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Treatment of cattle slaughterhouse wastewater by sequential coagulation-flocculation/electrooxidation process

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ABSTRACT

This study aimed to investigate the applicability and efficiency of sequential coagulationflocculation (CF) and electrooxidation (EO) processes for cattle slaughterhouse wastewater by evaluating treatment efficiency and total operating cost values together. The effect of two different coagulant dosages (FeCl3 and alum) in the CF process and operating parameters such as current density (5 to 30 mA/cm²), wastewater flow rate (0.9 to 3.6 L/h), and supporting electrolyte concentration (1 to 3 g NaCl/L) in the EO process on chemical oxygen demand (COD) and turbidity removal were investigated. During the first part of the study, the FeCl₃ coagulant dosage worked better than other coagulants, eliminating 50% of the COD and 68% of the turbidity. Due to the insufficient removal efficiencies of COD and turbidity, a secondary treatment was required. In the second part of the study, a boron-doped diamond (BDD) electrode was used to treat the coagulated effluent in a continuous EO reactor. The COD and turbidity removal efficiency under optimum treatment conditions ($j = 30 \text{ mA/cm}^2$, Q = 0.9 L/h, pH = 8.5, SE = 3.0 g NaCl/L, and hydraulic retention time = 1 hour) were calculated as 97.2% and 99.9%, respectively. At these optimum conditions, the energy consumption and total operating cost were calculated as 91.1 kWh/m³ (73.5 kWh/kg COD) and 3.50 US \$/m³ (1.5 US \$/kg COD), respectively. As a result, combined coagulation-flocculation and electrooxidation processes have proven to be very successful and cost-efficient for treating cattle slaughterhouse wastewater.

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1. Introduction

The United States Environmental Protection Agency (USEPA) has identified effluent from slaughterhouses as one of the most dangerous types of wastewater. The Organization for Economic Co-operation and Development has projected that the annual meat output will reach 366 million metric tons by 2029 [1]. A slaughterhouse produces wastewater from both the slaughtering process and the cleaning of the intestines. This wastewater contains high levels of organic substances, including chemical oxygen demand (COD), biological oxygen demand (BOD), total phosphorus (TP), total nitrogen (TN), total suspended solids (TSS), and oil and greases [2,3].

Efficiently treating slaughterhouse wastewater is crucial for both human well-being and environmental preservation. Commonly, slaughterhouse wastewater treatment approaches use a physicochemical treatment process [4,5], a biological (aerobic and anaerobic) treatment process [6-8], and an advanced treatment process [9,10]. Typically, biological processes are the primary techniques used to treat slaughterhouse effluent. Nevertheless, aerobic treatment requires a substantial oxygen supply of >100 mg O₂/L h, generates excessive quantities of biomass at around 8 kg/m³, and demands significant energy consumption. The accumulation of suspended particles and floating oils in the anaerobic treatment reactor has a negative impact on SWW treatment, making the aerobic and anaerobic processes for SWW treatment costly and time-consuming [11-13].

Among physicochemical processes, the coagulationflocculation (CF) process is often used in slaughterhouse wastewater treatment due to its notable efficiency and low cost, proving to be effective in removing COD, BOD, TSS, and turbidity [2]. Three distinct steps, namely coagulation, flocculation, and sedimentation, comprise the CF process. The coagulation phase's objective is to induce colloidal suspension destabilization by introducing coagulant chemicals. This is achieved by disrupting the repulsive forces arising from the presence of similar polarity charges on the colloids' surface. Flocculation makes colloidal substances even less stable, mostly by bringing together particles that aren't stable, which makes micro-flakes and then bigger flakes that settle. The often used coagulants in the CF process are aluminum and iron salts, namely aluminum chloride (AlCl₃), aluminum sulfate (Al₂(SO₄)₃), ferric chloride (FeCl₃), and ferric sulfate (Fe₂(SO₄)₃), due to their effectiveness, cost-effectiveness, and ease of use. Nevertheless, the CF process alone may not be sufficiently effective in eliminating organic compounds from slaughterhouse effluent, necessitating additional procedures to eliminate the residual organic matter down [14].

At present, electrochemical oxidation has emerged as a very effective approach for the treatment of several categories of wastewater. Electrooxidation (EO) offers several benefits as compared to traditional and alternative advanced oxidation process (AOP) treatment procedures. These steps prevent the production of extra wastes such as used sorbents, contaminated retentate, waste ferric hydroxide, or wasteactivated sludge, which typically occur during adsorption, filtering, Fenton reactions, and biological processes, respectively. EO has the advantage of being easy to examine during operation and does not require additional chemicals for process control [15]. When exposed to an electric current, dangerous or non-biodegradable chemicals experience oxidation, leading to their transformation into biodegradable substances or complete oxidation into water (H₂O), carbon dioxide (CO_2) , and simpler organic molecules [16]. In environmental science, the degradation of organic compounds occurs primarily via direct and/or indirect oxidation. The EO process generates hydroxyl radicals (•OH) and other reactive oxidant species (H₂O₂, Cl₂, and O₃) that are produced at the anode surface (A) during the treatment. These species then proceed to oxidize the contaminants directly at the anode surface. Anodes with high reactivity, such as RuO₂, IrO₂, or Pt, promote the partial and selective oxidation of pollutants.

In contrast, anodes with low reactivity, such as BDD, PbO₂, or SnO₂, may achieve total combustion. As a result, inactive anodes are considered favorable electrodes for the full conversion of organic compounds into CO₂ during the wastewater treatment process [17]. There has been a notable surge of interest in using BDD electrodes for chemically degrading pollutants in recent years. The BDD electrode exhibits a wide range of potential values in both water-based and non-water-based solutions, demonstrating good stability and corrosion resistance. Furthermore, it has a non-reactive surface that exhibits little adsorption and a low baseline current [18]. However, as with other procedures, EO has drawbacks. The mentioned phenomena include abrasion, passivation, polarization of electrodes, and significant energy consumption. Electrochemical technologies exhibit much higher energy consumption as compared to other technologies. To reduce the excessive energy consumption of the EO process, it is advantageous to treat the wastewater beforehand using other methods. There are many studies in the existing literature on single and sequential electrochemical processes for the treatment of cattle SWW; to the authors' knowledge, there is no research in the existing literature on the use of sequential coagulation-flocculation/electrooxidation processes for cattle SWW treatment.

Therefore, in light of the existing research gap, this study investigates the treatment of cattle slaughterhouse plant wastewater by pre-treatment, including coagulationflocculation (CF), in a continuous electrooxidation reactor using BDD electrodes. Initially, we used the CF process to remove colloidal and suspended particles from the cattle SWW. Then, a continuous electrooxidation reactor was used to remove the remaining persistently dissolved organic compounds. The CF process involved testing two types of coagulant dosages to investigate their effect on COD and turbidity removal. The effectiveness of the BDD anode was investigated in the CEO process. In particular, the impact of the applied current density, wastewater flow rate, and supporting electrolyte concentration on the treatment performance was studied. Furthermore, the specific energy consumption for COD, the energy consumption of the process, and the total operating costs were calculated.

2. Materials and methods

2.1. The Bishkek slaughterhouse plants

Bishkek serves as the capital and most populous city of Kyrgyzstan (Kyrgyz Republic). The city is located at

coordinates 42°52′29″N and 74°36′44″E, with an altitude of around 800 meters. It is positioned at the northern edge of the Kyrgyz Ala-Too Range, which is an extension of the Tian-Shan mountain range. The elevation of these mountains reaches 4,895 meters. The city spans a total area of 170 square kilometers. The mean annual precipitation is around 440 mm. The average daily maximum temperatures vary from 3 °C in January to around 31 °C in July (almost -10°C in winter). The official population of Bishkek in 2021 is about 110,472 individuals, with a population growth rate of roughly 2.1%, above the national average of 1.7%. Nevertheless, a significant number of individuals (~400.000 people) reside in Bishkek without formal recognition. The meat industry has a prominent position in Kyrgyzstan, similar to other Central Asian nations. The rural population relies heavily on livestock farming as their primary source of income and subsistence. In 2020, the per capita consumption of beef in Kyrgyzstan was 14.9 kilograms. The present number signifies a decline of 3.19% in comparison to the value seen in the preceding year. Within the historical framework, the average individual consumption of beef in Kyrgyzstan reached its highest level at 21.0 kg in 2002 and then dropped to its lowest point at 14.0 kg in 2010. Kyrgyzstan ranks 42nd out of 165 countries in terms of per capita consumption of beef. Currently, there are about 12 functioning slaughterhouses in Bishkek. These slaughterhouses have a daily average of slaughtering about 70

Table 1. Characteristics of the wastewater used in this study.

cattle and over 410 sheep and goats. The slaughterhouses produce an estimated volume of 80 m^3 of effluent every day.

2.2. Materials

The cattle slaughterhouse wastewater was obtained from the slaughterhouse plant located in Bishkek City (Kyrgyzstan). The slaughterhouse plant has a daily slaughter capacity of 20 cattle and produces an average of 10-15 m³ of wastewater. Samples were collected in polypropylene bottles, following the protocols outlined in the Standard Method [19]. The obtained samples were then transported under cold conditions and thereafter kept at a temperature of 4 °C until they were ready for use. The characteristics of dissolved oxygen, pH, electrical conductivity, and temperature were investigated while collecting wastewater from the slaughterhouse plant. (Table 1) presents the primary characteristics of cattle slaughterhouse wastewater. In the CF process, two different coagulant dosages were used (FeCl₃ and Al₂(SO₄)₃). All chemicals used in this study were analytical quality Merck products. The BDD, which consists of a layer of boron diamond film applied over a 1.5 mm niobium plate, was acquired from DiaCCon GmbH in Germany. Also, the cathode was comprised of a rectangular electrode made of stainless steel (316 AISI).

Parameters	SWW	Effluent of CF + CEO		
pH	7.4 ± 0.20	8.35 ± 0.10		
Temperature (T, °C)	17 ± 2	<30		
Turbidity (NTU)	175 ± 5	<1		
Dissolved Oxygen (DO, mg/L)	7.25 ± 0.25	7.85 ± 0.10		
Electrical Conductivity (EC, µS/cm)	1800 ± 10	11000 ± 100		
Total dissolved solids (TSS, mg/L)	2350 ± 30	<3		
Volatile suspended solids (VSS, mg/L)	450 ± 10	<1		
Total dissolved solids (TDS, mg/L)	1100 ± 20	6000 ± 100		
Oil and grease (mg/L)	250 ± 10			
Chemical oxygen demand (COD, mg/L)	4100 ± 100	<100		
Total phosphorus (TP, mg/L)	110 ± 5			
Total nitrogen (TN, mg/L)	330 ± 10			

2.3. Experimental apparatus and procedure

A series of standard jar test tests evaluated the coagulationflocculation (CF)process. Six beakers, each with a volume of 1000 mL, were used for this purpose. The experiments were conducted under controlled conditions at a room temperature of 25 ± 1 °C. In this experimental study, a total of six distinct coagulant dosages (100, 500, 800, 1000, 2000, and 3000

mg/L) were examined for each coagulant to ascertain the optimum coagulant dosage. In CF studies, after adding a coagulant dosage to wastewater, pH was adjusted to 6.5 for alum and 8.5 for FeCl₃. pH levels were adjusted using 1 M HCl and NaOH solutions. The CF included three consecutive stages: an initial phase of quick mixing at a speed of 220 rpm for 3 minutes, which was then followed by a gradual mixing stage lasting for 25 minutes at a speed of 40 rpm. Finally, the process concluded with a settling step that lasted for an additional hour. Subsequently, water samples were collected from the water surface (supernatant) to analyze COD and turbidity. These samples were then subjected to filtration using a membrane filter with a pore size of 0.45 μ m. The determination of the volume and quantity of the settled sludge was also conducted.

The experimental investigation included the use of a cylindrical plastic reactor for conducting experiments on the CEO reactor. The capacity of the reactor was 0.9 L, while the diameter and height were 110 mm and 140 mm, respectively. In each experimental trial, a volume of 0.8 L of wastewater was introduced into the reactor. The wastewater was then consistently supplied to the reactor using a peristaltic pump (Masterflex, Model NO: 7553-76). The wastewater was subjected to continuous stirring at a rate of 250 rpm using a magnetic stirrer of the (Heidolph MR Hei-End) type to achieve a homogenous solution in the reactor. In CEO reactor investigations, the anode used was BDD and the cathode was stainless steel. The dimensions of the used anode and cathode electrodes were as follows: 200 mm (length) x 60 mm (width) x 3 mm (thickness). The electrodes were arranged in a vertical parallel configuration inside the reactor. In the CEO reactor,

the contact area between the electrodes in the effluent is measured to be 100 mm, with a separation distance of 15 mm. The positioning of the electrodes in the CEO reactor was set at a distance of 15 mm from the bottom. The anode electrode has an active surface area of 120 cm². The electrodes were linked to a digital (DC) power source (Gwinstek DC SPS-60: 0-60 V, 0-6 A) via the galvanostatic connection. The direct current (DC) power source was used to establish the specific electrical currents required for each experimental procedure. The CEO reactor output was sampled at various time intervals, and each sample underwent filtration using a 0.45 µm filter paper before analysis. Subsequently, studies were conducted to determine the levels of COD and turbidity.

2.4. Analytical methods

The analysis of all samples was conducted by the Standard Method [19]. The dichromate open reflux technique (5220-B standard) was used for COD analysis, (ZBK Turb 550 IR) for turbidity, (Thermo Scientific Eutech pH 150) for pH, and (YSI 30) for conductivity.

The assessment of total operating cost (OC) is important in evaluating a wastewater treatment methodology. The energy consumption for the CEO reactor and specific energy consumption for removed kg COD from 1 m³ wastewater was determined by the use of Eqs (1) and (2), respectively. The energy consumption of the peristaltic pump was determined using Eq (3), while the energy consumption of the magnetic stirrer was determined using Eq (4), and the overall energy consumption was computed by using Eq (5) [20].

$$C_{\text{CEO, energy}}(kWh/m^3) = \frac{l \times l_{\text{CEO}} \times b}{v}$$
(1)

$$C_{\text{CEO, energy}}(kWh/kg \ COD) = \frac{l \times t_{\text{CEO}} \times i}{\Delta C \times v}$$
(2)

$$C_{\text{CEO, pump energy}}(kWh/m^3) = \frac{P \cdot t_{\text{CFEO}}}{v}$$
(3)

$$C_{\text{magnetic stirrer, energy}}(kWh/m^3) = \frac{N \times t}{\eta \ (\%) \times v}$$
(4)

$$C_{\text{total, energy}} = C_{\text{CEO, energy}} + C_{\text{magnetic stirrer, energy}} + C_{\text{CEO, pump energy}}$$
(5)

In this context, the variable (i) represents the applied current in amperes (A), U denotes the cell voltage in volts (V), t_{CEO} signifies the operating duration of the CEO reactor in hours, v represents the volume of treated wastewater in the CEO reactor in cubic meters (m³), and ΔC denotes the difference in COD between the influent and effluent to the reactor, measured in kg/m³, the variable P (kW) represents the power of the peristaltic pump, with the motor power of the pump utilized denoted as 760 W, and N representing the magnetic stirrer power, η is the magnetic stirrer power efficiency. The total operating cost for the CF, CEO, and CF+CEO processes was determined by using Eqs (6), (7), and (8) [20].

$$OC_{CF} (US \s/m^3) = \alpha C_{energy} + \beta C_{chemical} + \gamma C_{sludge}$$
(6)

$$OC_{CEO} (US \s/m^3) = \alpha_{Cenergy} + \beta C_{chemical}$$
(7)

$$OC_{total} (US \s/m^3) = OC_{CF} + OC_{CEO}$$
(8)

In this context, α represents the unit cost of electrical energy, Cenergy denotes the total energy consumption associated with the processes, β signifies the unit prices of chemicals used in the CF and CEO processes, Cchemical represents the dosage of coagulant (measured in kg), and γ denotes the unit cost of created sludge, with Csludge representing the quantity of sludge formed per unit volume (kg/m³). In December 2023, the Kyrgyzstan market reported an electrical energy (α) price of 0.033 US \$/kWh. The unit prices for the chemicals employed in the study were as follows: FeCl₃ had a unit price of 0.15 US \$/kg, alum had a unit price of 0.22 US \$/kg, NaOH had a unit price of 0.40 US \$/kg, HCl had a unit price of 0.45 US \$/kg, and NaCl had a unit price of 0.05 US \$/kg. The cost of disposing of the CF process sludge, with a moisture content of 25%, in a typical landfill location is estimated to be 0.15 US \$/kg of sludge.

The COD and turbidity removal efficiency (Re), are calculated by using Eq (9).

Removal efficiency (Re, %) = $\frac{C_i - C_t}{c_i} \times 100$ (9) Where C_i is the initial concentration of COD (mg/L) and turbidity (NTU) and C_i is the final concentration of COD and

3. Results and discussion

turbidity.

The present research aimed to examine the removal of COD and turbidity from slaughterhouse wastewater (SWW). The investigation was conducted in two distinct stages. During the first stage, the SWW underwent coagulation-flocculation treatment to eliminate suspended organic matter. This process included the use of two distinct coagulant dosages (alum and FeCl₃) to identify the most efficient one. During the second part of the study, different current densities (5-30 mA/cm²), different wastewater flow rates (0.9-3.6 L/h), and different supporting electrolyte concentrations (1-3 g/L) were used in the continuous electrooxidation reactor to determine the most efficient one in terms of removing COD and turbidity from SWW.

3.1. The impact of coagulant dosage on the COD and turbidity removal efficiency

Coagulation-flocculation (CF) is a chemical-physical treatment process that is extensively used for the treatment of wastewater generated by slaughterhouses. The use of precise

amounts of coagulants enables the precipitation of colloidal suspended particles that do not settle naturally, as well as the removal of small sedimentable solids that are difficult to eradicate by conventional physical treatment methods because of their prolonged sedimentation rate [21]. In the CF process, metal salts for hydrolysis, such as aluminium and iron, are prevalent as key coagulants in the treatment of wastewater. These coagulants play a crucial role in diminishing the levels of particulate matter and dissolved organic compounds within the wastewater. Upon dissolution in water, the aluminium and iron salts undergo a process whereby the metal (M) ions undergo hydration and hydrolysis, resulting in the formation of both monomeric and polymeric species, as well as the precipitation of solid compounds: MOH^{2+} , $M(OH)_{2^+}$, $M_2(OH)_2^{4+}$, M(OH)_4^{5+}, M(OH)_3(s), M(OH)_4. The quality of treated wastewater is influenced by the quantity of coagulant used and the appropriate pH level. Consequently, these elements have significant importance in the coagulation process, since each coagulant necessitates a certain pH range to facilitate effective coagulation [22].

This study conducted jar tests to examine the effects of various coagulant dosages (alum and FeCl₃), including 100, 500, 800, 1000, 2000, and 3000 mg/L, at constant pH levels (8.5 for FeCl₃ and 6.5 for alum), on the removal efficiencies of COD and turbidity from cattle slaughterhouse effluent. As shown in (Figure 1(a)), the effluent COD concentration was found to be 2000-2500 mg/L for FeCl₃ and 2100-2800 mg/L for alum, respectively. On the other hand, the effluent turbidity concentration was found to be 56-102 NTU for FeCl3 and 73-109 NTU for alum, respectively (Figure 1(b)). The effluent COD and turbidity concentrations under optimum coagulant dosages were determined to be 2100 mg/L and 79 NTU for alum and 2000 mg/L and 65 NTU for FeCl₃, respectively. The removal effectiveness of both coagulants remained rather stable for COD and turbidity at coagulant doses above 800 mg/L. According to these results, the optimum coagulant dosage for FeCl₃ and alum in the CF process was taken at 800 mg/L because they hypothesized that a portion of the dissolved COD would become adsorbed onto the metal hydroxyl flocs. However, they observed minimal variation in the COD elimination effectiveness after a specific coagulant dosage.



Figure 1. (a) The COD removal efficiency for $FeCl_3$ and alum coagulant dosages (b) The turbidity removal efficiency for $FeCl_3$ and alum coagulant dosages (conditions; pH = 6.5 for alum and 8.5 for $FeCl_3$).

The total operating cost for electrical energy (the energy consumption of the magnetic stirrer), chemicals (FeCl₃ and alum coagulant dosages, HCl, and NaOH), and sludge disposal were calculated with Eq (<u>6</u>). for the CF process. The total operating costs for the optimum coagulant dosages for the CF process were calculated as 0.45 US \$/m³ (0,43 US \$/kg COD) for FeCl₃ and 0.53 US \$/m³ (0,49 US \$/kg COD) for alum, respectively. Based on the findings, it is evident that the FeCl₃ dosage used in the CF process was determined to be the optimum coagulant dosage for treating SWW wastewater, considering both treatment effectiveness and minimal overall operating cost.

The quantity and properties of the sludge generated during the coagulation-flocculation (CF) process are primarily influenced by the particular coagulant used and the operational parameters. The volume of wet sludge at the base of the vertical-graded glass sedimentation columns, which had an internal diameter of 55 mm and a height of 350 mm, was measured to determine the amount of sludge produced in this

research after the CF process. The measurements were taken at an ambient temperature of 20 °C. The height of the liquid/sludge contact was measured at regular time intervals. Figure 2 shows the mean height fluctuations at the interface between the suspension and the supernatant, represented as the volume (mL) of settled sludge. The process of sludge settling was observed for one hour. This specific time frame was chosen to ensure that the settling process was close to completion after the observation period. A flat horizontal interface is seen in each coagulation process, exhibiting a clear discontinuity between the suspension and supernatant phases that are created after 1 hour (referred to as SV60, representing the sludge volume in mL at the 1-hour mark). The optimum coagulant dosage for SV60 was discovered to be 15 mL for both FeCl₃ and alum sludge (Figure 2). The settling speed of sludge was determined to be 0.0236 cm/s for both FeCl₃ sludge and alum sludge when a coagulant dosage concentration of 800 mg/L was used.



Figure 2. Sludge settling for the coagulation-flocculation process (conditions; 800 mg/L FeCl₃, and alum coagulant dosages, pH = 6.5 for alum and 8.5 for FeCl₃).

3.2. The impact of current density on the COD and turbidity removal efficiency

Significant advancements have been achieved in recent decades in the field of electrochemical technology used for the treatment of effluents that include organic contaminants. Electrochemical technologies are a viable alternative for addressing several environmental challenges within the process sector. This is mostly due to the diverse nature of electrons, which offer efficient, cost-effective, readily automatable, and environmentally friendly reagent options [10,23].

Current density has a significant impact on the formation rate of oxidizing species, making it a significant factor in all electrochemical processes. Due to its ability to regulate the reaction rate, electrooxidation procedures often regard the current density as the primary variable to manipulate (Can et al., 2019). The resulting data showed a COD output of 2000 mg/L and a turbidity output of 65 NTU when determining the optimum coagulant dosage in the CF process. The results show that the best coagulant dosage (800 mg/L of FeCl₃) used for coagulation-flocculation (CF) before the CEO reactor had a big effect on how well the BDD anode electrode worked. An investigation was conducted to analyze the influence of current density on the treatment of SWW using the CEO reactor, which was examined at four different current densities $(5, 10, 20, \text{ and } 30 \text{ mA/cm}^2)$ under optimum conditions: a $BDD + H_2O \rightarrow BDD(\bullet OH) + H^+ + e^-$

wastewater pH value of 8.5, a treatment duration of 3 hours (hydraulic retention time ($HRT = \frac{V_{reactor}}{Q}$) = 1 hour), a supporting electrolyte concentration of 3 g/L NaCl, and a wastewater constant flow rate of 0.9 L/h. The effluent COD concentrations achieved were 600 mg/L (70.0%) for 5 mA/cm², 480 mg/L (76.1%) for 10 mA/cm², 280 mg/L (86.0%) for 20 mA/cm², and 50 mg/L for mg/L (97.2%) for 30 mA/cm², respectively (Figure 3(a)). The effluent turbidity concentration exceeding <1 NTU (100%) was observed across all current densities, as illustrated in (Figure 3(b)).

As shown in Figure 3, there is a direct correlation between an increase in current density and an increase in the effectiveness of removing COD and turbidity. This phenomenon suggests That a higher current density significantly enhances the elimination of contaminants by generating a greater quantity of electrons in the SWW. However, many factors, such as the composition of the pollutants, the density of the electric current, the anode materials, and the pH level of the wastewater, influence the process of mineralization and/or oxidation of pollutants in electrooxidation [24].

In recent years, there has been a significant focus on the utilization of BDD electrodes to chemically eliminate pollutants. The distinguishing characteristic of a BDD electrode compared to conventional anodes is its high oxygen evolution potential [25]. The electrochemical incineration procedure for organic contaminants in wastewater utilizing a BDD electrode is often explained by Eqs. (10-12).

(10)

$$BDD(\bullet OH) + R \rightarrow BDD + oxidation \ products + H^+ + e^-$$
(11)

$$BDD(\bullet OH) \rightarrow BDD + H^+ + O_2 + e^-$$
(12)

The first step in the process entails the generation of hydroxyl radicals (•OH) through the electrolysis of SWW molecules. Organic oxidation is indirectly facilitated by the hydroxyl radical (•OH), which exhibits weak adsorption onto the BDD electrode surface. This adsorption leads to subsequent oxidative degradation as a result of the hydroxyl radical's exceptionally high reactivity. The reaction competes with the

side reaction of oxygen evolution that is generated from the adsorbed hydroxyl radical (•OH). Therefore, the quantification of electrogenerated oxidation radicals, specifically the reactive BDD(•OH), has the potential to serve as an indicator of the indirect electrochemical oxidation capability of the electrode materials [26]



Figure 3. (a) COD removal efficiency for BDD electrodes (b) The turbidity removal efficiency for BDD electrodes (conditions, initial COD $C_0 = 2000 \text{ mg/L}$, initial turbidity $C_0 = 65 \text{ NTU}$, initial pH = 8.5, HRT = 1 hour, Q = 0.9 L/h, SE = 3 g/L NaCl, stirring speed = 300 rpm, and distance between the electrodes = 15 mm).

3.3. The impact of effluent flow rate on the COD and turbidity removal efficiency

The wastewater flow rate (FR) is a crucial factor. FR calculates the duration of time that effluent remains in the EO process. To comprehend An investigation was conducted to examine the effect of different wastewater flow rates on the COD and turbidity removal from the SWW. The study included a flow rate range of 0.9 L/h to 3.6 L/h while maintaining a constant current density of 30 mA/cm². The pH value of the wastewater was 8.50, the supporting electrolyte concentration was 3 g/L NaCl, and the reaction duration was

3 hours. According to (Figure 4(a)), the removal efficiencies increased as the FR decreased. The effluent COD concentrations for wastewater flow rates of 0.9, 1.5, 2.5, and 3.6 L/h were determined as 50 mg/L (97.2%) for 0.9 L/h, 100 mg/L (95.2%) for 1.5 L/h, 152 mg/L (92.4%) for 2.1 L/h, and 216 mg/L (89.2%) for 3.6 L/h, respectively, as shown in Figure 4 (a). Conversely, the turbidity concentration for all FR was determined to be <1 NTU (~99.9 %) according to (Figure 4(b)). The maximum removal effectiveness for COD and turbidity parameters was recorded at the wastewater flow rate of 0.9 L/h.



Figure 4. (a) COD removal efficiency for BDD electrodes (b) The turbidity removal efficiency for BDD electrodes (conditions, initial COD $C_0 = 2000 \text{ mg/L}$, initial turbidity $C_0 = 65 \text{ NTU}$, $j = 30 \text{ mA/cm}^2$, initial pH = 8.5, HRT = 1 hour, SE = 3 g/L NaCl, stirring speed = 300 rpm, and distance between the electrodes = 15 mm).

3.4. The impact of supporting electrolyte concentration on the COD and turbidity removal efficiency

Supporting electrolytes (SE) in the electrochemical process may generate oxidation species and interact with organic contaminants in solutions, significantly impacting the oxidation capacity. Conversely, the addition of supportive electrolytes to wastewater is often seen to result in a reduction in electrical consumption and a significant improvement in removal efficiencies. This is achieved by the formation of radicals and stable oxidants [27,28]. Electrolysis was facilitated by the use of electrolytes since SWW typically includes a certain amount of salt. Electrolytes serve the purpose of facilitating electron transport and ionic conduction. The solution was subjected to different amounts of NaCl (1, 2, and 3 g/L) to examine the impact on COD and turbidity removal.

The oxidation procedure was conducted under optimum conditions, applying a wastewater pH value of 8.5, a current density of 30 mA/cm², using a BDD anode, a treatment duration of 3 hours, and a wastewater constant flow rate of 0.9 L/h. The effluent COD concentrations were determined to be 300 mg/L (85.0%) for 1 g/L of NaCl, 180 mg/L (91.1%) for 2 g/L of NaCl, and 50 mg/L (97.2%) for 3 g/L of NaCl, respectively, as shown in (Figure 5(<u>a</u>)). Furthermore, the turbidity concentration was determined to be <1 NTU (~99.9

%) using the same optimum conditions shown in (Figure 5(b)). The results show that a supporting electrolyte concentration of 3 g/L NaCl achieved the highest COD and turbidity removal efficiency The process of indirect electrochemical oxidation involves the degradation of contaminants by the use of a very potent oxidant on the surface of the anode. The current study included conducting CEO experiments with the incorporation of a concentration of 1-3 g/L NaCl into the SWW. Chlorine is often used as a potent oxidizing agent, derived from the anodic oxidation of chloride. The chloride reactions that occur during electrochemical oxidation are elucidated in Eqs (13-14). Hypochlorous acid (HOCl), chlorine (Cl₂), and hypochlorite (ClO⁻) are recognized as potent oxidizing agents often known as active chlorine species. Hypochlorite acid may facilitate the indirect elimination of organic substances Eq (15) [18]. During indirect oxidation, increasing the concentration of NaCl leads to a gradual shift in the oxygen alteration process towards more favorable potentials, as compared to electrolysis performed without an SE [10,29].

 $2\mathrm{Cl} \to \mathrm{Cl}_2 + 2\mathrm{e}^{-} \tag{13}$

$$Cl_2 + H_2O \rightarrow HOCl + H^+ + Cl^-$$
 (14)

$$HOCl \rightarrow ClO^- + H^+ \tag{15}$$



Figure 5. (a) COD removal efficiency for BDD electrodes (b) The turbidity removal efficiency for BDD electrodes (conditions, initial COD $C_0 = 2000 \text{ mg/L}$, initial turbidity $C_0 = 65 \text{ NTU}$, $j = 30 \text{ mA/cm}^2$, initial pH = 8.5, HRT = 1 hour, Q = 0.9 L/h, stirring speed = 300 rpm, and distance between the electrodes = 15 mm).

3.5. Operating cost and kinetic analysis for CF+CEO reactor

The attainment of cost-effectiveness is a fundamental prerequisite for the practical implementation of wastewater treatment technologies within industrial contexts. Hence, the assessment of the treatment expenses associated with the SWW was conducted to ascertain the viability of the suggested treatment approach. The total operating costs for the CF+CEO reactor include the expenses associated with electrical energy, chemicals, and sludge dewatering and disposal. (Table $\underline{2}$) presents a summary of the relationship between the parameters of the electrooxidation process, such as removal efficiency, energy consumption for the process, overall operating cost, and the effect of the current density, wastewater flow rate. and supporting electrolvte concentration. It is evident that when the current density rises, the removal efficiencies also improve. However, this has a detrimental impact on energy consumption and performance characteristics. Increasing the current density from 5 mA/cm² to 30 mA/cm² results in a 2.5-3.0 times drop in the BBD anode efficiency (η) . Conversely, energy consumption and total operating costs rise by 5.5-6.0 times and 3.0-3.5 times, respectively. When the flow rate of wastewater is raised from 0.9 L/h to 3.6 L/h, the efficiency of the anode (η) decreases by a factor of 1.0-1.5. Additionally, energy consumption and total operating costs rise by a factor of 0.5-1.0 and 1.0-1.5, respectively. On the contrary, when the concentration of the supporting electrolyte is raised from 1 g/L to 3 g/L, the anode efficiency (η) rises by a factor of 1.5–2.0. In contrast, energy consumption and total operating costs fall by a factor of 1.5–2.0 and 1.5–2.0, respectively. The findings suggest that operating at a lower current density is more advantageous in terms of energy efficiency and anode performance. Table 2. Table 2 demonstrates a strong association between the decline in COD concentration with time and the first-order reaction kinetics, as seen by the ideal linear correlations. The study's optimum conditions had an r² value of 0.99 and a k value of 18.7×10^{-3} (min⁻¹), respectively.

j	COD _{rem}	Turbidity _{rem}	SEC		Total OC		η	First order kinetic $\left(-\frac{dC}{dt} = k(Ct)\right)$	
mA/cm ²	%	%	kWh/kg COD	kWh/m ³	US \$/m ³	US \$/kg COD	gCOD/Ahm ²	r^2 k (min ⁻¹)	
5	70.0	98.0	10.2	15.1	1.1	0.27	35.5	0.98	6.3 ×10 ⁻³
10	76.1	99.9	26.4	32.7	1.7	0.54	21.5	0.96	7.2 ×10 ⁻³
20	86.0	99.9	52.3	65.3	2.7	1.10	13.5	0.98	9.8 ×10 ⁻³
30	97.2	99.9	73.5	91.1	3.5	1.50	11.9	0.99	18.7 ×10 ⁻³
Q	CODrem	Turbidity _{rem}	SEC		Total OC		η	First order kinetic $(-\frac{dC}{dt} = k(Ct))$	
L/h	%	%	kWh/kg COD	kWh/m ³	US \$/m ³	US \$/kg COD	gCOD/Ahm ²	r ²	k (min ⁻¹)
0.9	97.2	99.9	73.5	91.2	3.5	1.5	11.9	0.99	18.7 ×10 ⁻³
1.5	95.0	99.9	74.8	91.7	3.5	1.6	11.7	0.97	14.8 ×10 ⁻³
2.1	92.4	99.9	85.2	105.3	4.1	1.8	10.2	0.97	12.6 ×10 ⁻³
3.6	89.2	99.9	97.1	121.4	4.6	2.1	8.9	0.98	11.1 ×10 ⁻³
SE	COD _{rem}	Turbidity _{rem}	SEC		Total OC		η	First order kinetic $\left(-\frac{dC}{dt} = k(Ct)\right)$	
g/L	%	%	kWh/kg COD	kWh/m ³	US \$/m ³	US \$/kg COD	gCOD/Ahm ²	r ²	k (min ⁻¹)
1	85.0	99.9	110.4	136.4	5.0	2.33	7.9	0.97	10.8 ×10 ⁻³
2	91.0	99.9	86.8	106.5	4.1	1.82	11.2	0.99	13.1×10 ⁻³
3	97.2	99.9	73.5	91.2	3.5	1.50	12.0	0.99	18.7 ×10 ⁻³

Table 2. The total operating cost and kinetic analysis for the operation parameters.

4. Conclusion

In this study, the aim was to treat cattle slaughterhouse wastewater (SWW) with sequential coagulation-flocculation (CF) + continuous electrooxidation reactor (CEO) using a BDD anode under different operation conditions.

The prior application of CF eliminated the presence of suspended organic matter and facilitated the achievement of effective electrooxidation. The pre-treatment stage successfully removed 50% of the COD and 68% of the turbidity from the effluent. The COD and turbidity removal efficiencies in the CEO reactor under optimum treatment conditions (j = 30 mA/cm^2 , Q = 0.9 L/h, pH = 8.5, SE = 3.0 gNaCl/L and HRT = 1 h) were calculated to be 97.2% and ~99.9%, respectively. Therefore, using a CF + EO reactor with BDD electrodes was the most suitable method to meet the discharge limit of >125 mg/L COD for SWW. The specific energy consumption, process energy consumption, and total operating cost were 73.5 kWh/kg COD, 91.1 kWh/m³, and 3.5 US \$/m³, respectively. Current density and operating time significantly affected the EO process's removal efficiency. Moreover, both processes showed optimum removal

efficiency at near-neutral pH. Therefore, under optimum process conditions, there is no need for much chemical addition for pH regulation in both systems. However, the addition of electrolytes during EO was necessary as it led to a reduction in electricity consumption. This study demonstrated that the combination of CF and EO processes is a powerful alternative for the treatment of wastewater with high turbidity and organic content, with many advantages such as easy-tooperate equipment, a short operating time, and low chemical requirements. In addition, future studies can be carried out using the initial pH of the wastewater, type of supporting electrolytes, and different anode materials, where some of the parameters investigated in this study (current density, wastewater flow rate, concentration of supported electrolyte) are kept at optimum values. The authors are planning such a study.

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