Sustainable Chemistry for the Environment 2 (2023) 100014

Contents lists available at ScienceDirect

Sustainable Chemistry for the Environment

journal homepage: www.editorialmanager.com/scenv

Industrial wastewater treatment using batch recirculation electrocoagulation (BRE) process: Studies on operating parameters

Perumal Asaithambi[a,*,1], Mamuye Busier Yesuf[a], Rajendran Govindarajan[b], Periyasamy Selvakumar[c,2], Subramaniapillai Niju[d], T. Pandiyarajan[e], Abudukeremu Kadier[f,g,**], D. Duc Nguyen[h], Esayas Alemayehu[a,i]

[a] Faculty of Civil and Environmental Engineering, Jimma Institute of Technology, Jimma University, Po Box - 378, Jimma, Ethiopia
[b] Department of Chemical Engineering, Hindustan Institute of Technology and Science, Rajiv Gandhi Salai, Padur, Chennai 603103, Tamilnadu, India
[c] Department of Chemical Engineering, School of Mechanical, Chemical and Materials Engineering, Adama Science and Technology University, Adama 1888, Ethiopia
[d] Department of Biotechnology, PSG College of Technology, Coimbatore 641004, Tamilnadu, India
[e] Department of Sciences, Indian Institute of Information Technology Design and Manufacturing, Dinnedevarapadu , Kurnool 518007, Andhra Pradesh, India
[f] Laboratory of Environmental Science and Technology, The Xinjiang Technical Institute of Physics and Chemistry, Key Laboratory of Functional Materials and Devices for Special Environments, Chinese Academy of Sciences, Urumqi 830011, China
[g] Center of Materials Science and Optoelectronics Engineering, University of Chinese Academy of Sciences, Beijing 100049, China
[h] Department of Environmental Energy Engineering, Kyonggi University, South Korea
[i] Africa Center of Excellence for Water Management, Addis Ababa University, Po Box-1176, Addis Ababa, Ethiopia

ARTICLE INFO

Keywords:
Industrial effluent
Batch recirculation electrocoagulation
Pollutant removal
Energy use
Clean water
Solid sludge formed

ABSTRACT

The environmental industry has demonstrated an increasing interest in employing electrocoagulation (EC) process to treat industrial wastewater/effluent for recycling/drinking purposes. An iron (Fe)/aluminum (Al) plate-based batch recirculation electrocoagulation technology for wastewater treatment in the distillery industry is discussed in this work. The impact of different operational parameters, including COD, wastewater pH, current, inter-electrode spacing, combination of electrodes, recirculation flow rate, concentration of electrolytes, and treatment duration on % color, % COD reduction efficiency, and energy consumption was examined. The experimental outcomes demonstrated that, the color removal was 100 %, COD removal was 99.90 %, and energy consumption was 7.73 kWh m^{-3} for COD of 3600 mg L^{-1}, current of 0.56 Amp, combination of electrodes of Fe/Fe, inter-electrode spacing of 1 cm, wastewater pH of 7, flow rate of 15 L h^{-1}, concentration of electrolytes of 5 g L^{-1}, and treatment time of 180 min, respectively. It was found that, a longer treatment period, higher electrolyte concentrations and current, lower COD concentrations and recirculation flow rates, Fe/Fe electrode pairings, a pH of 7, and a smaller inter-electrode spacing all contributed to increased % COD reduction efficiency. The quantity of solid sludge formed were studied with the help of operational parameters, and the results were reported. Under the optimized process conditions, the wastewater treated can be fully recovered as clean water. As a consequence of this, the results of the experiments have shown that the batch recirculation electrocoagulation process has the potential to be a more promising solution to the problem of eliminating contaminants from wastewater and industrial effluent.

1. Introduction

In recent years, electrocoagulation (EC) technologies has garnered recognition as a water and wastewater treatment method that is both effective and environmentally friendly, and it is able to remove a wide variety of contaminants [1,2]. It has been suggested that EC is an efficient approach for the treatment of a variety of wastewaters, including those from the dairy wastewater [3], slaughterhouse wastewater [4],

* Corresponding author.
** Corresponding author at: Laboratory of Environmental Science and Technology, The Xinjiang Technical Institute of Physics and Chemistry, Key Laboratory of Functional Materials and Devices for Special Environments, Chinese Academy of Sciences, Urumqi 830011, China.
E-mail addresses: asaithambi.perumal@ju.edu.et, drasaithambi2014@gmail.com (P. Asaithambi), abudukeremu@ms.xjb.ac.cn (A. Kadier).
1 http://orcid.org/0000-0002-0533-0178
2 https://orcid.org/0000-0002-7326-8206

https://doi.org/10.1016/j.scenv.2023.100014
Received 26 February 2023; Received in revised form 19 May 2023; Accepted 23 June 2023

metal-plating wastewater [5], municipal wastewater [6], reactive dye bath wastewater [7], marble processing wastewater [8], heavy metal wastewater [9], mining industry [10], etc. Most EC research has so far been done in batch reactors that are small enough to operate in laboratories to wastewater treatment [11–13]. This is due to the ease with which these reactors can be installed and operated. The benefits and capabilities of coagulation, flotation, and adsorption, in addition to electrochemistry, are included into the EC process [2,14,15]. As a consequence of this, it provides a number of benefits that are not available with the more traditional approaches to treatment, and it is distinguished from other approaches by its eco-friendliness, simplicity of operation, adaptability, compactness, and low chemical demand [15,16].

The EC method, on the other hand, is plagued by excessive energy and electrode consumption, in addition to electrode passivation [17]. Thus, the batch-electrocoagulation method showed worse outcomes in terms of pollutant removal, performance with high energy consumption, and operating costs, the process was denied [18]. To prevent the drawback of the batch electrocoagulation process, it is possible to use the batch recirculation electrocoagulation technique for the treatment of industrial effluent and wastewater [1,18].

However, a batch reactor cannot successfully treat vast amounts of wastewater in the same manner that a continuous flow reactor can [19]. As a result, implementing a continuous flow electrocoagulation reactor is a viable choice. Currently, continuous flow electrocoagulation is utilized across the world in a range of industries to treat wastewater, including metal working wastewater [20], moquette industry wastewater [21], removal of fluoride [22], removal of azo dyes [23], removal of arsenic [24]. Changing the mode of operating is one of the many ways that have been suggested to cope with passivation. Of these, it has garnered the most attention. Over the last several years, there has been an increasing interest in running EC with a process that utilizes continuous flow[25]. During the last decades, there has been a significant amount of study conducted in this area, and several research papers have been published as a result [1,20,26–28].

In order to remediate wastewater from printing ink, Zampeta et al. [1] assessed a continuous flow electrocoagulation reactor technology and stated that it had the first research. Additionally, they discovered that the chemical oxygen demand (COD), color, and total suspended solids (TSS) removal rates were 82 %, 98 %, and 85 %, respectively, under the optimal operating parameters of flow rate of 6 mL min^{-1} and current density of 21 mA cm^{-2}. In addition, they came to the conclusion that using a continuous flow electrocoagulation reactor to treat wastewater with high COD concentrations was successful and affordable.

Continuous electrocoagulation using iron electrodes to remove textile dye from wastewater was shown to be a quite efficient treatment method by Hendaoui et al. in 2021 [25]. They succeeded in doing so; under optimum operating conditions, the color removal efficiency was 93.97 % at a cost of 0.0927 USD m^{-3}. Furthermore, they claimed that a continuous electrocoagulation technique could effectively be used on an industrial scale to quickly and cheaply detoxify actual wastewater. Kobya et al. [20] employed a continuous EC technique to treat wastewater from a metalworking plant. In order to establish their efficiency, operational factors were examined. Under optimum circumstances, they acquired the highest total organic carbon (TOC), COD, and turbidity removals via the EC process.

According to Abbasi et al., 2020 [29] used a continuous EC reactor to treat effluent from the manufacturing of licorice. They achieved elimination of COD of 91.50 %, color of 91 %, turbidity of 82 %, and alkalinity of 73.3 % while operating at optimal conditions. Continuous electrocoagulation (CEC) technique to remove COD, TS, turbidity, and color from pulp and paper mill effluent was explored by S. Maheshab et al. in 2016 [30]. Overall, they came to the conclusion that the CEC process can be utilized to treat the pulp and paper effluent in conjunction with other treatment processes after observing maximal COD and color removal were about 82.2 % and 89.9 % throughout the process. According to [31] Caizares et al., at the optimum conditions of flow rate of 10.5 L h^{-1}, oil content of 3000 mg L^{-1}, supporting electrolyte of 3000 mg L^{-1} NaCl, initial pH of 8.5, current density of 105 of A m^{-2}, and temperature of 25 °C, a continuous EC reactor removed 80 % COD from oil-water emulsions using Al electrodes.

Studies that have already been published have shown that the continuous electrocoagulation method can reduce color, COD, TOC, TS, and turbidity in wastewater [25,29]. From a practical and economic standpoint, electrical energy consumption is an essential part of the continuous electrocoagulation process in addition to removing contaminants from wastewater and industrial effluent. To the authors' knowledge, no research article has been published specifically regarding the role of continuous batch recirculation electrocoagulation process in reducing pollutant with consumed of electrical energy. With this in mind, the purpose of our study is to attempt to fill in this gap. To the best of the authors' knowledge, no research has been published on the continuous batch recirculation EC process's ability to treat actual distillery industrial wastewater. For the treatment of industrial wastewater from distilleries, continuous batch recirculation EC process studies are crucial for applicability.

Therefore, the goal of the present research was to give a detailed assessment of continuous batch recirculation electrocoagulation (BRE) process for the removal of color and COD together with a calculation of the electrical energy consumption from industrial effluent from distilleries. The effects of operational factors such as treatment time (30–240 min), COD (1800–6000 mg L^{-1}), wastewater pH $(1-12)$, current (0.11–0.65 Amp), inter-electrode spacing (1–4 cm), electrode combinations (Fe/Fe, Fe/Al, Al/Fe, and Al/Al), recirculation flow rate (15–90 L h^{-1}), and concentration of electrolytes (1–7 g L^{-1}) were investigated and reported.

2. Material and methods

2.1. Wastewater characterization

Distillery industrial wastewater, or DIW, was used in experiments; it was collected from distilleries in Addis Ababa, Ethiopia. Wastewater quality characteristics for the distillery's industrial effluent were measured, including color: dark brown, odor: burned sugar, pH: 4.2–4.4, COD: 80,000–90,000 mg L^{-1}, BOD: 7000–8000 mg L^{-1}, TDS: 14.44 g L^{-1}, and TDS: 5450–6650 mg L^{-1}. In the investigations, chemicals such as $K_2Cr_2O_7$, $(NH_4)_2Fe(SO_4)_2 0.6\ H_2O$, H_2SO_4 and NaOH, HCl, Ag_2SO_4, $HgSO_4$, etc. were utilized. Each and every chemical was bought from Merck in India, and it was utilized precisely as it had been provided.

2.2. Experimental set-up

The batch recirculation electrocoagulation (BRE) process experimental setup is depicted schematically in Fig. 1. With an entrance at the bottom of one side and an exit at the top of the other, the electrocoagulation reactor is built of acrylic sheet and has a 2.50 L capacity. The 2.0 L is the wastewater's active working volume. The anode and cathode used in the electrocoagulation process had inter-electrode spacing that ranged from 1 to 4 cm. Electrical connections were made to form an electrocoagulation cell with a constant current of 0.56 Amp using a direct current (DC) power source (AMETEK Model: EC 1000 S; 0–5 A, 0–270 V, 50 Hz).

The magnetically powered self-priming centrifugal pump and a 5000 mL reservoir were the other parts of the setup, and they were linked via silicone rubber tubing. A total of 5000 mL of wastewater with the necessary beginning COD content and supporting electrolyte concentrations of 5 g L^{-1} were added to the reservoir. Prior to starting the experiments, the original pH of the effluent was determined using a pH meter (Elico: Model LI120) and adjusted with 0.1 N H_2SO_4 and 0.1 N

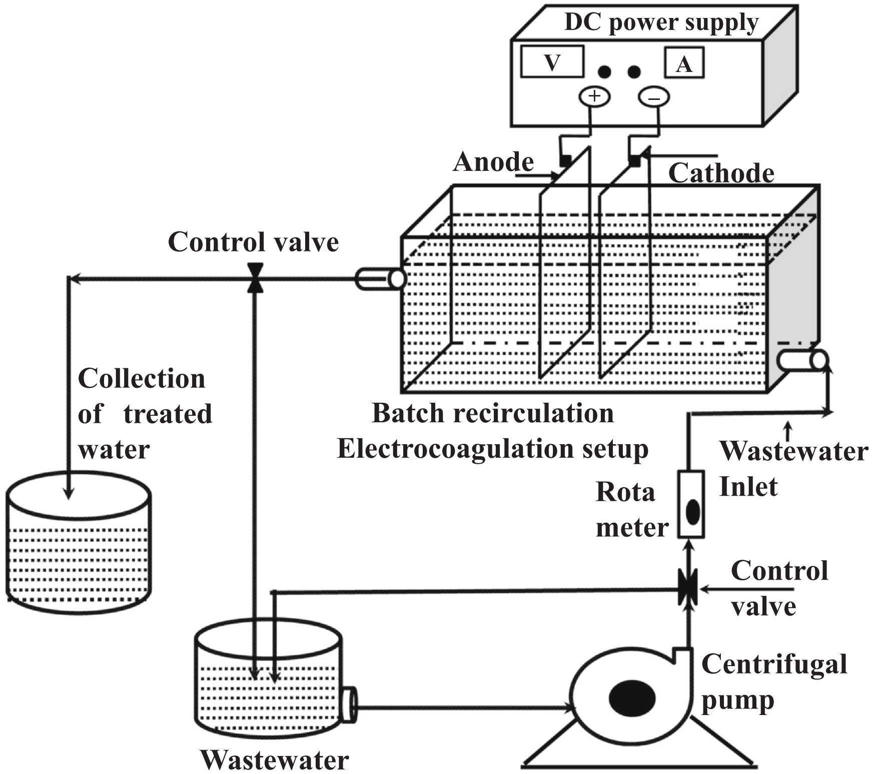

Fig. 1. The continuous batch recirculation electrocoagulation (CRE) process is shown in a schematic.

NaOH solution to the proper value in the range of 1–12. Pumping and regulating the valves allowed for the establishment of the necessary flow rate through the reactor. Using a calibrated Rota meter, the effluent recirculation flow rate into the electrochemical reactor was adjusted to 15, 30, 45, 60, 75, and 90 $L h^{-1}$. The performance of the electrolytic cell was further investigated when the concentration of the NaCl electrolyte varied from 1 g to 7 $g L^{-1}$ at a fixed flow rate. For the combinations of electrode (Fe/Fe, Fe/Al, Al/Fe, and Al/Al), electrodes such as iron (Fe: grade MS 104) and aluminum (Al) plates were used, and the dimensions were 10 × 13 × 0.1 cm. Effective electrode surface area was 10 × 10 × 0.1 cm. A multimeter was used to measure the cell voltage in order to calculate the amount of electricity used throughout the needed treatment period. At regular intervals, samples were taken out of the reactor and centrifuged (REMI, Model: R-24) for 30 min at 15,000 rpm before being examined for COD and color removal. The COD and color were measured using the closed reflux technique (Spectroquant® TR 320) and UV/Vis-Spectrophotometer (Spectroquant® Pharo 300).

2.3. Analysis

2.3.1. Efficiency of removal, (%)

The pollutant reduction efficacy (%) was determined using COD and color of distillery industry effluent before and after the batch recirculation electrocoagulation process.

Eqs. (1–2) were used to compute the color and COD reduction efficiency.

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

Where, A_i and A_t represent the effluent from distilleries' absorbance at a constant wavelength (λ_{ma}) before and after treatment, respectively.

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

Where, C_i and C_t are, respectively, the COD (mg/L) values of the distillery effluent before and after treatment.

2.3.2. Energy consumption

Electrical energy consumption is a crucial factor in the batch recirculation electrocoagulation (BRE) process that must be considered from a practical and economic standpoint. Eq. (3) was used to calculate this electrical energy usage [20–22].

$$Energy \quad consumption = \frac{VIt}{V_R} + C_{pump}, \left(kWhr \quad m^{-3}\right) \quad (3)$$

Where, V is the voltage of the cell (volts), I is the applied current (amps), t is the BRE treatment time (hours), C_{pump} is the energy consumed pump, and V_R is the volume of wastewater utilized (L).

2.3.3. Solid sludge formed

Solid sludge formed was determined using the Eq. (4) and is defined as the proportion of the mass of the sludge generated (m_p) to the volume of the reactor (m^3) and the treatment duration (hr)[32–35].

$$Y_{STY} = \frac{m_p}{V^*t}, \left(kg \quad m^{-3}hr^{-1}\right) \quad (4)$$

3. Results and discussion

3.1. Operating parameters for the BRE process

In order to find the best parameter conditions for the BRE process to remove the maximum percentage of COD with the least amount of energy, process parameters including COD concentration, wastewater pH, current, inter-electrode spacing, combination of electrodes, recirculation flow rate, concentration of electrolytes, and treatment time were examined. Following is a discussion of how various operational parameters affect the BRE process.

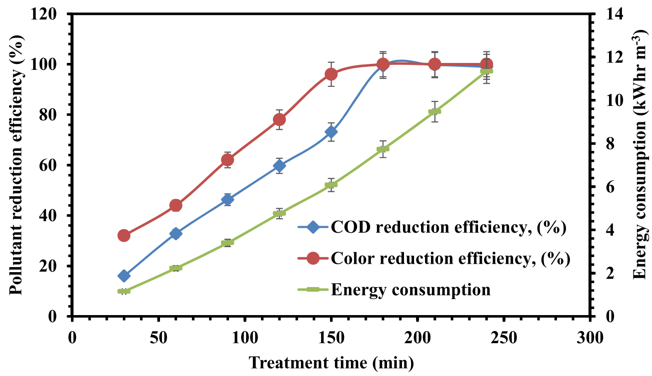

Fig. 2. Effect of treatment time on (a) % color, % COD reduction efficiency, and energy consumption by using BRE process (Experimental conditions: COD - 3600 mg L^{-1}; wastewater pH - 7; current - 0.56 Amp; inter-electrode spacing - 1 cm; combination of electrodes - Fe/Fe; recirculation flow rate - 15 L h^{-1}; concentration of electrolytes - 5 g L^{-1}).

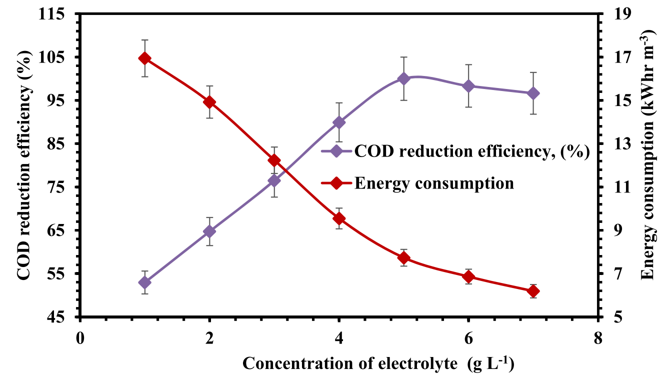

Fig. 3. Effect of concentration electrolyte on % COD reduction efficiency and energy consumption by using BRE process (Experimental conditions: COD - 3600 mg L^{-1}; wastewater pH - 7; current - 0.56 Amp; inter-electrode spacing - 1 cm; combination of electrodes - Fe/Fe; recirculation flow rate - 15 L h-1 and treatment time - 180 min).

3.1.1. *Effect of treatment time*

One of the most important factors in industrial wastewater treatment employing batch recirculation EC methods is the effect of treatment time [21,29]. Using a BRE process with COD of 3600 mg L^{-1}, combination of electrodes of Fe/Fe, current of 0.56 Amp, inter-electrode spacing of 1 cm, wastewater pH of 7, recirculation flow rate of 15 L h^{-1}, concentration of electrolytes of 5 g L^{-1}, and the results is shown in Fig. 2, this study examined the effects of treatment time on energy consumption and efficiency in reducing color and COD. The figure shows that as treatment time has increased from 30 to 240 min, energy usage (1.15–11.3 $kWhr^{-3}$) and the percentage color (32–100 %) and COD (16–100 %) reduction have both increased. A consistent quantity of electrode ions and associated oxidizing species are produced as the treatment duration increases. As a result, the effectiveness of removing color and COD rises as treatment duration increases. Because of the increased production of hydroxyl radicals, there is an improvement in both the color and the COD removal when there is an increase in the energy consumption through cell voltage.

3.1.2. *Effect of concentration electrolyte*

In electrochemical wastewater treatment technology, the concentration of the electrolyte is a key factor that not only affects the reaction result but also significantly affects the amount of electricity used and the cost of operation [36,37] The addition of various sodium chloride (NaCl) concentrations as a supporting electrolyte improves the conductivity of wastewater [37].

$$2Cl^{-}_{(aq)} \rightarrow Cl_{2(g)} + 2e^{-} \tag{5}$$

$$Cl_2 + H_2O \rightarrow HOCl + Cl^{-} + H^{+} \tag{6}$$

$$\mathrm{HOCl} \leftrightarrow \mathrm{OCl^{-}} + \mathrm{H^{+}} \tag{7}$$

When employing a batch recirculation EC method, a NaCl concentration of between 1(2.036 mS/cm) and 7(6.936mS/cm) g L^{-1} was utilized to raise the solution's conductivity (2.036–6.936 mS/cm) in order to save energy and increase the ability to remove COD from industrial wastewater. The results are given in Fig. 3. With an increase in NaCl content from 1(2.036 mS/cm) to 5(3.60 mS/cm) g L^{-1}, the energy consumption reduced from 17 to 7.73 kWhr m^{-3} and the percentage of COD elimination increased from 52.96 % to 100 %. By producing oxychloride (OCl^-) and chlorine gas, more Cl^- ions may contribute to this by acting as a stronger oxidizing agent [38]. The amount of passivation effect is reduced and the current efficiency is increased when chloride ions scavenge oxygen in the electrode to generate OCl^- ions [39]. However, a further increase in NaCl concentration from 5(3.60 mS/cm) to 7(6.936 mS/cm) g L^{-1} marginally reduces COD removal efficiency from 100 % to 96.64 %, whereas an increase in NaCl concentration from 1 to 7 g L^{-1} significantly reduces energy consumption from 17 to 6.19 kWhr m^{-3}, respectively. Due to better conductivity, this can be achieved by increasing the dose of the coagulant, but at higher NaCl concentrations, insufficient Fe dissolution results in unfavorable interactions between the coagulant and the particles. Using NaCl as an electrolyte results in two processes that may be utilized to treat wastewater: electrocoagulation with coagulant production and electrochemical degradation as a result of indirect hypochlorine oxidation. Furthermore, the treated DIW was tested for Cl^- (25 mg L^{-1}) and the findings revealed that the water is safe for both public and environmental health.

3.1.3. *Effect of recirculation flow rate*

Fig. 4 shows that, the impact of recirculation flow rate on percentage COD removal and energy consumption at fixed COD of 3600 mg L^{-1}, combination of electrodes of Fe/Fe, current of 0.56 Amp, inter-electrode spacing of 1, wastewater pH of 7, concentration of electrolytes of 5 g L^{-1}, and treatment time of 180 min, respectively. As the recirculation flow rate is increased, the percentage of COD removal declines. The COD elimination percentage fixed time at 180 min of operation has been observed to range from 99.38 % to 46.24 %, respectively, when the recirculation flow rate is adjusted from 15 to 90 L h^{-1}. Degradation of organic pollutants depends on their bulk concentration. The rate of COD elimination increases noticeably at lower flow rates [22]. This could be as a result of the process' enhanced production of hydroxyl radicals. As recirculation flow rates rise, less Fe^{2+} is transported from the bulk to the electrode surfaces [20]. This

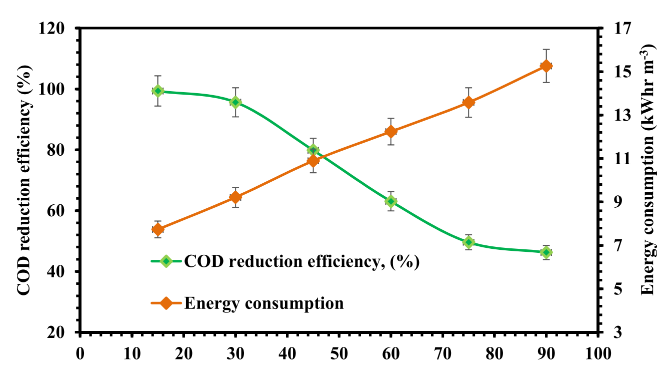

Fig. 4. Effect of recirculation flow rate on % COD reduction efficiency and energy consumption by using BRE process (Experimental conditions: COD - 3600 mg L^{-1}; wastewater pH - 7; current - 0.56 Amp; inter-electrode spacing - 1 cm; combination of electrodes - Fe/Fe; concentration of electrolytes - 5 g L^{-1} and treatment time - 180 min).

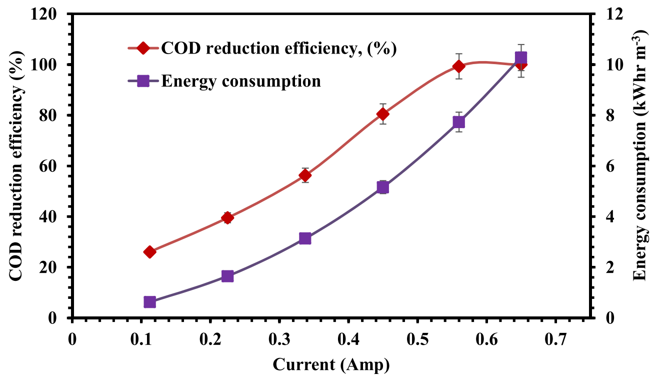

Fig. 5. Effect of current on % COD reduction efficiency and energy consumption by using BRE process (Experimental conditions: COD - 3600 mg L^{-1}; wastewater pH - 7; inter-electrode spacing - 1 cm; combination of electrodes - Fe/Fe; recirculation flow rate - 15 L h^{-1}; concentration of electrolytes - 5 g L^{-1} and treatment time - 180 min).

would make it easier to produce Fe(OH) and dissolve Fe^{2+} to create hydroxyl OH for reactions that aid in the greater elimination of COD. By increasing the recirculation flow rate of the effluent recirculation from 15 to 90 L h^{-1}, the energy consumption was significantly improved from 7.73 to 15.3 kWhr m^{-3}.

3.1.4. Effect of current

By performing the BRE process for industrial wastewater treatment at various currents between 0.1125 and 0.65Amp, the impact of current on energy usage and COD reduction efficiency was examined. The findings, as shown in Fig. 5, revealed that an increase in current resulted to a significant removal of COD. It was evident that during the course of the 180 min treatment, the removal efficiencies for COD was increased from 26.08 % to 100 % for applied current from 0.1125 to 0.65 Amp, respectively. According to the findings, the current played a significant role in boosting COD elimination during batch recirculation EC treatment. Two factors, including an increase in current, may improve COD removal: (1) The direct proportionality between the direct current field and potential electrolysis was impacted by the higher current, which meant that there was a greater formation of iron hydroxides; (2) the hydraulic condition in batch recirculation. The electric field's direct impact on particle movement and response during the EC treatment, including collisions between the coagulants that were produced and the bubbles that were created, might have an impact on the elimination of COD [20,22,40,41].

Since the cell potential rises with increasing current, which is directly proportional to the electrical energy consumption, Fig. 5 indicates that energy consumption increased from 0.63 to 10.3 kWhr m^{-3} with the increasing current from 0.1125 to 0.65 Amp. In order to prevent the generation of heat, the evolution of extra O_2 at a higher current density, and accomplish maximal COD elimination with the lowest amount of electric energy consumption, the current must be maintained at an optimal level. Operation with the proper current not only increases the effectiveness of COD removal but also reduces energy consumption. The beginning of excess sludge generation in the reaction system would rely on the reactor's circumstances, particularly the current, and may be used to ascertain the best time to operate the reactor in concern.

3.1.5. Effect of pH

According to earlier research, the batch recirculation EC process's treatment effectiveness was impacted by the wastewater's starting pH [41]. To examine the impact on energy use and COD reduction efficiency utilizing a batch recirculation EC method, the starting pH of the wastewater was changed from 1 to 11. The efficiency of COD reduction and energy use are shown in Fig. 6(a) as functions of the pH of the

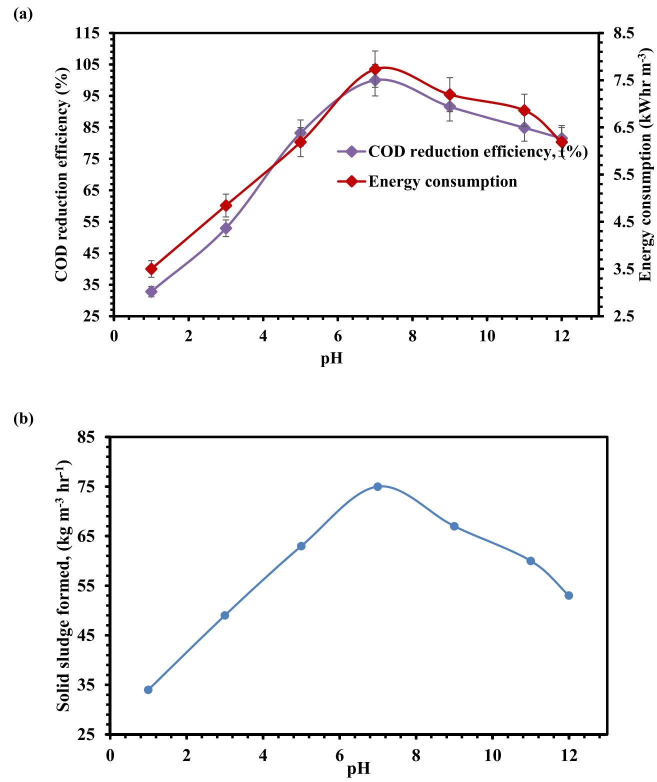

Fig. 6. Effect of pH on (a) % COD reduction efficiency and energy consumption and (b) Solid sludge formed by using BRE process (Experimental conditions: COD - 3600 mg L^{-1}; current - 0.56 Amp; inter-electrode spacing - 1 cm; combination of electrodes - Fe/Fe; recirculation flow rate - 15 L h^{-1}; concentration of electrolytes - 5 g L^{-1} and treatment time - 180 min).

initial wastewater. With an increase in the starting pH of wastewater from 1 to 7, the percentage COD reduction and energy consumption were continuously increased from 32.80 % to 100 % and 3.5–7.73 kWhr m^{-3}, respectively. However, when the initial pH of the wastewater increased from 7 to 12, the COD removal percentage and the energy consumption reduced from 100 % to 81.52 % and 7.73–6.19 kWhr m^{-3}, respectively. The highest percentage of COD removal was seen at a pH of 7 for the real wastewater. The majority of investigations discovered that when wastewater had a pH that was more acidic or alkaline than neutral, removal effectiveness was reduced [41]. The reason for the high percentage COD removal efficiency at neutral pH may be due to the precipitation of their hydroxides at the cathode. When the pH of wastewater changes to an acidic condition, ferrous iron (Fe^{2+}) is oxidized to ferric iron (Fe^{3+}), which reduces the efficiency of COD removal. The neutral and alkaline pH of wastewater seems to promote complex polymerization and the oxidation of Fe^{2+} to Fe^{3+}. In the end, hydroxylated colloidal polymers and an insoluble precipitate of hydrated ferric oxide were produced, increasing the COD removal efficiency by %.

The effect of pH on solid sludge formed for distillery wastewater was examined using a batch recirculation EC technique; the results are shown in Fig. 6. (b). The solid sludge formed increased from 34 to 75 kg $m^{-3}hr^{-1}$ with a rise in wastewater pH from 1 to 7, but decreased from 75 to 53 kg $m^{-3}hr^{-1}$ with a further increase in pH from 7 to 12, as can be seen in Fig. 6(b). The reason might be that the greatest COD removal rate and the highest solid sludge formed were reported at the starting pH of 7 for the wastewater and the lowest loading levels [33–35].

3.1.6. Effect of COD concentration

With the remaining operating parameters held constant, such as wastewater pH - 7, current - 0.56 Amp, inter-electrode spacing - 1 cm,

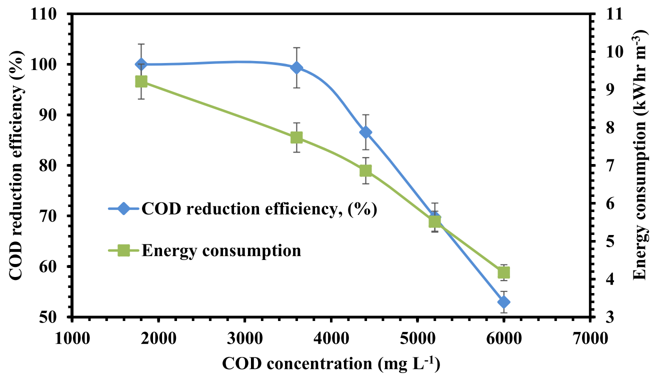

Fig. 7. Effect of COD on % COD reduction efficiency and energy consumption by using BRE process (Experimental conditions: wastewater pH - 7; current - 0.56 Amp; inter-electrode spacing - 1 cm; combination of electrodes - Fe/Fe; recirculation flow rate - 15 L h^{-1}; concentration of electrolytes - 5 g L^{-1} and treatment time - 180 min).

combination of electrodes - Fe/Fe, flow rate - 15 L h^{-1}, concentration of electrolytes - 5 g L^{-1}, and treatment time - 180 min, five experiments with COD concentrations of 1800, 3600, 4400, 5200, and 6000 mg L^{-1}of distillery wastewater solutions were conducted. Fig. 7 demonstrates that when COD content increased from 1800 to 6000 mg L^{-1}, energy consumption and COD elimination reduced from 9.21 to 4.17 kWhr m^{-3} and 100–53 %, respectively. This is due to the fact that the electrode consistently created the same number of iron hydroxide complexes under a particular operating environment, which led to the production of the same amount of hydroxide radicals [42,43]. With rising COD concentrations in wastewater, the amount of hydroxyl radicals needed to degrade all organic and inorganic compounds was insufficient into H_2O, CO_2, and certain other organic ions [44]. It is evident that, in contrast to higher COD concentrations, lower COD concentrations had a high% COD removal efficiency under the given experimental conditions.

3.1.7. *Effect of inter-electrode spacing*

In Fig. 8, a schematic representation of the outcomes of tests using the batch recirculation EC approach is shown. The distance between the anode and cathode ranged from 1 to 4 cm. When the distance between the anode and cathode was extended, the percentage of COD removal efficiency was decreased from 95.27 % to 66.40 %, while the energy consumption increased from 7.73 to 18.3 kWhr m^{-3}. When the distance between the anode and the cathode was increased, electrical current reduced, voltage and IR-drop increased [45,46]. Furthermore, less ion interaction with hydroxide polymer and electrostatic attraction,

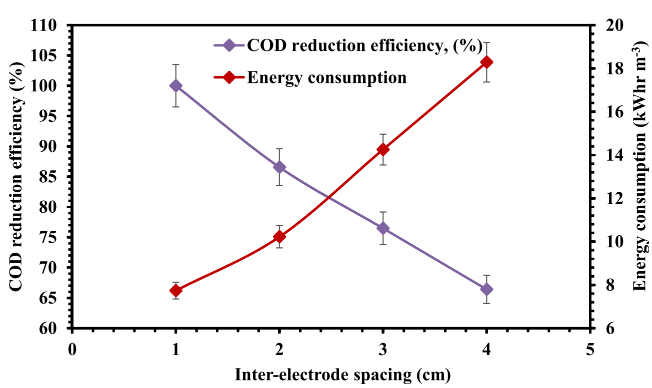

Fig. 8. Effect of inter-electrode spacing on % COD reduction efficiency and energy consumption by using BRE process (Experimental conditions: COD - 3600 mg L^{-1}; wastewater pH - 7; current - 0.56 Amp; combination of electrodes - Fe/Fe; recirculation flow rate - 15 L h^{-1}; concentration of electrolytes - 5 g L^{-1} and treatment time - 180 min).

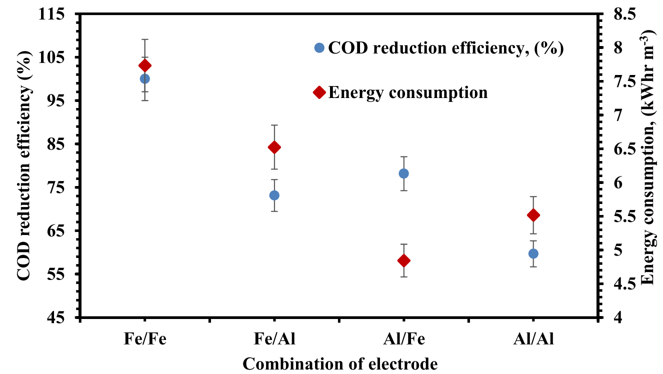

Fig. 9. Effect of combination electrode on % COD reduction efficiency and energy consumption by using BRE process (Experimental conditions: COD - 3600 mg L^{-1}; wastewater pH - 7; current - 0.56 Amp; inter-electrode spacing - 1 cm; recirculation flow rate - 15 L h^{-1}; concentration of electrolytes - 5 g L^{-1} and treatment time - 180 min).

respectively, result in a reduction in the percentage of COD removal and an increase in energy consumption of distillery effluent [47,48]. Therefore, the optimum value of the inter-electrode spacing between the anode and cathode was chosen as 1 cm in order to achieve a higher percentage of COD removal while using the least amount of energy possible during the batch recirculation EC process.

3.1.8. *Effect of combination electrode*

Electrode material is a crucial design element since the electrode is the electrocoagulation process' core aspect[49]. Iron (Fe) and aluminum (Al) electrodes are the most often utilized electrode materials in the EC process because they are affordable, easily available, and have a history of efficiency [50]. One of the most crucial elements in batch recirculation EC treatment was the material of the combination electrode[20]. Combination electrodes such as Fe/Fe (anode/cathode), Fe/Al, Al/Fe, and Al/Al were evaluated in batch recirculation EC treatment to achieve the optimum COD reduction performance, as shown in Fig. 9. As shown in fig, batch recirculation EC - Fe/Fe had the maximum COD removal efficiency at 180 min (100 %), whereas Fe/Al, Al/Fe, and Al/Al had respective removal efficiencies of 73.12 %, 78.16 %, and 59.68 %, respectively. Due to the higher adsorption capacity of hydrous ferrous and iron hydroxides than aluminum hydroxides, batch recirculation EC treatment with a Fe anode produced the highest COD removal performance [51]. Similar results were made by other authors, who claimed that the generated iron hydroxides had outstanding adsorption capacity and good removal efficacy when used in the EC treatment of metal-containing wastewater [52]. Due to the metal hydroxides' ability to successfully adsorb the arsenate, Kumar et al., 2004 discovered that a Fe-based electrode had a considerably greater removal efficiency for removing arsenate than an Al electrode [53]. Because it was more successful at eliminating COD, subsequent studies employed batch recirculation EC treatment with Fe anode/cathode. In addition, the treated DIW was tested for iron concentration (1.50 mg L^{-1}) and found to be safe to public and environmental health.

4. Conclusion

As a potential solution for treating distillery industrial effluent, an electrocoagulation process with continuous recirculation was tested and assessed. It has been found, on the basis of the experimental investigations that were carried out, that batch recirculation electrocoagulation (BRE) utilizing Fe electrodes is an effective and low-cost solution for the treatment of distillery industrial effluent. The effectiveness of the process as well as the influence of operating parameters such as concentration of chemical oxygen demand (COD), pH of wastewater, current, inter-electrode spacing, combination of electrodes, recirculation flow rate, concentration of electrolytes, and treatment time of distillery industrial wastewater have been investigated. It has been noticed that both the percentage of color and COD that was removed and the amount of energy used dramatically increased when the treatment time was raised to 240 min and the current was increased to 0.56 Amps. When the starting pH of the effluent is neutral, it may be possible to accomplish more pollutant removal while maintaining the needed level of energy consumption. The percentage of COD removal decreased from 100 % to 53 % when the COD concentration increased from 1800 to 6000 ppm. Additionally, the Fe/Fe electrode combination with a minimum inter-electrode distance between the anode and cathode of 1 cm had high color and COD removal. This combination had high pollutant removal. The quantity of solid sludge formed were evaluated and reported, also it is totally recover the treated wastewater as clean water under the optimal process conditions. In conclusion, the batch recirculation electrocoagulation technique is a productive and cost-effective way to treat industrial effluent and wastewater.

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. **S 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.

Data availability

The authors do not have permission to share data.

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.

Acknowledgements

The authors are very thankful and acknowledge the administration of Faculty of Civil and Environmental Engineering, Jimma Institute of Technology, Jimma University, Jimma, Ethiopia, Po Box – 378, as well as the authors are grateful for the support from the Tianchi Doctor Program of Xinjiang Uygur Autonomous Region [Grant Number: E33H6301], and the Natural Science Foundation of Xinjiang Uygur Autonomous Region [Grant Number: 2022D01A335].

References

[1] C. Zampeta, M. Mastrantonaki, N. Katsaouni, Z. Frontistis, P.G. Koutsoukos, D.V. Vayenas, Treatment of printing ink wastewater using a continuous flow electrocoagulation reactor, J. Environ. Manag. 314 (2022) 115033, , https://doi.org/10.1016/j.jenvman.2022.115033
[2] P.P. Das, M. Sharma, M.K. Purkait, Recent progress on electrocoagulation process for wastewater treatment: a review, Sep. Purif. Technol. 292 (2022) 121058, , https://doi.org/10.1016/j.seppur.2022.121058
[3] D. Ankoliya, A. Mudgal, M.K. Sinha, V. Patel, J. Patel, Application of electrocoagulation process for the treatment of dairy wastewater: a mini review, Mater. Today Proc. (2022), https://doi.org/10.1016/j.matpr.2022.10.254
[4] 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. Afr. 16 (2022) e01238, , https://doi.org/10.1016/j.sciaf.2022.e01238
[5] F. Ilhan, K. Ulucan-Altuntas, Y. Avsar, U. Kurt, A. Saral, Electrocoagulation process for the treatment of metal-plating wastewater: kinetic modeling and energy consumption, Front. Environ. Sci. Eng. 13 (2019) 73, https://doi.org/10.1007/s11783-019-1152-1
[6] Z. Qi, S. You, R. Liu, C.J. Chuah, Performance and mechanistic study on electrocoagulation process for municipal wastewater treatment based on horizontal bipolar electrodes, Front. Environ. Sci. Eng. 14 (2020) 40, https://doi.org/10.1007/s11783-020-1215-3
[7] A. Aygun, B. Nas, M.F. Sevimli, Treatment of reactive dyebath wastewater by electrocoagulation process: optimization and cost-estimation, Korean J. Chem. Eng. 36 (2019) 1441–1449, https://doi.org/10.1007/s11814-019-0334-7
[8] F. Ozyonar, B. Karagozoglu, Systematic assessment of electrocoagulation for the treatment of marble processing wastewater, Int. J. Environ. Sci. Technol. 9 (2012) 637–646, https://doi.org/10.1007/s13762-012-0093-z
[9] F. Zhang, C. Yang, H. Zhu, Y. Li, W. Gui, Optimal setting strategy of electrocoagulation process in heavy metal wastewater treatment plant, J. Environ. Manag. 310 (2022) 114724, , https://doi.org/10.1016/j.jenvman.2022.114724
[10] A.G. Merma, B.F. Santos, A.S.C. Rego, R.R. Hacha, M.L. Torem, Treatment of oily wastewater from mining industry using electrocoagulation: fundamentals and process optimization, J. Mater. Res. Technol. 9 (2020) 15164–15176, https://doi.org/10.1016/j.jmrt.2020.10.107
[11] A. Shahedi, A.K. Darban, F. Taghipour, A. Jamshidi-Zanjani, A review on industrial wastewater treatment via electrocoagulation processes, Curr. Opin. Electrochem. 22 (2020) 154–169, https://doi.org/10.1016/j.coelec.2020.05.009
[12] P. Asaithambi, Studies on various operating parameters for the removal of COD from pulp and paper industry using electrocoagulation process, Desalin. Water Treat. 57 (2016) 11746–11755, https://doi.org/10.1080/19443994.2015.1046149
[13] M. Ebba, P. Asaithambi, E. Alemayehu, Development of electrocoagulation process for wastewater treatment: optimization by response surface methodology, Heliyon 8 (2022) e09383, , https://doi.org/10.1016/j.heliyon.2022.e09383
[14] X. Chen, P. Ren, T. Li, J.P. Trembly, X. Liu, Zinc removal from model wastewater by electrocoagulation: processing, kinetics and mechanism, Chem. Eng. J. 349 (2018) 358–367, https://doi.org/10.1016/j.cej.2018.05.099
[15] B.K. Zaied, M. Rashid, M. Nasrullah, A.W. Zularisam, D. Pant, L. Singh, A comprehensive review on contaminants removal from pharmaceutical wastewater by electrocoagulation process, Sci. Total Environ. 726 (2020) 138095, , https://doi.org/10.1016/j.scitotenv.2020.138095
[16] A. Shokri, M.S. Fard, A critical review in electrocoagulation technology applied for oil removal in industrial wastewater, Chemosphere 288 (2022) 132355, , https://doi.org/10.1016/j.chemosphere.2021.132355
[17] N.G. Yasri, M. Ingelsson, M. Nightingale, A. Jaggi, M. Dejak, K. Kryst, T.B.P. Oldenburg, E.P.L. Roberts, Investigation of electrode passivation during electrocoagulation treatment with aluminum electrodes for high silica content produced water, Water Sci. Technol. 85 (2022) 925–942, https://doi.org/10.2166/wst.2022.012
[18] A.S. Fajardo, R.C. Martins, D.R. Silva, C.A. Martínez-Huitle, R.M. Quinta-Ferreira, Dye wastewaters treatment using batch and recirculation flow electrocoagulation systems, J. Electroanal. Chem. 801 (2017) 30–37, https://doi.org/10.1016/j.jelechem.2017.07.015
[19] M. Mousazadeh, E.K. Niaragh, M. Usman, S.U. Khan, M.A. Sandoval, Z. Al-Qodah, Z. Bin Khalid, V. Gilhotra, M.M. Emamjomeh, A critical review of state-of-the-art electrocoagulation technique applied to COD-rich industrial wastewaters, Environ. Sci. Pollut. Res. 28 (2021) 43143–43172, https://doi.org/10.1007/s11356-021-14631-w
[20] M. Kobya, P.I. Omwene, Z. Ukundimana, Treatment and operating cost analysis of metalworking wastewaters by a continuous electrocoagulation reactor, J. Environ. Chem. Eng. 8 (2020) 103526, , https://doi.org/10.1016/j.jece.2019.103526
[21] H. Ehsani, N. Mehrdadi, G. Asadollahfardi, G.N. Bidhendi, G. Azarian, Continuous electrocoagulation process for pretreatment of high organic load moquette industry wastewater containing polyvinyl acetate: a pilot study, Int. J. Environ. Anal. Chem. 102 (2022) 2260–2276, https://doi.org/10.1080/03067319.2020.1753716
[22] N.S. Graça, A.M. Ribeiro, A.E. Rodrigues, Removal of fluoride from water by a continuous electrocoagulation process, Ind. Eng. Chem. Res. 58 (2019) 5314–5321, https://doi.org/10.1021/acs.iecr.9b00019
[23] Z. Wu, J. Dong, Y. Yao, Y. Yang, F. Wei, Continuous flowing electrocoagulation reactor for efficient removal of azo dyes: kinetic and isotherm studies of adsorption, Environ. Technol. Innov. 22 (2021), https://doi.org/10.1016/j.eti.2021.101448
[24] P. Parthasarathy, S.K. Narayanan, Effect of hydrothermal carbonization reaction parameters on, Environ. Prog. Sustain. Energy 33 (2014) 676–680, https://doi.org/10.1002/ep
[25] K. Hendaoui, M. Trabelsi-Ayadi, F. Ayari, Optimization and mechanisms analysis of indigo dye removal using continuous electrocoagulation, Chin. J. Chem. Eng. 29 (2021) 242–252, https://doi.org/10.1016/j.cjche.2020.07.065
[26] M. Kobya, E. Gengec, E. Demirbas, Operating parameters and costs assessments of a real dyehouse wastewater effluent treated by a continuous electrocoagulation process, Chem. Eng. Process. Process. Intensif. 101 (2016) 87–100, https://doi.org/10.1016/j.cep.2015.11.012
[27] S. Zodi, B. Merzouk, O. Potier, F. Lapicque, J.P. Leclerc, Direct red 81 dye removal by a continuous flow electrocoagulation/flotation reactor, Sep. Purif. Technol. 108 (2013) 215–222, https://doi.org/10.1016/j.seppur.2013.01.052
[28] S. Hu, D. Li, C. Huang, D. Sun, X. Yuan, A continuous electrocoagulation system with pH auto-adjusting by endogenous products to treat Cr(VI)-contaminated soil flushing solution, Sep. Purif. Technol. 189 (2017) 213–219, https://doi.org/10.1016/j.seppur.2017.07.081
[29] S. Abbasi, M. Mirghorayshi, S. Zinadini, A.A. Zinatizadeh, A novel single continuous electrocoagulation process for treatment of licorice processing wastewater: optimization of operating factors using RSM, Process Saf. Environ. Prot. 134 (2020) 323–332, https://doi.org/10.1016/j.psep.2019.12.005

[30] S. Mahesh, K.K. Garg, V.C. Srivastava, I.M. Mishra, B. Prasad, I.D. Mall, Continuous electrocoagulation treatment of pulp and paper mill wastewater: operating cost and sludge study, RSC Adv. 6 (2016) 16223–16233, https://doi.org/10.1039/c5ra27486a
[31] P. Cañizares, F. Martínez, J. Lobato, M.A. Rodrigo, Break-up of oil-in-water emulsions by electrochemical techniques, J. Hazard. Mater. 145 (2007) 233–240, https://doi.org/10.1016/j.jhazmat.2006.11.018
[32] P. Asaithambi, R. Govindarajan, M.B. Yesuf, P. Selvakumar, E. Alemayehu, Investigation of direct and alternating current-electrocoagulation process for the treatment of distillery industrial effluent: studies, Oper. Parameters, J. Environ. Chem. Eng. 9 (2021) 104811, , https://doi.org/10.1016/j.jece.2020.104811
[33] P. Asaithambi, M. Susree, R. Saravanathamizhan, M. Matheswaran, Ozone assisted electrocoagulation for the treatment of distillery effluent, Desalination 297 (2012) 1–7, https://doi.org/10.1016/j.desal.2012.04.011
[34] A.S. Koparal, Y.Ş. Yildiz, B. Keskinler, N. Demircioğlu, Effect of initial pH on the removal of humic substances from wastewater by electrocoagulation, Sep. Purif. Technol. 59 (2008) 175–182, https://doi.org/10.1016/j.seppur.2007.06.004
[35] Y.Ş. Yıldız, A.S. Koparal, B. Keskinler, Effect of initial pH and supporting electrolyte on the treatment of water containing high concentration of humic substances by electrocoagulation, Chem. Eng. J. 138 (2008) 63–72, https://doi.org/10.1016/j.cej.2007.05.029
[36] P. Maha Lakshmi, P. Sivashanmugam, Treatment of oil tanning effluent by electrocoagulation: influence of ultrasound and hybrid electrode on COD removal, Sep. Purif. Technol. 116 (2013) 378–384, https://doi.org/10.1016/j.seppur.2013.05.026
[37] J.N. Hakizimana, B. Gourich, M. Chafi, Y. Stiriba, C. Vial, P. Drogui, J. Naja, Electrocoagulation process in water treatment: a review of electrocoagulation modeling approaches, Desalination 404 (2017) 1–21, https://doi.org/10.1016/j.desal.2016.10.011
[38] K. Thirugnanasambandham, V. Sivakumar, J.P. Maran, Response surface modelling and optimization of treatment of meat industry wastewater using electrochemical treatment method, J. Taiwan Inst. Chem. Eng. 46 (2015) 160–167, https://doi.org/10.1016/j.jtice.2014.09.021
[39] R. Sridhar, V. Sivakumar, V. Prince Immanuel, J. Prakash Maran, Treatment of pulp and paper industry bleaching effluent by electrocoagulant process, J. Hazard. Mater. 186 (2011) 1495–1502, https://doi.org/10.1016/j.jhazmat.2010.12.028
[40] J. Lu, Q. Zhuo, X. Ren, Y. Qiu, Y. Li, Z. Chen, K. Huang, Treatment of wastewater from adhesive-producing industries by electrocoagulation and electrochemical oxidation, Process Saf. Environ. Prot. 157 (2022) 527–536, https://doi.org/10.1016/j.psep.2021.10.035
[41] B. Merzouk, B. Gourich, A. Sekki, K. Madani, C. Vial, M. Barkaoui, Studies on the decolorization of textile dye wastewater by continuous electrocoagulation process, Chem. Eng. J. 149 (2009) 207–214, https://doi.org/10.1016/j.cej.2008.10.018
[42] J. Behin, N. Farhadian, M. Ahmadi, M. Parvizi, Ozone assisted electrocoagulation in a rectangular internal-loop airlift reactor: application to decolorization of acid dye, J. Water Process Eng. 8 (2015) 171–178, https://doi.org/10.1016/j.jwpe.2015.10.003
[43] S. Song, J. Yao, Z. He, J. Qiu, J. Chen, Effect of operational parameters on the decolorization of C.I. Reactive Blue 19 in aqueous solution by ozone-enhanced electrocoagulation, J. Hazard. Mater. 152 (2008) 204–210, https://doi.org/10.1016/j.jhazmat.2007.06.104
[44] M. Sedaghat, B. Vahid, S. Aber, M.H. Rasoulifard, A. Khataee, N. Daneshvar, Electrochemical and photo-assisted electrochemical treatment of the pesticide imidacloprid in aqueous solution by the Fenton process: effect of operational parameters, Res. Chem. Intermed. 42 (2016) 855–868, https://doi.org/10.1007/s11164-015-2059-5
[45] W.-L. Chou, C.-T. Wang, K.-Y. Huang, Effect of operating parameters on indium (III) ion removal by iron electrocoagulation and evaluation of specific energy consumption, J. Hazard. Mater. 167 (2009) 467–474, https://doi.org/10.1016/j.jhazmat.2009.01.008
[46] A. Dalvand, M. Gholami, A. Joneidi, N.M. Mahmoodi, Dye removal, energy consumption and operating cost of electrocoagulation of textile wastewater as a clean process, Clean. Soil Air Water 39 (2011) 665–672, https://doi.org/10.1002/clen.201000233
[47] N. Daneshvar, A.R. Khataee, A.R. Amani Ghadim, M.H. Rasoulifard, Decolorization of C.I. Acid Yellow 23 solution by electrocoagulation process: investigation of operational parameters and evaluation of specific electrical energy consumption (SEEC), J. Hazard. Mater. 148 (2007) 566–572, https://doi.org/10.1016/j.jhazmat.2007.03.028
[48] 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
[49] G.K. Akkaya, Treatment of petroleum wastewater by electrocoagulation using scrap perforated (Fe-anode) and plate (Al and Fe-cathode) metals: optimization of operating parameters by RSM, Chem. Eng. Res. Des. 187 (2022) 261–275, https://doi.org/10.1016/j.cherd.2022.08.048
[50] M. Ebba, P. Asaithambi, E. Alemayehu, Investigation on operating parameters and cost using an electrocoagulation process for wastewater treatment, Appl. Water Sci. 11 (2021) 175, https://doi.org/10.1007/s13201-021-01517-y
[51] T.S. Kazeem, B.A. Labaran, H.U.R. Ahmed, T. Mohammed, M.H. Essa, M.S. Al-Suwaiyan, M.S. Vohra, Treatment of aqueous selenocyanate anions using electrocoagulation, Int. J. Electrochem. Sci. 14 (2019) 10538–10564, https://doi.org/10.20964/2019.11.51
[52] T. Li, H. Xu, Y. Zhang, H. Zhang, X. Hu, Y. Sun, X. Gu, J. Luo, D. Zhou, B. Gao, Treatment technologies for selenium contaminated water: a critical review, Environ. Pollut. 299 (2022) 118858, , https://doi.org/10.1016/j.envpol.2022.118858
[53] P. Ratna Kumar, S. Chaudhari, K.C. Khilar, S.P. Mahajan, Removal of arsenic from water by electrocoagulation, Chemosphere 55 (2004) 1245–1252, https://doi.org/10.1016/j.chemosphere.2003.12.025