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Research

Combined ozone, photo, and electrocoagulation technologies-An innovative technique for treatment of distillery industrial wastewater

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ABSTRACT

Electrocoagulation with ozone and photo is a hybrid electrochemical and AOP techniques used to treat wastewater and industrial effluent. In the present study, the effects of combining O_3, UV, and EC processes on color removal as well as the related COD removal and EEC from the treatment of DIW were assessed. The results showed that, compared to other hybrid processes like O_3/EC, UV/EC, UV/O_3, and single processes of EC, O_3, and UV, the combined process of O_3/UV/EC is significantly more effective for treating DIW in terms of COD removal (98.99%) and complete color removal with a required EEC of 9.7 kWhr m^{-3}. Number of process variables, including reaction time (1 to 5 hr), O_3 (0.8 to 4 g L-1), UV (8 to 32 W), current density (0.08 to 0.23 A dm^{-2}), pH (1 to 11), COD (1000 to 6000 mg L^{-1}), inter-electrode distance (0.75 to 3.75 cm), and combination of electrodes were studied to analyze how they affected. The optimum operating circumstances underwent testing and shown outstanding effectiveness in eliminating the COD and determination of EEC from DIW. Overall, it was clear from the results that combined treatment approaches could be more effective in removing the pollutant successfully from DIW.

Keywords: COD reduction, decolorization, distillery industrial wastewater, electrical energy usage, ozone-photo-electrocoagulation, space time yield

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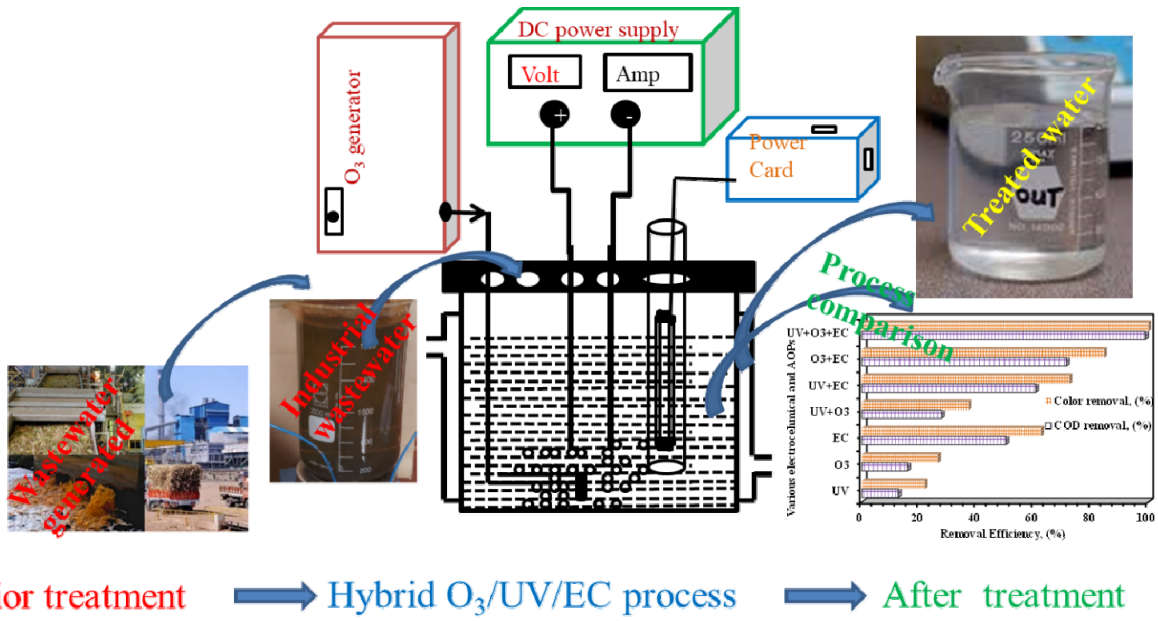
Graphical Abstract



1. Introduction

The treatment of wastewater produced by industries is one of the most serious environmental issues, and it poses a huge challenge to environmentalists [1, 2]. Distillery wastewater treatment presents a significant challenge for environmentalists and the cane molasses is the only raw material used entirely by distilleries for fermentation. The wastewater from these industries contains highly colored, chemical oxygen demand (COD), biochemical oxygen demand (BOD), total dissolved solids (TDS) and other organic matter [2, 3]. Unless the wastewater is properly treated and then disposed of, it may significantly stress waterways, harming aquatic life on a large scale [3]. It can also lower soil alkalinity, which has an impact on groundwater quality [4]. The necessity for introducing new methods to handle this wastewater effectively and affordably has been prompted by the continuous production of wastewater from distilleries and the stringent regulations controlling its disposal.

Distillery industrial wastewater decolorizaion and removing COD, BOD, total organic carbon (TOC), and other contaminants are accomplished through the use of physical, chemical, and biological processes [5–8]. The main disadvantages of these treatment systems are: inability to mineralize pollutants, phase transfer of contaminants from one medium to another, significant amounts of sludge generated, lower level pollutant removal efficiency, and higher operational costs. There is an urgent need to develop more efficient, cost-effective treatment technologies that use the least amount of chemicals and energy while removing the maximum of pollutants. Electrochemical and advanced oxidation processes (AOPs), such as electro-oxidation [9], electrocoagulation (EC)[10–13], electro-disinfection [11], electro-dialysis [14], photo-catalytic degradation [15–19], ozonation [20, 21], ozone-catalytic oxidation [22, 23], sono-catalytic [24, 25], solar-powered electrocoagulation [26], etc., because of their simplicity of operation, convenience of setup, high oxidation ability, and complete pollutant removal have garnered a lot of attention for the removal of contaminants from wastewater and industrial effluent. Recently, there has been a significant uptick in the amount of focus placed by researchers on the integration of electrocoagulation techniques into hybrid processes [5, 27–31].

According to Biliska et al. [32] electrocoagulation and O_3 techniques, which worked well in salty conditions, could be combined to produce extremely good color removal (95%) for both aqueous solution and actual industrial effluent containing Reactive Black 5 in a short treatment time, this was possible because of combined the electrocoagulation and O_3 techniques. In order to improve greywater treatment, a system that combined ozonation and electrocoagulation technologies was the study's focus by Barzegar et al. [33]. The study examined how ozone dosage, current density, pH, and contact time all affected the results. They obtained 85% and 70% of COD and TOC removals, respectively, after 60 minutes using iron electrodes with operating variables. The sequential EC and UV treatment processes were demonstrated by Jallouli et al. [34] to be efficient at reducing COD in tannery effluent. According to their findings, the combined process reduced COD by 94.10%, as opposed to the individual EC and UV treatments' respective reductions of 85.7% and 55.9%. In order to remove COD, total chromium (Cr(III) and Cr(VI)), and sulfide from tannery effluent was accomplished by Moradi and Moussavi [35] using the electrocoagulation technique in conjunction with a UVC/VUV-photoreactor. The elimination efficiency of these contaminants was found to be 99.52%, 100%, 100%, and 98.27% for acidic and neutral pH solutions, respectively. When UV irradiation and

Table. 1. Physiochemical properties of raw and diluted distillery industrial wastewater

Parameters	Raw	Diluted	Unit
pH	4.1 - 4.3	5.5	
COD	80,000 - 90,000	6000	mg L^{-1}
BOD	7,000 - 8,000	850	mg L^{-1}
TSS	15.44	5.75	g L^{-1}
TDS	5550-5750	600	mg L^{-1}
Colour	dark brown		-
Odour	burnt sugar		--

electrocoagulation (UV-EC) were used at low current densities, Cotillas et al. [36] stated that a synergistic impact was found for the decrease of turbidity and disinfection rate. They also discovered that when UV light is utilized to encourage production.

Aziz et al. [37], evaluated the O_3, EC, O_3-EC, peroxi-EC, UV-EC, and peroxi-UV-EC processes in terms of colour and COD reduction from distillery industrial effluent. They confirmed that the O_3-EC method accomplished complete color and COD removal with the required power usage. Asaithambi et al. [38], treated real distillery industrial effluent with the O_3, EC, and O_3/EC processes and compared the % color and COD reductions. Moreover, the hybrid electrocoagulation method achieved complete color removal and 95% COD elimination.

Our thorough review of the available literature revealed that hybrid techniques based on electrochemical and AOPs were successful in removing the pollutant and toxins from synthetic wastewater [39–41]. The AOPs and hybrid electrochemical processes have been applied in very few studies that have concentrated on actual industrial effluent and wastewater [42, 43]. In addition to eliminating pollutants from industrial effluent and wastewater, the electrical energy consumption (EEC) of the hybrid process is critical from an economic standpoint. The drawbacks of traditional treatment methods must also be addressed by environmental engineers via the development of simple, effective, and affordable hybrid techniques.

According to the authors' knowledge, no prior study has been done on the use of O_3 and UV in conjunction with an EC process to figure out the electrical energy consumption (EEC) connected with the removal of color and COD from DIW. This investigation's goal was to establish and develop innovative treatment methods that would allow for the measurement of EEC and the removal of color and COD from DIW. The primary goal of this study is to assess the color and COD reduction using O_3, UV, EC, O_3/UV, O_3/EC, EC/UV and O_3/UV/EC processes as well as to calculate the EEC from DIW and choose the best one. It was examined how several process variables, including treatment duration, pH, COD and O_3 concentration, electrode combination, inter-electrode spacing, and UV power, affected the COD removal and EEC of DIW treated with an O_3/US/EC process. Additionally, for the system of O_3/UV/EC, the space-time yield was evaluated and reported.

2. Material and Methods

2.1. Wastewater Collection

Experiments made use of distillery industrial wastewater (DIW), which was collected from distilleries located in Erode, Tamil Nadu, India. To avoid any degradation in wastewater quality, the effluent was maintained in a cold room (+4℃). The raw and diluted wastewater was analyzed for various physicochemical properties of water quality parameters and the results are given in Table 1. Chemicals like H_2SO_4, NaOH, $K_2Cr_2O_7$, $Na_2S_2O_3$, $(NH_4)_2Fe(SO_4)_2$, etc. were utilized in the investigation. The analytical reagent (AR) grade chemicals were provided by Merck, India, and were utilized straight from the container without further purification.

2.2. Process Hybrid O_3+UV+EC Experimental Setup

2.2.1. Electrocoagulation (EC)

Fig. 1 illustrates the investigative setup of configuration for the combined Ozone+photo+electrocoagulation (O_3/UV/EC) for the treatment of DIW. In the integrated process, the O_3, UV, and EC process units were used. An acrylic sheet-based batch electrochemical reactor with a 2.5 L capacity was used to conduct the EC (Fig. 1). A 2.0 L DIW operating capacity was used for each experiment. The DIW had a COD range from 1,000 to 6,000 mg L^{-1}, and the solution was made by using dilution factor using distilled water from the raw DIW. The Fe/Fe (Iron/Iron), Fe/Al (Iron/Aluminum), Al/Fe, and Al/Al electrode materials were used for the anode and cathode. Both electrodes had the same size, 0.1 cm of thickness, and a 10 cm by 15 cm effective electrode surface area (width x height). A parallel connection between the anodes and cathodes, and a direct current (DC) power source was used (APLAB Ltd; Model L1606). The electrodes used in the procedures were manually cleaned with a solution of 35% HCl, rinsed with distilled water, and dried before each experiment. With the use of a pH meter (Elico: Model LI120) and a solution of NaOH and H_2SO_4, the pH of DIW was determined. After ensuring that the conditions of the experiment were as expected, a steady direct current (DC) was passed between the electrodes with the help of DC power supply.

2.2.2. Ozone+electrocoagulation (O_3/EC)

An O_3 generator (Exclusive Heritgae (M) SDN BHD) produced ozone with a 4 g hr^{-1} capacity, and at a steady flow rate of 20 L min^{-1}, it was continually purged to the electrocoagulation reactor's bottom (Fig.1). Iodometric titration was used to calculate the concentration of O_3. A gas-absorbing bottle solution containing 2% KI has been used to store the extra O_3.

2.2.3. Ozone+photo+electrocoagulation (O_3/UV/EC)

The reaction contents were further subjected to UV irradiation, but the operating circumstances for the experiment were then

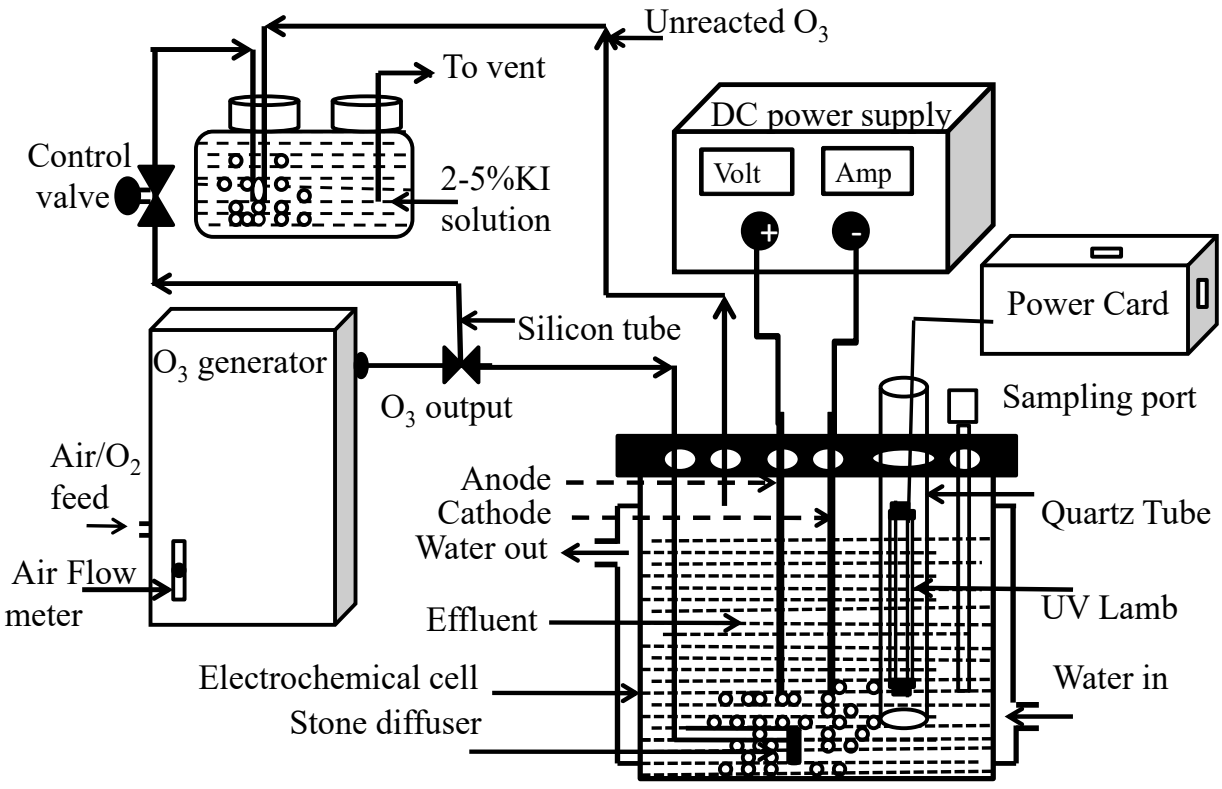


Fig. 1. Hybrid O_3/UV/EC process experimental setup.

similar to those for the O_3/EC process. A low-pressure mercury lamp producing mostly at 254 nm with an output of 8–32 Watts was employed as the UV light source. The O_3/EC method is also referred to as hybrid O_3/UV/EC since the UV lamp was added to it (Fig.1). Using a multimeter, the cell voltage and current were measured during the EC, UV/EC, O_3/EC, and O_3/UV/EC processes. Periodically during the operation, the samples were taken from the reactor and transferred to an Erlenmeyer flask containing a $Na_2S_2O_3$ solution, thereby put an end to the reaction. The liquid supernatant is immediately evaluated for COD (Spectroquant ® TR320) and color (Spectroquant Pharo ® 300) after the samples were centrifuged at 15,000 rpm for 10 min to separate the liquid from the solid components.

2.3. Analysis

2.3.1. Efficiency in removing color and COD, (%)

The color and COD reduction efficiencies were determined using an Eq. (1) and (2) below.

The color was measured using a UV/Vis spectrophotometer (Spectroquant Pharo ®300) at the wavelength that corresponded to the maximum absorbance (λ_{max} -300 nm).

$$\text{Removal of color, (\%)} = \left(1 - \frac{Abs_t}{Abs_i}\right) * 100 \quad (1)$$

where Abs_i and Abs_t represent the DIW's absorbances at the relevant wavelengths (λ_{max}) before and after the treatment process, respectively.

$$\text{Removal of COD, (\%)} = \left(1 - \frac{COD_t}{COD_i}\right) * 100 \quad (2)$$

where COD_i and COD_t are the COD (mg/L) are before and after the treatment of DIW, respectively.

2.3.2. Electrical energy consumption (EEC)

When comparing the UV, O_3, EC, O_3/UV, EC/UV, O_3/EC, and O_3/UV/EC processes to DIW, the removal efficiency of color and COD as well as EEC is assessed. The integrated system's treatment cost was principally driven by the EEC-kWhm^{-3} of UV, O_3, and EC [37]. To calculate the EEC, the following Eq (3-5) were used.

$$EEC_{O3/UV/EC} = EEC_{O3} + +EEC_{UV} + EEC_{EC} \quad (3)$$

where $EEC_{UV/03/EC}$, EEC_{UV}, EEC_{03} and EEC_{EC} are the electrical energy consumption for the O_3/UV/EC, UV, O_3 and EC processes, respectively.

2.3.3. Electrocoagulation (EC)

$$EEC_{EC} = \frac{UIt}{V_R}, (kWhr\ m^{-3}) \quad (4)$$

In this case, the variables *U*-cell voltage (V), *I*-current (A), *t*-reaction time (h), and V_R - DIW volume are employed.

2.3.4. Photo (UV) and ozonation (O_3)

$$EEC_{UV} + EEC_{O3} = \frac{P_{et}t1000}{V_R 60\, log(\frac{COD_t}{COD_i})}, (kWhr\, m^{-3}) \quad [44] \qquad (5)$$

where rated power in kW for P_{et} (0.008, 0.016, and 0.032 kW for photo (UV) and 0.100 kW for ozone (O_3)) and V_R - DIW volume are employed.

2.3.5. Space time yield (YSTY)

The space-time yield ($kgm^{-3}h^{-1}$) can be expressed in terms of the mass of a product (m_p) generated per unit of reactor volume (m^3) and time (h). The Eq. (6) below can be used to calculate Y_{STY}.

$$Y_{STY} = \frac{m_p}{vt}, (kgm^{-3}h^{-1}) \qquad (6)$$

where v is the volume of reactor and t is the time.

3. Results and Discussion

3.1. Electrochemical and AOPs Comparisons

In this part, to assess the rates of color and COD elimination with the necessary EEC, tests were carried out while employing the individual O_3, UV, and EC processes as well as various combinations of O_3, UV, and EC processes, such as O_3/UV, O_3/EC, UV/EC, and O_3/UV/EC for the treatment of DIW. The results are shown in Fig. 2 (a) and (b). According to the figures, electrochemical and advanced oxidation methods are coupled to remove color and COD from DIW while determining EEC. The following processes are given in increasing order: UV alone < O_3 only < UV/O_3 only < EC only < UV/EC < O_3/EC < UV/O_3/EC. Fig. 2 (a) showed that, the color and COD removal efficiency for the UV/O_3/EC, O_3/EC, UV/EC, and EC alone procedures were much higher than for the UV, O_3 solo, and UV/O_3 processes. The aforementioned results demonstrated that a considerable increase in color and COD removal percentages occurred as a consequence of the addition of O_3/EC to the UV process. It could be as a result of parallel pathways being formed to successfully produce enough $^{\bullet}OH$ radicals from the UV, O_3, and EC processes to effectively remove color and COD from DIW [31, 45–49]. Hernández-Ortega et al. [50] observed similar results for COD removal from mixed industrial wastewater by combineds electrocoagulation–ozone process.

Electrical energy consumption (EEC) is related to the economic viability of using UV and O_3 with the EC process. The Eq. (3-5) was used to compute it, and the results are presented in Fig. 2b, EEC of 9.70 kWhr m^{-3} was needed to complete removal color and the COD removal of 98.99% from DIW using a hybrid UV/O_3/EC process. In comparison to the O_3/UV/EC process for removing color and COD, various combinations and single processes like UV/EC, O_3/EC, UV/EC and EC, O_3, UV method needed considerable energy usage. As a result, wastewater and industrial effluents can be treated utilizing this hybrid (UV/O_3/EC) approach that is based on electrochemical and AOPs.

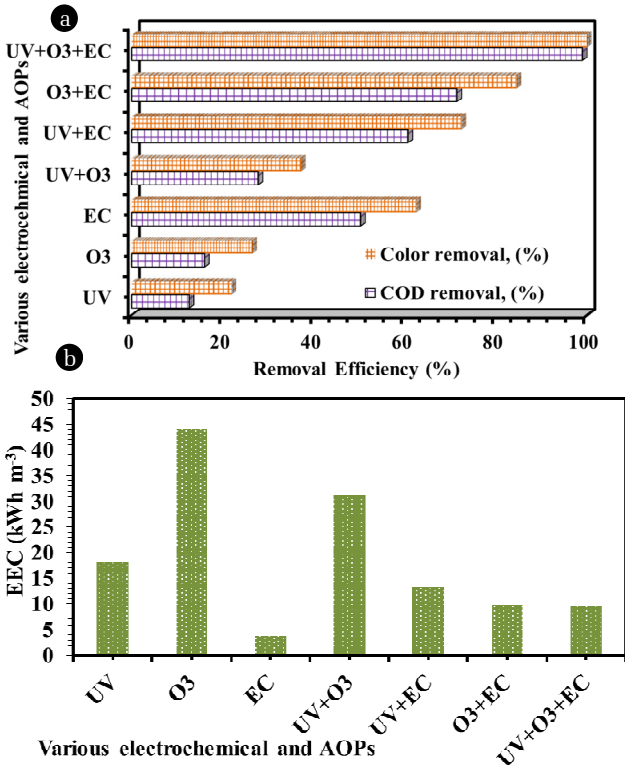


Fig. 2. Comparison of EC only, O_3 only, UV only, O_3/EC, O_3/UV, UV/EC, and O_3/UV//EC process on the (a) color, (%) and COD removal, (%) and (b) EEC from DIW (Conditions: current density = 0.20 A dm^{-2}, pH = 7, COD concentration = 3500 mg L^{-1}, distance between electrodes = 0.75 cm, electrode combination = Fe/Fe, UV power = 32 W, reaction time = 4 h, and O_3 concentration and flow rate = 4 g L^{-1} and 20 L min^{-1}).

3.2. Process Optimization for O_3/UV/EC

The effects of treatment time, current density, COD and O_3 concentration, pH, UV, inter-electrode distance, electrode combination were examined and addressed in depth below in order to enhance the O_3/UV/EC process performance.

3.2.1. Operational parameter studies

3.2.1.1. Treatment time

When it comes to treating industrial wastewater utilizing a hybrid technique, one of the most important factors to consider is treatment time [31, 51]. The results of the O_3/UV/EC process with the following parameters: pH = 7, COD concentration = 3500 mg L^{-1}, inter-electrode spacing = 0.75 cm, electrode combination = Fe/Fe, UV power = 32 W, and O_3 concentration and flow rate = 4 g L^{-1} and 20 L min^{-1} are shown in Fig. 3(a). According to Fig, as treatment time has risen, the percentage color removal, COD removal and EEC was increased from 65 to 100%, 55 to 99.66% and 4.84 to 10.7 kWhr m^{-3}, respectively. These findings are consistent with those reported by Garca-Morales et al. [52]. The amount of color and COD eliminated is directly related to the amount of ions produced by the electrodes. An increasing number of electrode ions

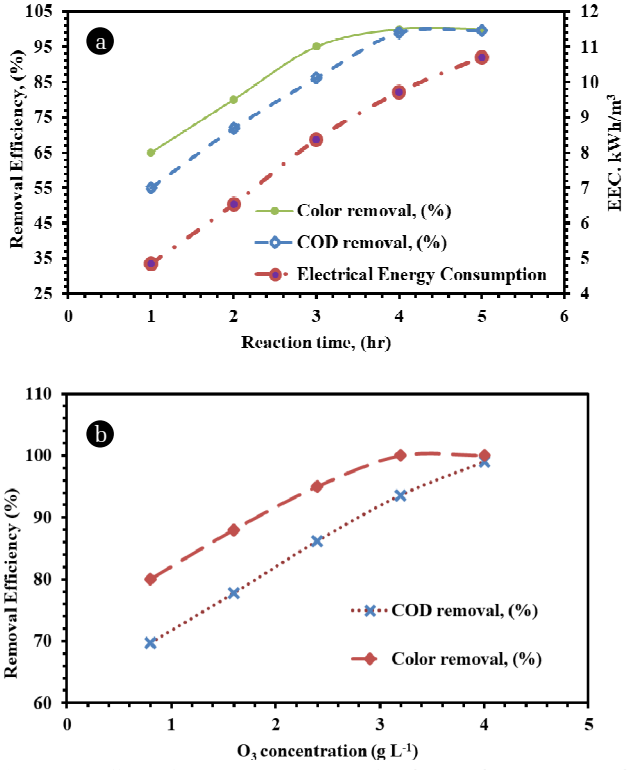


Fig. 3. (a) Effect of treatment time on % color, and COD removal, and EEC from DIW by hybrid O_3/UV//EC process (Conditions: distance between electrodes = 0.75 cm, current density = 0.20 A dm^{-2}, electrode combination = Fe/Fe, pH = 7, COD concentration = 3500 mg L^{-1}, UV power = 32 W, O_3 concentration and flow rate = 4 g L-1 and 20 L min^{-1}). (b). Effect of O_3 concentration on the (%) color, and (%) COD removal from DIW by hybrid O_3/UV//EC process (Conditions: current density = 0.20 A dm^{-2}, pH = 7, COD concentration = 3500 mg L^{-1}, distance between electrodes = 0.75 cm, electrode combination = Fe/Fe, UV power = 32 W, reaction time = 4 h and O_3 flow rate = 20 L min^{-1}).

and associated oxidizing species are created as the treatment period increases [53]. As a result, as treatment duration is increased, the efficiency of eliminating color and COD increases. When EEC is increased by cell voltage, more ˙OH radicals are created, which improves color and COD elimination [31].

3.2.1.2. O_3 concentration

The percentage color and COD removal efficiency and requirement of EEC utilizing the O_3/UV/EC method for DIW were examined in order to determine the effects of varying the concentration of O_3 and the findings are shown in Fig. 3(b). As the O_3 concentration increased from 0.8 to 4 g L^{-1}, the removal of color increased from 80 to 100% and the elimination of COD increased from 69.75 to 98.99%, as shown in Fig. 3(b). The results show that the amount of color and COD removal is affected by the O_3 dissolution, which also impacts the transfer of O_3 to the reaction solution and the amount of ˙OH production [33]. As increasing in the concentration of O_3 makes it possible for there to be a better mass transfer of O_3, and as a result, a higher efficiency. As O_3 concentration in air bubbles increases, the driving force for O_3 transport to the solution also rises. This increase in O_3 concentration leads to a greater rate of color and COD elimination from the DIW. But when the concentration of O_3 increases to its maximum in the liquid phase, the pace of chemical reaction progressively controls the process. In order to reduce both the concentration of exhaust O_3 and the use of electrical energy, it is crucial to regulate the dose of O_3.

3.2.1.3. UV power

During the O_3 plus EC process, an additional UV exposure was made. During a 4 hr of O_3/UV/EC method for DIW, as shown in Fig. 4(a), a range of UV power from 8 to 32 W was investigated using UV-C lamps. In addition, the UV power was raised from 8 to 32 W, increasing the COD removal efficiency and EEC from

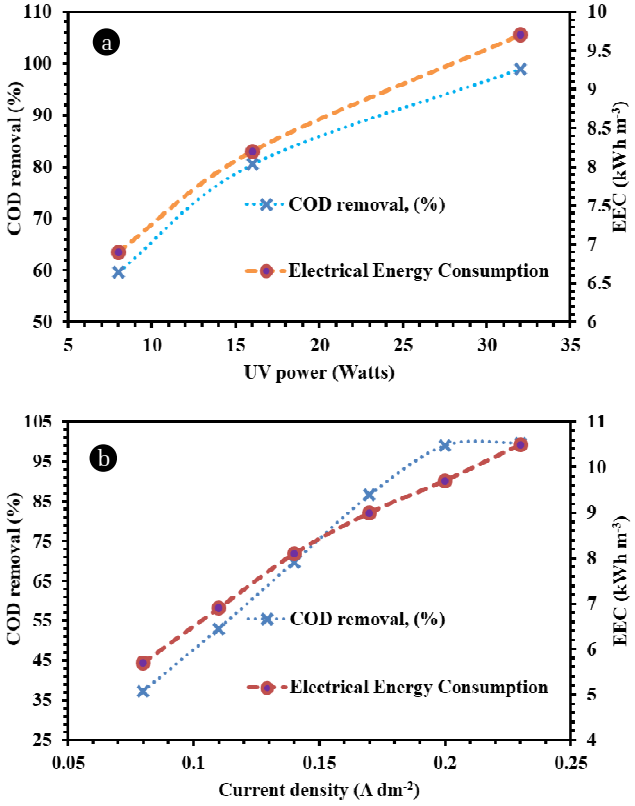


Fig. 4. (a). Effect of UV power on % COD removal and EEC from DIW by hybrid O_3/UV//EC process (Conditions: pH = 7, electrode combination = Fe/Fe, current density = 0.20 A dm^{-2}, COD concentration = 3500 mg L^{-1}, distance between electrodes = 0.75 cm, reaction time = 4 h and O_3 concentration and flow rate = 4 g L^{-1} and 20 L min^{-1}). (b). Effect of current density on % COD removal and EEC from DIW by hybrid O_3/UV//EC process (Conditions: distance between electrodes = 0.75 cm, pH = 7, COD concentration = 3500 mg L^{-1}, electrode combination = Fe/Fe, UV power = 32 W, reaction time = 4 h, and O_3 concentration and flow rate = 4 g L^{-1} and 20 L min^{-1}).

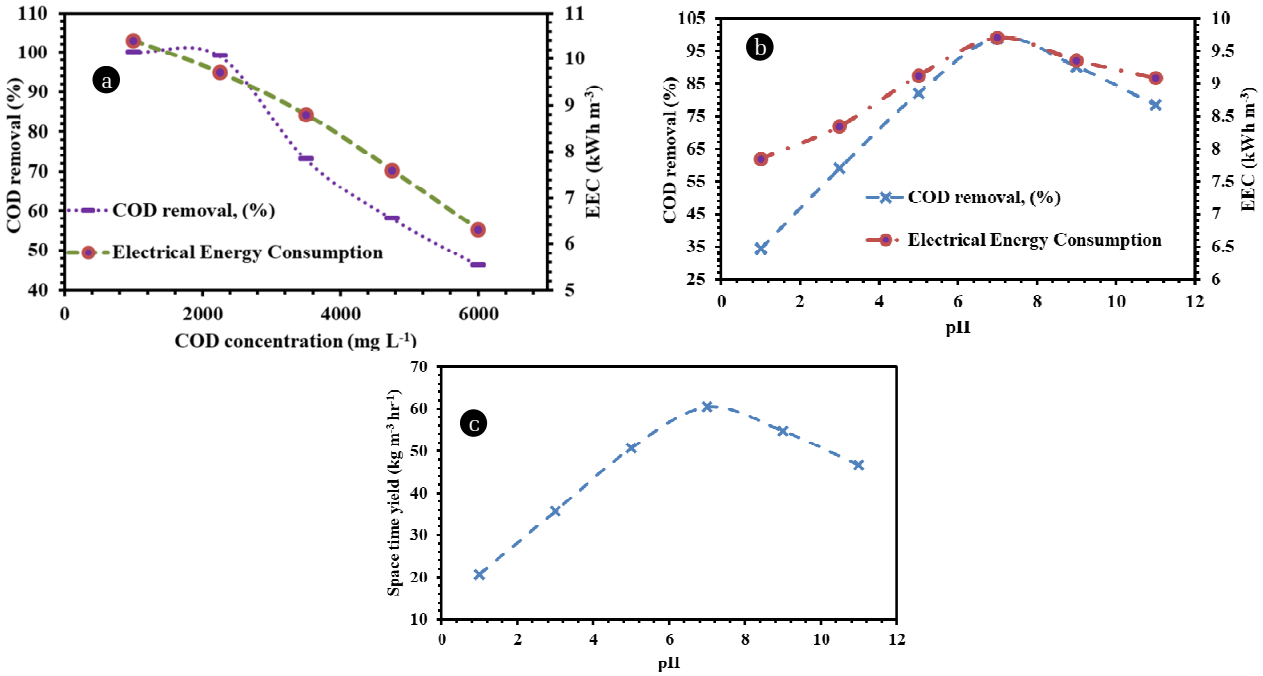


Fig. 5. (a) Effect of COD concentration on % COD removal and EEC from DIW by hybrid O_3/UV//EC process (Conditions: distance between electrodes = 0.75 cm, current density = 0.20 A dm^{-2}, pH = 7, electrode combination = Fe/Fe, UV power = 32 W, reaction time = 4 h, and O_3 concentration and flow rate = 4 g L^{-1} and 20 L $amin^{-1}$). (b). Effect of pH on % COD removal and EEC, and (c) space time yield from DIW by hybrid O_3/UV//EC process (Conditions: current density = 0.20 A dm^{-2}, COD concentration = 3500 mg L^{-1}, distance between electrodes = 0.75 cm, electrode combination = Fe/Fe, UV power = 32 W, O_3 concentration and flow rate = 4 g L^{-1}, and reaction time = 4 h and 20 L min^{-1}).

59.68 to 98.99% and 6.50 to 9.70 kWh m^{-3}, respectively. These findings are consistent with those reported by Keramati et al. [54]. Production of more ˙OH radicals is influenced by changes in lamp power and light source irradiation intensity per unit area. The O_3/EC was positively impacted by the increasing accessibility of photoactive areas. This improvement was attributable to an increase in the production of ˙OH radicals via photo-decomposition and photo-reduction reactions [37].

$$Fe(OH)^{2+} + hv \rightarrow Fe^{2+} + \bullet OH \quad (7)$$

$$R(CO_2) - Fe^{3+} + hv \rightarrow R(\bullet CO_2) + Fe^{2+} \rightarrow \bullet R + CO_2 \quad (8)$$

Due to this, the O_3/UV/EC procedure for DIW is more effective in removing color and COD and using electrical energy.

3.2.1.4. Current density

The current density is a crucial factor in an integrated EC process because it regulates floc development, coagulant production rate, and bubble creation [55, 56]. To ascertain the ideal current density, studies were conducted at various current densities ranging from 0.08 to 0.23 A dm^{-2}. Fig. 4(b) shows the COD removal efficiency and EEC progression during the O_3/UV/EC process throughout the course of treatment. The COD removal efficiencies increased from 37.17 to 98.99% with an increase in current density from 0.08 to 0.20 A dm^{-2}, although were slightly reduced from 0.20 to 0.23 A dm^{-2} from 98.99 to 97%, respectively. Additionally, the EEC increased from 5.7 to 10.50 kWhr m^{-3} with current densities rising from 0.08 to 0.23 A dm^{-2}. For all subsequent studies, the optimum current density was set at 0.20 A dm^{-2}, which produced the highest COD removal rates and a low EEC value of 9.70 kWhr m^{-3}. These findings agree with those published by Yazici Guvenc et al. [57]. According to Faraday's Law, current density is directly proportional to the coagulant dosage. Therefore, the dissolution rate of iron electrode increases with current density and consequently more ferrous hydroxyl radicals are produced [58]. The heat production, unwanted reactions, excess oxygen evaluation, and cell voltage all increase with increasing current density; cell voltage is directly proportional to power consumption [59]. Due to the high power consumption and operating expenses, the current density should be kept at its optimum level.

3.2.1.5. COD concentration

At constant current densities, the impact of an initial COD concentration in the range of 1000 to 6000 mg L^{-1} on elimination COD % and EEC was investigated. As can be seen in Fig.5 (a), increasing the starting COD concentrations from 1000 to 6000 mg L^{-1} for a 4 hr treatment duration resulted in a drop in the elimination of COD % from 100 to 46.24% and EEC from 10.4 to 6.3 kWhr m^{-3}, respectively. The amount of COD was removed 1000 and 3225 mg L^{-1} for the initial concentration of 1000 and 6000 mg L^{-1}, respectively. When the initial COD concentration of solutions increased, the removal COD dropped, and the quantities of pollutant concentration eliminated rose. Faraday's law states that a

constant quantity of Fe^{2+} is passed to the solution for increasing initial effluent concentration at a constant galvanostatic value of current density and duration. With the same current density and treatment duration, the rate at which $^{\bullet}OH$ radicals were released and the quantity of flocs that formed with $Fe(OH)_3$ were almost constant [60, 61]. Ion adsorption on iron hydroxide flocs served as the primary method of pollutant removal in the hybrid O_3/UV/EC process, however the flocs' adsorption capability was limited. The higher initial COD content indicates that there were more pollutants present in the wastewater solution. As a result, the hydroxide matrices have a greater potential to catch and remove (sweep coagulation) this higher COD from the solution. However, the amount of COD removed is higher at constant current density, which reduces electrical energy consumption as the initial COD content of effluent increases.

3.2.1.6. pH

The physiochemical characteristics of the coagulant and the pollutants in the wastewater are both impacted by pH, and it is a crucial element in the hybrid electrocoagulation wastewater treatment process [50, 60, 62]. To achieve the required starting pH range of 1 to 11, the DIW sample was operated by adding 0.1 M NaOH and 0.1 M H_2SO_4 solution. The obtained results from the pH investigation are showed in Fig. 5(b), where it can be seen that the removal of % COD (34.40 to 9%) and EEC increases (7.85 to 9.70 kWhr m^{-3}) with increasing pH up to 7, and that the removal of % COD (99 to 78.50%) and EEC (9.70 to 9.08 kWhr m^{-3}) then steadily decreases as pH increases above 7, with the maximum COD removal being attained at pH 7 with the required EEC of 9.70 kWhr m^{-3}. The removal effectiveness of COD may decline at extremely low and high starting pH values as a result of the predominance of soluble iron compounds such $Fe(OH)_4$ formation. Pollutants are removed from the wastewater in this process through adsorption, precipitation, or flotation, with the creation of iron hydroxides flocs serving as the main mechanism for color and COD removal [63]. It should be noted that different monomers, such as $Fe(OH)_3$, and hydroxyl complexes and polymer compound such as $Fe(H_2O)^{3+}{}_6$, $Fe(H_2O)_5(OH)^{2+}$, $Fe(H_2O)_4(OH)^{2+}$, $Fe(H_2O)_8(OH)^{2+}$, and $Fe(H_2O)_6(OH)^{2+}$ are generated from iron hydroxides when the pH varies, which affects the color and COD removal efficiency [64, 65]. The observed results agreed with the findings of Asaithambi et al. [66], who determined that removal effectiveness was low at acidic or alkaline pH values.

Fig. 5(c) illustrates how initial pH affects space-time yield. The results show that the increasing pH value from a 1 to 7, increases the space time yield from 20.68 to 60.466 kg $m^{-3}h^{-1}$, while further increasing from 7 to 11, reduces the space time yield from 60.466 to 46.69 kg $m^{-3}h^{-1}$. At initial pH 7, when charge loading is at its lowest, the space-time yield is at its highest.

3.2.1.7. Inter-electrode spacing

The Ohmic potential of the hybrid EC process and energy consumption are known to be influenced by inter-electrode distance for the treatment of wastewater/industrial effluent [67,68]. The elimination effectiveness of COD and EEC is shown in Fig. 6(a) for various inter-electrode distances, including 0.75, 1.5, 2.25, 3, and 3.75 cm. It can be revealed in Fig. 6(a) with an inter-electrode gap of 0.75 cm, the highest COD removal efficiency is 98.99% with an EEC of 9.70 kWhr m^{-3}. At inter-electrode distances of 1.5, 2.25, 3, and 3.75 cm, hybrid O_3/UV/EC performance exhibits COD elimination efficiencies of 84.88, 73.12, 70.77, and 60.52% as well as EEC efficiencies of 13.10, 15.50, 18.60, and 22.80 kWhr m^{-3}, respectively. The findings unambiguously show that for inter-electrode distances greater than 1.5 cm, particle collisions may result from strong electrostatic charges between the electrodes, and the formation of flocs significantly increases the number of gas bubbles that cause turbulence, which is largely responsible for lower COD removal efficiency and higher EEC [45, 69].

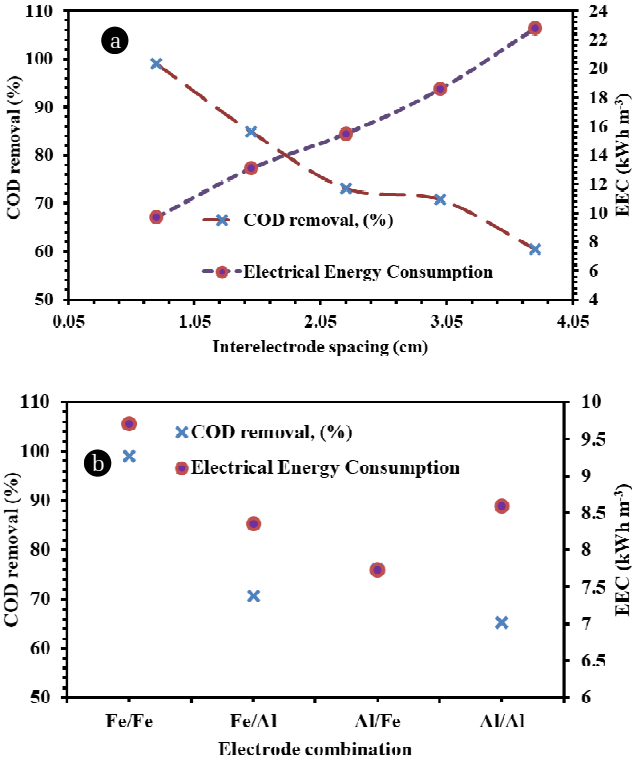


Fig. 6. (a). Effect of distance between electrodes on % COD removal and EEC from DIW by hybrid O_3/UV//EC process (Conditions: current density = 0.20 A dm^{-2}, pH = 7, COD concentration = 3500 mg L^{-1}, electrode combination = Fe/Fe, UV power = 32 W, O_3 concentration and flow rate = 4 g L^{-1} and 20 L min^{-1}, and reaction time = 4 h). (b). Effect of electrode combination on % COD removal and EEC from DIW by hybrid O_3/UV//EC process (Conditions: current density = 0.20 A dm^{-2}, pH = 7, COD concentration = 3500 mg L^{-1}, distance between electrodes = 0.75 cm, UV power = 32 W, O_3 concentration and flow rate = 4 g L^{-1} and 20 L min^{-1}, and reaction time = 4 h).

3.2.1.8. Combination of electrode

Under constant experimental conditions, including current density of 0.20 A dm^{-2}, pH of 7, COD concentration of 3500 mg L^{-1}, inter-electrode distance of 0.75 cm, UV power of 32 W, O_3 concentration and flow rate of 4 g L^{-1} and 20 L min^{-1}, and reaction time of 4 hr for DIW using the O_3/UV/EC process, the effects of electrode types on COD removal and EEC were examined. As shown in

Fig. 6(b), compared to Fe/Al, Al/Fe, or Al/Al, utilizing Fe/Fe resulted in a greater removal of COD. The kinds of electrode materials used as well as the composition of the anode and cathode electrodes had an impact on the hybrid system's performance, which are the main parts of the hybrid O_3/UV/EC process. Due to the Fe electrode's substantially higher oxidation potential (-0.447 V) than the Al electrode's (-1.662 V), three times as much Fe coagulant is created during pollutant removal [70, 71]. The primary reason is that $Fe(OH)_3$-generated particles may settle more readily than $Al(OH)_3$-produced ones. For these reasons, in the following investigation using the O_3/UV/EC technique for DIW, employed Fe electrodes as the anode and cathode.

3.3. Instrumental Analysis

The UV/visible absorption spectra before and after treatment with the hybrid UV/O_3/EC process were analyzed to investigate color removal from distillery industrial wastewater (Fig. 7). The absorbance spectrum of the distillery industrial wastewater contained an absorbance peak at 300 nm that belonged to the coloring agent. The absorbance of peaks decreased significantly after the treatment time, as shown in fig. 7. It might be due to the production of intermediates during the removal of color and COD.

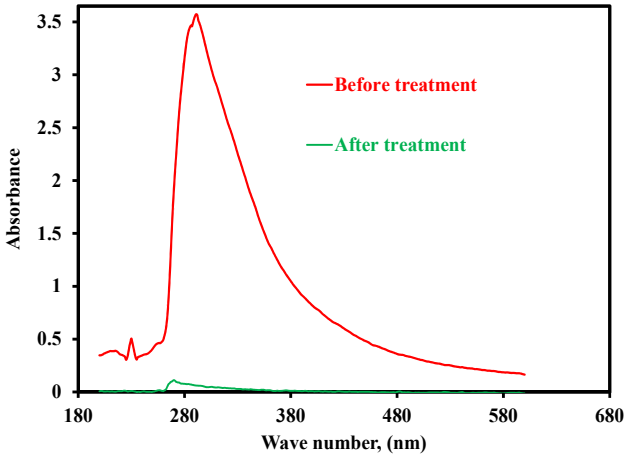


Fig. 7. The characteristic of UV – Vis spectra for treatment of distillery industrial wastewater by combined O_3/UV/EC process (Conditions: current density = 0.20 A dm^{-2}, pH = 7, COD concentration = 3500 mg L^{-1}, distance between electrodes = 0.75 cm, electrode combination = Fe/Fe, UV power = 32 W, reaction time = 4 h, and O_3 concentration and flow rate = 4 g L^{-1} and 20 L min^{-1}).

4. Conclusions

The treatment of industrial wastewater from distilleries was investigated in this work using a variety of single processes as well as a combination of electrochemical and advanced oxidation processes, such as O_3 only, UV only, EC only, and UV/EC, O_3/UV, O_3/EC, and UV/O_3/EC processes for the removal of color and COD while taking electrical energy consumption into account. The experimental findings indicate that distinct processes, such as UV, O_3 and EC, each perform less well in the treatment of effluent from diluted distilleries industrial wastewater. However, when combined into a hybrid O_3/UV/EC model, the efficiency is greater than that of the individual processes. According to the findings of our study, the hybrid O_3/UV/EC process is superior to the other process in terms of its ability to effectively remove the color of 100% and COD of 98.99%, while using the required electrical energy consumption of 9.70 kWhr m^{-3}. These findings suggest that combining UV and O_3 with an EC method may be a viable alternative to conventional physico-chemical procedures for wastewater treatment.

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Conflict-of-Interest Statement

The authors declare no conflicts of interest with regard to the publication of this research article.

Author Contribution

P.A (Assistant Professor) investigation, data curation, Resources, Writing the original draft. M.B.Y (Assistant Professor) conceptualization, methodology, validation and supervision. R.G (Assistant Professor) investigation, data curation, formal analysis and resources. S. N (Assistant Professor) investigation, data curation, formal analysis and resources. P. S (Assistant Professor) Formal analysis and resources. Z.A.R (Assistant Professor) formal analysis and resources. T. P (Assistant Professor) conceptualization, methodology, validation and supervision. A.K (Professor) conceptualization, methodology, validation and supervision, D.M (Assistant Professor) Formal analysis and resources. E.A (Professor) conceptualization, methodology, validation and supervision.

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