Some research [52,53] reported the use of the PEC process to remove PTM from water and found that the modification of certain operating variables or intrinsic properties of the electrodes produced significant effects on the performance and efficiency of the PEC oxidation of this drug. In this sense, it is important to take into account that the efficiency of the PEC process will depend both on the nature and treatment of the electrodes, as well as on a series of variables inherent to the technique. Therefore, to develop new technologies that are efficient in the removal of pharmaceuticals from water, it is necessary to understand how the different variables affect the process. Thus, this review gathers information about the main variables that affect the performance of different photoelectrodes in the removal of pharmaceuticals from water using photoelectrocatalysis as a remediation method. The main goal of the present review paper is to facilitate future researchers to design their experiments concerning the PEC processes for the degradation of pharmaceuticals, especially, PTM, as well as the existing limitations of each system.

1.3.1. TiO₂-Based Photoelectrodes

Among the metal oxide semiconductors, TiO₂ is one of the most attractive photocatalytic materials due to its high stability in a wide pH range and robustness to various irradiation wavelengths, high efficiency and highly oxidizing photo-generated holes [54]. TiO₂ presents a wide band gap (~3.2 eV) [55], and due to this and its optical absorption in the UV region, many efforts have been focused on improving the PEC activity of this material. Eid et al. [56] reported that the formation of defects, such as monovacancies, vacancy clusters, and a few voids inside a mixture of Ti-nitride and oxynitride, decreased the bandgap energy to 2.4 eV, increased visible-light response and enhanced the incident photon-to-current collection efficiency (IPCE) and the photocurrent density. Eid et al. [57] also observed that the functionalization of titanium oxynitride nanotubes with iridium nanoparticles entails abundant defects, such as small nitrogen vacancies, larger-size vacancy clusters and small voids, increasing the photocurrent density with respect to unmodified material.

Titania in its crystalline anatase phase is the most used catalyst for PEC water decontamination, owing to its low toxicity, band gap near the visible range and excellent chemicals and photochemical stability [58], and it is generally used either as a thin film or a nanostructure.

Xie et al. [53] studied the degradation of PTM by EC, PC and PEC processes in a three-electrode cell using a nanoporous TiO₂ electrode that was grown directly on titanium plates for 3 h. The best results were achieved by the PEC technique. Indeed, PTM was rapidly eliminated by PEC degradation with UV-visible light irradiation, although total removal of PTM was not obtained when separate PC and EC methods were used. The highest efficiency of the PEC method in PTM removal can be attributed to the synergistic effect of combining both light and electrical energy. The photoanode absorbs pho-

tons and photo-excited electron–hole pairs are generated, which can either directly oxidize the PTM molecules or produce hydroxyl radicals, after reacting with water, then attacking the PTM molecule. On the other hand, the application of current assists in the driving of electrons from the photoanode, thus inhibiting the spontaneous recombination of electrons and holes.

Table 1 summarizes the main results of acetaminophen degradation using EC and PEC techniques [53]. The performance of the untreated electrode was compared to that of an electrochemically treated electrode (EC reduction treatment was conducted on the nanoporous TiO₂ in 0.1 mol·L⁻¹ H₂SO₄ at a cathodic current of -5 mA/cm² for 10 min) in the PEC degradation of PTM (see Figure 3). The electrochemically treated electrode enhanced the efficacy of PEC degradation by 87% due to the formation of Ti³⁺, Ti²⁺ and oxygen vacancies as well as an enhancement in the electrical conductivity during the EC reduction process. However, this behavior was not observed when PC (under UV-Vis irradiation, but without an applied electrode potential) or EC (at an applied electrode potential of 1.0 V vs. Ag/AgCl, but in the absence of UV-Vis light irradiation) were carried out, whether or not the treated or untreated electrode was used. In addition, the effect of the temperature was evaluated by varying it from 0 °C to 60 °C. The rise in temperature increased the rate constant of PTM degradation by PEC treatment, indicating that this parameter can significantly affect PTM removal.

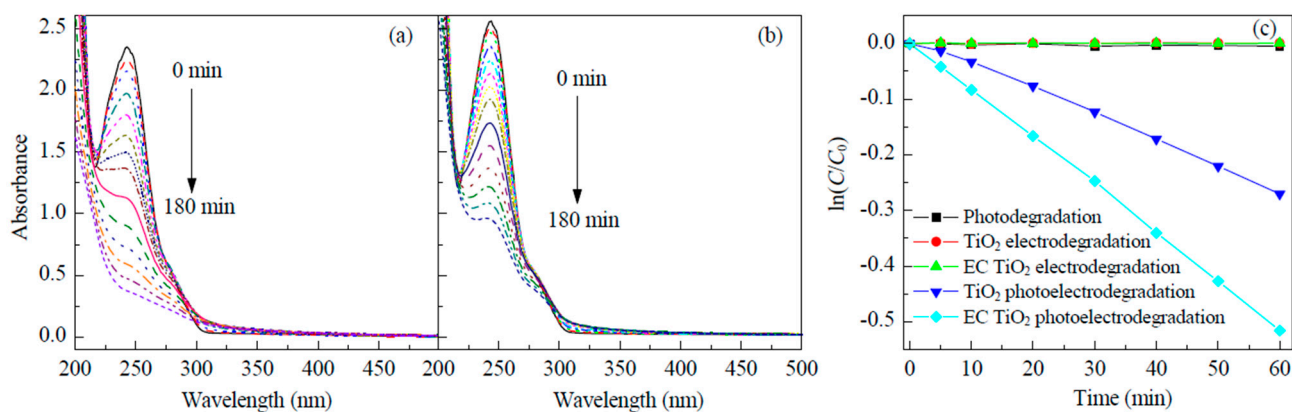


Figure 3. Absorbance spectra for the photoelectrochemical degradation of 40 mg·L⁻¹ of acetaminophen in 0.1 mol/L Na₂SO₄ at treated (a) and untreated (b) TiO₂ nanotube electrode with a potential of 1 V. (c) Plot of ln(C/C₀) versus time of the degradation of acetaminophen by different means. Reproduced with permission [53].

Danish et al. [59] studied the PTM degradation under PC and PEC processes using TiO₂, ZnS QDs, Fe@ZnS QDs and Fe@ZnS QDs/TiO₂ nanocomposites, comprising titania, iron and zinc sulfide quantum dots. PC evaluation was carried out both under UV-light and Vis-light sources in the presence of atmospheric oxygen. PEC evaluation was assessed in a three-electrode quartz cell assembly in a 0.1 M Na₂SO₄ aqueous solution. The working electrode was prepared by drop-casting the photocatalysts on a Fluorine-doped Tin Oxide (FTO) substrate. A saturated calomel and a Pt wire were used as reference and counter electrodes, respectively.

Table 1. PEC acetaminophen degradation using TiO₂-based photoelectrodes.

Photoelectrode	Conditions		Rate Constant (min ⁻¹)	Degradation Time	Removal Efficiency (%)	TOC Removal (%)	Ref.
TiO ₂ /Ti plate	0.1 mol·L ⁻¹ Na ₂ SO ₄	20 °C	0.0046		-	-	
	40 mg·L ⁻¹ PTM solution	0 °C	0.0065		-	-	
EC Treated-TiO ₂ /Ti plate	UV-Vis	20 °C	0.0086		79.72	59.3	[53]
	130 mW/cm ²	40 °C	0.0116		-	-	
TiO ₂ /Au/TiO ₂	1.0 V vs. Ag/AgCl	60 °C	0.0137		-	-	
		39 mg·L ⁻¹ PTM solution	-	300 min	47	-	
	EC oxidation	78 mg·L ⁻¹ PTM solution	-	300 min	37	-	
	0.05 mol·L ⁻¹ Na ₂ SO ₄	157 mg·L ⁻¹ PTM solution	-	300 min	29	-	
		39 mg·L ⁻¹ PTM solution	-	360 min	95	-	
	PEC oxidation	78 mg·L ⁻¹ PTM solution	-	360 min	69	-	
	0.05 mol·L ⁻¹ Na ₂ SO ₄	157 mg·L ⁻¹ PTM solution	-	360 min	61	-	
	36 W LED UVA, 88 W·m ⁻²	39 mg·L ⁻¹ PTM solution	-	120 min	100	-	[58]
		0.035 mol·L ⁻¹ Na ₂ SO ₄	-	150 min	100	-	
		0.015 mol·L ⁻¹ NaCl	157 mg·L ⁻¹ PTM solution	-	150 min	50	-
	36 W LED UVA, 88 W·m ⁻²	39 mg·L ⁻¹ PTM solution	-	3–4 min	100	-	
	PEC + PEF oxidation	Continuous H ₂ O ₂ generation					

The order of PC performances in the PTM degradation under Vis-light radiation was Fe@ZnSQDs/TiO₂ > TiO₂ > Fe@ZnSQDs > ZnSQDs (see Figure 4). It could be seen that the degradation of paracetamol was insignificant in the absence of irradiation, which demonstrate that this compound is quite stable and could not be degraded under direct photolysis. The authors also evaluated the degradation of PTM under analogous conditions using a UV light source with Fe@ZnSQDs/TiO₂ and ZnSQDs catalysts, finding an improved efficiency for the Fe@ZnSQDs/TiO₂ catalyst, which was attributed to better UV-light absorbing ability and efficient charge carrier separation (due to the generation of heterojunction between ZnSQDs and TiO₂ in the composite material) [59].

Regarding the PEC performances in PTM removal, the order was Fe@ZnSQDs/TiO₂ > Fe@ZnSQDs > ZnSQDs > TiO₂. This variation in catalytic performance with respect to PC assays was correlated with optical properties, electrochemical impedance and generation of hydroxyls and superoxide radicals. Concerning PTM mineralization, the change in TOC content was evaluated for PC processes using Fe@ZnSQDs/TiO₂ nanocomposite. After 40 min of illumination under a UV-light source, 61% of mineralization was found in the PC process [59].

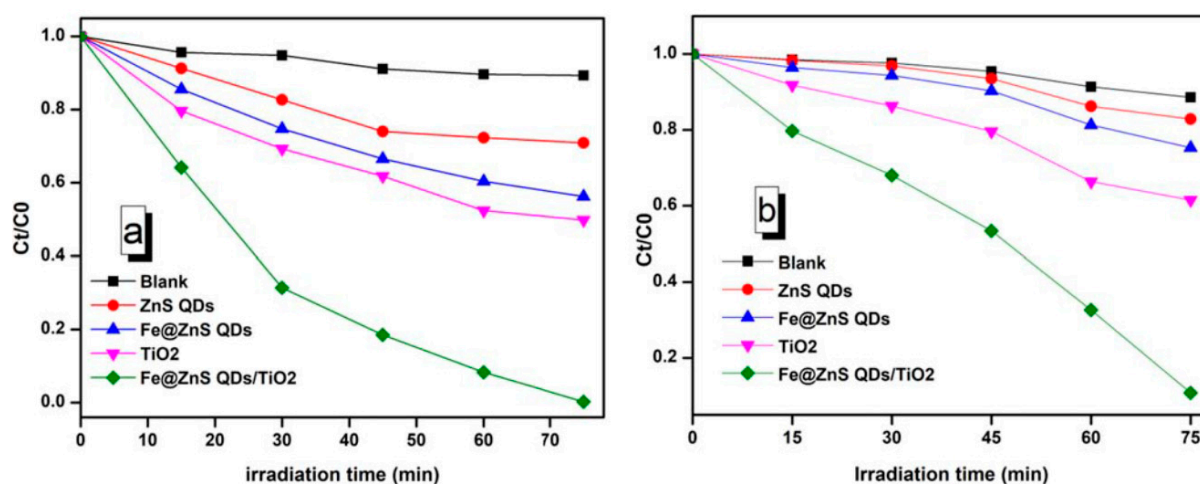


Figure 4. Change in concentration of Rhodamine B (RhB) (a) and paracetamol (b) as a function of time in the presence of different catalysts under visible light illumination. Reproduced with permission [59].

Rodríguez et al. [58] studied the degradation of PTM employing the PEC process and compared the removal efficiency with a hybrid process involving PEC + photo-electro Fenton (PEF) processes with the air-diffusion cathode (Figure 5). A thin-film sandwich-type composite of $\text{TiO}_2/\text{Au}/\text{TiO}_2$ (band gap: 3.22 eV vs. 3.44 eV for TiO_2 film) was used as a photoanode and was combined with a 316 L stainless-steel plate (SSP) cathode in a sulfate medium in the absence and presence of chloride ions. EC and PEC oxidation processes were carried out in galvanostatic mode and the effect of the applied current and PTM initial concentration (C_0) was evaluated in the PEC process. In the hybrid process (PEC + PEF), the cathode (stainless-steel plate) was replaced with a carbon-polytetrafluoroethylene (C-PTFE) air-diffusion cathode and fed with air for continuous H_2O_2 generation in a 0.50 mM Fe_2SO_4 solution.

For EC experiments (Figure 5A), until 300 min of treatment, PTM removal percentages of 47%, 37% and 29% were obtained for initial drug concentrations of 39 $\text{mg}\cdot\text{L}^{-1}$, 78 $\text{mg}\cdot\text{L}^{-1}$ and 157 $\text{mg}\cdot\text{L}^{-1}$, respectively (Table 2). While the normalized percentage of drug removed decreases with increasing initial drug concentration, the total content of degraded PTM increased for the same EC process conditions, which was attributed to the enhancement in the quantity of available $\cdot\text{OH}$ due to the deceleration of the parasitic reactions of this radical (oxidation to O_2). The same behavior was observed for PEC treatment (Figure 5B), but in this case, the large number of additional hydroxyl radicals formed under UVA irradiation resulted in a faster degradation as compared to EC. Further, 95%, 69% and 61% PTM was degraded at the end of the PEC treatment of 39 $\text{mg}\cdot\text{L}^{-1}$, 78 $\text{mg}\cdot\text{L}^{-1}$ and 157 $\text{mg}\cdot\text{L}^{-1}$, respectively. Regarding the influence of Cl^- ions (Figure 5C), the PTM decayed much more rapidly in the presence of chloride, reaching total removal at 120 min and 150 min for 39 $\text{mg}\cdot\text{L}^{-1}$ and 78 $\text{mg}\cdot\text{L}^{-1}$, respectively, but only completing 50% for the most concentrated solution. This behavior was ascribed to the quicker destruction of PTM by active chlorine species formed from oxidation of Cl^- to $\text{Cl}_2(\text{dis})$ at the photoanode, followed by hydrolysis to HClO (strongest active chlorine species at the treatment pH). However, it is well known that hydroxyl radicals may be scavenged by chlorine ions, and this partial destruction of generated radicals could explain the inhibition of PTM removal at 157 $\text{mg}\cdot\text{L}^{-1}$ in the presence of chlorine [58].

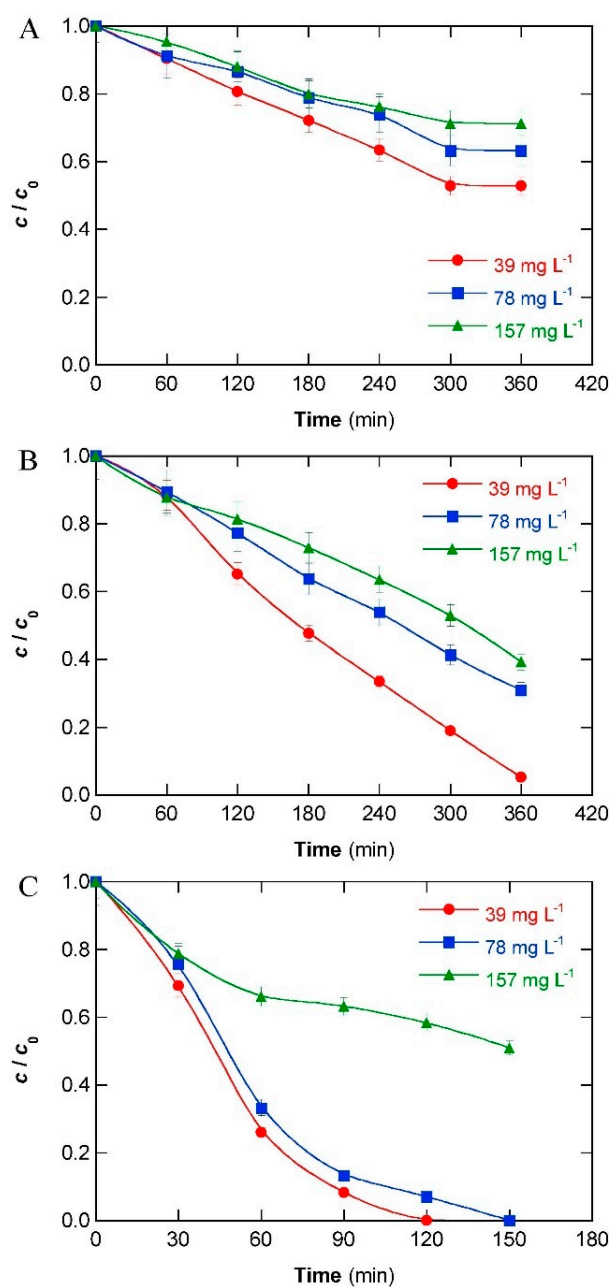


Figure 5. Normalized paracetamol concentration decay vs. electrolysis time for (A) EC, (B) PEC and (C) PEC + Cl⁻. Reproduced with permission [58].

In the hybrid process (PEC + PEF), total PTM removal was obtained after 4–5 min of treatment, a time much shorter than by single PEC. This result can be explained by the fast destruction of the molecule with the high amounts of $\cdot\text{OH}$ formed from Fenton's reaction in the bulk, superior to the action of the other oxidizing agents formed at the photoanode [58].

1.3.2. Bismuth Vanadate (BiVO₄)-Based Photoelectrodes

BiVO₄, an n-type semiconductor, like TiO₂, is one of the most promising photoanode materials. It has a band gap of 2.4 eV and is composed of inexpensive materials. BiVO₄ presents the valence band at around 2.4 V vs. RHE provides sufficient overpotential for holes to photo-oxidize water, a good visible light responsive activity. Bismuth vanadate has been widely applied in visible light in PC and PEC water treatment applications due

to its attractive properties, such as low toxicity, excellent physicochemical stability and narrow band gap [60].

Unfortunately, the performance of pristine bismuth vanadate is hampered by the relatively fast recombination of photo-excited electron–hole pairs as well as the poor transport of charge carriers. Therefore, doping with metallic and/or non-metallic impurities, formation of heterojunctions with other semiconductors and morphology control have been applied to overcome this drawback. It was reported that the heterojunction between p-type and n-type semiconductors can improve PEC performance by improving the effective separation of photogenerated electron–hole pairs and light harvesting [60].

Another bismuth-based semiconductor is BiOI (p-type semiconductor), with a small band gap (ca. 1.8 eV) and a great prospect as a photocatalyst for visible-light applications. However, the use of BiOI could promote the rapid recombination of photogenerated electron–hole pairs. This problem could be solved by combining with some n-type semiconductors (TiO₂, ZnO). A p–n heterojunction between BiVO₄ and BiOI, deposited on FTO glass, was used as a photoanode in the PTM degradation by PEC [61]. EC and PEC experiments were carried out in a three-electrode configuration cell. Pt foil and Ag/AgCl were used as a counter and reference electrode, respectively. In the PEC process, a solar simulator (100 W, Xe lamp) was used as the light source, and the experiments were carried out in a 0.1 M Na₂SO₄ solution. Thus, 68% of degradation efficiency and 59% of TOC removal were achieved using a bias potential of 1.5 V within 2 h (see Table 2). PEC degradation of PTM was also evaluated with BiVO₄ and BiOI electrodes, and results demonstrated the formation of a p–n heterojunction in the BiVO₄/BiOI electrode since degradation efficiencies of BiVO₄ and BiOI electrodes were 44% and 53%, respectively (significantly lower than 85% obtained with BiVO₄/BiOI electrode). The authors also observed that the efficiency of both PTM removal and TOC conversion decreased in the presence of ciprofloxacin (see Table 2).

Orimolade et al. [52] coupled a cathodic electro-Fenton (EF) process (thermally treated carbon felt (T-CF) and hydroxyl radicals electrochemically produced from spontaneous reactions of iron catalyst and H₂O₂ by oxygen reduction at the cathode) and an anodic PEC oxidation process (bismuth oxyiodide electrodeposited on fluorine-doped tin oxide glass—FTO-BiVO₄/BiOI) into a single electrochemical advanced oxidation system for the mineralization of PTM. Table 3 compares the effect of applying the techniques individually with the use of the techniques in combination with PTM removal efficiency. The coupled process resulted in higher mineralization of PTM than EF and PEC processes. This behavior was attributed to the spontaneous degradation of PTM occurring simultaneously at both the cathode and anode. Total removal of PTM and 92% of TOC removal after 4 h were achieved in under 2 h, with an EF/PEC system and a current density of 10 mA cm⁻². The effect of the current density in the PTM and TOC removal in the PEC process, within a 5–20 mA cm⁻² current density range, was studied (Figure 6). As the current density increased from 5 mA cm⁻² to 10 mA cm⁻², the PTM removal improved. However, with the application of 15 mA cm⁻², no significant increase in the PTM removal was observed, whereas the application of 20 mA cm⁻² decreased the percentage removal since it affected the stability of the semiconducting materials. Therefore, an increase in current density may not always lead to improved degradation.

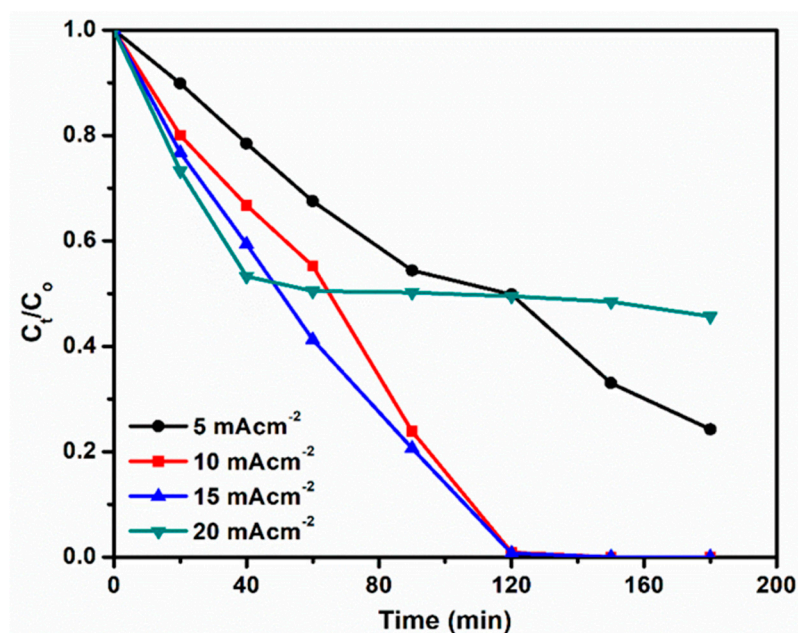


Figure 6. Effect of current densities on the PEC degradation of paracetamol on FTO-BiVO₄/BiOI (pH 3; 0.1 mM paracetamol). Reproduced with permission [52].

Table 2. PEC acetaminophen degradation using BiVO₄-based photoelectrodes.

Photoelectrode	Conditions	Degradation Time	Removal Efficiency (%)	TOC Removal (%)	Ref.
BiVO ₄ /BiOI	PEC oxidation 0.1 mg·L ⁻¹ Na ₂ SO ₄ Cathode: Pt foil	120 min	62	44	[61]
	10 mg·L ⁻¹ PTM solution 10 mg·L ⁻¹ ciprofloxacin solution Xe lamp 100 W Visible irradiation				
BiVO ₄	0.1 mg·L ⁻¹ Na ₂ SO ₄	120 min	68	59	[61]
	Cathode: Pt foil	120 min	44	-	
BiOI	10 mg·L ⁻¹ PTM solution Xe lamp 100 W Visible irradiation	120 min	53	-	[61]
	PEC oxidation 50 mmol·L ⁻¹ Na ₂ SO ₄ , pH: 3 0.1 mmol·L ⁻¹ PTM solution Cathode: Thermally treated-Carbon Felt (TCF) Current density: 5 mA cm ⁻²	180 min	76	-	
FTO-BiVO ₄ /BiOI	Current density: 10 mA cm ⁻²	180 min	98	59	[52]
	Current density: 15 mA cm ⁻²	180 min	98	-	
	Current density: 20 mA cm ⁻²	180 min	56	32	
	EC + EF oxidation 50 mmol·L ⁻¹ Na ₂ SO ₄ , pH: 3 0.1 mmol·L ⁻¹ PTM solution Cathode: Thermally treated-Carbon Felt (TCF) 0.2 mmol·L ⁻¹ FeSO ₄ ·7H ₂ O	180 min	56	73	

FTO-BiVO ₄ /BiOI	Current density: 10 mA cm ⁻² PEC + EF oxidation 50 mmol·L ⁻¹ Na ₂ SO ₄ , pH: 3 0.1 mmol·L ⁻¹ PTM solution Cathode: (TCF) 0.2 mmol·L ⁻¹ FeSO ₄ ·7H ₂ O 150 W linear Halogen lamp (VIS, 420–600 nm)	120 min	98	92	
	Current density: 10 mA cm ⁻² Constant O ₂ saturation EF oxidation 50 mmol·L ⁻¹ Na ₂ SO ₄ , pH: 3 0.1 mmol·L ⁻¹ PTM solution Cathode: Thermally treated-Carbon Felt (TCF) 0.2 mmol·L ⁻¹ FeSO ₄ ·7H ₂ O	240 min	28	71	
	Current density: 20 mA cm ⁻² Constant O ₂ saturation 50 mmol·L ⁻¹ Na ₂ SO ₄ , pH: 3 0.1 mmol·L ⁻¹ PTM solution Cathode: Thermally treated-Carbon Felt (TCF) 0.2 mmol·L ⁻¹ FeSO ₄ ·7H ₂ O	240 min	-	28	
	Current density: 10 mA cm ⁻² Constant O ₂ saturation 50 mmol·L ⁻¹ Na ₂ SO ₄ , pH: 3 0.1 mmol·L ⁻¹ PTM solution Cathode: Carbon Felt (CF) 0.2 mmol·L ⁻¹ FeSO ₄ ·7H ₂ O	240 min	-	27	[52]
	Current density: 20 mA cm ⁻² Constant O ₂ saturation EC ox. 50 mmol·L ⁻¹ Na ₂ SO ₄ , pH: 3 0.1 mmol·L ⁻¹ PTM solution Cathode: Thermally treated-Carbon Felt (TCF)	120 min	61	40	
	Current density: 10 mA cm ⁻² PC oxidation 50 mmol·L ⁻¹ Na ₂ SO ₄ , pH: 3 0.1 mmol·L ⁻¹ PTM solution Cathode: Thermally treated-Carbon Felt (TCF) 150 W linear Halogen lamp (VIS, 420–600 nm)	120 min	13	-	

1.3.3. Zinc-Oxide-Based Photoanodes

ZnO, considerably cheaper than the above photocatalysts, is extensively found in nature and considered an environmental-friendly material due to its innocuous character over living beings' health. Hexagonal wurtzite and cubic zinc blende are two crystalline structures of zinc oxide. Wurtzite is the most thermodynamically stable structure at ambient conditions, with a band gap value of 3.4 eV, with possible use under UV irradiation. The light penetration into the material and, therefore, the photogeneration of electron/hole pairs are improved by the use of ZnO thin films.

Hierarchical branched ZnO nanorod grown on FTO was used as an electrode for the PEC degradation of PTM [62]. The PEC process was carried out using a three-electrode configuration, with a saturated Ag/AgCl reference electrode and a Pt wire counter electrode. The conversion fraction of PTM increased from 32% over ZnO nanoparticles to 62% over branched ZnO nanorod arrays, attributed to the increase in the surface area. EC and PC degradation efficiencies were 2% and 8%, respectively, indicative of the synergistic effect between the EC and PC processes. Mineralization of the degraded PTM was 50–60%.

Table 3. PEC acetaminophen degradation using Bi₂WO₆-CNP-TiO₂ NTA-based photoelectrodes.

Photoelectrode	Conditions	Degradation Time	Removal Efficiency (%)	TOC Removal (%)	Ref.
Bi ₂ WO ₆ -CNP-TiO ₂ NTA	EC ox. 0.1 mol·L ⁻¹ Na ₂ SO ₄ , pH: 7 5 mg·L ⁻¹ PTM solution Cathode: Pt foil Current density: 10 mA cm ⁻²	180 min	57	30	[63]
	PC ox. 0.1 mol·L ⁻¹ Na ₂ SO ₄ 5 mg·L ⁻¹ PTM solution LCS-100 W solar simulator UV cut-off ($\lambda < 400$ nm)	180 min	42	22	
	PEC ox. 0.1 mol·L ⁻¹ Na ₂ SO ₄ , 5 mg·L ⁻¹ PTM solution Cathode: Pt foil Current density: 10 mA cm ⁻² LCS-100 W solar simulator UV cut-off ($\lambda < 400$ nm)	180 min	84	72	
	pH: 7	180 min	4	-	
	pH: 2	180 min	4	-	
	pH: 10	180 min	18	-	

Pd-loaded ZnO/carbon nanofibers (CZnO-Pd) were used in the PEC degradation of PTM [63]. The prepared material was deposited onto FTO glass and used as a photoanode. Pt mesh was used as the cathode and a 150 W linear halogen lamp (420–600 nm) as a light source. After 150 min of treatment, 56.40 % PTM removal was achieved using the CZnO anode (Figure 7a), while total removal of PTM was achieved with CZnO-Pd100 anode (see Table 3). Thus, the incorporation of Pd on the surface of CZnO improved the PEC performance of the photoanode. This behavior was attributed to the fact that transition metals increase light absorptivity and limit spontaneous recombination, therefore, improving the PC efficiency of semiconductors. However, an increase in the Pd load (CZnO-Pd200) produced a decrease in PEC performance (80% of PTM removal after 150 min).

In the absence of light (Figure 7b), the percentage removal of PTM decreased to about 82% after 3 h, which showed that irradiation enhanced the performance of the electrode. The experiment without applying an external current (PC process) drastically reduced percentage removal (14.30%), which demonstrates that the applied current in a PEC process improves the performance of the electrode by promoting efficient charge separation, as it provides a driving force for the photogenerated electron to detach from the anode surface. On the other hand, PEC treatment was carried out with 5 mA cm⁻² to confirm the dependence of the PEC process on the applied current (Figure 7c). In comparison with results obtained at 10 mA cm⁻², the percentage of PTM degradation was reduced to 78.11% after 150 min [63].

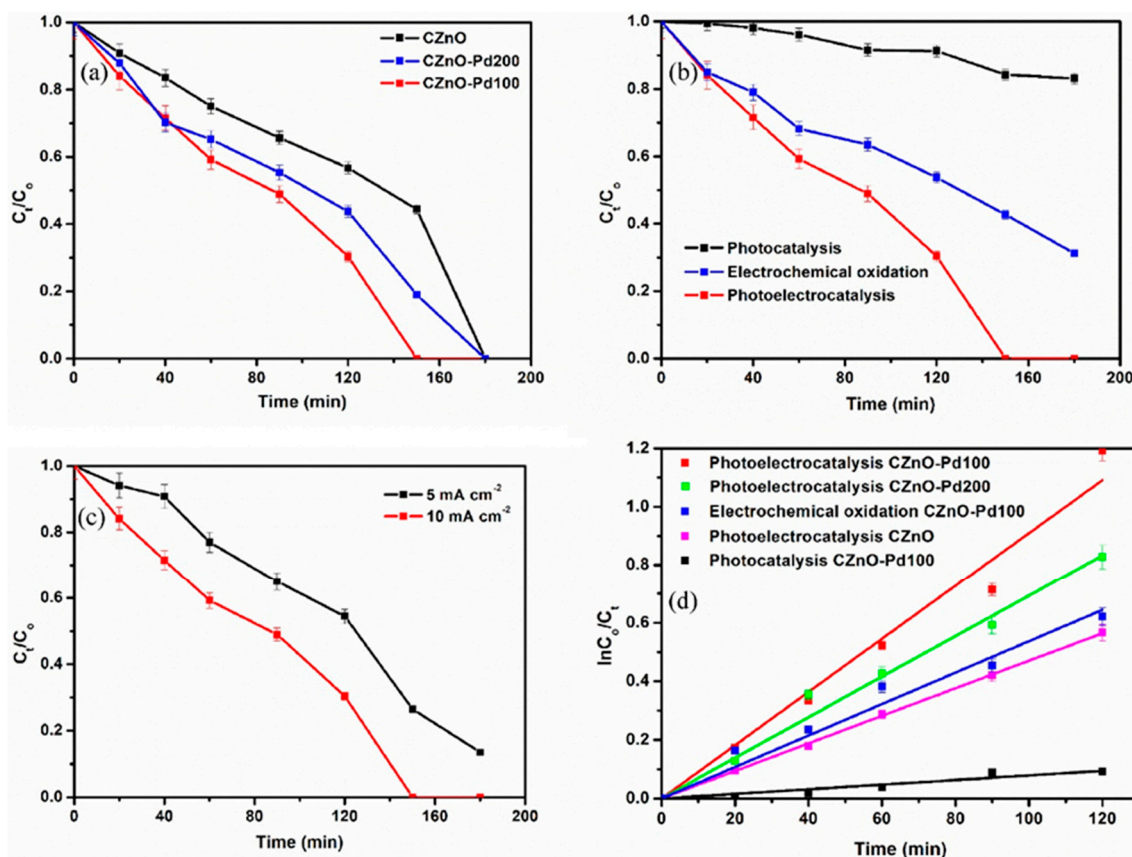


Figure 7. Normalized concentration decay versus time plot for PEC degradation of paracetamol using (a) CZnO, CZnO–Pd100 and CZnO–Pd200; (b) photocatalysis, electrochemical oxidation and photoelectrocatalysis using CZnO–Pd100; (c) photoelectrocatalysis at different current densities using CZnO–Pd100; (d) corresponding kinetics plot for the processes at 10 mAcm⁻² (0.1 mM paracetamol; pH 7; 50 mM Na₂SO₄). Reproduced with permission [63].

Despite the numerous publications related to ZnO as a photoanode in the PEC process, there is a lack of information on its combination with the Fenton process.

1.3.4. Tungsten-Oxide-Based Photoelectrodes

Tungsten trioxide (WO₃) is an n-type semiconductor whose crystal structure is dependent on the temperature, being the monoclinic I, gamma-WO₃ (17–330 °C), which exhibits the greatest stability at room temperature and a moderate band gap energy (2.4–2.8 eV) [64]. WO₃ is one of the visible-light active materials widely explored for water purification [65]. Among the main characteristics that make WO₃ interesting for its application in the removal of organic compounds present in water are its relatively high optical absorption in the visible range of the solar spectrum, deep valence band (3.1 eV), stability in low pH solutions, non-toxicity and low susceptibility to corrosion [64]. However, pure

WO₃ has a lower light energy conversion efficiency than the more widely used TiO₂, as the reduction potential of the electrons in WO₃ is low due to its low conduction band level.

Despite the excellent performance of WO₃-based photocatalysts in the removal of a certain organic compound, such as orange II dye [66,67], 2-nitrophenol [67] and Rhodamine B (RHB) [68], there is still no relevant information on their application in the removal of PTM present in water.

Bi₂WO₆, with a perovskite layered structure and high surface area, has been used in the PEC removal of organic pollutants [69]. Although it has a narrow band gap (2.7 eV) [70], which means it has strong light-harvesting capacity compared to TiO₂, it is limited by its low separation efficiency and its incapacity to completely utilize solar energy [71]. The heterojunction via two semiconductors and one electron conductor was proposed to overcome the problem of low separation efficiency and improve the photocatalytic efficiency of Bi₂WO₆ [70].

Mahhumane et al. [70] studied the PEC degradation of PTM performance of a visible-light-driven ternary z-scheme heterojunction of Bi₂WO₆ with carbon nanoparticles (CNPs) and TiO₂ nanotube arrays (NTAs) (Bi₂WO₆-CNP-TiO₂ NTA). At optimal conditions, a degradation efficiency of 84% and a TOC removal of 72% were achieved within 180 min. EC and PC processes yielded 57% and 42% of PTM removal and 30% and 22% of TOC removal, respectively. These results confirm that the synergy between applied bias potential and solar energy is a key factor for breaking down the PTM molecules. Regarding the effect of current density, with an increase in this parameter, ranging from 5 mA cm⁻² to 8 mA cm⁻², the degradation efficiency increased, which indicates that the hydroxyl radical species production depends on the applied current density. Nevertheless, working at higher current densities than the optimum values may give rise to oxygen evolution, preventing the generation of hydroxyl radicals and, therefore, suppressing the degradation efficiency [50]. The PEC process was also studied under different pH conditions [70]. At an acidic pH value, the surface of the catalyst becomes positively charged since the point zero of charge of TiO₂ and Bi₂WO₆ is 6.2 and 5.5, respectively. At pH 2, a 4% degradation efficiency was obtained, which could be related to the repulsion between the catalyst surface and the PTM molecules, since in this condition, PTM is in its protonated form. At alkaline conditions (pH 10), 18% PTM degradation was obtained. At this pH, more hydroxyl radical species may be formed, but PTM is in its deprotonated form and, therefore, the repulsion between the negatively charged solution and the PTM molecule might diminish the degradation performance. At pH 7, where PTM is in its neutral form, a maximum degradation efficiency was obtained (84%) [70]. Table 3 summarizes the results obtained with the photoelectrode Bi₂WO₆-CNP-TiO₂.

Optical, electrochemical and photoelectrical characterization of the materials was conducted. UV-vis diffuse reflectance spectroscopy (Figure 8a) showed that the Bi₂WO₆-CNP-TiO₂ NTA electrode has higher absorbance than the TiO₂-NTA electrode in the visible region, which suggests that the formation of the z-scheme heterojunction has enhanced visible light activity. The photocurrent transients (Figure 8b) exposed that the Bi₂WO₆-CNP-TiO₂ NTA electrode exhibits a more marked photocurrent response with a higher current density of 0.789 mA cm⁻² when compared to bare TiO₂ NTA with a current density of 0.172 mA cm⁻². This behavior could result from the z-scheme of heterojunction between Bi₂WO₆, CNP and TiO₂ NTA, leading to an improved separation of electron hole pairs (Figure 8c). To better understand the improved performance of the prepared electrodes, an experiment was conducted using electrochemical impedance spectroscopy (Figure 8d). The charge transfer resistance of TiO₂ NTA and Bi₂WO₆-CNP-TiO₂ NTA was 92.14 Ω and 31.32 Ω, respectively, which suggests that the fabrication of the heterojunction improved the charge mobility of the electrode and lowered the rate of instantaneous recombination of electron-hole pairs [70].

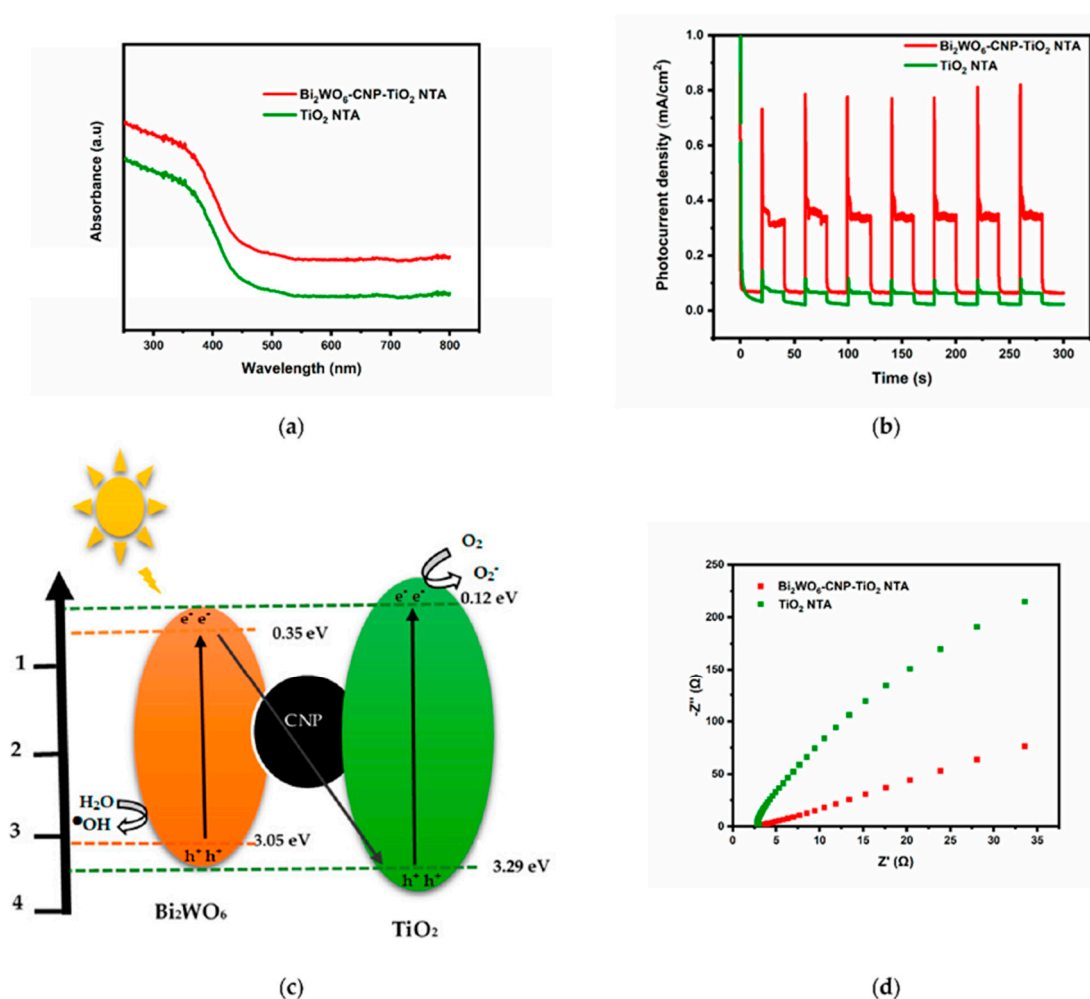


Figure 8. (a) UV-Visible diffuse reflectance spectra TiO₂ NTA Bi₂WO₆-CNP-TiO₂ NTA; (b) photocurrent responses of TiO₂ NTA and Bi₂WO₆-CNP-TiO₂ NTA in 0.1 M Na₂SO₄; (c) schematic diagram of Z-scheme mechanism of Bi₂WO₆-CNP-TiO₂ NTA electrode; (d) Nyquist plot TiO₂ NTA and Bi₂WO₆-CNP-TiO₂ NTA photoanodes in 5 mM [Fe(CN)₆]^{3-/4-} in 0.1 M KCl (pH 7). Reproduced with permission [70].

2. Conclusions and Prospects

One of the most prevalent problems bothering people throughout the world is derisory access to clean water and sanitation. Problems with water are likely to grow worse in the near-term years. Addressing these problems calls for research to identify robust new methods of purifying water at a lower cost and with less energy, while at the same time minimizing the use of chemicals and their impact on the environment. Several studies point to the development of semiconductor materials and PEC degradation of pharmaceuticals.

This review highlights the PEC process applied in the removal of PTM from water, the main photocatalysts used and the parameters affecting their performance. Among the different photocatalysts reported, large differences in their efficiency were observed, which are strongly associated with the experimental conditions. Bandgap energy plays an important role in determining the photoactivity of the different materials, so strategies, such as heterojunction between different semiconductors and coupling of nanocarbon, can improve the efficiency of the PEC.

It was demonstrated that the coupling of the electrochemical and photochemical processes, as well as Fenton reaction and consequently photoelectrocatalysis, results in higher efficiency than when these processes are applied separately, which highlights the synergistic effect of the coupled processes. On the other hand, experimental variables, such as

temperature, pH, current density, nature of the photocatalysts, as well as their preparation methods, are parameters that considerably influence the photoelectrocatalytic activity of each system, determining its efficiency in paracetamol removal.

Among the different systems reported, there are large differences in the PMT percentage removal as well as in the percentage of mineralization. Although the comparison between these systems is not simple, considering that the experimental conditions used in each case are somewhat different (electrode types and morphology, fixed currents or applied potentials, etc.), it is important to highlight that for each type of material, the modification of these variables leads to different results. While reported studies show a very good performance in the removal of PTM by PEC processes, demonstrating the promising application of these systems in the removal of pharmaceuticals present in water, it would be of great relevance to be able to corroborate the efficiency for more complex matrices, close to real operating conditions. On the other hand, the efficiency of a drug removal method is also linked to the degree of drug mineralization it achieves. In this sense, it is very important that research in this field reports the percentage of TOC removed after testing, to ensure that the compound of interest has not been broken down into more harmful compounds. Furthermore, studies using in situ spectroelectrochemical techniques, such as differential electrochemical mass spectrometry (DEMS) and electrochemical Raman (EC-Raman), among others, are necessary to elucidate the operating reaction mechanism of each drug under study.

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

Abbreviations

CECs	Contaminants of emerging concern
PEC	Photoelectrocatalytic
PTM	Paracetamol
APIs	Active pharmaceutical ingredients
AOPs	Advanced Oxidation Processes
EAOPs	Electrochemical Advanced Oxidation Processes
EC	Electrocatalytic
PC	Photocatalytic
PE	Photoelectrode
EF	Electro-Fenton
PEF	photo-Electro-Fenton
SMC	Semiconductor
SSP	Stainless-steel plate
C-PTFE	Carbon-polytetrafluoroethylene

CNP
NTACarbon nanoparticles
nanotube arrays

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