



Evaluation of Magnetic Stirring and Aeration on Electrocoagulation Performance in Actual Industrial Treatment

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Agitation was a significant factor in achieving the high performance of the electrocoagulation (EC) system. Three EC systems with four parallel monopolar Al electrodes were established to clarify the influence of agitation methods on pollutants removal efficiency; magnetic stirring, continuous aeration, and combination of magnetic stirring and aeration. The aim of this work was to maximize industrial wastewater treatment in a short detention time and to understand the mechanisms that occurred in different EC systems. The coolant wastewater from the aluminum product industry was represented as industrial wastewater. The hybrid stirring-aeration EC system obtained a lower COD removal compared to the stirring EC system and the aeration EC system. Although aeration can cause an increase in COD removal due to complete circulation and effective coagulant formation of Fe (OH)₃, however, the combination of aeration and stirring negatively affected the performance of EC. The possible reason was that the excessive agitation led to a rapid mixing of the solution, and then the coagulants and pollutants obtained insufficient time to form flocs to precipitate. The best EC performance was observed in the aeration EC system, followed by the stirring EC system, control system (without agitations), and the stirring aeration EC system, respectively, in the short detention time of 15 min. Furthermore, all EC systems could achieve an excellent COD removal of 91% when the detention time was sufficient (eg, 45 min for the stirring aeration EC system). Furthermore, the decreasing number of electrodes affected the COD removal efficiency, whereas the NaCl additive was insignificantly affected.

Keywords: electrocoagulation, coolant wastewater, Fe-Al electrodes, stirring, aeration

INTRODUCTION

Coolant is a form of industrial waste and wastewater, and it is widely used in various metal-working industries for cooling systems and lubricating the interfaces of machines and workpieces. Its significant characteristic is oily wastewater containing high nonbiodegradable organic content (as represented in a low biodegradable index), high turbidity, and some levels of metals (Hilal et al., 2004; Yu et al., 2017). Coolant waste and wastewater are considered hazardous and require special treatment and disposal. Due to waste management in Thailand, coolant wastewater is separated from other industrial wastewaters which can be treated by industrial treatment plants. Coolant wastewater is stored and sent to a private waste disposal service for further treatment and disposal. Meanwhile, illegal disposal of coolant waste and wastewater into the environment has been observed (Marfe and Di Stefano, 2016).

The electrocoagulation (EC) process is known as one of the outstanding techniques with many advantages, such as high efficiency in a short treatment time, simplicity of design and operation, mild condition for treatment, and low cost of material and operation (Coimbra et al., 2021; Lucakova et al., 2021). The CE process was widely utilized to treat various types of industrial wastewater, such as tannery wastewater (Azarian et al., 2018a; Eryuruk et al., 2018), poultry slaughterhouse and egg processing wastewater (Godini, 2012; Azarian et al., 2018a; Azarian et al., 2018b; Rahmani et al., 2018; Ehsani et al., 2020), and crude vegetable oil refinery wastewater (Preethi et al., 2020). Due to the conceptual EC process, iron (Fe) or aluminum (Al) was introduced as electrodes to generate iron hydroxide or aluminum hydroxide flocs [that is, Fe (OH)₂, Fe (OH)₃, Al (OH)₃] to destabilize pollutants and allow them to coagulate. However, a better performance of the EC process, including a techno-economic trade-off, was observed in the system combining Fe and Al as electrodes rather than the Fe/Fe and Al/Al systems (Chezeau et al., 2020; Kobya et al., 2014).

In addition, several other factors were mentioned as key factors influencing the EC performance and treatment efficiency, such as electrode number and supporting electrolyte type and concentration. The increase in the number of electrodes from two to six improved the performance of EC, while it also increased the energy consumption during the EC process (Gusa et al., 2020). Among the most favorable supporting electrolyte of NaCl, Na₂SO₄, NaNO₃, and HClO₄ (Yildiz et al., 2008; Rahmani et al., 2018), the NaCl significantly achieved the highest pollutants removal rate. Recently, external air addition has been reported to enhance CE performance for organic content (as presented in COD) and arsenite removals (Kumar et al., 2018; Syam Babu et al., 2021; Akansha et al., 2020), due to the presence of adequate dissolved oxygen necessary for the conversion of pollutants and chemical reactions. However, the concept of the hybrid electrochemical reaction and aeration system to improve wastewater treatment efficiency was unclearly explained.

The objective of this present work was to clarify the pollutants removal that occurred in the hybrid stirring-aeration EC system using Fe as anode and Al as cathode. The performance of the hybrid system to treat the wastewater from the coolant was

compared with that of the traditional EC system using magnetic stirring and the alternative system using aeration. Moreover, the other operating factors, including electrode number and salt additive, were simply discussed.

METHODOLOGY

Wastewater

The actual coolant wastewater was collected from an aluminum product industry, located in Amata City Chonburi (Thailand). The appearance of the wastewater was a light white solution and slightly suspended, and its characteristics were listed as the following; pH of 6, 1,333 mg/L of total suspended solids, 56,400 mg/L of chemical oxygen demand (COD), 16,800 mg/L of biochemical oxygen demand (BOD), and 19,600 mg/L of oil and grease. The coolant wastewater without any pretreatments and pH adjustments was used in this work.

Electrocoagulation System

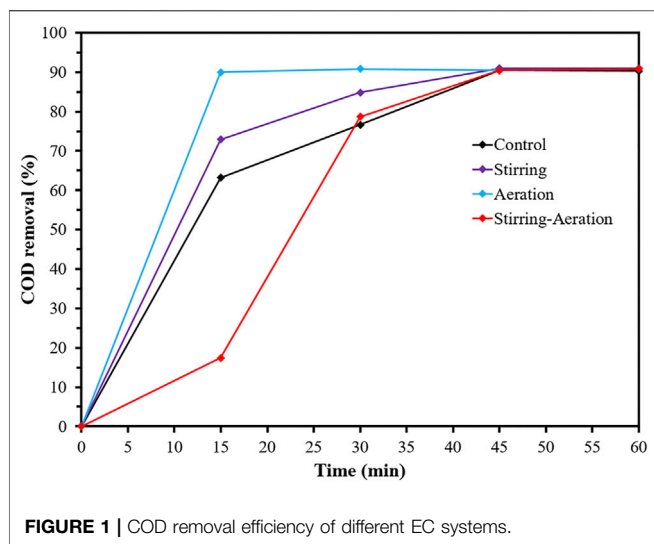
In the present work, the EC reactor was made of acrylic material with a dimension of 8 cm (length) x 7.5 cm (width) x 15 cm (height). The Fe and Al plates were used as anode and cathode, respectively, with a size of 7.5 cm (width) x 10 cm (height) x 0.1 cm (thickness). The electrodes were dipped in the solution to maintain an effective area of 45 cm² per electrode and the interelectrode distance between electrodes was controlled at 1 cm. In the stirring EC system, the EC reactor was placed on a magnetic stirrer and stirred at ~150 rpm which was the optimal stirring rate as suggested by Bayer et al. (2011). In the EC aeration system, two aquatic aerators were placed on the reactor base and aerated at ~0.5 L/min. In the stirring-aeration EC system, both stirring and aeration were supplied to the reactor at ~150 rpm and ~0.5 L/min respectively. Another reactor was set up as a control system with no stirring and no aeration. For all EC systems, the electric current from the DC power supply was supplied to the electrodes to provide the current density of approximately 20 mA/cm² which was suggested by Ehsani et al. (2020) and Pantorlawn et al. (2017).

Experimental Procedure

First, the coolant was mixed with 5 g/L of NaCl to increase the conductivity (Pantorlawn et al., 2017) before treating in different types of EC systems. Subsequently, the number of electrodes was reduced from four (2 Fe and 2 Al) to two (1 Fe and 1 Al) with the same distance between electrodes. Finally, the NaCl additive was varied from 1 to 10 g/L to optimize the conductivity of the wastewater and the efficiency of coolant treatment.

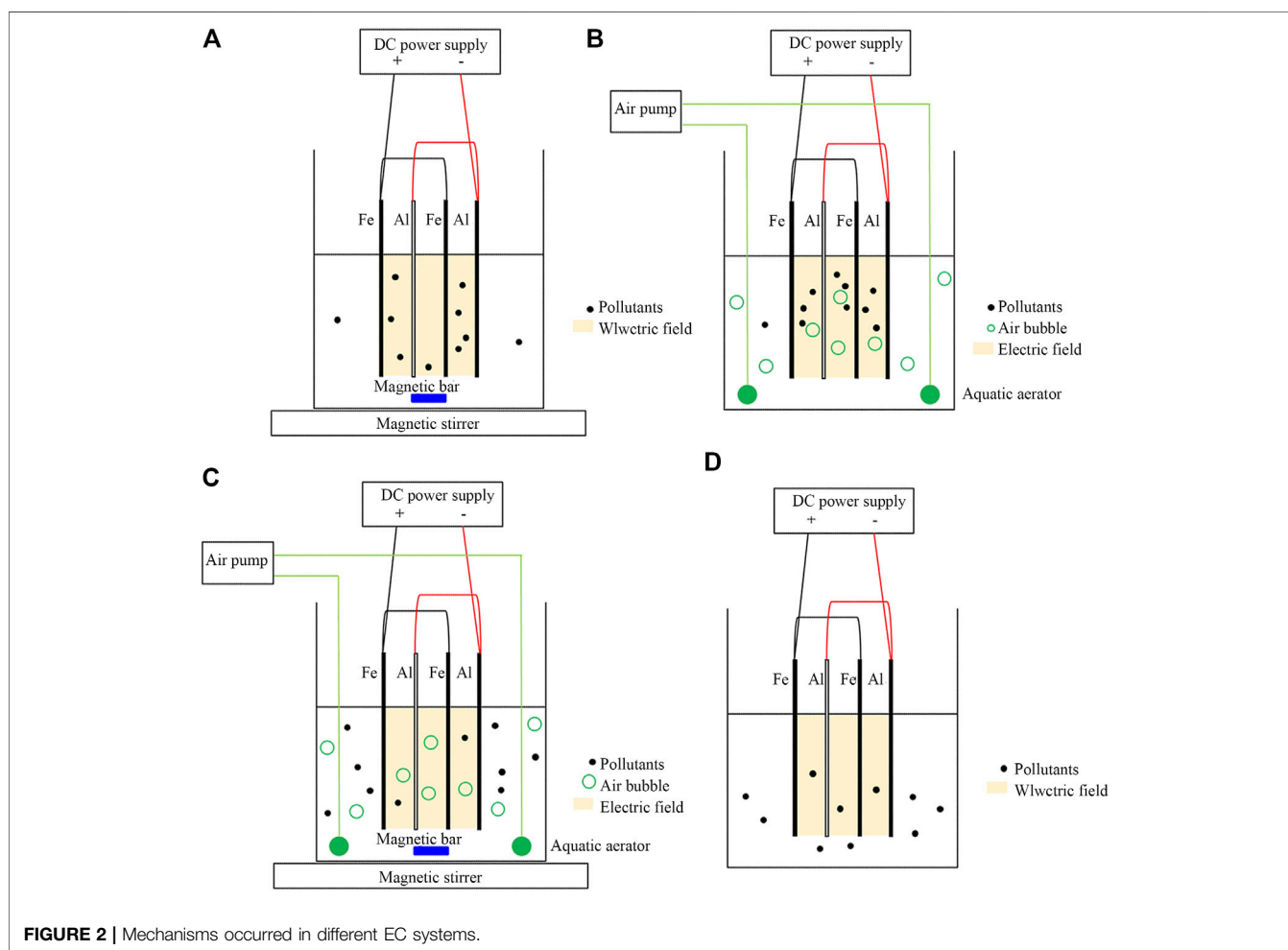
The performance of the EC system was identified by reduction of organic content, which was represented in the COD value. The COD measurement method corresponded to DIN ISO 15705 and was analogous to EPA 410.4, APHA 5220 D, and ASTM D1252-06 B. The COD solutions A + B were purchased from Sigma-Aldrich Canada Co. Ltd. The efficiency of COD removal was calculated as the following equation:

$$COD\ removal\ (\%) = \frac{COD_0 - COD_t}{COD_0} \times 100 \quad (1)$$



RESULTS AND DISCUSSIONS

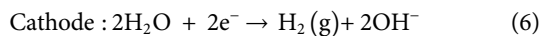
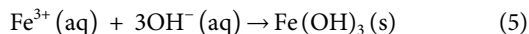
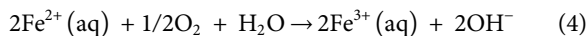
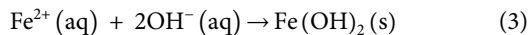
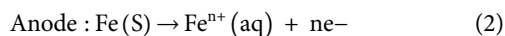
A significant factor for achieving wastewater treatment by the EC process was agitation to maintain a uniform condition and avoid the concentration gradient in the system. The EC system was normally agitated by a magnetic stirrer. From **Figure 1**, the traditional stirring EC system obtained a COD removal efficiency of 72.9%; the COD value was decreased from the initial 56,400 mg/L to 16,800 mg/L in 15 min of detention time. The COD removal efficiency was increased to a maximum of 91.0% in 45 min. The main reactions were that the iron cations were dissolved into ferrous ion (Fe^{2+}) and/or ferric ion (Fe^{3+}) from the sacrificial Fe electrode (anode), and hydroxide ion (OH^-) occurred on the Al electrode (cathode) from water hydrolysis, and eventually coagulants of $Fe(OH)_2$ and/or $Fe(OH)_3$ were formed (as explained in equations **Eqs. 2–6**) (Ghanbari et al., 2014; Hakizimana et al., 2017). The organic content in the coolant wastewater was removed through



where COD_0 was the initial concentration of COD in the coolant wastewater and COD_t was the concentration of COD in the treated water at the detention time of t .

destabilization, coagulation, and sorption processes by $Fe(OH)_2$ or $Fe(OH)_3$. The dispersion of pollutants in the stirring EC system is illustrated in **Figure 2A**. As a result of

the agitation, the pollutants were mainly exiting in the electric field, which was a reactive area for the electrocoagulation process.



The aeration method was another widely used agitation method in a biological treatment system and was applied in the EC aeration system. The COD removal efficiency of the EC aeration system reached the maximum efficiency of 90% in 15 min of detention time. The explanation was that aeration provided excellent homogeneous conditions and also served as an oxygen source, which caused an improving oxidation reaction in the system. Aeration initiated sufficient dissolved oxygen for converting generated Fe^{2+} into Fe^{3+} (Kumar et al., 2018). Since $\text{Fe}(\text{OH})_3$ was a more effective coagulant than $\text{Fe}(\text{OH})_2$ (Naje et al., 2017), therefore the performance of the EC aeration system was greater than that of the EC stirring system. In addition, continuous aeration can improve the EC performance through vertical movement and complete circulation between the electric field and the external electric field. **Figure 2B** shows that air was diffused along the system and pollutants were easily separated to the surface due to air bubbles.

In addition, the control system without stirring and without aeration achieved a lower COD removal efficiency of 63.2% in 15 min of detention time. Only pollutants existing in the electric field was coagulated and removed (**Figure 2D**). During treatment, the pollutants were slowly transported to the electric field by H_2 gas generated at the cathode. Consequently, complete circulation occurred in the control EC system, as indicated by the high COD removal of 90.6% in 45 min of detention time. It should be noted that the homogenous mixture can occur under no agitation in the small-scale system.

The stirring-aeration EC system was established with the aim of enhancing the performance of the EC system by double stirring and aeration; the stirring speed and aeration rate were the same values as the stirring EC system and the aeration EC system, respectively. However, a relatively low COD removal was observed in 15 min; its efficiency was worse than the control EC system. Double agitations led to a rapid mixing of the solution; then coagulants and pollutants obtained insufficient time in the electric field to form flocs, as illustrated in **Figure 2C**. However, the performance of the stirring-aeration EC system can reach a similar maximal COD removal efficiency of 90.5% in the longer detention time of 45 min. Although the floc formation of coagulants and pollutants was reduced, the EC performance can be recovered eventually by increasing the collision of coagulants and pollutants due to the extended detention time. Meanwhile, the rapid mixing solution affected the consumption of electricity from ions transfer (Ilhan et al., 2008). It should be noted that the stirring-aeration EC system was operated under a specific speed of stirring and a specific air flow rate. Therefore, the performance of the stirring-aeration EC system is possibly increased when the

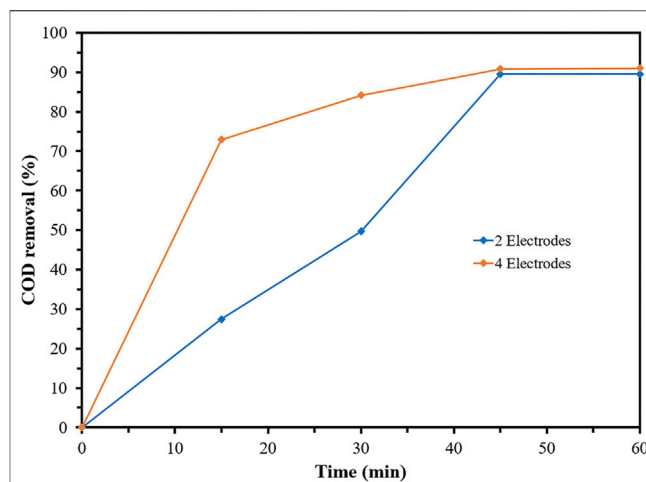
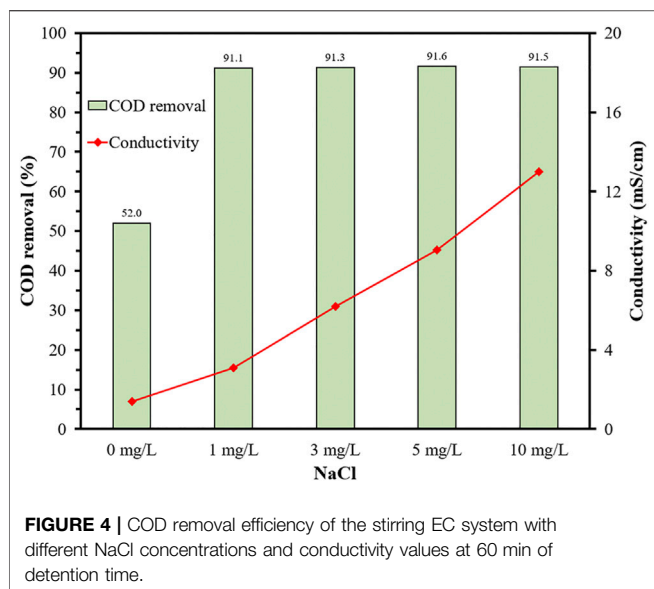


FIGURE 3 | COD removal efficiency of the stirring EC system with different numbers of electrodes.

stirring speed and air flow rate were decreased to an optimal combination; coagulants and pollutants were perfectly dispersed and collided in the solution (Bayar et al., 2011). The optimum of the aeration and stirring values should be further studied.

The stirring EC system was continuously operated at a decreasing electrode number of two in order to reduce the operating cost and electric consumption. From **Figure 3**, the stirring EC system with two electrodes obtained the lower COD removal efficiency in 15–30 min rather than that with four electrodes; ranging 27–50% for the EC system of two electrodes and 73–84% for the EC system of four electrodes. Since the distance between the electrodes of both systems was unchanged, there was no effect of electrostatic force to degrade the flocs (very short distance between the electrodes and strong electrostatic force) or develop the lower flocs (long distance between the electrodes and weak electrostatic force) (Naje et al., 2017). The lower COD removal efficiency was due to the decrease in the electric field which referred to the decrease in $\text{Fe}(\text{OH})_2$ and flocs formation. When sufficient flocs were produced, the organic compounds containing in the coolant wastewater were effectively removed, as illustrated by 90% of the COD removal in 45 min. The number of electrodes was negatively correlated with the operating time of the EC system. The supplied electrical current could be lowered by decreasing the number of electrodes (under constant current density condition); however, the operating cost was increased by extending the detention times to achieve the excellent EC performance and wastewater treatment efficiency.

The maximum efficiency of COD removal in this study was found in the longer detention time of 45 min rather than in the literature of 20–30 min (Azarian et al., 2018a; Ehsani et al., 2020). The main reason was related to a different source of wastewater. The coolant wastewater that contains high oil and grease and nonbiodegradable organic carbon. Therefore, it might take longer to degrade the large complex molecule to smaller compounds, which are easier to coagulate. An increasing biodegradability index (BOD_5/COD) and a stable COD concentration were found



during the CE process in the previous study (Pantorlawn et al., 2017). Other possible reasons were reactor design, electrode material, number of electrodes, and submerged area of electrode. When these parameters mentioned are kept in the best condition, the excellent treatment efficiency can be obtained at a short detention time, leading to an acceptable treatment cost and a practical system at the site.

In the stirring EC system, the pollutants removal efficiency and energy consumption were related to the solution conductivity due to the movement of generated ions and the flow of electric current. NaCl was one of the common salt additives to improve the solution conductivity. The increase in NaCl concentration was a direct function of conductivity, as presented in **Figure 4**. The stirring EC system with four electrodes was operated at various NaCl concentrations of 1–10 mg/L and 45 min of detention time. Coolant wastewater containing 1.4 mS/cm of conductivity obtained a COD removal efficiency of 52%. The COD removal efficiency was immediately to 91% after adding 1 g/L NaCl and achieving a conductivity of 3.1 mS/cm. At higher NaCl concentrations and conductivities, the COD removal efficiency was approximately 91%, which was the highest efficiency for coolant wastewater in this work.

In principle, the cell voltage decreases with an increase in conductivity at a constant current density; the total resistance in the solution decreases, and thus the electrical energy consumption decreases. However, excessive NaCl possibly caused an overconsumption of electrodes due to corrosion pitting and irregular electrode dissolution (Calvo et al., 2003).

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The optimized NaCl additive (e.g., <1 g/L) and the minimal detention time (e.g., <45 min) should be maintained for a cost-efficient coolant treatment using the CE process. Furthermore, metal ions were normally found in coolant waste and wastewater (Yeon and Moon, 2003; Teodosiu et al., 2014), the metal types and concentrations significantly influenced the solution conductivity as well as optimized NaCl additive. Therefore, it should be another concerning factor for different industrial wastewater.

CONCLUSION

The actual coolant wastewater was treated by EC systems with different agitations, stirring, aeration, and hybrid stirring and aeration. The aeration EC system achieved the maximal COD removal efficiency of 90% in a short detention time of 15 min, while the stirring EC system required 45 min of detention time. The significant reasons were that the complete circulation in the aeration EC system led to a homogeneous dispersion of coagulants and flocs, and dissolved oxygen also played a role in converting Fe^{2+} to Fe^{3+} and generated effective coagulant of $\text{Fe}(\text{OH})_3$. However, the combination of stirring and aeration could not provide the best performance in this work. In addition, the optimal stirring speed and aeration rate should be considered to further improve the EC process.

DATA AVAILABILITY STATEMENT

The raw data supporting the conclusion of this article will be made available by the authors, without undue reservation.

AUTHOR CONTRIBUTIONS

DT has performed experiment and data analysis. TT has did data analysis and revised manuscript. QB has did data analysis and revised manuscript. DC did experimental design, data analysis and wrote manuscript. AN design, data analysis, and wrote main manuscript. WK initiated the project, designed the experiment, did data analysis and wrote manuscript.

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