Sono-alternating current-electro-Fenton process for the removal of color, COD and determination of power consumption from distillery industrial wastewater

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ABSTRACT

In this study, a variety of electrochemical and advanced oxidation processes (AOPs), such as sono (US), US/ hydrogen peroxide (H_2O_2), direct/alternating current-electro-Fenton (DCEF / ACEF), and sono-direct/alternating current-electro-Fenton (US + DCEF / US + ACEF) processes were compared in terms of their ability to remove a certain percentage of color and chemical oxygen demand (COD) from distillery industrial wastewater (DIW), as well as their impact on the amount of power required to treat the wastewater. According to experimental findings, the hybrid US + ACEF process produced complete color-100% and COD-100% removal efficiencies with a lower power consumption of 3.40kWhr m^{-3} than single like US, DCEF, ACEF processes, and hybrid US/H_2O_2 and US + DCEF processes. The consequences of significant operational parameters such as treatment time (30–210 min), sonication power (20–100 W) current density (0.1–0.6A dm^{-2}), pH (1–5), COD concentration (1500–7500 mg L^{-1}), inter-electrode distance (1–4 cm), H_2O_2 concentration (100–350 mg L^{-1}), pulse duty cycle (0.14–1.00) and combination of electrode (Al/Fe, Al/Al, Fe/Fe, Fe/Al) on the % COD removal efficiency and power consumption of DIW were investigated by using hybrid US + ACEF process. The synergistic index and water recovery between US and ACEF process were also investigated and reported in this work. When compared to the other processes, the US + ACEF method is the most appropriate since it can be used effectively and efficiently to remove pollutants from wastewater and industrial effluent.

1. Introduction

Waste management by the industries is one of the most serious environmental issues confronting the globe today [1–3]. Environmentalists have a significant difficulty in the treatment of industrial wastewater produced by distilleries [4–6]. Therefore, more efficient and practical advanced treatment techniques are required to reduce the negative environmental effects and potential risks associated with distillery industrial wastewater (DIW) discharges. These technologies must also be able to meet current regulatory standards. For the deterioration of DIW, several remediation studies have been described, based on microalgae [7,8], photo (UV)/ hydrogen per oxide (H_2O_2) [9], integrated aerobic biological oxidation and ozonation (O_3) [10], microbial fuel cell [11,12], photo-catalytic degradation [13], ultrasound (US) and
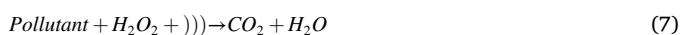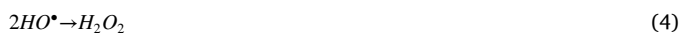
enzyme [14], adsorptive treatment [15], electrochemical (EC) [16–18], and others. Advanced Oxidation Processes (AOPs) have garnered the greatest attention and acceptance of these approaches as treatment methods for organic waste removal since they have a high organic load in terms of chemical oxygen demand (COD) and a low five days-biochemical oxygen demand (BOD₅) [19,20]. But for many AOPs, including photo-catalyst and Fenton (Fe^{2+}/H_2O_2), photo-Fenton (UV/Fe^{2+}/H_2O_2) [21], and Fenton-like AOPs [22], pre-and post-treatment of wastewater is necessary. This include controlling the pH, mixing and separating the wastewater, forming sludge, regeneration of the catalyst, and activation. As a result of integrating the two approaches (EC & AOPs), i.e., hybrid Electrochemical Advanced Oxidation Processes (EAOPs) [23–25], it is possible to overcome these shortcomings of the individual AOPs.

Combining sono-chemistry with electro-chemistry is an efficient technique to overcome the limitations of using either process alone [26–30]. The two processes have a unique synergism when they are employed together, and ultrasound's high mixing and turbulence, which increases the production of •OH radicals in the reaction media, considerably improves the electrochemical reaction kinetics [26]. In general, the sono-electrochemical (US-EC) process is able to overcome the constraints of electrode passivation and electrode fouling, as well as boost both the rate of mass transfer and the rate at which pollutants are degraded [31]. During the process of degrading persistent organic pollutants, the US-EC approach speeds up the reaction rate while also extending the life of the electrodes, improving mineralization efficiency, and allowing for in-situ cleaning of the electrode surfaces [28]. Electrochemical conversion involves either direct oxidation of pollutants at the electrode surface or mediation of these reactions by hydroxyl radicals (•OH), whereas sonolysis involves direct oxidation of pollutants through a thermolysis process occurring inside the microbubble [32]. Thus, the synergistic effect (SE) between the EC and AOPs depends on the types of pollutants, their degradative processes, advancements in mass transfer, and the role that microjets play in in-situ electrode activation [33].

1.1. Mechanisms of US, DC/ACEF and US + DC/ACEF process

1.1.1. Sono (US)

Water is subjected to sonolysis (US), which yields OH radicals and hydrogen atoms. However, because of recombination, there is a considerable loss of •H and •OH species [34,35].

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

$$H^• + {}^•\!H \rightarrow H_2O \tag{2}$$

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

$$2HO^• \rightarrow H_2O_2 \tag{4}$$

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

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

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

1.1.2. Direct/alternating current-Electro-Fenton (DC/ACEF)

The electro-Fenton (EF) method, which uses iron (Fe) electrodes to remove contaminants from industrial effluent, can be expressed as follows [36,37].

Anodic reaction:

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

Cathodic reaction:

$$2H_2O + 2e^- \rightarrow H_2 + 2OH^- \tag{9}$$

A sacrificial Fe anode is utilized as a source of Fe^{2+}, and external hydrogen peroxide (H_2O_2) is supplied to the electrochemical system [30,34,38–40].

$$Fe^{2+} + H_2O_2 \rightarrow Fe^{3+} + HO^• + OH^- \tag{10}$$
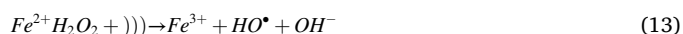
This reaction is mostly propagated by the regeneration of ferrous ions by reducing the ferric species that are formed with H_2O_2 [41].

$$Fe^{3+} + H_2O_2 \rightarrow Fe^{2+} + HO_2^• + H^+ \tag{11}$$

In order to maintain a reasonable level of HO• generation, higher ferrous ion dose is required. As a consequence, the Fenton and EF processes generate a significant quantity of ferric hydroxide sludge, which need extra separation steps and disposal.

1.1.3. Sono-direct/alternating current-Electro-Fenton (US + DC/ACEF)

Fenton's reagent (Fe^{2+}/H_2O_2) and sonication (US) work together synergistically in the US + EF process to decompose $Fe–O_2H^{2+}$ to Fe^{2+}, which then interacts with H_2O_2 to form HO•, as shown in the Eqs. (12)–(13) [30,35]. So, the addition of EF to US improves both the creation of HO• and the replenishment of Fe^{2+} [41].

$$Fe - O_2H^{2+} +))) \rightarrow Fe^{2+} + OOH^• \tag{12}$$

$$Fe^{2+} H_2O_2 +))) \rightarrow Fe^{3+} + HO^• + OH^- \tag{13}$$

Ghjair and Abbar, 2023, for the purpose of removing COD from hospital wastewater, investigated the viability of employing electro-Fenton (EF) and sono-electro-Fenton (SEF) techniques. They discovered that the EF system significantly improved when paired with the US approach. Finally, they confirmed that using SEF was a more effective and cost-efficient approach of treating hospital wastewater [42]. Mahmoudi et al., 2022 examined into the decolorization of two widely used textile dyes, such as Acid Black 172 and Disperse Blue 56, utilizing the combined sono-photo-electro-Fenton (SPEF) method. They also conducted a variety of comparisons of Fenton-based processes, including electro-Fenton (82–88%), sono-electro-Fenton (89.5–91%), photo-electro-Fenton (91.5–92.3%), and SPEF (95.5–97.4%). Additionally, they confirmed that the Fenton-based methods for water and wastewater purification were quite successful [39].

The efficiency of the sono-electro-Fenton method for removing the antibiotic cefixime from aqueous solutions was evaluated by Hasani et al. in 2020. In optimum experimental conditions, Cefixime antibiotic elimination by electro-Fenton was 81.7%, ultrasonic removal was 9%, and sono-electro-Fenton removal was 97.5%. Finally, they draw the conclusion that the sono-electro-Fenton process significantly affects the elimination of Cefixime [43]. Yousefi et al., 2019 explored the Sonoelectro-activated persulfate oxidation process for the treatment of petrochemical wastewater. They found that the combined system of sonoelectro-persulfate, electro-persulfate, sono-persulfate, persulfate, and ultrasound effectively removed COD by 82.31%, 68.97%, 21.11%, 13.06%, and 2.6%, respectively. They concluded that the sonoelectro-activated persulfate oxidation process was a successful and effective method for treating petrochemical wastewater [44].

However, in order to compare the hybrid EAOPs to traditional treatment methods, one must also take into account of power consumption as one of the key EAOP parameters in addition to pollutant removal effectiveness [45]. There is a dearth of information on the potential use of the sono-direct/alternating current-electro-Fenton technique to remove pollutants from industrial wastewater while calculating power consumption. Based on the reviewed literature, the current study compared the effectiveness of color and COD removal with power consumption from DIW using US: Sonolysis, US + H_2O_2: Sono-Hydrogen peroxide, DCEF: Direct Current-Electro-Fenton, ACEF: Alternating Current-Electro-Fenton, US + DCEF: Sono-Direct Current-Electro-Fenton, and US + ACEF: Sono-Alternating Current-Electro-Fenton process. To increase the percentage of color and COD removal efficiency with the

least amount of power consumption, operational parameters including sonication power (P), COD, pH, H$_2$O$_2$, inter-electrode distance (IED), current density (CD), pulse duty cycle (PDC), and combination of electrode (CE) have been studied. In addition, the findings of an examination into the synergistic index and water recovery utilizing sono and alternating current-electro-Fenton process are reported in this study.

2. Materials and methods

2.1. Wastewater collection

The DIW used in this investigation was collected from distillery industries in Erode, Tamilnadu, India. The wastewater sample was done using the grab sampling approach, then it was stored in a cold chamber at a temperature of +4 °C to prevent any deterioration in the quality of the wastewater. Experiments were carried out using a variety of chemicals, including H$_2$SO$_4$, NaOH, K$_2$Cr$_2$O$_7$, Na$_2$S$_2$O$_3$, and (NH$_4$)$_2$Fe(SO$_4$)$_2$, etc. The analytical reagent (AR) grade chemicals were provided by Merck, India, and the chemicals were used in their unaltered form without any additional purification being performed.

2.2. Characterization

Standard methods were used to characterize the distillery's industrial wastewater. Burnt sugar odor, dark brown color, pH: 4.1–4.3, 7000–8000mgL^{-1} of Biochemical Oxygen Demand (BOD), 80,000–90,000mgL^{-1} of Chemical Oxygen Demand (COD), 5550–5750mgL^{-1} of Total Dissolved Solids (TDS), and 15.44gL^{-1} of Total Suspended Solids (TSS) were all characteristics of the DIW.

2.3. Methods

Fig. 1 depicts the experimental setup used to study the sono-alternating current electro-Fenton (US + ACEF) process. The sonication (US) and electro-Fenton (EF) procedures were part of the experimental setup. The 1100 mL electrochemical setup was constructed using perpexiglass. Electrode configurations utilized as the anode and cathode included Al/Al, Fe/Al, Al/Fe and Fe/Fe. With a desired inter electrode spacing of 1–4 cm, the electrodes were arranged vertically and parallel to one another. After filling the reactor with 1000 mL of the wastewater, electricity was supplied to the electrodes by a carefully regulated DC/AC

Fig. 1. Experimental set up of sono-alternating current-electro-Fenton (US + ACEF) process.

current (APLAB Ltd; Model L1606). The pH of the wastewater was measured using a pH meter (Elico; Model LI120) and adjusted using NaOH (0.1 N) and H$_2$SO$_4$ (0.1 N) solution. Following the setup for the experiment, using the DC/AC power supply, a constant DC was provided across the electrodes. Distilled water was used to optimally fill the US water bath (Power Sonic 405; 40 kHz, 350 W). To create an ultrasonic environment, the electro-Fenton apparatus was submerged in the ultrasonic water bath.

During both the DC/AC + EF and US + DC/AC + EF processes, the voltage and current of the electrochemical cells were measured using a multimeter at regular intervals. The samples were taken at predetermined intervals during the processes and centrifuged in a REMI Model: R-24 (15,000 rpm, 10 min). After that, the samples were examined for color retention and COD elimination. The COD of the sample was assessed using the dichromatic closed reflux method (Spectroquant ® TR320), in accordance with the APHA's recommendations. Using a UV/Vis–spectrophotometer (Spectroquant Pharo ® 300), the samples colors were determined at the wavelength corresponding to the maximum absorbance max (λ_{max} – 350 nm).

To investigate the synergistic effects of US and ACEF process were conducted. The treatment time (30–210 min), sonication power (20–00 W), current density (0.1–0.6Adm^{-2}), pH (1–5), COD (1500–7500 mg L^{-1}), inter-electrode distance (1–4 cm), H$_2$O$_2$ (100-350mgL^{-1}), pulse duty cycle (0.14–1.00) and combination of electrode (Fe/Fe, Al/Al, Fe/Al, Al/Fe) were adjusted to study how adjusting these variables affected output COD removal efficiency, and power usage.

2.4. Analysis

2.4.1. Removal efficiency
Using the following Eqs. (14) and (15), the percentage color and COD removal efficiency was determined [41,44].

$$Color\,removal\,efficiency,(\%) = \left(1 - \frac{A_t}{A_i}\right)*100 \quad (14)$$

Where,

The absorbances of wastewater from DIW before and after treatment are A_i and A_t at the appropriate wave length (λ_{max}), respectively.

$$COD\,removal\,efficiency,(\%) = \left(1 - \frac{COD_t}{COD_i}\right)*100 \quad (15)$$

Where,

The Chemical Oxygen Demand (COD) of industrial effluent from distilleries before and after treatment is known as COD_i and COD_t, respectively.

2.4.2. Power consumption
The power usage, chemical, and equipment costs are factors that affect the operational costs of EC and AOPs. The following Eqs. (16)–(18) were used to determine the power requirements for the direct/alternating-electro-Fenton (PC$_{DC/AC+EF}$), sono (PC$_{US}$), and sono-direct/alternating current electro-Fenton (PC$_{US+DC/AC+EF}$) processes [41,46].
Direct/Alternating current-Electro-Fenton process

$$PC_{DC/AC+EF} = \frac{VIt}{V_R}, (kWhrm^{-3}) \quad (16)$$

Where,

The average voltage (Volt) of a cell, denoted by V, and the current (Amp) being applied, denoted by I, V_R is the amount of wastewater used (in liters), and t is the amount of time it takes for the reaction to occur (hour).
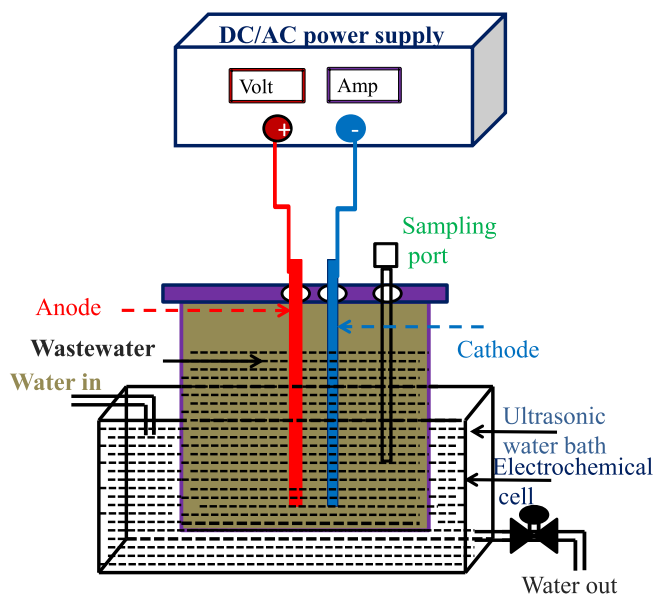**Sono (US) process**

$$PC_{US} = \frac{P_{el}*t}{V_R*60*log\left(\frac{COD_i}{COD_i}\right)}*1000, \, (kWhrm^{-3}order^{-1}) \tag{17}$$

Where,

In this equation, P_{el} is the amount of power consumed electrically (in kiloWatts).

Sono-direct/alternating current-electro-Fenton process

$$PC_{US+DC/AC+EF} = PC_{US} + PC_{DC/AC+EF}, \, (kWhrm^{-1}) \tag{18}$$

3. Results and discussion

3.1. Effect of operating parameters

3.1.1. Reaction time (RT)

The ability of EAOPs is significantly impacted by increasing the contact between the contaminant and the oxidative treatment agents when the electrolysis duration is increased [42], hence the focus of this research is on how well the US + ACEF process performs when the treatment time is changed. Different electrolysis periods (30–210 min) were used to study the effects of the US + ACEF process on the degradation, and power consumption of DIW with the Sonication Power (P)-100 W, Chemical Oxygen Demand (COD)-3000mgL^{-1}, pH-3, Hydrogen per Oxide (H$_2$O$_2$) -250mgL^{-1}, Inter-Electrode Distance (IED)-1 cm, Current Density (CD)-0.40 A dm^{-2}, Pulse duty Cycle (PDC)-0.50, Combination of Electrode (CE) -Fe/Fe) and the results are depicted in the Fig. 2. As demonstrated in Fig. 2, increasing the electrolysis time from 30 to 180 min enhanced the efficiency of % COD removal from 33 to 100%. These findings are consistent with those reported by Garca-Morales [47]. This is because the US + ACEF process generates more ferrous ions, which in turn generates more hydroxyl radicals in the aqueous bulk, making it more effective at removing pollutants. As the rate of time electrolysis increased further from 180 to 210 min, their rates of % COD removal was decreased dramatically from 100 to 96%, respectively. The increase in time reaction corresponds to the release of an excessive amount of ferrous ions from the anode electrode's surface, which may devour the created hydroxyl radicals and produce less reactive oxidizing agents [41,48].

In terms of application, power consumption is a significant measurement in electrochemical and AOPs for pollutant removal from wastewater. The power consumption was calculated using Eq. (18)

based on the COD removal and the results was given in Fig. 2. The Fig. 2 indicates that, the power consumption has increased from 1.9 to 4.1 kWh m^{-3} with an increase in treatment time from 30 to 210 min. When the treatment time increases, a constant amount of electrodes ions and their oxidizing species are produced. Increased power consumption via cell voltage results in an increase in hydroxyl radicals, which improves the COD removal [49].

3.1.2. Ultrasonic power (P)

Ultrasonic power (P) is an important parameter of the combined process of US-EC because it has a direct relationship with the amount of cavitation bubbles collapsing on the electrode's surface and the redox process current value [44]. While maintaining the other parameters values such as COD-3000 mg L^{-1}, pH-3, H$_2$O$_2$-250mgL^{-1}, IED-1 cm, CD-0.40 Adm^{-2}, PDC-0.50, CE-Fe/Fe, and RT-180 min, the influence of ultrasonic irradiation power on degrading efficiency and power consumption was examined in the range of 20–100 W. With an increase in the ultrasonic power value from 20 to 100 W, it was noticed that COD removal effectiveness improved from 84 to 100%, as shown in Fig. 3. This is due to fact that when ultrasonic power increases, more active cavitation bubbles are generated, more of them implode on the electrode surface, releasing more energy, increasing the sonochemical impact. In addition, the rate of •OH radical production increases, which is deteriorates the pollutants [50,51]. Additionally, it was examined how ultrasonic power affected the amount of power consumption needed to degrade DIW and the results are depicted in Fig. 3. It was found that the power usage was increased from 3.1 to 3.4 kWh m^{-3} when ultrasonic power increased from 20 to 100 W. This is because, with greater P, more current is needed, and hence more power is consumed. As reported by Thokchom et al., 2015 [51] energy usage increases from 0.033 to 0.127 kWh while going from 40 to 100 W of ultrasonic power.

3.1.3. Pulse duty cycle (PDC)

Whenever an alternating current (AC) is used in an electrochemical process, the pulse duty cycle (PDC) is a crucial variable [40,52–54]. A pulse duty cycle (θ) is the proportion of total of power-on time to the whole reaction cycle time.

$$\theta = \frac{Powerontime}{Wholereactioncycletime} \tag{19}$$

The Eq. (20) can be used to determine the impact of pulse duty cycle (Fig. 4(a)).
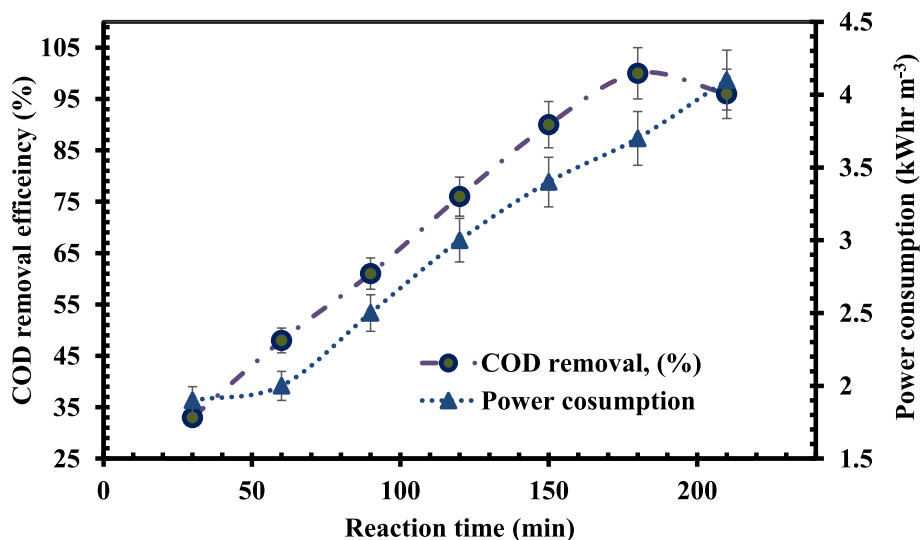

Fig. 2. Effect of treatment time (RT) on the % COD removal and energy consumption in US + ACEF process (Conditions: Sonication Power (P)–100 W, Chemical Oxygen Demand (COD)–3000mgL^{-1}, pH–3, Hydrogen per oxide (H$_2$O$_2$)–250mgL^{-1}, Inter–electrode distance (IED)–1 cm, current density (CD)–0.40 A dm^{-2}, Pulse duty cycle (PDC)–0.50, and Combination of electrode (CE)–Fe/Fe).
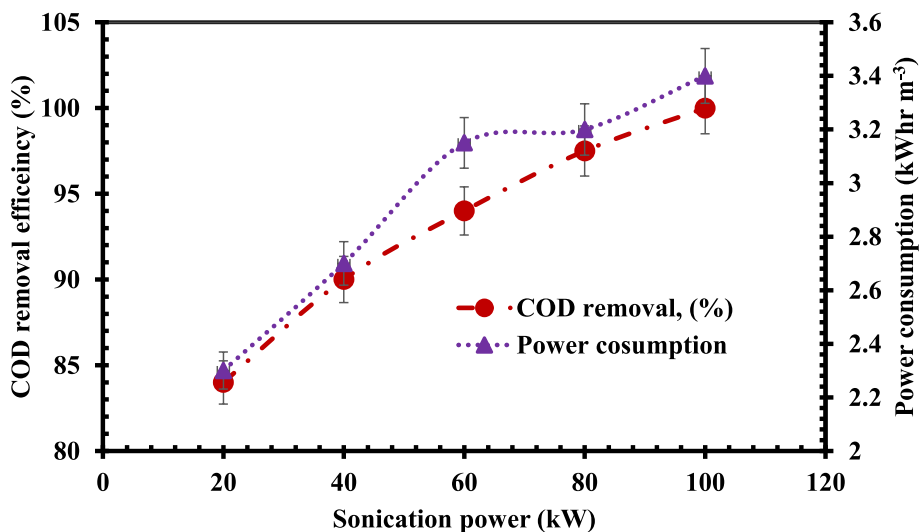
Fig. 3. Effect of US power on the % COD removal and energy consumption by US + ACEF process (Conditions: COD–3000mgL^{-1}, pH–3, H$_2$O$_2$–250mgL^{-1}, IED–1 cm, CD–0.40Adm^{-2}, PDC–0.50, CE–Fe/Fe, and RT–180 min).
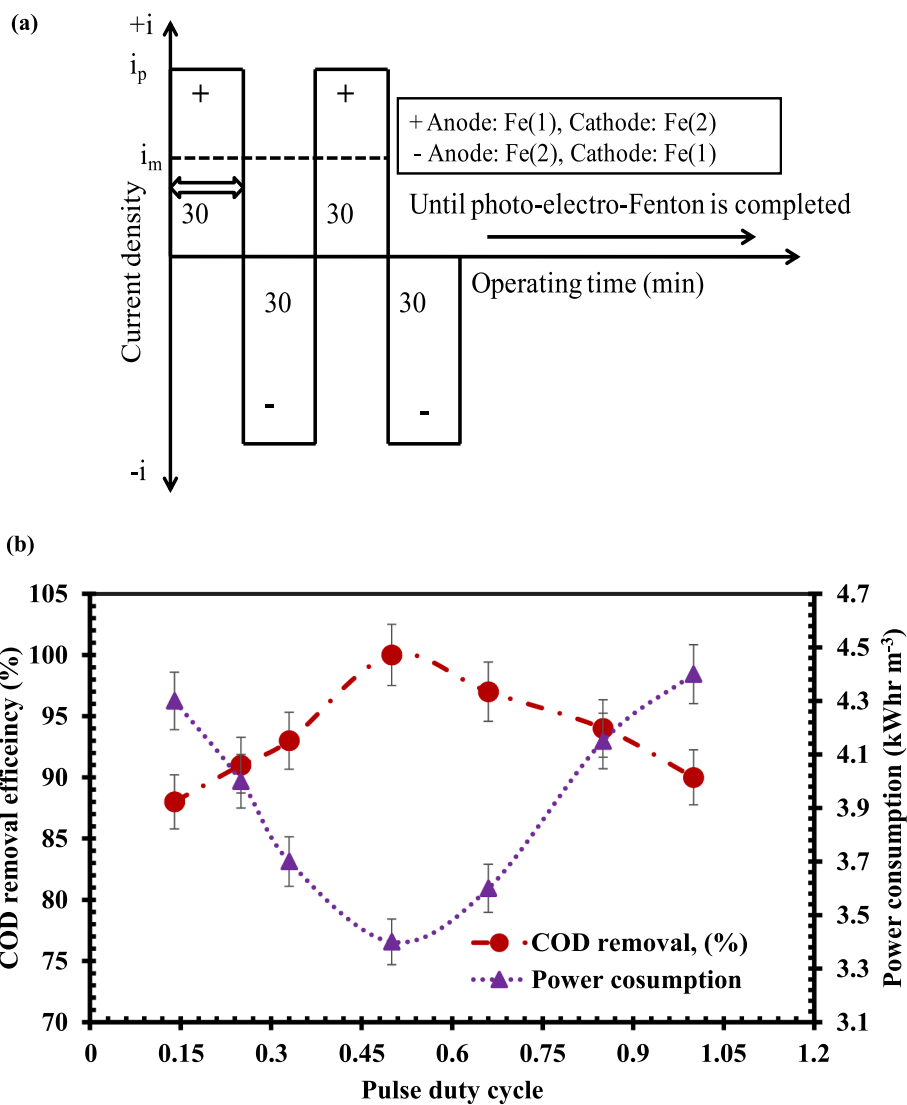

Fig. 4. (a) Schematic diagram of US + ACEF process for the distillery industrial wastewater treatment (Anode or cathode operating is operating for 30 min, then replaced or interchanged with each other until sono–electro–Fenton process completed). **(b)** Effect of pulse duty cycle on % COD removal and power consumption in US + ACEF process (Conditions: COD–3000mgL^{-1}, pH–3, H$_2$O$_2$–250mgL^{-1}, IED–1 cm, CD–0.40Adm^{-2}, CE–Fe/Fe, P–100 W, and RT–180 min).
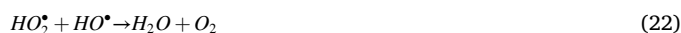
$$\theta = \frac{t_{on}}{(t_{on} + t_{off})} = \frac{t_{on}}{T} \tag{20}$$

The evaluation of PDC impact on % COD elimination and power consumption was shown in Fig. 4 (b) with the operating conditions such as COD-3000 mgL^{-1}, pH-3, H$_2$O$_2$-250 mgL^{-1}, IED-1 cm, CD-0.40 Adm^{-2}, CE-Fe/Fe, P-100 W, and RT-180 min, respectively. Increased the PDC from 0.14 to 0.50, increased the percentage of COD removal from 81 to 100% and reduced power consumption from 4.30 to 3.40kWhrm^{-3}. However, when the PDC was raised from 0.50 to 1, the percentage of COD elimination dropped from 100% to 85% while the energy consumption went up from 3.40 to 4.40kWhr m^{-3}. These findings are consistent with those reported by Ren [52].The experimental results showed that at a lower and higher value of the PDC, the % COD removal and the power consumption of the US + ACEF process were similar to that of the US + DCEF process [52]. Based on the findings of the experiments, the optimal range for the pulse duty cycle to provide maximum pollution eradication with minimal power usage is between 0.45 and 0.55.

3.1.4. H$_2$O$_2$ concentrations

When using the sono-electro-Fenton method (US + EF), the concentration of H$_2$O$_2$ is the primary source of $^\bullet$OH production [38,48]. A scavenging effect is exerted on the OH that is created by the presence of excess H$_2$O$_2$ in the reaction solution. Additionally, COD content of wastewater is increased by residual H$_2$O$_2$ in the effluent. Rising H$_2$O$_2$ dose also increases treatment costs since it is one of the key operating expenses. Therefore, optimizing H$_2$O$_2$ is crucial for the US + EF reaction. The influence of H$_2$O$_2$ on the treatment efficiencies of US + ACEF process was investigated at a fixed COD of 3000mgL^{-1}, pH of 3, IED of 1 cm, CD of 0.40Adm^{-2}, PDC of 0.50, CE of Fe/Fe, P of 100 W, and RT of 180 min. As seen in Fig. 5 raising the H$_2$O$_2$ concentration from 100 to 250 mg L^{-1}, increased the COD removal efficiencies from 65 to 100%. The observed results agreed with the findings of Serkan S¸ahinkaya [38].

Therefore, more $^\bullet$OH was produced by increasing the dose of H$_2$O$_2$, leading to improved treatment efficiencies [48]. In contrast, the rate at which COD eliminated was decreased from 100 to 90 % with further increasing H$_2$O$_2$ concentrations from 250 to 350 mg L^{-1}, respectively. This is due to the recombination of hydroxyl radicals, scavenging effect of H$_2$O$_2$ and inhibition of iron corrosion by H$_2$O$_2$ as shown in Eqs. (21)–(23) [55].

$$HO^\bullet + H_2O_2 \rightarrow HO_2^\bullet / O_2^{\bullet-} + H_2O \tag{21}$$

$$HO_2^\bullet + HO^\bullet \rightarrow H_2O + O_2 \tag{22}$$

$$HO^\bullet + HO^\bullet \rightarrow H_2O_2 \tag{23}$$

After examining different H$_2$O$_2$ concentrations, it was determined that 250 mg L^{-1} is the optimum for US + ACEF methods. Fig. 5 displays the results of an analysis of power usage based on COD elimination, showing a significant drop from 4.2 to 2.8kWhrm^{-3} when H$_2$O$_2$ concentration was increased from 100 to 350 mg L^{-1}. Perhaps as a result, the voltage of the cells dropped as the concentration of H$_2$O$_2$ increased. There is a one-to-one relationship between cell voltage and the associated running costs and energy requirements.

3.1.5. Initial pH

Particularly in Fenton chemistry, pH plays a significant influence in the electrochemical process [35,38]. The effectiveness of electrochemical processes like electro-oxidation and EF is greatly influenced by the pH of the solution. The pH of a solution directly affects the stability of H$_2$O$_2$, the rate at which OH radicals are produced, and the type and shape of iron that is precipitated out of the solution [48]. For optimal results from the EF method, acidic pH levels are preferred (pH from 2 to 4) [38]. Due to the accelerated HO$^\bullet$ formation, which results in the development of the oxidation potential at this pH range, it is possible to efficiently speciate iron and produce hydrogen that are suited for the Fenton reaction [56]. The effectiveness of the process reduces at high pH levels, notably pH over 5, since H$_2$O$_2$ decomposes quickly into H$_2$O and CO$_2$ in alkaline environments owing to structural instability. Additionally, increasing pH lowers $^\bullet$OH potential for oxidation [48].

Variations in COD removal efficiency were analyzed between pH 1 and 5 (Fig. 6). The following parameters were used to carry out this section of the study: COD-3000 mg L^{-1}, H$_2$O$_2$-250mgL^{-1}, IED-1 cm, CD-0.40 Adm^{-2}, PDC-0.50, CE-Fe/Fe, P-100 W, and RT-180 min. Results show that pH has a significant impact on both COD removal efficiency and electricity usage. The pH was raised from 1 to 3, increased the COD removal efficiency and power consumption from 87 to 100% and 2.4 to 3.4 kWh m^{-3}, respectively. Further increasing the pH from 3 to 5 (Fig. 6) decreased the COD removal efficiency and power consumption from 100 to 80% and 3.4 to 2.75 kWh m^{-3}, respectively. Our finding could be explained by the fact that the oxidation potential of $^\bullet$OH decreased with increasing pH. Because of the formation of insoluble ferric hydroxo complexes at high pH, $^\bullet$OH production is suppressed. Above a pH of 3, Fe^{3+} began to precipitate in the form of amorphous $Fe(OH)_3$. The production of $Fe(OH)_3$ not only reduced the concentration of dissolved Fe^{3+},
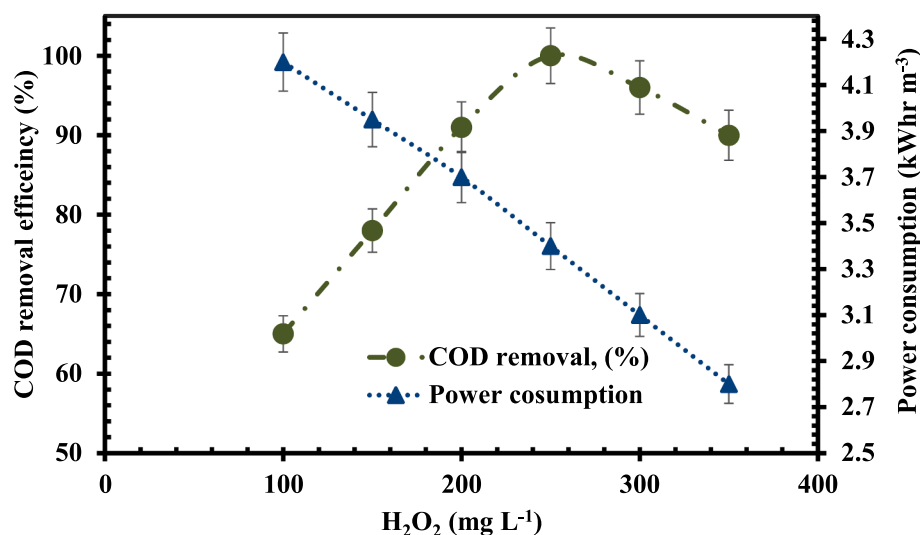

Fig. 5. Effect of H$_2$O$_2$ on the % COD removal and energy consumption by US + ACEF process (Conditions: COD–3000mgL^{-1}, pH–3, IED–1 cm, CD–0.40Adm^{-2}, PDC–0.50, CE–Fe/Fe, P–100 W, and RT–180 min).
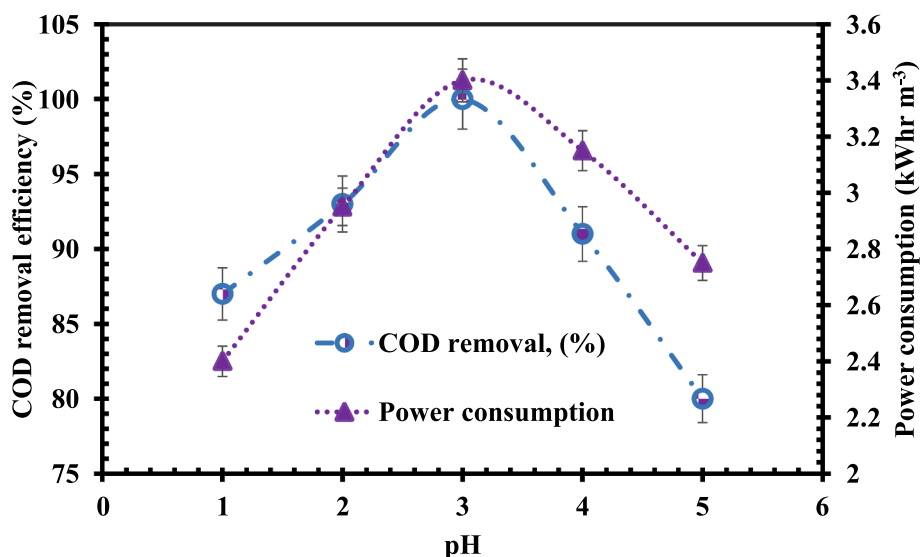
Fig. 6. Effect of pH on the % COD removal and power consumption by US + ACEF process (Conditions: COD–3000mgL^{-1}, H$_2$O$_2$–250 mg L^{-1}, IED–1 cm, CD–0.40Adm^{-2}, PDC–0.50, CE–Fe/Fe, P–100 W, and RT–180 min).

but it also prevented *Fe^{2+}* regeneration by partly covering the electrode surface. The optimal pH in EAOPs for maximum pollution removal was shown to be 3.

3.1.6. Current density (CD)

Studies evaluating the impact of the applied current density on the EAOPs system are regarded crucial since it affects the efficacy of the electrochemical process and the cost of the treatment [42]. Up to a certain value, increasing the applied current will enhance the efficiency of electrochemical processes, which is also correlated with an increase in the amount of •OH generated in the solution and the speedy reduction of ferrous ion. However, if the applied current is increased at higher, parasitic reactions of HO will become visible. Therefore, using appropriate current levels to achieve the desired efficiency is a significant needed. As shown in Fig. 7, the current density was changed from 0.1 to 0.60 A dm^{-2}, which is resulted in an increase in the percentage of COD elimination from 32 to 100%. These findings agree with those published by Yazici Guvenc [57]. This is because of an increase in *Fe^{2+}* generation

and electroreduction of *Fe^{2+}* from *Fe^{3+}* ion, both of which contributed to improved HO• formation [41,42]. In order to increase the percentage of COD removed, raising the current density increases the quantity of oxidized iron produced from the electrode. However, voltage increases as current density rises. It seems that the anodic scarification and production of *Fe^{2+}* ions, the primary element required to perform the Fenton reaction, are accelerated at higher current densities, in accordance with Faraday's law [58].

Fig. 7 implies that power consumption increased from 1.10 to 5.3 kWh m^{-3} with the rising current density from 0.1 to 0.6 A dm^{-2} because the cell potential increased with the increasing current density, directly proportional to the power consumption. To minimize heat generation, extra O$_2$ evolution at a higher current density, and to ensure maximum% COD elimination at the lowest level of power consumption, the current density must remain at an optimal level [59].

3.1.7. Inter-electrode distance (IED)

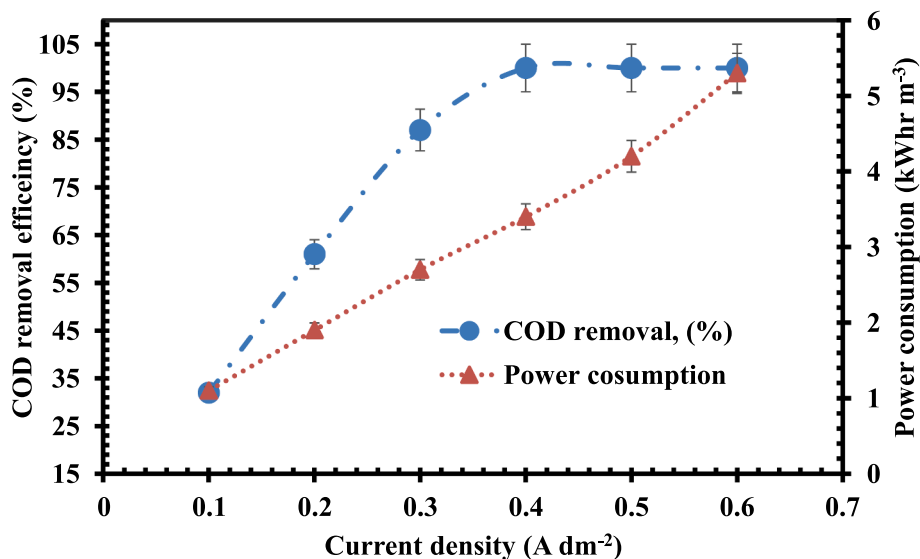According to the research available [29], the electrical resistance

Fig. 7. Effect of current density on the % COD removal and energy consumption in by US + ACEF process (Conditions: COD–3000mgL^{-1}, pH–3, H$_2$O$_2$–250mgL^{-1}, IED–1 cm, PDC–0.50, CE–Fe/Fe, P–100 W, and RT–180 min).

within the electrochemical units and the development of the passive layer on the anode surfaces are both affected by the distance between the electrodes [60]. As a result, research has begun to establish if the IED impacts the effectiveness with which the US + ACEF can remove COD. These tests were initiated with an IED of 1 to 4 cm, and the following parameters were COD-3000 mg L^{-1}, pH-3, H$_2$O$_2$-250mgL^{-1}, CD-0.40Adm^{-2}, PDC-0.50, CE-Fe/Fe, P-100 W, RT-180 min. When shown in Fig. 8, the findings revealed that as the IED was increased from 1 to 4 cm, the percentage of COD removed was reduced from 100 to 60%. Fig. 8 also shows that an increased in IED from 1 to 4 cm resulted in a increase in power usage from 3.4 to 8.2kWhrm^{-3}. The growth of a passive layer on the anode and the increase in electrical resistance are to held responsible for the negative effects of increased IEDs on the performance maintained throughout: of the electro-Fenton process [29]. This limits the production of coagulants and, as a result, reduces the electro-Fenton process's effectiveness. As a result, an IED of 1 cm was determined to be the optimum value for computing the operating cost, taking into account the findings of power consumption and COD reduction.

3.1.8. COD concentration

Since pollutant concentration has been presented as effective factors in electrochemical processes, this investigation additionally assessed the impact of COD initial concentration on removal efficiency [44,48]. Optimal conditions were used in this study, which included operating the process with COD ranging from 1500 to 7500 mg L^{-1} at pH-3, H$_2$O$_2$-250mgL^{-1}, IED-1 cm, CD-0.40Adm^{-2}, PDC-0.50, CE-Fe/Fe, P-100 W, and RT-180 min. The results showed that in Fig. 9 the degrading efficiency dropped from as the concentration of starting COD increased from 1500 to 7500 mg L^{-1}, with the US + ACEF process achieving efficiencies of 100% and 46.5% at its best and lowest. These findings are consistent with those studied by Dargahi et al. (2021) [48]. Our study's conclusions in this section can be summed up as follows: Since all experiments were conducted in the same operating condition, the same number of HO$^\bullet$ are generated for each initial concentration of COD, and the HO$^\bullet$ produced efficiently removes the significant quantity of COD at its low concentrations. However, the hydroxyl ions generated are insufficient for the breakdown of the additional pollutant present in the higher COD content. Additionally, the possibility of competition between generated intermediates molecules increases when COD concentrations are high, which is contributes to a decrease in COD removal efficiency [61]. As shown in Fig. 9, the use of the US + ACEF method resulted in a decrease in the amount of power required, which is reduced

from 4.8 to 1.9kWhr m^{-3} as the COD concentration increased from 1500 to 7500 mg L^{-1}, respectively.

3.1.9. Combinations of electrode (CE)

One of the hybrid EAOP's control criteria that influences not just the process' effectiveness and performance but also its running costs is the electrode material chosen. Under constant experimental conditions, including P-100 W, COD-3000mgL^{-1}, pH-3, H$_2$O$_2$-250mgL^{-1}, IED-1 cm, CD-0.40Adm^{-2}, PDC-0.50 and RT-180 min for DIW using the US + ACEF process, the effect of electrode types on COD removal and power usage was studied using two anode and cathode materials such as Fe and Al. As shown in Fig. 10, the use of Fe/Fe led to a greater reduction in the amount of COD that was eliminated compared to the use of Fe/Al, Al/Fe, or Al/Al. Because the anode and cathode electrodes are the key components of the hybrid US + ACEF process, the performance of the hybrid system was impacted by the kind of electrode materials as well as the component composition of the anode and cathode electrodes. Since the oxidation potential of the *Fe* electrode is substantially higher than that of the *Al* electrode (–0.447 V compared to –1.662 V), this results in three times the quantity of *Fe* coagulant being formed during the process of contaminant removal [62,63]. The primary justification for this hypothesis is based on the possibility that particles made by *Fe(OH)$_3$* have a greater capacity for settling than those formed by *Al(OH)$_3$* [63]. Because of this considerations, in this investigation continued to utilize Fe electrodes both as the anode and cathode on DIW by employing the US + ACEF process.

3.2. Comparison of US, US + H₂O₂, DCEF, ACEF, US + DCEF, US + ACEF processes

Fig. 11. (a) shows that the colour, COD elimination efficiency, and (b) power consumption time-dependent trends, which was developed utilizing the US alone, US + H$_2$O$_2$, DCEF, ACEF, US + DCEF, and US + ACEF techniques. It is abundantly clear that the US + ACEF method resulted in a greater removal efficiency of color and COD while also resulting in a lower power usage than other methods. This phenomenon may be explained as the result of the reduction of Fe^{2+} anions in the US + ACEF, which results in the generation of more powerful hydroxyl radicals [30]. However, compared to other processes, the combination of US and electro-Fenton has a larger synergistic impact to produce strong hydroxyl radicals, and ultrasonic irradiation may function as an activator for hydroxyl [38,41,48]. An increase in the mass transfer
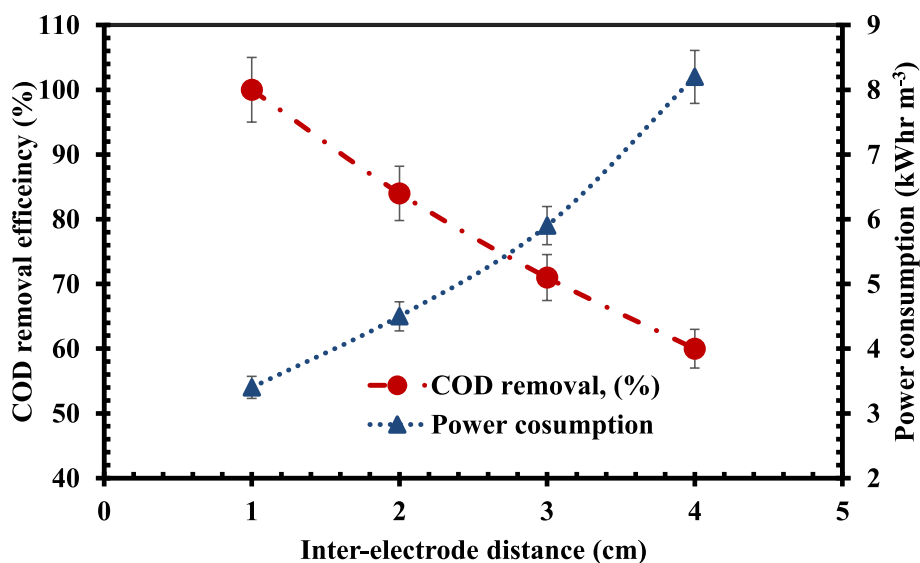

Fig. 8. Effect of inter-electrode distance on the % COD removal and energy consumption by US + ACEF process (Conditions: COD–3000mgL^{-1}, pH–3, H$_2$O$_2$–250mgL^{-1}, CD–0.40Adm^{-2}, PDC–0.50, CE–Fe/Fe, P–100 W, and RT–180 min).
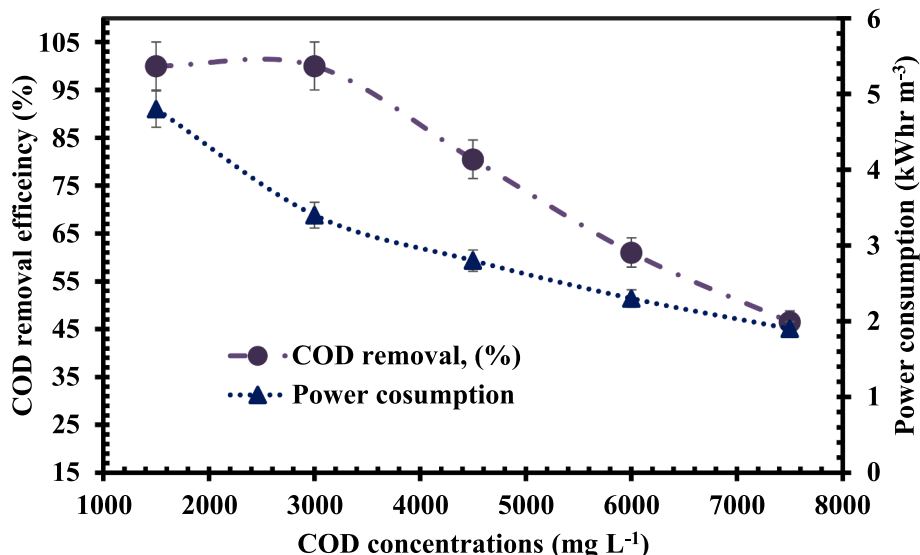
Fig. 9. Effect of COD on the % COD removal and energy consumption by US + ACEF process (Conditions: pH–3, H_2O_2–250mgL^{-1}, IED–1 cm, CD–0.40 Adm^{-2}, PDC–0.50, CE–Fe/Fe, P–100 W, and RT–180 min).
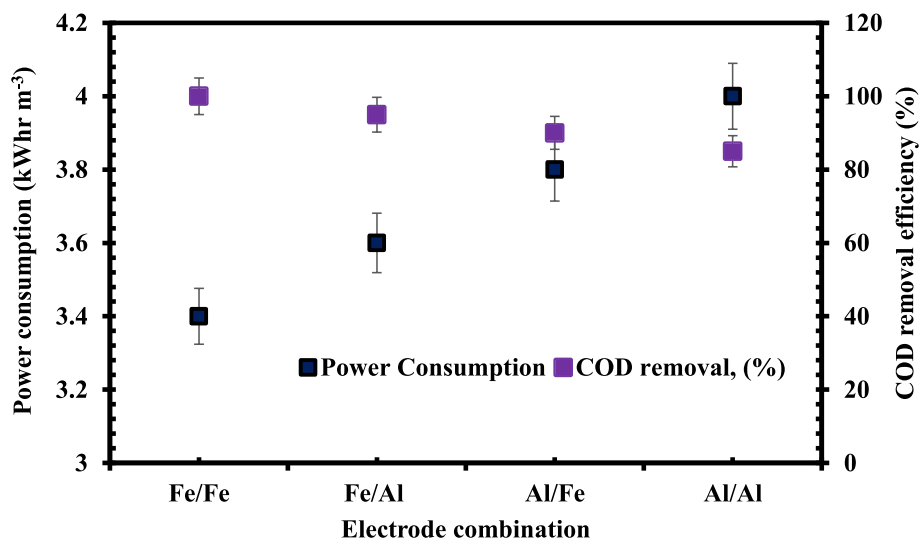

Fig. 10. Effect combination of electrode on the % COD removal and energy consumption in US + ACEF process (Conditions: P–100 W, COD–3000mgL^{-1}, pH–3, H_2O_2–250mgL^{-1}, IED–1 cm, CD–0.40Adm^{-2}, PDC–0.50 and RT–180 min).

reaction and the generation of hydroxyl radicals in the solution would be caused by a mechanical effect caused by ultrasonic radiation [35]. However, utilizing US/H_2O_2 and US irradiation did not significantly improve the removal efficiency of color or COD. Due to the fact that the cavitation process brought on by ultrasound only produced a minimal quantity of H_2O_2, the removal effectiveness of the US method for treating wastewater is practically nonexistent [44]. Since H_2O_2 chemical reaction rate is quite slow, the COD removal efficiency was not drastically reduced when using just US/H_2O_2 and US process. The formation of more powerful oxidants of hydroxyl radicals accounts for the increased rate at which the direct and alternating current-electro-Fenton proceeds in comparison to US/H_2O_2 and US process alone. Babuponnusami and Muthukumar, 2012 observed similar results for COD removal from phenol degradation by combined sono-electro-Fenton process [64]. Serkan Şahinkaya, 2013 similar results were obtained for color and COD removal from synthetic textile wastewater using an ultrasound assisted electro-Fenton oxidation method [38]. The removal efficiency of combined US and AC/DCEF process is compared with other electrochemical and advanced oxidation process and given in the Table. 1. As observed from the results (Table. 1), the combined processes gives higher removal efficiency than the individual, and also there are only few studies on the removal of pollutant with determination of power consumption from distillery industrial wastewater using EF process.

The combination of US with DC/AC + EF process for the removal of color and COD from DIW is particularly concerned with power consumption in order to establish the hybrid process's economic feasibility. The total power consumption of the hybrid US + DC/AC + EF process was computed using equation (16–18), and the results are displayed in Fig. 11. (b). The hybrid US + ACEF process eliminated 100% of color and 100% of COD while utilizing 3.4 kWhr m^{-3} of power for the DIW, as shown in Fig. 1b. To remove the color and COD, single process such as US alone, US + H_2O_2, DCEF, ACEF, and US + DCEF needed substantially more power than the US + ACEF method. As a result, the hybrid US + ACEF technique can be used to remove pollutants from almost any form of wastewater or industrial effluent, regardless of the source of the contaminants. The treated distillery industrial wastewater was tested in

Table 1
Comparison of removal efficiency and initial conditions of present study with other process applied for treatment of distillery industrial wastewater.

Method	Optimum conditions	Removal efficiency	Literature
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	[17]
Electrocoagulation	Electrodes- iron, inter-electrode distance − 0.5 cm, pH − 7, current − 5 A, agitation speed − 300 rpm, electrolysis time − 2 h.	Decolorization- 79%	[68]
EF	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%	[68]
Fenton	pH- 3, Fenton's reagent − 40 mg/L, agitation - and at 500 rpm, treatment time − 4 h.	Decolorization- 66%	[68]
EF	applied voltage −2.86 V, H_2O_2/wastewater ratio −15.8%, treatment time - and 90.7 min.	Color − 88.50%.	[69]
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%	[69]
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	[70]
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%	[71]
Ozone-Fenton	COD − 4500 mg L^{-1}, H_2O_2/COD − 0.015	Color − 92% COD − 25%	[72]
US + H_2O_2	P–100 W, COD–3000mgL^{-1}, pH–3, H_2O_2–250 mg L^{-1} and RT–180 min	Color − 35% COD − 27% PC – 7.26 kWhrm^{-3}	Present study
DCEF	COD–3000mgL^{-1}, pH–3, H_2O_2–250 mg L^{-1}, IED–1 cm, CD–0.40Adm^{-2}, CE–Fe/Fe, and RT–180 min	Color – 86 COD – 73 PC-4.1 kWhrm^{-3}	Present study
ACEF	COD–3000mgL^{-1}, pH–3, H_2O_2–250 mg L^{-1}, IED–1 cm, CD–0.40Adm^{-2}, PDC–0.50, CE–Fe/Fe, and RT–180 min	Color – 95% COD – 86% PC – 3.3 kWhrm^{-3}	Present study
US + DCEF	P–100 W, COD–3000mgL^{-1}, pH–3, H_2O_2–250 mg L^{-1}, IED–1 cm, CD–0.40Adm^{-2}, PDC–0.50, CE–Fe/Fe, and RT–180 min	Color – 100% COD – 90% PC – 4.5 kWhrm^{-3}	Present study
US + ACEF	P–100 W, COD–3000mgL^{-1}, pH–3, H_2O_2–250 mg L^{-1}, IED–1 cm, CD–0.40Adm^{-2}, PDC–0.50, CE–Fe/Fe, and RT–180 min	Color – 100% COD – 100% PC – 3.4 kWhrm^{-3}	Present study

accordance with local public health reuse regulations, and the findings revealed that the treated water is safe for public and environmental health.

3.3. Synergistic COD degradation achieved in the US + ACEF system

The synergistic effects were assessed by comparing the COD removal rates of the two processes separately (UF and ACEF) and together (UF + ACEF). R, the enhancement factor, was used to calculate these properties using the equation (24) [30]. When R equals to 1, the sum of the impacts of the combined processes is equal to the sum of the effects of the individual. If the value of R is greater than one, it indicates that the sum of the impacts of the combined processes is greater than the sum of the effects of the individual. Finally, if R is less than one, it means that the sum of the impacts of the combined processes is less than the sum of the effects of the individual (opposite the synergetic effect) [44].

$$R = \left(\frac{The\,rate\,of\,COD\,removal\,in\,the\,hybrid\,technique}{The\,rate\,of\,COD\,removal\,in\,US + The\,rate\,of\,COD\,removal\,in\,ACEF} \right)$$
(24)

Results revealed an R > 1 synergistic impact between ultrasonic irradiation and ACEF, with an enhancement factor for COD elimination of 6.50 being calculated.

This confirms that the findings of earlier studies, which is revealed that the combined process of generating radicals was far more active than either process alone, or that the energy needed for oxidation processes could be readily provided by US irradiation. However, the synergistic impact is made obvious by the fact that the combined process was significantly more efficient in COD removal than either approach alone. Conclusions stronger $^{•}OH$ were produced during oxidation processes and anion reduction [38,41,48]. In addition, Fig. 11 illustrates that the combined impact of the US and ACEF process in COD elimination, it was showed that the combination technique (US + ACEF) was much more effective at removing COD than either US irradiation or ACEF alone. The combination systems (US + ACEF and US + DCEF) removed COD by a percentage of 100% and 90%, respectively, whereas ACEF removed of 86%, DCEF removed of 73%, US/H_2O_2 removed of 27%, and a solo US system removed of 11%, respectively. As a result, it stands to reason that the synergistic effect has taken place in the integrated process. The findings showed that although individual US had a little influence on COD elimination, US generated strong mechanical vibrations inside the system, which accelerated mass transfer in the aqueous medium.

3.4. Water recovery

The water recovery rate continues to be an essential criterion in all of the treatment techniques that are available for removing pollutants from industrial effluent and wastewater [40]. One definition of water recovery is the proportion of treated water to the total volume of wastewater. In order to determine this, used the equation (25).

$$Water\,recovery = \left(\frac{(Volume\,of\,product\,water)_{after\,treatment}}{(Initial\,volume\,of\,effluent)_{before\,treatment}} \right), \left(\frac{m^3}{m^3} \right)$$
(25)

Following the US + EF process, the clear supernatant is either product water or treated water; the bottom of the US + EF reactor contains the settled sludge, and the results are shown in the Table. 2. Based on the data, the US + ACEF process mode (0.97) significantly outperformed then the US + DCEF mode (0.90) in terms of water recovery.

3.5. Kinetic studies

The color and COD removal rates in the US + ACEF process revealed a first-order process that was proportional to the COD content in the
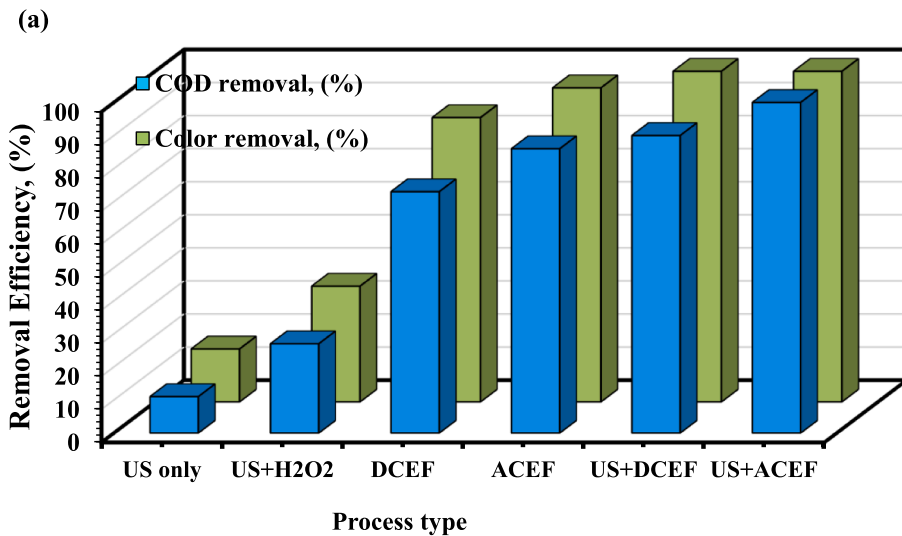
(a)

(b)

Fig. 11. Comparative analysis of different methods (US: Sonolysis, US + H$_2$O$_2$: Sono–Hydrogen peroxide, DCEF: Direct current–Electro–Fenton, ACEF: Alternating current–Electro–Fenton, US + DCEF: Sono–Direct current–Electro–Fenton, US + ACEF: Sono–Alternating current–Electro–Fenton) on the (a). % color and % COD removal, (b). Power consumption (Conditions: P–100 W, COD–3000mgL^{-1}, pH–3, H$_2$O$_2$–250 mg L^{-1}, IED–1 cm, CD–0.40Adm^{-2}, PDC–0.50, CE–Fe/Fe, and RT–180 min).
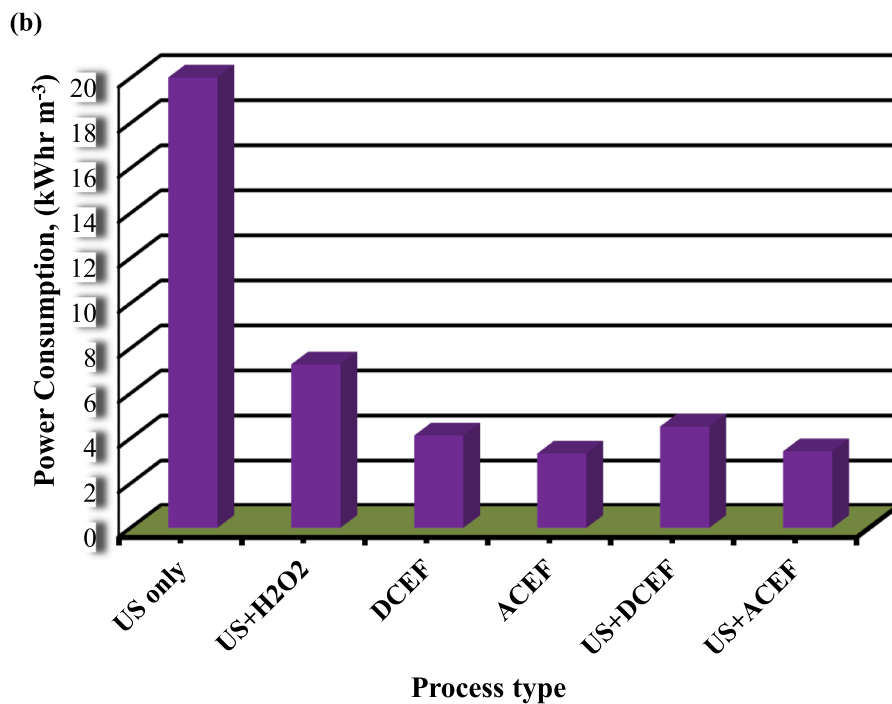
Table 2
Experimental conditions with different initial COD concentration for % color, % COD removal and power consumption in US + DCEF and US + ACEF processes.

COD, mg L^{-1}	CD, A dm^{-2}	RT, min	Final pH	Color removal efficiency (%)	COD removal efficiency (%)	Power consumption (kWhr m^{-3})	Water recovery (m^3 / m^3)
1000	0.20	180	7.1	100	100	2.70	0.97
2000	0.30	180	7.3	100	100	3.0	0.95
3000	0.40	80	6.8	100	100	3.40	0.94

solution. As a result, the COD removal kinetics are as follows.

$$-\frac{d}{dt}[COD] = k[COD] \qquad (26)$$

When, the Eq. (26) is rearranged and integrated, it yields.

$$ln\left[\frac{COD_t}{COD_i}\right] = -kt \qquad (27)$$

Plotting $ln\left[\frac{COD_t}{COD_i}\right]$ on the *y*-axis versus reaction time on the x-axis

resulted in a straight line with the slope of *k*. According to Fig. 2 and Fig. 12, the rate constant *k* and R^2 values for COD removal were 0.0195 min^{-1} and 0.83, respectively.

3.6. Economic analysis

Cost estimation is a critical economic analysis parameter in the hybrid electro-Fenton process, as it is in all other electrochemical (EC) and advanced oxidation processes (AOPs). The running cost comprises
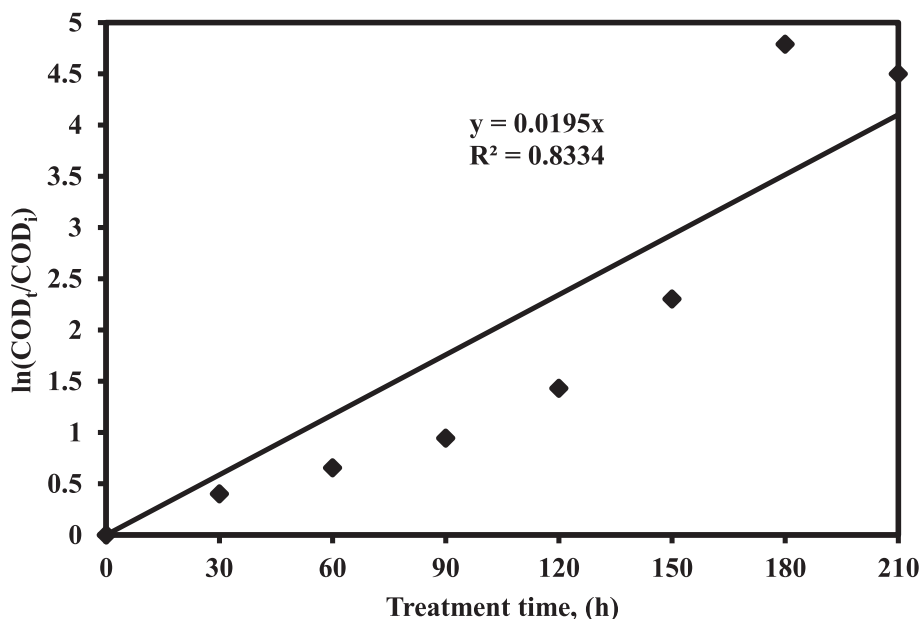
Fig. 12. Degradation kinetics of distillery industrial wastewater.

the cost of electrodes, power usage, chemical consumed, labour, maintenance, sludge disposal charges, and other fixed expenditures. As a result, the operational cost can be determined using the Eq. (28) [65–67].

$$Operating cost = A_{PC} + B_{ELC} + C_{CC} \qquad (28)$$

Where, A_{PC} = power consumed – kWh/m^3, B_{ELC} = electrode consumed– kg/m^3 and C_{CC} = chemical consumed – kg/m^3. Under optimum condition such as P-100 W, COD-3000 mg L^{-1}, pH-3, H_2O_2-250 mg L^{-1}, IED-1 cm, CD-0.40 A dm^{-2}, PDC-0.50, CE-Fe/Fe, and RT-180 min, the power consumed, electrode and chemical consumptions were found to be 3.40 kWh/m^3, 0.20 kg/m^3 and 1.50 kg/m^3, respectively. The operating cost under optimum conditions was computed using equation (28) and found to be 2.75 US $/m^3$.

4. Conclusion

It has been demonstrated that the US + ACEF method is a more effective treatment technique for the removal of pollutants from distillery industrial wastewater than the US only, US + H_2O_2, DCEF, ACEF, and US + DCEF process. When processing distillery industrial wastewater with an US + ACEF process, a number of different operating parameters were investigated in order to identify the optimal condition, which resulted in the highest possible percentage of color and COD removal efficiency with the lowest possible amount of power consumption. Using the US + ACEF process, the parameters: P-100 W, COD-3000mgL^{-1}, pH-3, H_2O_2-250mgL^{-1}, IED-1 cm, CD-0.40Adm^{-2}, PDC-0.50, CE-Fe/Fe, and RT-180 min were found to be optimal for achieving higher levels of color (100%) and COD (100%) elimination with lower power consumption of 3.40$kWhr$ m^{-3}. This study also examined and reported on the synergistic index as well as the water recovery that can be achieved by combining US and ACEF process. When compared to conventional wastewater treatment methods, the newly developed US + ACEF approach could be considered a more technologically sophisticated option for the elimination of contaminants from wastewater and industrial effluents.

CRediT authorship contribution statement

Perumal Asaithambi: Investigation, Data curation, Resources, Writing – original draft. **Mamuye Busier Yesuf:** Conceptualization,

Methodology, Validation, Supervision. **Rajendran Govindarajan:** Investigation, Data curation, Formal analysis, Resources. **P. Selvakumar:** Investigation, Data curation, Formal analysis, Resources. **Subramaniapillai Niju:** Investigation, Data curation, Formal analysis, Resources. **T. Pandiyarajan:** Conceptualization, Methodology, Validation, Supervision. **Abudukeremu Kadier:** Conceptualization, Methodology, Validation, Supervision. **D. Duc Nguyen:** Conceptualization, Methodology, Validation, Supervision. **Esayas Alemayehu:** Conceptualization, Methodology, Validation, Supervision.

Declaration of Competing Interest

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

The authors do not have permission to share data.

References

[1] V. Innocenzi, S.B. Zueva, N.M. Ippolito, F. Ferella, M. Prisciandaro, F. Vegliò, A review of the existing and emerging technologies for wastewaters containing tetramethyl ammonium hydroxide (TMAH) and waste management systems in micro-chip microelectronic industries, Chemosphere 307 (2022), 135913, https://doi.org/10.1016/j.chemosphere.2022.135913.

[2] L. Fillaudeau, P. Blanpain-Avet, G. Daufin, Water, wastewater and waste management in brewing industries, J. Clean. Prod. 14 (2006) 463–471, https://doi.org/10.1016/j.jclepro.2005.01.002.

[3] P. Asaithambi, L. Garlanka, N. Anantharaman, M. Matheswaran, Influence of experimental parameters in the treatment of distillery effluent by electrochemical oxidation, Sep. Sci. Technol. 47 (2012) 470–481, https://doi.org/10.1080/01496395.2011.621160.

[4] J. Fito, N. Tefera, H. Kloos, S.W.H. Van Hulle, Physicochemical properties of the sugar industry and ethanol distillery wastewater and their impact on the environment, Sugar Tech. 21 (2019) 265–277, https://doi.org/10.1007/s12355-018-0633-z.

[5] S.K.R. Namasivayam, M. Kavisri, G.P. Avinash, K. Samrat, R.S.A. Bharani, M. Moovendhan, Effect of distillery industry discharge wastewater and dye in aqueous solution treated by chemical modified Penicillium biomass on *Arachis hypogaea* growth parameters and its antioxidants efficiency, Biomass Convers. Biorefinery. (2022), https://doi.org/10.1007/s13399-022-03394-0.

[6] S. Ghosh Ray, M.M. Ghangrekar, Comprehensive review on treatment of high-strength distillery wastewater in advanced physico-chemical and biological degradation pathways, Int. J. Environ. Sci. Technol. 16 (2019) 527–546, https://doi.org/10.1007/s13762-018-1786-8.

[7] D.K. Amenorfenyo, X. Huang, C. Li, F. Li, Q. Zeng, N. Zhang, L. Xie, P. Wang, A review of microalgae and other treatment methods of distillery wastewater, Water Environ. J. 34 (2020) 988–1002, https://doi.org/10.1111/wej.12552.

[8] Y. Ravikumar, S.A. Razack, J. Yun, G. Zhang, H.M. Zabed, X. Qi, Recent advances in Microalgae-based distillery wastewater treatment, Environ. Technol. Innov. 24 (2021), 101839, https://doi.org/10.1016/j.eti.2021.101839.

[9] M. Mohajerani, M. Mehrvar, F. Ein-Mozaffari, Photoreactor design and CFD modelling of a UV/H2O2 process for distillery wastewater treatment, Can. J. Chem. Eng. 90 (2012) 719–729, https://doi.org/10.1002/cjce.20569.

[10] F.J. Beltrán, P.M. Álvarez, E.M. Rodríguez, J.F. García-Araya, J. Rivas, Treatment of high strength distillery wastewater (cherry stillage) by integrated aerobic biological oxidation and ozonation, Biotechnol. Prog. 17 (2001) 462–467, https://doi.org/10.1021/bp010021c.

[11] S. Naina Mohamed, R. Thota Karunakaran, M. Manickam, Enhancement of bioelectricity generation from treatment of distillery wastewater using microbial fuel cell, Environ. Prog. Sustain. Energy. 37 (2018) 663–668, https://doi.org/10.1002/ep.12734.

[12] V. Jaswal, Y. Kadapakkam Nandabalan, Rice husk-derived silicon nanostructured anode to enhance power generation in microbial fuel cell treating distillery wastewater, J. Environ. Manage. 328 (2023), 116912, https://doi.org/10.1016/j.jenvman.2022.116912.

[13] A. Wani, J.B. Bhasarkar, R.W. Gaikwad, Photocatalytic degradation of sugar and distillery industry effluent, J. Inst. Eng. Ser. E. 103 (2022) 79–86, https://doi.org/10.1007/s40034-020-00190-1.

[14] P.C. Sangave, A.B. Pandit, Ultrasound and enzyme assisted biodegradation of distillery wastewater, J. Environ. Manage. 80 (2006) 36–46, https://doi.org/10.1016/j.jenvman.2005.08.010.

[15] H. Patel, Characterization and adsorptive treatment of distillery spent wash using bagasse fly ash, Arab. J. Sci. Eng. 47 (2022) 5521–5531, https://doi.org/10.1007/s13369-021-05497-8.

[16] S.V. Krishna, P.K. Kumar, K. Verma, D. Bhagawan, V. Himabindu, M.L. Narasu, R. Singh, Enhancement of biohydrogen production from distillery spent wash effluent using electrocoagulation process, Energy Ecol. Environ. 4 (2019) 160–165, https://doi.org/10.1007/s40974-019-00122-9.

[17] I. Johnson, M. Kumar, Electrochemical oxidation of distillery wastewater by dimensionally stable Ti-RuO2 anodes, Environ. Technol. Innov. 20 (2020), 101181, https://doi.org/10.1016/j.eti.2020.101181.

[18] A.K. Prajapati, S. Mehra, T. Dewangan, D. Sharma, S. Sen, S. Dubey, R.K. Kaushal, P.K. Chaudhari, D. Pal, Treatment of rice grain based distillery biodigester effluent using iron metal and salt: chemical oxidation and electro-oxidation combined study in batch mode, Environ. Nanotechnol., Monit. Manag. 16 (2021), 100585, https://doi.org/10.1016/j.enmm.2021.100585.

[19] S. Talwar, A.K. Verma, V.K. Sangal, U.L. Štangar, Once through continuous flow removal of metronidazole by dual effect of photo-Fenton and photocatalysis in a compound parabolic concentrator at pilot plant scale, Chem. Eng. J. 388 (2020), 124184, https://doi.org/10.1016/j.cej.2020.124184.

[20] W. Zheng, Y. Liu, W. Liu, H. Ji, F. Li, C. Shen, X. Fang, X. Li, X. Duan, A novel electrocatalytic filtration system with carbon nanotube supported nanoscale zerovalent copper toward ultrafast oxidation of organic pollutants, Water Res. 194 (2021), 116961, https://doi.org/10.1016/j.watres.2021.116961.

[21] B. Sarı, S. Türkeş, H. Güney, O. Keskinkan, The utilization and modeling of photo-fenton process as a single unit in textile wastewater treatment, Clean – SoilAir, Water 51 (2023) 2100328, https://doi.org/10.1002/clen.202100328.

[22] A. Olabi, Ultrasound-assisted photo-Fenton process for treatment of pulp and paper mill wastewater and reduction of phytotoxicity, Water Environ. J. n/a (2023), https://doi.org/10.1111/wej.12841.

[23] S.A. Hien, C. Trellu, N. Oturan, A.S. Assémian, B.G.H. Briton, P. Drogui, K. Adouby, M.A. Oturan, Comparison of homogeneous and heterogeneous electrochemical advanced oxidation processes for treatment of textile industry wastewater, J. Hazard. Mater. 437 (2022), 129326, https://doi.org/10.1016/j.jhazmat.2022.129326.

[24] H. Zazou, H. Afanga, S. Akhouairi, H. Ouchtak, A.A. Addi, R.A. Akbour, A. Assabbane, J. Douch, A. Elmchaouri, J. Duplay, A. Jada, M. Hamdani, Treatment of textile industry wastewater by electrocoagulation coupled with electrochemical advanced oxidation process, J. Water Process Eng. 28 (2019) 214–221, https://doi.org/10.1016/j.jwpe.2019.02.006.

[25] R. Raj, A. Tripathi, S. Das, M.M. Ghangrekar, Removal of caffeine from wastewater using electrochemical advanced oxidation process: a mini review, Case Stud. Chem. Environ. Eng. 4 (2021), 100129, https://doi.org/10.1016/j.cscee.2021.100129.

[26] A. Dargahi, H. Rahimzadeh Barzoki, M. Vosoughi, S. Ahmad Mokhtari, Enhanced electrocatalytic degradation of 2,4-dinitrophenol (2,4-DNP) in three-dimensional sono-electrochemical (3D/SEC) process equipped with Fe/SBA-15 nanocomposite particle electrodes: degradation pathway and application for real wastewater, Arab. J. Chem. 15 (2022), 103801, https://doi.org/10.1016/j.arabjc.2022.103801.

[27] A. Raschitor, C.M. Fernandez, I. Cretescu, M.A. Rodrigo, P. Cañizares, Sono-electrocoagulation of wastewater polluted with Rhodamine 6G, Sep. Purif. Technol. 135 (2014) 110–116, https://doi.org/10.1016/j.seppur.2014.08.003.

[28] N. Tran, P. Drogui, S.K. Brar, Sonoelectrochemical oxidation of carbamazepine in waters: optimization using response surface methodology, J. Chem. Technol. Biotechnol. 90 (2015) 921–929, https://doi.org/10.1002/jctb.4399.

[29] A. Arka, C. Dawit, A. Befekadu, S.K. Debela, P. Asaithambi, Wastewater treatment using sono-electrocoagulation process: optimization through response surface methodology, Sustain. Water Resour. Manag. 8 (2022) 61, https://doi.org/10.1007/s40899-022-00649-6.

[30] F. Ghanbari, A. Hassani, S. Wacławek, Z. Wang, G. Matyszczak, K.Y.A. Lin, M. Dolatabadi, Insights into paracetamol degradation in aqueous solutions by ultrasound-assisted heterogeneous electro-Fenton process: key operating parameters, mineralization and toxicity assessment, Sep. Purif. Technol. 266 (2021), https://doi.org/10.1016/j.seppur.2021.118533.

[31] R. Patidar, V.C. Srivastava, Ultrasound-induced intensification of electrochemical treatment of bulk drug pharmaceutical wastewater, ACS ES&T Water. 1 (2021) 1941–1954, https://doi.org/10.1021/acsestwater.1c00159.

[32] F.C. Moreira, R.A.R. Boaventura, E. Brillas, V.J.P. Vilar, Electrochemical advanced oxidation processes: a review on their application to synthetic and real wastewaters, Appl. Catal. B Environ. 202 (2017) 217–261, https://doi.org/10.1016/j.apcatb.2016.08.037.

[33] M. Shestakova, M. Vinatoru, T.J. Mason, E. Iakovleva, M. Sillanpää, Sonoelectrochemical degradation of formic acid using Ti/Ta2O5-SnO2 electrodes, J. Mol. Liq. 223 (2016) 388–394, https://doi.org/10.1016/j.molliq.2016.08.054.

[34] M.A. Oturan, I. Sirés, N. Oturan, S. Pérocheau, J.L. Laborde, S. Trévin, Sonoelectro-Fenton process: a novel hybrid technique for the destruction of organic pollutants in water, J. Electroanal. Chem. 624 (2008) 329–332, https://doi.org/10.1016/j.jelechem.2008.08.005.

[35] P. Menon, T.S. Anantha Singh, N. Pani, P.V. Nidheesh, Electro-Fenton assisted sonication for removal of ammoniacal nitrogen and organic matter from dye intermediate industrial wastewater, Chemosphere 269 (2021), 128739, https://doi.org/10.1016/j.chemosphere.2020.128739.

[36] E. Keshmirizadeh, S. Yousefi, M.K. Rofouei, An investigation on the new operational parameter effective in Cr(VI) removal efficiency: a study on electrocoagulation by alternating pulse current, J. Hazard. Mater. 190 (2011) 119–124, https://doi.org/10.1016/j.jhazmat.2011.03.010.

[37] K.E. Adou, A.R. Kouakou, A.D. Ehouman, R.D. Tyagi, P. Drogui, K. Adouby, Coupling anaerobic digestion process and electrocoagulation using iron and aluminium electrodes for slaughterhouse wastewater treatment, Sci. African 16 (2022) e01238.

[38] S. Şahinkaya, COD and color removal from synthetic textile wastewater by ultrasound assisted electro-Fenton oxidation process, J. Ind. Eng. Chem. 19 (2013) 601–605, https://doi.org/10.1016/j.jiec.2012.09.023.

[39] N. Mahmoudi, M. Farhadian, A.R. Solaimany Nazar, P. Eskandari, K.N. Esfahani, Investigation and optimization of the performance of sono-photo-electro-Fenton process for removal of Acid Black 172 and Disperse Blue 56 from polluted water: comparison of the degradation activity with electro-Fenton-based processes, Int. J. Environ. Sci. Technol. 19 (2022) 1671–1682, https://doi.org/10.1007/s13762-021-03296-0.

[40] P. Asaithambi, R. Govindarajan, M.B. Yesuf, E. Alemayehu, Removal of color, COD and determination of power consumption from landfill leachate wastewater using an electrochemical advanced oxidation processes, Sep. Purif. Technol. 233 (2020), 115935, https://doi.org/10.1016/j.seppur.2019.115935.

[41] M. Amarzadeh, S. Salehizadeh, S. Damavandi, N.M. Mubarak, M. Ghahrchi, B. Ramavandi, Y.D. Shahamat, N. Nasseh, Statistical modeling optimization for antibiotics decomposition by ultrasound/electro-Fenton integrated process: non-carcinogenic risk assessment of drinking water, J. Environ. Manage. 324 (2022), 116333, https://doi.org/10.1016/j.jenvman.2022.116333.

[42] A.Y. Ghjair, A.H. Abbar, Applications of advanced oxidation processes (Electro-Fenton and sono-electro-Fenton) for COD removal from hospital wastewater: optimization using response surface methodology, Process Saf. Environ. Prot. 169 (2023) 481–492, https://doi.org/10.1016/j.psep.2022.11.039.

[43] K. Hasani, A. Peyghami, A. Moharrami, M. Vosoughi, A. Dargahi, The efficacy of sono-electro-Fenton process for removal of Cefixime antibiotic from aqueous solutions by response surface methodology (RSM) and evaluation of toxicity of effluent by microorganisms, Arab. J. Chem. 13 (2020) 6122–6139, https://doi.org/10.1016/j.arabjc.2020.05.012.

[44] N. Yousefi, S. Pourfadakari, S. Esmaeili, A.A. Babaei, Mineralization of high saline petrochemical wastewater using Sonoelectro-activated persulfate: degradation mechanisms and reaction kinetics, Microchem. J. 147 (2019) 1075–1082, https://doi.org/10.1016/j.microc.2019.04.020.

[45] P. Asaithambi, M.B. Yesuf, R. Govindarajan, N.M. Hariharan, P. Thangavelu, E. Alemayehu, Distillery industrial wastewater(DIW) treatment by the combination of sono(US), photo(UV) and electrocoagulation(EC) process, J. Environ. Manage. 320 (2022), 115926, https://doi.org/10.1016/j.jenvman.2022.115926.

[46] A.R.A. Aziz, P. Asaithambi, W.M.A.B.W. Daud, Combination of electrocoagulation with advanced oxidation processes for the treatment of distillery industrial effluent, Process Saf. Environ. Prot. 99 (2016) 227–235, https://doi.org/10.1016/j.psep.2015.11.010.

[47] M.A. García-Morales, G. Roa-Morales, C. Barrera-Díaz, B. Bilyeu, M.A. Rodrigo, Synergy of electrochemical oxidation using boron-doped diamond (BDD)

electrodes and ozone (O3) in industrial wastewater treatment, Electrochem. Commun. 27 (2013) 34–37, https://doi.org/10.1016/j.elecom.2012.10.028.

[48] A. Dargahi, M. Moradi, R. Marafat, M. Vosoughi, S.A. Mokhtari, K. Hasani, S. M. Asl, Applications of advanced oxidation processes (electro-Fenton and sono-electro-Fenton) for degradation of diazinon insecticide from aqueous solutions: optimization and modeling using RSM-CCD, influencing factors, evaluation of toxicity, and degradation pat, Biomass Convers. Biorefinery. (2021), https://doi.org/10.1007/s13399-021-01753-x.

[49] A. Somayajula, P. Asaithambi, M. Susree, M. Matheswaran, Sonoelectrochemical oxidation for decolorization of Reactive Red 195, Ultrason. Sonochem. 19 (2012) 803–811, https://doi.org/10.1016/j.ultsonch.2011.12.019.

[50] R. Patidar, V.C. Srivastava, Ultrasound enhanced electro-Fenton mineralization of benzophenone: kinetics and mechanistic analysis, ACS ES T. Water (2022) 1–18, https://doi.org/10.1021/acsestwater.2c00364.

[51] B. Thokchom, K. Kim, J. Park, J. Khim, Ultrasonically enhanced electrochemical oxidation of ibuprofen, Ultrason. Sonochem. 22 (2015) 429–436, https://doi.org/10.1016/j.ultsonch.2014.04.019.

[52] M. Ren, Y. Song, S. Xiao, P. Zeng, J. Peng, Treatment of berberine hydrochloride wastewater by using pulse electro-coagulation process with Fe electrode, Chem. Eng. J. 169 (2011) 84–90, https://doi.org/10.1016/j.cej.2011.02.056.

[53] M.S. Bhatti, A.S. Reddy, A.K. Thukral, Electrocoagulation removal of Cr(VI) from simulated wastewater using response surface methodology, J. Hazard. Mater. 172 (2009) 839–846, https://doi.org/10.1016/j.jhazmat.2009.07.072.

[54] S. Aber, A.R. Amani-Ghadim, V. Mirzajani, Removal of Cr(VI) from polluted solutions by electrocoagulation: modeling of experimental results using artificial neural network, J. Hazard. Mater. 171 (2009) 484–490, https://doi.org/10.1016/j.jhazmat.2009.06.025.

[55] S. Meddah, M. El Hadi Samar, M. Bououdina, L. Khezami, Outstanding performance of electro-Fenton/ultra-violet/ultra-sound assisted-persulfate process for the complete degradation of hazardous pollutants in contaminated water, Process Saf. Environ. Prot. 165 (2022) 739–753, https://doi.org/10.1016/j.psep.2022.08.002.

[56] S. Vasudevan, An efficient removal of phenol from water by peroxi-electrocoagulation processes, J. Water Process. Eng. 2 (2014) 53–57, https://doi.org/10.1016/j.jwpe.2014.05.002.

[57] S. Yazici Guvenc, G. Varank, E. Can-Güven, H. Ercan, D. Yaman, E. Saricam, O. K. Türk, Application of the hybrid electrocoagulation–electrooxidation process for the degradation of contaminants in acidified biodiesel wastewater, J. Electroanal. Chem. 926 (2022), 116933, https://doi.org/10.1016/j.jelechem.2022.116933.

[58] M. Teymori, H. Khorsandi, A.A. Aghapour, S.J. Jafari, R. Maleki, Electro-Fenton method for the removal of Malachite Green: effect of operational parameters, Appl. Water Sci. 10 (2019) 39, https://doi.org/10.1007/s13201-019-1123-5.

[59] S. Fekadu, E. Alemayehu, P. Asaithambi, B. Van der Bruggen, Removal of phosphate from the healthcare wastewater through peroxi-photoelectrocoagulation process: effect of process parameters, Int. J. Environ. Res. 16 (2022) 8, https://doi.org/10.1007/s41742-021-00388-0.

[60] V.K. Sandhwar, B. Prasad, Comparison of electrocoagulation, peroxi-electrocoagulation and peroxi-coagulation processes for treatment of simulated purified terephthalic acid wastewater: optimization, sludge and kinetic analysis,

Korean J. Chem. Eng. 35 (2018) 909–921, https://doi.org/10.1007/s11814-017-0336-2.

[61] A. Dargahi, D. Nematollahi, G. Asgari, R. Shokoohi, A. Ansari, M.R. Samarghandi, Electrodegradation of 2,4-dichlorophenoxyacetic acid herbicide from aqueous solution using three-dimensional electrode reactor with G/β-PbO2 anode: Taguchi optimization and degradation mechanism determination, RSC Adv. 8 (2018) 39256–39268, https://doi.org/10.1039/C8RA08471H.

[62] M. Moradi, Y. Vasseghian, H. Arabzade, A. Mousavi Khaneghah, Various wastewaters treatment by sono-electrocoagulation process: a comprehensive review of operational parameters and future outlook, Chemosphere 263 (2021) 128314. 10.1016/j.chemosphere.2020.128314.

[63] C.A. Igwegbe, O.D. Onukwuli, J.O. Ighalo, C.J. Umembamalu, Electrocoagulation-flocculation of aquaculture effluent using hybrid iron and aluminium electrodes: a comparative study, Chem. Eng. J. Adv. 6 (2021), 100107, https://doi.org/10.1016/j.ceja.2021.100107.

[64] A. Babuponnusami, K. Muthukumar, Advanced oxidation of phenol: a comparison between Fenton, electro-Fenton, sono-electro-Fenton and photo-electro-Fenton processes, Chem. Eng. J. 183 (2012) 1–9, https://doi.org/10.1016/j.cej.2011.12.010.

[65] S. Abbasi, A.A. Zinatizadeh, M. Mirghorayshi, S. Zinadini, T. McKay, Electrocoagulation technique for continuous industrial licorice processing wastewater treatment in a single reactor employing Fe-rod electrodes: process modeling and optimization and operating cost analysis, J. Environ. Chem. Eng. 10 (2022), 106686, https://doi.org/10.1016/j.jece.2021.106686.

[66] P. Patel, S. Gupta, P. Mondal, Electrocoagulation process for greywater treatment: statistical modeling, optimization, cost analysis and sludge management, Sep. Purif. Technol. 296 (2022), 121327, https://doi.org/10.1016/j.seppur.2022.121327.

[67] M. Bayramoglu, M. Kobya, O.T. Can, M. Sozbir, Operating cost analysis of electrocoagulation of textile dye wastewater, Sep. Purif. Technol. 37 (2004) 117–125, https://doi.org/10.1016/j.seppur.2003.09.002.

[68] C. David, M. Arivazhagan, F. Tuvakara, Decolorization of distillery spent wash effluent by electro oxidation (EC and EF) and Fenton processes: a comparative study, Ecotoxicol. Environ. Saf. 121 (2015) 142–148, https://doi.org/10.1016/j.ecoenv.2015.04.038.

[69] A. Thanapimmetha, P. Srinophakun, S. Amat, M. Saisriyoot, Decolorization of molasses-based distillery wastewater by means of pulse electro-Fenton process, J. Environ. Chem. Eng. 5 (2017) 2305–2312, https://doi.org/10.1016/j.jece.2017.04.030.

[70] S. Dubey, A. Joshi, R. Trivedi, D. Pal, A.K. Prajapati, Electro-peroxone treatment of rice grain based distillery biodigester effluent: COD and color removal, Water Resour. Ind. 25 (2021), 100142, https://doi.org/10.1016/j.wri.2021.100142.

[71] M.M. Arimi, Modified natural zeolite as heterogeneous Fenton catalyst in treatment of recalcitrants in industrial effluent, Prog. Nat. Sci. Mater. Int. 27 (2017) 275–282, https://doi.org/10.1016/j.pnsc.2017.02.001.

[72] M. Coca, M. Peña, G. González, Chemical oxidation processes for decolorization of brown-colored molasses wastewater, Ozone Sci. Eng. 27 (2005) 365–369, https://doi.org/10.1080/01919510500250689.