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Review

Use of Electrocoagulation for Treatment of Pharmaceutical Compounds in Water/Wastewater: A Review Exploring Opportunities and Challenges

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Abstract: Increasing dependency on pharmaceutical compounds including antibiotics, analgesics, antidepressants, and other drugs has threatened the environment as well as human health. Their occurrence, transformation, and fate in the environment are causing significant concerns. Several existing treatment technologies are there with their pros and cons for the treatment of pharmaceutical wastewater (PWW). Still, electrocoagulation is considered as the modern and decisive technology for treatment. In the EC process, utilizing electricity (AC/DC) and electrodes, contaminants become coagulated with the metal hydroxide and are separated by co-precipitation. The main mechanism is charge neutralization and adsorption of contaminants on the generated flocs. The range of parameters affects the EC process and is directly related to the removal efficiency and its overall operational cost. This process only could be scaled up on the industrial level if process parameters become optimized and energy consumption is reduced. Unfortunately, the removal mechanism of particular pharmaceuticals and complex physiochemical phenomena involved in this process are not fully understood. For this reason, further research and reviews are required to fill the knowledge gap. This review discusses the use of EC for removing pharmaceuticals and focuses on removal mechanism and process parameters, the cost assessment, and the challenges involved in mitigation.

Keywords: remediation; pharmaceuticals; polyaromatics; electrocoagulation; co-precipitation; optimisation

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1. Introduction

Pharmaceuticals compounds have now become a necessary component of a sophisticated lifestyle in many ways. Every day, we extensively use pharmaceuticals such as antibiotics, hormones, analgesics, and antidepressants. The presence of these contaminants in the environment either directly or due to their metabolites/after chemical modifications are very toxic. In the last decade, the detection of several groups of antibiotics such as Norfloxacin, Ciprofloxacin, Clarithromycin, Ofloxacin, Levofloxacin, and Oxytetracycline in the environment has imposed anxiety on researchers [1,2]. Even they are present in drinking water in varying concentrations [3,4]. These common pharmaceuticals can easily enter into water bodies via drug manufacturing companies [5], hospitals [6], medical waste, and prescribed consumption of different medicines. These pharmaceuticals upon consumption are partially metabolized by the human body and are mainly discharged through urine and faeces [7].

Wastewater from pharmaceutical companies is associated with a high amount of COD and nitrogenous compounds that need additional treatment. Discharging such effluent into

a nearby environment without proper treatment can cause a detrimental effect on flora and fauna [8]. The presence of a high concentration of oestrogen could enhance the mortality rate of a fish [9] and the feminization of male fish [10]. They also have an adverse effect on the human body. The long-term exposure of some complex pharmaceutical compounds present in water bodies may cause chronic as well as acute contamination [11,12], behavioural changes [13], reproductive harm [14], and cell proliferation inhibition [15]. These effluents further create microbial hazards [16,17], chemical hazards [18], and agricultural implications by entering into the food chain [19,20].

Pharmaceutical wastewater includes different chemical compounds and functional groups, which inhibit microbial activity [21,22]. This characteristic makes it unfit for biological treatment. Therefore, major treatment methods for pharmaceutical wastewater are based on physio-chemical treatment. Unfortunately, the high cost of the physiochemical process makes it less popular for pharmaceutical wastewater [23]. Thus, optimized treatment by electrocoagulation process is comparatively less costly [24]. The benefit of using an EC method is that the dissolute metal hydroxides ions eliminate the inorganic contaminant present in wastewater [25–27].

Even after going through recent existing literature on pharmaceutical removal via EC, the removal mechanism of particular pharmaceuticals and complex physiochemical phenomena involved in the process is not fully understood. In this review paper, the removal of the pharmaceutical compounds from water/wastewater via EC and their mechanisms are proposed discussed. The potential sources and different characteristics of PWW are reviewed. Additionally, their efficiencies on various operating parameters, integrated EC approach, techno-economic analysis, and opportunities and future challenges associated are discussed. In addition, the problems and mitigations in EC scale-up, software aided optimization, and different integrated EC techniques are summarized.

2. Sources and Characteristics of Pharmaceuticals in Wastewater

Pharmaceutical compounds enter the water bodies from several scattered sources. Pharmaceutical production companies, wastewater treatment plants, hospitals, landfills, and even graveyards are some of the main sources of contamination [28].

2.1. Pharmaceutical Production Companies

Pharmaceutical companies produced many types of medicines and cosmetics, including antibiotics, polyaromatics, and phenols [29]. Additionally, they discharge microplastics, gene-altering substances, and endocrine disruptors, threatening human health [30]. According to a study [31], pharmaceutical companies in Hyderabad, India, failed to treat discharged contaminants properly. As a result, a high level of antibiotic and antifungal drug residue was found in nearby water bodies [32–34]. However, a recent draft bill published on 23 January 2020 by the Indian Government limits the antibiotic concentration present in discarded waste by pharmaceutical companies [35].

2.2. Wastewater Treatment Plants

Pharmaceutical compounds eventually meet industrial/municipal wastewater from different sources and are collected at a nearby wastewater treatment plant. The most probable entry point of pharmaceuticals in the environment is the municipal wastewater treatment plant effluent. The human body partially metabolizes pharmaceuticals; therefore, after excretion, they are collected in the wastewater treatment plant [7,36]. Here, by conventional water treatment, including biodegradation and adoption, the pharmaceutical compounds remain in the effluent or concentrated on the sludge in a significant concentration [37–39].

2.3. Hospitals

Hospitals are the nucleus of pharmaceutical activities. A various range of drugs is found in the hospital wastewater. According to the study of Zhang et al. (2016), hospital wastewater contains a higher concentration of central nervous system drugs such as sulpiride and clozapine [40]. It is also seen that there is a strong correlation of pharmaceuticals in hospital waste to the population density, economy, and consumption rate [41].

2.4. Landfills

Waste from hospitals and pharmaceutical companies are generally disposed of in engineered landfills. However, still, there are chances of escape of these contaminants when concentrated landfill leachates reach groundwater. In the study conducted by Zhang et al. (2016) [40], the authors reported the presence of chloramphenicol, which was used in typhoid treatment, in the landfill leachate, livestock excrement, and aquaculture water [42].

2.5. Characteristic of Wastewater Associated with Pharmaceutical Industry

The characteristic of PWW depends on the types of prevalent drugs found in PWW. Petrovic et al. (2009) [43] reviewed different samples of PWW from different sources and gave an overview of prevalent drugs found in PWW. It is shown in Table 1.

Table 1. Prevalent drugs in PWW.

Drugs	Chemical Class	Pharmacological Class
Citalopram	SSRIs	Antidepressant
Cocaine	Tropane alkaloid	CNS stimulant/narcotic
Ibuprofen	Propionic acid derivative	NSAIDs
Propranolol	Beta blocker	Antihypertensive
Clotrimazole	Imidazoles	Antifungal
Diclofenac	Acetic acid derivative	NSAIDs
Metoprolol	Beta blocker	Antihypertensive
Indomethacin	Indole derivative	NSAIDs
Atenolol	Beta blocker	Antihypertensive
Paracetamol	Para-aminophenol derivative	NSAIDs
Ranitidine	H2 receptor blocker	Antihistaminic
Gemfibrozil	Fibric acid derivative	Lipid and cholesterol regulating
Sulfadiazine	Sulfonamide	Antibiotic
Clofibric acid	Clofibrate metabolite	Lipid and cholesterol regulating
Norfloxacin	Fluoroquinolone	Antibiotic
Carbamazepine	Tricyclic anti-depressant	Psychiatric/Anticonvulsant
Amoxicillin	Penicillin	Antibiotic
sulfamethoxazole	Sulfonamide	Antibiotic
Chloramphenicol	Amphenicol-class antibacterial	Antibiotic
Ofloxacin	Fluoroquinolone	Antibiotic
Trimethoprim	Aminopyrimidine	Antibiotic
Ciprofloxacin	Fluoroquinolone	Antibiotic
Fibrates	Amphipathic carboxylic acid	Blood and lipid regulating

Wastewater characterization is necessary to select the appropriate processing method [44]. However, often, pharmaceutical effluents contain a variety of chemicals, including carcinogenic, mutagenic, and others. All possible pollutants are classified in Figure 1.

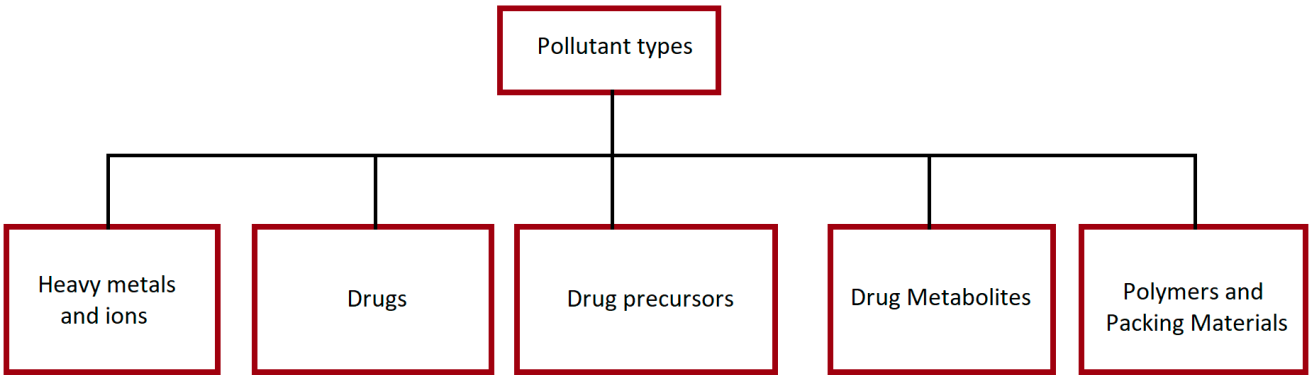

Figure 1. Classification of possible pollutants in PWW.

Various physicochemical parameters such as pH, BOD, COD, and TSS, among others are all affected by these pollutants. Characterization of different pharmaceutical wastewater is shown in Table 2.

Table 2. Range of parameters in different PWW.

Parameter	References						
	[45]	[46]	[47]	[48]	[49]	[50]	[51]
BOD (mg/L)	120	304	900	22,000	200	-	263–330
COD (mg/L)	490	420	4000	34,400	1753	-	2565–28,640
pH	6.9	7	5.2	7.2	7.3	5.65–6.89	5.8–6.9
TSS (mg/L)	370	57	68	6250	-	29.67–123.03	761–1202
TDS (mg/L)	1550	-	-	-	-	136.33–193.05	1443–3788
TS (mg/L)	1920	484	-	29,150	-	-	-
Ammonium nitrogen (mg/L)	-	52	-	-	220	-	-
TP (mg/L)	-	7.5	1.7	-	17	-	-
Chloride (mg/L)	-	132	-	-	4.2	-	-
Turbidity (NTU)	-	-	18	-	-	17.22–28.78	-
Conductivity (mS cm^{-1})	-	-	0.5	-	20	157–119.36	-
Temperature (°C)	-	22	25	29–36	-	32–46	31–34

3. Different Removal Technologies for Pharmaceutical Contaminants

Various chemical, physical, and biological methods can be used for the extermination of pharmaceutical contaminants from PWW. The presence of a high COD amount [52], dissolved organic nitrogen [53], and salinity [54] make PWW more toxic and usually unfit for biological treatment. Therefore, a physio-chemical treatment is generally applied.

3.1. Physiochemical Treatment Technologies

Physicochemical processes are the most common treatment method for PWW. Usually, processes such as activated carbon adsorption, carbon nanotubes, electrocoagulation, and ozone treatment are used.

3.1.1. Activated Carbon

Briefly, activated carbon is a type of carbon that provides a larger surface area for adsorption by having low volume pores. Carbon can be powdered (PAC), granular (GAC), or bead-shaped (BAC). The removal capacity of pharmaceuticals by activated carbon depends on hydrophobicity, charges of pharmaceutical contaminants, and water matrix [55,56]. A variety of precursors is used to prepare AC out of biomass, such as cocoa shells [57], coffee residuals [58], palm leaflets [59], and fruit stones [60]. Sheng et al. (2016) reported an efficient pharmaceutical contaminants removal using integrated adsorption and filtration [61].

3.1.2. Carbon Nanotubes

Carbon nanotubes are hollow materials made up of single/multiple layer graphite sheets. It varies in terms of length and diameter according to the synthesis and requirement [62]. The presence of higher surface area and larger microspore volume makes it efficient for the removal of pharmaceuticals, heavy metals [63,64], dyes [65], phenol [66], and others. Shan et al. (2016) prepared novel granular carbon nanotubes by improving the surface area and pore volume and providing easy separation [67]. Apart from the numerous data available in this field, a knowledge gap still exists to understand better pharmaceuticals removal from PWW using nanotubes.

3.1.3. Electrocoagulation

In the electrocoagulation process, dissolved metal hydroxides remove the contaminants. The removal mechanism follows metal dissolution on anode and production of hydroxyl ion on the cathode [68]. This method is more economical and efficient than other physicochemical methods.

3.1.4. Ozone Treatment

Ozone treatment is considered an effective oxidant and disinfectant for a long time. It can oxidize many pharmaceuticals and organic matters present in wastewater and enhance biodegradability at natural pH [69]. Gome and Upadhyay (2013) applied ozone treatment on PWW under acidic or alkaline conditions. The authors reported that an ozone dose of 32.7 mg/L for acidic and 30.0 mg/L for alkaline was required [45].

3.1.5. Advanced Oxidation Technologies

Advanced oxidation technologies (AOTs) are being used to treat drinking water for a long time. Nowadays, it is also used for industrial effluents and PWW. AOTs include the Fenton process [70], UV/H_2O_2 process [71], sonolysis [72], electrochemical oxidation [73], radiation [74], and combined advance oxidation technologies [75]. Since these technologies are based on the production of highly reactive oxygen species generated during this process, they are often used to treat pharmaceutical contaminants. In addition, the final products of AOTs are lesser harmful such as CO_2, water, and acids [76].

3.2. Bioremediation

In general, bioremediation is applied to treat wastewater rich in carbon and nitrogen. Nevertheless, bioremediation is usually not sufficient in pharmaceutical contaminants, since PWW has high COD and many toxic substances. Bioremediation depends on several factors and operating conditions, e.g., toxicity, pollutant concentration, microbial strains' efficiency, climatic condition, and retention time [77]. Several studies have been performed on the removal of pharmaceuticals by different bioremediation technologies, e.g., bacterial bioremediation [78], active sludge process [79], membrane bioreactors [80], sequence batch reactor [81], and fungal bioremediation [82].

These methods have certain limitations, i.e., they require higher specificity [83], while some drugs show no removal [84].

3.3. Membrane Technology

Membrane-based treatment is a pressure-driven process that has become popular due to its energy efficiency and environmentally friendly nature [85]. The effectiveness of this method depends upon membrane fouling, selectivity, flux, and hydrophilicity [86]. Previously a myriad amount of work has been done for pharmaceuticals, including analgetics [87,88], antibiotics [89–91], and antidepressants [92] based on different membrane technology. Still, the limitations associated with the membrane fouling and high cost exist, thus preventing the wide industrial application.

4. Applicability of Electrocoagulation in Removal of Pharmaceuticals

4.1. Theory and Mechanism of EC

EC technology is an old approach [93,94]. However, its application for the separation of contaminants is new [95,96]. Lately, it was found that electro-generated flocs are helpful in water decolorization [97]. Nowadays, this method is used to efficiently treat various wastewaters, including industrial, agricultural, and pharmaceuticals [98]. In a broad sense, EC is based on various mechanisms including physical (sorption, coagulation/flocculation), chemical (redox reaction, hydroxide precipitation), and electro-chemical (metal disintegration, water reduction, pollutant electro-oxidation/reduction) [95,99], which are summarized in Figure 2.

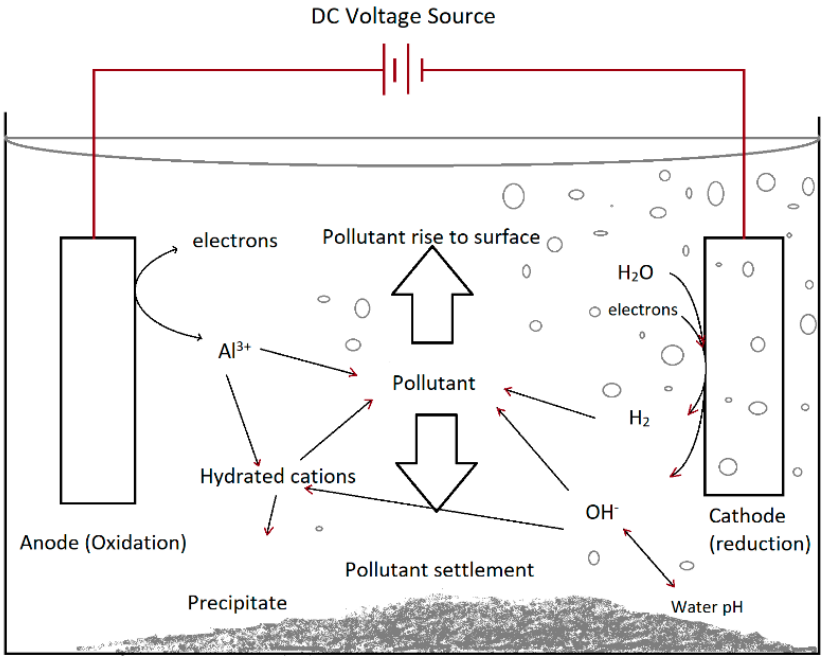

Figure 2. Interactions occurring within the EC reactor.

In this process, electric current (DC/AC) is used as the main power source. Utilizing the current and the appropriate electrodes, the contaminants become coagulated and separated by precipitation producing clean water. Generally, Al/Fe electrodes are taken as sacrificial electrodes, and their disintegration occurs into the electrolyte. Coagulant species further react with dissolve/suspended pollutants, resulting in flocs production that can be easily isolated from the water [100,101].

4.2. EC Mechanisms

The primary mechanism behind the EC process is the generation of in situ coagulant species by electro-dissolution of sacrificial electrodes usually aluminium or iron.

A general chemical equation would be as follows:

Metal is oxidized, and cations are formed at the anode according to Equation (1)

$$M \rightarrow M^{Z+} + Ze^- \quad (1)$$

where Z is the number of electrons transferred in this process; there are also chances of secondary reactions in case of the high potential anode.

At cathode: water is reduced, resulting in the production of hydroxyl ions and hydrogen gas.

$$3H_2O + 3e^- \rightarrow \frac{3}{2} H_2 + 3OH^- \quad (2)$$

The amount of dissolved metal is solved with the help of Faraday's law [102,103].

$$m = \phi \frac{It}{ZF} M \quad (3)$$

In Equation (3), M represents the atomic weight electrode of metal and F represents Faraday's constant [99]. Sometimes, a parallel reaction occurs, and all electrons do not participate in metal dissolution at the electrode. In this case, a correction factor, ϕ is required to compensate the difference between theoretical and experimental disintegration of the metallic anode [104,105]. Generally, the value of ϕ is less than 1. However, when chemical and electrochemical oxidation happens on anode simultaneously, the value of ϕ exceeds 1 [106–108]. The released metal cation in the system undergoes several equilibrium reactions such as precipitation, acid/base, and redox reaction. However, the formation of metal hydroxides is the most common phenomenon that exhibits poor solubility and separates easily. Some of the metal hydroxides reactions with soluble pollutants are listed in Table 3.

Table 3. Extraction mechanism of distinct soluble pollutant via metal hydroxides.

Soluble Pollutants in WW	Mechanism of Removal	References
Organic Compounds	Complexation, co-precipitation	[109]
Phosphate Anions	Precipitation, Adsorption, Complexation	[110]
Sulphide Anions	Precipitation	[111]
Calcium Cations	Co–precipitation	[112]
Fluoride Anions	Complexation, Precipitation	[113]

The main mechanisms of pollutants removal through hydroxide precipitations are adsorption and complexation [114]. For some pollutants, the direct adsorption of pollutants on electrode occurs where pollutants are attracted toward anode due to electric forces [115]. A summary of these mechanisms is shown in Figure 3.

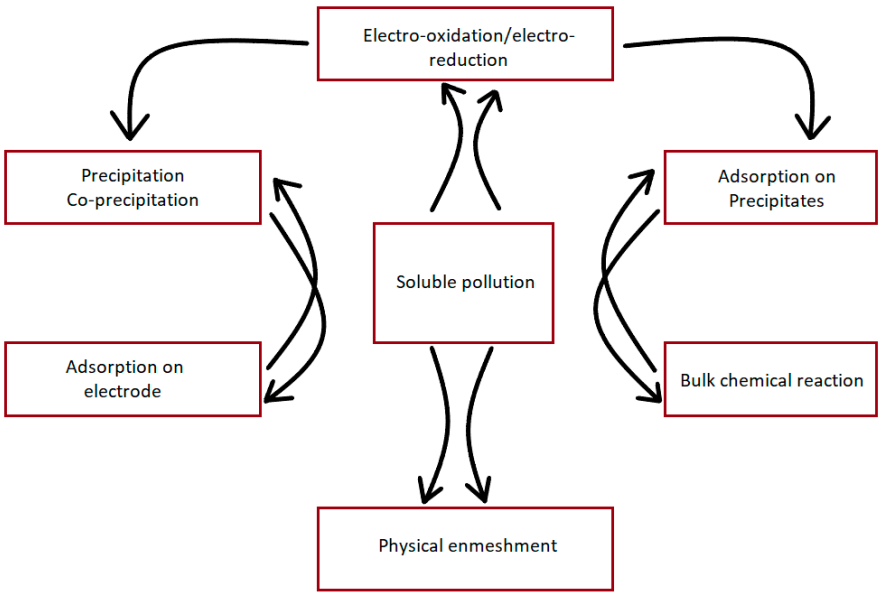

Figure 3. The main mechanism of soluble pollutant abatement using EC [99].

4.3. EC Using Al Electrodes

While using Al electrodes, Al^{3+}/Al follows Equation (1), where Z = 3. Here, apart from the formation of metal hydroxides, other monomeric species are also formed, which follow acid/base reactions (Equations (4) and (7)).

$$Al^{3+} + H_2O \rightarrow Al(OH)^{2+} + H^+ \quad (4)$$

$$Al(OH)^{2+} + H_2O \rightarrow Al(OH)_2{}^+ + H^+ \quad (5)$$

$$Al(OH)_2{}^+ + H_2O \rightarrow Al(OH)_3 + H^+ \quad (6)$$

$$Al(OH)_3 + H_2O \rightarrow Al(OH)_4{}^- + H^+ \quad (7)$$

The role of pH prevails in aluminium speciation. $Al(OH)_3$ predominates between pH 4–10. Otherwise, pH above 10, soluble aluminate anions prevail, and pH below 4, soluble Al^{3+} cations prevail.

4.4. EC Using Fe Electrodes

Electrochemical reactions become more complex using iron electrodes than aluminium as Fe^{2+}/Fe^{3+} cations form after the anodic reaction.

$$Fe \rightarrow Fe^{2+} + 2e^- \quad (8)$$

$$Fe \rightarrow Fe^{3+} + 3e^- \quad (9)$$

However, Fe^{2+} dissolution mainly prevails, as Fe^{3+} dissolution is negligible [116–118]. These ions become hydrolysed and form several monomeric/polymeric species depending on pH and Fe^{3+} concentration. Further, acid/base equilibrium reactions make it more complex. Anodic oxidation strongly depends on O_2 concentration and pH [117,119]. In alkaline/neutral media, Fe^{2+} species directly turn to ferrous hydroxide and further oxidizes into ferric hydroxide Equations (11) and (12). While at low pH, they slowly react with oxygen and turn into Fe^{3+} (10).

$$Fe^{2+} + O_2 + 2H_2O \rightarrow Fe^{3+} + 4OH^- \quad (10)$$

$$Fe^{2+} + 2OH^- \rightarrow Fe(OH)_2 \quad (11)$$

$$4Fe(OH)_2 + O_2 + 2H_2O \rightarrow 4Fe(OH)_3 \quad (12)$$

Similar to Al dissolution, Fe dissolution on anode also follows Faraday's law where the value of ϕ is around 0.8–1.0 [106–108]. Despite this, the value is greater than one at lower pH [116]. Finally, several monomeric/polymeric species become amorphous precipitate ($Fe(OH)_3$) that assist adsorption of soluble and trapping of insoluble contaminants [120,121].

5. Parameters Affecting EC

5.1. Initial pH

One of the most significant factors influencing the EC process is the initial pH. It mainly depends on the type of contaminants, anode material, and reaction mechanisms. However, it increases as the reaction proceeds [122]. Thus, a reaction mechanism is needed to optimize pH for effective removal [123]. Pharmaceutical plants produce effluents with a broad pH range, from very acidic to alkaline. Lots of studies show that pH has a significant impact on the EC process and treatment effectiveness. The type of EC-causing species and the solubility all depend on pH [124,125].

Apart from a general process, pH also has a vital role for specific pharmaceuticals such as tetracycline-related compounds. For acidic pH (less than 3.3), tetracycline is a cation due to the presence of protonated dimethyl ammonium group. At medium pH (3.3–7.7), the phenolic diketone moiety loses a proton and forms zwitterion. Lastly, at high pH (greater than 7.7), tetracycline is present as a monovalent/divalent anion [126].

5.2. Duration of Electrocoagulation Treatment

The duration of EC treatment is another critical factor that directly affects the EC process. Regardless of the current density used, removal efficiencies are directly proportional to the time of electrolysis [127]. Increased electrolysis time leads to the increment of generation of complex aluminium ions, hydrogen bubbles through electrodissolution of the anode, and reduction in the cathode. However, the removal efficiency only increases with time until an optimal value. The removal efficiency becomes constant outside the optimum time limit due to excess coagulant [128]. Although a slight increment in the duration could increase the removal efficiency in light of the price and process suitability, the optimum duration for the EC is usually considered 20–30 min [118,129].

5.3. Current Density (CD)

CD has a significant impact on the EC process, since it controls the rate at which coagulant is added, the evolution of gas, the formation of bubbles, the size of the floc, and the EC operating costs resulting in faster pollutant removal [130,131]. BOD, COD, and colour removal increase with the current density, and hence, the treatment time decreases. Increased current causes faster anode disintegration, results in increased precipitation, and more bubble production, which accounts for more flocculation and coagulation, resulting in higher removal [132]. The number of flocs produced is a function of the amount of treatment time and the CD used. High CD increases the quantity and rate of floc generation that collects a lot of suspended solids and increases the amount of sludge generated. In addition, excess metal hydroxide flocs are hard to float, resulting in operational shortfalls [133]. In addition, using high CD, much of the energy is lost in the water electrolysis reaction and heating due to the joule effect. Therefore, it is crucial to operating the device at an optimum CD [21]. For removing oxytetracycline hydrochloride, Nariyan et al. (2017) reported that the optimum current density was 20 $mA.cm^{-2}$ using Fe/Al [134].

5.4. Mode of Electricity Application

The constant supply of electricity ensures the continuous supply of Al^{3+} species, which are responsible for the charge neutralization and eventual precipitation of suspended micropollutants and simultaneous adsorption of dissolved organic matter and other chemical species. However, intermittent electricity supply can reduce the consumption of energy and electrode material, since the metal dissolution stops between the gaps [135]. As a consequence, intermittent electrical supply limits anode passivation. The energy consumption is calculated by Equation (13).

$$Q = V.I.t.\theta^2 \quad (13)$$

where Q denotes consumption (Wh); V denotes peak voltage (V); I denotes peak current (A); t denotes reaction time (s); θ denotes intermittent cycle, which is 1 in case of continuous current supply.

Ensano et al. (2017) studied the extraction of pharmaceutical waste by intermittent coagulation (5 min ON/20 min OFF) and reported 90% removal of diclofenac at a current density of 0.5 mA/cm^2 for HRT of 38 h [24].

5.5. Electrode Material

The electrocoagulation process revolves around the vital role of electrodes. Consequently, parameters and configurations, including electrode materials, spacing, shape, and arrangement, are highly significant. Several materials such as Al, Fe, Zn, Ag, Na, Mg, Si, Ca, Sr, and Cd are used as electrodes in EC [136]. In addition, using graphite and lead oxide as electrode materials for EC is also reported [137]. These materials are different in terms of their chemical and physical properties. Al and Fe electrodes are the most frequently used materials for the electrode in electrocoagulation applications because they are inexpensive, non-toxic, readily available, and proven reliable. Researchers applied electrocoagulation to treat olive mill wastewater with both iron and sacrificial aluminium electrodes [138]. They observed that the aluminium electrode was more efficient at reducing COD in wastewater.

5.6. Electrode Spacing

Electrode spacing is directly linked to the functioning of the EC, as it can alter electrostatic attraction, the in-between residence time, turbulence, mass transfer, and finally, pollutant removal efficiency. For instance, the electrostatic force between metal hydroxide flocs would be very high at low spacing; therefore, degradation of flocs will occur due to intensive collision [139]. On the other hand, at higher electrode spacing, the time for agglomeration of produced metal hydroxide is enough with fewer flocs degradation, and hence, the removal efficiency increased. However, if the spacing increased beyond the optimum range, the removal efficiency decreased due to the decreased flocs formation and increased potential drop [140].

5.7. Electrode Arrangement

Irrespective of the ease of the general EC set-up, they are not widely applicable for industrial wastewater treatment, as they require a large surface area of the electrode. However, this drawback can be solved by using series/parallel monopolar or bipolar systems of electrodes. In a complex EC system, three different electrode arrangements are reported [141,142].

1. Monopolar parallel electrodes (MP-P): In the MP-P setup, both oppositely charged electrodes are joined to one another and farther to the outer circuit. As the currents divide in this arrangement, the potential difference becomes lower [143] (Figure 4a).
2. Monopolar-series electrodes (MP-S): In the MP-S setup, each internal pair of sacrificial electrodes are joined to one another making an equal amount of current supply in each. However, the voltage is additive here (Figure 4b).
3. Bipolar series electrodes (BP-S): In the BP-S arrangement, two of the outer monopolar electrodes are joined to the external circuit, and internal bipolar sacrificial electrodes are without a connection. Here, on the positive side, oxidation of meal takes place, and the cathodic reaction takes place on the negative side [144] (Figure 4c).

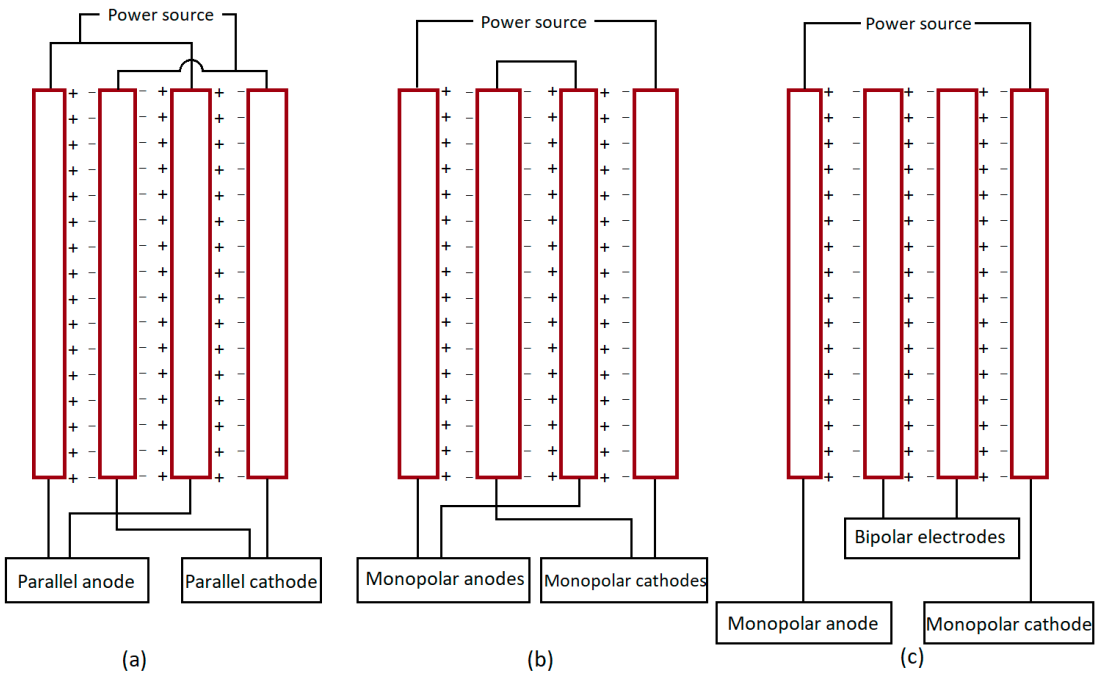

Figure 4. Different electrodes arrangements in an electrochemical cell: (**a**) MP-P; (**b**) MP-S; (**c**) BP-S.

5.8. Electrode Shape

Electrodes have been used in several shapes such as rectangular, circular, cylindrical, punched hole type in which rectangular shape is the most common. Generally, they are placed vertically. However, in some studies, horizontal placement of electrodes is also reported, which enhanced the mixing efficiency [145,146]. Another study based on a comparison of punched vs. plane electrodes claimed that punched electrodes have higher collection efficiency and higher electric field on the punched edges [147].

5.9. Mode of Power Supply

Supplies in the form of DC power are generally used for electro-coagulation cells. Several studies have been carried out using DC sources except for a very few studies where the AC source had been implemented [128,136]. However, the use of DC means that the anode is oxidized, and a layer of oxide is developed on the cathode called as cathode passivation. This passive layer results in the decrease in the current flow between both electrodes, and the efficiency of EC decreases. Passivity results in overpotential, which leads to a large amount of energy utilization. Several study results with supply in the form of AC were promising. Mollah et al. (2004) noticed that AC supply ensures an adequate life of electrodes by cyclic energy that hinders DC's conventional consumption [142]. The AC and DC supply reaction on cadmium removal from water through electrical coagulation has been examined by Vasudevan et al. (2011) [134]. The results have shown that less energy and greater removal efficiency were achieved using an AC power supply.

Ahmadzadeh et al. (2017) studied optimal conditions for the elimination of ciprofloxacin from hospital wastewater with the help of response surface methodology. The optimal conditions were pH 7.78, electrode spacing 1 cm, CD 12.5 mA/cm^2, reaction time 20 min, initial concentration of ciprofloxacin 32.5 mg/L, and 0.07 M NaCl as the electrolyte [148]. Some of the previous studies based on pharmaceutical removal by EC are summarized in Table 4 below.

Table 4. Few past studies carried out for pharmaceutical removal by EC.

Source of Wastewater	Pharmaceutical Contaminants	Experimental Conditions	%Removal Efficiency	References
Pharmaceutical Wastewater	Oxytetracycline Hydrochloride	Iron/aluminium anode (70 × 50 mm); Stainless steel cathode (70 × 50 mm); The gap between electrodes: 5 cm; 20 mAcm^2 current density; Time to react: 120 min; 0.19 kWh/L power consumption; 200–50 mg/L as a starting concentration.	82.96–93.17	[134]
Hospital Wastewater	Ciprofloxacin (CIP)	Aluminium anode and cathode; pH- 7.78; Inter-electrode distance: 1 cm; Time to react: 20 min; 12.5 mAcm^2 current density; 32.5 mg/L1 as a starting point.	88.57	[148]
Pharmaceutical Wastewater	COD	Aluminium anode and cathode (150 cm^2); Distance between electrodes: 25 mm; pH: 3–9; 1.7–1.9 mAcm^2 current density; The electrical voltage is 40 volts. 60-min response time.	34.2	[21]
Water Containing Heavy Metals	Arsenic	Aluminium (both the anode and the cathode); Bipolar electrode configuration.	93	[149]
	Chromium (VI)- Pb	Electrodes SS–SS; Current density (A/m$_2$) 73.5; pH 3.5; Content: 55.3–3.5 [mg/L]; Electrodes SS–SS; Current density (A/m^2) 73.5; pH 3.5;	91.7–91.3	[150]
	Perfluorobutane sulfonate (PFBS) Perfluorohexane sulfonate (PFHxS) Perfluorooctane sulfonate (PFOS) Perfluoroalkane sulfonic acids (PFSAs)	Electrode: Al-Zn Voltage supply:12v pH: 7	87.4 95.6 100 100	[151]

6. Combined Electrocoagulation Processes

Several combined EC processes have been applied to increase pollutant removal efficiency and compensate for the cost. In these processes, EC can be applied with an-

other treatment method as a pre/post-treatment unit depending on the type of pollutants present [47–49,152]. In addition, an overview of selected studies on different PWW treatment using EC/combined EC with pollutant removal efficiency at optimum condition is summarized in Table 5.

6.1. Combined EC/adsorption (CEA) Process

CEA is a relatively new technique and efficient in terms of cost and performance [153]. The authors analysed the removal efficiency by applying the EC treatment process followed by bioadsorption using Al as an electrode and Ectodemis of Opuntia as bioadsorbent. It reports that removal of COD was 50% by EC and a further 60% by bioadsorption. The colour removal was also higher in the combined EC/bioadsorption process. Recently, several works have been published on combined EC/adsorption, mainly on industrial wastewater. Aouni et al. reported removing COD, colour, and turbidity by 98.33%, 98.37%, and 100%, respectively, using a combined EC/adsorption process from textile wastewater [154].

6.2. Combined Chemical Coagulation (ECCC)/EC Process

Several studies were done on the removal of pollutants by combining EC/chemical coagulation with the help of suitable coagulants before EC to enhance performance and reduce the cost [155,156]. Can et al. (2006) studied the impact of two different coagulants before EC for removing COD and achieved a maximum removal of 80% [157]. Recently, Muharam et al. (2017) investigated the performance of EC, CC, and combined ECCC on medical wastewater. Authors reported removing total organic compound (TOC) using CC, combined ECCC, and EC processes by 41%, 92.21%, and 62.51%, respectively [158]. These results indicate that the combined EC/chemical coagulation enhances different pollutants' removal efficiency, including pharmaceuticals from wastewater.

Table 5. Summary of EC applied for wide range of PWW with pollutant removal efficiency at optimum condition.

Type of Wastewater	Electrode Material(Anode-Cathode)	Optimal Current Density	Mode of Electricity Application/Type	Electrode Arrangement	Electrodes Spacing	Initial Concentration	Initial pH and Temperature	Treatment Time	Pollutant Removal	Source
Synthetic PWW	Al-Fe	0.5 mA/cm^2	Intermittent (5 min ON/20 min OFF)	-	5 cm	10 mg/L	pH = 7.5, T = 25 °C	38 h	Diclofenac = 90%, carbamazepine (CBZ) = 70%, Amoxicillin (AMX) = 77%	[24]
Pharmaceutical industry wastewater	Fe-Fe	15 mA/cm^2	Combined EC (EC/electro-Fenton)	MP-P	2 cm	COD = 4000 mg/L, TOC = 1200 mg/L, BOD = 900 mg/L	pH = 7 T = 25 °C	2 h	COD = 70.2%, TOC = 64%, BOD = 97%	[47]
Pharmaceutical effluents	Al-Al	46.83 mA/ cm^2	Continuous electricity application	-	1 cm	Conductivity = 784 µS/cm, turbidity = 784 NTU, COD = 525 mg/L	pH = 5.31,	18 min	Turbidity = 96.7%, COD = 70.8%	[159]
Synthetic PWW (Oxytetracycline hydrochloride)	Al-Al Fe-Fe	20 mA/ cm^2	Continuous electricity application	MP-P	5 cm	50 mg/L	-	120 min	87.75% 93.20%	[134]
Drug industry	Al-Al	80 A/m^2	Combined EC (EC/anaerobic process)	-	1 cm	COD = 34,400 mg/L, BOD = 22,000 mg/L	pH = 7.2	25 min	COD = 24%, BOD = 35%, Colour = 70.25%	[48]
Pharmaceutical factory wastewater	Fe-Fe	763 A/m^2	Combined EC (EC/photocatalysis)	MP-P	2 cm	-	pH = 6.0, T = 25 °C	90 min	Turbidity = 91%, COD = 86%	[49]
Artificial PWW (Amoxicillin)	Al-Al	0.7 A	Combined EC (EC/nanofiltration)	MP-P	1 cm	50 mg/L	pH = 2.5, T = 25 °C	60 min	52.7%	[160]
Pharmaceutical effluent	Fe-Al	0.04 A	Continuous electricity application	-	1 cm	COD = 7692 mg/L, TDS = 16,290 mg/L, chloride = 9017 mg/L	T = 25–27 °C	15 min	COD = 92.3%, TDS = 91.5%	[161]
Synthetic PWW (Amoxicillin)	Al-Al	-	Continuous electricity	BP-S	2.5 cm	10 mg/L	pH = 7	75 min	98.8%	[162]
Berberine hydrochloride (BH) wastewater	Fe-Fe	19.44 mA/cm^2	Pulse EC	MP-P	2 cm	BH = 1500 mg/L	pH = 7	3.5 h	BH = 72.8%	[163]

6.3. Combined Membrane/EC Process

Regardless of the several types of EC combination, there are few studies on the combination of membrane separation with EC [164,165]. Hakizimana et al. (2017) studied pollutant removal in seawater using EC pretreatment in combined membrane/EC. The authors concluded that this method was cost effective, particularly for the seawater, due to high conductance [99]. Another study on the membrane-assisted EC process was investigated by Sardari et al. (2018). In this study, EC followed by direct contact membrane distillation (DCMD) was investigated. The authors concluded that the combined EC-DCMD process was efficient for the removal of organic matter and suspended solids. Further, they reported that this method could maximize water recovery and minimize the concentration of brine [166].

6.4. Combined Sono/Electrocoagulation Process

It is a combined process of EC with ultrasound irradiation that promotes agitation resulting in homogeneity in the system. However, simultaneous ultrasonication sometimes produces undesirable effects such as destruction of the produced colloidal hydroxides and their adsorption layer by ultrasound waves, reducing the removal efficiency [167]. However, the combined process has several advantages: the production of free radicals intense mixing and the reduction in anodic passivation effect if a controlled frequency is applied [168,169]. Raschitor et al. (2014) reported 95% removal by combined sono-EC, while the removal was only 60% using EC alone [169].

7. Cost Analysis

Any wastewater treatment method must be cost effective. Therefore, techno-economic analysis plays a significant role in proposing a process of treatment. Limited work has been done on the cost analysis during electrocoagulation, particularly for contaminants removal from PWW. The utilization of electricity is an essential aspect in influencing the operational cost in the EC process. To be industrially scalable, this technique must be economically viable [170]. Thus, a low-cost EC technology is required to scale up on an industrial level [171]. Cell voltage, reaction time, and current can all be used to calculate electricity utilization in an EC operation. When electric current density exceeds the optimum amount, undesirable responses will arise, extra oxygen will expand, and cell energy will be increased, resulting in increased power utilization and operating costs [172]. Apart from the costs of electricity, the expenses of chemicals and sacrificial electrodes are the other main components used in calculating the cost of an electro-chemical device [130,173,174]. Thus, energy usage, complete dissolution of the electrode, any cost add up of chemical substances (adjustment of the conductivity or altering solution pH), and cost of disposal and transportation of sludge are all part of the EC process costs. However, the use of a renewable energy source and process optimization could reduce the overall cost. Several studies have been conducted on the reduction in EC process cost. For instance, [175] reduced the overall EC cost for Cr (VI) treatment using multivariate optimization of a process variable. In another study, using response surface methodology (RSM), all process affecting variables optimized to make the treatment cost effective [176]. Some of the typical cost analysis on the EC process are included in this review. The operational expense of treating real dye house wastewater with continuous flow electrocoagulation was calculated by M Kobya et al. (2016) [133]. The cost of operation was shown as energy expenses, cost of electrode materials, and chemical utilization expenses. Expenses of utilizing electrode of aluminium were estimated as 1.851 USD/m^3 in comparison to 1.562 USD/m^3 using electrodes of iron at the optimum working conditions of 80 min of operation, current density of 65 A/m^2, and 0.010 L/min of flow.

Espinoza et al. (2009) examined the efficacy of electro-coagulation in tannery wastewater treatment [177]. Electrocoagulation was more cost effective compared to other conventional methods and achieved the required removal. More specifically, the estimated expenses of electro-coagulation were 1.7 dollars per m^3 of treated effluent versus 3.5

dollars per m^3 using the conventional process. Khaled et al. (2019) looked into the effects of different design parameters of reactors on the performance and operating costs of electro-coagulation to remove cadmium from wastewater [178]. Electrodes distance, electrode attachment mode, stirring speed, surface area to volume ratio (S/V), and initial temperature were all investigated. The expenses of electrodes (aluminium), electricity, and chemicals applied to alter pH were all considered in this study's operating costs. At specific conditions of 0.5 cm inter-electrode distance, 50 degrees Celsius initial temperature, stirring speed of 300 revolution/min, ratios of S/V as 13.5, and monopolar connection mode, 100 percent cadmium extraction was achieved with less power utilization and a minimum operating expenses of 0.116 TND (Tunisian National Dinar, equal to 0.06 USD). The authors also stated a cost of 4.36 TND (2.1 USD) for chemical coagulation, demonstrating that electro-coagulation is more cost effective. Some of the operating costs in different wastewater using EC are summarized in Table 6.

Table 6. Few EC based studies indicating operation cost in different condition.

Type of Wastewater	Initial Conc.	Electrode Material	Removal Efficiency	Current Density	Operating Cost	Reference
Synthetic Wastewater	100 mg/L	Al	80–95%	208–310 A/m^2	0.34–0.52 USD/kg dye	[179]
Synthetic Wastewater	50 mg/L	Al Fe	87.5–93.4% 90.7–98.1%	155–350 A/m^2 155–350 A/m^2	7.04–17.4 USD /kg dye 4.01–13.8 USD/kg dye	[180]
Coal Mine Drainage	–	Fe	28.7–99.96%	200–500% A/M2	1.09–2.184 USD/m^3	[181]
Textile Dye Wastewater	3422 mg/L COD	Al Fe	15–62% 57–78%	50–200 A/m^2 50–200 A/m^2	0.32–0.58 USD/kg COD 0.7–0.175 USD/kg COD	[182]
Textile Dye Wastewater	2031 mg/L COD	Al (MP-P) Fe (MP-P)	- -	30–60 A/m^2 30–60 A/m^2	0.4–0.65 USD/m^3 0.25–0.4 USD/m^3	[183]
Wastewater from Metal Industries	3155 mg/L TOC, 17,312 mg/L COD	Al Fe	93% COD 92% COD	60 A/m^2 60 A/m^2	0.768 USD/m^3 0.479 USD/m^3	[184]
Metal Removal from Soil Leachate	-	Fe	99.4% Zn^{2+} 99.7% pb^{2+}	68 A/m^2	35.38 USD tst-1	[185]

8. Challenges and Suggested Mitigations

Though EC is not a new technology for wastewater treatment, most of these pollutant removal studies have been performed on a laboratory scale with pollutant centred approach. To make a feasible treatment method on the industrial level, EC technologies should be scaled-up [143]. EC scale-up has several challenges, such as deterioration of cathode performance by chemicals, anodic biofilm reduction, short-term stability, fouling, distortion of chemicals, optimization of the process, and system clogging by solid contaminants. Still, several attempts have been made to optimize the transition from bench scale to a pilot plant [186,187]. In addition, most of the work has been done on synthetic wastewater, which creates ideal conditions without the interference of other contaminants. For this reason, some extensive research is required using real pharmaceutical wastewater to investigate actual performance in the field and representative conditions [188].

Another problem with the EC technology is the high operating cost, including costly electrode material and electricity required [189]. Thus, future research is needed to improve anodic consumption and cathodic passivation [190]. However, this problem could be alleviated to some extent using scrap materials from the aluminium and iron industries. To compensate for the operational costs, the electric current recovery and hydrogen production possibilities are being explored. In addition, renewable sources such as windmills, solar energy, and biofuel could be used for cost-effective EC processes [191].

Even after several software aided optimization, the EC process parameters are not fully optimized yet [143]. In order to prove this method reliable for large-scale pharmaceutical wastewater treatment, more exhaustive studies are required in the field of pharmaceutical range and metabolic, process optimization, modelling, and scale-up. The sludge generated during the PWW treatment contains various harmful and toxic substances, which need sustainable end-use or extensive sludge management to prevent negative environmental impact. However, sludge reuse from the EC process is becoming attention in the last few years due to the presence of useful compounds such as metallic hydroxides [123].

In addition, the combination of the EC process with other technologies as pre/post-treatment might enhance the system performance. Several studies have been conducted on these combinations, such as EC adsorption [154], combined sono-EC [168,169], combined EC/chemical coagulation [161], and combined membrane/EC technologies [166]. They all observed enhanced efficiency. Therefore, future work must focus on this direction.

9. Conclusions

The drastic rise in pharmaceutical load in the water/wastewater has compelled researchers to investigate various treatment methods. Numerous pharmaceutical removal techniques such as advanced oxidation technology, membrane technology, bioremediation, activated carbon, and carbon nanotubes have been applied in the past. Out of these modern treatment methods, the EC treatment method is one of the best efficient methods for pharmaceutical wastewater. The simplicity in operation and ease in sludge handling also make it more persuasive. In addition, a combined EC treatment method is also possible as a pre/post treatment unit depending on the type of pollutant present, e.g., combined membrane/EC process, combined Sono/EC process, and combined EC/adsorption process. However, some constraints such as energy consumption, processing cost, and sludge handling are present. Other drawbacks such as electrode passivation and inconsistent coagulant dosing on long-period operation are also there. Still, this technology could be one of the best treatment options for pharmaceuticals after a potential advancement in process parameters.

For this reason, further investigations are required in the field of EC mechanism for the target pollutant, understanding of complex physiochemical phenomena, process parameter, possibilities of the scalable reactor, and sustainable energy sources. Different combinations of pretreatment and post-treatment with EC are more effective for various pollutants and need further investigation. Moreover, more studies are suggested in the field of pharmaceutical removal from wastewater by electrocoagulation using real wastewater.

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