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

Perumal Asaithambi (✉ drasaithambi2014@gmail.com)

Jimma University College of Engineering and Technology: Jimma University Institute of Technology

<https://orcid.org/0000-0002-0533-0178>

Rajendran Govindarajan

Hindustan Institute of Technology and Science: Hindustan University

Research Article

Keywords: Distillery industrial wastewater, Sono-Photo-Electrocoagulation, COD removal, color removal, electrical energy consumption, synergy index

Posted Date: March 1st, 2022

DOI: <https://doi.org/10.21203/rs.3.rs-1330571/v1>

License: © ⓘ This work is licensed under a Creative Commons Attribution 4.0 International License.

[Read Full License](#)

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

Perumal Asaithambi^{a,*}, Rajendran Govindarajan^b

^aFaculty of Civil and Environmental Engineering,
Jimma Institute of Technology, Jimma University, Jimma, Ethiopia, Po Box - 378.

^bDepartment of Chemical Engineering,
Hindustan Institute of Technology and Science,
Rajiv Gandhi Salai, Padur, Chennai – 603103, Tamilnadu, India.

***Corresponding author:** Tel.: +251–472115547; Fax.: +251–471111450

Email address: asaithambi.perumal@ju.edu.et (Perumal Asaithambi)

drasaithambi2014@gmail.com (Perumal Asaithambi)

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 ($0.075\text{--}2.0 \text{ A dm}^{-2}$), COD ($1800\text{--}6000 \text{ mg L}^{-1}$), 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.

Keywords: Distillery industrial wastewater; Sono-Photo-Electrocoagulation; COD removal; color removal; electrical energy consumption; synergy index.

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 (Deghles and Kurt 2016; Cotillas et al. 2020). 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 (Nigri et al. 2020; Preethi et al. 2020; Chezeau et al. 2020; Moussavi et al. 2021). 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 (Raschitor et al. 2014; Deghles and Kurt 2016). 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 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. 2020).

70 Additionally, if not properly managed, the generated sludge may contain a variety of components,
71 including potentially dangerous compounds to the environment (Romani et al. 2020). The EC
72 technology as a stand-alone treatment process may have significant practical constraints,
73 particularly if the wastewater is extremely polluted/contaminated (Aziz et al. 2016). Hence, an
74 efficient and relatively inexpensive treatment techniques are required. In response to this, various
75 studies have demonstrated, combining EC with other treatment methods such as advanced
76 oxidation processes (AOPs) improve its performance.

77 Numerous research studies have demonstrated that EC works cohesively with other
78 treatment modalities, including ozone (O₃) assisted EC (He et al. 2007; Das et al. 2021; Mehralian
79 et al. 2021), EC/Adsorption, EC/Chemical coagulation, EC/Magnetic field, EC/reverse osmosis
80 (Al-Qodah et al. 2020), US-EC (Emerick et al. 2020; Moradi et al. 2021), alternating current EC
81 (Alimohammadi et al. 2019; Payami Shabestar et al. 2021), peroxi-EC (Sandhwar and Prasad
82 2018), photo-EC (Farhadi et al. 2012; Asaithambi et al. 2016; Moradi and Moussavi 2019), aerated
83 EC (Akansha et al. 2020), EC-nanofiltration and membrane filtration (Tavangar et al. 2019;
84 Gönder et al. 2020; Uceqli and Kaya 2021) for the elimination of contaminants from industrial
85 effluent and wastewater.

86 Farhadi et al. (2012) compared the various processes for removal of COD from
87 pharmaceutical wastewater. Their findings show that the following order of COD removal
88 efficiency was established under optimal operating conditions for each process: peroxi-EC
89 >peroxi-photo-EC >photo- EC > EC. Cotillas et al. (2014) reported that, when UV irradiation and
90 electrocoagulation (UV-EC) were utilized at low current densities, a synergistic effect was shown
91 for the reduction of turbidity and disinfection rate. Additionally, they observed that performance
92 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 (Zazou et al. 2019; Chanikya et al. 2021). Meanwhile, only a few studies have focused on real wastewater and industrial effluent utilizing hybrid electrochemical and AOPs (Valero et al. 2017; da Costa et al. 2019). 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 characterization

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 DIW 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.

The chemicals used in the experiments were H₂SO₄, NaOH, K₂Cr₂O₇, Na₂S₂O₃, (NH₄)₂Fe(SO₄)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. 1(a), (b) & (c). The US, UV, and EC process units were used in the integrated process. The EC (Fig. 1(a)) 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 x 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. Using a pH meter, the pH of effluent was measured and corrected with H₂SO₄ and NaOH solution (Elico: Model LI120). 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.1 (b)).

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.1 (c)). 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 reactor and added to an Erlenmeyer flask holding $\text{Na}_2\text{S}_2\text{O}_3$ solution to quench the process. After centrifuging the samples at 10,000 rpm for 15 minutes to separate the solid particles and liquid, the supernatant liquid is immediately analyzed for COD (Spectroquant ® TR320) and colour (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}

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)

$$EEC_{EC} = \frac{U \times I \times t}{V_R}, (kWh\ m^{-3}) \quad (4)$$

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)

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

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 Studies on operating parameters

3.1.1 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.

3.1.1.1 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 the Fig. 2. The Fig. 2 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.

3.1.1.2 Current density

The current density is an important factor in the hybrid EC process for the treatment of industrial effluent (Sandhwar and Prasad 2018; Mehralian et al. 2021; Moradi et al. 2021). 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. 3. As illustrated in Fig. 3, 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. 3, 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 3,600 mg L⁻¹ concentration of DIW.

According to Fig. 3, 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).

3.1.1.3. 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 1,800 to 6,000 mg L⁻¹, as shown in Fig. 4. 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.

3.1.1.4. UV power

During the US+EC process, additional UV irradiation was carried out. As illustrated in Fig. 5, 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 [•]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.

3.1.1.5. 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. 6. 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 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.

3.1.1.6. 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 – $3,600 \text{ mg L}^{-1}$, 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. 7, 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 $\text{Fe}(\text{OH})_3$ 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.

3.1.1.7. 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 (Sandhwar and Prasad 2018; Moradi et al. 2021). 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 – $3,600 \text{ mg L}^{-1}$, electrode pairing – Fe/Fe, UV – 32 W, US – 100 W, and reaction time – 4 h. As shown in Fig. 8, 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.

3.2 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. 9.) were determined using the following experimental conditions: current density – 0.75 A dm^{-2} , inter-electrode distance – 0.75 cm, COD – 3600 mg L^{-1} , pH – 7, electrode

pairing of Fe/Fe, UV – 32 W, US – 100 W, and reaction time – 4 h. As illustrated in Fig. 9 (a), 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 (Farhadi et al. 2012; Dizge et al. 2018; Moradi and Moussavi 2019; Moradi et al. 2021). 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 $\cdot\text{OH}$ radicals via UV, US, and EC processes have been devised to enable efficient color and COD removal from DIW (Dizge et al. 2018; Moradi and Moussavi 2019; Moradi et al. 2021).

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 $\cdot\text{OH}$ from $\text{Fe}(\text{OH})^{2+}$ photoreduction and photodecomposition of complexes formed during Fe^{3+} reactions (Aziz et al. 2016):

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

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. 9(b). As illustrated in Fig. 9(b), 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. 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.

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 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 industrial wastewater.

Ethics approval and consent to participate

Not applicable.

Consent for publication

Not applicable.

Authors Contributions

Perumal Asaithambi: Investigation; Data curation; Resources; Writing - original draft.

Rajendran Govindarajan: Investigation; Methodology; Data curation; Formal analysis; Resources.

Funding:

This research did not receive any specific grant from funding agencies in the public, commercial, or not-for-profit sectors.

Availability of data and materials

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

Competing interests

The authors declare that they have no competing interests.

Reference

- Ahlawat R, Srivastava VC, Mall ID, Sinha S (2008) Investigation of the Electrocoagulation Treatment of Cotton Blue Dye Solution using Aluminium Electrodes. CLEAN – Soil, Air, Water 36:863–869. <https://doi.org/https://doi.org/10.1002/clen.200800019>
- Akansha J, Nidheesh P V, Gopinath A, et al (2020) Treatment of dairy industry wastewater by combined aerated electrocoagulation and phytoremediation process. Chemosphere

253:126652. <https://doi.org/https://doi.org/10.1016/j.chemosphere.2020.126652>

Al-Qodah Z, Tawalbeh M, Al-Shannag M, et al (2020) Combined electrocoagulation processes as a novel approach for enhanced pollutants removal: A state-of-the-art review. *Sci Total Environ* 744:.. <https://doi.org/10.1016/j.scitotenv.2020.140806>

Alimohammadi M, Mesdaghinia A, Shayesteh MH, et al (2019) The efficiency of the electrocoagulation process in reducing fluoride: application of inductive alternating current and polarity inverter. *Int J Environ Sci Technol* 16:8239–8254. <https://doi.org/10.1007/s13762-019-02297-4>

Asaithambi P, Sajjadi B, Abdul Aziz AR, Wan Daud WMAB (2016) Performance evaluation of hybrid electrocoagulation process parameters for the treatment of distillery industrial effluent. *Process Saf Environ Prot* 104:406–412. <https://doi.org/10.1016/j.psep.2016.09.023>

Asaithambi P, Susree M, Saravanathamizhan R, Matheswaran M (2012) Ozone assisted electrocoagulation for the treatment of distillery effluent. *Desalination* 297:1–7. <https://doi.org/10.1016/j.desal.2012.04.011>

Aziz ARA, Asaithambi P, Daud WMABW (2016) Combination of electrocoagulation with advanced oxidation processes for the treatment of distillery industrial effluent. *Process Saf Environ Prot* 99:227–235. <https://doi.org/10.1016/j.psep.2015.11.010>

Chanikya P, Nidheesh P V., Syam Babu D, et al (2021) Treatment of dyeing wastewater by combined sulfate radical based electrochemical advanced oxidation and electrocoagulation processes. *Sep Purif Technol* 254:.. <https://doi.org/10.1016/j.seppur.2020.117570>

Chen M, Dollar O, Shafer-Peltier K, et al (2020) Boron removal by electrocoagulation: Removal mechanism, adsorption models and factors influencing removal. *Water Res* 170:115362. <https://doi.org/https://doi.org/10.1016/j.watres.2019.115362>

410 Chezeau B, Boudriche L, Vial C, Boudjemaa A (2020) Treatment of dairy wastewater by
 411 electrocoagulation process: Advantages of combined iron/aluminum electrodes. *Sep Sci*
 412 *Technol* 55:2510–2527. <https://doi.org/10.1080/01496395.2019.1638935>

413 Cotillas S, Llanos J, Miranda OG, et al (2014) Coupling UV irradiation and electrocoagulation for
 414 reclamation of urban wastewater. *Electrochim Acta* 140:396–403.
 415 <https://doi.org/10.1016/j.electacta.2014.04.037>

416 Cotillas S, Llanos J, Moraleda I, et al (2020) Scaling-up an integrated electrodisinfection-
 417 electrocoagulation process for wastewater reclamation. *Chem Eng J* 380:122415.
 418 <https://doi.org/10.1016/j.cej.2019.122415>

419 da Costa PRF, de A. Costa ECT, Castro SSL, et al (2019) A sequential process to treat a cashew-
 420 nut effluent: Electrocoagulation plus electrochemical oxidation. *J Electroanal Chem* 834:79–
 421 85. <https://doi.org/10.1016/j.jelechem.2018.12.035>

422 Dalvand A, Gholami M, Joneidi A, Mahmoodi NM (2011) Dye Removal, Energy Consumption
 423 and Operating Cost of Electrocoagulation of Textile Wastewater as a Clean Process. *Clean -*
 424 *Soil, Air, Water* 39:665–672. <https://doi.org/10.1002/clen.201000233>

425 Das PP, Anweshan, Mondal P, et al (2021) Integrated ozonation assisted electrocoagulation
 426 process for the removal of cyanide from steel industry wastewater. *Chemosphere* 263:128370.
 427 <https://doi.org/10.1016/j.chemosphere.2020.128370>

428 Deghles A, Kurt U (2016) Treatment of tannery wastewater by a hybrid
 429 electrocoagulation/electrodialysis process. *Chem Eng Process Process Intensif* 104:43–50.
 430 <https://doi.org/10.1016/j.cep.2016.02.009>

431 Dizge N, Akarsu C, Ozay Y, et al (2018) Sono-assisted electrocoagulation and cross-flow
 432 membrane processes for brewery wastewater treatment. *J Water Process Eng* 21:52–60.

<https://doi.org/10.1016/j.jwpe.2017.11.016>

Emerick T, Vieira JL, Silveira MHL, João JJ (2020) Ultrasound-assisted electrocoagulation process applied to the treatment and reuse of swine slaughterhouse wastewater. *J Environ Chem Eng* 8:104308. <https://doi.org/10.1016/j.jece.2020.104308>

Farhadi S, Aminzadeh B, Torabian A, et al (2012) Comparison of COD removal from pharmaceutical wastewater by electrocoagulation, photoelectrocoagulation, peroxi-electrocoagulation and peroxi-photoelectrocoagulation processes. *J Hazard Mater* 219–220:35–42. <https://doi.org/10.1016/j.jhazmat.2012.03.013>

Gönder ZB, Balcioğlu G, Vergili I, Kaya Y (2020) An integrated electrocoagulation–nanofiltration process for carwash wastewater reuse. *Chemosphere* 253:126713. <https://doi.org/10.1016/j.chemosphere.2020.126713>

He CC, Hu CY, Lo SL (2016) Evaluation of sono-electrocoagulation for the removal of Reactive Blue 19 passive film removed by ultrasound. *Sep Purif Technol* 165:107–113. <https://doi.org/10.1016/j.seppur.2016.03.047>

He ZQ, Song S, Qiu JP, et al (2007) Decolorization of C.I. Reactive Yellow 84 in Aqueous Solution by Electrocoagulation Enhanced with Ozone: Influence of Operating Conditions. *Environ Technol* 28:1257–1263. <https://doi.org/10.1080/09593332808618884>

Igwegbe CA, Onukwuli OD, Ighalo JO, Umembamalu CJ (2021) Electrocoagulation-flocculation of aquaculture effluent using hybrid iron and aluminium electrodes: A comparative study. *Chem Eng J Adv* 6:100107. <https://doi.org/10.1016/j.ceja.2021.100107>

Jallouli S, Wali A, Buonerba A, et al (2020) Efficient and sustainable treatment of tannery wastewater by a sequential electrocoagulation-UV photolytic process. *J Water Process Eng* 38:101642. <https://doi.org/10.1016/j.jwpe.2020.101642>

- Keramati M, Ayati B (2019) Petroleum wastewater treatment using a combination of electrocoagulation and photocatalytic process with immobilized ZnO nanoparticles on concrete surface. *Process Saf Environ Prot* 126:356–365. <https://doi.org/10.1016/j.psep.2019.04.019>
- Li J, Song C, Su Y, et al (2013) A study on influential factors of high-phosphorus wastewater treated by electrocoagulation–ultrasound. *Environ Sci Pollut Res* 20:5397–5404. <https://doi.org/10.1007/s11356-013-1537-9>
- Li Y-F, Hu C-Y, Lee Y-C, Lo S-L (2021) Effects of zinc salt addition on perfluorooctanoic acid (PFOA) removal by electrocoagulation with aluminum electrodes. *Chemosphere* 132665. <https://doi.org/https://doi.org/10.1016/j.chemosphere.2021.132665>
- Maha Lakshmi P, Sivashanmugam P (2013) Treatment of oil tanning effluent by electrocoagulation: Influence of ultrasound and hybrid electrode on COD removal. *Sep Purif Technol* 116:378–384. <https://doi.org/10.1016/j.seppur.2013.05.026>
- Mehralian M, Khashij M, Dalvand A (2021) Treatment of cardboard factory wastewater using ozone-assisted electrocoagulation process: optimization through response surface methodology. *Environ Sci Pollut Res* 28:45041–45049. <https://doi.org/10.1007/s11356-021-13921-7>
- Moradi M, Moussavi G (2019) Enhanced treatment of tannery wastewater using the electrocoagulation process combined with UVC/VUV photoreactor: Parametric and mechanistic evaluation. *Chem Eng J* 358:1038–1046. <https://doi.org/https://doi.org/10.1016/j.cej.2018.10.069>
- Moradi M, Vasseghian Y, Arabzade H, Mousavi Khaneghah A (2021) Various wastewaters treatment by sono-electrocoagulation process: A comprehensive review of operational

479 parameters and future outlook. Chemosphere 263:128314.
 480 <https://doi.org/10.1016/j.chemosphere.2020.128314>

481 Moussavi M, Pendashteh A, Alinia H (2021) Treatment of a natural gas refinery effluents by
 482 electrocoagulation. Environ Challenges 3:100036.
 483 <https://doi.org/https://doi.org/10.1016/j.envc.2021.100036>

484 Nawarkar CJ, Salkar VD (2019) Solar powered Electrocoagulation system for municipal
 485 wastewater treatment. Fuel 237:222–226.
 486 <https://doi.org/https://doi.org/10.1016/j.fuel.2018.09.140>

487 Negarestani M, Motamedi M, Kashtiaray A, et al (2020) Simultaneous removal of acetaminophen
 488 and ibuprofen from underground water by an electrocoagulation unit: Operational parameters
 489 and kinetics. Groundw Sustain Dev 11:100474.
 490 <https://doi.org/https://doi.org/10.1016/j.gsd.2020.100474>

491 Nigri EM, Santos ALA, Rocha SDF (2020) Removal of organic compounds, calcium and
 492 strontium from petroleum industry effluent by simultaneous electrocoagulation and
 493 adsorption. J Water Process Eng 37:101442.
 494 <https://doi.org/https://doi.org/10.1016/j.jwpe.2020.101442>

495 Oza H, Anantha Singh TS, Sasikumar Jampa S (2021) Removal of arsenic from aqueous solution
 496 using combined ultrasonic and electrocoagulation process. Mater Today Proc 47:728–732.
 497 <https://doi.org/https://doi.org/10.1016/j.matpr.2021.01.569>

498 Özyonar F, Gökkuş Ö, Sabuni M (2020) Removal of disperse and reactive dyes from aqueous
 499 solutions using ultrasound-assisted electrocoagulation. Chemosphere 258:127325.
 500 <https://doi.org/https://doi.org/10.1016/j.chemosphere.2020.127325>

501 Payami Shabestar M, Alavi Moghaddam MR, Karamati-Niaragh E (2021) Evaluation of energy

and electrode consumption of Acid Red 18 removal using electrocoagulation process through RSM: alternating and direct current. *Environ Sci Pollut Res*. <https://doi.org/10.1007/s11356-021-15345-9>

Prajapati AK (2021) Sono-assisted electrocoagulation treatment of rice grain based distillery biodigester effluent: Performance and cost analysis. *Process Saf Environ Prot* 150:314–322. <https://doi.org/https://doi.org/10.1016/j.psep.2021.04.030>

Preethi V, Ramesh ST, Gandhimathi R, Nidheesh PV (2020) Optimization of batch electrocoagulation process using Box-Behnken experimental design for the treatment of crude vegetable oil refinery wastewater. *J Dispers Sci Technol* 41:592–599. <https://doi.org/10.1080/01932691.2019.1595640>

Raschitor A, Fernandez CM, Cretescu I, et al (2014) Sono-electrocoagulation of wastewater polluted with Rhodamine 6G. *Sep Purif Technol* 135:110–116. <https://doi.org/10.1016/j.seppur.2014.08.003>

Romani M, Espinoza-Quiñones FR, Módenes AN, Borba CE (2020) New insights into the improvement of electrocoagulation performance on the basis of a time-integrated performance index: The pivotal role of electrical conductivity. *J Environ Chem Eng* 8:103902. <https://doi.org/https://doi.org/10.1016/j.jece.2020.103902>

Sandhwar VK, Prasad B (2018) 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* 35:909–921. <https://doi.org/10.1007/s11814-017-0336-2>

Tavangar T, Jalali K, Alaei Shahmirzadi MA, Karimi M (2019) Toward real textile wastewater treatment: Membrane fouling control and effective fractionation of dyes/inorganic salts using

a hybrid electrocoagulation – Nanofiltration process. Sep Purif Technol 216:115–125.
<https://doi.org/https://doi.org/10.1016/j.seppur.2019.01.070>

Uceвли O, Kaya Y (2021) A comparative study of membrane filtration, electrocoagulation, chemical coagulation and their hybrid processes for greywater treatment. J Environ Chem Eng 9:104946. <https://doi.org/https://doi.org/10.1016/j.jece.2020.104946>

Valero P, Verbel M, Silva-Agredo J, et al (2017) Electrochemical advanced oxidation processes for Staphylococcus aureus disinfection in municipal WWTP effluents. J Environ Manage 198:256–265. <https://doi.org/https://doi.org/10.1016/j.jenvman.2017.04.070>

Zazou H, Afanga H, Akhouairi S, et al (2019) Treatment of textile industry wastewater by electrocoagulation coupled with electrochemical advanced oxidation process. J Water Process Eng 28:214–221. <https://doi.org/https://doi.org/10.1016/j.jwpe.2019.02.006>

List of Figures

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

Fig. 2. 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).

Fig. 3. 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).

Fig. 4. 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).

Fig. 5. 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).

Fig. 6. 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).

Fig. 7. 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).

Fig. 8. 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).

Fig. 9. 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).

Fig. 1. (a)

Fig. 1. (b)

Fig. 1. (c)

Fig. 2

Fig. 3

Fig. 4

Fig. 5

Fig. 6

Fig. 7

721
722
723

724

Fig. 8

Fig. 9