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# **Research article**

# Studies on electrode combination for COD removal from domestic wastewater using electrocoagulation



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ARTICLE INFO	ABSTRACT
Keywords: COD Domestic wastewater Electrode and factors affecting COD removal	Electrocoagulation is an electrochemical method that uses sacrificial electrodes to remediate wastewater. The combination of electrodes for the treatment of domestic wastewater is the factor that influences the removal efficiency of COD (Chemical Oxygen Demand) by using the electrocoagulation process. Aluminum and Iron electrodes are combined as anode-cathode and cathode-anode in Al–Al, Fe–Fe, Al–Fe, and Fe–Al. Different factors are considered to evaluate the removal efficiency of COD like; pH (3–9), reaction time (15–60 min), and current density (9.23–45 A/m <sup>2</sup> ). Based on this influencing factor Al–Al and Fe–Fe can remove COD up to 87.5 % and 90 % respectively. Similarly, 87.5 % and 88.89 % of COD were removed, when aluminum and iron were combined as Al–Fe and Fe–Al respectively. In addition, the effects of different operating parameters were discussed on the removal percentage of COD. This indicated that the combination of electrode influence the removal efficiency of

COD using the electrocoagulation process under different operating parameters.

# 1. Introduction

Nowadays, wastewater is the most source of environmental pollution that alters the physical and biological environmental condition when directly discharged into an environment without any treatment (Koyuncu and Ariman, 2020). This is a severe problem, especially in developing countries that is a massive problem for a million people due to the lack of, inadequate water supply, improper sanitation, and hygiene that related to water contamination with wastewater (Khan et al., 2019). Domestic wastewater discharges a high volume of wastewater and that contains different pollutants which affect the quality of water (Okadera et al., 2020). Domestic wastewater consists of large volumes of wastewater generated by human activities, especially household wastewater like; baths, showers, hand basins, washing machines, dishwashers, and kitchens, and that harm the natural environment and the health of human beings (Gorgich et al., 2019). Electrocoagulation (EC) is a water and wastewater treatment process that uses electrochemical, chemical, and physical mechanisms to electrochemically dissolve cationic metallic ions in situ by oxidizing a sacrificial anode using just electric current and the dissolved electrodes produce a coagulant species that destabilizes and traps pollutants in the form of suspended particles in water and wastewater, forcing them to form flocs and precipitate, resulting in the removal of pollutants (Ebba et al., 2021). The electrocoagulation process was a system when only electric current was applied to the required wastewater under treatment with other appurtenances, and to increase the motion of suspended and colloidal pollutants that resulted in producing sensitive coagulants from the reaction of electrode reactions (Esfandyari et al., 2019; Garcia-Segura et al., 2017; Qi et al., 2020). Recent researchers suggested that electrocoagulation is the most important technology implemented for water and wastewater treatment due to its environmental friendly (Koyuncu and Arıman, 2020; Nugroho et al., 2019), the high removal efficiency of pollutants (Koyuncu and Arıman, 2020; Nugroho et al., 2019), high energy efficiency (Nagarajappa and Impa, 2015), cost-effectiveness (Nagarajappa and Impa, 2015), avoiding the usage of chemicals (Koyuncu and Arıman, 2020) and a small amount of sludge produced compared to chemical treatment methods (Papadopoulos et al., 2019). In addition to this, the study indicated electrocoagulation is an effective treatment method for removal of pollutants from wastewater; medical (Dehghani et al., 2014; Gökkuş and Yildiz, 2015), domestic (Nagarajappa and Impa, 2015), printing wastewater (Safwat, 2020), textile (Vidal et al., 2017), petroleum industry (Dermentzis, 2016), electroplating wastewater (Melnik, 2016; Moersidik et al., 2020; Tezcan Un et al., 2017), industrial wastewater (chromium removal) (Thirugnanasambandham and Shine, 2018), dairy wastewater

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(Bazrafshan et al., 2013; Chakchouk et al., 2017; Chezeau et al., 2019; Smoczynski et al., 2013), etc.

According to (Dermentzis, 2016), the anodic and Cathodic reactions is formed for Aluminum and Iron electrodes respectively look like as follows;

Anode reaction for Aluminum:

$$Al \to Al^{3+} + 3e^{-} \tag{1}$$

Cathode reaction for Aluminum:

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

Overall reaction:

 $2Al + 6H_2O \rightarrow 2Al(OH)_3 + 3H_2 \tag{3}$ 

Anode reaction for Iron:

 $Fe \rightarrow Fe^{2+} + 2e^-$  (4)

Cathode reaction for Iron:

$$2H_2O + 2e^- \rightarrow 2OH^- + H_2$$

Overall reaction:



The generated Aluminum hydroxide from  $Al^{3+}$  and  $OH^-$  and Iron Hydroxide Fe<sup>2+</sup> and OH<sup>-</sup> ions were used as a coagulant for destabilizing the pollutants from wastewater. Amorphous metal hydroxide precipitates are formed by these coagulants. They have a great affinity for dispersed particles and dissolved contaminants due to their excellent adsorption characteristics. Coagulation can then separate the contaminants from the aqueous phase. The bubbles generated from hydrogen at the cathode increase turbulence in the system and bond with contaminants, lowering their relative specific weight. As a result, they improve the flotation separation process (An et al., 2016). Electrocoagulation was chosen for the treatment of domestic wastewater because it is a simple procedure to perform, has a higher removal efficiency with less energy consumption, does not require the use of chemicals in the treatment process, and produces less sludge after treatment. In this study, the removal percentage of COD was determined under the consideration of pH, current density  $(A/m^2)$ , and electrolysis time (minutes) utilizing the electrocoagulation process.

This was investigated by arranging electrodes as Al–Al, Fe–Fe, Al–Fe, and Fe–Al.



(5)

Figure 1. Setup of electrocoagulation.

## 2. Materials and methods

### 2.1. Materials

Domestic wastewater was collected from the Jimma university teachers' apartment, Jimma zone, Oromiya regional state, Ethiopia. Wastewater is characterized by a pH of 6.3–9.5, temperature 22–28.5 °C, and electrical conductivity of 950–1800  $\mu$ S/cm. COD reactor, pH meter, copper wires, electrical clips, magnetic stirrer, magnetic bar stirrer, power source (DC power), electrocoagulation cell, and Aluminum and Iron electrodes that bought from the local market with 90 Ethiopian birr that means around 2\$ USD were materials used up during the study.

# 2.2. Methods

In the electrocoagulation process, proper setup is important to make a reasonable investigation that indicated in Figure 1 with all parts and materials. Iron and Aluminum electrodes were used in the study since they are easily available and cost-effective and are prepared with 32g and 47g as well as with dimensions of 6cm, 12cm, and 0.9cm length, width, and thickness respectively for both electrodes. A sample of wastewater prepared was added to the electrocoagulation cell and placed on the magnetic stirrer. The gap between anode and cathode was adjusted to 1cm for all numbers of experimental runs and all parameters were adjusted accordingly and an electrode connected to DC power was dipped into the electrocoagulation cell. Then the power was supplied to the cell and based on the fixed electrolysis time and other parameters the study was conducted for the determination of COD for electrode combinations of Al–Al, Fe–Fe, Al–Fe, and Fe–Al.

#### 2.3. Analysis

Based on the electrode combinations of Al–Al, Fe–Fe, Al–Fe, and Fe–Al the removal efficiency of COD was determined by considering pH, current density, and electrolysis time.

The percentage removal of COD (Hamada et al., 2018; Rahman and Borhan, 2014) was determined according to the following formula shown in Eq. (7).

$$COD (\%) = \frac{COD_o - COD_t}{COD_o} *100$$
(7)

where,  $COD_0$  and  $COD_t$  are the chemical oxygen demand at time = 0 (initial) and at t (reaction time, t) respectively.



Figure 2. Removal efficiency of COD at pH (3), and current density (9.23  $\mbox{\rm A/m}^2\mbox{\rm )}.$ 

# 3. Results and discussion

### 3.1. Removal efficiency of COD

The percentage removal efficiency of Chemical Oxygen Demand (COD) was determined for the electrode combinations of Al-A, Fe–Fe, Al–Fe, and Fe–Al. This was done by fixing the parameters such as pH, current density, and electrolysis time such that they have their impacts on the removal efficiency of COD.

# 3.1.1. Al-Al (anode-cathode/cathode-anode)

The elimination effectiveness of COD was measured during the Al–Al combination. When the pH was 3 and the current density was  $9.23 \text{ A/m}^2$ , chemical oxygen demand (COD) was removed to the maximum of 61.54% at the reaction time of 60 min Figure 2 also indicates the removal percentages of COD in 15, 30, and 45 min were 15.38%, 23.08%, and 38.46% respectively. In Figure 3, the removal percentage of COD was 25%, 37.5%, 50% and 62.5% in 15, 30, 45 and 60 min respectively, when pH was 6, and current density was  $20 \text{ A/m}^2$ . Similarly, when the pH was 9 and the current density was  $45 \text{ A/m}^2$ , the maximum removal efficiency of COD was 87.5% when the reaction time was 60 min which is shown in Figure 4. This indicates that the removal efficiency of COD was increased by increasing pH, reaction time, and current density using the electrocoagulation process for domestic wastewater.



Figure 3. Removal efficiency of COD at pH (6), and current density  $(20 \text{ A/m}^2)$ .



Figure 4. Removal efficiency of COD at pH (9), and current density (45 A/m<sup>2</sup>).

# 3.1.2. Fe-Fe (anode-cathode/cathode-anode)

This was also another electrode combination method used to test COD elimination effectiveness. In Figure 2, keeping the pH was 3, and current density was 9.23 A/m<sup>2</sup> good removal efficiency of COD was evaluated such that it can be removed up to 63.33 % when the reaction time was 60 min. Similarly, around 18.18, 27.27, and 45.45 % of COD removal were achieved in 15, 30, and 45 min respectively. Increasing the electrolysis time resulted in a high removal percentage of COD by keeping the pH was 6, and current density  $(20A/m^2)$  which was indicated in Figure 3. This indicates good progress of COD removal percentages such that 33.33, 50, 66.67, and 83.33 % removal was achieved in 15, 30, 45, and 60 min respectively which are shown in Figure 3. On the other hand, when pH was 9, and current density was (45 A/m<sup>2</sup>), the removal degree of COD was 40, 60, 80, and 90 % in 15, 30, 45, and 60 min respectively which has shown in Figure 4. In Fe-Fe, good removal percentage of COD was achieved by increasing the reaction time by keeping other factors constant at the different number of experimental investigations.

# 3.1.3. Al-Fe (anode-cathode)

Aluminum and iron were combined as Al-Fe, and the elimination efficiency of chemical oxygen requirement was calculated, as shown in Figures 2, 3, and 4. When the pH was 3, and the current density was 9.23  $A/m^2$ , 20, 40, 60, and 80 % of COD was removed at an electrolysis time in 15, 30, 45, and 60 min. Similarly, when the increasing the pH to 6, current density to 20  $A/m^2$  the percentage removal of COD was 16.67, 40, 66.67, and 83.33 % at the electrolysis time of 15, 30, 45, and 60 min respectively that shown in Figure 3. In the first 15 min when pH was 3, COD removal was 20 %, which was higher than that of a pH 6 which was 16.67 %, but when the reaction time was 30 min the removal percentage was equal. However, in all cases when pH was 6 higher removal than at pH was 3. This was also true when the pH was 9, and current density 45  $A/m^2$ , COD removal were 25, 50, 62.5 and 87.5 % at the reaction time of 15, 30, 45, and 60 min respectively as indicated in Figure 4. This was indicated that Al-Fe was effective in the removal of chemical oxygen demand in domestic wastewater.

# 3.1.4. Fe-Al (anode-cathode)

The combination of Iron and Aluminum, Fe–Al the removal efficiency was effective, especially when the pH was 9, and the current density was 45 A/m<sup>2</sup>, 33.33, 55.56, 77.78, and 88.89 % of COD was removed at the reaction time of 15, 30, 45 and 60 min respectively that shown in Figure 4. In addition to this, the percentage removal of COD from domestic wastewater for Fe–Al (Anode-Cathode) was shown in detail in Figures 2 and 3 clearly when pH was 3, 6, and 9 for both figures separately at different reaction times. As shown in Figures 2, 3, and 4 the removal percentage of COD was increased gradually from 28.57 to 88.89 % as a pH increase (3–9), reaction time from (15–60 min), and current density from (9.23–45 A/m<sup>2</sup>). Moreover, keeping constant the value of current density and reaction time, when the pH was 3 and 6, Fe–Al was more effective than other combinations and also when pH was 9, Fe–Fe was the best effective compared to others.

Generally, by what form Aluminum and Iron were combined the removal efficiency of COD was increased when the pH, current density, and reaction time increased.

# 3.2. Effects of pH on COD removal

One aspect that affects COD removal effectiveness from wastewater is pH. Since there was a variation of wastewater pH, it was adjusted to acidic, basic, or neutral condition based on the desired level using sulfuric acid or sodium hydroxide. In this study pH of wastewater was adjusted to 3, 6, and 9 that were shown in Figures 2, 4, and 4 respectively. As the value of pH value of the wastewater sample increased from 3 to 6 and 6 to 9, the removal degree of COD was enhanced while wastewater was undergone an electrocoagulation process for different electrode combinations under the same operating parameters. This is because the

increase in pH during the electrocoagulation process seems to be mostly due to the rapid evolution of hydrogen gas at the cathode, and the concentration of hydroxyl ions in the solution increases as a result of electrochemical processes that lead to high percentage removal (Ebba et al., 2021). A variety of metal hydroxide species are formed in electrocoagulation treatments depending on the sample pH, and the stability of insoluble hydroxides are similarly regulated by sample acidity and basicity (Bener et al., 2019; Niazmand et al., 2019).

# 3.3. Effects of current density on COD removal

Current density is an electric current applied per an effective area of electrode for electrocoagulation. It is a critical parameter in electrocoagulation because it regulates the size and growth of the flocs, which affects removal rates and quantity of metal ion dissolved from sacrificial electrodes, resulting in a large amount of precipitate for the removal of pollutants from wastewater (Mohamud et al., 2018). As shown in Figures 2, 3, and 4 current density was 9.23 A/m<sup>2</sup>, 20 A/m<sup>2</sup>, and 45 A/m<sup>2</sup> respectively. This indicates increasing the current density as an operating parameter, increases the removal percentage of electrocoagulation. The current density flow decides the coagulant created from the anode, which is identified with the complete electric charge going through the terminal, as characterized by Faraday's standards of electrolysis (Mohamud et al., 2018). Thus, the current density increased, and more coagulant was made to weaken colloidal particles, bringing about more accelerates that settled effectively in the electrocoagulation cell (Mohamud et al., 2018).

# 3.4. Effects of reaction time on COD removal

The electrolysis time is a crucial metric in Electrocoagulation since it dictates how long water needs to be treated to meet the required criteria. As shown in Figures 2, 3, and 4, the reaction time of wastewater in electrocoagulation was 15, 30, 45, and 60 min. At 15 min, certain COD removal was achieved and as the time progress from 15 to 30 better COD was achieved in all electrode combinations even if there is a certain degree of variation. Similarly, when the reaction time was enhanced to 45 and 60 min, a high degree of COD removal was achieved which indicates as pH increases the COD removal was increased which was shown in Figures 2, 3, and 4. Anodic electro-dissolution causes the release of coagulant species during electrolysis.

Furthermore, the effectiveness with which pollutants are removed is directly proportional to the concentration of metal ions formed on the electrodes. The concentration of metal ions and associated hydroxide flocs increases as the electrolysis time increases (Niazmand et al., 2019).

The formation of adequate quantities of various ions from electrodes, which are required for the generation of adsorbents, such as  $Al(OH)_3$  or  $Fe(OH)_3$  in the case of aluminum or Iron electrodes, as well as the discharging of gases bubbles from both electrodes, which are essentially provided with more assistance to carry the destabilized pollutants toward the surface of the solution, is dependent on electrolysis time (Yasir et al., 2020).

## 3.5. Effects of electrode type and combination on COD removal

The performance of the electrocoagulation system is influenced by the electrode material, particularly the anode, which determines the type of cations released into the solution. Because there was some disagreement over the anode's dissolving mechanism, it was necessary to investigate the effect of the anode material on the extent of electrocoagulation (El-ashtoukhy et al., 2020). For different electrode materials, different reactions occur, and the properties of the metal hydroxides generated during the reactions are critical for the electrocoagulation process' efficiency (Bener et al., 2019). In electrocoagulation, different materials are used as electrodes. In electrocoagulation, several metallic materials have been utilized as the sacrificial electrode or anode. Al, Ag, As, Ba, Ca, Cd, Cr, Cs, Fe, Mg, Na, Si, Sr, and Zn are some of these materials.

Iron in the form of mild steel (MS) or stainless steel (SS), aluminum (Al), zinc (Zn), copper (Cu), and magnesium are the most commonly utilized metallic electrode materials (Mg) (Al-Qodah and Al-Shannag, 2017). Many chemical and physical properties of these elements differ, including oxidation potential, ion size and charge, migration speed in solution, the polarity of the ion-OH connection, and hydroxide compound structure and size (Al-Qodah and Al-Shannag, 2017). Al and Fe were used as an electrode due to low cost and locally available and using Fe as electrodes is better than Al as the electrode for the removal of COD.

Electrodes made of aluminum and iron plates in various combinations Al–Al, Fe–Fe, Al–Fe, and Fe–Al were used to investigate the electrocoagulation process's proficiency as shown in Figures 2, 3, and 4. While Al–Al, Fe–Fe, Al–Fe, and Fe–Al combined COD removal efficiency achieved was 87.5, 90, 87.5, and 88.89 % respectively.

# 4. Conclusion

The electrocoagulation process is an effective system for the removal of COD from domestic wastewater by using aluminum and iron electrodes by different combinations either at anode or cathode. This was done by combining aluminum and iron electrodes in the form of Al–Al, Fe–Fe, Al–Fe, and Fe–Al by considering different operating parameters such as pH, current density, and electrolysis time. Increasing the pH (3–9), current density (9.23–45  $A/m^2$ ), and electrolysis time (15–60 min) resulted in the increase of COD removal percentages from domestic wastewater in all electrode combinations even if there were variation in values. Fe–Fe combination was the best electrode combination compared to others under the same fixed operating parameters for the removal of COD (90 %) from domestic wastewater. Generally, different ways of aluminum and iron combination can affect the removal efficiency of COD from domestic wastewater by considering different operating parameters using the electrocoagulation process.

#### Declarations

#### Author contribution statement

Million Ebba Bote: Conceived and designed the experiments; Performed the experiments; Analyzed and interpreted the data; Contributed reagents, materials, analysis tools or data; Wrote the paper.

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# Data availability statement

Data included in article/supp. material/referenced in article.

## Declaration of interests statement

The authors declare no conflict of interest.

# Additional information

No additional information is available for this paper.

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