

Recent Trends and Advancement in the Removal of Persistent Organic Pollutants from Wastewater by Hybrid Electrocoagulation Chemical Degradation Processes

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Abstract

EC is an emerging technique used for various applications, due to its benefits, such as coagulation, flocculation and Ec treatment. Emerging pollutants are a real concern, and many researchers have focused on their removal, using EC, due to its several advantages, such as easiness of operation, less space requirements, versatile pollutants removal, lower sludge production, cost effectiveness, lower energy consumption and environment friendliness. Literature has also determined that EC technology is a proven efficient technology for treating various nature industrial effluents. However, those emerging pollutants, such as pesticides, Polyfluoroalkyl substances, cyproflyxin, amoxicillin and surfactants are not efficiently treated by simple EC technology, and their separation is not the solution, since they require degradation and safe disposal. Therefore, waste disposal from this process is a major concern, and the choice of advanced EC-degradation technology depends upon the nature and strength of the pollutant. To address these issues, various processes coupled with EC have been used, such as EF, PDS, EO, ozonation, photochemical degradation, PC, aeration and sonication. Thus, this paper focuses on the basics of EC technology, several degradation systems and application of combined degradation-EC technologies, for treating various WW.

Keywords: di-sulfate oxidation; Fenton process; ozonation; PC; per-sulfate.

Introduction*

The shortage of water resources, and polluted WW, are undeniable global challenges of XXI century (Uncertainty 2012). The treatment of water bodies is the only solution to reduce WW pollution. In the recent past, physical and chemical technologies have been combined to produce water free from pollutants [20]. Nowadays, due to technological advancement and industrial activities, pollutants are different from those of ancient times. The presence in trace amounts of emerging pollutants from personal care

* The abbreviations list is in pages 410-411.

products, surfactants, pharmaceutical active compounds, hospitals [2, 46], industries [7, 44, 58], municipal WW treatment plants [27], run-off or seepage from sewers [50], micro pollutants [77, 86] and landfill leachates [88] affects the environment [45]. So, the treatment of emerging pollutants through modern technologies is paramount. Most researchers have focused on the principle, parameters of influence, treatment mechanism and different applications [37, 48, 73] of EC process, which has some limitations, such as: rapid consumption of sacrificial anodes, which requires electrodes replacement, after some time; reduced process efficiency, due to passivation; limited removal of organic compounds; and EEC cost. WW contains POP that are resistant to degradation by photolytic, chemical and biological processes, due to their persistence, semi volatility, hydrophobicity, bioaccumulation and toxicity, as identified by the Stockholm convention [17]. Disposal of untreated pollutants into the environment can have harmful impacts. To address this issue, EC is often combined with other processes [75], such as chemical degradation technologies, ozonation, Fenton, photo-Fenton and anodic Fenton processes, photolysis, photo-catalysis, PC, PCM and PDS techniques, self-power air cathodes, aerated EC and use of sustainable power sources, to meet energy requirements. These combinations have several advantages, such as improved removal of contaminants and decreased energy consumption. EC is also effective in treating emerging contaminants like POP, micro plastics and nanoparticles. This review focuses on the fundamentals of EC technologies, including advanced processes with *in situ* generation of free radicals or any other oxidant species, to enhance pollutants RE. It also discusses the combination of other processes with EC, in order to develop the performance and applicability of novel and revolutionary ideas that may increase emerging pollutants RE.

EC principle

EC has achieved momentous attention in the area of synthetic and natural WW treatment. This technique splits the stable emulsion and suspension in a solution by salt polymers or polyelectrolytes. The metal ions generated at the anode help to attract pollutants, after the reduction of water molecules at the cathode. H₂ gas generated at the cathode lifts coagulant particles at the surface, which are then separated when they settle down at the bottom by gravity action (Fig. 1 and eqs. 1-3).

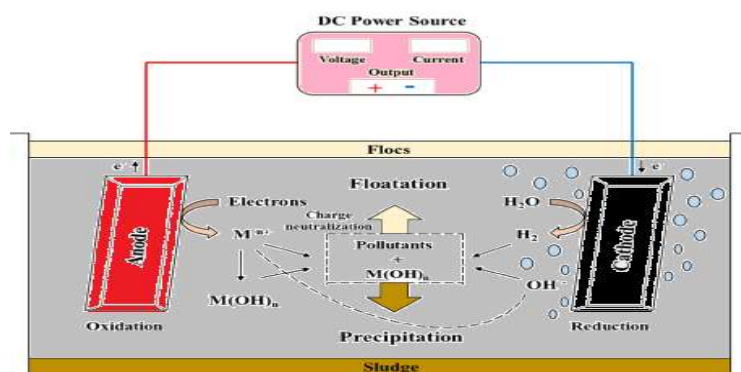
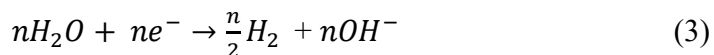
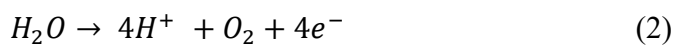


Figure 1: EC setup.



where M is metal to be oxidized, n is number 0,1,2,3... and e⁻ is electron.

Concept of chemical coagulation and EC

Chemical coagulation is a process carried out when small particles in the size range from 1 nm to 2 μm are dispersed in a solution, and do not settle by gravity, because they are so tiny and also pass through filter paper. Two forces are responsible for colloidal particles stability: repulsive force; and weak Van der Waals force of attraction. Permanent induced dipoles generated by Van der Waals forces require short distances. Electric double layer, stern layer and diffuse layers are generated by repulsive forces [61, 81].

Electric double layer

The presence of charge particles in ions suspensions will electrostatically attract opposite charge ions. The primary surface charge attracts opposite charge ions and, then, diffusing counter ions, to produce an electric double layer [6, 90]. When colloids are exposed to an electric field, they tend to move towards a positive electrode. The particles in the solution with opposite charges move towards the electrode, leaving the diffuse external layer (Fig. 2) [39, 79].

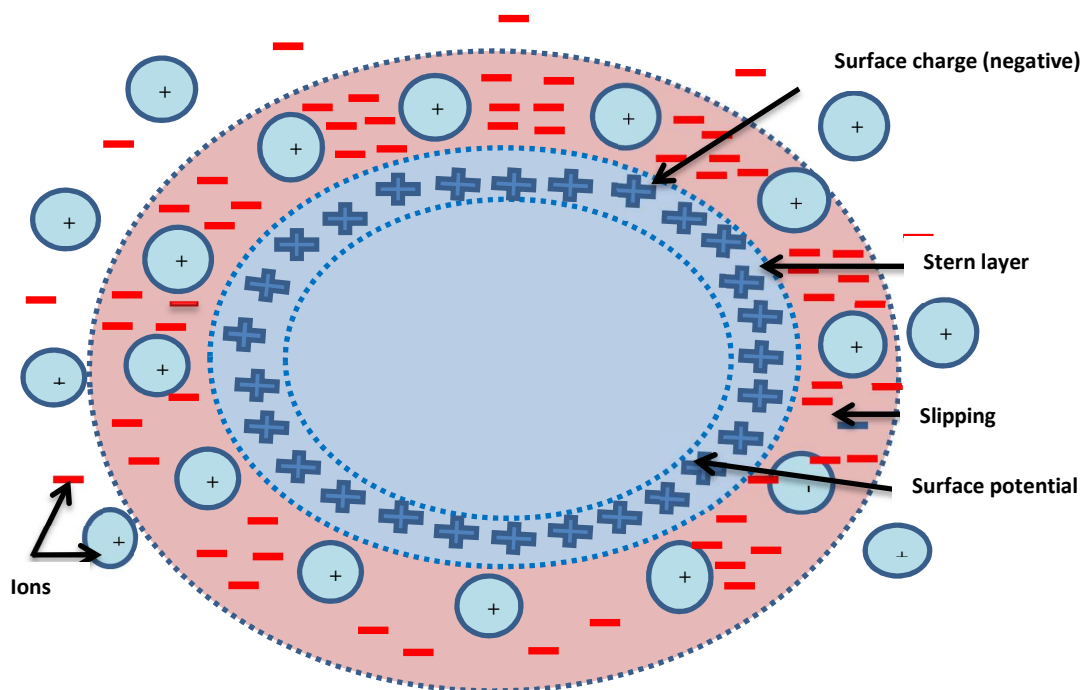


Figure 2: Electric double layer [39].

The difference between the charge at the electrode surface internal and external layers is known as zeta E. The interaction among colloids is due to the charges.

Derjaguin–Landua–Verwey–Overbeek theory

This theory is used to study colloids in suspension [61]. The net force that acts on colloids is due to the electric double layer formation, which explains the equilibrium of colloid particles in suspension [81]. Opposite charges of equal magnitude balance the initial charge present on colloids. Net force is the addition of Van der Waals attraction and electrostatic repulsion energies, which depends upon distance between particles [61, 73].

Factors affecting EC process

EC process performance depends on the types of electrodes, their configuration, the distance between them, the solution initial pH and J. The operational parameters variation allows for WW treatment. So, their optimization is a main issue during EC process [9]. The electrolysis effect is exerted during the process, causing the pollutants separation from contaminated water. The efficiency of the pollutants EC treatment process depends on the optimization of those parameters.

Initial PH

The solution initial pH has a major influence on EC process. When pH changes from acidic to alkaline, there is an increase in RE, because metal OH^- are produced. Using an Al electrode, [15] studied pH and TOC impact on RE. They found that maximum RE of 65% PO_4^{3-} occurred at pH 5, and that it decreased at higher pH values, due to H gas production at the cathode. The outcome demonstrated that optimum pH value was from 6 to 7, due to $\text{Fe}(\text{OH})_2$ production, as shown in Fig. 3.

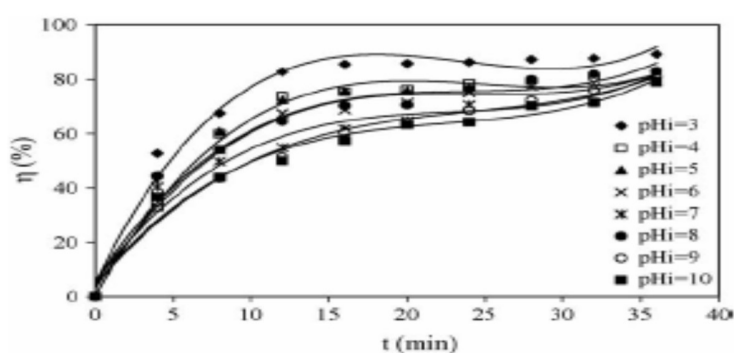


Figure 3: Effect of pH on PO_4^{3-} removal from WW. Reprinted with permission from [41] (Copyright 2006, Elsevier).

Electrolysis time

The key factor affecting RE is the length of the electrolysis process. As the electrolysis time lengthens, RE also increases up to a point, before becoming constant, as more metal OH^- are formed over time. In under 50 min, [28] were able to remove 92% cefazolin and COD from hospital WW (Fig. 4).

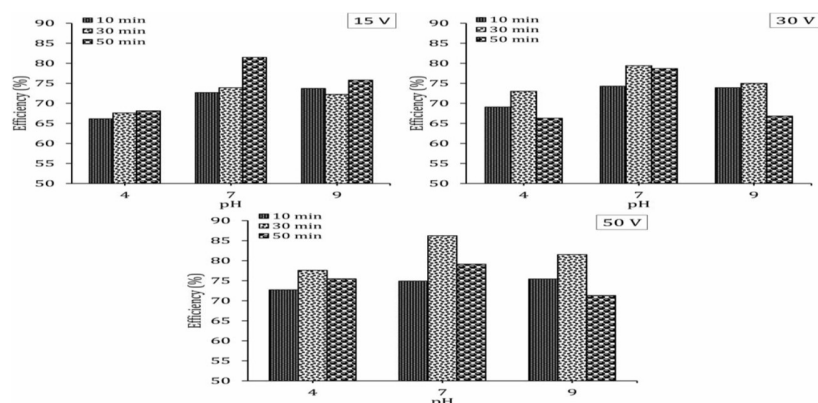


Figure 4: Effect of operating parameters (pH, V and reaction time) on cefazolin RE by EC process. Open access [28].

[9] removed more than 90% PO_4^{3-} , for 140 min, at an electrode distance of 0.5 cm, pH 7, 6 mM Ct of NaCl, temperature of 30 °C and J of 10 mA/cm². They determined that RE increased with stronger electrolysis (Fig. 5).

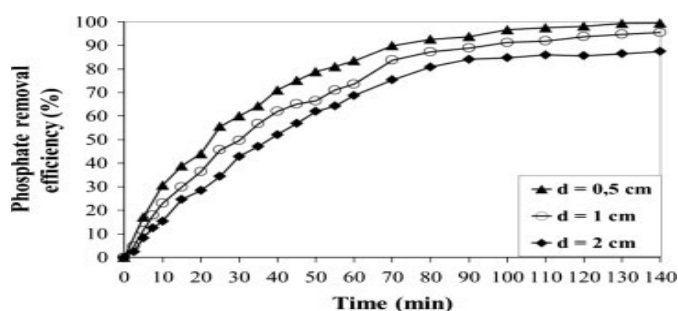


Figure 5: Effect of electrodes distance on RE of PO_4^{3-} . Reprinted with permission from [9] (copyright 2014, Elsevier).

J

With the anode higher dissolution, its reaction with the pollutant in the solution increased with increased J, during EC. [15] found a relationship between TOC removal and J (Fig. 5). The authors obtained maximum and minimum pollutant removal at 100 and 12.5 mA/cm², respectively. When J was changed from 50 to 100 mA/cm², RE improved from 28.5 to 34.42%. [63] worked with high J (605, 908, 1211, 1513 and 1816 A/m²), using EC to treat sewage WW. COD, BOD and suspended solids RE obtained were 98.07, 98.07 and 97.64%, respectively, at pH 7 [63].

Electrode distance

Electrode distance and surface area have great impact on RE. With a decrease in distance between electrodes for effluents with high electrical conductivity, J was kept constant, to reduce power consumption. [9] studied the effect of the distance between electrodes on RE of PO_4^{3-} from WW. They determined optimum operational

parameters as 5 mm electrode distance, initial pH 3 and 3.2 Ms conductivity, respectively (Fig. 6).

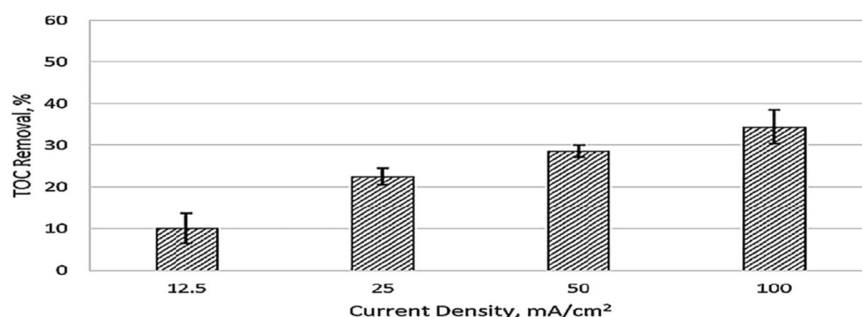


Figure 6: Effect of electrodes distance on RE of PO₄³⁻. Reprinted with permission from [15] (copyright 2019, Elsevier).

Advanced EC-degradation processes strategies

Nowadays, recent advances in industrial sector produces different emerging pollutants which cannot be treated by simple EC process. Therefore, research combines various EC processes with degradation methods. The combination has various advantages and disadvantages, depending upon the nature of the pollutant being treated. Herein, different advance degradation methods will be discussed in combination with EC processes. Table 1 shows the treatment of various emerging pollutants by combining EC-degradation technologies in different countries.

Table 1: Treatment of various emerging contaminants and POP by hybrid-EC-degradation technologies.

Reference	Country	WW	Contaminant	Operating conditions	RE	Treatment process
[3]	Turkey	Aqueous solution	Hydroquinone	J: 5 A/m ² ; UV light power: 24 W (W/dm ³ solution); TT: 60 min	91.5%	Photochemical oxidation + EC
[21]	Spain	Synthetic WW	Aniline	Ti/Pt anode, O diffusion cathode, time: 5 h, 20 A	95%	EF with PC
[40]	Pakistan	Natural veterinary pharmaceutical WW	Enrofloxacin and Amoxicillin	TT: 60 min; E difference: 5 V; pH 7; 0.4 mg/min O ₃	Enrofloxacin 100%, 90% Amoxicillin and 85.12% COD	Synergic photochemical ozonation + electro flocculation
[57]	Iran	Steel industry WW	Phenol	Fe ²⁺ /H ₂ O ₂ : 1.5; 29.15 kWh/kg COD; electrolyte Ct Na ₂ SO ₄ : 10 mg/L; TT: 25 min; J: 1.5 mA/cm ² ; pH: 4; Al anode and Fe cathode	100%	EC + photo-Fenton
[72]	China	Synthetic WW	Berberine hydrochloride	Reaction time: 3.5 h; pulse duty cycle: 0.3; pulse frequency: 1.0 kHz; J: 19.44 mA/cm ² ; distance between electrodes: 2.0 cm	72.8% and COD 69.6%	Pulse EC
[87]	Iran	Synthetic WW	Azithromycin	J: 20 mA/cm ² ; H ₂ O ₂ Ct: 2 mM/L ⁻¹ ; energy consumption: 2 kWh (kg COD) ⁻¹ ; pH 3.0	COD 95.6%	PC process
[92]	China	Aqueous solution	Bisphenol	Gr felt Ti cathode; Al anode; J: 2.3 mA/cm ² ; TT: 20 min; enzymatic activity 25 U, at 30 °C; 3.5 kWh/Kg bisphenol	90%	Electroenzymatic catalysis + EC

EF is a process in which OH^\bullet ion is generated at the cathode, by reaction of H_2O_2 with Fe electrode [59, 66]. It is an example of advanced Ec oxidation processes (Fig. 7). This process allows to degrade organic pollutants until complete demineralization, but if any by-product remains in the water, it can be separated from WW through EC, since H_2O_2 use does not increase much the costs. At the same time, the generated sludge can be safely disposed, because there is no toxicity. However, some researchers say that the time for degradation in combination with EC is a major factor to efficiently perform the process, such that the generated byproducts will not be toxic or less harmful than parent pollutants present in the water.

In addition to that, EF process increases the solution pH to alkaline values near the cathode, due to HER. The combination of electro-coagulation process with EF makes the solution pH to decrease near 7 through OH^- ion oxidation at the anode [65].

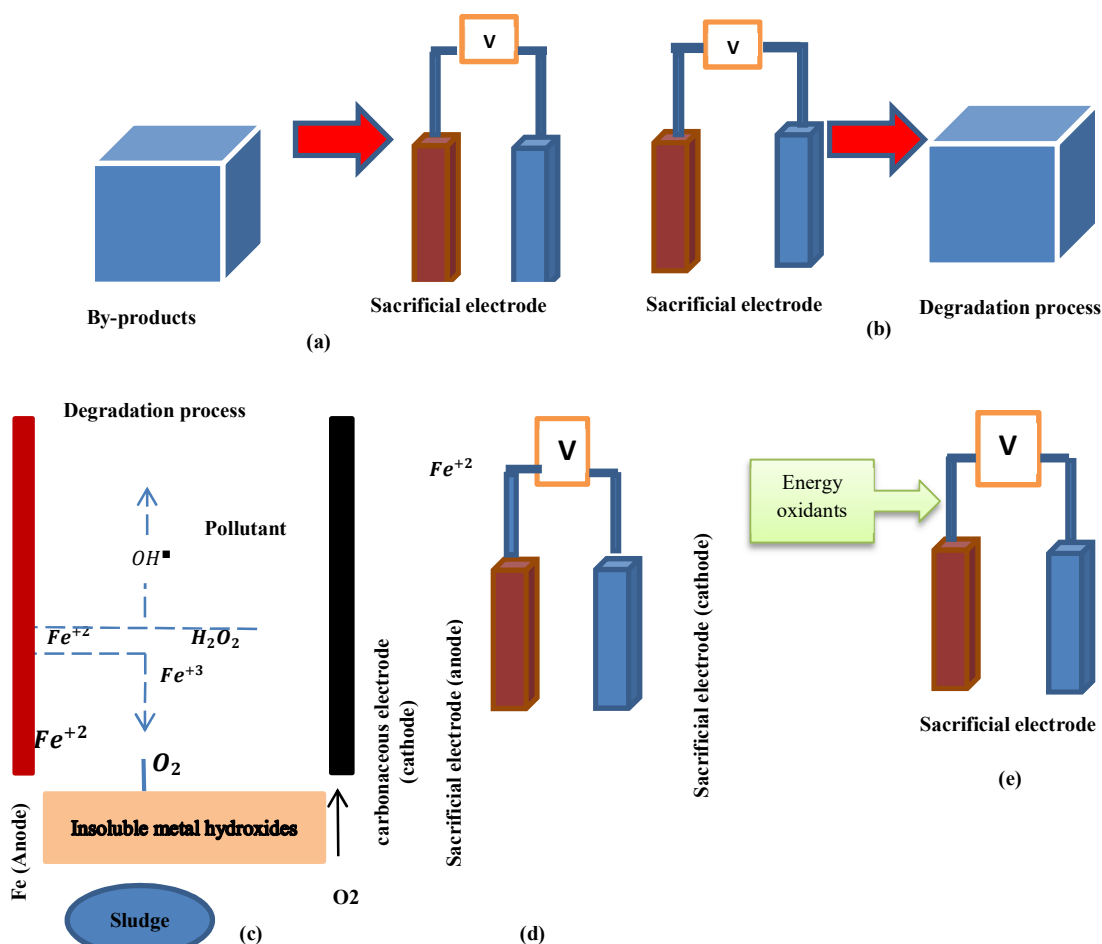


Figure 7: Pre-degradation before EC; EC followed by degradation; degradation and EC in one electrolytic cell [65].

Fig. 7(b) shows EC process carried out after the degradation process. Then, EC and degradation are combined. In this process, EC is carried out first than degradation,

which is effective for high strength contaminated WW. Treating high strength WW poses a significant challenge [83]. However, one approach to address this issue is by employing an initial treatment method such as the EC process, which reduces pollutant Ct in the WW. Pretreatment plays a vital role in facilitating the degradation process during subsequent stages. Pretreatment will support the degradation process for further treatment. In this combination, some toxic compounds are produced, due to the production of sludge. Figs. 7 (c-e) show the different methods of EC and degradation processes in an electrolytic cell. PC shown in Fig. 7(c) is the best example for these types. Fe anodes used in PC favor EC and EF treatments in a single electrolytic cell. To increase the degradation efficiency, some amount of acid is used, since pH increases during the process. An electrode with high O overpotential, or a stable anode and cathode as sacrificial electrode are another options. Fig 7(d) shows that this can be used for direct or indirect Ec oxidation. In the later, changing polarity decreases EC passivation leads. [91] has used arsenate from water.

Application of advanced EC-degradation treatment technologies

Aerated EC of Fe

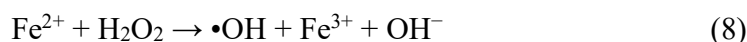
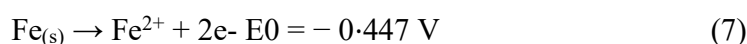
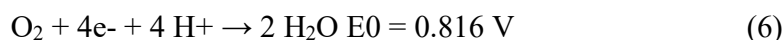
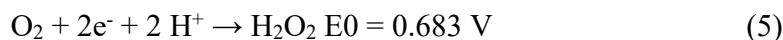
The presence of dissolved O in the electrolytic solution is essential for the EC method effectiveness. Without aeration, pollutants RE becomes lower over time, as dissolved O levels drop. According to studies, EC process performance and mechanism depend largely on dissolved O consumption [66]. [11] conducted further research on arsenate removal from groundwater that contained synthetic arsenite, employing continuous aeration with Fe. With an initial Ct of 1 mg/L arsenite, they performed trials at two different flow rates of 5 and 10 L/h, for twelve cycles of six h. Due to the development of a fouling layer on the electrode surface, after the fifth cycle, the removal performance declined.

EC effectiveness with and without aeration was compared by [12]. To treat the arsenite-contaminated solution, Fe electrodes were used in tests performed in a 5 L batch reactor. The study discovered that external aeration considerably improved the arsenic RE from aqueous solutions. [11] made a similar observation, noting an improvement in RE with external aeration.

Self-powered air cathode EC

The cost and environmental impact of EEC, particularly for low conductive streams, is one of the main issues with EC for water and WW treatment. The use of external aeration to enhance RE in EC leads to increase energy consumption [56]. To address this challenge, a new approach called ACEC has been introduced. ACEC replaces the traditional metal cathode with an air diffusion cathode, transforming the EC cell from an electrolytic into a galvanic system. This modification enables the EC system to become self-powered, generating its own electricity, while effectively removing contaminants. ACEC combines EC with Ec advanced oxidation processes, offering a promising solution for efficient contaminant removal with power generation, eliminating energy consumption issues associated with traditional EC. However, ACEC implementation still faces several challenges. The use of an air cathode makes it more difficult to scale up and design the reactor, since each cathode in an

EC stack needs to be supplied with air through a distribution system. Additionally, stability fouling degradation and lifespan of air cathodes in ACEC system require further investigations to ensure their long term performance.



Ozonation

The ozonation process degrades pollutants by direct oxidation or free radical generation. Ozonation combined with other methods enhances the process performance, by the degradation mechanism that originates a highly reactive species such as OH^\bullet . It can remove color, odor, TOC and COD of industrial effluents [13, 51]. The combined process of EC with ozonation involves direct interaction of Fe ion and O_3 for $(\text{FeO})^{2+}$ intermediate production. The intermediate species generates COH, which will reduce toxicants in industrial effluents, due to the presence of refractory compounds [8]. Therefore, EC assisted by O_3 enhances the performance of color decontamination and COD. [38] combined ozonation and EC. Initially, ozonation was applied, followed by EC, for azo dyes removal from aqueous media. 97% decolonization and 85% TOC removal were obtained at J of 15 mA/cm^2 , pH 6.5, salt Ct of 100 mg/L , O_3 flow rate of 20 mL/min^{-1} and electrode gap of 3 cm.

Fenton process

According to Fenton process, an OH radical is produced when Fe catalyzes H_2O_2 breakdown [67]. This technique is one of the most advanced oxidation methods for refractory materials degradation, due to its simple operation, short reaction times with ecologically friendly chemicals, and absence of energy needs for H_2O_2 creation [93]. In the classic Fenton reaction, the catalyst (Fe^{+2}) and reactant (H_2O_2) are in the same liquid phase, and the catalyst is composed of particles that are insoluble in Fe [64].



Most researchers have worked in a sequence form in which EC is initially carried out for pretreatment. Afterwards, Fenton activity will be carried out, enhancing the target pollutant RE. [43] used sequential process for treating yarn dyed WW [14, 43]. 20% enhancement in COD removal and complete Zn removal was achieved using $20 \text{ mM H}_2\text{O}_2$ in the system. The limitation of using combined EC with Fenton process occurs when inorganic elements are present in WW, because they suppresses OH radical generation, creating other toxic compounds that will affect the overall removal of heavy metals and organic pollutants [22].

Photolysis, photo-catalysis, and photo-Fenton process

Photo-Fenton is the process in which UV irradiation increases OH radical generation, Fe^{+2} production by Fe^{+3} reduction (visible irradiation $< 580 \text{ nm}$) and

H_2O_2 photolysis (UV-C < 310 nm) [24, 85]. Photo-catalysis is a process in which water generates free radicals that will react with the pollutant to degrade it, mostly employing semiconductor materials. Semiconductors are excited by light energy greater or equal to the band gap (Fig. 8). This will generate the hole pair of which further reaction will take place in the aqueous phase [47].

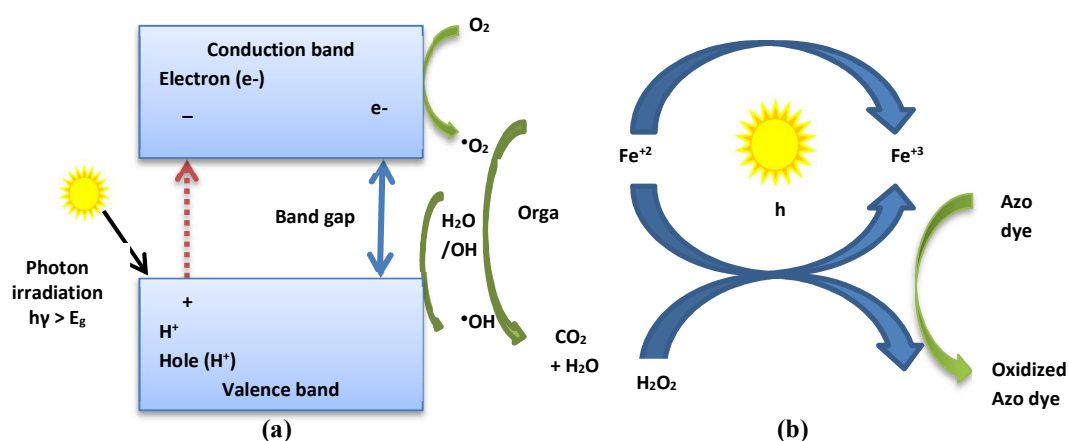


Figure 8: (a) Photo-catalytic oxidation process on the semiconductor surface. Reused with permission from [91] (copy right 2015, Elsevier); (b) Azo dye degradation in photo Fenton process. Reused with permission from [10] (copy right 2019, Elsevier).

For tannery effluents treatment, [60] investigated the use of EC and photo oxidation processes in succession. Flow through mode with SS anode and cathode electrode surfaces of 115.5 cm^2 was used. Photo-oxidation was supplied by a radiation source of 12 UV lamps, with maxima emission at 254 nm, and analyzed in circulation mode. The results showed that EC alone removed 71.8% suspended solids and 71.8% COD, at pH 7, in 20 min. EC-UV-visible photo reactor, at 254 and 185 nm, removed 99.52% COD, 100% Cr and 98.27% sulfide, in 40 min.

A number of processes are involved in the mechanism of EC-UV photo-catalytic oxidation, including the creation of metal flocs and OH^- , Cr (III) reduction to Cr (VI), followed by co-precipitation, and S^{2-} , because Al_2S_3 may be trapped in flocs and eliminated from the supernatant. EC effluents are subjected to photolysis. According to [60], the vacuum UV plays a critical role in degradation, particularly in the direct photolysis of organic pollutants, and in the formation of ROS via H_2O hemolysis/photoionization [1, 60].

Anodic Fenton processes

Indirect Ec oxidation is a process in which the pollutant is oxidized by intermediate species generation from water and other contaminants. Most researchers select anodes with high O_2 evolution E for Ec oxidation, because these electrodes are able to generate $\cdot\text{OH}$ radical [1]. Most frequently used materials for the anode are Pt and BDD, since, as they are very effective in OH radical generation, they have better

capacity to oxidize organic pollutants and treat industrial effluents. Most frequently, chlorine is produced during Ec process by oxidizing chloride, which is extremely insoluble in water [20-21, 59]. Chlorine diffuses over the anode, producing HClO and Cl^- [12]. [16] combined EC and EO for color and COD removal from tannery WW. The anode was made of Fe/Al plates with an effective surface area of 120 cm^2 . Metal OH^- production and COD elimination linearly depend upon J up to 17 A/m^2 . Above this value, metal OH^- and COD removal will become constant. The results showed that COD removal with Fe and Al electrodes was 62 and 82%, respectively. The hybrid process of EC and EO, at J of 42 A/m^2 and time of 40 min, reduced COD to 5 mg/L [16]. [54] has done research combining EC and EO, to produce a synergic effect for POP decontamination [54]. This work also compared individual EC and EO. EC process, at operating conditions of pH 8 and J of 800 A/m^2 , using Fe electrodes, eliminated 100% suspended solids, but it was ineffective against COD removal. Electro oxidation with a dimensionally stable BDD anode and Fe cathode, at pH 7.2 and J of 800 A/m^2 removed 100% COD in 21 h. However, since EO alone took more time to remove COD, the authors focused on developing a synergic effect. First, they performed EC for 30 min, and then EO, for 90 min, at pH 8 and J of 800 A/m^2 , which completely removed COD, BOD_5 , color, turbidity and coliforms. [62] worked on developing a synergic effect by combining EC and EO for treating textile industry effluents, and estimated RE of EC. The used electrodes were Al and Ir coated Ti oxides plates, at I of 0.6 A and constant pH 6. The obtained RE results were 90% COD, 87% BOD, 95% oil and grease, 98% suspended solids, 98% turbidity and 96% sulfate [62].

PC process

PC is the process of developing a synergic effect between EC and EF methods in a single reactor [1]. The process comprises a Fe electrode anode that releases Fe ions into the EC system, after combination with OH^\bullet ions generated at the cathode. The cathode comprises a gas diffusion electrode, where O is externally supplied. Then, O will react with H ions generated at the cathode, to form H_2O_2 (Fig. 9). H ions generation will depend upon the material nature used as cathode. The main reaction of the process is shown in Fig. 9 [33].

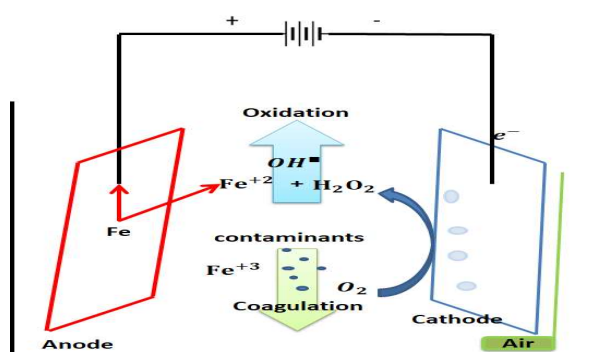
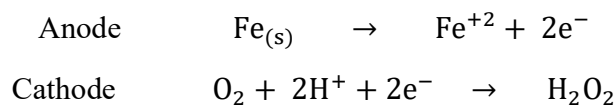


Figure 9: PC and EF process.

Fenton reagent allows the degradation of organic pollutants through $\bullet\text{OH}$ mediated Fenton process. At the same time, Fe ions produce Fe OH^- to make Fe OH^- pollutants complex [66].



Four main steps are performed in the process: oxidation, neutralization, flocculation and sedimentation. The first two steps occur in EF process, and the last two in EC. [11] conducted an experiment for olive oil mill WW treatment, combining EC and electro-flotation approaches. Initially, the treatment was carried out by EC, using Fe electrodes, which gave 40% TOC removal in 20 min. Then, electro-flotation was done using a BDD/air diffusion cell. The operating conditions for electro-flotation were pH 3, J of 25 mA/cm^2 and time of 36 min. The procedure was able to remove 81.5% TOC. The pH has little effect on electro-flotation performance, because BDD is responsible for free radical generation, rather than for Fenton activity [29, 32]. A similar study obtained 21% increase in TOC removal using Fenton, for 60 min. PC results were worse [67]. Literature has determined that PC process has some limitations, such as pH dependency, high energy consumption and huge Fe sludge production [52].

Persulfate and disulfate oxidation

$\text{SO}_4^{\cdot-}$ radical has oxidation E close to that of $\bullet\text{OH}$ ($E_0 = 1.8 \text{ V} - 2.7 \text{ V}$). It has better stability dispersion to greater distances, long working pH and longer span life. $\text{SO}_4^{\cdot-}$ radical is an interesting option for advanced oxidation processes like driven advanced OH radical [91, 74]. $\text{SO}_4^{\cdot-}$ radical activation can be ignited by various methods, such as temperature, UV light, ultrasound waves, transition metals and zero valent metals [62, 80]. $\text{SO}_4^{\cdot-}$ radical generation can also be carried out by ferrous ions production by Ec reaction [42]. EC combination with $\text{SO}_4^{\cdot-}$ radical advanced oxidation processes is the better option for this type of practices. $\text{SO}_4^{\cdot-}$ radical can be produced from PMS or PDS oxidation (Fig. 10) [91].

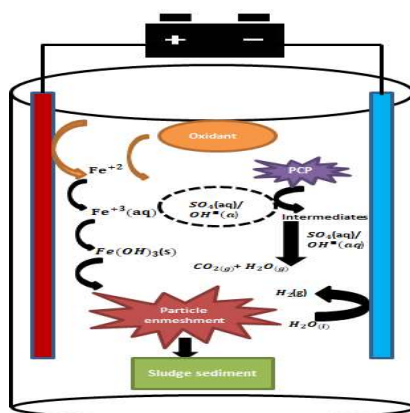


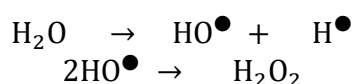
Figure 10: PDS addition for pollutants degradation. Reused with permission from [53] (Copyright 2016, Elsevier).

[49] determined that PMS has more promising results for pollutants degradation, due to the formation of hypochlorous acid and chlorine gas, if the chloride ion is activated by PMS. With PDS addition, contaminants are broken down. On the other hand, maintaining lower Ct of PMS in the system has a radical scavenging effect that can be recovered [49].

Fe⁺² ions can destroy SO₄²⁻ radical, which can be recovered by providing higher Ct of S₂O₈²⁻. According to [35], the hybrid technique of EC and SO₄²⁻ radical-based oxidation was found to be promising for pentachlorophenol, in an aqueous medium, and followed the order EC < PDS-EC < PMS-EC [35].

Sonication

Sonication is the process in which sound waves are employed for pollutants degradation. Normally, US alone is not the best option for contaminants removal. Various authors have shown that US combined with advanced oxidation processes is a better approach for removing pollutants from aqueous media [78]. The chemical reaction includes formation of •OH radical by hemolytic breaking of water molecules [36], when organic compounds are present in WW. •OH radical breaks organic compounds, and sometimes, it breakdowns into H₂O₂, by dimerization [36].



Physical effects include cavitation to overcome electrode weight loss, due to passivation, and a decrease in I performance with time. There are some main advantageous of combining US and EC: (i) byproducts or gases on the electrode surface avoid its passivation; (ii) crystal effects generation helps activating the electrode surface; (iii) diffusional resistance thickness at the electrode is reduced by lowering the reaction time and increasing I efficiency [69]; iv) and the friction between liquids and the electrode surface is increased by US, which causes its local amplification [4].

Nowadays, US is more employed by researchers for WW treatment [18]. Less investigators have worked on radical production in US driven EC. Most of them have used both EC and US, in a single reactor, at alkaline pH, which favors the combined process [84]. It is evident that US combined with electro-chemical treatments enhance the synergic effect for contaminants removal from WW [70]. [84] worked on synthetic WW by OH radical generation containing azo dyes as pollutant. US assisted process showed improvement in oxidants removal. They also worked on the removal of textile dyes contaminants, such as congo red, erichrome black T and methyl red. At electrolysis time of 15, 25 and 35 min, 34 Khz US almost removed 100% textile dyes using EC of Al combined with US.

Hybrid or advanced EC processes are also effectively used for removing emerging and micro-pollutants from WW. US and EC continuous and simultaneous application, in short term batch mode, produces unwanted effects in the cell, and decreases EC performance. These side effects include the destruction of colloidal

OH^- *in situ* generation [4]. Table 2 shows a summary of EC technology for various industrial effluents, under optimized operating conditions.

Table 2: Treatment of various industries WW by EC.

Author	WW	Electrodes	Optimum conditions	RE
[19]	Tannery industry WW	Al	Uncontrolled pH: 3; controlled pH: 5; J: 1.2 mA/cm ² ; stirring speed: 150 RPM; TT: 60 min	COD: 83.5 %; total cost: 0.86 \$m ⁻³
[20]	Tannery industry WW	Al	Retention time: 60 min; J: 1.2 mA/cm ² ; initial pH: 3; stirring speed: 150 RPM	COD: 83.33; controlled pH: 5; total cost: 1.0899 \$/m ³ ; 97% oil-grease; 98% Cr; 68% total N; 100% PO ₄ ³⁻ ; 84% COD; 98% turbidity; 79% SO ₄ ²⁻
[23]	Swine WW	Fe anode	Reaction time: 35 min; electrode gap: 2 cm; J: 30 mA/cm ² ; no stirring; initial pH: 6.3	Suspended solids 98.5%; total P: 99.5%; COD: 50.6%; BOD: 10%
[25]	Industrial oil and soap WW	Al electrodes	Initial pH: 3; J: 73.33 A/m ² ; TT: 30 min; initial COD Ct: 19750 mg/L	COD: 94.93%; oil and grease: 90.20%; energy consumption: 1.68 KWh/Kg COD for Al
[26]	Synthetic WW	SS	Electrode gap: 1-2 cm; initial pH: 8.5; 30 V; TT: 20 min; total cost: 7.04 to 8.13 US\$/m ³	99% Cr
[31]	Battery industry WW	Al	pH: 9; TT: 60 min	99.8% Pb
[34]	Synthetic Cr polluted WW	MS	TT: 60 min; pH: 3.4; NaCl electrolyte Ct: 1 g/L; J: 32.78 mA/cm ²	99.9 % Cr
[55]	Synthetic WW	Al	J: 11.57 A/m ² ; electrode gap: 1 cm; pH: 3-6; electrical conductivity: 899-2300 $\mu\text{S}/\text{cm}$	Cu (II): 97.9%; Cr (VI): 96.4%; total Cr: 94.9%
[72]	Textile industry WW	Fe	I: 4A; TT: 5 min,	COD: 64%; color: 60%
[76]	Electroplating industry effluent	Fe	J: 73.5 A/m ² ; pH: 3.5; TT: 90 min	91.7% Cr i(VI); 91.3% Pb

EC energy requirement and cost analysis

The economic part of EC advanced oxidation process is one major concern, because higher pollutants RE requires greater costs. The total operation involves various costs shown by the following formula.

$$\text{Total cost of operation} = \text{electricity price} \times \text{energy consumption} + \text{anode price} \times \text{electrode consumption} + \text{sludge transportation and disposal cost} + \text{chemicals price} \times \text{chemical added} + \text{maintenance cost} + \text{depreciation} + \text{labour cost} + \text{amortization} \quad [30]$$

The total operation costs depend upon the market price of consumables, expenditures on sludge management and technology used for the treatment. Ec process parameters that influence major portion in costs are EEC and electrode consumption.

$$EEC = \frac{EIt_{EC}}{V_s} \quad (10)$$

where V is electric E , and t_{EC} is time of electrolysis. EEC unit is $W/h/m^3$ or $Kw/h/m^3$. It is broadly used for determining energy consumption by water treatment technologies. One other relevant term also shown in literature is SEEC.

$$SEEC = \frac{nFE}{3.6 \times 10^{-3} M_w \times \varphi} \quad (11)$$

where n is the number of electrons consumed in the anode sacrificial oxidation ($n = 2$ and 3 mol for Fe and Al), F is Faraday constant ($96,487 \text{ C/mol}^{-1}$), M_w is anodic material molecular weight (55.84 and 26.98 g/mol^{-1} for Fe and Al) and φ is efficiency of I , which can be determined from the following equation:

$$\varphi = \frac{\Delta m_{exp}}{\Delta m_{theo}} \quad (12)$$

where Δm_{exp} and Δm_{theo} are electrode mass loss during TT, and determined by Faraday law, respectively.

$$\Delta m_{theo} = \frac{M_w I t_{EC}}{nF} \quad (13)$$

There is also other relation used for determining specific energy consumption on the basis of removed pollutant mass.

$$SEEC = \frac{EIt_{EC}}{V_s \sum (Y M_{pol} \cdot [pollutant]_o)} \quad (14)$$

where Y is removed pollutant during EC process, M_{pol} is pollutant molecular weight in $g/mole^{-1}$ and $[pollutant]_o$ is initial pollutant Ct in mol/m^3 [89].

[94] worked on energy consumption requirements of three different Ec processes: EC, EO, EF and photo-EF process, for the treatment of leather tannery effluents. Fe electrodes were used as anode and cathode for EC experiment. BDD was used as anode and Fe as cathode for EO. Both BDD for EF and photoelectron-Fenton processes were operated at J of 65 and 111 mA/cm^2 . The reactor volume was 250 mL . It was revealed from results that energy consumption requirement for various Ec process followed the order: $EO < EC < EF < \text{Photo-EF process}$.

Conclusion and future perspectives

This critical review analyzed EC process, mechanism and application in treatment of various industrial WW for the removal of various conventional and emerging contaminants. Literature has determined that EC is effectively employed for various industrial WW treatment. This technology is more economical and environment friendly than conventional processes. It has several advantageous, such as versatility in pollutants treatment, easy operation and maintenance, less costs and environment friendliness. Researches indicated that various advanced technologies are obtained by coupling EC with different advanced oxidation techniques, such photo-Fenton, O_3 , EF, US, PC and EF processes. These technologies are more efficient than EC in

removing pollutants. If WW contains more toxic recalcitrant compounds, various advanced techniques in combination with EC processes are feasible to treat them, since they produce strong degrading agents, such as H_2O_2 $\text{OH}\cdot$ free radical and O_3 . Electrode passivation is one of the main disadvantages of EC and other Ec methods. However, it can be minimized by combining them with Fenton process, proper cleaning, AC use and higher Ct of NaCl, to improve indirect Ec oxidation. High chloride content in WW increases sacrificial metal ions during EC process. In some cases, EC is less effective than aerated EC. Auspicious results were obtained when EC was combined with advanced oxidation processes. These combined processes proved to be effective in eco-environmental and economic terms for pollutants removal.

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Authors' contributions

All authors contributed to the study procedures, manuscript writing and proofreading.

Conflict of interest

Authors declare that there was no conflict of interest.

Abbreviations

AC: alternating current
ACEC: air cathode EC process
 Al_2S_3 : aluminum sulphides
BDD: boron doped diamond electrode
BOD: biochemical oxygen demand
COD: chemical oxygen demand
Ct: concentration
E: potential
Ec: electrochemical
EC: electrocoagulation
EEC: electrical energy consumption
EF: electro-Fenton
EO: electro-oxidation
 H_2O_2 : hydrogen peroxide
 H_2SO_4 : sulfuric acid
HER: hydrogen evolution reaction
J: current density
MS: mild steel

OH•: hydroxyl
OH⁻: hydroxide
PC: peroxi-coagulation
PDS: Peroxy-disulfate
PMS: peroxy-monosulfate
PO₄³⁻: phosphate
POP: persistent organic pollutant
RE: removal efficiency
ROS: reactive oxygen species
SEEC: specific electrical energy consumption.
SO₄²⁻: sulfate
SS: stainless steel
TOC: total organic carbon
TT: treatment time
US: ultra-sonication
UV: ultraviolet
WW: wastewater

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