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Review Article

A Review of Hybrid Process Development Based on Electrochemical and Advanced Oxidation Processes for the Treatment of Industrial Wastewater

Perumal Asaithambi,[1] **Mamuye Busier Yesuf**,[1] **Rajendran Govindarajan**,[2] **N.M. Hariharan**,[3] **Perarasu Thangavelu**,[4] **and Esayas Alemayehu**[1,5]

[1]*Faculty of Civil and Environmental Engineering, Jimma Institute of Technology Jimma University, Jimma, Ethiopia*
[2]*Department of Chemical Engineering, Hindustan Institute of Technology and Science Rajiv Gandhi Salai Padur, Chennai 603103, Tamil Nadu, India*
[3]*Department of Biotechnology, Sree Sastha Institute of Engineering Chembarambakkam, Chennai 600123, Tamil Nadu, India*
[4]*Department of Chemical Engineering, AC Tech Campus Anna University, Chennai 600 025, Tamil Nadu, India*
[5]*Africa Center of Excellence for Water Management, Addis Ababa University, Addis Ababa, Ethiopia*

Correspondence should be addressed to Perumal Asaithambi; asaithambi.perumal@ju.edu.et

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Nowadays, increased human activity, industrialization, and urbanization result in the production of enormous quantities of wastewater. Generally, physicochemical and biological methods are employed to treat industrial effluent and wastewater and have demonstrated high efficacy in removing pollutants. However, some industrial effluent and wastewater contain contaminants that are extremely difficult to remove using standard physicochemical and biological processes. Previously, electrochemical and hybrid advanced oxidation processes (AOP) were considered a viable and promising alternative for achieving an adequate effluent treatment strategy in such instances. These processes rely on the production of hydroxyl radicals, which are highly reactive oxidants that efficiently break down contaminants found in wastewater and industrial effluent. This review focuses on the removal of contaminants from industrial effluents and wastewater through the integration of electrochemical and advanced oxidation techniques. These processes include electrooxidation, electrocoagulation/electroflocculation, electroflotation, photo-Fenton, ozone-photo-Fenton, sono-photo-Fenton, photo-electro-Fenton, ozone/electrocoagulation, sono-electrocoagulation, and peroxi/photo/electrocoagulation. The data acquired from over 150 published articles, most of which were laboratory experiments, demonstrated that the hybrid process is more effective in removing contaminants from industrial effluent and wastewater than standalone processes.

1. Introduction

Recent years have seen a progressive decline in rainfall rates due to the global problem of environmental contamination. Globally, scarcity of fresh water is increasing due to a plethora of factors such as population growth, rapid industrialization, urbanization, and climate change. Water scarcity and environmental degradation have a direct effect on civilization, economic growth, and human life around the world [1–3]. The demand for clean water has increased as a result of daily human activities and industrialization, necessitating the efficient use of water resources such as rainwater, stormwater, treated wastewater, and industrial effluents.

Due to rapid industrialization, enormous amounts of polluted water are generated each year by various industries such as petroleum and oil [4–6], textile [7–9], pulp and paper [10, 11], leather [12], pharmaceutical [13], paint, distillery [14, 15], tannery [16, 17], fertilizer [18, 19], sugar [20], landfill leachate [21, 22], hospital [22, 23], municipal [22] and domestic [24, 25], and agricultural wastewater [26]. The

aforementioned industrial pollutants contain a variety of complex pollutants like nitrogen, phosphorous, sulphur, and food scraps, including refractory or hazardous organic molecules like fulvic acid, humic acids, phenolic compounds, soap, oil, pathogens, and other harmful substances [27, 28].

Polluted water from industry causes a variety of different types of water pollution, which are hazardous to human health and aquatic life in the environment. For instance, when heavy metal industries dump wastewater into adjacent lakes and rivers, it can aggregate and cause birth defects, as well as cancer, immunological suppression, and acute poisoning. The utilization of microorganisms in industry leads to contamination, which contributes to the infant mortality rate via cholera, typhoid, and so on. Numerous industries, such as distillery, paint, and pharmaceutical, contribute to the depletion of oxygen in bodies of water, posing problems for aquatic organisms and human health. Additionally, they reduce the amount of sunlight reaching the water, impairing the growth of photosynthetic plants and microorganisms.

In the coming decades, the central concern for environmental research and development will be to create new sources of water, particularly in nations with limited water resources.

1.1. Industrial Effluents. Wastewater management is one of the most significant environmental issues confronting industries. Environmentalists face a significant difficulty in treating wastewater created by industries. The liquid is discharged as effluent from industries and contains a significant proportion of organic matter, both suspended and dissolved materials [29]. If this effluent is not properly treated, it can put significant stress on the water body, resulting in extensive damage to aquatic life. It also depletes soil alkalinity, affecting the quality of groundwater [30]. The ever-increasing output of wastewater by industries and the rigorous regulatory regulations governing its disposal have accelerated the need for the development of innovative technologies for efficiently and economically processing effluent.

Physical, chemical, and biological approaches are used to remove pollutants such as heavy metals, colour, COD, TOC, BOD, and emerging contaminants from wastewater and industrial effluent [31, 32]. However, these approaches may eventually generate a substantial amount of sludge, resulting in secondary contamination.

2. Wastewater Treatment Techniques

Industrial effluents and wastewater have been treated using a variety of physical, chemical, and biological techniques [32]. The efficiency of the treatment process can be increased by combining processes or by applying them independently. The system must be designed with both technical (treatment efficiency, plant simplicity, etc.) and economic considerations in mind (investment and operating cost). The disadvantages of conventional treatment systems are (a) high running costs and (b) pollutant transfer from one phase to another. When biooxidation is used, the organic compound should be biodegradable and have low toxicity.

To address the limitations of traditional treatment, numerous developing technologies such as electrooxidation [15], advanced oxidation [33], and membrane processes [34] have been applied to wastewater treatment. Industrial wastewater was treated using a variety of approaches, including biooxidation, electrochemical oxidation, AOPs, and physicochemical processes.

Each of the existing techniques has significant limits in terms of their own benefits and demerits for the treatment of industrial wastewater. The key problems for environmental researchers in this situation are to design and create a novel procedure that overcomes restrictions such as operational costs, treatment efficiency, and secondary pollutant generation.

2.1. Electrochemical Processes. Electrochemical technology is a viable and attractive means of treating industrial wastewater [35, 36]. The principal advantages of this technology over other conventional processes include cost-effectiveness, ease of automation, adaptability, environmental compatibility, energy efficiency, selectivity, and minimal maintenance costs [37, 38]. Electrochemical approaches for industrial effluent and wastewater treatment are cost-effective and environmentally benign [38].

The application of electrochemistry in the treatment of environmental pollution has been examined in recent years, with numerous researchers investigating the potential of electrochemical systems in the destruction of organic materials in effluent. Different electrochemical techniques (Figure 1) are considered for the treatment of industrial effluent such as electrocoagulation [39, 40], electrooxidation [41, 42], electrodeposition [43], electroflotation [44], electrodialysis [43, 45], electroflocculation [46], electrophoresis [47], and electroreduction.

The development of electrochemical technology and its use in industrial effluent have been addressed through several approaches such as electrooxidation, electrocoagulation, electroflotation, and electroflocculation.

2.1.1. Electrooxidation. Electrochemical oxidation for wastewater treatment began in the early 19th century [48], with the investigation of the electrochemical degradation of cyanide [49]. An extensive study on this technique began in the 1970s [50] when it was realized that phenolic compounds could be anodized. Papouchado et al. (1975) investigated the electrochemical oxidation of phenol compounds in aqueous solutions.

Koile and Johnson (1979) investigated electrochemical purification of phenol-containing wastes in a pilot plant [51] and described the anodic oxidation pathways of phenolic chemicals. The electrochemical elimination of phenolic coatings from platinum anodes was investigated by [52]. Comninellis et al. [53] achieved significant results in electrooxidation of phenol during wastewater treatment.

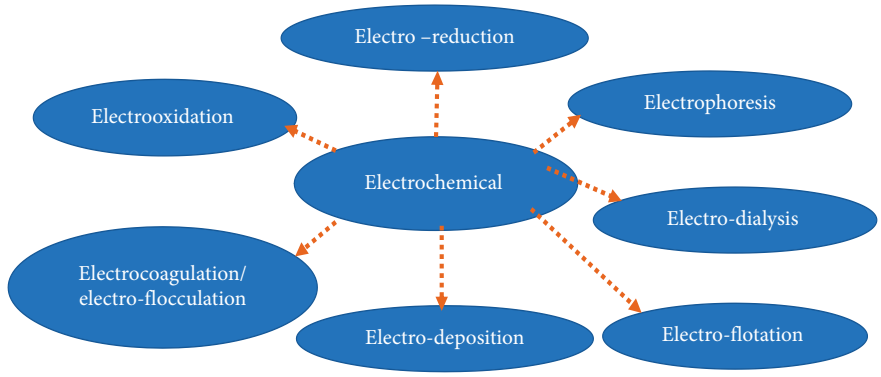

Figure 1: Types of electrochemical processes.

Electrochemical oxidation of industrial effluent can be accomplished in two ways, directly or indirectly, employing a suitable anodically generated oxidant. The pollutant can be oxidized directly at anodes via physically adsorbed or chemisorbed active oxygen. This is commonly referred to as "anodic oxidation" or "direct oxidation."

The contaminants are oxidized electrochemically using powerful oxidizing agents created at the anode surface, such as Fenton reagent, H_2O_2, O_3, and chlorine. The oxidant reacts with the organic substance, which is required to convert CO_2, water, and other inorganic compounds completely [54]. In the electrochemical cell, indirect oxidation process utilizes strong oxidizing agents such as chlorine/hypochlorite generated in situ during electrolysis of wastewater.

The following reactions involved are

$$At \cdot \text{anode} \cdot 2Cl^- \longrightarrow Cl_2 + 2e^-, \quad (1)$$

$$At \cdot \text{cathode} \cdot 2H_2O + 2e^- \longrightarrow H_2 + 2OH^-, \quad (2)$$

$$Bulk \cdot \text{solution} \cdot Cl_2 + H_2O \longrightarrow HOCl + H^+ + Cl^-, \quad (3)$$

$$HOCl \longrightarrow H^+ + OCl^-, \quad (4)$$

$$Organic\ matter + OCl^- \longrightarrow CO_2 + H_2O + Cl^- + Product. \quad (5)$$

Several industry effluents can be treated with an electrooxidation process, including olive mills [55], distillery [56, 57], petroleum exploration of the Petrobras [58], removal of Rhodamine 6G, azo dye, acid black 210 dye, reactive red 120 [59], and pharmaceutical residue [60].

Manisankar et al. [56] reported maximum colour removal using distillery effluent through electrochemical oxidation using two types of anodes such as PbO_2/Ti and RuO_2/Ti electrodes. They discovered that the maximum removal of COD, BOD, and colour was 92%, 98.10%, and 99.50%, respectively, in the case of RuO_2/Ti electrode. Chen et al. [61] examined the electrochemical oxidation of ammonia in wastewater using RuO_2–IrO_2–TiO_2/Ti electrode. They observed that the direct anodic oxidation efficiency of ammonia and the current efficiency were, respectively, less than 5% and 10%. Radha et al. [62] determined the percentage removal of COD, TS, TDS, and TOC from textile effluents using an electrochemical oxidation process to be 68%, 49.2%, 50.7%, and 96.8%, respectively.

Panakoulias et al. [59] demonstrated electrochemical oxidation of reactive red 120 using DSA type (Ti/IrO_2–RuO_2) and BDD anodes. The BDD was shown to be capable of completely mineralizing the organic pollutant to CO_2 and also of completely removing the colour. Rocha et al. [58] proved that the anodic oxidation method can be used to remove COD from real petroleum exploration at the Petrobras plant in Brazil, utilizing Ti/Pt and BDD anodes, and that this technology can also be employed as a pretreatment procedure to minimize treatment costs and time.

Numerous researchers have examined electrochemical oxidation of various types of wastewaters with the purpose of removing pollutants from industrial effluent. Apart from colour and COD removal efficiency, one must consider power consumption and current efficiency as crucial metrics when comparing electrochemical technology to conventional treatment techniques.

2.1.2. Electrocoagulation and Electroflocculation. Electrocoagulation is used for treatment of industrial effluent in its simple form of electrochemical cell, when DC voltage is applied between the aluminium or iron electrodes, which generates a coagulant by the dissolution of metal from the anode with simultaneous formation of hydroxyl ions and hydrogen gas at the cathode [63, 64]. Additionally, these methods can create aluminium or iron hydroxide and/or polyhydroxide [65]. Moreover, the production of H_2 gas aids in the flocculation of particles near the water's surface [65].

Before the turn of the twentieth century, electrocoagulation was proposed. In 1889, a sewage treatment plant in London was developed to treat sewage by mixing it with seawater and electrolyzing it [66]. Dietrich developed electrocoagulation in 1906 for the treatment of ship bilge water. Harries received a patent in the United States in 1909 for wastewater treatment using electrolysis using sacrificial

aluminium and iron anodes. In the 1990s, Matteson et al. [67] described an "electronic coagulator" that electrochemically dissolved aluminium (from the anode) into the solution, where it reacted with the hydroxyl ion (from the cathode) to generate aluminium hydroxide. The hydroxide precipitates and coagulates suspended materials, effectively purifying the water. The electrochemical reaction involved in the reactor for anode and cathode when Al was used [68].

$$\text{At} \cdot \text{Anode} : \cdot Al \longrightarrow Al^{3+} + 3e^{-}, \quad (6)$$

$$\text{At} \cdot \text{Cathode} : \cdot 3H_2O + 3e^{-} \longrightarrow 3/2H_2 + 3OH^{-}, \quad (7)$$

$$\text{Overall} : \cdot Al^{3+} + 3OH^{-} \longrightarrow Al(OH)_3. \quad (8)$$

Electrocoagulation has been successfully used to remove pollutants from a variety of industrial wastewaters, including metal plating [69], livestock [70], pulp and paper [71], laundry wastewater [72], greywater [73], nanoparticles [74], and industrial wastewater [75]. This procedure is also utilized to remove soluble ionic species from heavy metals, defluoridation of septentrional Sahara water [76], and removal of arsenic, nitrate [77, 78], urea [79], sulfide, sulphate, and sulfite [80], boron [81], phosphate, chromate, and so on. This approach has also been used successfully to remove other mineral cations and anions.

Vasudevan et al. [82] conducted studies to remove iron from drinking water using the electrocoagulation technique and observed a 98.4% iron removal efficiency. Saravanan et al. [83] investigated electrocoagulation techniques for the treatment of Acid Blue 113 with an iron anode and observed that 91% of COD was removed.

However, the high-energy consumption and anode passivation phenomenon have hampered the electrochemical process's further applicability. Fortunately, the introduction of alternating current electrocoagulation had a tremendous impact on electrochemical processes; it not only saved energy but also demonstrated increased activity and efficiency throughout the process [84, 85]. Thus, in recent years, the development of the alternating current electrocoagulation process has been investigated [84]. Eyvaz et al. [86] investigated the dye and TOC removal effects of direct and alternating pulse current electrocoagulation. They concluded that alternating pulse current electrocoagulation performed better than direct current electrocoagulation. Vasudevan et al. [84] examined the effect of alternating and direct current electrocoagulation on cadmium removal and found that the removal efficiency was 97.80% and 96.90%, respectively, with energy consumption of 0.665 and 1.236 kWh/m^3. Keshmirizadeh et al. [85] used aluminium/iron electrodes to perform an electrocoagulation procedure for the removal of chromium. Ren et al. [87] evaluated the pulse electrocoagulation method for the treatment of refractory berberine hydrochloride wastewater, estimating that energy costs might be reduced by up to 90% with this technique.

2.1.3. Electroflotation. Electroflotation was proposed for the first time in 1904 to separate precious minerals from ores (Elmore et al. (1905)). Electroflotation is a promising technique for removing pollutants from industrial wastewater. The performance of an electroflotation system is determined by the efficiency with which pollutants are removed, which is dependent on the size of the generated bubble, and the power consumption, which in turn is dependent on the cell design and operating circumstances and electrode materials.

This technology has been used to remove heavy metals [88], ammonia and phosphate [89], decontamination of groundwater, metal finishing effluent, algae removal [90], and defluoridation of drinking water [91].

According to the literature review, a single method was ineffective for total pollutant removal, but when combined with other treatment processes, it resulted in a high pollutant removal rate. Thus, the establishment and construction of a new model is a crucial issue concerning pollution removal, energy consumption, and operational costs. To maximize pollutant removal, reduce operation time and expense, and mitigate the high-energy need for electrochemical treatments, hybrid treatment procedures such as combining electrochemical treatments with advanced oxidation processes may be used.

2.2. Hybrid Advanced Oxidation Processes. Advanced oxidation processes (AOPs) have emerged as an extremely promising method for treating materials with high chemical stability and/or low biodegradability [92]. The treatment of industrial effluents containing chemicals with AOPs can result in the production of CO_2, water, and inorganic compounds [93]. Additionally, partial degradation of a biodegradable organic contaminant might result in the formation of biodegradable intermediates.

The AOPs occur in two distinct ways: by oxygen oxidation and through the utilization of high-energy oxidants such as O_3 and H_2O_2, which contribute to the generation of •OH radicals. The free •OH is a potent nonselective chemical oxidant that interacts with organic molecules in solution. AOP is typically operated at temperatures and pressures close to ambient. The AOPs using different reagents systems include photochemical degradation processes (UV/O_3; UV/H_2O_2) [94, 95], photocatalysis (TiO_2/UV; photo-Fenton reactions) [96], and chemical oxidation processes (O_3, O_3/H_2O_2, and H_2O_2/Fe^{2+}) [97]. Mechanisms of AOPs-based on the UV and O_3 are given in Figures 2(a) and 2(b).

$$\% \; AOPs \longrightarrow HO^{\bullet} \overset{Pollu\tan t}{\longrightarrow} CO_2 + H_2O + inorganicions. \quad (9)$$

Additionally, AOPs can be classed as heterogeneous or homogenous (Figure 3). Homogenous was further classified into those who used energy and those who did not. A homogenous procedure typically uses UV light that is utilized to degrade organic substances with a correspondingly broad spectrum and also for the generation of hydroxyl radicals at elevated temperatures and pressures. In the homogenous process, electrical energy is employed to break

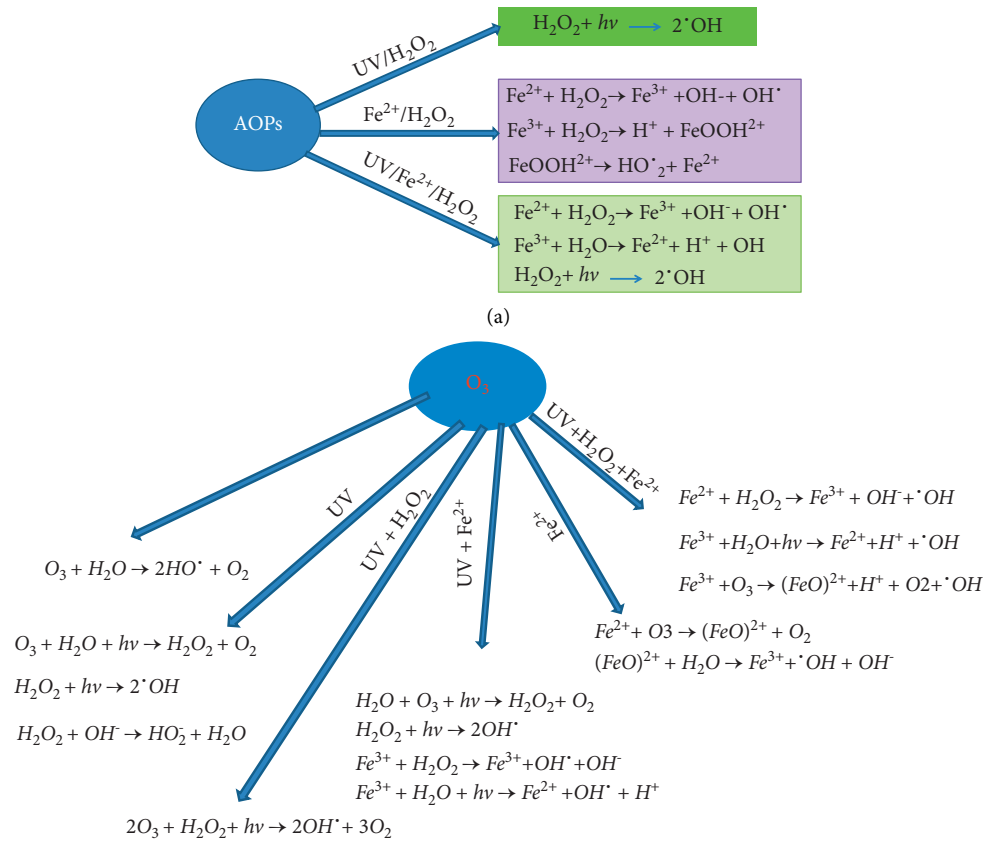

Figure 2: Mechanisms of AOPs.

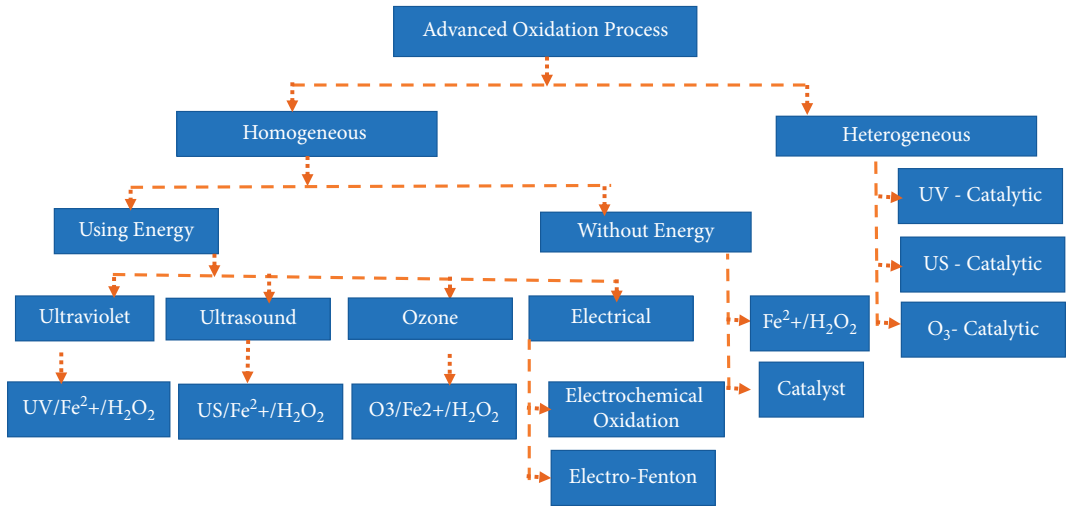

Figure 3: Types of advanced oxidation processes.

up molecules and substances in preparation for degradation. Electrons are transferred as a result of the participation of hydroxyl radicals. These approaches have a number of advantages, including increased process effectiveness and reduced reliance on additional reagents. Likewise, the energy cost and the lifetime of the electrodes must also be considered.

A heterogeneous advanced oxidation process that is frequently utilized with a catalyst in the decomposition of organic molecules. The process can be carried out using a catalyst such as a metal catalyst, a metal oxide catalyst, or an organometallic catalyst. Catalytic ozonation, photocatalytic ozonation, and photocatalysts are all heterogeneous processes that can be used to remediate industrial wastewater.

However, this technique has limitations in terms of energy transmission and scarcity of photocatalysts.

Advanced oxidation techniques have demonstrated potential as a treatment and purification technology for industrial wastewater, including the elimination of naturally occurring toxins, chemicals of emerging concern, pesticides, and other deleterious substances. Glaze et al. [98] made one of the earliest references to AOPs as processes involving the creation of hydroxyl radicals in sufficient quantity to effect water purification. Since the 1990s, the definition and development of AOPs have grown to encompass a number of ways for producing hydroxyl radicals and other reactive oxygen species, such as superoxide anion radicals, H_2O_2, and single oxygen. The AOPs include UV/H_2O_2 [99], H_2O_2/Fe^{2+}, $UV/H_2O_2/Fe^{2+}$ [100–102], O_3 [103], O_3/Fe^{2+} [104], O_3/UV, O_3/US [104], $O_3/UV/H_2O_2$, $O_3/UV/Fe^{2+}$ [105, 106], nonthermal plasmas [107], sonolysis [103, 108–110], H_2O_2/UV/US [110], photocatalysis [108], radiolysis [111], and supercritical water oxidation processes [112, 113].

Kusic et al. [114] compared the efficiency of numerous ozone- and/or UV-based techniques for phenol mineralization. The $UV/H_2O_2/O_3$ process procedure resulted in total mineralization. Catalkaya et al. [115] treated pulp mill wastewater using a variety of AOPs. They recognized that TiO_2-assisted photocatalysis removes more TOC and toxicity than the other AOPs. Wu et al. [116] investigated the breakdown of isopropyl alcohol and its major degradation intermediate, acetone formation. On the basis of TOC removal efficiency, the performance of various O_3, O_3/UV, H_2O_2/UV, H_2O_2/O_3, and H_2O_2/O_3/UV systems under various physicochemical circumstances was examined. Ozonation was significantly accelerated in the presence of UV light, H_2O_2, or both oxidants. The results indicated that when compared to the other AOPs, the $UV/H_2O_2/O_3$ procedure was the most successful and efficient. The decolorization of C.I. Reactive Red 2 was explored by Wu et al. [117] using ozone and photo-based AOPs. They observed that the $UV/O_3/H_2O_2/Fe^{3+}$ system was the best suitable way to remove more pollutants while consuming the least amount of energy. Lucas et al. [106] investigated the use of Fenton's reagent (H_2O_2/Fe^{2+}) in a batch reactor to remove COD from olive mill wastewater. Chandrasekara Pillai et al. [118] described a procedure for treating terephthalic acid wastewater using ozonation catalyzed process with Fe^{2+}, H_2O_2, and UV irradiation. When compared to other AOPs, the combined $O_3/H_2O_2/Fe^{2+}$/UV process removes pollutants at a rate of around 90% after 240 minutes. Hadavifar et al. [119] proved the efficacy of Fenton and photo-Fenton procedures in the alcohol distillery industry. They concluded that a photo-Fenton (18 to 97%) technique achieved a better removal efficiency than Fenton (5–47%) alone. Numerous research studies have been conducted to determine the efficacy of ozonation in decolorizing and degrading textile industrial effluents such as anthraquinone dye, bezafibrate, and others. The results indicated that ozonation was an extremely efficient method of decolorization and degradation [120, 121].

According to the results of the previous literature review, the $O_3/UV/H_2O_2/Fe^{2+}$ system consumes less energy while removing more pollutants than other photo- and ozone-based AOPs.

Asaithambi et al. [122] compared the removal of% colour and COD with minimal electrical energy per order from landfill leachate wastewater using various AOPs such as O_3, US, O_3/H_2O_2, US/H_2O_2, $O_3/Fe^{2+}/H_2O_2$, $US/Fe^{2+}/H_2O_2$, and $O_3/US/Fe^{2+}/H_2O_2$. They found that the $O_3/US/Fe^{2+}/H_2O_2$ process was more effective compared to the other processes. Asaithambi et al. [123] evaluated the colour removal and COD removal effectiveness of UV, Fe^{2+}/H_2O_2, $UV/Fe^{2+}/H_2O_2$, and $O_3/UV/Fe^{2+}/H_2O_2$ processes, as well as the associated electrical energy per order, from distillery industrial effluent. They noticed that, in comparison to all other combinations of the AOPs, the $O_3/UV/Fe^{2+}/H_2O_2$ procedure removed 100% colour and 95.50% COD with an electrical energy consumption of 0.015 kWh/m^3. Asaithambi et al. [124] examined the effectiveness of the US, UV, UV/H_2O_2, US/H_2O_2, Fe^{2+}/H_2O_2, $UV/Fe^{2+}/H_2O_2$, $US/Fe^{2+}/H_2O_2$, and $UV/US/Fe^{2+}/H_2O_2$ process in removing% colour and COD from landfill leachate using minimal electrical energy per order. They found that, as compared to the separate processes, the hybrid $UV/US/Fe^{2+}/H_2O_2$ method greatly boosted colour reduction (100%) and COD reduction (96%).

2.3. Electrochemical Advanced Oxidation Processes. The AOPs such as chemical, photocatalytic, photochemical, and electrochemical approaches, among others, are potential environmentally acceptable solutions for treating industrial wastewater containing hazardous and biorefractory contaminants. Electrochemical advanced oxidation processes are one type of AOP system used to treat industrial wastewater. This procedure is inexpensive and highly effective and does not require the addition of toxic chemicals or the generation of hazardous waste [125].

Anodic oxidation is a process used in EAOPs that is carried out in an electrolytic cell under the action of hydroxyl intermediates formed during water oxidation [126].

$$M + H_2O \longrightarrow M(^{\bullet}OH) + H^+ + e^-, \quad (10)$$

where M($^{\bullet}$OH) denotes the hydroxyl radical adsorbed on the anode M or remaining near its surface.

Recently, EAOPs have been extensively explored and employed for the treatment of industrial effluent and wastewater due to their high oxidation capacity, rapid reaction rate, complete elimination of pollutant, low overall cost, and ease of handling. Simple AOPs and/or electrochemical processes are incapable of mineralizing the organics and inorganics found in industrial effluent to a significant extent. Combining AOPs with other treatment technologies, like electrochemical technologies, would be an efficient and cost-effective way to obtain high organic and inorganic removal efficiencies while consuming less energy.

Ruiz et al. [127] investigated the electro-Fenton and solar photoelectron-Fenton-Fenton (SPEF) degradation of Acid Yellow 36 azo dyes utilizing a recirculation flow plant with undivided cells containing boron-doped diamond. They reported that entire mineralization is nearly obtained with SPEF, but the electro-Fenton procedure results in poor TOC

removal. The SPEF approach maximizes current efficiency and reduces energy costs. Módenes et al. [128] combined the widely utilized photo-Fenton and electrocoagulation techniques to treat tannery industrial wastewater. The results indicated that an integrated photo-Fenton and electrocoagulation method might be used to remove organic and inorganic contaminants from tannery industrial effluent with less environmental impact and at a lower cost than the existing technique. Babuponnusami et al. [129] compared the degradation of phenol using Fenton, electro-Fenton, sono-electro-Fenton, and photo-electro-Fenton treatment techniques. They discovered that simultaneous UV irradiation and electrolysis with Fenton's reagent resulted in improved performance. The degrading efficiency of photo-electro-Fenton was observed to be greater than that of sono-electro-Fenton, electro-Fenton, and Fenton.

Vahid et al. [130] designed the photoassisted electrochemical system for the treatment of C.I. Acid Blue 92 in a recirculation mode and with UV irradiation (AB92). They found that the degradation efficiency of AB92 was 27.89, 37.65, and 95.86% after 45 minutes, respectively, using photolysis, electrochemical oxidation, and photo-assisted electrochemical methods. Ding et al. [131] demonstrated that combining anodic photo-electrochemical/electro-Fenton (PEC) and cathodic electro-Fenton (EF) cells results in a dual-cell wastewater treatment system (D-PEC-EF). Not only did the results demonstrate that they provide a very successful and energy-efficient advanced oxidation technology for wastewater treatment, but also they contributed to a better understanding of the synergic effect in dual-cell electrochemical processes. Asaithambi et al. [132] compared the removal of % colour and COD from landfill leachate wastewater using photo (UV), electro-Fenton, and photo-electro-Fenton processes. Their findings indicated that the hybrid photo-electro-Fenton process achieved greater colour removal efficiencies of 100% and COD removal efficiencies of 97% while consuming less energy (3.10 $kWhr/m^3$) than the photo- and electro-Fenton processes alone.

3. Hybrid Process Based on the Electrochemical and AOPs

Electrocoagulation systems are used in tandem with UV, O_3, US, and H_2O_2 [133, 134] to remove a variety of organic and inorganic pollutants and to guarantee that the effluent is of high quality prior to discharge into the aquatic environment.

By combining electrochemical and advanced oxidation technologies, a hybrid system can be built (Figure 4). Hybrid systems such as ozone-assisted electrocoagulation [135–138], peroxi-electrocoagulation [139, 140], photo-electrocoagulation [128, 141], peroxi-photo-electrocoagulation [133], sono-electrocoagulation [140, 142, 143], peroxi-photo-ozone-electrocoagulation [133], electro-oxidation and Oxone [144], electro-peroxone-ultrasound irradiation [145], coagulation-electro-activated H_SO_5 [146], and ozone-photo-Fenton [133] were applied to improve the quality of wastewater.

The advantages of hybrid systems include the following: (a) high oxidation capacity, (b) rapid reaction rate, (c) total pollutant removal in organic and inorganic pollutants while minimizing the total cost, and (d) coupling electrochemical processes with AOPs which is conceptually advantageous because it results in increased overall efficiencies when compared to the efficiency of a single process. There is also active research underway to develop hybrid [147].

The selection of the optimal system combination should take several factors into account, both technical (e.g., plant simplicity, treatment efficiency, flexibility, and operating parameters) and economic (e.g., capital and operating costs, including reagent and energy consumption, sludge and gas disposal, and maintenance) considerations [133].

Numerous researchers have attempted to build a hybrid method that combines AOP with an electrochemical process to remove contaminants from diverse industries' wastewater.

3.1. Ozone-Electrocoagulation. Song et al. [148] investigated the removal of azo dye and C.I. Reactive Blue 19 using a combination of ozonation and electrocoagulation. This procedure demonstrates an increase in the percentage of colour and COD removed. Hernández-Ortega et al. [149] treated real industrial wastewater using electrocoagulation and ozonation pretreatments, enhancing the effectiveness of the existing biological treatment. Although biological treatment alone was ineffective in treating these industries' raw effluents, when combined with the electrocoagulation-ozonation process, it resulted in a better treated effluent.

Bernal-Martínez et al. [150] demonstrated the efficacy of electrochemical, ozone, and combined electrochemical-ozone procedures for industrial wastewater treatment. The results indicated that when electrochemical-ozone processes were combined, the elimination of COD, BOD_5, colour, turbidity, and total coliforms increased dramatically to 99%, 84%, 79%, 95%, 96%, and 99%, respectively. Bernal-Martínez et al. [151] investigated the performance of electrochemical, ozone, and combined electrochemical-ozone methods in the treatment of industrial wastewater. Due to the quantity of sludge and electrode passivation, the integrated electrochemical-ozone process with energy pulses was a suitable process, as the amount of sludge and electrode passivation was less than that with the electrochemical approach alone.

Behin et al. [136] used a rectangular internal-loop airlift reactor to perform ozone-assisted electrocoagulation for the decolorization of synthetic wastewater containing acid brown 214. They discovered that when electrocoagulation and ozonation were combined, the combined method was more successful. Asaithambi et al. [125] examined the efficacy of ozonation, electrocoagulation, and ozone-assisted electrocoagulation in treating real distillery industrial effluent. The results indicated that the hybrid electrocoagulation technique removed 100% of the colour and 95% of the COD. Bilińska et al. [152] investigated the combination of EC and ozonation as a single-step ($EC + O_3$) and two-step ($EC \longrightarrow O_3$) treatment method for the removal of colour from RB5 aqueous solution containing dye effluent

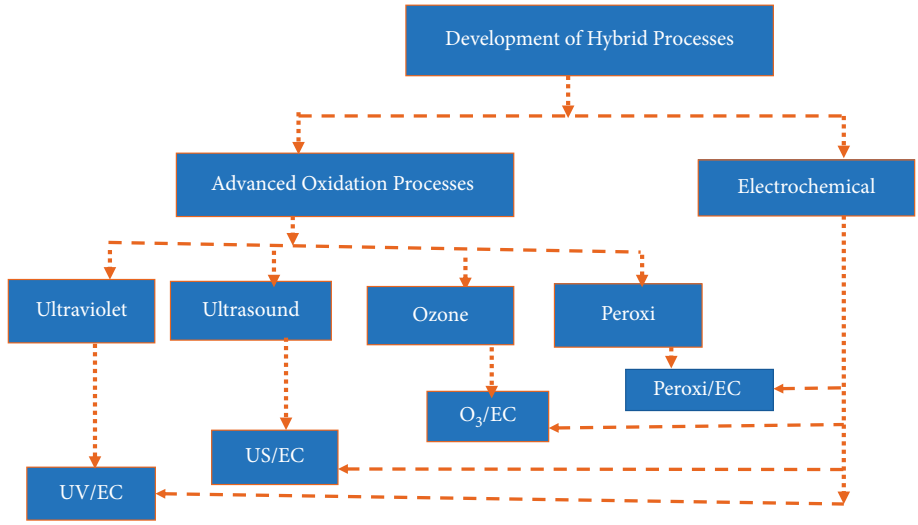

Figure 4: Development of hybrid processes.

and reported that the system achieved excellent results. Additionally, even after a prolonged treatment time, the application of EC as a single process did not result in sufficient colour removal. Ozonation performed in a single operation necessitated a lengthy treatment period in order to obtain a high level of colour removal.

Das et al. [153] explored the removal of cyanide, chemical oxygen demand (COD), biological oxygen demand (BOD), and chloride from the steel industry's biological oxidation treatment (BOT) effluent using an integrated ozonation-assisted electrocoagulation process. Their findings indicated that when electrocoagulation and ozonation were used independently, the contaminants were removed inefficiently. However, when ozonation and electrocoagulation were combined, a relatively good outcome was obtained. Mehralian et al. [154] examined the removal of COD from real cardboard effluent using the EC/O_3 process with the Box-Behnken Design (BBD). Their findings indicated that the EC/O_3 procedure might be used to rapidly and effectively remove colour and COD from wastewater.

This could be attributed to the enhanced EC process due to O_3 and the formation of more active species, such as $^{\bullet}OH$ radicals. The mechanism involves hydroxide radicals ($^{\bullet}OH$) production by ozone, and Fe^{2+} catalyzes ozone decomposition to generate hydroxyl radicals, as shown in the following equations:

$$O_3 + Fe^{2+} \longrightarrow FeO^{2+} + O_2, \tag{11}$$

$$FeO^{2+} + H_2O \longrightarrow Fe^{3+} + HO^{\bullet} + OH^{-}, \tag{12}$$

$$FeO^{2+} + Fe^{2+} + 2H^{+} \longrightarrow 2Fe^{3+} + H_2O. \tag{13}$$

3.2. Peroxi- and Photoelectrocoagulation. In an electrocoagulation system, H_2O_2 is added to the electrocoagulation system, and a sacrificial Fe anode is used as the Fe^{2+} source.

$$Fe^{2+} + H_2O_2 \longrightarrow Fe^{3+} + {}^{\bullet}OH + OH^{-}. \tag{14}$$

This reaction is propagated from ferrous ion regeneration mainly by reduction of the produced ferric species with hydrogen peroxide.

$$Fe^{3+} + H_2O_2 \longrightarrow Fe^{2+} + HO_2^{\bullet} + H^{+}. \tag{15}$$

Ferrous ions are consumed more rapidly than they are produced. In addition, ferrous ions can be rapidly destroyed by hydroxyl radicals with a rate constant in the range of 3.2–4.3 X $10^8 M^{-1}\ s^{-1}$.

$$Fe^{2+} + {}^{\bullet}OH \longrightarrow Fe^{3+} + OH^{-}. \tag{16}$$

Therefore, a more ferrous ion dosage is needed to maintain hydroxyl radical production in a moderate amount. This results in a large amount of ferric hydroxide sludge during the neutralization stage of the Fenton process, which requires an additional separation process and disposal.

The performance of the electrocoagulation and peroxi-electrocoagulation process can be improved by using UV radiation, which is known as the photo-electrocoagulation and peroxi-photo-electrocoagulation process. This process is improved due to the higher production rate of •OH from the photoreduction of $Fe(OH)^{2+}$ and photodecomposition of complexes from Fe^{3+} reactions, as shown in the following equation:

$$Fe(OH)^{2+} + h\nu \longrightarrow Fe^{2+} + {}^{\bullet}OH, \tag{17}$$

$$\begin{aligned} R(CO_2) - Fe^{3+} + h\nu &\longrightarrow R({}^{\bullet}CO_2) \\ +Fe^{2+} &\longrightarrow {}^{\bullet}R + CO_2. \end{aligned} \tag{18}$$

The peroxi-electrocoagulation procedure was developed by Yüksel et al. [155] for the removal of sodium dodecyl sulphate (SDS) surfactant. The research findings indicated that the overall sodium dodecyl sulphate removal efficiency was 81.60% with a 1.63 kWh $(kgSDS)^{-1}$ energy

consumption. Farhadi et al. [13] compared the removal of COD from pharmaceutical wastewater using electrocoagulation, photo-electrocoagulation, peroxi-electrocoagulation, and peroxi-photo-electrocoagulation. They observed the COD removal efficiency for the treatment of pharmaceutical wastewater as follows: peroxi-electrocoagulation > peroxi-photo-electrocoagulation > photo-electrocoagulation > electrocoagulation process.

Barışçı et al. [139] indicated that by comparing three distinct procedures for the cholesterol-lowering medication atorvastatin, including electrically synthesized ferrate (VI), electrocoagulation, and peroxi-electrocoagulation. They determined that the particular drug removal efficiencies for the Fe(VI)-77.6%, EC-77.1%, and p-EC-82%. According to the results, the p-EC procedure had the highest removal efficiency and was also shown to be the most economical. Asaithambi et al. [156] studied the performance of electrocoagulation, peroxi-electrocoagulation, photo-electrocoagulation, and peroxi-photo-electrocoagulation procedures for the removal of colour and COD, as well as the energy consumption of the distillery industry effluent. They reported that the peroxi-electrocoagulation process was the most efficient and effective in removing colour and COD when compared to other electrochemical advanced oxidation procedures. The % colour and COD removal indicated that the peroxi-electrocoagulation process achieved 100 and 86% removal, respectively, with a minimum electrical energy usage of 1.2 kWh/m^3.

3.3. Sono-Electrocoagulation. He et al. [142] investigated the EC and sono-EC processes for the treatment of synthesized wastewater containing RB19, utilizing aluminium electrodes. They discovered that sono-EC has a greater removal efficiency than EC alone. Additionally, ultrasound has been shown to be beneficial in reducing the thickness of the electrode passive film and the size of the particles in the solution. The continuous ultrasonic method is more effective in dissolving the passive film than the intermittent ultrasonic process. Thus, the intermittent ultrasonic technique was found to be compatible with sono-EC. Asaithambi et al. [157] concluded that sonication, electrocoagulation, and sono-assisted electrocoagulation methods were successfully used to treat pulp and paper mill effluent and that the results were compared in terms of colour removal, COD removal, and energy consumption. Their findings indicated that electrocoagulation assisted by ultrasound was more successful than electrocoagulation and sonication performed independently.

According to Dizge et al. [158], combining treatment techniques is a popular issue in environmental engineering, particularly for severely polluted wastewater. They evaluated the treatment performance of brewery wastewater using electrocoagulation (EC), ultrasonication (US), and sono-electrocoagulation (SEC) procedures and found that the suggested integrated SEC process removed more pollutants than the EC and US processes used alone. Prajapati et al. [159] conducted the research to compare the performance and cost of sonication, electrocoagulation, and sono-electrocoagulation processes for the removal of colour and COD from biodigester effluent (BDE). The results indicate that the sono-electrocoagulation process provides higher colour and COD removal efficiencies than sonication and electrocoagulation alone.

Based on a survey of the literature, the new model was created and developed with the goal of removing pollutants while consuming little energy and functioning at a low cost. By developing a hybrid method based on electrochemical and advanced oxidation processes, the obstacles associated with process development and conditions were overcome.

3.4. Synergistic Effect of the Combined Process. The synergistic effect (SE) is an important factor [160, 161] in the creation of a hybrid method for the removal of pollutants from wastewater. It can be defined as the increase in effect resulting from integrating two or more processes, variables, and so on, compared to the sum of their individual effects. The SE can be calculated using (19), which compares the pollutant removal efficiency of the hybrid process with the sum of the pollutant removal efficiency of the two individual processes.

$$SE = \left(\frac{X_{US+EC}}{X_{US} + X_{EC}} - 1 \right) 100. \quad (19)$$

A positive SE value indicates a positive synergistic effect. The use of the hybrid process has some advantages which contribute to its enhanced efficiency, namely, (i) increased mass transport and activation of the electrode, (ii) lack of passive layer on the surface of the electrode, and (iii) generation of more hydroxyl radicals.

4. Conclusion

Health concerns are becoming more prevalent because of environmental challenges caused by wastewater and industrial effluent. One of the major issues facing the modern era is the efficient removal of pollutants from wastewater and industrial effluent that are difficult to remove using standard treatment methods. As a result, it is necessary to design novel techniques for removing the contaminant via an altogether new treatment process. At the moment, electrochemical and hybrid AOPs are the most widely used methods for treating contaminated wastewater, and these processes have the potential for treating refractory, organic, and inorganic wastewater. Their interconnection is a solution that has the ability to provide high removal rates with minimal expenditure. In such situation, recent advancements in electrochemical, AOPs, EAOPs, and hybrid processes indicate that their application to the treatment of industrial effluents should be pursued aggressively. There is an urgent need for hybrid techniques that strike a good balance between economic viability, high removal efficiency, and ecological sustainability. Further research on these technologies should focus on real-world wastewater experiments and

continuous operation, which will permit progressive scaling up.

Data Availability

Data will be available upon request.

Conflicts of Interest

The authors declare that they have no conflicts of interest.

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