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Research Article

Treatment of Distillery Industrial Wastewater Using Ozone Assisted Fenton's Process: Color and Chemical Oxygen Demand Removal with Electrical Energy per Order Evaluation

Perumal Asaithambi,[1] **Mamuye Busier Yesuf**,[1] **Seifu Kebede Debela**,[1] **Rajendran Govindarajan**,[2] **N. M. Hariharan**,[3] **and Esayas Alemayehu**[1,4]

[1]*Faculty of Civil and Environmental Engineering, Jimma Institute of Technology, Jimma University, P.O. Box—378, 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 and Technology, Chembarambakkam, Chennai 600123, Tamil Nadu, India*
[4]*Africa Center of Excellence for Water Management, Addis Ababa University, P.O. Box-1176, Addis Ababa, Ethiopia*

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

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Ozonation is one of the most effective and efficient advanced oxidation processes (AOPs) and has shown great potential in the treatment of industrial effluent and wastewater. In the present work, the ozone-Fenton process for % COD and color removal together with electrical energy per order (EE/O) determination for distillery industrial wastewater (DIW) was established. The process was developed by combining the ozone (O_3) with the Fenton (Fe^{2+}/H_2O_2) process. The ozone-Fenton ($O_3/Fe^{2+}/H_2O_2$) was compared with other treatment processes such as O_3, Fe^{2+}, H_2O_2, O_3/Fe^{2+}, O_3/H_2O_2, and Fe^{2+}/H_2O_2 for EE/O together with % COD and color removal efficiency for DIW. The removal of color at 100% and chemical oxygen demand (COD) of 96.875% were achieved with a minimum of EE/O of 0.5315 kWh/m^3 using the $O_3/Fe^{2+}/H_2O_2$ process by operating at optimum conditions. The % COD and color values obtained using $O_3/Fe^{2+}/H_2O_2$ were significantly higher than those obtained using O_3, Fe^{2+}, H_2O_2, O_3/Fe^{2+}, O_3/H_2O_2, and Fe^{2+}/H_2O_2 processes. The % color, % COD removal, and its associated EE/O were evaluated by varying Fe^{2+}, H_2O_2, O_3 inlet and COD concentration, and initial wastewater pH using the $O_3/Fe^{2+}/H_2O_2$ process. The synergy effect of the O_3 and Fe^{2+}/H_2O_2 processes was evaluated and reported. Our experimental findings suggest that combining O_3 with the Fe^{2+}/H_2O_2 process could effectively treat industrial effluent and wastewater.

1. Introduction

Nowadays, the ozonation (O_3) process is commonly used to treat water, wastewater, and industrial effluents in a variety of applications, including disinfection of swimming pool [1], drinking [2], domestic [3], distilleries [4, 5], fermentation [6], dyeing [7], slaughterhouse [8], laundry [9], municipal [10], pharmaceutical [11], synthetic textile [12], RB 5 dye [13], laboratory [14], and petrochemical [15]. The use of O_3 in wastewater treatment to disinfect, deodorize, decolorize, and oxidize is becoming essential, particularly when a high degree of treatment is required [6, 16]. It has high efficiency of pollutant removal, an absence of secondary contamination, and a short residence time for wastewater treatment. In comparison to other methods of wastewater treatment such as physical, chemical, biological process, electrochemical [17, 18], and advanced oxidation processes (AOPs) [18, 19], the O_3 process has advantages such as high oxidizing ability, nonselective simple reaction conditions, without the need for high temperature or pressure [20], etc.

Ozone has a high oxidation potential and produces the hydroxyl radical ($^\bullet$OH), a highly reactive oxidative species [21, 22]. While O_3 has high oxidizing power, its conversion to $^\bullet$OH is inefficient, and thus O_3 alone has a lower capacity for pollutant removal than $^\bullet$OH [23]. Consequently, in recent years, new O_3 hybrid processes based on AOPs and electrochemical processes have been developed, including catalytic, nanocatalyzed, photocatalytic, and sonolytic ozonation, O_3/UV, O_3/H_2O_2, O_3/Fenton, O_3/electrochemical, and $O_3/UV/H_2O_2$ [21, 24]. The addition of Fe^{2+} and H_2O_2 to O_3, referred to as the $O_3/Fe^{2+}/H_2O_2$ process [25], improves the decolorization/degradation/mineralization of organic/inorganic matter present in effluent and wastewater [26, 27]. The $O_3/Fe^{2+}/H_2O_2$ process produced additional $^\bullet$OH radicals via the reaction of O_3 with Fe^{2+}/H_2O_2, and the reactions are described elsewhere [28, 29].

Owing to the synergic effect, the combination of O_3 with Fe^{2+}/H_2O_2 reagent exhibits a high oxidation rate. The $O_3/Fe^{2+}/H_2O_2$ process produces excessive $^\bullet$OH via the reaction of O_3 and Fe^{2+}/H_2O_2, resulting in increased pollutant removal at a reduced treatment time. The $O_3/Fe^{2+}/H_2O_2$ has been used to treat water present in various types of wastewater, such as urban [30] and landfill leachate [31]. A comparison showed that O_3 and Fe^{2+}/H_2O_2 processes were both less effective than the $O_3/Fe^{2+}/H_2O_2$ combination [32].

Feng et al. examined the degradation of spent resin using the O_3–Fe^{2+}/H_2O_2 and Fe^{2+}/H_2O_2 processes and concluded that both processes run according to first-order kinetic equations, and the O_3–Fe^{2+}/H_2O_2 process had the maximum removal competence and cost savings [32]. Li et al. compared the Fe^{2+}/H_2O_2 and $O_3/Fe^{2+}/H_2O_2$ processes for amoxicillin degradation. The COD–65% removal rate was achieved in the O_3–Fe^{2+}/H_2O_2 process as opposed to the Fe^{2+}/H_2O_2 process. The synergistic effects worked in harmony with each other, thereby giving the $O_3/Fe^{2+}/H_2O_2$ process the edge [33]. Goi et al. evaluated individual processes such as Fe^{2+}/H_2O_2, O_3, and the combination of these treatment processes using leachate waste. They discovered that the coupled Fe^{2+}/H_2O_2 and O_3 processes eliminated the most COD (77 %) as compared to other approaches [34].

Substantial research has concentrated on the elimination of pollutants from simulated wastewater using O_3 and AOPs, with only a few studies using real industrial effluent and wastewater. Furthermore, prior research has concentrated on the efficiency of hybrid O_3-based AOPs in terms of % COD and color removal (%) but has not put an emphasis on pollutant elimination together with electrical energy per order (EE/O). It was crucial to establish the EE/O of O_3-based AOPs to determine the process's operating costs and feasibility. As a result, the current research concentrated on the determination of EE/O while removing color and COD from DIW utilizing the O_3 and Fe^{2+}/H_2O_2 coupled AOPs process.

The primary goal of this study is to assess the efficiency of coupled O_3 and Fe^{2+}/H_2O_2 based AOPs in terms of % COD and color removal, as well as the determination of EE/O in distillery industrial wastewater (DIW). This study investigated the influence of various operating conditions on the $O_3/Fe^{2+}/H_2O_2$ process such as Fe^{2+} (5–30 mM) and H_2O_2 concentration (20–140 mM), initial pH of wastewater (1–11), COD (800–4800 ppm), and O_3 inlet concentration (0.80–4 g/hr). The synergy between the combined O_3 and Fe^{2+}/H_2O_2 processes was investigated and recorded.

2. Materials and Methods

2.1. Wastewater Collection and Characterization. Distillery industrial wastewater was collected from distilleries in Erode, Tamil Nadu, India. The wastewater had the following characteristics: dark brown color, burn sugar odor, pH: 4.1–4.3, chemical oxygen demand (COD) of 80,000–90,000 mg/L, biochemical oxygen demand (BOD) of 7000–8000 mg/L, total dissolved solids (TDS) of 5550–5750 mg/L, and total suspended solids (TSS) of 15.44 g/L.

The chemicals used in the experiments were H_2O_2–50% (w/w), $FeSO_4.7H_2O$, NaOH, H_2SO_4, $Na_2S_2O_3$, KI, $K_2Cr_2O_7$, etc. The analytical reagent (AR) grade chemicals were purchased from Merck, India. They were used as received without any purification.

2.2. Experimental Setup. The experimental setup for the $O_3/Fe^{2+}/H_2O_2$ process is depicted in Figure 1. The O_3 was generated with the aid of an ozone generator (Ozonetek Limited, Chennai). The air was pumped at a rate of 20 liters per minute (LPM) and generated up to 4 g/hr of O_3. The generated O_3 is directed into the reactor, which has a capacity of 500 mL. The O_3 was purged via a diffuser at the reactor's bottom. The residual O_3 in the gas stream leaving the reactor was destroyed by the 2–5% KI solution. The DIW was adjusted to the necessary pH and COD concentrations and was loaded into the reactor along with the measured amount of Fe^{2+} and H_2O_2.

The concentration of O_3 was determined using the iodometric titration process. After the process, the samples were taken from the reactor and immediately added to $Na_2S_2O_3$ to stop the reaction. After centrifuging for 15 minutes at 15000 rpm, the supernatant was collected and analyzed for color using a UV/Vis–Spectrophotometer (TR300 Spectroquant®) and for COD using the principle of the closed reflux method (TR320, Spectroquant®).

2.3. Analysis

2.3.1. COD Removal (%). The COD removal efficiency was measured using the following equation:

$$\text{COD removal efficiency (\%)} = \left(\frac{\text{COD}_{Ini} - \text{COD}_{Fin}}{\text{COD}_{Ini}}\right)100, \tag{1}$$

where COD_{Ini} and COD_{Fin} are the chemical oxygen demand (mg/L) values of DIW before and after the treatment process.

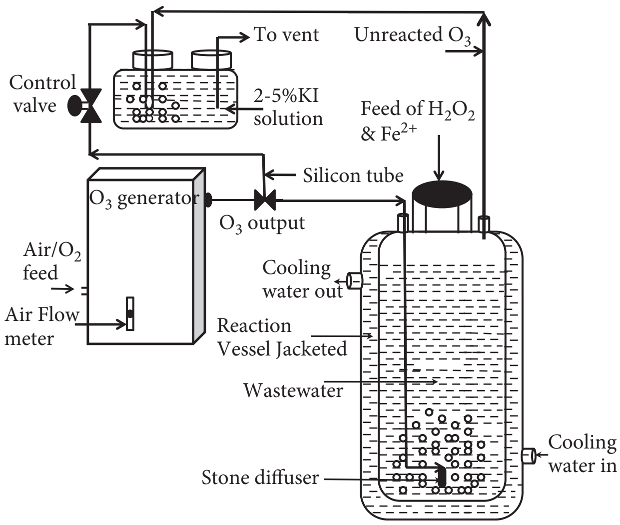

Figure 1: Experimental setup of the O_3/Fe2+/H_2O_2 process.

2.3.2. Color Removal (%). The color removal efficiency was calculated using the following equation:

$$\text{color removal (\%)} = \left(1 - \frac{Abs_{Fin}}{Abs_{Ini}}\right)100, \quad (2)$$

where Abs_{Ini} and Abs_{Fin} are the absorbances of the before and after treatment process of DIW.

2.3.3. Electrical Energy per Order (EE/O). Electrical energy per order has emerged as an additional and effective way to determine the suitability of wastewater treatment, and it must be economical for both individual and combined processes [35].

The equation proposed for the determination of EE/O for COD removal is as follows:

$$\frac{\mathrm{EE}}{\mathrm{O}}\left(\mathrm{kWh/m^3order^1}\right) = \frac{P * t * 1000}{V * 60 * \log\left(COD_{Ini}/COD_{Fin}\right)}, \quad (3)$$

$$\log\left(\frac{COD_{Ini}}{COD_{Fin}}\right) = kt, \quad (4)$$

where P is the power (kW) for O_3, t is the treatment time (min), V is the volume of the reactor (L), k is the pseudo-first-order rate constant ($\min^{-1}$) for the deterioration of the pollutant concentration.

Combining equations (3) and (4), EE/O becomes

$$\frac{\mathrm{EP}}{\mathrm{O}}\left(\mathrm{kWh/m^3order^1}\right) = \frac{38.4 * P}{V * k}. \quad (5)$$

2.3.4. Synergistic Effect (SE). The synergy effects (SE) of the $O_3/Fe^{2+}/H_2O_2$ process can be determined from the COD and/or color removal rate constants of the coupled and standalone processes using the following equation (6) [36]:

$$SE = \left(\frac{k_{O_3/Fe^{2+}/H_2O_2}}{k_{O_3} + k_{Fe^{2+}/H_2O_2}} - 1\right)100, \quad (6)$$

where, $k_{O_3/Fe^{2+}/H_2O_2}$, k_{O_3}, k_{Fe^{2+}/H_2O_2} are the rate constants of the $O_3/Fe^{2+}/H_2O_2$, O_3, and Fe^{2+}/H_2O_2 system, respectively.

A *SE* value ≥ 1 indicates that the coupled process surpasses the sum of the individuals, instead of $SE \leq 1$ means that the coupled process produces a negative effect in combining the individuals.

3. Results and Discussion

3.1. Process Comparisons. The individuals such as Fe^{2+}, H_2O_2, O_3, and hybrid processes such as Fe^{2+}/H_2O_2, O_3/Fe^{2+}, O_3/H_2O_2, and $O_3/Fe^{2+}/H_2O_2$ was carried out under the following operating conditions: COD—1600 mg/L, pH—6, Fe^{2+}—20 mM, H_2O_2—100 mM, and O_3 flow rate and concentration of 20 LPM and 4 g/hr, respectively. The efficiency of these processes was compared in terms of % COD and color reduction, accompanied by an estimate of EE/O for DIW, and the findings are depicted in Figures 2(a) and 2(b). As represented in Figure 2(a), the single Fe^{2+}, H_2O_2, and O_3 methods were ineffective at removing color and COD. The Fe^{2+}/H_2O_2 process removed % COD and color at a moderate rate. The % COD and color removed by combining O_3 with Fe^{2+} and H_2O_2 processes such as O_3/Fe^{2+}, O_3/H_2O_2, and $O_3/Fe^{2+}/H_2O_2$ were approximately 46.87%, 68.75%, and 96.87%, and 57.43%, 79.74%, and 100%, respectively. As predicted, the hybrid $O_3/Fe^{2+}/H_2O_2$ process is more efficient at removing COD and color than the O_3/Fe^{2+} and O_3/H_2O_2 treatment processes. The abovementioned results suggested that adding Fe^{2+} and H_2O_2 to the O_3 process significantly increases COD and color removal. Perhaps this is due to the influence of concurrent pathways capable of producing plenteous $^{\bullet}$OH radicals for COD and color removal from DIW [37, 38].

The suitability of the $O_3/Fe^{2+}/H_2O_2$ process for wastewater treatment is primarily determined by the EE/O, which relies on COD removal using equation (3). A minimum of 0.2 kWh/m^{3}order^1 of EE/O was needed to remove COD and color from DIW using the $O_3/Fe^{2+}/H_2O_2$ process. In comparison to the $O_3/Fe^{2+}/H_2O_2$ process, the other single and combined processes such as O_3 and O_3/Fe^{2+}, O_3/H_2O_2 required a high amount of EE/O to remove COD and color.

3.2. Various Operating Parameters. Experimental operating parameters including the Fe^{2+}, H_2O_2, COD, and O_3 inlet concentration, and initial wastewater pH [39, 40], and so on, are found to have a major effect on the efficacy of the combined $O_3/Fe^{2+}/H_2O_2$ process in terms of COD, color removal, and EE/O of DIW.

3.2.1. Effect of Fe^{2+}. The concentration of Fe^{2+} and H_2O_2 is an important operating parameter, affecting the efficiency of pollutant removal through the $O_3/Fe^{2+}/H_2O_2$ process and preventing the excessive use of Fe^{2+} and H_2O_2 [29, 41]. Figure 3 illustrates the significance of the amount of Fe^{2+} on the % COD reduction and EE/O in the $O_3/Fe^{2+}/H_2O_2$

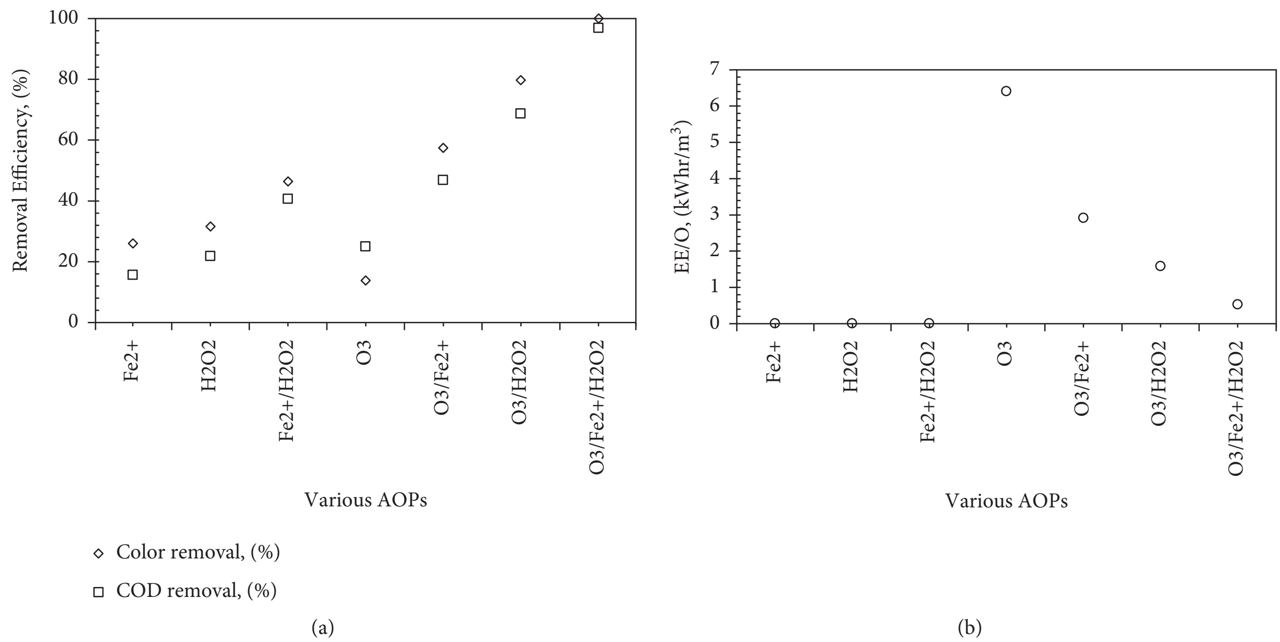

Figure 2: Comparison of various AOPs such as Fe^{2+}, H_2O_2, Fe^{2+}/H_2O_2, O_3, O_3/Fe^{2+}, O_3/H_2O_3, and $O_3/Fe^{2+}/H_2O_2$ on (a) % color and % COD removal and (b) EE/O (experimental conditions: COD–1600 mg/L; reaction time–4 h; O3 flow rate and production–20 LPM and 4 g/h; H_2O_2–100 mM; Fe^{2+}–20 mM; and pH–6).

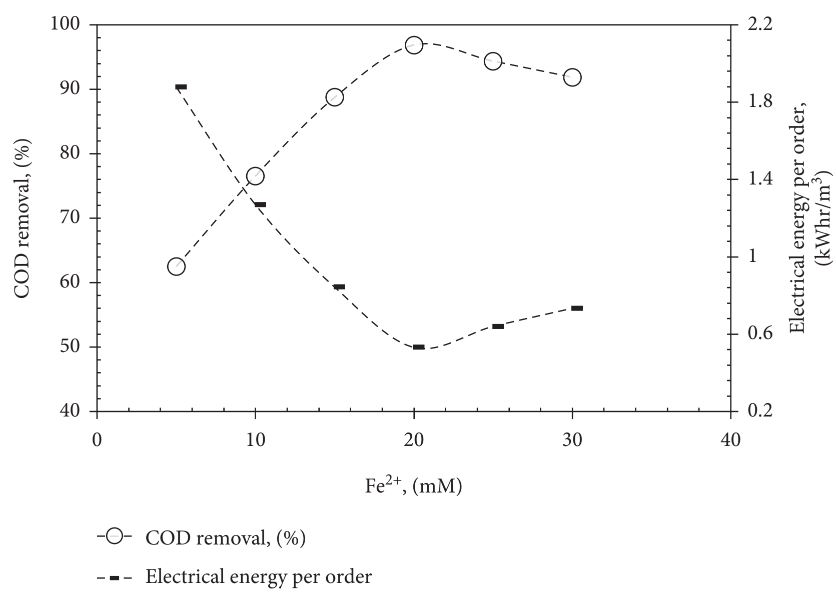

Figure 3: Effect of Fe^{2+} on % COD removal and EE/O in the $O_3/Fe^{2+}/H_2O_2$ process (experimental conditions: COD–1600 mg/L; reaction time–4 h; O_3 flow rate and production–20 LPM and 4 g/h; H_2O_2–100 mM; and pH–6).

method. As illustrated in Figure 3, around 96.87% COD removal and 0.5315 kWhr/m^3 EE/O were observed in the $O_3/Fe^{2+}/H_2O_2$ process at a Fe^{2+} concentration of 20 mM in comparison to other Fe^{2+} concentrations. The combination of Fe^{2+} and H_2O_2 increases the development of $^{\bullet}OH$ and thus the oxidation efficiency. This may be because the increased Fe^{2+} concentration facilitated the formation of $^{\bullet}OH$ radicals, thereby accelerating DIW degradation [41]. As Fe^{2+} concentrations exceeded 20 mM, the excess Fe^{2+} absorbed $^{\bullet}OH$ radicals, resulting in a small decline in % COD reduction and a rise in EE/O of DIW [40]. Thus, the optimal Fe^{2+} concentration was estimated to be 20 mM.

3.2.2. *Effect of H_2O_2.* The influence of varying the H_2O_2 concentration from 20 to 140 mM on the effectiveness of the $O_3/Fe^{2+}/H_2O_2$ process in terms of % COD reduction and EE/O for DIW was studied, with the findings shown in Figure 4. The H_2O_2 dose added had a major effect on the studied process. According to Figure 4, the % COD removal

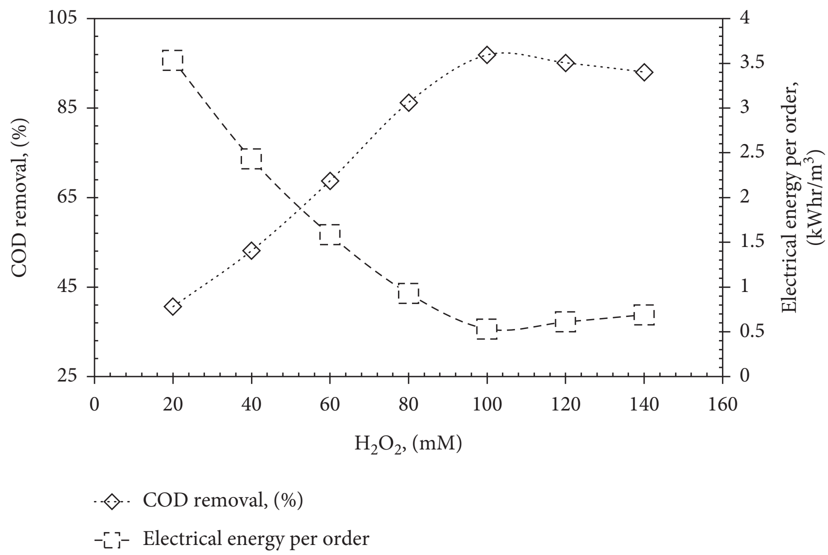

FIGURE 4: Effect of H_2O_2 on % COD removal and EE/O in the $O_3/Fe^{2+}/H_2O_2$ process (experimental conditions: COD–1600 mg/L; reaction time–4 h; O_3 flow rate and production–20 LPM and 4 g/h; Fe^{2+}–20 mM; and pH–6).

increases from 40.63 to 96.87%, and the EE/O decreases from 3.53 to 0.53 kWhr/m^3 as the initial H_2O_2 dose increases to a certain point, reaching a maximum at an initial H_2O_2 dose of about 100 mM, resulting in a substantial increase of process performance. At high concentrations of H_2O_2, it functions as an efficient $^{\bullet}OH$ scavenger [41, 42], depending on the pollutant in concern. The observation is consistent with the following empirical equation (7):

$$H_2O_2 + \bullet OH \longrightarrow HO_2^{\bullet} + H_2O \quad (7)$$

$$H_2O_2 \longrightarrow H_2O + \frac{1}{2}O_2. \quad (8)$$

While $HO_2^{\bullet}$ promotes radical chain reactions and is an effective oxidant in its own right, it has a much lower oxidation potential than $^{\bullet}OH$. Thus, when H_2O_2 concentrations are too high, the treatment efficiency is reduced, and its concentration must be tailored for each form of wastewater. The increase in H_2O_2 dosage increases the number of active sites on the surface, facilitating the decomposition of O_3 molecules into additional $^{\bullet}OH$. Thus, the $O_3/Fe^{2+}/H_2O_2$ process demonstrates an improvement in COD efficiency as the H_2O_2 dose is increased.

3.2.3. Effect of Wastewater pH. The pH of wastewater at its initial state is critical because it affects the reaction of organic/inorganic compounds with the $O_3/Fe^{2+}/H_2O_2$ system [25]. Figure 5 shows the results of an investigation into the impact of the initial values of pH on the % COD reduction and EE/O in the $O_3/Fe^{2+}/H_2O_2$ process for the DIW. As shown in Figure 5, with the $O_3/Fe^{2+}/H_2O_2$ process, the highest % COD removal efficiency and the lowest EE/O occurred at an initial pH of 6. As the pH increased up to 6, COD removal increased and EE/O decreased. Lower values of pH reveal the H^+ ion's scavenging effect on $^{\bullet}OH$, the creation of $H_3O_2^+$ by reaction between H_2O_2 and H^+, resulting in increased H_2O_2 stability and decreased % COD removal efficiency. The % COD reduction improved as the pH increased from 1 to 6, owing to the increased formation of $^{\bullet}OH$ (a more powerful oxidant than O_3) via the hydroxylation reaction with O_3.

Furthermore, raising the wastewater pH to 6 in the combined $O_3/Fe^{2+}/H_2O_2$ process resulted in coagulation, which increases contaminants through the complexation reaction induced by the transformation of Fe^{2+} and Fe^{3+} to $Fe(OH)_n$-type structures [25]. When the pH was greater than 6, COD removal decreased marginally with increasing pH. It is deduced that COD removal exists as a eupterotid that is energetic and easily reacts with the hydroxyl ion in acidic conditions, but it acts as a stable molecule in basic conditions, resulting in a decrease in % COD reduction and an increase in EE/O as the pH increases.

3.2.4. Effect of COD Concentration. The initial pollutant content is critical in the $O_3/Fe^{2+}/H_2O_2$ process wastewater treatment process [43, 44]. The increased initial COD concentration from 800 to 4800 mg/L, then decreased the % COD elimination from 100 to 45.83%, with a rise in the EE/O from 0.07 to 3 kWh/m^3order[1] for DIW using the $O_3/Fe^{2+}/H_2O_2$ process, as presented in Figure 6. Since, raising the initial COD content promoted the creation of intermediate products that compete with the O_3-consuming pollutant, thus decreasing % COD removal performance, and increase of EE/O for DIW [45].

3.2.5. Effect of O_3 Inlet Concentration. It is critical to select the optimal O_3 inlet concentration for pollutant removal from wastewater when using the $O_3/Fe^{2+}/H_2O_2$ process [40, 46]. The effect of O_3 inlet concentration on the % COD removal efficiency and EE/O with the $O_3/Fe^{2+}/H_2O_2$ process was explored by bubbling ozone into the DIW solution at different gas concentrations ranging from 0.8 to 4 g/h. As

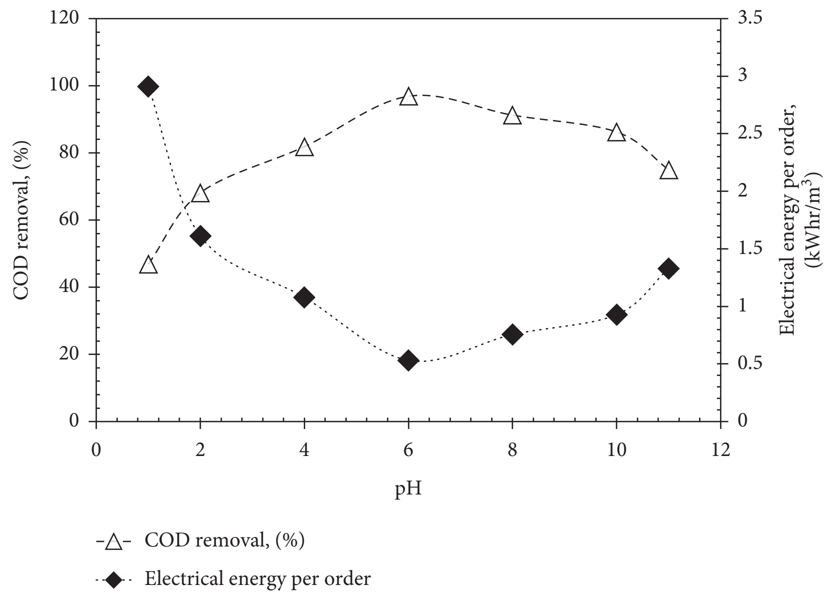

Figure 5: Effect of pH on % COD removal and EE/O in the $O_3/Fe^{2+}/H_2O_2$ process (experimental conditions: COD–1600 mg/L; reaction time–4 h; O_3 flow rate and production–20 LPM and 4 g/h; H^2O^2–100 mM; Fe^{2+}–20 mM; and pH–6).

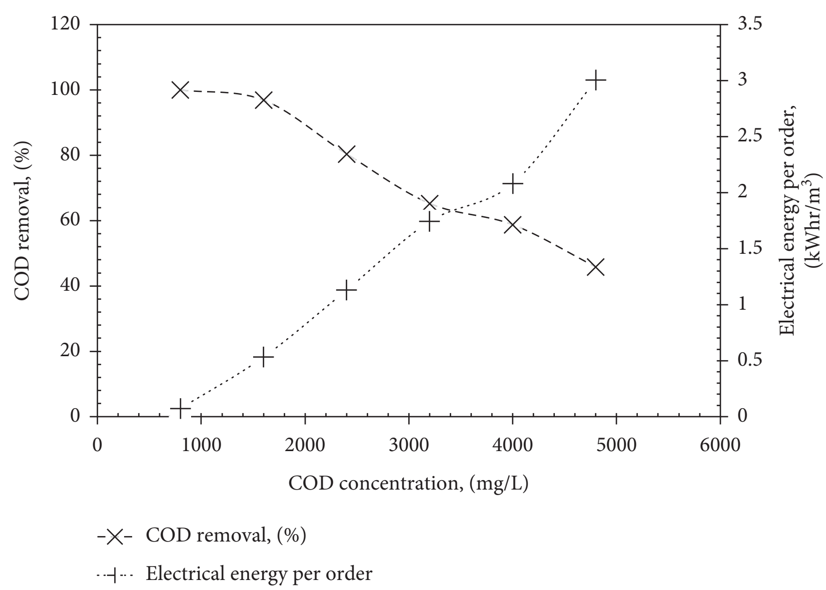

Figure 6: Effect of COD on % COD removal and EE/O in the $O_3/Fe^{2+}/H_2O_2$ process (experimental conditions: reaction time–4 h; O_3 flow rate and production–20 LPM and 4 g/h; H_2O_2–100 mM; Fe^{2+}–20 mM; and pH–6).

shown in Figure 7, the % COD reduction improved from 52.18 to 96.87% and the EE/O reduced from 2.49 to 0.535 kWh/m^3 order[1] as the $O_3/Fe^{2+}/H_2O_2$ system's O_3 concentration increased from 0.8 to 4 g/hr. This can be explained by the fact that the two key parameters, O_3 concentration and O_3–liquid mass transfer resistance, have a massive effect on the mass transfer rate of O_3 [40, 46]. With a higher O_3 concentration, the driving factor for O_3 mass transfer is increased, allowing the DIW solution to absorb more O_3. As a consequence, the excess O_3 in the solution interacted with radical initiators (Fe^{2+}, H_2O_2, $^{\bullet}OH$, etc.) to produce more $^{\bullet}OH$, which eventually improves COD and color removal efficiency while lowering EE/O. Zhao et al. [46] reported similar results for the removal of Ni-EDTA using O_3-based oxidation processes.

3.3. Synergy Effect. The O_3 and Fe^{2+}/H_2O_2 processes were carried out under the best experimental conditions to determine the synergy among each process for the removal of % color and % COD from DIW. The experimental findings confirmed a synergy index among O_3 and Fe^{2+}/H_2O_2 processes for COD removal of DIW. Thus, as opposed to other processes (O_3 and Fe^{2+}/H_2O_2), the $O_3/Fe^{2+}/H_2O_2$ process greatly improves COD and color removal. Equation (6) was used to determine the synergy index of the O_3 and Fe^{2+}/

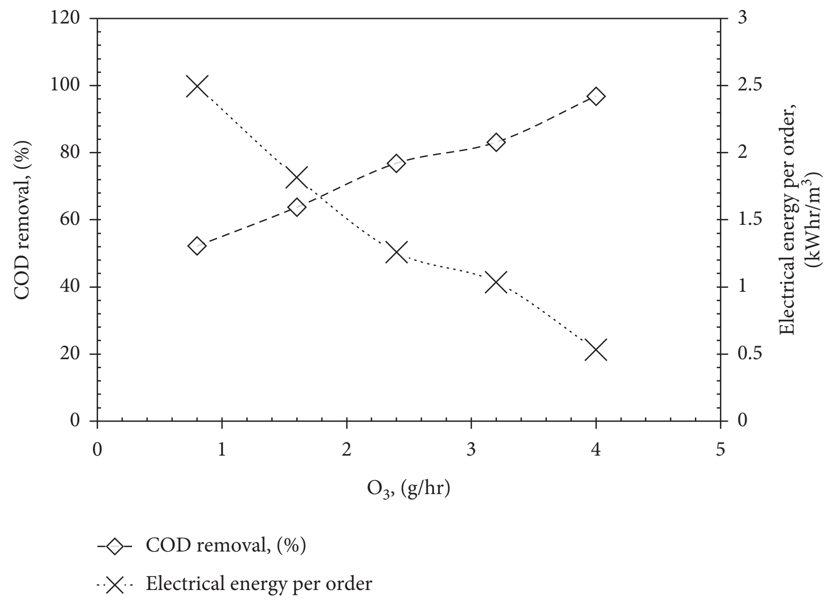

Figure 7: Effect of O_3 inlet concentration on % COD removal and EE/O in the $O_3/Fe^{2+}/H_2O_2$ process (experimental conditions: COD–1600 mg/L; reaction time–4 h; O_3 flow rate–20 LPM; H2O2–100 mM; Fe^{2+}–20 mM; and pH–6).

H_2O_2 processes [36]. The synergy index was 21.50%, suggesting that the effectiveness in terms % color and % COD removal was sufficiently greater for the combined process than for the individual O_3 and Fe^{2+}/H_2O_2 processes. The reported synergy index is due to the fact that combining O_3 and Fe^{2+}/H_2O_2 provides a larger volume of •OH, which enhances the rate of % color and COD removal from DIW. Thus, the coupled O_3 and Fe^{2+}/H_2O_2 processes offer an alternative and novel approach for industrial wastewater treatment.

4. Conclusion

The developed $O_3/Fe^{2+}/H_2O_2$ process is compared to O_3, Fe^{2+}, H_2O_2, O_3/Fe^{2+}, and O_3/H_2O_2, Fe^{2+}/H_2O_2 processes, and the outcomes indicate that the $O_3/Fe^{2+}/H_2O_2$ process is a promising method for achieving high % color and % COD removal efficiencies while consuming a minimal amount of EE/O from DIW. The findings suggest that to ensure an effective treatment process, operating parameters such as Fe^{2+}, H_2O_2, COD, and O_3 inlet concentration, and initial wastewater pH should be considered. A synergy effect was calculated to exist between the O_3 and Fe^{2+}/H_2O_2 processes at 21.50 percent. The $O_3/Fe^{2+}/H_2O_2$ method was found to be capable of eliminating pollutants from a range of industrial effluents and wastewater.

Data Availability

The datasets analyzed during the study are available from the corresponding author on request.

Conflicts of Interest

The authors acknowledge that they have no conflicts of interest.

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