JURNAL SAINS MATERI INDONESIA (JUSAMI)
ISSN: 1411-1098 (print), 2614-087X (online)
VOL. 25, ISSUE 2, 2024
DOI:10.55981/jsmi.2024.3120

ORIGINAL ARTICLE

Aluminum Waste as Electrode for Home Textile Industry Wastewater Treatment Using Batch Electrocoagulation Process: Studies on Operating Parameters

C. Irawan[1,2*], R. Mu'minah[1], A. Purnawilda[1] and I. F. Nata[1]

[1]Department of Chemical Engineering, Faculty of Engineering, Universitas Lambung Mangkurat, Indonesia
[2]Professional Engineering Education Study Program, Faculty of Engineering, Universitas Lambung Mangkurat, Indonesia

ABSTRACT – The manufacture of the Sasirangan home textile industry involves coloring and dyeing processes using synthetic dyes in large quantities. These contaminants of dyes and organic materials would cause high color and chemical oxygen demand (COD) contaminants values. This study aims to characterize the wastewater of batik-modified Sasirangan and determine the effect of current density and length of operating time on color removal and reduction of COD in Sasirangan home textile industry wastewater through the batch electrocoagulation process. The method used in this research is an electric current flowing in the same direction to the Sasirangan home textile industry wastewater in a reactor with dimensions of 310 × 180 × 240 mm^3. The electrode used is aluminum alloy type 1100. The aluminum/aluminum (Al/Al) electrode is used in this electrocoagulation (EC) process, then connected to a direct current (DC) power supply. The experiment was carried out at room temperature using an electrode distance of 2 cm with variations in the time of the electrocoagulation process for 15–120 minutes. The experiment was repeated for variations in pH (4–9) with a current density of 3.5–5.5 mA/cm^2. Furthermore, an analysis of the color removal and the decrease in the concentration of COD was carried out. The results showed that the contaminant content in Sasirangan home textile industry wastewater decreased significantly, whereas the optimal conditions for the EC reaction were determined using color and COD removal efficiency parameters. The decrease in color and COD concentrations occurred at a current density of 5.5 mA/cm^2 with a pH of 4 for 120 minutes, around 1110 PtCo and 90.4 mg/L of COD, respectively.

ARTICLE HISTORY
Received: 12 Jan 2024
Revised: 29 Feb 2024
Accepted: 05 Mar 2024

KEYWORDS
Electrocoagulation
Sasirangan textile wastewater
Color
COD
Aluminum electrode

INTRODUCTION

The growth of the Sasirangan home textile industry in South Kalimantan, particularly Tanah Laut Regency, continues to increase until it reaches around 78 small and medium industries (*industri kecil menengah*, IKM entrepreneurs) with the largest distribution at the Sasirangan IKM Center in Tambang Ulang District. During their process activity, the Sasirangan home textile industry involves a coloring and dyeing process using synthetic dyes, such as naphthol, indigosol, and indanthrene dyes, which produce large amounts of wastewater. This dye wastewater contains high color, total suspended solids, and organic materials, which cause high chemical oxygen demand (COD) concentration [1]–[4]. Therefore, if it is not handled properly, it poses a serious risk to both human health and the sustainability of the environment.

Several methods have been carried out to treat textile wastewater that contains high color, total suspended solid (TSS), and COD concentration, such as filtration, chemical coagulation, ozonation, oxidation-reduction, ion exchange, neutralization, and adsorption methods [1], [4]–[8]. Even though these methods are effective, they require quite a long time of treatment, using chemical substances that create secondary pollution in their wastewater treatment process. Therefore, the electrocoagulation method can be an effective and environmentally friendly alternative with advantages including being economical because it does not use additional chemicals, but the efficiency is relatively high, efficient in terms of time, the flocs are formed electrically so that the sedimentation process is faster, its use can be done on a large scale, and easy to implement [9].

Electrocoagulation is the process of destabilizing suspensions, emulsions, and solutions containing contaminants by passing a direct electric current through water, causing the formation of clots that are easily separated [2], [4], [10], [11]. Many operating factors, such as current or voltage, operation time, pH, electrode material, conductivity, and electrode spacing, influence the potential for an efficient treatment. The electrocoagulation (EC) equipment usually uses aluminum, iron, or combined aluminum and iron as electrodes in their EC process [11]–[13], and even a little bit uses other metal materials such as Mn, Ti, stainless steel, and Cu [12]. Iron, since it is less expensive, but in certain applications, such as color and COD removal, it has been observed that aluminum electrodes are more efficient than iron electrodes, possibly due to the higher adsorption capacity and electrostatic interaction of aluminum oxides. [12]–[15].

Recently, the electrocoagulation process has been increasingly interested in treating textile wastewater [2], [4], [6], [14], [16], [17]. One effort to minimize the negative impact of contaminants in this research is to treat the Sasirangan textile wastewater before disposing it to the environment using the electrocoagulation method. Although some of the electrocoagulation process for textile wastewater is presented in previous research, other data such as the type of real textile of wastewater, modified electrocoagulation reactor with hot plate magnetic stirrer, and use the aluminum electrodes (Al 1100) from the waste scraps of the aluminum furniture industry, are observed to this research. The type of real textile wastewater used in this research is Sasirangan textile wastewater, which contains such as naphthol, indigo sol, and indanthrene dyes. Aluminum alloy 1100 is almost entirely made of pure aluminum, with tiny amounts of other metals added to improve corrosion resistance and machinability, typically copper or iron. Another important thing in Sasirangan textile wastewater contaminants is color and COD, which causes high turbidity in water due to undissolved solids and compounds in water not chemically breaking down [15]. In addition, the effect of operation time, initial pH, and current density of the electrode were also investigated in reducing color and COD concentration of the Sasirangan textile wastewater.

EXPERIMENTAL METHOD

Materials and Instruments

The electrocoagulation reactor

The electrocoagulation reactor was made by assembling an acrylic vessel with dimensions of 310 × 180 × 240 mm. The electrodes (anode and cathode) used are Aluminum (Al)/Aluminum (Al) (type Al 1100) from the waste scraps of the aluminum furniture industry with dimensions of 8 × 8 cm and a thickness of 2 mm. The electrodes are connected to a direct current (DC) power supply with a controllable current density regulator, as shown in Figure 1.

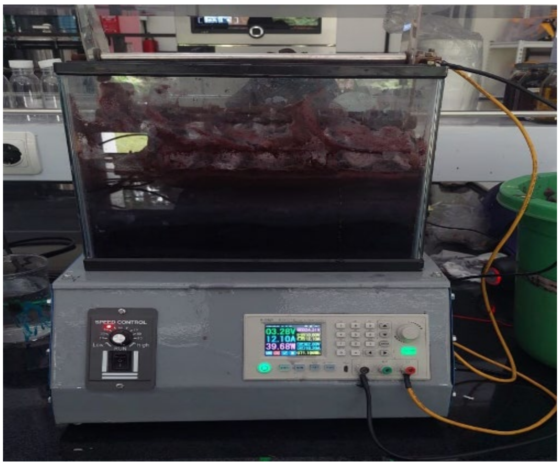

Figure 1. Electrocoagulation experimental reactor with Al/Al electrodes

Characterization of Sasirangan textile wastewater

The sasirangan textile wastewater-modified batik pattern used in this research was taken at the Sasirangan IKM (small and medium industries) center in Tambang Ulang District, Tanah Laut Regency, South Kalimantan. Sasirangan textile production involves a coloring and dyeing process using synthetic dyes such as naphthol, indigosol, reactive, and indanthrene, which will generate abundant amounts of wastewater that is dark in color and contains contaminants that usually exceed the effluent quality standards for wastewater for the textile industry.

Table 1. The quality results of Sasirangan textile wastewater based on the effluent liquid before the electrocoagulation process

Parameter	Unit	Quantity	Standard
pH	-	11.0	6.0–9.0
Color	PtCo	23950.0	200.0
Chemical oxygen demand (COD)	mg/L	1355.9	150.0

In Table 1, it is shown that the results of Sasirangan textile wastewater before the electrocoagulation process was carried out, all parameters did not meet the wastewater quality standards for the textile industry [18]. These results are considered dangerous for the environment. Thus, it must be processed before the Sasirangan textile wastewater is discharged into water bodies or reused for the Sasirangan textile production process.

Method and Procedure

A total of 8 L of Sasirangan textile wastewater samples were placed in an electrocoagulation batch reactor. The pH of the experiment was observed with a pH probe and set at the initial pH using HCl 5 N or NaOH 5 N. The Al/Al electrode was connected to a DC power supply with a varying measured current density. During electrocoagulation, the sample was stirred with a magnetic bar measuring 80 mm. The experiment was carried out at room temperature using an electrode distance of 2 cm and varying times for the electrocoagulation process of 15, 30, 60, 90, and 120 minutes. The experiment was repeated for initial pH variations (4, 7, 9, and 11) using 5 N NaOH with current densities of 3.5, 4.5, and 5.5 mA/cm3. Furthermore, the samples resulting from the process are periodically analyzed for color and COD concentration using a Spectrophotometer analyzer. The color of wastewater was analyzed by the Platinum-Cobalt (PtCo) method at 455 nm using a Thermo Scientific Orion AQ8000 spectrophotometer and the COD analysis method based on the Indonesian National Standard (SNI) of SNI 06-6989.2-2004 with closed reflux spectrophotometrically.

RESULT AND DISCUSSION

Process parameters, including initial textile wastewater pH, current density, and treatment time, were examined to determine the optimal parameter conditions for the electrocoagulation process to decrease the concentration of color and chemical oxygen demand (COD).

Effect of initial pH during Electrocoagulation (EC) time on Color and COD treatment

Theoretically, aluminum (Al) ions are released during the electrocoagulation process. Dissolution of aluminum occurs at the cathode and anode. At the cathode, there is absorption of the electrode surface, while at the anode, there is a decrease in positive ions [2], [11], [19], [20]. EC time, initial pH of wastewater sample, and current density are important variables that influence the electrocoagulation process [2], [6], [21]. In general, the pH of the solution can change during the electrocoagulation process. The pH change depends on the electrode material used and the initial pH of the sample. Changes in the pH of the solution during the electrocoagulation process indicate a transfer of Al ions from the electrode to the solution, resulting in a flocculation formation process [2], [4], [16]. In the electrocoagulation process, aluminum hydroxide [Al(OH)3] is formed, which is alkaline in the overall oxidation and reduction reaction, and the pH of the wastewater will be increased. The decrease in color and COD has the same pattern at a current density of 3.5 mA/cm2, as seen in Figure 2 and Figure 3.

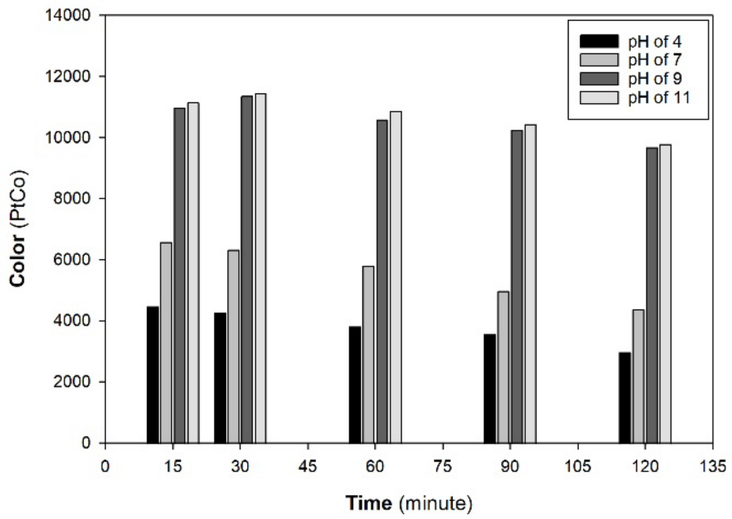

Figure 2. Profile of decreasing color concentration with various initial pH during the EC process at ambient temperature and current density of 3.5 mA/cm^2

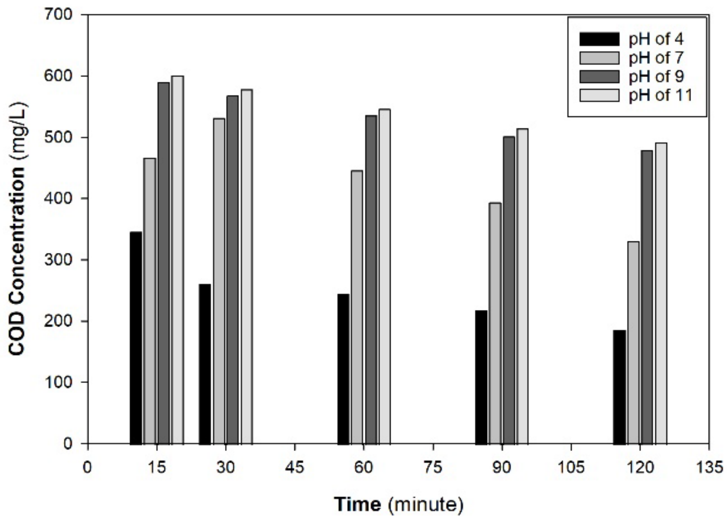

Figure 3. Profile of decreasing COD concentration with various initial pH during the EC process at ambient temperature and current density of 3.5 mA/cm^2

During the EC process, aluminum hydroxide formed from Al^{3+} and OH^- was used as a coagulant to destabilize pollutants in wastewater. The coagulants produce amorphous metal hydroxide precipitates. They have a strong affinity for dispersed particles and dissolved contaminants due to their excellent adsorption properties. Coagulation then separates the contaminants from the aqueous phase. Hydrogen bubbles at the cathode increase turbulence and bond with contaminants, reducing their specific weight [4], [6], [13]–[16]. The optimum color concentration was obtained around 3000 PtCo based on the profile graph shown in Figure 2 after 120 minutes of EC time at an initial pH of 4. In contrast, in Figure 3, the optimum COD concentration was 187.6 mg/L. The initial Sasirangan textile wastewater pH was 4 during 120 minutes of EC time, given the lower concentration of color and COD. However, the ionization reaction between the electrodes will be lowered during the 120 minutes of EC time due to the alkaline pH of the wastewater, making the reduction in color and COD concentrations ineffective [22], [23]. The optimum color and COD reduction efficiency occurred at pH 4 with an operating EC time of 120 minutes, while efficiency slightly increased at times higher than 120 minutes, though this was not statistically meaningful. This is due to the pH of the solution changing and tending to increase because, in acidic conditions, the Al cathode will experience an acid reduction, producing H_2. In contrast, the aluminum anode will experience oxidation, producing Al^{3+} ions according to equation (1) [11], [19].

$$\begin{array}{rl} 6H^+ + 6e^- & \rightarrow 3H_2 \\ 2Al & \rightarrow 2Al^{3+} + 6e^- \\ \hline 6H^+ + 2Al & \rightarrow 3H_2 + 6e^- \end{array} \quad (1)$$

While electrolytic flotation occurs, the presence of H2 generated produces hydroxide ions that bind pollutants in the wastewater into insoluble compounds that float to the surface of the EC reactor. When the pH is initially set to an alkaline state during the electrocoagulation process, the efficacy of the pollutants in wastewater binding decreases [4], [20], [21]. In all cases in this research, the initial sample pH of 4 demonstrated the highest removal efficiency (Figure 2 and Figure 3). Al electrodes have previously been shown to have an optimum working pH of 4-6 using the EC process to remove organic compounds in wastewater [24], [25].

Effect of current density during EC time on Color and COD treatment

The current applied to the electrode (anode) divided by the electrode's active area is the definition of current density. The measurement unit is mA/cm3 [14]. The current density determines the release rate of metal cations from the anode and the productivity of air bubbles (hydrogen/oxygen). The contribution of current density to the response variable occurs because the current given to the anode will dissolve the anode metal into solution to become metal cations (M+). The metal cations formed then interact with hydroxy ions (OH-) to form the coagulant Al(OH)x¬. The decrease in color and COD have the same pattern at pH 4, as seen in Figure 4 and Figure 5.

Figure 4 shows the decreasing color, and Figure 5 shows the decreasing COD concentration with increasing EC time and current density to become 1110 PtCo and 90.4 mg/L, respectively. This value is found at a current density of 5.5 mA/cm2 and an EC time of 120 minutes during the EC process at ambient temperature and initial pH of 4. The color and concentration of contaminants decrease during the electrocoagulation process as more and more large Al(OH)3 flocs form as a result of colloidal particles binding together throughout the electrocoagulation reaction [4], [14], [17].

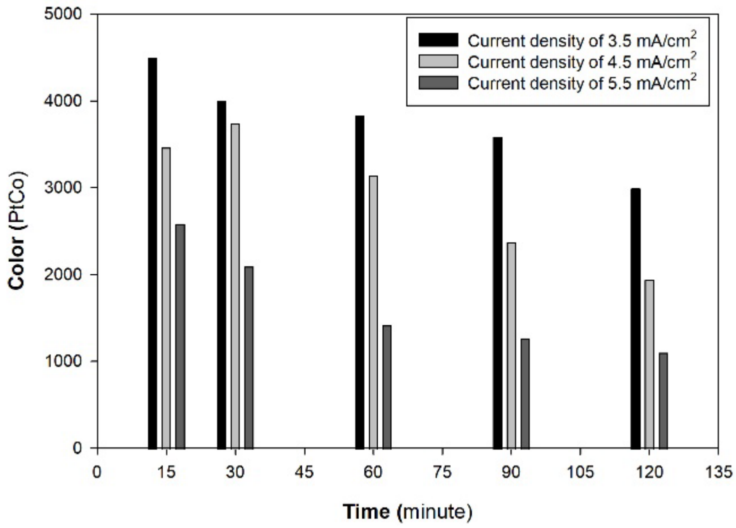

Figure 4. Profile of decreasing color concentration with various current densities during the EC process at ambient temperature and initial pH of 4

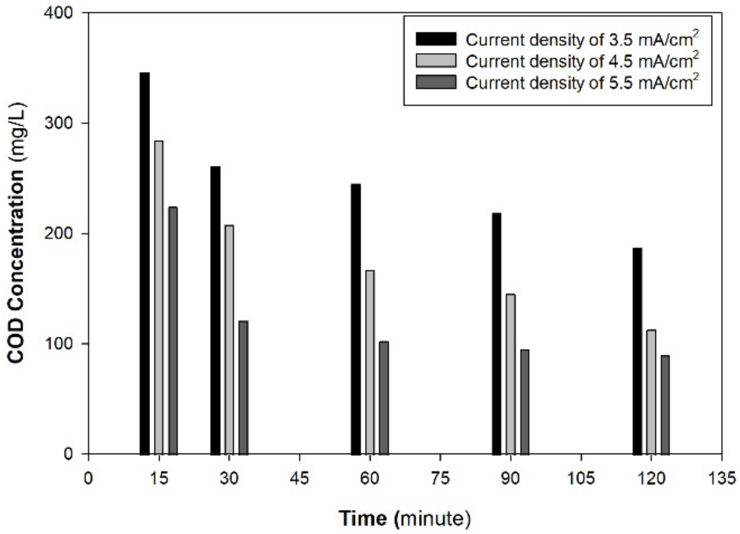

Figure 5. Profile of decreasing COD concentration with various current densities during the EC process at ambient temperature and initial pH of 4

This is consistent with Faraday's law, which states that the amount of anode metal present as a source of dissolved coagulant directly correlates with the increase in current. Al(OH)x coagulant, created by the Al anode and OH-ions that come from H2O molecules, is formed during the contaminant reduction process. The impurities will then be absorbed by Al(OH)x coagulant and enter its molecular cavity [16], [26].

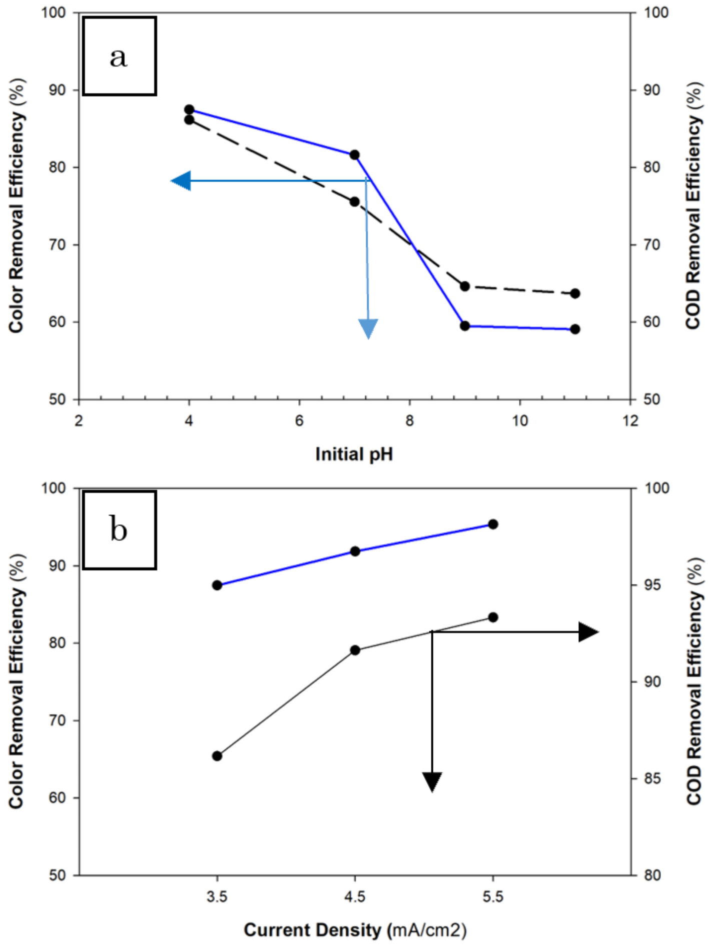

Figure 6. Profile of color and COD removal efficiency concentration with (a) various initial pH and (b) current density during the EC process at ambient temperature

The percentage reduction in contaminants in Sasirangan textile wastewater can be seen in Figure 6. Figure 6 (a) shows that it is well understood that the EC process heavily relies on the wastewater's initial pH. The generation of metallic hydroxides is pH-dependent, and the initial pH of the wastewater influences electrocoagulation performance. During electrocoagulation treatments, various metal hydroxide species are formed based on the sample pH, and the stability of insoluble hydroxides is similarly regulated by sample acidity and basicity [2], [25]. Besides, Figure 6 (b) shows at high current densities, the dissolution of aluminum in the anode increases, producing greater amounts of Al3+ and Al(OH)n(s). Increasing the concentration of Al3+ can increase the neutralization reaction of contaminant loads, which then form flocs. The higher percentage efficiency values for color and COD removal were found at a pH of 4, while in the case of current density, they were found at 5.5 mA/cm2 with a contact time of 120 minutes at ambient temperature. The most effective removal pattern with this electrocoagulation method is a decrease in color, followed by a decrease in COD. Although the initial pH was set to 4, the final pH increased to 7.2 ± 0.3 by the end of the experiment, as reported in the literature. This increase can be attributed to the formation of hydrogen gas and the accumulation of hydroxide following water reduction in the cathode [25], [27]. Figure 6 shows that in this study, it was concluded that increasing the initial pH of Sasirangan textile wastewater reduced color (87.47% to 59.66%) and COD (86.16% to 63,68%) removal efficiency, as seen in Figure 6 (a), whereas increasing current density increased color (87.47% to 95.37%) and COD (86.16% to 93.33%) removal efficiency as seen in Figure 6 (b).

CONCLUSION

The contaminant content in Sasirangan textile wastewater, such as color and chemical oxygen demand (COD), has decreased significantly using the electrocoagulation (EC) process. The decrease in color and COD concentrations occurred at a current density of 5.5 mA/cm2 with a pH of 4 at 120 minutes, becoming 1110 PtCo and 90.4 mg/L, respectively. The longer the EC operation time, the more effective it is in reducing contaminant content in Sasirangan textile wastewater. The optimum EC time for reducing contaminant content in this study occurred at 120 minutes. The degree of acidity also affects the electrocoagulation process. The optimum initial pH of Sasirangan textile wastewater for this research occurs in acidic conditions, at pH 4. The current density used is directly proportional to the electric current flowing. The higher the electric current density used and the longer the contact time, the greater the reduction in contaminants in Sasirangan textile wastewater.

ACKNOWLEDGEMENT

We would like to thank the Directorate of Research, Technology, and Community Services of the Ministry of Education, Culture, Research and Technology (Kemendikbudristek), Republic of Indonesia, for supporting this research through the Graduate Research Project and the Chemical Engineering study program for helping and facilitating the laboratory of Water and Wastewater Treatment and equipment during this research.

REFERENCES

[1] F.J. Ariza-Pineda, I.F. Macías-Quiroga, D.F. Hinojosa-Zambrano, J.D. Rivera-Giraldo, D.M. Ocampo-Serna, and N.R. Sanabria-González. "Treatment of textile wastewater using the Co(II)/$NaHCO_3$/H_2O_2 oxidation system." *Heliyon*, vol. 9, p. e22444, 2023.
[2] S. Bener, Ö. Bulca, B. Palas, G. Tekin, S. Atalay, and G. Ersöz. "Electrocoagulation process for the treatment of real textile wastewater: Effect of operative conditions on the organic carbon removal and kinetic study." *Process Safety and Environmental Protection*, vol. 129, pp. 47–54, 2019.
[3] J.M. Bidu, K.N. Njau, M. Rwiza, and B. Van der Bruggen. "Textile wastewater treatment in anaerobic reactor: Influence of domestic wastewater as co-substrate in color and COD removal." *South African Journal of Chemical Engineering*, vol. 43, no. 1, 112–121, 2023.
[4] A. Negash, D. Tibebe, M. Mulugeta, and Y. Kassa. "A study of basic and reactive dyes removal from synthetic and industrial wastewater by electrocoagulation process." *South African Journal of Chemical Engineering*, vol. 46, no. 1, 122–131, 2023.
[5] S.S.A. Norrahma, N.H.A. Hamid, N.H.H. Hairom, L. Jasmani, and D.A.B. Sidik. "Industrial textile wastewater treatment using Neolamarckia cadamba NFC filter paper via cross-flow filtration system." *Journal of Water Process Engineering*, vol. 55, p. 104188, 2023.
[6] J. Núñez, M. Yeber, N. Cisternas, R. Thibaut, P. Medina, and C. Carrasco. "Application of electrocoagulation for the efficient pollutants removal to reuse the treated wastewater in the dyeing process of the textile industry." *Journal of Hazardous Materials*, vol. 371, pp. 705–711, 2019.
[7] S. Raj, H. Singh, and J. Bhattacharya. "Treatment of textile industry wastewater based on coagulation-flocculation aided sedimentation followed by adsorption: Process studies in an industrial ecology concept." *Science of The Total Environment*, vol. 857, no. 2, p. 159464, 2023.
[8] M. Sanavi Fard, A. Ehsani, and F. Soleimani. "Treatment of synthetic textile wastewater containing Acid Red 182 by electro-Peroxone process using RSM." *Journal of Environmental Management*, vol. 344, p. 118379, 2023.
[9] J.N. Hakizimana, B. Gourich, M. Chafi, Y. Stiriba, C. Vial, P. Drogui, and J. Naja. "Electrocoagulation process in water treatment: A review of electrocoagulation modeling approaches." *Desalination*, vol. 404, pp. 1–21, 2017.
[10] Y.G. Asfaha, F. Zewge, T. Yohannes, and S. Kebede. "Application of hybrid electrocoagulation and electrooxidation process for treatment of wastewater from the cotton textile industry." *Chemosphere*, vol. 302, p. 134706, 2022.
[11] A.S. Naje, S. Chelliapan, Z. Zakaria, M.A. Ajeel, and P.A. Alaba. "A review of electrocoagulation technology for the treatment of textile wastewater." *Reviews in Chemical Engineering*, vol. 33, no. 3, pp. 263–292, 2017.
[12] S. Boinpally, A. Kolla, J. Kainthola, R. Kodali, and J. Vemuri. "A state-of-the-art review of the electrocoagulation technology for wastewater treatment." *Water Cycle*, vol. 4, pp. 26–36, 2023.
[13] T. Rangseesuriyachai, K. Pinpatthanapong, J. Boonnorat, S. Jitpinit, T. Pinpatthanapong, and T. Mueansichai. "Optimization of COD and TDS removal from high-strength hospital wastewater by electrocoagulation using aluminium and iron electrodes: Insights from central composite design." *Journal of Environmental Chemical Engineering*, vol. 12, no. 1, p. 111627, 2024.
[14] M.E. Bote. "Studies on electrode combination for COD removal from domestic wastewater using electrocoagulation." *Heliyon*, vol. 7, no. 12, p. e08614, 2021.
[15] M. Nor El houda, M. Chabani, S. Bouafia-Chergui, and A. Touil. "Removal of chemical oxygen demand from real petroleum refinery wastewater through a hybrid approach: Electrocoagulation and adsorption." *Chemical Engineering and Processing - Process Intensification*, vol. 196, p. 109680, 2024.
[16] S. Najari, M. Delnavaz, and D. Bahrami. "Application of electrocoagulation process for the treatment of reactive blue 19 synthetic wastewater: Evaluation of different operation conditions and financial analysis." *Chemical Physics Letters*, vol. 832, p. 140897, 2023.
[17] A. Yaqub, H. Raza, H. Ajab, S.H. Shah, A. Shad, and Z.A. Bhatti. "Decolorization of reactive blue-2 dye in aqueous solution by electrocoagulation process using aluminum and steel electrodes." *Journal of Hazardous Materials Advances*, vol. 9, p. 100248, 2023.
[18] MOEF, Wastewater effluent standard for industries, Decree No. 19, The Indonesian Ministry of Environment and Forestry, Jakarta, Indonesia, 2019.
[19] C.E. Barrera-Díaz, P. Balderas-Hernández, and B. Bilyeu. "Chapter 3-Electrocoagulation: Fundamentals and Prospectives," in: C.A. Martínez-Huitle, M.A. Rodrigo, O. Scialdone (Eds.), *Electrochemical Water and Wastewater Treatment*, *Butterworth-Heinemann*, 2018, pp. 61–76.

[20] M.Y.A. Mollah, P. Morkovsky, J.A.G. Gomes, M. Kesmez, J. Parga, and D.L. Cocke. "Fundamentals, present and future perspectives of electrocoagulation." *Journal of Hazardous Materials*, vol. 114, no. 1–3, pp. 199–210, 2004.
[21] P. Asaithambi, M.B. Yesuf, R. Govindarajan, P. Selvakumar, S. Niju, T. Pandiyarajan, A. Kadier, D.D. Nguyen, and E. Alemayehu. "Industrial wastewater treatment using batch recirculation electrocoagulation (BRE) process: Studies on operating parameters." *Sustainable Chemistry for the Environment*, vol. 2, p. 100014, 2023.
[22] S. Garcia-Segura, M.M.S.G. Eiband, J.V. de Melo, and C.A. Martínez-Huitle. "Electrocoagulation and advanced electrocoagulation processes: A general review about the fundamentals, emerging applications and its association with other technologies." *Journal of Electroanalytical Chemistry*, vol. 801, pp. 267–299, 2017.
[23] A. Kothai, C. Sathishkumar, R. Muthupriya, K.S. sankar, and R. Dharchana. "Experimental investigation of textile dyeing wastewater treatment using aluminium in electro coagulation process and Fenton's reagent in advanced oxidation process." *Materials Today: Proceedings*, vol. 45, no. 2, pp. 1411–1416, 2021.
[24] N. Adhoum and L. Monser. "Decolourization and removal of phenolic compounds from olive mill wastewater by electrocoagulation." *Chemical Engineering and Processing: Process Intensification*, vol. 43, no. 10, pp. 1281–1287, 2004.
[25] R. Niazmand, M. Jahani, and S. Kalantarian. "Treatment of olive processing wastewater by electrocoagulation: An effectiveness and economic assessment." *Journal of Environmental Management*, vol. 248, p. 109262, 2019.
[26] M. Nasrullah, S. Ansar, S. Krishnan, L. Singh, S.G. Peera, and A.W. Zularisam. "Electrocoagulation treatment of raw palm oil mill effluent: Optimization process using high current application." *Chemosphere*, vol. 299, p. 134387, 2022.
[27] B. Karagozoglu and R. Malkoc. "The investigation of the removal of reactive orange 16 DYE from textile wastewater by using electrocoagulation process. *Cumhuriyet Science Journal*, vol. 38, no. 3, pp. 544–556, 2017.