Tuijin Jishu/Journal of Propulsion Technology
ISSN: 1001-4055
Vol. 45 No. 4 (2024)

A Review on The Effectiveness of Electrocoagulation, Advanced Oxidation Process, and Aeration-Enhanced Electrocoagulation for Treating Distillery Spent Wash

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Abstract:- For every liter of alcohol generated in the distillery sector, around 8-15 liters of byproduct known as spent wash are released as wastewater; exhibiting extremely elevated levels of chemical oxygen demand as well as biological oxygen demand. In the absence of proper treatment, discharging spent wash into water-receiving bodies could result in potential harm to aquatic ecosystems. A comprehensive Systematic literature review is presented on Electrocoagulation, Aerated Electrocoagulation Process and Advanced Oxidation in treating distillery spent wash. The review encompasses several sections viz., treatment options and work done by various researchers using different electrodes, operating parameters, and variables. A cumulative 101 articles were chosen due to their content validity, relevance to the research question, strength of evidence, publication year (2003–2023), and alignment with the research area. The electrocoagulation method is acknowledged as an outstanding technique that gains various advantages viz., high efficiency over a short treatment period and cost-effectiveness in materials and operation conditions. Aerated Electro-Coagulation (AEC) is a hybrid method that merges aeration and electrocoagulation techniques within the scope of physical-chemical treatment processes. This approach eliminates the need for chemical oxidants, as the supplied oxygen is the oxidizing agent. Advanced oxidation processes are distinguished by the production of a significant amount of hydroxyl radicals, which play a pivotal role in the treatment of spent wash.

Keywords: *Distillery Spent Wash, Electrocoagulation, Electro-Fenton, Aerated Electrocoagulation..*

1. Introduction

The rise of global industrialization and urbanization has resulted in the contamination of water bodies, necessitating the treatment of industrial wastewater before its release into the environment [1,2]. Industrial activities produce a substantial amount of hazardous effluents containing numerous bio-recalcitrant compounds [3]. Ethanol is a crucial primary ingredient for manufacturing various intermediary chemicals and pharmaceuticals

[4]. The primary sources for ethanol production are sugarcane and sugar beet [5,6]. After Brazil, India is the world's second-largest sugar-producing nation [7]. Following the pulp and paper industry, primary metal industry, and chemical industry, the sugar industry sector ranks fourth in terms of water consumption [7] [8] [9]

Worldwide, there is a rising demand for alcohol and alcoholic products [10]. Distilleries represent a significant sector within the secondary agro-based industries [11]. The alcohol distillery industry is making significant efforts to meet this growing demand, with cane bagasse being a widely used feedstock for distilleries around the globe [12]. In the ethanol production process, the distillery industry generates a significant amount of liquid waste effluent known as Distillery Spent Wash (DSW) [13]. In India, there are 319 distilleries with a 3.25 billion liter installed capacity for alcohol, producing 48 billion liters of raw distillery spent wash a year [14] having a high amount of organic content, acidic pH, color [15], dissolved metallic impurities and depleted dissolved oxygen (DO) [16]. The spent wash is often released into water bodies and on the ground [14], therefore, the effluent is a significant source of water and soil contamination [17]. Distilleries have been added to the Central Pollution Control Board of India's "red category" list for this reason [8] [13].

The rise of global industrialization and urbanization has resulted in the contamination of water bodies, necessitating the treatment of industrial wastewater before its release into the environment [1,2]. Industrial activities produce a substantial amount of hazardous effluents containing numerous bio-recalcitrant compounds [3]. Ethanol is a crucial primary ingredient for manufacturing various intermediary chemicals and pharmaceuticals [4]. The primary sources for ethanol production are sugarcane and sugar beet [5,6]. After Brazil, India is the world's second-largest sugar-producing nation [7]. Following the pulp and paper industry, primary metal industry, and chemical industry, the sugar industry sector ranks fourth in terms of water consumption [7] [8] [9].

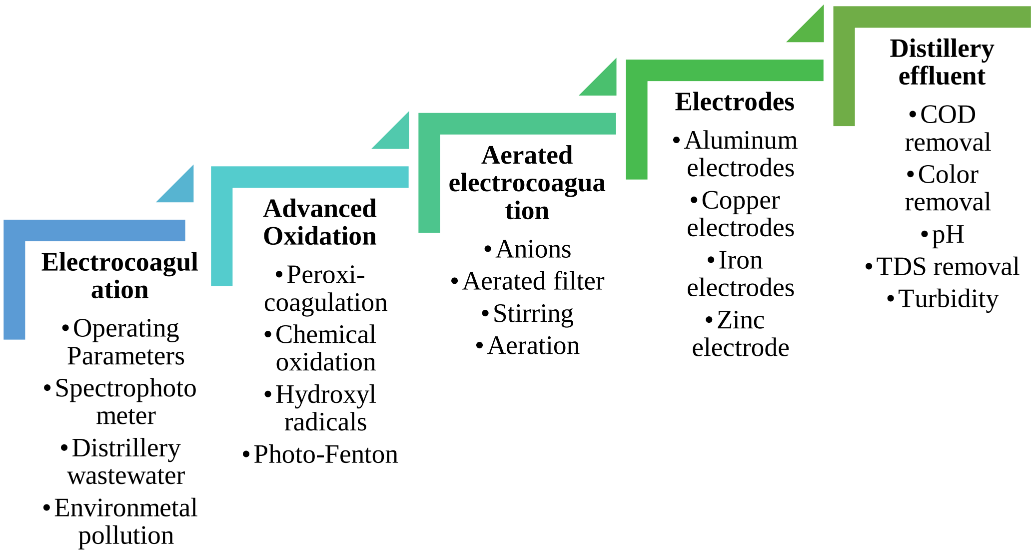

Figure 1: Milestone of the Keywords in Publications Surveyed

As observed by [26] a low BOD/COD ratio was noted, which led to the preference for physio-chemical treatment over biological treatment for Distillery Spent Wash (DSW). Within the scope of this review, physicochemical techniques such as Electrocoagulation (EC), Advanced Oxidation Process (AOP), and Aerated Electrocoagulation (AEC) are outlined in this paper. Information gathered from a systematic review of the literature is instrumental in furnishing definitive answers to particular research inquiries.

Systematic Literature Review

Researchers consistently turn to conducting a systematic literature review when aiming to determine if there exists sufficient evidence to support a particular practice [27] [28]. A Systematic Literature Review (SLR) is usually developed by thoroughly exploring relevant scientific literature to select, differentiate, and analyse specific published studies [29, 30]. Systematic reviews are instrumental in identifying evidence gaps and proposing pathways for future studies. The present study effectively illustrates the systematic literature approach in Figure 2. The initial step involves formulating the research question and establishing the scope or parameters of the literature review. The web search entailed utilizing keywords to identify and retrieve articles associated with the research question and qualitative variables. The utilized keywords comprised; distillery spent wash, industrial wastewater treatment, Electrocoagulation, Electro-Fenton, Advanced Oxidation, Aerated Electrocoagulation, and Electrodes. Establishing inclusion and exclusion criteria ensures that only pertinent articles aligned with the study's objectives are considered for inclusion, maintaining the relevance of the selected literature. In this Systematic Literature Review, we specifically focused on research articles, published between 2003-2023. The number of studies related to the research area has shown a healthy growth since 2003**.**

A tollgate approach was employed in the study selection process. Hence, after keyword searches across all subsequent databases, a total of 250 research studies were extracted. Out of the 250 studies identified, 45 were duplicates and were consequently removed. Subsequently, the inclusion and exclusion criteria were applied to the remaining 205 studies based on title, abstracts, keywords, and publication type. This screening process led to the exclusion of an additional 30 studies, resulting in a final selection of 175 studies. Following the initial screening, additional selection criteria were applied, resulting in the exclusion of another 28 studies. Consequently, only 147 studies met the refined criteria and were retained for further analysis. The subsequent and concluding stage involved the application of the quality assessment criteria (QAC) to the penultimate list of 147 studies. The quality assessment criteria were implemented at the conclusion as the final step in deducing the ultimate list of studies for the Systematic Literature Review (SLR). Typically, the QAC identifies studies whose quality does not significantly contribute to addressing the research question. Following the application of the QAC, 46 studies were excluded, resulting in a final selection of 101 primary studies. Distribution of databases of selected studies after applying selection process is shown in Figure 3.

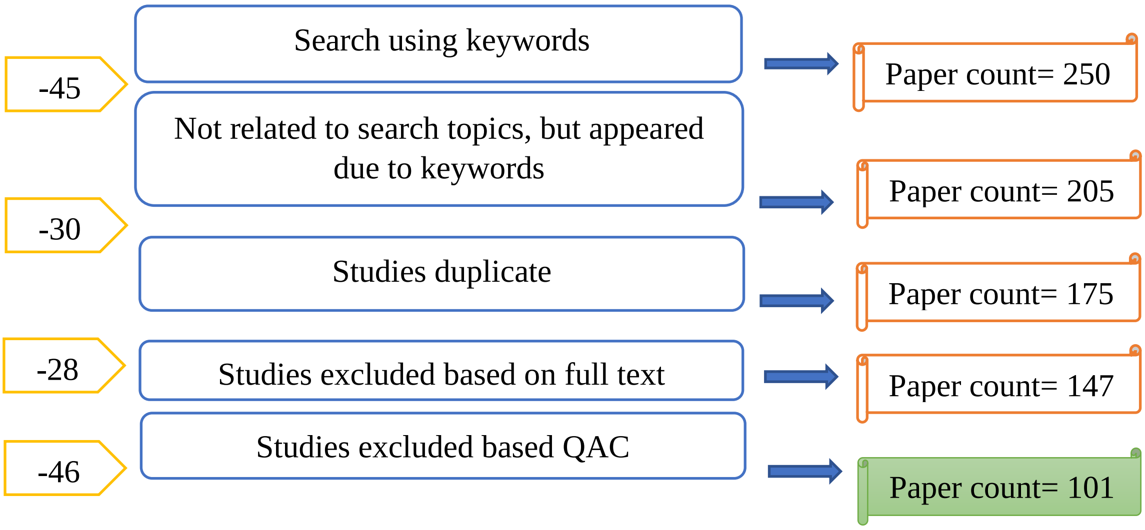

Figure 2: Systematic Overview of Selection Process

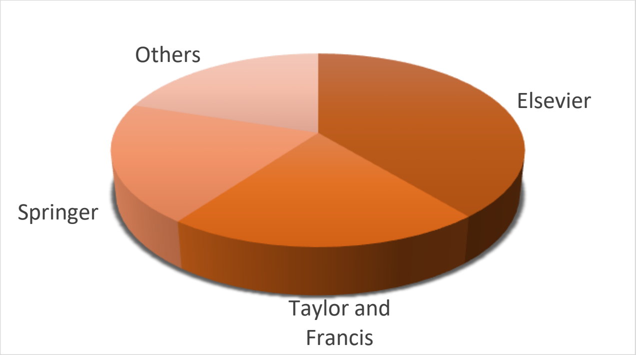

Figure 3: Distribution of Databases of Selected Studies

2.1 Research questions.

The research questions hold significant importance in a systematic literature review because they dictate the search queries and keywords that will be employed to explore research publications. The review article addresses the research questions outlined in Table 1.

Table 1: Research questions along with motivation.

Research Question	Motivation
RQ1: What different treatment processes are used?	To identify the trends in the Physio-chemical process in treating DSW over almost two decades.
RQ2: Which combination of Electrodes is used in the treatment of DSW?	An in-depth literature review will give the option the select the best electrode combination in treating DSW.
RQ3: What different operating parameters are used to obtain maximum removal efficiency?	To highlight the operating parameters that have been investigated and to identifying parameters, which need more research attention.

Figure 4 illustrates the distribution of the number of studies throughout the study period (2003-2023). A Flow diagram representing different treatment options for treating DSW is shown in Figure 5.

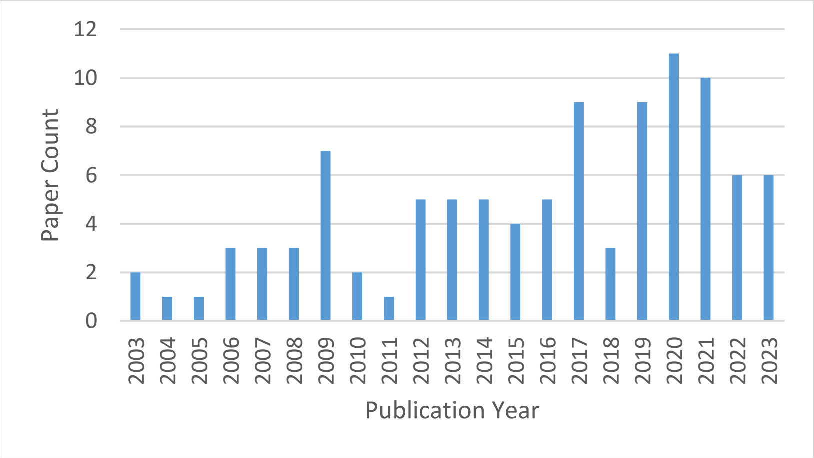

Figure 4: Publications over the years.

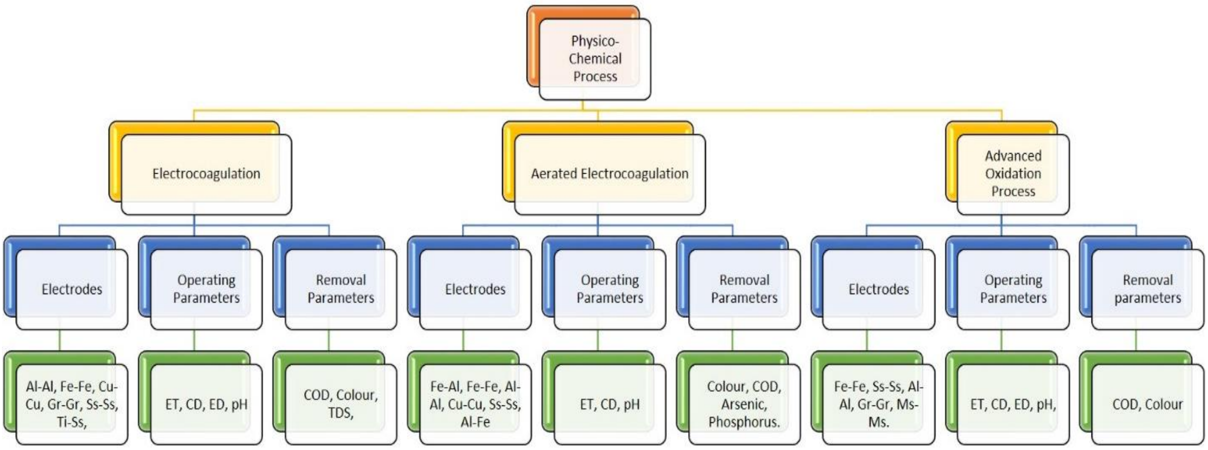

Figure 5: Flow Diagram Representing Different Treatment Process and its Protocol.

3. Characteristics of Distillery Spent Wash.

The liquid waste produced during alcohol production is called Distillery Spent Wash (DSW) [3] [8] [9] [31]. The dark brown colour of Distillery Spent Wash is due presence of Melanoidins and smells strongly of burnt sugar [4] [5] [6] [7] [12] [13] [15] [19].The vibrant components it possesses result in decreased sunlight penetration within waterbodies, subsequently lowering photosynthetic activity and the concentration of dissolved oxygen [20] [32] this situation poses a hazard to aquatic life. The pH of the untreated spent wash is between 3.8 to 4.2, making it acidic [33] [34]. A series of concurrent chemical reactions involving amino compounds and carbohydrates known as the Maillard reaction is responsible for the formation of melanoidins [2]. Melanoidins can only be degraded to a maximum of 6% to 7% using conventional anaerobic-aerobic treatment techniques [5] [10] [35]. To remove the color from spent wash, it is therefore required to research further treatments [15]. Various Physicochemical treatments can eliminate melanoidins [10] and these processes produce a lot of sludge and require high reagent dosages [15]. Treated DSW from distilleries, enriched with nutrients like nitrates and phosphates, has proven valuable for enhancing agricultural output through its utilization as fertilizers and irrigation [33]. Nevertheless, caution must be exercised regarding the extensive use of nutrient-rich distillery wastewater on land, as it could adversely impact plant growth and soil physico-chemical attributes [36]. The amount of organic matter in spent wash enhances the appeal of anaerobic treatment when contrasted with direct aerobic treatment [31]. As a result, bio-methanation becomes the principal treatment stage, frequently succeeded by a two-stage aerobic treatment before being released into a water body or utilized for land irrigation [19] [12]. Additionally, the potential contamination of ground and surface water poses a threat to aquatic life and ecosystems [37]. The typical characteristic of distillery spent wash is shown in Table 2.

Table 2: Typical Characteristics of distillery spent wash [5]

Sl.No	Parameter	Range (mg/L, except pH)
1	pH	3-4.5
2	BOD_5	50000-60000

3	COD	80000-150000
4	TS	110000-190000
5	TSS	13000-15000
6	Chlorides	8000-8500
7	Phenol	8000-10000
8	Sulphate	7500-9000
9	Phosphate	2500-2700

4. Impact of Distillery Spent Wash on Water and Soil Environment

Untreated spent wash can have implications for human health, acting as a potential origin of waterborne illnesses, attracting flies and mosquitoes, harboring germs, and emitting unpleasant odors [7]. The release of spent wash containing elevated levels of total dissolved solids (TDS) will have negative impacts on the aquatic environment, is unsuitable for consumption, and can also lead to the corrosion of pipelines [15] [19]. Suspended solids induce turbidity, leading to a reduction in the water bodies ability to transmit light effectively [3] [8] [12].

The physical, chemical, and biological characteristics of soil could drastically change as a result of the spent wash with high content of organic and inorganic materials [13]. When planning for spent wash disposal on agricultural land, soil permeability, and porosity are crucial factors to take into account [8]. Heavy amounts of organic carbon compounds from distillery waste disposal may result in high oxygen demands from bacterial activity in soils developing anaerobic conditions, which would ultimately result in a reduction in infiltration rate and a reduction in hydraulic permeability due to the buildup of particles [21] [32] [37].

However, investigations made soon after spent wash was applied to soils revealed an initial decline in the microbial populations and enzyme activity, despite the distillery effluent's richness in nutrients and organic matter being found to improve the soil microbial populations as quoted by [26]. [13] revealed that adding spent wash at a rate of 250 m^3 ha/L to the soil boosted its dehydrogenase activity.

5. Hindrance of Other Treatment Process

The wet oxidation treatment process entails the oxidation of both organic and inorganic pollutants in the presence of water and oxygen. However, it is associated with various hindrances, including elevated energy consumption, challenges related to corrosion, the generation of undesirable by-products, restricted applicability, and complexity of its operational procedures [38]. The treatment of wastewater using membrane filtration processes attains challenges such as membrane fouling, elevated energy consumption, restricted tolerance to specific substances, sensitivity to chemicals and substantial capital costs [7]. The coagulation process in wastewater treatment has specific setbacks in the generation of sludge, the utilization of chemicals, sensitivity to pH, settling time for floc formation, and the impact of variability in water quality [39]. To overcome all these hindrances electrocoagulation, advanced oxidation process, and aerated electrocoagulation for the treatment of DSW may be suitable options for treatment.

6. Electrocoagulation

The electrocoagulation method is recognized as an exceptional technique offering numerous benefits, including its high efficacy within a brief treatment duration, straightforward design, operation, gentle treatment conditions, and cost-effectiveness in terms of materials and operations [12] [18] [40] [41] [42]. Electrocoagulation technology is a treatment process that involves the application of electrical current to effectively treat aggregate contaminants without the need to introduce additional coagulants [43] [44]. Extensive research has been carried out, yielding successful applications in the removal of heavy metals, organic compounds, anions, and particulates from both domestic sewage and industrial wastewater [32] [40]. The process of electrocoagulation involves a complex series of parallel redox reactions taking place simultaneously within the system [3] [45]. Anodic oxidation leads to the liberation of metal ions from the sacrificial electrodes, leading to the creation of adsorbents by using principles of electrochemistry [20] [26] [46]. The anode electrode is responsible for generating ions to facilitate flocculation, while the cathode produces hydrogen gas to aid in the flotation contaminants [18] [43] [47] [48].

The particulates are neutralized through the formation of hydroxide complexes, to create agglomerates [46] [49]. These agglomerates start to develop at the bottom of the tank and can be siphoned out through filtration [45]. The characteristics of flocs, including their size, structure, and strength, play a vital role in the subsequent separation stages of sedimentation and filtration [43], as a result, these attributes are pivotal in determining the effectiveness of the EC process. Electrons serve as the primary reagent in these processes, yet the presence of supporting electrolytes is also essential [32] [50]. The degradation of pollutants through an electrochemical process occurs through either a direct or indirect oxidation process [48] [51]. In the case of direct anodic oxidation, pollutants initially adhere to the surface of the anode before undergoing destruction through electron transfer reactions at the anode [45]. Within an indirect oxidation process, potent oxidants like hypochlorite/chlorine, ozone, and hydrogen peroxide are electrochemically generated [52]. Subsequently, the pollutants undergo degradation within the solution volume through oxidation reactions involving the generated oxidant [53] and are promptly utilized by the process [32].

The widespread application of the Electrocoagulation process encompassed the treatment of diverse industrial wastewater streams [35] [53], notably including those originating from the tannery [46], textile [45] [53], dairy industry, Rice Grain-Based Biodigester effluent [1] [12], Bio-Treated Distillery Wastewater [19] [37] [54], Sugar processing industry wastewater (SPIW) [7], Petroleum refinery Wastewater [55], Pulp and Paper Mill Wastewater [56], Fruit-juice production wastewater [23], Mineral Processing Wastewater [57], Petrochemical Wastewater [58].

The process of EC treatment incorporates a blend of different techniques [1] [8] [31] [37] [53] [59] which involves,

- Electrochemical reactions occurring at the electrode interface.
- Creation of coagulants (anodic metal hydroxides) within the aqueous medium.
- The binding of soluble or colloidal pollutants to the coagulant surfaces through adsorption.
- Elimination through electrocoagulation, followed by sedimentation and attachment to bubbles.

The equation of the electrocoagulation process [41] [44] is given below,

At anode: $M \rightarrow Mn^{+} + ne^{-}$ Eq1

At cathode: $2H_2O + 2e^{-} \rightarrow H_2 + 2OH^{-}$ Eq2

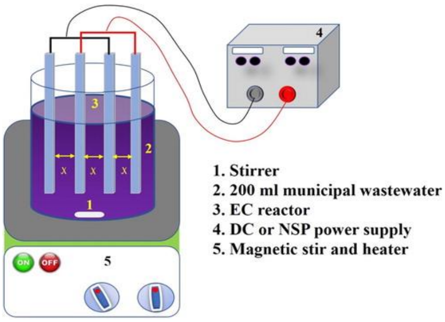

Figure 6 Schematic Diagram of Electrocoagulation Setup (Source: [23])

Through the conceptual implementation of the electrocoagulation process, the introduction of pair of electrodes choice is of fundamental importance [32] [43] and made from materials viz., Iron observed by [53], Aluminium [46] [53] [60], Copper [7] [4], Zinc [51], Graphite [23], Carbon, Iron [61] [1], Iron and aluminum [42], Ferum and Aluminium [65] or Stainless steel facilitated the generation of hydroxide flocs to destabilize the pollutants and enable them to undergo coagulation [42]. Dimensionally stable anode materials have undergone extensive research to apply organic oxidation [32].

As per the research work carried out by [63], increasing the electrode count from 2 to 6 resulted in an enhancement of electrocoagulation performance, however, this adjustment also led to a rise in energy consumption during the EC process. The findings of the experiments conducted by [1], the potential use of electrocoagulation in rice grain-based distilleries to mitigate pollution impact, the COD degradation was observed 93% at current density (CD) 99 Am^{-2} and Colour removal was 77%. As the current density rises, there is a corresponding improvement in COD removal efficiency. This occurs because heightened current density leads to a greater release of Al^{3+} ions, resulting in the formation of increased aluminum hydroxide cations. As observation made by [7], the COD degradation at CD 89.23 Am^{-2} in Sugar Processed Industry Wastewater (SPIW) was 73%, The primary reactions involved the dissolution of iron cations from the sacrificial Fe electrode (anode) into ferrous ion (Fe^{2+}) and/or ferric ion (Fe^{3+}), alongside hydroxide ion (OH^-) generation on the Al electrode (cathode) due to water hydrolysis, ultimately coagulants in the form of $Fe(OH)_2$ and/or $Fe(OH)_3$ were produced [42]. Table 3 displays the collection of Research work concerning operating parameters and the effectiveness of electrocoagulation systems in eliminating different pollutants from diverse industrial wastewater sources. The Schematic diagram of the electrocoagulation setup is shown in Figure 6 [23].

Table 3: Works done by various researchers on the Electrocoagulation process on various industrial wastewater using different electrodes.

Reference	Wastewater type	Treatment	Electrode combination (anode-cathode)	Reactor type	Operating parameters	Removal Parameters	Removal Efficiency
[1]	Rice Grain-Based Distillery Biodigester effluent	EC	Fe-Fe	Batch	DC- 5A	COD Color	93% 77%
[2]	Distillery effluent	EC		Batch Continuous	Energy-9.77$kWkg^{-1}$	COD	52.94% 83.2%
[3]	Distillery wastewater	EC	Poly-acrylic sheet and Fe	Batch	pH- 2-10 CD-13,26 and 40Am^{-2}	TOC TP	73% 85%

[4]	Rice Grain-Based Distillery Biodigested effluent	EC	Cu-Cu	Batch	CD- 89.2 Am^{-2} pH- 3.5	COD Colour	80% 65%
[12]	Bio-digester effluent	EC	Fe-Fe	Batch	pH-5.6	COD Color	50.5% 95.2%
[14]	Distillery spent wash	EC	Al-Al Fe-Fe	Batch	pH-8 Vol- 25V	COD Color	85.71% 99.78%
[16]	Sugar industry effluent	EC	Cu-Cu	Batch	CD-133Am^{-2} pH- 6.5	Color	95.67%
[20]	Distillery spent wash	EC	Al-Al Fe-Fe	Batch	pH 3 CD- 0.187 Acm^{-2}	COD	81.3%
[22]	Reverse Osmosis reject from distillery	EC	Al-Al Fe-Fe	Batch	pH- 7.2 current density(CD)- 1.79 Am^{-2} Electrolysis time (ET)- 3hr	COD	Al-Al=84.66% Fe-Fe=76.9%
[26]	Distillery Wastewater	EC	Fe-Fe Al-Al Ss-Ss	Batch	Vol-80V	COD BOD TDS OM	Fe-Fe
[32]	Distillery spent wash	EC	Ti-Ss	Batch	CD-0.01428 Am^{-2} pH- 5.5	Color COD	81% 37%
[33]	Distillery spent wash	EC	Fe-Ss	Batch	CD-0.031 Am^{-2} ET – 4hrs	Color	93.5%
[37]	Bio-Treated Distillery Wastewater	EC	Al-Al	Batch	pH-3 CD- 35.9 Am^{-2}	COD Nitrate PO_4^{3-}	50% 26% 61%
[53]	Distillery spent wash	EC	Fe-Fe Al-Al	Batch	pH 6-12	COD	88% 64%

[64]	Anaerobically digested distillery spent wash	EC	Al-SS-Al	Batch	ET-150min CD-0.088 Am^{-2} Electrode distance (ED)-20mm	COD Color Turbidity TDS TSS	84.64% 91.37% 86.92% 29.2% 77.62%
[65]	Distillery Spent Wash	EC	Aluminium foil scraps	Batch	ET-150min	COD Color TDS Sulfate Chloride	77.29% 76.55% 58.32% 64.72% 20.44%
[66]	Sugar industry Wastewater	Combination Thermal and EC	Al, Fe, and Cu	Batch	pH-6 CD-156Am^{-2} ED-20mm ET-150min	COD Color	Thermal 75.6% 79.2% Combined 97.8% 99.7%
[67]	Bio-digest distillery spent wash	EC	Al-Al	Batch	CD-120 Am^{-2} pH 6 ED- 1cm	COD	52.23%

7. Aerated Electrocoagulation

Aerated electro-coagulation (AEC) can be defined as a hybrid approach that combines both aeration and electrocoagulation methods within the realm of physical-chemical treatment processes [62]. In aerated EC reactors, there is no requirement for the addition of chemicals for oxidation since the oxygen supplied serves as the oxidation agent [68]. In the Aerated Electrocoagulation setup, aquatic aerators were positioned at the bottom of the reactor and activated for aeration [69]. Aeration can result in an elevated COD removal rate because it facilitates thorough circulation and the effective formation of $Fe(OH)_3$ coagulants [42]. Increasing the aeration rate will result in an enhanced efficiency of the AEC process [62]. Moreover, aeration within the EC reactor has the dual benefit of reducing the formation of passivation layers on the electrode surfaces and facilitating effective mixing within the reactor [68]. Optimizing the parameters influencing system performance is crucial in achieving an efficient aerated iron electrocoagulation (EC) mechanism [69].

The widespread application of the Aerated Electrocoagulation process encompassed the treatment of diverse industrial wastewater streams [59], notably including those originating from municipal wastewater [25], groundwater [70], Leachate [62], Dairy Industrial Wastewater [71], Oily industry [59], Natural water and wastewater [69].

Electrocoagulation and aerated electrocoagulation experiment as observed by [42], and results were compared and observed that the COD removal was increased from 72.9% (EC) to 90% in the AEC process, this is due to the introduction of aeration at the reactor's base improves solution mixing and generates turbulence around the electrodes [62] [72]. This facilitates the escape of reactive species from the anode, enhancing the overall effectiveness of the process [59].

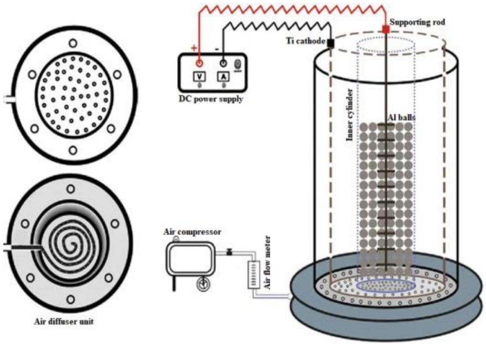

Figure 7: Schematic Diagram of Aerated Electrocoagulation Setup (Source: [68]).

Aeration introduced an ample amount of dissolved oxygen, enabling the conversion of generated Fe^{2+} into Fe^{3+} [59] [73] due to the rapid production of oxygen in the solution, $Fe(OH)_3$ demonstrated greater coagulation effectiveness compared to $Fe(OH)_2$ [25]. Furthermore, continuous aeration contributed to the improvement of mixing and collisions between pollutants and electro-coagulants. Aeration played a pivotal role in enhancing the process's performance, underscoring the significant potential of electrocoagulation processes for treating oily wastewater [59]. [25] Carried out aerated electrocoagulation experiments employing Cu anodes and concluded that aeration had a positive impact on the anodic reaction, leading to an increased generation of Cu^{2+} ions in the solution, thereby promoting coagulation. Continuous aeration can enhance EC performance by promoting vertical movement and facilitating complete circulation between the internal and external electric fields. Figure 7 illustrates how air diffuses throughout the system resulting in effective pollutant separation at the surface, facilitated by the presence of air bubbles [42]. In non-aerated experiments, a faint scum layer was observed by [59], on the surface at a lower current density, and it became more substantial at 90 and 120 Am^{-2}. Additionally, in aerated experiments, there was a distinct absence of foam formation behavior and this can be attributed to the continuous stream of air bubbles mixing the solution, preventing the development of foam [70]. The findings of [68] indicated that, the utilization of aerated EC reactors with sphere-shaped electrodes for arsenic removal from groundwater proved to be more effective than traditional EC reactors. Table 4 displays the collection of literature concerning operating parameters and the effectiveness of Aerated Electrocoagulation in eliminating different pollutants from diverse industrial wastewater sources.

Table 4: Works done by various researchers on the Aerated Electrocoagulation process on various industrial wastewater using different electrodes.

Reference	**Wastewater type**	**Treatment**	**Electrode combination (anode-cathode)**	**Reactor type**	**Operating parameters**	**Parameters**	**Removal Efficiency**
[70]	Groundwater	AEC	Fe-Fe	Batch	Vol- 5V ET- 60min	Arsenite	98%
[24]	Composite Wastewater	AEC	Fe-Fe	Batch	Vol- 1V	COD	50%

		Peroxi-coagulation			pH- 3 ET- 1h	Color	60% 77.7% 97%
[25]	Municipal wastewater	Biological Aerated Filter with EC	Cu-Cu	Batch	ET- 1hr CD- 3A Vol- 24V	Phosphorus	98%
[42]	Coolant wastewater	AEC EC	Fe-Al	Batch	ET- 15min	COD	90% 72.9%
[59]	Oily wastewater	AEC EC	Ss-Ss	Batch	CD-120Am^{-2}	COD	93.3% 84%
[62]	Leachate	AEC	Ferum and Al	Batch	CD- 200 Am^{-2}	NH_3-N Color	40.66% 85.91%
[70]	Groundwater	AEC	Fe-Fe	Batch	Flow rate- 10 L/h ET 6hr	Arsenite	99%
[71]	Dairy Industry	AEC	Al-Fe	Batch	pH- 7 Vol- 5V	COD	97%
[74]	Perfluorooctanoic acid (PFOA) in aqueous solution	AEC Peroxi-coagulation	Fe-Fe	Batch	ET – 60min CD- 78.34 Am^{-2} pH-3.6	PFOA	56% 90%
[75]	Groundwater	AEC	3D Al-Al	Batch	ET-30min	Arsenic	98.6%

8. Operational Parameters during Electrocoagulation and Aerated Electrocoagulation Treatment.

8.1 Effect of pH

The effectiveness of electrocoagulation treatment and Aerated Electrocoagulation treatment for effluents is significantly influenced by the solution's pH [53]. Initial pH plays a pivotal role in influencing the charge of particles, and consequently, it greatly impacts the overall performance of the process [59]. Within the aerated electrocoagulation process, a greater consumption of H^+ ions occurs in conjunction with oxygen, resulting in more favorable effluent pH values when compared to the non-aerated EC process [7]. At a lower pH, the electrodes experienced passivation due to the formation of oxide layers [76]. The primary cause of neutralization in EC solutions is the generation of OH^- ions at the cathode, leading to an increase in the solution's pH [25] [59]. Also, oxygen reduction can take place at the cathode by consuming H^+ ions. For instance, textile effluents are typically optimized within a pH range of 3–5 [45] [53], while effluents from distillery industries are usually optimized at pH levels between 4.5 and 5.5 [4]. In the experiment carried out by [1], the impact of pH on the treatment of Bio Digested Effluent (BDE) was examined while maintaining a constant current density of 99 Am^{-2}, enhanced COD and color reduction of BDE solution were observed as pH increased from 3.5 to 8, yet diminished when pH exceeded 9.5 due to characteristics and volume of hydroxide ions produced at particular pH. The pH significantly impacted the chemical breakdown of iron in wastewater within the pulp and paper industry as quoted by [56]. In an investigation by [37] [53], the final pH consistently exceeded its initial pH in EC as it demonstrates a degree of pH buffering, particularly in alkaline conditions.

Within the aerated iron electrocoagulation process, a greater consumption of H^+ ions occurs in conjunction with oxygen, resulting in more favorable effluent pH values when compared to the non-aerated EC process [69]. Following a 90-minute reaction period with an initial pH of 3.8, it was observed by [59] that the pH of non-aerated solutions rose within the range of pH 3.9 to 4.9, the aerated solution maintained a neutral pH within the range of 6.9 to 7.6 until the end of the process, when subjected to current density ranging from 30 to 120 Am^{-2}. Neutralization in the solution primarily occurs due to the generation of OH^- ions at the cathode. The anodes within the aerated cells of the EC underwent a more evenly distributed consumption. The introduction of aeration notably enhanced the process's efficiency.

8.2 Effect of Current Density.

Current density refers to the amount of electric current flowing through a given unit of the electrode's cross-sectional area [3]. The current density holds significant importance in electrocoagulation processes and AEC as it governs factors like the rate at which coagulation dosage is applied, reaction rate, the rate of bubble generation [1] [56], formation, size and development of flocs [37]. [77] reported that elevated current density levels led to an increased concentration of iron ions, consequently causing higher precipitation of pollutants. The study also noted a positive correlation between the rate of hydrogen bubble production and diffusion with higher current density. The effectiveness of pollutant removal from wastewater is significantly influenced by the quantity of metal ions liberated during the process. The release of these metal ions is contingent upon the current density applied. Faraday's law establishes a connection between current density and the quantity of anode material that undergoes dissolution within the sample [12] [37] [78].

Research work conducted by [45] [51] concluded that, when using higher cell currents, there was an increase in the amount of metal oxidation, leading to the generation of a larger quantity of hydroxide flocs for pollutant removal. Conversely, as the conductivity level increased, a slight enhancement in the efficiency of COD (Chemical Oxygen Demand) was achieved using Fe-Fe electrodes [53]. The degradation of COD and Colour increases with an increase in current density [1] [4], supported by Faraday's law, the highest degradation of COD in Bio-Treated Distillery Wastewater (BTDW) was 60% at CD 36 Am^{-2} and CD was as significant factor as observed by [37]. At 73.75 $Am^{-2,\ a}$ COD degradation of 77% was observed by [56] for a four-plate setup. [8] [53] revealed that higher ionic strength typically leads to a rise in current density under the same cell voltage, or the cell voltage decreases with a rise in wastewater conductivity while maintaining a constant current density. In the study carried out by [22], it was observed that the highest efficiency in COD (Chemical Oxygen Demand) removal was achieved when utilizing a current density of 1.79 Am^{-2} during a 3-hour electrolysis period, the rise in current density results in an escalated anode dissolution. In the findings reported by [58], it was noted that the efficiency increased as the current density was raised to 0.8A. However, beyond this point, a further increase in current density resulted in a decrease in efficiency, the reduction could potentially be attributed to the existence of a passive layer forming on the electrode's surface.

Introducing aeration at the reactor's base, a comparison was made by [59], between the COD removal in cells with AEC and those with EC, the COD removal efficiency for the aerated cell was higher at the same current density, demonstrating its superior performance compared to the non-aerated cell, by introducing aeration at the reactor's base improves solution mixing and induces turbulence around the electrode, this, in turn, facilitates the escape of reactive species from the anode. According to the findings presented by [62] review, the optimal range for current density in AEC experiments for the effective removal of NH_3-N and color falls between 50 Am^{-2} and 300 Am-4e5re3aw^{2}. In the study conducted by [74], it was observed that an efficient Aerated EC process was established at a pH of 3.6. Under optimal conditions, a 58% degradation of PFOA, a 51% reduction in TOC, and a 38% defluorination were achieved, with a current density of 78.34 Am^{-2}. Faraday's law establishes a connection between current density and the quantity of anode material that undergoes dissolution within the sample [1] [3] [12] [37] [59] [100].

8.3 Effect of Electrolysis Time.

Even with the pH changes within the system, the retention time in the bipolar electrocoagulation reactor and AEC remained a crucial factor [23] [69]. The efficiency rises with prolonged duration, with increased time, there is a concurrent rise in the concentration of hydroxyl ions. However, after a certain point of prolonged electrolysis, the pollutant removal efficiency stabilizes and doesn't continue to increase despite further extension of time. This phenomenon could be attributed to an excess quantity of ions, which leads to the destabilization of particles [3]. With an extended electrolysis duration, the energy consumed per unit volume (per cubic meter) increases by Joule's law [69]. It was noticed by [12] that an extended electrolysis time leads to a higher concentration of iron ions and the formation of hydroxyl flocs, as a result, an increase in electrolysis time enhances the efficiency of COD removal. To investigate the impact of operating time, the current density was maintained at a constant value of 100 A/m^2 [53], the aluminum electrode exhibited effective removal efficiencies after 15 minutes, which is 50% longer compared to the time required for the iron electrode.

Within the AEC process, the operational duration stands out as a crucial parameter directly influencing the removal of arsenic as observed by [68], additionally, factors such as energy consumption, electrode wear, sludge formation, and operational costs are primarily contingent on the duration of were also affects on the removal efficiency. As the results indicated as the operating time increased, there was a corresponding rise in the removal of both anions and arsenic, aeration at the bottom of the reactor enhances the mixing of the solution and initiates turbulence around the electrodes. [3] noted that a retention time of 12 minutes resulted in higher removal efficiencies for both TOC and TP. Longer retention times were found to augment the dosage of adsorbents in the system, thereby improving the efficiency of pollutant removal. The kinetics of turbidity removal were investigated by [58], by altering the concentration of NaCl within the range of 0.5 to 2 g/L. The total treatment time for these experiments was set at 30 minutes. The enhancement in turbidity removal with NaCl is attributed to the generation of chloride ions. These ions effectively eliminate the passive oxide layer that forms on the electrode surface, thereby increasing the presence of hydroxide in the solution and improving overall efficiency.

9. Advanced Oxidation Process (AOP)

The combination of electrochemical processes and advanced oxidation techniques presents an interesting solution for treating industrial wastewater [79] [80]. Advanced oxidation processes (AOPs) are characterized by the generation of a substantial quantity of hydroxyl radicals, which play a crucial role in the treatment of wastewater [9] [81]. Adopting this interdependent approach, maximum removal efficiency can be achieved by keeping operating costs to a minimum [82]. Advanced Oxidation Processes (AOPs) are wastewater treatment methods conducted at room temperature and atmospheric pressure [81] these processes entail the production of a significant quantity of hydroxyl radicals, leading to effective wastewater purification [76]. Advanced oxidation processes (AOP) can be implemented through various methods, including combinations such as hydrogen peroxide/ultraviolet irradiation (H_2O_2/UV), ozone/ultraviolet irradiation (O_3/UV), and ozone/hydrogen peroxide (O_3/H_2O_2) [2] [83]. These combinations harness the power of ozone and hydroxyl radicals, potent oxidizing agents that can efficiently oxidize a diverse array of organic compounds when dissolved in wastewater [82]. Hydroxyl radicals are highly reactive species that typically exhibit rate constants of 10^6 to 10^9 L/mol-sec when interacting with a wide range of organic molecules [76]. The Fenton oxidation remains a highly employed advanced oxidation technique [84].

The widespread application of the Advanced Oxidation process encompassed the treatment of diverse industrial wastewater streams [82], notably including those originating from Biomethanated spent wash [19], real dyeing wastewater [5] [85], Textile wastewater [52] [86], Distillery spent wash [87] olive oil mill wastewater [79], Synthetic Dyes [88], Brewery industry [82]. Anaerobically digested effluent [9], Dye Intermediate Wastewater [75], Fruit-Juice production Wastewater [23], Textile industry, Synthetic dyes [88] and Tannery industry [101]. Selecting an electrode material that offers both excellent performance and affordability holds significant importance for the practical implementation of the electro-Fenton process [78]. Advanced Oxidation Processes (AOP) are characterized by the production of radicals, that can oxidize most of the organic and inorganic content to produce hydrogen peroxide (H_2O_2) [89] [90]. The potential of Fenton's reagent in effectively breaking down organic compounds, either partially or entirely, shows promise [5].

In the traditional Fenton procedure, hydroxyl radicals arise through the interaction of hydrogen peroxide (H_2O_2) with a catalyst consisting of Fe^{2+} ions [6] [19] [76]. The merits of the Fenton method encompass its remarkable effectiveness and straightforward operational process, ensuring its extensive utilization in the treatment of sewage and industrial wastewater [90] and relies on several key factors viz., the concentration of H_2O_2, the ratio of Fe^{2+} to H_2O_2, the pH level, and the duration of the reaction [18] [82]. Furthermore, the initial concentration and nature of the pollutants, along with temperature, play a significant role in determining the ultimate efficiency of the process [83]. Figure 8 illustrates the oxidation mechanism utilized in the Fenton process [91].

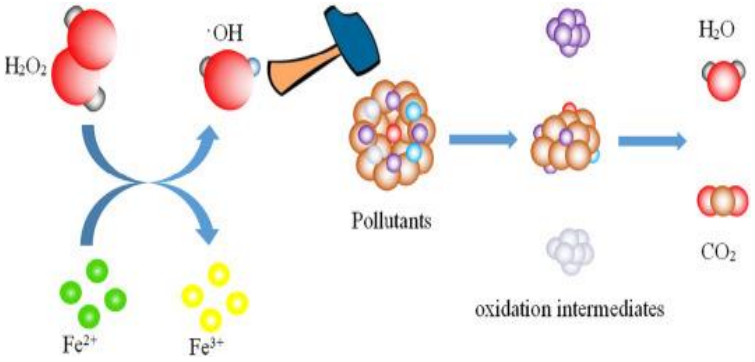

Figure 8: Reaction Mechanism for the Fenton Process (Source: [91]).

The primary benefit of the Fenton process lies in the fact that the reagent components are both safe to manage and environmentally friendly [92]. Although the Fenton process has shown success on a laboratory scale, its application on an industrial scale remains limited [84]. This is primarily because it is not very effective in reducing certain recalcitrant pollutants, such as acetic acid, oxalic acid, and trichloride-ethane [93]. Additionally, the process generates a high amount of total dissolved solids, further complicating its industrial use [82]. However, drawbacks associated with the conventional Fenton process have also been documented, including issues like the build-up of iron sludge, elevated expenses, and the potential hazards linked to the storage and conveyance of H_2O_2 [90] [93]. To enhance the management and the quality of sludge produced through the application of the Fenton reagent, the electro-Fenton (EF) process was developed [94]. This process is based on the principles of ionization, oxidation, and the separation of wastewater constituents at the atomic level through the use of electric currents [82]. The work carried out by [95] found that pre-treating diluted wastewater with Fenton's reagent resulted in enhanced biomethane production and improved removal of COD (Chemical Oxygen Demand).

The Electro-Fenton process is an advanced oxidation technique [82] that utilizes the external addition of ferrous ions and in-situ generated hydrogen peroxide to create highly reactive hydroxyl radicals within a water medium, as described in Equation 4 [78] [90] [93]. In an acidic environment, hydrogen peroxide is produced at the cathode surface through a two-electron reduction of oxygen, as illustrated in Equation 5 [24].

$H_2O_2 + Fe^{2+} \quad Fe^{3+} + \cdot OH + OH^{-} \rightarrow$ Eq 4

$O_2 + 2H^{+} + 2e^{-} \quad H_2O_2 \quad \rightarrow$ Eq 5

Furthermore, the Fe^{2+} catalyst has the potential to be restored through electro-reduction of Fe^{3+} occurring at the cathode's surface, leading to a reduction or even elimination of iron sludge formation [90]. Although Fenton's reagent boasts high oxidative efficiency, its practical use is constrained by challenges related to storing and transporting concentrated H_2O_2 and the formation of Fe(III) sludge [5]. One of the key benefits of Electo-Fenton (EF) is its ability to generate hydrogen peroxide (H_2O_2) on-site, thus eliminating the need for potentially hazardous transportation and storage of this chemical [85] [93]. The effectiveness of oxygen reduction at the cathode plays a pivotal role in the Electro-Fenton process [36]. Oxygen can undergo reduction at the cathode through two distinct pathways, resulting in the formation of two different products [90] [96]. At the anode, the formation of heterogeneous hydroxyl radicals through the process of water oxidation plays a role in contributing to the degradation of pollutants [82] [90]. Table 5 displays the collection of literature concerning operating parameters and the effectiveness of AOP. The Electro-Fenton process is emerging as a promising substitute for wastewater decontamination, with growing interest in comparison to the conventional Fenton method [90].

Table 5: Investigative Studies carried out by Various Researchers on the Advanced Oxidation Process on Various Industrial Wastewaters using Combination of Electrodes.

Reference	**Wastewater type**	**Treatment**	**Electrode combination (anode-cathode)**	**Reactor type**	**Operating parameters**	**Parameters**	**Removal Efficiency**
[9]	Anaerobically Digested Distillery spent wash	EF	Fe-Fe	Batch	pH-3 EC-18W Speed-500rpm ET-120min	COD	24.9%
[10]	Distillery spent wash	EC EF Fenton	Fe-Fe Graphite	Batch	pH-3-7 CD- 4-5 A ET 2-4 hr	Color	79% 44% 66%
[11]	Biomethanated distillery spent wash	EC Adsorption FO	Al-Al	Batch	-	COD	45%
[19]	Biomethanated spent wash	Fenton	-	-	pH-3	COD	70%
[61]	Industrial Wastewater	EC EO EC-EO	Fe-Fe BDD-Fe	Batch	ET- 30min ET-3hrs ET-90min	COD	50%
[87]	Distillery Spent wash	Fenton	-	-	pH-3.45	COD EC TDS	93.45% 85% 82%

						Turbidity	92%
[97]	Distillery Spent wash	EF	Fe-Fe	Batch	pH-4 ET-55min Dosage-25ml/L Vol- 3V	COD Color	35.31% 88.55%
[98]	Spent wash, Biomethanated spent wash	EF	Fe-Fe, Ss-Ss	Batch	pH-3.4 $V \le 1.23V$	COD	60% 50%

10. Operational Parameters during Advanced Oxidation Process (AOP)

10.1 Effect of H_2O_2 Concentration

Optimizing the quantity of H_2O_2 employed in the electro-fenton procedure is crucial, considering both cost and the potential adverse consequences associated with H_2O_2 utilization [76] [82]. The Fenton reaction commences by oxidizing the ferrous ion (Fe^{2+} cation) to the ferric ion (Fe^{3+} cation) in the presence of hydrogen peroxide, which serves as an oxidizing agent. Consequently, this process leads to the creation of a hydroxide ion and a hydroxyl free radical as by-products [94]. The significant influence of H_2O_2 concentration on pollutant degradation within the Fenton process, there is a strong need to innovate and create an effective cathode that enhances H_2O_2 production [90]. A desirable cathode should exhibit excellent chemical stability, minimal catalytic activity for H_2O_2 decomposition, and substantial over potential for H_2 evolution [78]. An experiment carried out by [76], revealed that with an increase in the dosage of hydrogen peroxide, there was a corresponding elevation in the percentage degradation of the pollutant. However, the excess amount of unused hydrogen peroxide in the Fenton process contributes to the chemical oxygen demand (COD), rendering an excessive quantity undesirable. [82] investigated the removal of BOD and COD in the electro-Fenton (EF) process in three separate sample replicates, the results indicated that the maximum BOD and COD removal rates reached up to 96.9% and 93.16% respectively. Importantly, these removal rates remained consistent without any statistically significant changes ($p < 0.05$) [82]. In the research conducted by [36], it was observed that the oxidizing capability of H_2O_2 alone was insufficient for the degradation of large molecules. To address this, a series of experiments were conducted to determine the optimal amount of Fe(II) to be added. The introduction of ferrous ions led to a significant enhancement in color removal efficiency, increasing it from 9% to 46%, the surplus ferrous ions tend to react with the hydroxyl free radicals, which possess a high oxidative potential, consuming them in the process.

10.2 Effect of pH AOP

pH plays a pivotal role in influencing the charge of particles, and consequently, it greatly impacts the overall performance of the process [59]. The optimal solution pH for the degradation of pollutants in both Fenton and Electro-Fenton processes was reported to be pH 3 [24] [36] [82] for the generation of hydrogen peroxide since the conversion of dissolved oxygen into hydrogen peroxide consumes protons in an acidic solution, as quoted in Equation 2 [78]. An acidic environment, characterized by a low pH, promotes the generation of hydrogen peroxide [52], this may be because the conversion of dissolved oxygen into hydrogen peroxide involves the consumption of protons in an acidic solution [36]. The research work conducted by [85], investigated the impact of the initial pH of the solutions, the study was carried out with an initial COD concentration of 2500 ppm and a current density of 13 Am^{-2}, while 234 mg/L of H_2O_2 was added. The research indicated that the most effective pH value was 5. The creation of $Fe(OH)_3$ not only decreased the concentrations of dissolved Fe^{3+} but also impeded the regeneration of Fe^{2+} by partially covering the electrode surface. [10], observed that the highest color reduction, reaching up to

44%, was achieved at pH 3 after a treatment duration of 4 hours. However, as the pH was increased towards neutrality, the efficiency of color removal decreased, because the fenton process exhibits higher efficiency under acidic conditions. Several researchers have consistently identified the optimal pH for the electro-Fenton process to be around pH 2.8–3. This pH range allows for maintaining the catalytic behavior of the Fe^{2+}/ Fe^{3+} coupling at its best [36] [24] [76] [82] [93].

10.3 Effect of Current Density on AOP

Under a specific current density, the production of H_2O_2 exhibited a linear correlation with time [90] and shorter electrolysis time is needed when applying a higher current density [9]. The applied current density generates a driving force that leads to the reduction of oxygen at the cathode, ultimately resulting in the production of hydrogen peroxide [52]. The elevated current leads to a rapid generation of H_2 bubbles, which hinders floc formation and consequently reduces the percentage of pollutant removal [37]. As the current is increased, it also enhances the electro-regeneration of ferrous ions and boosts the efficiency of Fenton chain reactions [52]. The lack of correlation between current efficiency and current density could be attributed to the brief residence time, which constrained the occurrence of side reactions involving H_2O_2 decomposition [90], it is important to highlight that the enhancement in removal efficiency was only marginal upon further elevating the current density from 100 to 200 Am^{-2}, the observation aligns with findings from a previous research carried out by [99]. The study conducted by [18], explored the impact of current density on the removal of color and Chemical Oxygen Demand (COD) in distillery wastewater, the experimental findings revealed that as the current density increased from 0.03 to 0.23Adm^{-2}, the efficiency of color and COD removal significantly improved, rising from 31% to 100% for color removal and from 25% to 97% for COD removal, the increased electrical current density results in the rapid formation of hydrogen gas bubbles, impeding the formation of flocs. [85], noted that as the current density increased, the production rate of hydrogen peroxide on the cathode also increased, consequently enhancing the efficiency of color removal.

Conclusion

A systematic review of the literature showcases how electro-coagulation (EC), Aerated electro-coagulation (AEC), and Advanced Oxidation processes can effectively treat industrial wastewater over several years. The choice of physicochemical treatment for industrial wastewater is contingent upon the characteristics of the effluent. It is anticipated that mechanisms change the process, and determining the primary mechanisms and their specific roles remains an unresolved aspect. It is essential to conduct an initial analysis of the wastewater beforehand. In terms of electrode specifications for various treatment processes, there is an enhancement in non-corrosiveness, increased capacity for hydroxyl radical formation, and the demonstration of effective oxidation properties. Optimizing a set of relevant input process parameters, viz., pH, current density, inter-electrode distance, agitation speed, and Fenton's reagent concentration, is required to achieve the highest possible removal efficiency for each process. However, the electrocoagulation method continues to face a deficiency in a fundamental and methodological approach at a mechanistic level, as indicated by the absence of uniformity in reactor design and application. A coherent and systematic strategy for gaining a fundamental comprehension of electrocoagulation remains difficult to find and necessitates further investigation. Combining electro-coagulation with aeration presents a viable method for treating industrial effluent. The introduction of aeration is expected to accelerate the oxidation rate, enhancing the effectiveness of the process and enabling efficient decolorization of the effluent. Exploring research in this field offers several advantages concerning system efficiency, size, initial investment costs, treatment adaptability, process stability, and operational expenses. The available literature indicates that the electro-Fenton process exhibits superior effectiveness in treating diverse types of industrial wastewater when contrasted with conventional Fenton processes. The Fenton process is capable of functioning at ambient temperature; however, the composition of the water matrix, particularly the concentration of alkalinity, significantly influences the oxidation process.

Acknowledgement

The author would like to express sincere thanks to Jnana Yaana Research Fellowship, Visvesvaraya Technological University, Belagavi, and UBDT College of Engineering Davanagere, A constituent college of VTU University, Department of Civil Engineering, Davanagere Karnataka, India.

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