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Hybrid Industrial Wastewater Treatment Using a Sono-Alternating Current-Electrocoagulation Technique with Power Usage Estimation

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Distillery industrial wastewater (DIW) was tested for color and COD removal percentages using an electrochemical and advanced oxidation processes (AOPs). Specifically, the study compared direct/alternating–current–electrocoagulation (DC-EC/AC-EC), sono (US), and direct/alternating current–electrocoagulation coupled with sono (US) (DC-EC/US and AC-EC/US) processes. Also evaluated were the effects of these procedures on the power needed to treat DIW. Experimental results showed that compared to single processes such as DC-EC, AC-EC, US, hybrid DC-EC/US, and the hybrid AC-EC/US process achieved a total color elimination efficiency of 100% and a COD elimination efficiency of 100% while using a lower power consumption of 4.76 $\mathrm{kWhrm^{-3}}$. The effects of important operational factors such treatment duration, cycle of pulse duty, sonication power, current density, chemical oxygen demand, electrode spacing, electrode pairing, pH, concentration of electrolyte on the % removal of COD and power usage of DIW were investigated using hybrid AC-EC/US process. When using a Fe/Fe electrode combination, the effectiveness of COD removal was shown to be enhanced by increasing the treatment duration, current, US power, and decreasing the COD concentration, electrode spacing. The study also provided the results of an investigation into the synergistic index between AC-EC and US process and operational cost. Based on its ability to efficiently and effectively remove contaminants from wastewater and industrial effluent, the AC-EC/US approach stands out among the other methods.

1. Introduction

The water crisis is a critical gLobal issue, particularly in urban regions,[1,2] despite the fact that the majority of the earth's surface is covered in water that is either polluted or unusable. Rapid industrialization in the last several decades has resulted in a corresponding rise in the demand for fresh water across a wide range of industries. The distillery,[3] pulp and paper mill,[4] textile,[5] pharmaceutical,[6] and sugar industries[7] are significant consumers of water. Because of this, these industries generate a significant amount of effluent, which is then added to surface or ground water and contaminates the water supply. Hence, the accessible source of drinking water suffers from severe pollution, which is the primary cause of the water crisis. Water is essential for the sustenance of all living things, including humans, animals, and plant life; therefore, industrial effluent must be treated appropriately.

During the past few decades, researchers have investigated various hybrid electrocoagulation (EC) techniques for treating wastewater.[8,9] These techniques include ozone (O_3) assisted EC,[10–13] sono(U)S–EC,[13–15] alternating current–EC,[16,17] peroxi–EC,[18] photo–EC,[19–21] aerated–EC,[22] EC/Magnetic field, EC/Adsorption, EC/reverse osmosis, EC/Chemical coagulation,[23] EC-nanofiltration and membrane filtration,[24–26] and biological treatment with EC.[27] There are a number of hybrid treatment approaches that have been developed and used for wastewater decolorization and degradation.[13–15] The EC treatment process has been considered for wastewater treatment among other developed methods because it is simple, cost-efficient, safe to operate, and environmentally friendly; it relies on coagulant species being produced by the application of a sacrificial anode.[28–31] Wastewaters with a high rate of organic loading can be effectively treated using hybrid electrochemical treatment technologies, such as EC.[9,32]

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For the treatment of different wastewaters, direct–current (DC) is extensively utilized in the hybrid EC method.[33,34] However, cathode passivation was one of the major issues with a DC-EC method that used continuous current.[35] Cathode passivation can impede the movement of metallic ions in a solution by trapping them in the surface layer of the electrode. This can result in a limited amount of metallic ions being produced due to secondary reactions taking place instead of electrode oxidation. As a consequence, there is a reduced amount of coagulant, increased energy consumption due to higher Ohmic resistance and over potential, and a lower efficiency in the removal process.[36] To alleviate the drawback of the DC-EC process with cathode passivation, either anode and cathode in DC mode operation may be replaced periodically with each other, or the alternating-current electrocoagulation (AC-EC) method may be selected.[35]

Research by Alimohammadi et al.,[16] revealed that, as compared to the DC method, the AC and polarity inverter EC approach was much more successful in removing fluoride from drinking water. Bian et al.,[37] used both DC and AC-powered electrochemical methods to demonstrate the efficacy of the high salinity bilge water treatment approach. Continuous AC was shown to be more effective than DC mode utilization in maintaining the same output at lower energy costs. Arabameri et al.,[38] examined the DC versus AC modes of the EC system for the removal of $Ni^{2+}$from aqueous solution. They found that the average amount of remaining nickel in both the DC and AC modes was 44.06 and 43.91 mgL^{-1}, the energy consumption was 34.9 and 29.2 kWh/kg Ni removed, and the electrode consumption was 2.3 and 1.2 kg Al/kg Ni removed respectively. Furthermore, they determined that the shift from traditional DC to AC coagulation resulted in a significant decrease of over 16% in energy usage and a substantial reduction of 47% in electrode consumption. Karamati-Niaragh et al.,[39] employed a continuous–electrocoagulation (CEC) technique to study the effects of AC and DC on nitrate removal and operating costs. The author discovered that the AC mode nitrate removal was marginally more effective than the DC mode. Furthermore, they found that switching from DC to AC lowered operational expenses by almost 40% on average.

According to Özyonar et al.,[40] aqueous dye solutions were significantly more effectively removed of color and COD by combining US and EC than by employing either US or EC alone. Additionally, when EC treatment was coupled with US irradiation, electrode passivation was much reduced compared to the EC technique. Using US, EC, and US+EC, Prajapati[41] analyzed the performance and cost of removing color and COD from biodigester effluent. They found that compared to using just the US and EC, the US+EC method considerably improved removal efficiencies for both color (61.60% efficiency) and COD (99.10% efficiency) with only 0.58 $kWhrm^{-3}$ of energy usage. The ECF and ECF/US technologies are practical options for treating wastewater, as demonstrated by Emerick et al..[14] Oza et al.[42] discovered that a viable strategy for eliminating hazardous compounds like arsenic involved combining US and EC treatment procedures.

One of the essential Electrochemical Advanced Oxidation Processes (EAOPs) criterion beyond pollutant removal performance is power utilization, which must be considered when comparing hybrid EAOPs to traditional treatment methods.[43] There is a lack of data regarding the possible application of the DC/AC-EC coupled with US method to quantify power consumption while removing contaminants from industrial wastewater. To the best of the authors' knowledge, no prior research has been undertaken on the coupling of AC-EC with the US method for estimating the power usage related with color and COD reduction from distillery industrial wastewater (DIW). Following a literature analysis, this study evaluated DIW power utilization with the efficacy of color and COD removal utilizing US: Sonolysis, DC-EC: Direct current–Electrocoagulation, AC-EC: Alternating current–Electrocoagulation, DC-EC/US: Direct current–Electrocoagulation coupled sono, AC-EC/US: Alternating current–Electrocoagulation coupled with sono process. In order to maximize COD removal effectiveness while consuming the least amount of power, operational parameters such as Treatment Duration (TD), Cycle of pulse duty (CPD), Sonication Power (SP), Current, Chemical Oxygen Demand (COD), Electrode Spacing (ES), Electrode Pairing (EP), Concentration of Electrolyte (CE) have been examined. Furthermore presented in this work are the results of an investigation into the synergistic index using US and AC-EC technique.

1.1. Mechanisms of US, DC/AC-EC and US+DC/AC-EC Process

1.1.1. Sono (US)

When sonolysis (US) is applied to water, hydrogen atoms and OH radicals are produced. However, there is a significant loss of $^\bullet H$ and $^\bullet OH$ species due to recombination.[44,45]

$$H_2O +))) \ (Ultrasound) \rightarrow HO^\bullet + H^\bullet \tag{1}$$

$$HO^\bullet + H^\bullet \rightarrow H_2O \tag{2}$$

$$H^\bullet + O_2 \rightarrow HO_2^\bullet \tag{3}$$

$$2HO^\bullet \rightarrow H_2O_2 \tag{4}$$

$$2HO_2^\bullet \rightarrow H_2O_2 + O_2 \tag{5}$$

$$H^\bullet + H_2O_2 \rightarrow HO^\bullet + H_2O \tag{6}$$

$$Pollutant + H_2O_2 +))) \rightarrow CO_2 + H_2O \tag{7}$$

1.1.2. DC-EC/AC-EC

When current flows through electrode material during DC-EC/AC-EC process, metal ion production takes place in–situ at the anode while typical H_2 generation happens at the cathode. The anode and the cathode undergo the subsequent reactions.

At the anode:

$$Fe \rightarrow Fe^{2+} + 2e^{-} \tag{8}$$

When the pH is alkaline

$$2OH^{-} + Fe^{2+} \rightarrow Fe(OH)_2 \tag{9}$$

Moreover, H^+attacks the electrode.

$$Fe^{2+} + {}^{1}\!/_{4}O_2 + {}^{5}\!/_{2}H_2O \rightarrow 2H^{+} + Fe(OH)_3 \tag{10}$$

Oxidation of $Fe^{2+}$to Fe^{3+}

$$Fe^{2+} + {}^{1}\!/_{4}O_2 + {}^{1}\!/_{2}H_2O \rightarrow OH^{-} + Fe^{3+} \tag{11}$$

At the anode, a process is followed to evolve the oxygen.

$$2H_2O \rightarrow O_2 + 4H^{+} + 4e^{-} \tag{12}$$

At the cathode

$$2H_2O + 2e^{-} \rightarrow H_2 + 2OH^{-} \tag{13}$$

Equation (10) shows the production of *Fe(OH)$_3$*, which allows for fast adsorption of organics and trapping of colloids particles.

1.1.3. US+DC-EC/AC-EC Process

Conversely, sonication provided both chemical and physical effects. Because of the action of cavitation bubbles, the volante shock wave and micro jet provide a physical effect that acts as a cleaning agent for the electrode, hence maintaining the in–situ generation of metal ions at the anode. Ultrasound produces in–situ formation of H_2O_2 at the cathode, which is then converted into •OH radicals. The intermediates in bulk solution react with •OH radicals and decompose into CO_2 and H_2O. The functioning mechanism of US+DC-EC/AC-EC is based on the combination of US and DC-EC/AC-EC processes. The authors proposed the equation below to represent the mechanism of the US+EC.[41]

$$\begin{gathered} Contaminants + H_2O_2 + \bullet OH + \\ Ultrasonic\ irradiation \rightarrow Intermediates \rightarrow nCO_2 \\ + nH_2O \end{gathered} \tag{14}$$

2. Materials and Methods

2.1. Wastewater Collection

The distillery industrial wastewater (DIW) utilized in this study was obtained from Addis Ababa, Ethiopia. Following the collection of a wastewater sample via the grab sampling approach, it was kept cold–at +4 °C–in a cold chamber to avoid any degradation in its quality. Various chemicals were used in the experiments such as $Na_2S_2O_3$, $K_2Cr_2O_7$, NaOH, H_2SO_4, KCl and $(NH_4)_2Fe(SO_4)_2$, etc. The chemicals utilized in the study were obtained from Merck, India, in their original form without undergoing any further purification. These chemicals were of analytical reagent (AR) grade.

2.2. Characterization

Standard procedures were utilized in order to explain the DIW. Table 1 show that the results of an examination of raw distillery effluent based on specific physicochemical properties of water quality properties.

2.3. Methods

This study experimental setup for an AC-EC coupled with US is shown in Figure 1. The experimental setup included the US and EC processes. Perpexi gLass was used to build the 1100 mL electrochemical setup with the active working volume of wastewater being 1.0 L. The necessary COD concentration of wastewater was prepared by adding distilled water to the raw distillery effluent using a dilution factor. The iron (Fe)/Fe, aluminum (Al)/Al, Fe/Fe, and Al/Fe were among the electrode combinations used as the anode and cathode. The electrodes were positioned vertically and parallel to one another, with a desirable inter electrode spacing of 0.75–3 cm. The electrodes were powered by a precisely controlled DC/AC current (APLAB Ltd; Model L1606), after the reactor was filled with 1000 mL of effluent. After measuring the effluent pH with a pH meter (Elico; Model LI120), a solution of 0.1 N NaOH and 0.1 N H_2SO_4 was added to bring the pH level up to 7. Following the experiment setup, a steady DC was applied across the electrodes using the DC/AC power source. The US water bath (Power Sonic 405; 40 kHz, 350 W) was filled with distilled water to provide optimal conditions. The EC apparatus was immersed in the US water bath in order to produce an ultrasonic environment.

Throughout the DC/AC-EC and DC/AC-EC+US processes, a multimeter was used to measure the current and voltage of the electrochemical cells. During the procedures, samples were

Table 1. Physicochemical wastewater quality parameters and the results.

Parameters	Raw	Unit
Colour	dark brown	–
Odour	burnt sugar	–
pH	4.1–4.3	–
EC	120,000–125,000	μS/cm
COD	80,000–90,000	mgL^{-1}
BOD	7,000–8,000	mgL^{-1}
TSS	15.44	gL^{-1}
TDS	5550–5750	mgL^{-1}

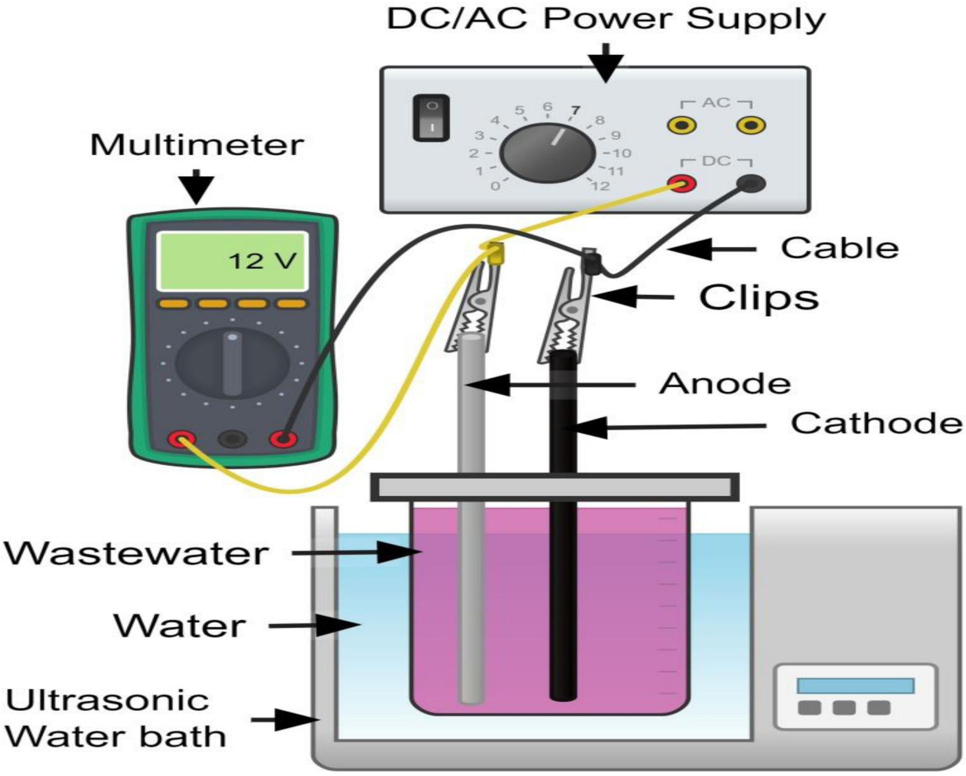

Figure 1. Alternating current–electrocoagulation coupled with sono (AC-EC/US) process setup for experimentation.

centrifuged in a REMI Model R-24 for 10 minutes at 15,000 rpm after being collected at regular intervals. The samples were then tested for COD removal and color retention. Following the recommendation of the APHA, the dichromatic closed reflux (Spectroquant ® TR320) was used to determine the COD of the sample. The Spectroquant Pharo ® 300-UV/Vis spectrophotometer was used to determine the sample colors at the wavelength that matched the maximum absorbance max (λmax−350 nm).

2.4. Analysis

2.4.1. Efficiency in Removing Color and COD

Equations (15) and (16) were used to compute the % COD removal and the efficacy of color removal.[46,47]

$$COD\ removal\ efficiency,\ (\%) = \left(\frac{COD_i - COD_t}{COD_i}\right)^{*} 100 \quad (15)$$

Where,

DIW COD before and after treatment are denoted by the COD_i and COD_t.

$$Color\ removal\ efficiency,\ (\%) = \left(\frac{A_i - A_t}{A_i}\right)^{*} 100 \quad (16)$$

Where,

A_i before treatment and A_t after treatment are the absorbances of DIW wastewater at the appropriate wave length (λmax).

2.4.2. Power Usage

The operational costs of AOPs and EC are influenced by factors like as power consumption, chemical expenses, and equipment costs. Using the following formulas (17–19), the power needs for sono (PU_{US}), direct/alternating–electrocoagulation ($PU_{DC/AC\text{-}EC}$), and direct/alternating–current–electrocoagulation coupled with sono ($PU_{DC/AC\text{-}EC+US}$) processes were determined.[47,48]

2.4.2.1. DC/AC+EC

$$PU_{DC/AC-EC} = \frac{VIt}{V_R},\ (kWhrm^{-3}) \quad (17)$$

Where, V_R stands for the used wastewater volume in liters, t for the reaction time in hours, and V and I for the average cell voltage and applied current, respectively.

2.4.2.2. US

$$PU_{US} = \frac{P_{el}^{*}t}{V_{R}^{*}60^{*}\log\left(\frac{COD_i}{COD_t}\right)}^{*}1000,\ \left(kWhrm^{-3}\ order^{-1}\right) \quad (18)$$

Where, P_{el} is the quantity of electrical power used (kW).

2.4.2.3. US + DC/AC + EC

$$PU_{US+DC/AC+EC} = PC_{US} + PC_{DC/AC-EC}\ \left(kWhr\ m^{-1}\right) \quad (19)$$

3. Results and Discussion

3.1. Comparative Investigation

Experiments were conducted to compare the US: Sonolysis alone, DC-EC: Direct current–Electrocoagulation and AC-EC: Alternating current–Electrocoagulation alone, DC-EC/US: Sono–Direct current–Electrocoagulation, and AC-EC/US: Sono–Alternating current–Electrocoagulation methods were applied to the effluent from the distillery industry and evaluated for % color and COD removal, as well as power consumption. The operational parameters, such as Treatment duration (TD)–3 hrs, Cycle of pulse duty (CPD)–0.6, Sonication Power (SP)–100 W, Current density (CD)–3.75 A dm^{-2}, Chemical Oxygen Demand (COD)–3200 mgL^{-1}, Electrode Spacing (ES)–0.75 cm, Electrode Pairing (EP)–Fe/Fe, pH–7, Concentration of Electrolyte (CE)–3 gL^{-1} were utilized, and the results are illustrated in Figures 2(a) and (b).

Figure 2 (a) shows that over the 3.5 hrs reaction time, US alone had a minor effect on color and COD removal. In contrast, applying DC-EC and AC-DE alone increased the removal efficiency to 81.50% and 91.50% of color and 71% and 83% of COD, respectively. In reality, the US alone lacks the strength necessary to allow the target pollutant to decompose in an acceptable manner. In order to degrade the target organic pollutant, the application of US alone causes a limited amount of free oxidizing radicals to be produced in the solution.[46] After 3 hours, the addition of US to the DC-EC (DC-EC/US) and AC-EC (AC-EC/US) processes resulted in a rapid increase in color removal efficiency (86% and 100%) and COD (78% and 100%). A possible explanation for this behavior is the production of stronger hydroxyl radicals due to a reduction of Fe^{2+}anions in the AC-EC/US.[49] On the other hand, when combined with AC-EC, ultrasonic irradiation has the potential to activate hydroxyl radicals, and US has a stronger synergistic effect in generating powerful hydroxyl radicals.[47,50,51] The mass transfer reaction and the solution hydroxyl radical generation would both be accelerated by the mechanical effect of ultrasonic waves.[45] Table 2 compares the removal efficiency of the combined US and DC-EC/AC-EC processes to other electrochemical and advanced oxidation processes (AOPs). As shown in the results (Table 2), combined processes have greater removal efficiency than separate processes, and there are only a few researches on pollutant removal with calculation of energy usage from distillery industrial effluent utilizing EC process.

This hybrid process needs to be evaluated economically, power utilization is a key problem when combining US with DC/AC-EC for COD and color removal from DIW. Equation (19) was used to compute the power usage for the hybrid DC/AC-EC/US process, and the results are shown in Figure 2 (b). Figure shows that the hybrid AC-EC/US process completely removed color and COD while only using 4.76 $kWhrm^{-3}$ of power for the wastewater. Power consumption was significantly higher whether using DC-EC, AC-EC, US alone, or DC-EC/US to remove color and COD compared to when using the AC-EC/US approach. Consequently, the hybrid AC-EC/US method is capable of removing contaminants from a wide variety of wastewater and industrial effluent types, regardless of their origin. In line with the local public health reuse requirements, tests were conducted on the treated industrial effluent from distilleries, and the results showed that the water was safe for both humans and the environment.

3.2. The US/AC-EC Process Operating Parameters

Process variables, including treatment duration, cycle of pulse duty, sonication Power, current density, chemical oxygen demand, electrode spacing, electrode pairing, wastewater initial pH, concentration of electrolyte on the distillery wastewater, AC-EC/US method was examined to establish the optimum parameters for maximum COD removal while using the least amount of power usage. The following section discusses the impact of these operational parameters on the AC-EC/US procedure.

3.2.1. Treatment Duration

The effectiveness of COD elimination and power utilization via the AC-EC/US technique are greatly affected by the treatment period, as shown in figure 3. According to the results of the experiments, the COD elimination efficiency (21.87 to 100%) and power usage (0.99 to 5.61 $kWhrm^{-3}$) at a constant current density of 3.75 A dm^{-2} are both increased when the treatment period increased from 0.5 to 3.5 hr. This could be because the amount of coagulant generated increases to a certain point.[43,52] However, it was discovered that prolonging the treatment period beyond optimum value (3 hr) had no effect on the process efficacy. It should be remembered that increasing the treatment duration increases the operating costs due to higher power and electrode material usage. Furthermore, it was noted that COD removal efficiencies of 83.20% were attained prior to the end of the 2.5–hrs AC-EC/US process, and that over 29% of that efficiency was accomplished in the first hour. On the other hand, as Figure 3 shows, a longer treatment period results in more power usage. This tendency can be attributable to the electrochemical cell increasing power consumption over time, which may not be economically favorable.[53]

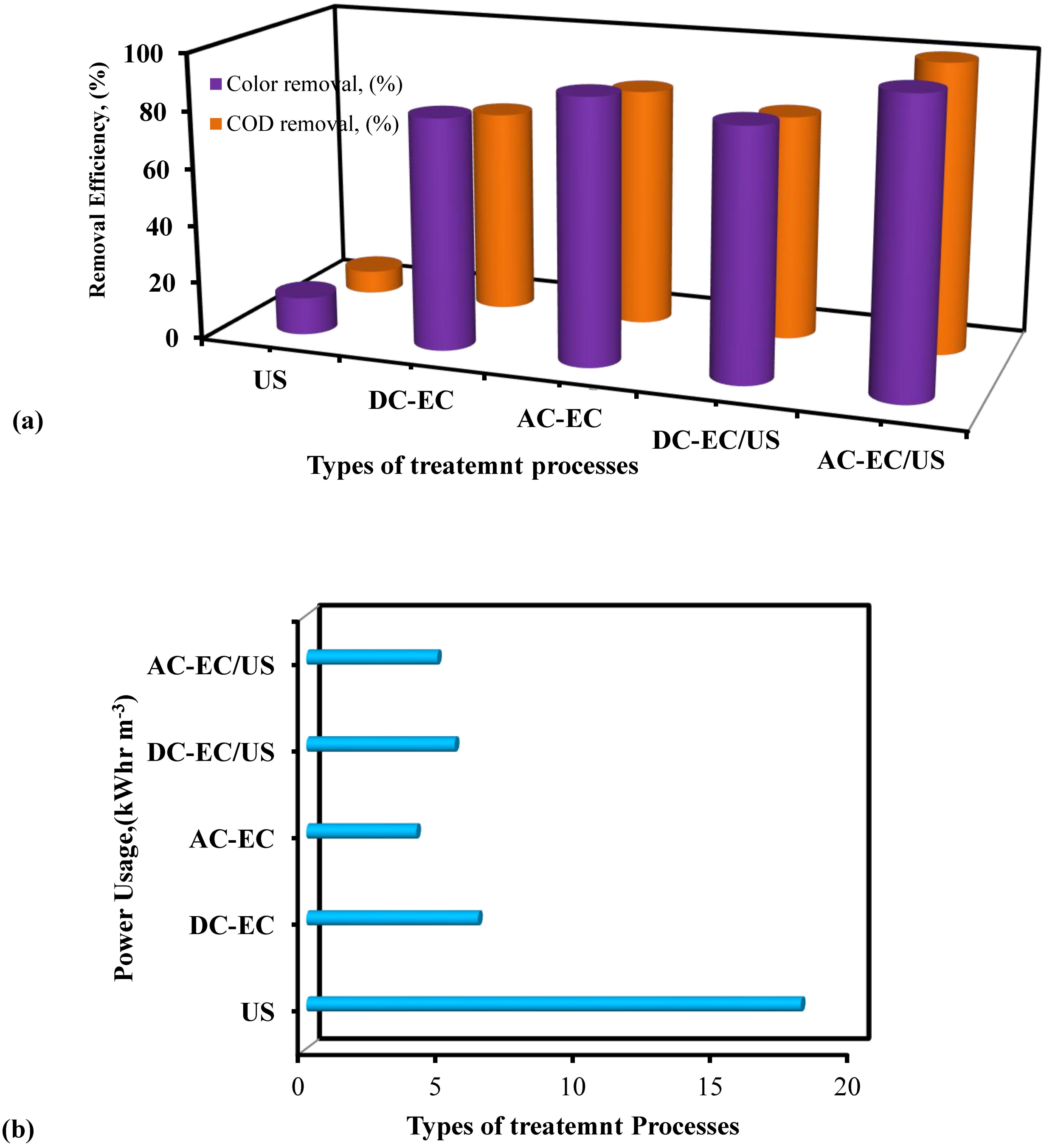

Figure 2. Comparison of various treatment methods (US: Sonolysis, DC-EC: Direct current–Electrocoagulation, AC-EC: Alternating current–Electrocoagulation, DC-EC/US: Direct current–Electrocoagulation coupled sono, AC-EC/US: Alternating current–Electrocoagulation coupled with sono) on the (a). Removal efficiency (%), and (b). Power Usage (Treatment duration (TD)–3 hrs, Cycle of pulse duty (CPD)–0.6, Sonication Power (SP)–100 W, Current Density (CD)–3.75 Adm^{-2}, Chemical Oxygen Demand (COD)–3200 mgL^{-1}, Electrode Spacing (ES)–0.75 cm, Electrode Pairing (EP)–Fe/Fe, pH–7, Concentration of Electrolyte (CE)–3 gL^{-1}).

3.2.2. Pulse Duty Cycle

The pulse duty cycle is an important characteristic to consider whenever EC is performed using AC.[54–57] The pulse duty cycle (20) describes the ratio of the total time the power is on to the total time of the reaction cycle.

$$\theta = \frac{\text{Power on time}}{\text{Whole reaction cycle}} \tag{20}$$

As demonstrated in Figure 4(a), the effect of pulse duty cycle can be calculated using the formula (21).

Table 2. The efficiency of removal and initial conditions of the present investigation are compared to previous methods used to treat industrial wastewater from distilleries.

Method	Optimum conditions	Removal efficiency	Literature
Ozone-Fenton	COD–4500 mg L^{-1}, H_2O_2/COD–0.015	Color–92 % COD–25 %	[77]
Natural Zeolite-Fe/H_2O_2	Pellet catalyst dosage–150 g/L, H_2O_2–2 g/L and 25 °C.	Color–90 % TOC– and 60 %	[78]
Electrocoagulation	Electrodes- iron, inter-electrode distance–0.5 cm, pH–7, current–5 A, agitation speed–300 rpm, electrolysis time–2 h.	Decolorization–79 %	[79]
Electro-peroxone	current density–32 A/m^2, COD - 6000 mg/dm^3, pH–6, O_3–135 mg/dm^3, Na_2SO_4–0.15 M, stirring speed–100 rpm, electrolysis time–50 min	COD–99.9 %, color–9.30 % Energy consumption–3.8 kWh/m^3	[80]
Electro-Fenton	Electrodes- carbon (graphite), Inter-electrode distance - 0.5 cm, pH–3, current –4 A, $FeSO_4$–20 mg/L and agitation speed–400 rpm, electrolysis time–3.	Decolorization–44 %	[79]
Electrochemical oxidation	Current–0.5 A, inter-electrode distance–0.5 cm, time–14 h	COD–52 % Colour–92.4 % Energy–177.4 mg COD/Wh	[81]
Fenton	pH–3, Fenton's reagent–40 mg/L, agitation- and at 500 rpm, treatment time–4 h.	Decolorization–66 %	[79]
Pulse-EF	The maximal resulted from conditions of 2.5 kHz pulse frequency, 25 % pulse duty cycle, 2 cm electrode distance and 15.8 H_2O_2/wastewater ratio.	Color–89.50 % COD–40.71 %	[82]
US	TD–3 hrs, SP–100 W, COD–3200mgL^{-1}, pH–7, CE–3 gL^{-1}.	Color–13 % COD–8 % PC–18 kWhrm^{-3}	Present study
DC-EC	TD–3 hrs, SP–100 W, CD–0.75 Adm^{-2}, COD–3200 mgL^{-1}, ES–0.75 cm, EP–Fe/Fe, pH–7, CE–3 gL^{-1}.	Color–80.50 % COD–71 % EU- 6.25 kWhrm^{-3}	Present study
AC-EC	TD–3 hrs, CD–0.75 Adm^{-2}, COD–3200 mgL^{-1}, ES–0.75 cm, EP–Fe/Fe, pH–7, CE–3 gL^{-1}.	Color–91.50 % COD–83 % EU–4 kWhrm^{-3}	Present study
US + DC-EC	TD–3 hrs, SP–100 W, CD–0.75 Adm^{-2}, COD–3200mgL^{-1}, ES–0.75 cm, EP–Fe/Fe, pH–7, CE–3 gL^{-1}.	Color–86 % COD–78 % EU–5.4 kWhrm^{-3}	Present study
US + AC-EC	TD–3 hrs, SP–100 W, CD–0.75 Adm^{-2}, COD–3200 mgL^{-1}, ES–0.75 cm, EP–Fe/Fe, pH–7, CE–3 gL^{-1}.	Color–100 % COD–100 % EU–4.76 kWhrm^{-3}	Present study

$$\theta = \frac{t_{on}}{(t_{on} + t_{off})} \tag{21}$$

Figure 4 (b) shows the results of an analysis of the effect of pulse duty cycle on COD removal percentage and power consumption under various operating conditions, such as TD–3 hrs, SP–100 W, CD–3.75 A dm^{-2}, pH–7, COD–3200 mgL^{-1}, ES–0.75 cm, EP–Fe/Fe and CE–3 gL^{-1}, respectively. Improved COD elimination from 87.90 to 100 %, decreased power consumption from 5.76 to 4.76 kWhrm^{-3}, and increased PDC from 0.15 to 0.60. On the other hand, power consumption increased from 4.76 to 5.705 kWhrm^{-3} and the percentage of COD removal decreased from 100 % to 90.59 % when the PDC was increased from 0.60 to 1.05. At both low and high PDC values, the AC-EC/US process achieved comparable COD removal percentages and power usage to the DC-EC/US method, according to the experimental data.[55] The best range for the PDC to provide maximal pollutant eradication with minimal power usage, according to the testing results, is between 0.50 and 0.60.

3.2.3. Sonication Power

When it comes to treating industrial wastewater utilizing the EC/US process, changed US intensities significantly impact pollutant elimination.[58] For industrial wastewater treatment, the AC-EC/US technique employs US power ranging from 20 to 100 W, a number of experiments examined the impact of US power on COD removal efficiency and power utilization. The results are shown in Figure 5. The COD elimination efficiency went up from 82.53 to 100 % and the power usage went up from 3.5 to 4.76 kWhm^{-3} as a result of raising the US power. With an increase in US intensity, the passive layer became thinner, which decreased resistance and improved the energy efficiency of the AC-EC/US process. Therefore, the passive coating is efficiently removed by the ultrasonic method, leading to an increase in the release of coagulant metal.[59,60] When Li et al. (2013)[42] compared the US and EC methods both alone and together, they found comparable outcomes for the efficiency of phosphate removal. In terms of phosphate removal efficiency, they discovered that the US + EC system worked

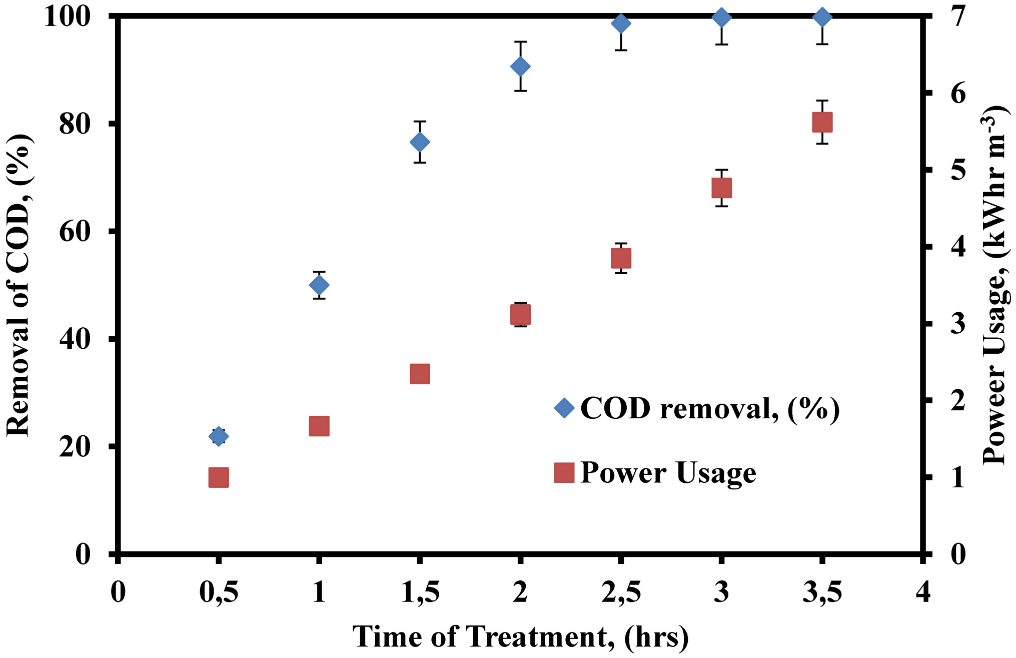

Figure 3. Effect of treatment duration on the removal of COD (%) and power usage during AC-EC/US process (CPD–0.6, SP–100 W, CD–3.75 Adm^{-2}, pH–7, COD–3200 mgL^{-1}, ES–0.75 cm, EP–Fe/Fe, CE–3 gL^{-1}).

better than the sum of the two systems alone. This illustrates how combining the US and EC processes has a beneficial result.

3.2.4. Current Density

The impact of applied current density on the effectiveness of COD removal and power usage was assessed within the range of 0.75 to 4.25 Adm^{-2}, taking into account that the US/EC process is the primary mechanism responsible for decolorization and degradation. Current density is a key variable in the AC-EC/US process because it impacts the amount and size of electro–generated ions, which influences the treatment process performance.[61] In this study, modest current density was used to prevent the overuse of electricity. When the applied current density is increased from 0.75 to 4.25 Adm^{-2}, as shown in Figure 6, the COD removal efficiency was increases from 26.08 to 100%. The AC-EC/US process removes more % COD when the current is increased to the specified value of 3.75 Adm^{-2}, which enhances the electro–generation of irons and their dissolution and release into the bulk solution.[62] The development of HO$^{\bullet}$ during US irradiation can be accelerated by increasing the current density, which speeds up the formation of cavitation bubbles and causes them to be of smaller size. A combination of a faster dissolving rate during electro–generation of irons and an increase in the development of cavitation bubbles enhances HO$^{\bullet}$ generation and, by extension degradation efficiency.[31,63,64] When sufficient quantities of HO$^{\bullet}$ are available for the sequestration of the target pollutant, as shown in Fig. 6, increasing the current density above the optimal value does not improve the COD removal efficiency.

As shown in Figure 6, the power consumption ranges from 0.60 to 7.1 kWhm^{-3} as the current density increases from 0.75 to 4.25 Adm^{-2}. This is due to the fact that the cell potential increases in direct proportion to the power consumption, which in turn increases the current density. Optimal current density is required to limit heat generation, additional O_2 development at higher current densities, and accomplish optimum COD removal with minimum power consumption.[65]

3.2.5. COD Concentration

Using experimental conditions such as TD–3 hrs, SP–100 W, pH–7, CD–3.75 Adm^{-2}, CPD–0.6, ES–0.75 cm, EP–Fe/Fe, and CE–3 gL^{-1}, the impact of COD concentration on the AC-EC/US process efficiency was assessed. As the concentration of COD increases from 1600 to 8000 mgL^{-1}, the efficiency of the AC-EC/US process drops from 100 to 49.60%, as shown in Figure 7. The AC-EC/US technology successfully breaks down organic and inorganic contaminants in highly polluted water sources. With an initial COD concentration of 4800 mgL^{-1}, the removal effectiveness was over 80%, as demonstrated in Figure 7. The treatment method was more effective in removing CODs when the starting concentration of the target effluent was lower. Power consumption decreased from 6.09 to 2.71 kWhrm^{-3} as shown in Fig 7, when the initial COD content increased from 1600 to 8000 mgL^{-1}. The oxidizing agents, free radical species, and electro–generated irons are ineffective at removing and degrading larger quantities of COD.[66,67] One alternative is that the amounts of COD in the solution cannot be adequately adsorbed and precipitated by the $Fe(OH)_3$ coagulant that is generated.

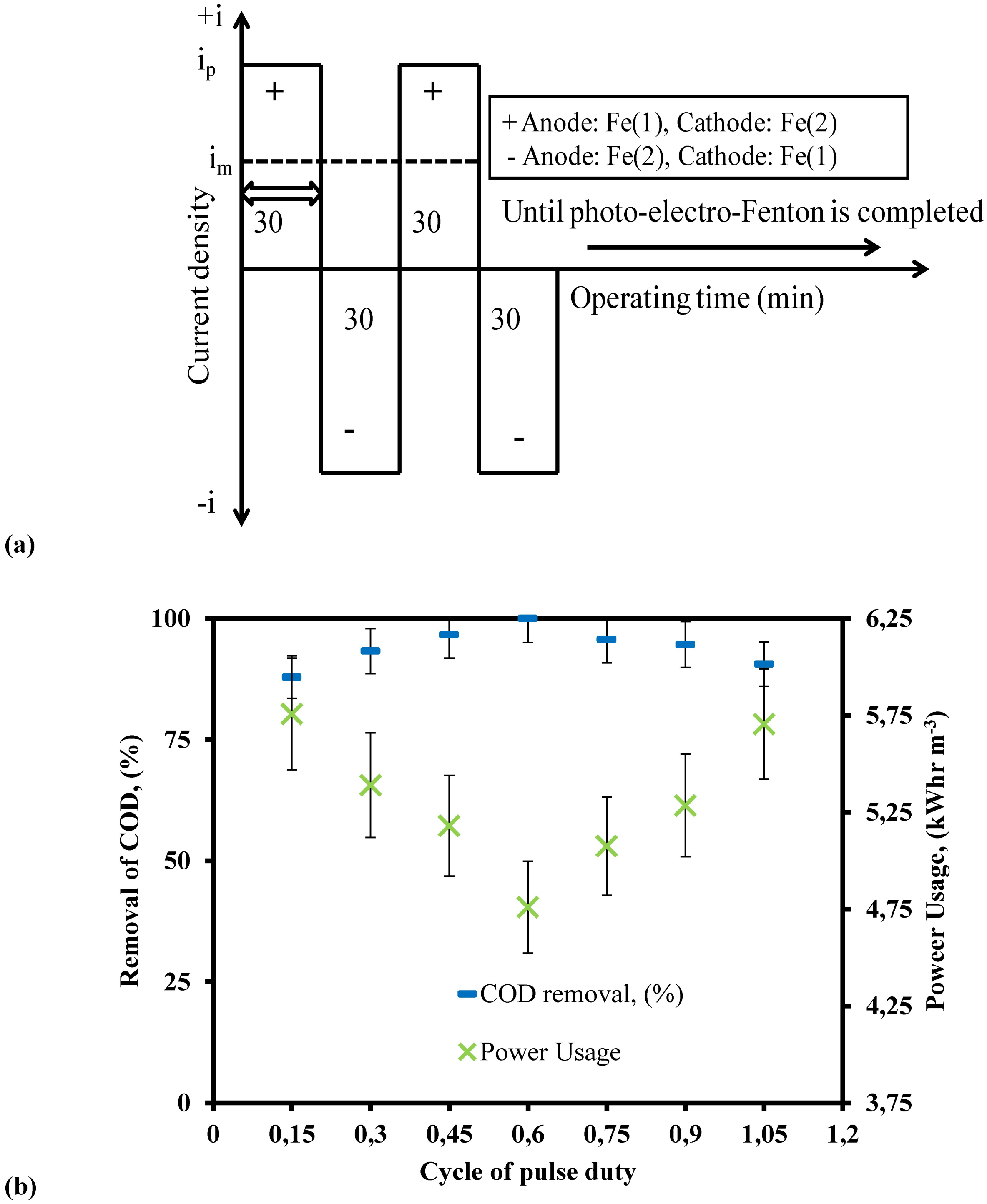

Figure 4. **(a)** Diagram showing the industrial wastewater treatment process for distilleries using AC-EC/US (Each anode and cathode will run for 30 minutes before being switched out or replaced until AC-EC/US is finished). **(b)** Pulse duty cycle impact on the removal of COD (%) and power usage during AC-EC/US process (TD–3 hrs, SP–100 W, CD–3.75 Adm^{-2}, pH–7, COD–3200 mgL^{-1}, ES–0.75 cm, EP–Fe/Fe, CE–3 gL^{-1}).

3.2.6. Electrode Spacing

Figure 8 is a schematic representation of the findings of experiments that were conducted utilizing the AC-EC/US procedure with anode and cathode spacing varied from 0.75 to 3 cm. As the distance between the anode and cathode increased, the power consumption went up from 4.76 to 14.53 $kWhrm^{-3}$, and the percentage of COD eliminated went down from 100 to 66.40%. Electrical current decreased, voltage increased, and IR-drop increased as the distance between the anode and cathode was increased. Furthermore, the reasons for the increase in power consumption and decrease in the percentage of COD removal of distillery effluent are reduced ion interaction with the hydroxide polymer and electrostatic

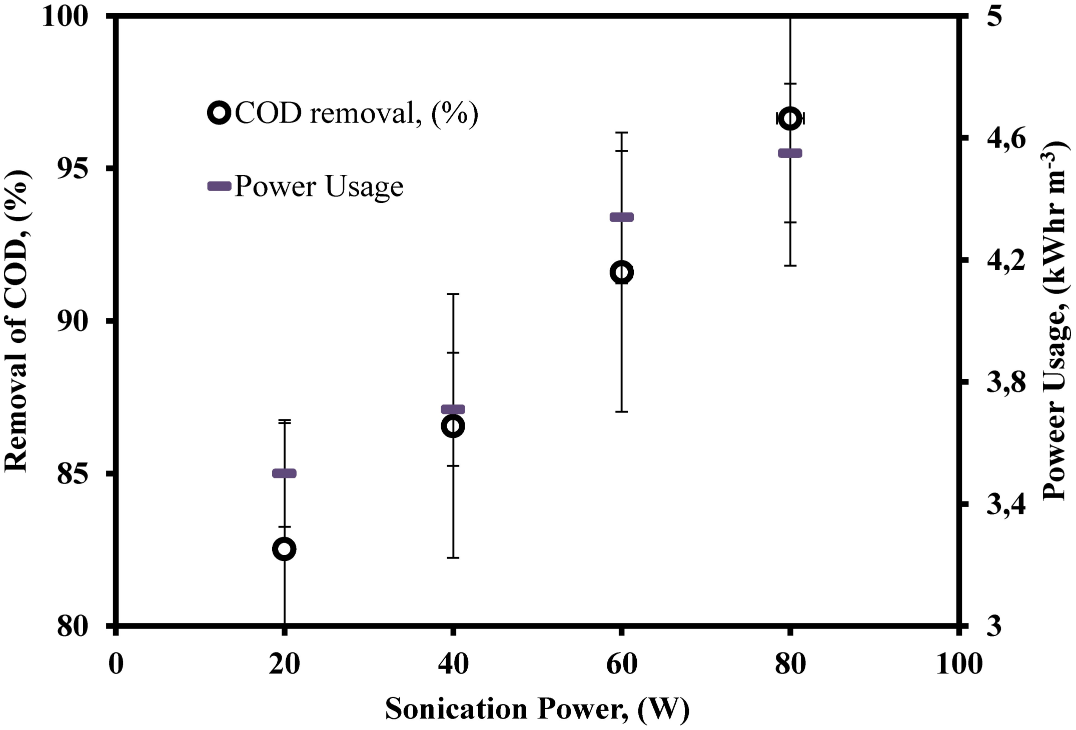

Figure 5. Effect of sonication power on the removal of COD (%) and power usage during AC-EC/US process (TD–3 hrs, CPD–0.6, CD–3.75 Adm^{-2}, pH–7, COD–3200 mgL^{-1}, ES–0.75 cm, EP–Fe/Fe, CE–3 gL^{-1}).

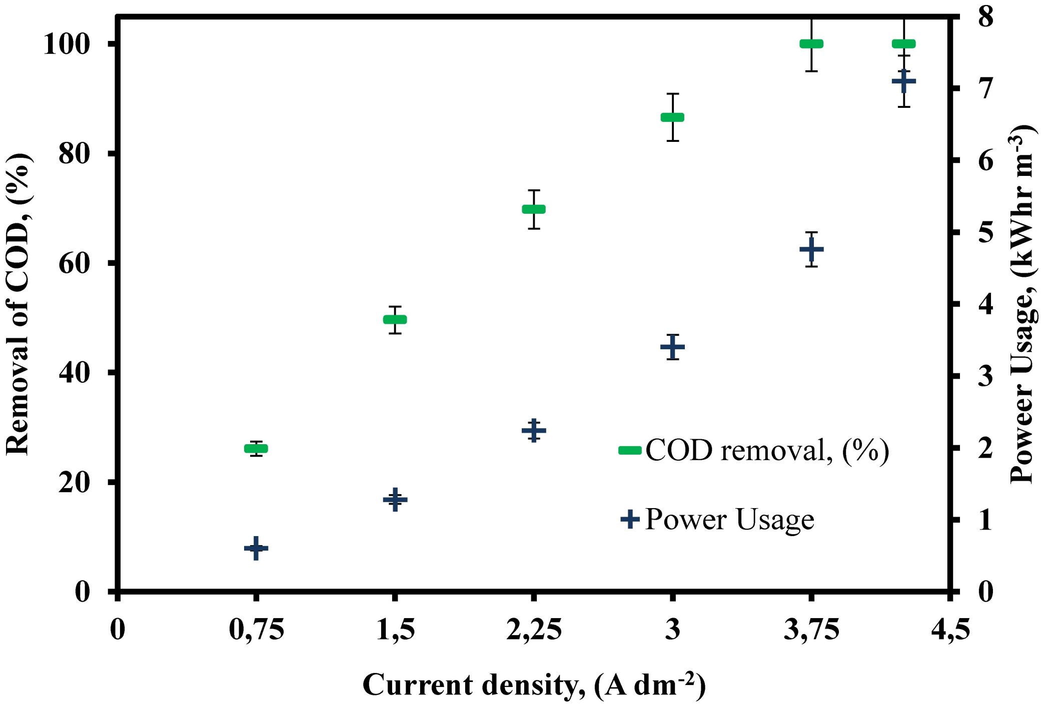

Figure 6. The influence of current density on COD elimination(%) and power usage during AC-EC/US process (TD–3 hrs, SP–100 W, CPD–0.6, pH–7, COD–3200 mgL^{-1}, ES–0.75 cm, EP–Fe/Fe, CE–3 gL^{-1}).

attraction.[68,69] In order to get a higher percentage of COD elimination with minimum power usage from the AC-EC/US process, therefore, 0.75 cm was found to be the ideal value for the inter–electrode distance between the cathode and anode.

3.2.7. Electrode Pairing

When treating wastewater with the EC method, the removal efficiency is greatly affected by the combination of electrode

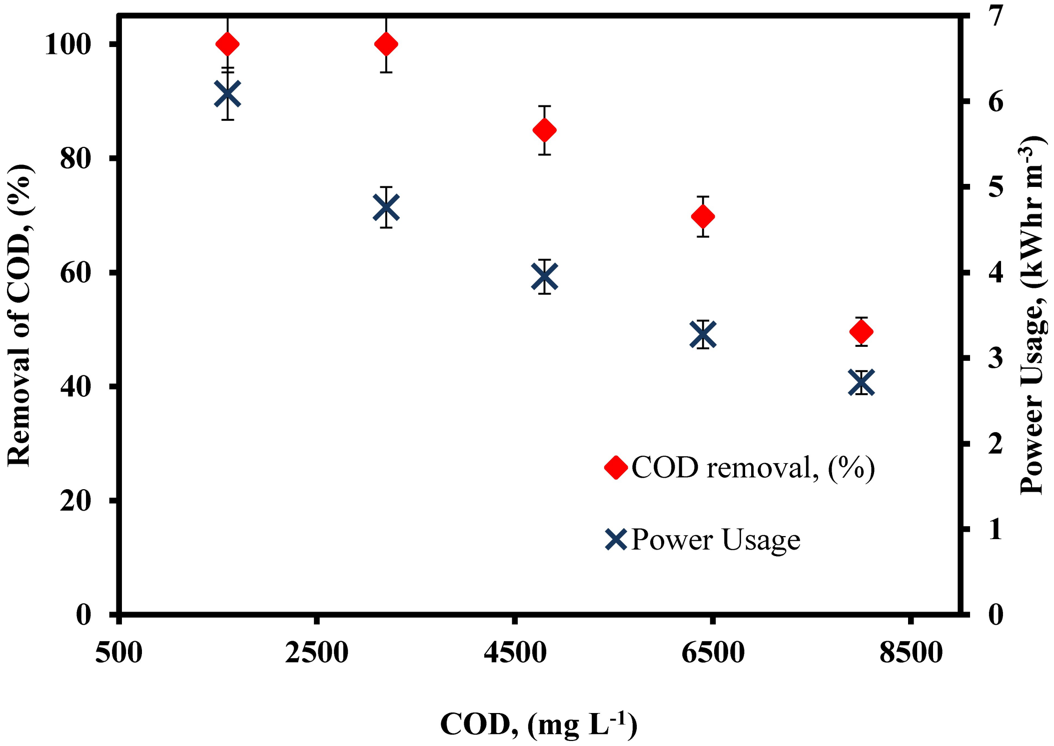

Figure 7. Influence of different COD concentration on the removal of COD (%) and power usage during AC-EC/US process (TD–3 hrs, SP–100 W, pH–7, CD–3.75 Adm^{-2}, CPD–0.6, ES–0.75 cm, EP–Fe/Fe, CE–3 gL^{-1}).

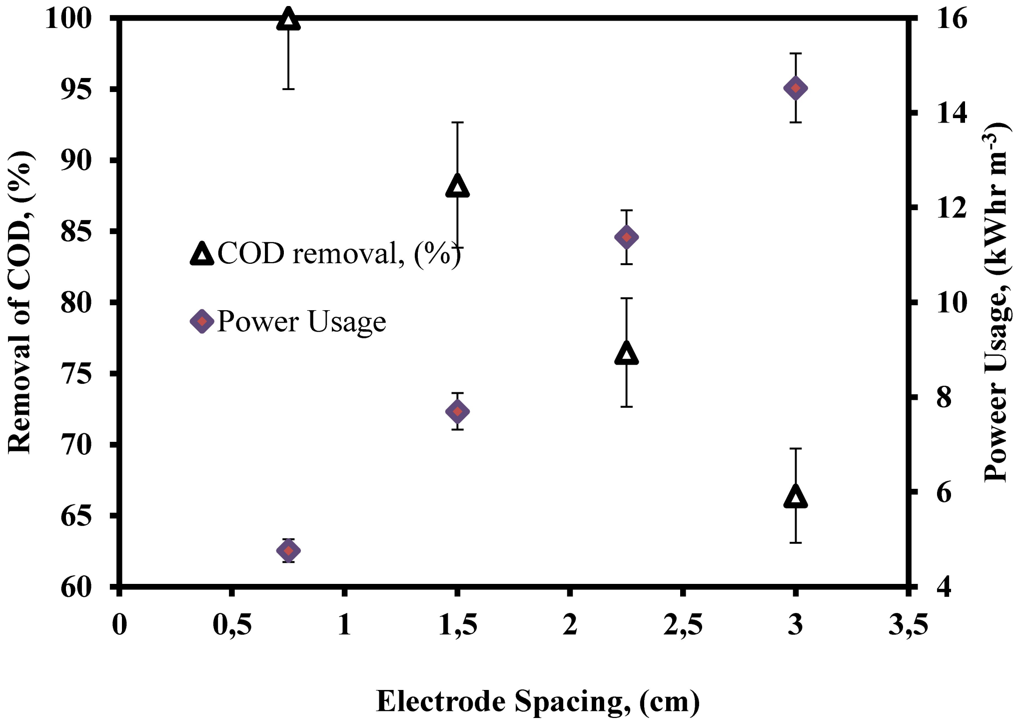

Figure 8. Effect of electrode spacing on the removal of COD (%) and power usage during AC-EC/US process (TD–3 hrs, SP–100 W, pH–7, COD–3200 mgL^{-1}, CD–3.75 Adm^{-2}, CPD–0.6, EP–Fe/Fe, CE–3 gL^{-1}).

materials.[4] To obtain a high degree of contamination removal in the AC-EC/US process, it is vital to properly select the electrode material. Figure. 9 displays the results of an investigation into the COD removal (%), and power consumption from distillery wastewater using the AC-EC/US process. The investigation included various combinations of electrodes (anode/cathode), including Fe/Fe, Fe/Al, Al/Fe, and Al/Al. For the Fe/Fe, Fe/Al, Al/Fe, and Al/Al anode/cathode electrode configurations, the COD removal and power consumption were around 100, 93.28, 89.92, and 86.56 %, and 4.76, 6.02, 5.6, and 5.81 kWhrm^{-3}, respectively, as shown in Figure 9. When compared to the other electrode configurations, the sacrificial combination of Fe/Fe was clearly superior in terms of COD elimination and power consumption. One possible explanation for why the percentage of COD removed by an Al electrode is lower than that of a Fe electrode is that hydrous aluminum oxide (HAO) cannot adsorb every organic or inorganic molecule. Furthermore, AC-EC and DC-EC both involve electrochemical processes at the Fe anode (equation.22) and Al anode (equation.23).

For Fe anode:

$$Fe_{(s)} \rightarrow Fe^{2+}_{(aq)} + 2e^{-} \tag{22}$$

For Al anode:

$$Al_{(s)} \rightarrow Al^{3+}_{(aq)} + 3e^{-} \tag{23}$$

Particles can be effectively flocculated by the nascent Fe and Al ions, which act as coagulants. Fe has a faster oxidation rate than Al because, according to the chemical reactions (Eq. 22 and 23), its oxidation potential is higher (−0.447 V) than Al (−1.662 V).[15,70] Compared to Al, the coagulant generation while employing Fe was three times higher, leading to a high COD removal percentage with minimal power consumption for the Fe anode, even when the two anodes were passed the same current.

3.2.8. Electrolyte Concentration

With respect to power consumption, the AC-EC/US process cost is directly affected by the conductivity of the wastewater.[71] Minimizing power consumption is achieved by increasing the solution's conductivity. Power consumption decreases as solution conductivity increases. Therefore, in order to improve process optimization, different concentrations of NaCl in the range of 0.75 to 4.5 gL^{-1} were used to investigate the impacts of electrolyte (NaCl) concentration on elimination COD % and power utilization at TD–3 hrs, SP–100 W, pH–7, COD–3200 mgL^{-1}, CD–3.75 Adm^{-2}, ES–0.75 cm, CPD–0.6, EP–Fe/Fe. The results are presented in Figure 10. It is observed that when NaCl concentration increases from 0.75 to 3 gL^{-1}, COD removal (%) increased from 88.24 to 100 % but further increased NaCl concentration from of 3 to 4.5 gL^{-1}, the COD removal (%) slightly decreased from 100 to 97.31 %, respectively. The lowest power consumption of 4.34 kWhrm^{-3} was recorded at the NaCl content of 4.5 gL^{-1}. This might be because adding NaCl to the solution causes anodic discharge, which results in Cl_2 and OCl^-. The process requires the least amount of power because the conductivity of the solution is increased by this OCl^-.[72] Moreover, OCl^- is a strong oxidant that eliminates organic compounds from industrial wastewater. Overdosing on NaCl causes an excess of OCl^-, which can destabilize organic particles, so the concentration of NaCl cannot rise any more.[73]

3.3. Synergistic Index (R)

The rate of COD removal for the two processes, AC-EC and US, as well as for the combined process (AC-EC/US), was compared

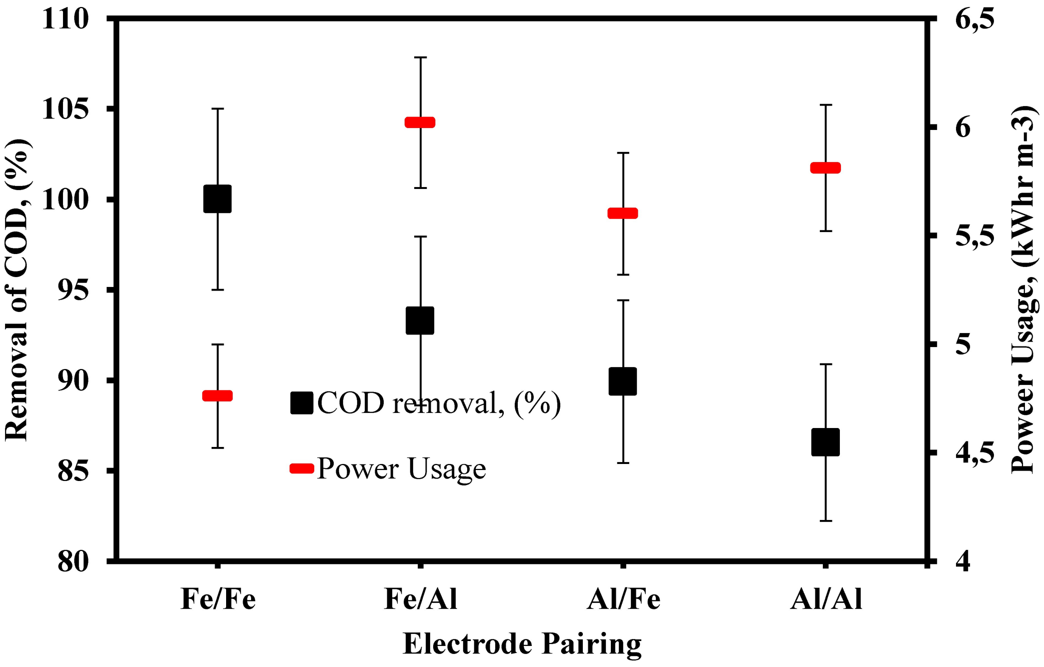

Figure 9. Effect of electrode pairing on the removal of COD (%) and power usage during AC-EC/US process (TD–3 hrs, SP–100 W, pH–7, COD–3200 mgL^{-1}, CD–3.75 Adm^{-2}, ES–0.75 cm, CPD–0.6, CE–3 gL^{-1}).

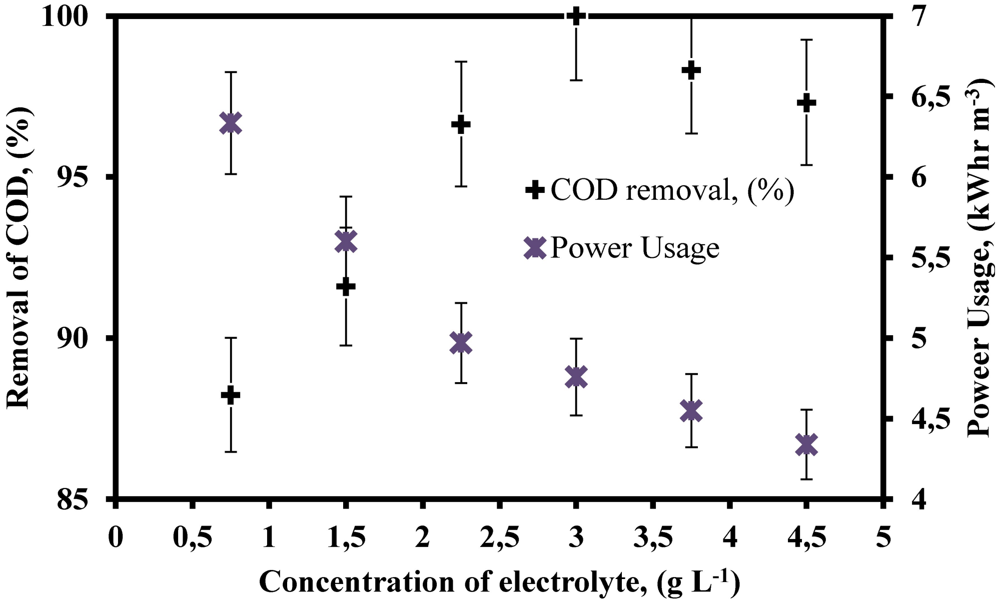

Figure 10. Influence of different electrolyte concentration on the removal of COD (%) and power usage during AC-EC/US process (TD–3 hrs, SP–100 W, pH–7, COD–3200 mgL^{-1}, CD–3.75 Adm^{-2}, ES–0.75 cm, CPD–0.6, EP–Fe/Fe).

to determine the synergistic effects. To determine these properties, the enhancement factor R was utilized in the calculation (equation 24).[49] When $R=1$, the total influence of all processes is equal to the individual effect total impact.

Each process has an effect, but when R is >1, its combined impact is larger than the sum of the individual impacts. As a last point, the synergistic effect is countered if R <1, which indicates that the combined influences are less than the sum of the individual effects.[46]

$$R=\left(\frac{\text{The rateof COD removal in the AC}-\text{EC/US}}{\text{The rateof COD removal in US }+\text{The rate of COD removal in AC}-\text{EC)}}\right) \tag{24}$$

After calculating an enhancement factor for COD removal of 7, the results showed that US irradiation and AC-EC had a synergistic impact with a R >1. This confirms the findings of earlier studies that showed that producing radicals together was much more effective than doing it individually, or that the energy needed for oxidation activities may be supplied by basically subjecting the subject matter to US irradiation. The combined technique was far more effective in removing CODs than either method alone, demonstrating the synergistic influence. In conclusion, the oxidation processes and anion reduction resulted in the production of stronger $^{\bullet}$OH.[47,50,51]

3.4. Kinetics

The AC-EC/US procedure color and COD removal rates showed a 1st-order process that was directly related to the solution's COD level. The subsequent rates of COD elimination are thus as follows.

$$-\frac{d}{dt}[COD]=k[COD] \tag{25}$$

The result is obtained by rearranging and integrating equation (26).

$$ln\left[\frac{COD_t}{COD_i}\right]=-kt \tag{26}$$

An equation with a slope of k was obtained by plotting the $\boldsymbol{ln}\left[\frac{COD_t}{COD_i}\right]$ on the y-axis against reaction time on the x-axis. Figure 11 shows the rate constant k for COD elimination to be 1.6323 min^{-1}, whereas Figure 11 shows the R^2 value to be 0.9067.

3.5. Economic Evaluation

The hybrid electrochemical process, like all other EC and AOP processes, relies heavily on cost estimation as an economic analysis parameter. The operating cost includes fixed expenditures such as electricity, chemicals, labor, maintenance, sludge disposal fees, and electrodes. Consequently, the equation (27) can be used to calculate the operational cost.[74–76]

$$Operating\ cost=A_{PC}+B_{ELC}+C_{CC} \tag{27}$$

In this case, A_{PC} stands for power consumed (kWhm^{-3}), B_{ELC} for electrode consumed (kgm^{-3}), and C_{CC} for chemical consumed (kgm^{-3}). Under optimum condition such as TD–3.5 hrs, CPD–0.6, SP–100 W, CD–3.75 Adm^{-2}, pH–7, COD–3200 mgL^{-1}, ES–0.75 cm, EP–Fe/Fe, CE–3 gL^{-1}, it was discovered that power consumption, electrode consumption, and chemical consump-

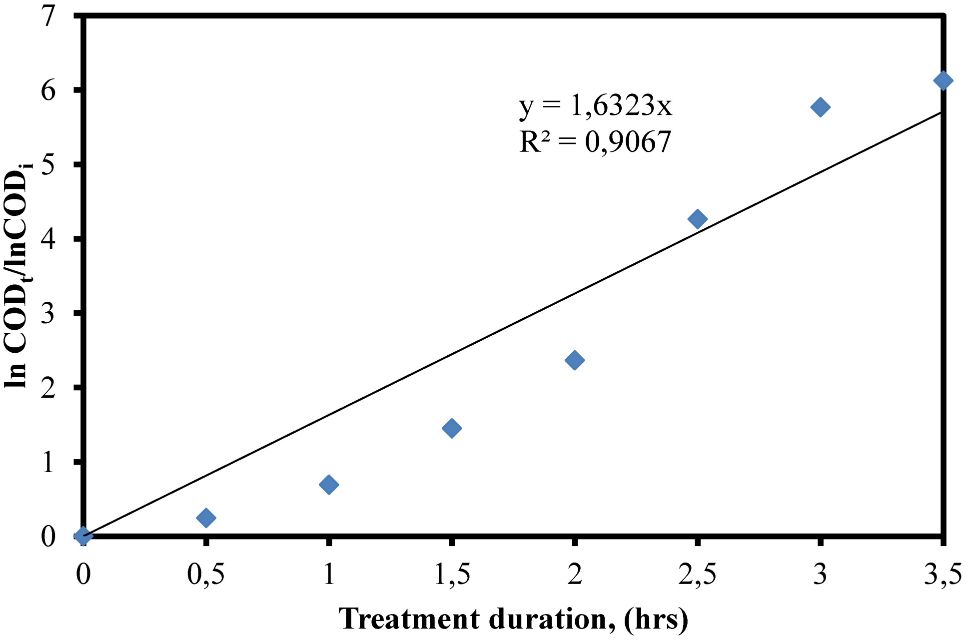

Figure 11. Degradation kinetics of distillery industrial wastewater.

tion were 4.76 kWhm^{-3}, 0.25 kgm^{-3}, and 1.25 kgm^{-3}, respectively. Equation (27) was used to calculate the running cost under optimal conditions, which came out to be 3 US $/m^3.

3.6. Sludge Analysis

The bomb calorimeter method can be used to determine the heating value of sludge, and it was found to be 4.10 MJ/kg. This sludge can be burned in furnaces or incinerators, or it can be used to make blended fuel briquettes with other organic fuels. Following its burning, the bottom ash can be combined with cementious compositions to be used in construction materials. Setting and leaching studies on cementious mixtures have revealed that bottom ash can be widely included into cementious matrices without causing unacceptable cement setting delays or excessive heavy metal leaching from solidified products.[73] This method of disposing of hybrid electrocoagulation residue recovers energy from the residues while also chemically fixing iron and other hazardous elements found in the hybrid electrocoagulation process sludge.

4. Conclusions

Removing color and COD from distillery industrial effluent using the AC-EC/US approach has been shown to be more effective than using DC-EC, AC-EC, US, or DC-EC/US process. Various operating parameters were tested when AC-EC/US process was used to treat industrial wastewater from distilleries. The goal was to determine the optimal operating condition that would remove the most color and COD with the least amount of power consumption. The following values were determined to be optimal for the AC-EC/US process: TD–3 hrs, CPD–0.6, SP–100 W, CD–3.75 A dm^{-2}, COD–3200 mgL^{-1}, ES–0.75 cm, EP–Fe/Fe, pH–7, and CE–3 gL^{-1}, leading to improved color–100 % and COD–100 % elimination levels and reduced power usage of 4.76 kWhr m^{-3}. Combining the US and AC-EC processes can result in a synergistic index, which was also examined and reported on in this work. Finally, the kinetic studies and operational costs were analyzed in this investigation. When it comes to removing pollutants from wastewater and industrial effluents, the recently developed AC-EC/US technology might be a more technologically sophisticated option than conventional wastewater treatment processes.

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Conflict of Interests

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data Availability Statement

The authors do not have permission to share data.

Keywords: COD and color removal · Sono · Direct/alternating–current · electrocoagulation · distillery industrial wastewater · power usage

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