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Research article

Distillery industrial wastewater(DIW) treatment by the combination of sono (US), photo(UV) and electrocoagulation(EC) process

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ABSTRACT

The color and Chemical Oxygen Demand (COD) reduction in distillery industrial effluent (DIW) was investigated utilizing photo (UV), sono (US), electrocoagulation (EC), UV + US, UV + EC, US + EC, and US + UV + EC technologies. The empirical study demonstrated that the UV + US + EC process removed almost 100% of color and 95.63% of COD from DIW while consuming around 6.97 kWh m^{-3} of electrical energy at the current density of 0.175 A dm^{-2}, COD of 3600 mg L^{-1}, UV power of 32 W, US power of 100 W, electrode pairings of Fe/Fe, inter–electrode distance of 0.75 cm, pH of 7, and reaction time of 4 h, respectively. The values found were much greater than those produced using UV, US, EC, UV + US, UV + EC, and US + EC methods. The influence of various control variables such as treatment time (1–5 h), current density (0.075–2.0 A dm^{-2}), COD (1800–6000 mg L^{-1}), inter-electrode distance (0.75–3.0 cm), electrode pairings (Fe/Fe, Fe/Al, Al/Fe, Al/Al), UV (8–32 W), and US (20–100 W) on the color and COD reduction were investigated to determine the optimum operating conditions. It was observed that, an increase in treatment time, current density, UV and US power, decrease in the COD, and inter-electrode distance with Fe/Fe electrode combination improved the COD removal efficiency. The UV and US + EC processes' synergy index was investigated and reported. The results showed that, the US + UV + EC treatment combination was effective in treating industrial effluent and wastewater.

1. Introduction

Numerous organizations have shifted their focus in recent years toward non-chemical treatments that are more sustainable, such as electrochemistry treatment technology, due to its advantages over conventional chemical treatments. Electrocoagulation (EC) is one such example of this technology, which is employed in place of conventional physical–chemical processes (Cotillas et al., 2020; Deghles and Kurt, 2016). This cutting-edge treatment approach often provides superior benefits to conventional chemical coagulation systems without the associated costs and adverse effects (Deghles and Kurt, 2016).

The EC process has gained recognition as a viable and successful method of treating effluent and wastewater from a wide variety of industries (Chezeau et al., 2020; Moussavi et al., 2021; Nigri et al., 2020; Preethi et al., 2020). The EC has the ability to remove and decrease undesirable contaminants/pollutants from a variety of sources, including wastewater and industrial effluents (Nigri et al., 2020). Such contaminants/pollutants include suspended and dissolved solids, pathogens, organic and inorganic chemicals, etc (Romani et al., 2020). EC units are not complicated to install and do not require a large amount of material, and chemical addition are simple to operate, require a short startup period, are easy to control, have a high capacity for pollutant removal, facilitate the collection of produced sludge, and are relatively inexpensive and easy to acquire (Deghles and Kurt, 2016; Raschitor et al., 2014). The electrodes that are frequently utilized in an EC system are constructed of iron (Fe) or aluminum (Al), both of which are readily available (Chen et al., 2020; Chezeau et al., 2020; Li et al., 2021).

The primary disadvantages of the EC process are the high anode

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consumption, electrode passivation on the cathode which results in reduction of current, and process and pollutant removal efficiency, and increased electrical energy consumption (EEC) (Al-Qodah et al., 2020b). Additionally, if not properly managed, the generated sludge may contain a variety of components, including potentially dangerous compounds to the environment (Romani et al., 2020). The EC technology as a stand-alone treatment process may have significant practical constraints, particularly if the wastewater is extremely polluted/contaminated (Aziz et al., 2016). Hence, an efficient and relatively inexpensive treatment techniques are required. In response to this, various studies have demonstrated, combining EC with other treatment methods such as advanced oxidation processes (AOPs) improve its performance.

Numerous research studies have demonstrated that EC works cohesively with other treatment modalities, including ozone (O_3) assisted EC (Al-Qodah and Al-Shannag, 2019; Das et al., 2021; He et al., 2007; Mehralian et al., 2021), EC/Adsorption, EC/Chemical coagulation, EC/Magnetic field, EC/reverse osmosis (Al-Qodah et al., 2020b), US-EC (Al-Qodah and Al-Shannag, 2019; Emerick et al., 2020; Moradi et al., 2021), alternating current EC (Alimohammadi et al., 2019; Payami Shabestar et al., 2021), peroxi–EC (Sandhwar and Prasad, 2018), photo–EC (Asaithambi et al., 2016; Farhadi et al., 2012; Moradi and Moussavi, 2019), aerated-EC (Akansha et al., 2020), EC-nanofiltration and membrane filtration (Gönder et al., 2020; Tavangar et al., 2019; Ucevli and Kaya, 2021), and biological treatment with EC (Al-Qodah et al., 2020a) for the elimination of contaminants from industrial effluent and wastewater.

Farhadi et al. (2012) compared the various processes for removal of COD from pharmaceutical wastewater. Their findings show that the following order of COD removal efficiency was established under optimal operating conditions for each process: peroxi-EC > peroxi-photo-EC > photo- EC > EC. Cotillas et al. (2014) reported that, when UV irradiation and electrocoagulation (UV-EC) were utilized at low current densities, a synergistic effect was shown for the reduction of turbidity and disinfection rate. Additionally, they observed that performance improves when UV radiation is used to promote the creation of hydroxyl and chlorine radicals. Jallouli et al. (2020) showed that the sequential EC and UV treatment processes were effective at reducing COD in tannery effluent, and their results indicated that the combined procedure reduced COD by 94.1%, compared to 85.7% and 55.9% for the individual EC and UV treatments respectively.

According to Özyonar et al. (2020) the removal of color and COD from aqueous dye solutions by US + EC process was significantly more than by US or EC alone. Additionally, they discovered that when US irradiation was combined with EC treatment, electrode passivation was greatly reduced compared to the EC method. In a more recent study by Prajapati (2021) performed a performance and cost analysis on the removal of COD and color from BDE using US, EC, and US + EC. Their findings indicated that when compared to US and EC alone, the US + EC process had much greater COD (99.1%) and color (61.6%) removal efficiencies with a 0.58 kWh m^{-3} energy consumption. Emerick et al. (2020) established that the ECF and ECF/US procedures are viable options for wastewater treatment. Oza et al. (2021) demonstrated that integrating US and EC was an effective treatment method for the removal of hazardous contaminants such as arsenic.

Our meticulous analysis of the literature established that hybrid approaches based on electrochemical and AOPs were effective at removing contaminants from synthetic wastewater (Chanikya et al., 2021; Zazou et al., 2019). Meanwhile, only a few studies have focused on real wastewater and industrial effluent utilizing hybrid electrochemical and AOPs (da Costa et al., 2019; Valero et al., 2017). Additionally, apart from the removal of contaminants from industrial effluent and wastewater, the hybrid process's EEC is critical from an economic standpoint. Moreover, environmental engineers must create and develop simple, efficient, and cost-effective hybrid procedures that address the shortcomings of conventional treatment systems.

To the authors' awareness, no previous research has been conducted on the combination of UV and US with an EC procedure for determining the EEC associated with the color and COD reduction from DIW. The purpose of this investigation was to create and develop novel treatment strategies for determining EEC while removing contaminants from DIW. The main objective of this work is to compare UV, US, EC, UV + US, UV + EC, US + EC, and US + UV + EC processes in terms of color and COD reduction, as well as to determine the EEC from DIW and to choose the best one. The effect of process parameters such as treatment time (1–5 h), current density (0.075–2.0 A dm^{-2}), COD (1800–6000 mg L^{-1}), inter-electrode distance (0.75–3.0 cm), combination electrode (Fe/Fe, Fe/Al, Al/Fe, Al/Al), UV (8–32 W), and US (20–100 W) on the COD removal and EEC of DIW treated with a UV + US + EC process was investigated. Additionally, the synergy index between UV and US/EC procedures was evaluated and reported.

2. Material and methods

2.1. Wastewater collection

The DIW used in this investigation was obtained from distilleries in Erode, Tamil Nadu, India. The grab sampling technique was employed for sampling. To ensure against any degradation in wastewater quality, the effluent was preserved in a cold room (+4 °C). The chemicals used in the experiments were H_2SO_4, NaOH, $K_2Cr_2O_7$, $Na_2S_2O_3$, $(NH_4)_2Fe(SO_4)_2$, etc. Merck, India supplied the analytical reagent (AR) grade chemicals and the chemicals were utilized in their original state without further purification.

2.2. Experimental setup of hybrid sono–photo–electrocoagulation (US + UV + EC) process

2.2.1. Electrocoagulation (EC)

For the treatment of DIW, the experimental setup scheme for the integrated US + UV + EC is depicted in Fig. 1a–c. The US, UV, and EC process units were used in the integrated process. The EC (Fig. 1a) was carried out in a batch electrochemical reactor composed of an acrylic sheet with a 2.5 L capacity. Each experiment was run with a 2.0 L DIW operational capacity. The anode/cathode electrodes were made of Fe/Fe, Al/Al, Fe/Al, and Al/Fe. Both electrodes had the same dimensions with 0.1 cm thickness and had an effective electrode surface area of 10 cm × 15 cm (width x height). The anodes and cathodes were coupled in parallel to a direct current (DC) power supply (APLAB Ltd; Model L1606). Before each experiment, the electrodes used in the processes were manually cleaned with a 35% HCl solution, washed with distilled water, and dried. The pH of effluent was measured using a pH meter (Elico: Model LI120) and adjusted with H_2SO_4 and NaOH solution. Using the direct current (DC) power source, a continuous DC was supplied across the electrodes when the required experimental condition was achieved.

2.2.2. Sono-electrocoagulation (US + EC)

Before beginning the ultrasonication process, the bath (Elma Ultrasonics type d-7822 K) was filled with distilled water to the recommended level. The ultrasonic water bath's distilled water was refilled before each experiment began. The EC reactor system was submerged in ultrasonic water bath to begin the US + EC process (Fig. 1b).

2.2.3. Sono-photo-electrocoagulation (US + UV + EC)

The operating conditions for the experiment were similar to those for the US + EC process, but the reaction contents were further exposed to UV irradiation. As a UV light source, an 8–32 Watts low–pressure mercury lamp emitting primarily at 254 nm was used. Due to the fact that the UV lamp was incorporated into the US + EC process, the procedure is referred to as US + UV + EC (Fig. 1c). Throughout the EC, US + EC, and US + UV + EC processes, the cell voltage and current are monitored using a multimeter. Regularly timed samples were collected from the

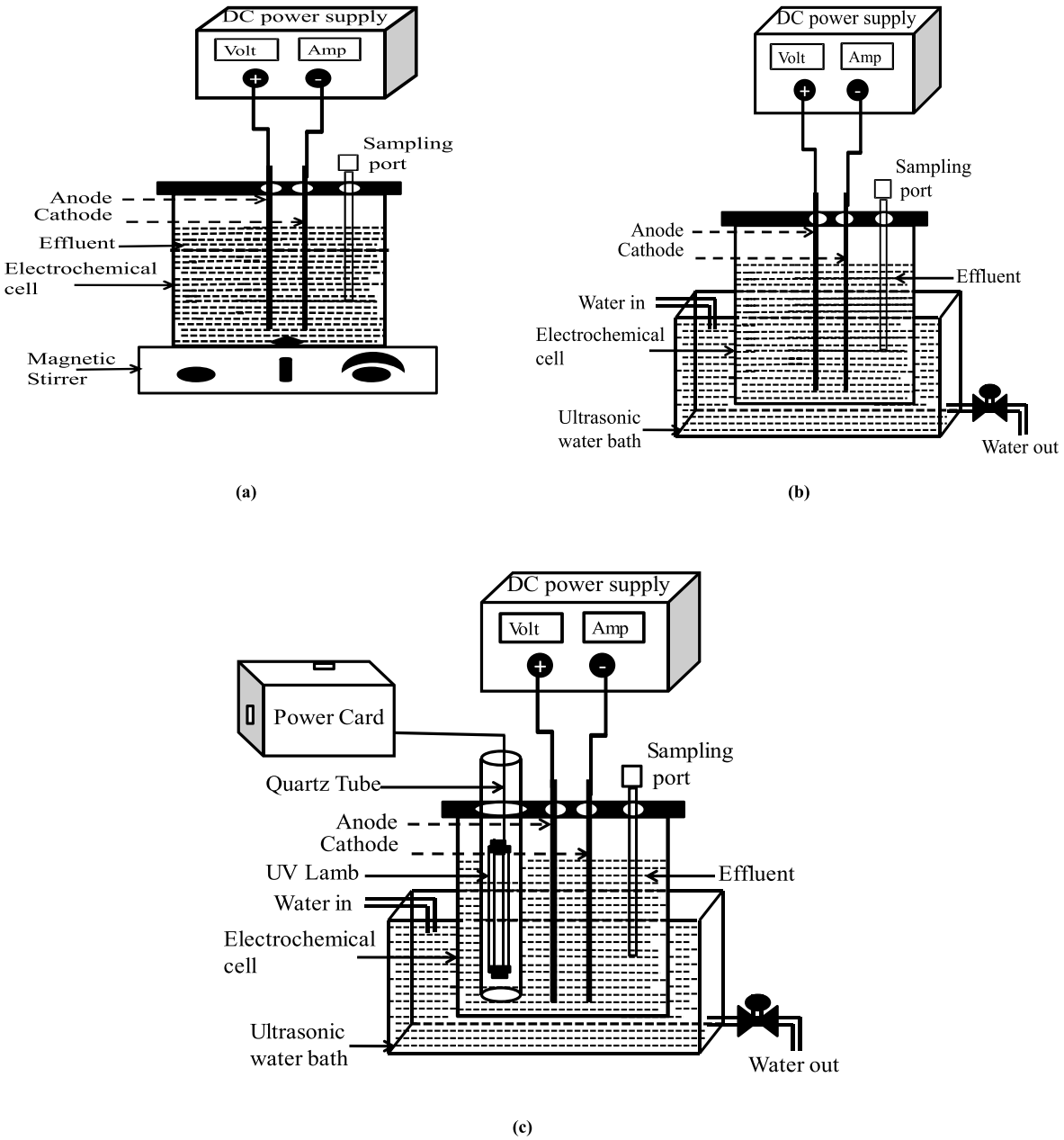


Fig. 1. a, b & c. Experimental setup of EC, US + EC & hybrid US + UV + EC process.

reactor and added to an Erlenmeyer flask holding $Na_2S_2O_3$ solution to quench the process. After centrifuging the samples at 10,000 rpm for 15 min to separate the solid particles and liquid, the supernatant liquid is immediately analyzed for COD (Spectroquant ® TR320) and color (Spectroquant Pharo ®300).

2.3. Analysis

2.3.1. Color and COD removal efficiency, (%)

The color and COD reduction efficiencies were determined using the following equations (1) and (2).

The color was determined using a UV/Vis spectrophotometer at the wavelength corresponding to the maximal absorbance λ_{max} (300 nm) (Spectroquant Pharo ®300).

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

where, Abs_i and Abs_t are the absorbances of the DIW at the corresponding wavelength (λ_{max}) before and after the treatment process.

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

where, COD_i and COD_t are the COD (mg L^{-1}) of DIW prior to and following the after treatment, respectively.

2.3.2. Electrical energy consumption (EEC)

The removal efficiency of color and COD along with EEC is evaluated for the comparison of UV, US, EC, UV + US, UV + EC, US + EC, and US + UV + EC process to DIW. The EEC–kWh m^{-3}) of UV, US, and EC primarily dominated the treatment cost of the integrated system. The equation was listed as follows.

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

where, $EEC_{UV/US/EC}$ EEC_{UV}, EEC_{US} and EEC_{EC} are electrical energy consumption for UV + US + EC, UV, US and EC process.

2.3.3. Electrocoagulation (EC)

$$EEC_{EC} = \frac{U\ x\ I\ x\ t}{V_R},\ \left(kWhr\ m^{-3}\right) \quad (4)$$

where, U represents the cell voltage (V), I represent the current (A), t represents the reaction time (h) and V_R represents the volume of DIW utilized.

2.3.4. Photo (UV) and sonication (US)

$$EEC_{UV} + EEC_{US} = \frac{P_{et}\ x\ t\ x\ 1000}{V_R\ x\ 60\ x \log\left(\frac{COD_f}{COD_i}\right)},\ \left(kWh\ m^{-3}\right) \quad (5)$$

where, P_{el} denotes the rated power in kW (0.008, 0.016, and 0.032 kW for UV and 0.02, 0.04, 0.06, 0.08 and 0.100 kW for sonication).

3. Results and discussion

3.1. Wastewater characterization

The distillery industrial wastewater was characterized using standard procedures. The DIW had the odor of burnt sugar, a dark brown color, a pH of 4.1–4.3, a Biochemical Oxygen Demand (BOD) of 7000–8000 mg L^{-1}, a Chemical Oxygen Demand (COD) of 80,000–90,000 mg L^{-1}, a Total Dissolved Solids (TDS) of 5550–5750 mg L^{-1}, and a Total Suspended Solids (TSS) of 15.44 g L^{-1}.

3.2. Studies on operating parameters

3.2.1. Optimization of US + UV + EC process

To optimize the US + UV + EC process performance, the impacts of treatment time, current density, COD, UV and US power, electrode combination, and inter-electrode distance were explored and addressed in detail below.

3.2.1.1. Treatment time. One of the most essential factors for industrial wastewater treatment employing hybrid procedures is the effect of treatment time (Maha Lakshmi and Sivashanmugam, 2013). The influence of reaction time on removal of COD and EEC from DIW using US + UV + EC process with the current density of 0.75 A dm^{-2}, inter-electrode distance of 0.75 cm, COD of 3600 mg L^{-1}, pH of 7, electrode pairing of Fe/Fe, UV of 32 W and US of 100 W the results are depicted in Fig. 2a. Fig. 2a indicates that, the % COD removal and EEC has increased with an increase in treatment time. The COD removed is proportional to the concentration of ions generated by the electrodes. When the treatment time increases, a constant amount of electrodes ions and their oxidizing species are produced. Consequently, an increase in the treatment time increases the COD removal efficiency. Increased EEC via cell voltage results in an increase in hydroxyl radicals, which improves the COD removal.

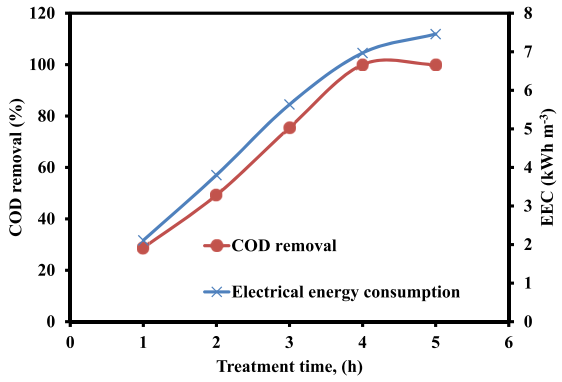
3.2.1.2. Current density. The current density is an important factor in the hybrid EC process for the treatment of industrial effluent (Mehralian et al., 2021; Moradi et al., 2021; Sandhwar and Prasad, 2018). The current density was varied between 0.07 and 0.20 A dm^{-2} in this investigation for the US + UV + EC system utilizing the DIW, and the findings are depicted in Fig. 2b. As illustrated in Fig. 2b, the EEC of the hybrid US + UV + EC process has increased from 3.11 to 7.01 kWh m^{-3} and the COD removal efficiency has increased from 37.84 to 95.63% for an increase in current density from 0.07 to 0.175 A dm^{-2} at constant treatment time of 4 h. According to Faraday's rule, the quantity of Fe dissolved by electrochemical reaction is proportionate to the charge loadings on the surface of electrode that promote flocculation process (Dizge et al., 2018). Generally, increasing current density results in an increase in coagulant formation with anode dissolution and H_2 gas production via water reduction at the cathode (Farhadi et al., 2012). Enormous amounts of coagulants increase the efficacy of COD removal via the adsorption and precipitation processes. Additionally, H_2 gas coalesces the lighter particles, resulting in an increase in the COD removal efficiency of pollutants via the flotation process (Nawarkar and Salkar, 2019; Negarestani et al., 2020). Additionally, as illustrated in Fig. 2b, after increasing the current density from 0.175 to 0.200 A dm^{-2}, EEC increased from 7.01 to 8.89 kWh m^{-3}, but COD removal efficiency declined from 95.63 to 91.60%. This might account for the production of necessary coagulants at a current density of 0.175 A dm^{-2}, and hence current density of 0.175 A dm^{-2} are considered to be the optimum values for a 3600 mg L^{-1} concentration of DIW.

According to Fig. 2b, the overall EEC increased from 3.11 to 8.89 kWh m^{-3} with an increase in current density from 0.075 to 0.20 A dm^{-2}. This could be because current density is directly related to cell voltage (Dizge et al., 2018).

3.2.1.3. COD. For wastewater and industrial effluent treatment, the initial pollutant concentration influences the hybrid EC process (Moradi et al., 2021). The US + UV + EC process was evaluated for its efficiency in removing COD and EEC from DIW by adjusting the initial COD concentration from 1800 to 6000 mg L^{-1}, as shown in Fig. 2c. The COD removal efficiency of 100%, 95.63%, 79.84%, 63.04%, and 49.60% were reached with initial COD concentrations of 1800, 3600, 4400, 5200, and 6000 mg L^{-1}, respectively. As the initial COD concentration raised from 1800 to 6000 mg L^{-1}, the EEC and COD removal rate dropped; hence, a longer time was required to attain a constant COD removal efficiency during the treatment process. This is because consistent current density and treatment time create the same amount of metal hydroxide (Moradi et al., 2021). This quantity is insufficient to coagulate and flocculate the polluted waters with increased COD concentration (Ahlawat et al., 2008). As a result, the amount of $Fe(OH)_3$ coagulant produced is insufficient for sedimentation as the COD concentration increases. Excessive surface oxidation reduces anode electrode's ability to release metal ions and generate hydroxyl radicals (Moradi et al., 2021). As a result, the efficiency with which COD is removed decreases.

3.2.1.4. UV power. During the US + EC process, additional UV irradiation was carried out. As illustrated in Fig. 2d, a range of UV power from 8 to 32 W was investigated using UV-C lamps during a 4 h US + UV + EC procedure for DIW. The efficiency of COD removal was enhanced from 63.04 to 95.63%, the EEC was increased from 4.16 to 6.97 kWh m^{-3}, and the UV power was increased from 8 to 32 W, respectively. Increases in lamp power and light source irradiation intensity per unit area result in the production of additional hydroxyl radicals (Keramati and Ayati, 2019). The increased availability of photoactive sites had a favorable influence on the US + EC. This improvement was attributed to increased $^{\bullet}OH$ generation via photoreduction and photodecomposition reaction (Aziz et al., 2016). This increases the efficiency of COD removal and electrical energy usage in the US + UV + EC process for DIW.

3.2.1.5. US power. Changed intensities of US have an important effect on the pollutant elimination by using US + EC process for treatment of industrial effluent (Dizge et al., 2018). In a series of studies, the influence of US power on EEC and COD elimination efficiency was investigated using US power ranging from 20 to 100 W for DIW utilizing the US + UV + EC method. The findings are illustrated in Fig. 2e. By increasing the US power, the COD removal efficiency has enhanced from 83.20 to 95.56% and the EEC was increased from 5.98 to 6.97 kWh m^{-3}, respectively. The passive layer became thinner as the US intensity increased, reducing resistance and enhancing the energy efficiency of



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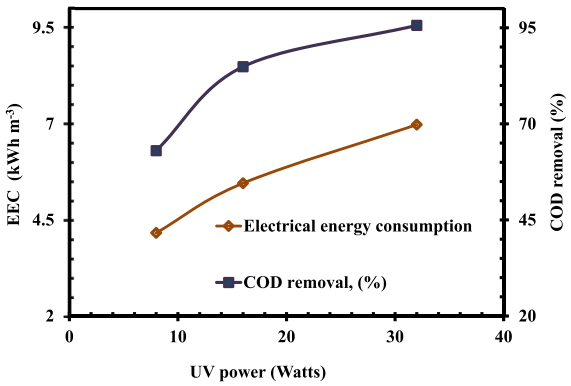

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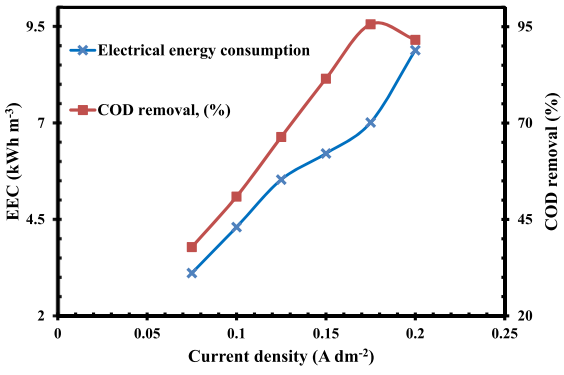

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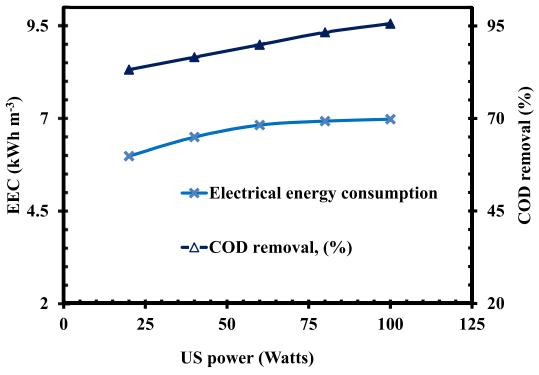

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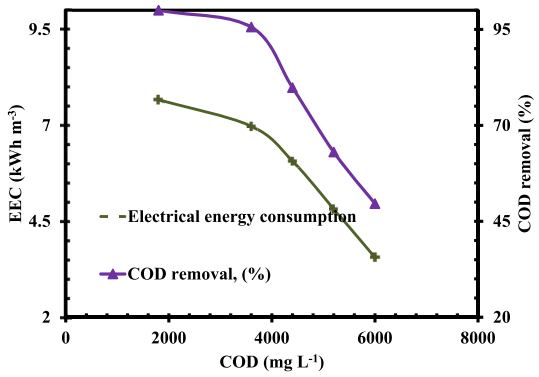

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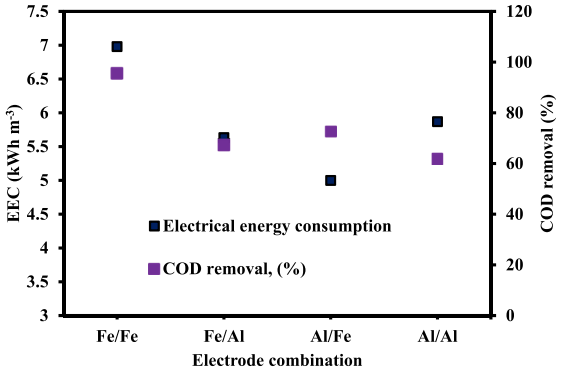

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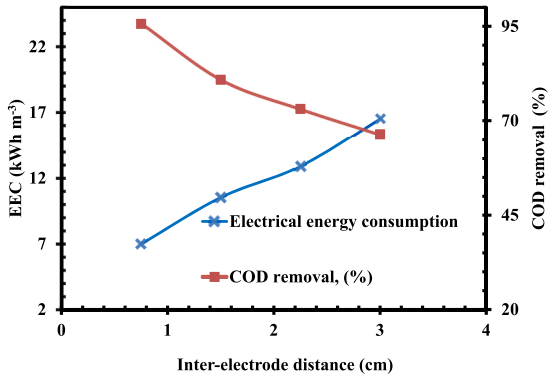

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Fig. 2. a. Effect of treatment time on COD removal and EEC by hybrid US + UV + EC process (current density = 0.175 A dm^{-2}, COD = 3600 mg L^{-1}, UV = 32 W, US = 100 W, electrode combination = Fe/Fe, inter–electrode distance = 0.75 cm and pH = 7). b. Effect of current density on EEC and COD removal by hybrid US + UV + EC process (COD = 3600 mg L^{-1}, UV = 32 W, US = 100 W, electrode combination = Fe/Fe, inter–electrode distance = 0.75 cm, pH = 7, and reaction time = 4 h). c. Effect of COD concentration on EEC and COD removal by hybrid US + UV + EC process (current density = 0.175 A dm^{-2}, UV = 32 W, US = 100 W, electrode combination = Fe/Fe, inter–electrode distance = 0.75 cm, pH = 7, and reaction time = 4 h). d. Effect of UV power on EEC and COD removal by hybrid US + UV + EC process (current density = 0.175 A dm^{-2}, COD = 3600 mg L^{-1}, US = 100 W, electrode combination = Fe/Fe, inter–electrode distance = 0.75 cm, pH = 7, and reaction time = 4 h). e. Effect of US power on EEC and COD removal by hybrid US + UV + EC process (current density = 0.175 A dm^{-2}, COD = 3600 mg L^{-1}, UV = 32 W, electrode combination = Fe/Fe, inter–electrode distance = 0.75 cm, pH = 7, and reaction time = 4 h). f. Effect of electrode combination on EEC and COD removal by hybrid US + UV + EC process (current density = 0.175 A dm^{-2}, COD = 3600 mg L^{-1}, UV = 32 W, US = 100 W, inter–electrode distance = 0.75 cm, pH = 7, and reaction time = 4 h). g. Effect of inter-electrode distance on EEC and COD removal by hybrid US + UV + EC process (current density = 0.175 A dm^{-2}, COD = 3600 mg L^{-1}, UV = 32 W, US = 100 W, electrode combination = Fe/Fe, pH = 7, and reaction time = 4 h).

the US + UV + EC process. As a result, the ultrasonic procedure effectively eliminates the passive coating, resulting in increased coagulant metal release (He et al., 2016). Li et al. (2013) observed similar results for the phosphate removal efficiency by comparing the US, EC processes individually and in combinations. They found that the combined US + EC system had a greater phosphate removal efficiency than the efficiency of EC and US individual systems added together. This demonstrates the synergistic effect of integrating the US and EC process.

3.2.1.6. Electrode combination/pairing. The electrode material selected is one of the EC control parameters that affects not only the process's performance and efficiency, but also its running cost (Moradi et al., 2021). In a hybrid system, the nature and extent of electrodes have a significant impact on the treatment system's capacity and the pace at which pollutants are eliminated. Due to their availability, low cost, and capacity to generate stable hydroxyl species, Fe and Al electrodes have been utilized more frequently than other electrodes for the elimination of contaminants from wastewater (Igwegbe et al., 2021).

The influence of electrode types on COD removal and EEC was investigated using two anode and cathode materials, Fe and Al, under constant experimental conditions, including current density – 0.175 A dm^{-2}, initial pH – 7, COD – 3600 mg L^{-1}, electrode distance – 0.75 cm, UV – 32 W, US – 100 W, and reaction time – 4 h for DIW using the US + UV + EC process. As illustrated in Fig. 2f, using Fe/Fe led in a higher elimination of COD than using Fe/Al, Al/Fe, or Al/Al. Because the anode and cathode electrodes are the primary components of the hybrid US + UV + EC process, the kind of electrode materials and their component composition had an effect on the hybrid system's performance. The Fe electrode's oxidation potential *(–0.447 V)* is significantly greater than the Al electrode's *(–1.662 V)*, resulting in three times the amount of Fe coagulant produced during contaminant removal. (Asaithambi et al., 2012; Igwegbe et al., 2021). The main rationale is that particles created by $Fe(OH)_3$ may have a higher settling ability than those formed by $Al(OH)_3$ (Igwegbe et al., 2021). For these reasons, we used Fe electrodes as the anode and cathode for our subsequent studies utilizing the US + UV + EC method for DIW.

3.2.1.7. Inter-electrode distance. To achieve total pollutant removal while lowering operating costs in a hybrid EC process, the inter-electrode spacing is a critical operating parameter (Moradi et al., 2021; Sandhwar and Prasad, 2018). The influence of inter-electrode spacing on the US + UV + EC process was investigated using values ranging from 0.75 to 3 cm under the following operating conditions: current density – 0.75 A dm^{-2}, pH – 7, COD – 3600 mg L^{-1}, electrode pairing – Fe/Fe, UV – 32 W, US – 100 W, and reaction time – 4 h. As shown in Fig. 2g, increasing the electrode spacing from 0.75 to 3 cm lowered the % COD removal efficiency from 95.63 to 66.40% and increased EEC from 6.97 to 16.56 kWh m^{-3}. With increasing inter-electrode distance, ohmic losses associated with anode and cathode over voltages and mass transfer resistance increase, inhibiting anodic oxidation and resulting in a decrease in the amount of Fe^{2+} at the anode (Dalvand et al., 2011). As a result, the production of coagulants in the middle will be slowed down, as would the adsorption of pollutants at a greater inter-electrode distance (Dalvand et al., 2011). Whereas, when the inter-electrode distance is kept to a minimum, the reduced resistance of current flow in solution facilitates the electrolytic process, increasing the percent COD elimination. Therefore, the selection of the optimal inter–electrode distance is 0.75 cm to minimize EEC while still increase COD removal efficiency.

3.3. Comparison of electrochemical and AOPs

The color and COD removal by UV, US, EC, UV + US, UV + EC, US + EC, and US + UV + EC processes for DIW (Fig. 3a) were determined using the following experimental conditions: current density of 0.75 A dm^{-2}, inter-electrode distance of 0.75 cm, COD of 3600 mg L^{-1}, pH of 7, electrode pairing of Fe/Fe, UV of 32 W, US of 100 W, and reaction time of 4 h. As illustrated in Fig. 3a, sole process of UV (21.94% and 12.64%), sole process of US (26.5% and 16%), and hybrid UV + US process (37.20% and 27.76%) were inefficient at removing % color and % COD. Additionally, there was a significant level of electrical energy use. The UV, US, and UV + US treatments were unable to create enough free radicals to remove color and COD from the DIW(Dizge et al., 2018; Farhadi et al., 2012; Moradi et al., 2021; Moradi and Moussavi, 2019). Additionally, a substantial color removal of 62.55% and a COD reduction of 50.27% for EC were accomplished. The color and COD removal by combining UV and US with EC, such as UV + EC, US + EC, and US + UV + EC hybrid processes, was around 72.45%, 84.57%, and 100%; 60.68%, 71.44%, and 95.63%, respectively. As expected, the hybrid US + UV + EC process outperformed the UV, US, EC, and their combination methods in aspects of removal efficiency of color and COD. This could be because parallel pathways for the generation of sufficient $^{\bullet}OH$ radicals via UV, US, and EC processes have been devised to enable efficient color and COD removal from DIW (Dizge et al., 2018; Moradi et al., 2021; Moradi and Moussavi, 2019).

During the EC step of the UV + EC process, supplemental UV irradiation was performed. The abundance of photoactive sites had a beneficial influence on the EC process. This improvement was achieved by increasing the rate of synthesis of $^{\bullet}OH$ from $Fe(OH)^{2+}$ photo-reduction and photo-decomposition of complexes formed during Fe^{3+} reactions (Aziz et al., 2016):

$$Fe(OH)^{2+} + hv \rightarrow Fe^{2+} + {}^{\bullet}OH \quad (6)$$

$$R(CO_2) - Fe^{3+} + hv \rightarrow R(\bullet CO_2) + Fe^{2+} \rightarrow \bullet R + CO_2 \quad (7)$$

The authors proposed the following equation to describe the mechanism of the US + EC process (Prajapati, 2021):

$$Contaminants + H_2O_2 + \bullet OH + Ultrasonic\ irradiation \rightarrow Intermediates \rightarrow nCO_2 + nH_2O \quad (8)$$

The combination of UV and US with EC for the removal of color and COD from DIW is primarily concerned with the consumption of electrical energy in order to establish the economic viability of the hybrid process. The hybrid process's total EEC was calculated using equation (3), and the results are shown in Fig. 3b. As illustrated in Fig. 3b, the hybrid UV + US + EC process removed 100% of color and 95.63% of COD while consuming 6.97 kWh m^{-3} of electrical energy for the DIW.

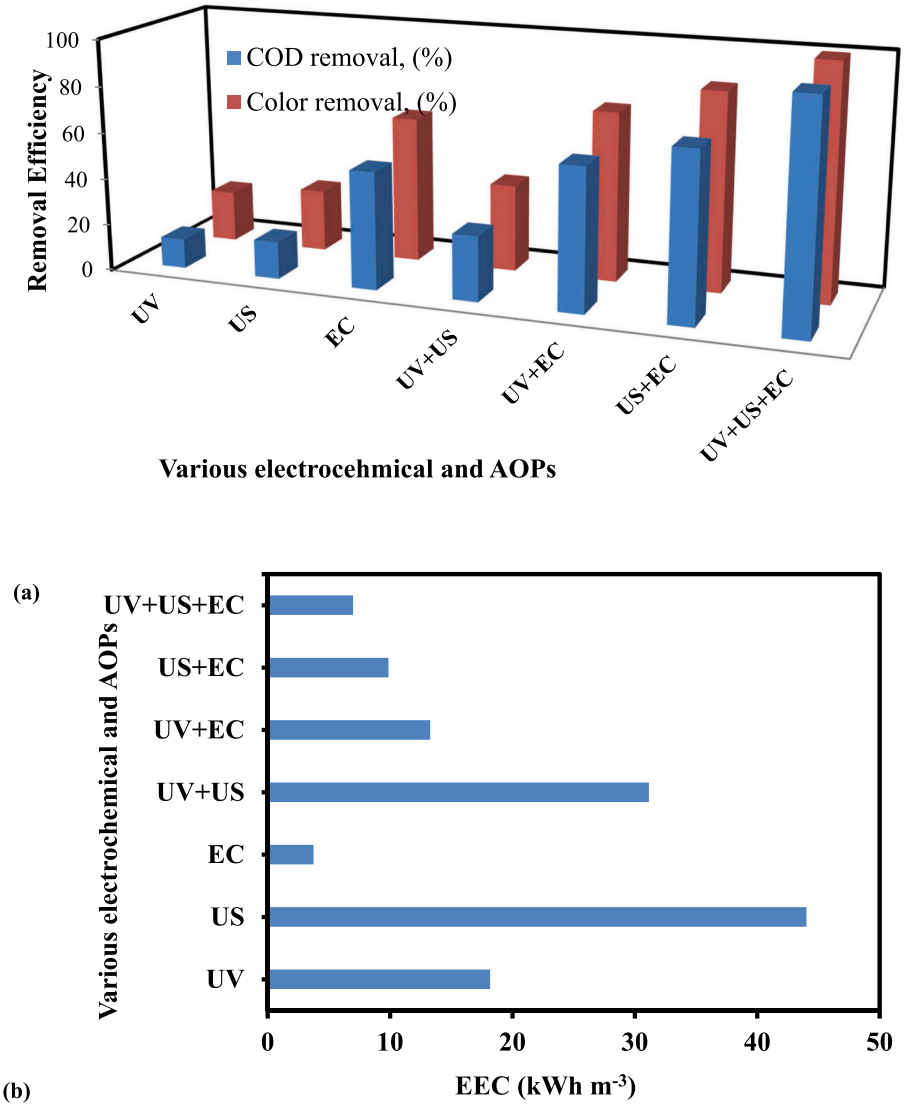

Fig. 3. Comparison of UV only, US only, EC only, UV + US, UV + EC, US + EC and US + UV + EC process on the (a) color, (%) and COD removal, (%) and (b) EEC from distillery wastewater (current density = 0.175 A dm^{-2}, COD = 3600 mg L^{-1}, UV = 32 W, US = 100 W, electrode combination = Fe/Fe, inter–electrode distance = 0.75 cm, pH = 7, and reaction time = 4 h). (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

The single processes such as EC, UV, US and combinations such as UV + US, UV + EC, US + EC required significantly more electrical energy to remove the color and COD than the US + UV + EC process. Consequently, the hybrid UV + EC approach can be utilized to remove contaminants from practically any type of wastewater or industrial effluent, regardless of the source of the contaminants.

3.4. Kinetic model

The COD and color removal rates in the US + UV + EC process showed that a 1st – order process, which was proportional to the COD concentration in the solution. Consequently, the kinetics for the COD removal is written as follows.

$$-\frac{d}{dt}[COD] = k[COD] \tag{9}$$

Rearranging and integrating equation (9) gives

$$1n\frac{COD_t}{COD_t} = -kt \tag{10}$$

Plotting $ln(COD_t/COD_i)$ on the *y*-axis versus reaction time on the x-axis resulted in a straight line with the slope of k. According to Figs. 2a and Fig. 4, the rate constant *k* and R^2 values for COD removal were 0.014 min^{-1} and 0.82, respectively.

4. Conclusions

The purpose of this research was to assess the effectiveness of UV and US combined with EC such as UV, US, EC, UV + US, UV + EC, US + EC, and US + UV + EC processes on the removal of color and COD, as well as the EEC from DIW. When compared to the separate and combined processes, the hybrid US + UV + EC method greatly boosted the color (100%) and COD (95.63%) elimination while using less electrical energy (6.97 kWh m^{-3}). To determine the US + UV + EC process performance, controlling variables including treatment time (1–5 h), current density (0.075–2.0 A dm^{-2}), COD (1800–6000 mg L^{-1}), UV (8–32 W) and US

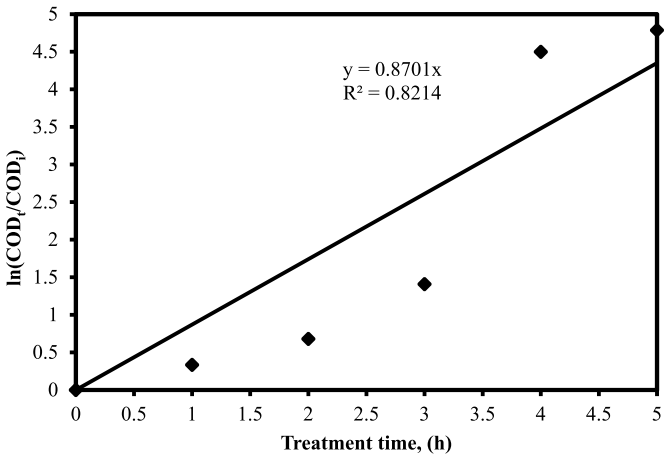

Fig. 4. Degradation kinetics of distillery industrial wastewater.

power (20–100 W), electrode pairings (Al/Al, Al/Fe, Fe/Al, Fe/Fe), and inter-electrode distance (0.75–3.0 cm) were evaluated and reported. The synergy index between the UV and US + EC processes was determined to be 20%. The hybrid US + UV + EC procedure demonstrated that it could be utilized to completely remove contaminants from distillery industrial wastewater.

Credit author statement

Perumal Asaithambi: Investigation; Data curation; Resources; Writing – original draft. **Mamuye Busier Yesuf**: Data curation; Formal analysis; Resources. **Rajendran Govindarajan**: Investigation; Methodology; Data curation; Formal analysis; Resources. **N. M. Hariharan**: Data curation; Formal analysis; Resources. **V. T. Perarasu**: Data curation; Formal analysis; Resources. **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 data that has been used is confidential.

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