Effect of High-Rate Aeration in the Electrocoagulation Treatment of Pharmaceutical and Synthetic Textile Industrial Wastewater Effluents

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Efecto de la aireación de alta tasa en el tratamiento de electrocoagulación de efluentes de aguas residuales industriales farmacéuticas y textiles sintéticas

Efecte de l'aireig d'alta taxa en el tractament d'electrocoagulació d'efluents d'aigües residuals industrials farmacèutiques i tèxtils sintètiques

SUMMARY

Despite the extensive literature on the effects of implementing aeration in the electrocoagulation (EC) treatment of industrial wastewater, few studies evaluate the performance of this technique at high aeration rates. In this work, we present the outcomes derived from EC treatment of pharmaceutical and synthetic textile wastewater effluents in two lab-scale reactors with and without aeration, each equipped with different electrode combinations of aluminium and iron (Al/Al, Al/Fe, and Fe/Fe). In the non-aerated EC reactor, maximum chemical oxygen demand (COD) reductions of 56% and 80% were achieved for the pharmaceutical and textile effluents, respectively, with up to 100% removal of turbidity and colour with the Al/Al combination. The installation of high-rate aeration to prevent sludge accumulation on the electrode surface markedly reduced residence times and enhanced the COD removal in the pharmaceutical effluent. However, this implementation potentially reduced the process efficiency for the synthetic textile effluent as prolonged operation led to flocculation breakdown and subsequent re-dissolution of the contaminants in the water.

Keywords: COD, Current Density, High-Rate Aeration, Industrial Wastewater, Lab-Scale Electrocoagulation

RESUMEN

A pesar de la extensa literatura sobre los efectos de implementar la aireación en el tratamiento por electrocoagulación (CE) de aguas residuales industriales, pocos estudios evalúan el desempeño de esta técnica a altas tasas de aireación. En este trabajo, presentamos los resultados derivados del tratamiento EC de efluentes de aguas residuales farmacéuticas y textiles sintéticas en dos reactores a escala de laboratorio con y sin aireación, cada uno equipado con diferentes combinaciones de electrodos de aluminio y hierro (Al/Al, Al/Fe y Fe/Fe). En el reactor EC no aireado, se lograron reducciones máximas de la demanda química de oxígeno (DQO) del 56% y el 80% para los efluentes farmacéuticos y textiles, respectivamente, con una eliminación de hasta el 100% de turbidez y color con la combinación Al/Al. La instalación de aireación de alta velocidad para evitar la acumulación de lodos en la superficie del electrodo redujo notablemente los tiempos de residencia y mejoró la eliminación de DQO en el efluente farmacéutico. Sin embargo, esta implementación redujo potencialmente la eficiencia del proceso para el efluente textil sintético, ya que la operación prolongada provocó la ruptura de la floculación y la posterior redisolución de los contaminantes en el agua.

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Palabras clave*: DQO, densidad de corriente, aireación de alta velocidad, aguas residuales industriales, electrocoagulación a escala de laboratorio

RESUM

Malgrat l'extensa literatura sobre els efectes de la implementació de l'aireació en el tractament d'electrocoagulació (EC) d'aigües residuals industrials, pocs estudis avaluen el rendiment d'aquesta tècnica a altes taxes d'aireació. En aquest treball, presentem els resultats derivats del tractament EC d'efluents d'aigües residuals tèxtils farmacèutiques i sintètiques en dos reactors a escala de laboratori amb i sense aireació, cadascun equipat amb diferents combinacions d'elèctrodes d'alumini i ferro (Al/Al, Al/Fe i Fe/Fe). En el reactor EC no airejat, es van aconseguir reduccions màximes de la demanda química d'oxigen (DQO) del 56% i del 80% per als efluents farmacèutics i tèxtils, respectivament, amb una eliminació de fins a un 100% de terbolesa i color amb la combinació Al/Al. La instal·lació d'aeració d'alta velocitat per evitar l'acumulació de fangs a la superfície de l'elèctrode va reduir notablement els temps de residència i va millorar l'eliminació de DQO a l'efluent farmacèutic. Tanmateix, aquesta implementació va reduir potencialment l'eficiència del procés per a l'efluent tèxtil sintètic, ja que el funcionament prolongat va provocar la ruptura de la floculació i la posterior redissolució dels contaminants a l'aigua.

Paraules clau*: COD, densitat de corrent, aireació d'alta velocitat, aigües residuals industrials, electrocoagulació a escala de laboratorio

INTRODUCTION

According to international estimates, 90% of the wastewater generated worldwide is directly discharged to the environment (Corcoran *et al.*, 2010; Rathoure and Dhatwalia, 2015; Khalaf, Mubarak and Nosier, 2016). This release of untreated or inadequately treated wastewater poses severe consequences due to the discharge of contaminant loads that are difficult to eliminate through conventional treatments. The textile industry, for example, produces wastewater effluents with high levels of chemical oxygen demand (COD), persisting colouring pollutants, biochemical oxygen demand (BOD), total dissolved solids, total organic carbon (TOC), and total suspended solids (TSS) (Bisschops and Spanjers, 2008; Kishor *et al.*, 2021). A similar concern is caused by the presence of non-biodegradable organic matter in pharmaceutical wastewater (Sirés and Brillas, 2012; Rana *et al.*, 2017) since the high COD/BOD ratios in this type of water limit its biodegradation (Hamaidi-Chergui and Brahim Errahmani, 2019).

While a multitude of treatments have been proposed to deal with these effluents, their design must cater to the specific wastewater characteristics. Predominant technologies in this group encompass biological treatment, coagulation, and chemical treatment. Despite their potential benefits, the use of these techniques implies huge installation surface areas, permanent use of chemicals, high amounts of sludge, and management of residues (Crini and Lichtfouse, 2019; Hashim *et al.*, 2020; Bahrodin *et al.*, 2021). Electrocoagulation (EC) stands as a promising alternative due to its lower operating costs, less production of sludge, and higher efficiencies in removing organic and inorganic contaminants from various water matrices without the addition of chemicals (Barrera-Díaz, Balderas-Hernández and Bilyeu, 2018; Islam, 2019; Shahedi *et al.*, 2020; Nidheesh *et al.*, 2022).

In contaminated water, the Van der Waals forces create a weak attraction between neutral molecules. These forces induce a temporary union and foster a balance that sustains the adhesion of contaminant particles to water molecules. EC aims to destabilize this suspension by reducing the repulsion force between particles and prompting their union to form larger conglomerates (Naje and Abbas, 2013). The elimination of contaminants occurs as a result of physical and chemical processes during electrolysis. The fundamental principle of the process is driven by reduction and oxidation reactions. These reactions result in oxidation at the anode and reduction at the cathode, while treated water serves as an electrolytic solution (Rusdianasari *et al.*, 2019). Specifically, the anode, often referred to as the sacrificial electrode, dissolves and releases metallic ions. In contrast, the cathode retains its plate without dissolving (Adhoum and Monser, 2004). Table 1 shows a general scheme of the reaction mechanisms occurring in the cathode and the anode. In these expressions, M is the metal, M^{n+} is the related cation, and n is the number of transferred electrons.

Various studies indicated effective reductions of COD and elimination of colloids with diameters between 1-1000 nm, salts, polychlorinated biphenyls, cyanides, nitrites, phenols, surfactants, dyes and colourants, fats, oils and emulsions, and heavy metals, including

Table 1. *General reaction mechanisms that take place in the cathode and the anode of an EC-system (Islam, 2019; López-Guzmán, Flores-Hidalgo and Reynoso-Cuevas, 2021).*

Cathode	Anode
$2H_2O_{(l)} + 2e^- \rightarrow H_{2(g)} + 2\,OH^-_{(aq)}$	$2H_2O_{(l)} \rightarrow 4H^+_{(aq)} + O_{2(g)} + 4\,e^-$
$M^{n+}_{(aq)} + n\,e^- \rightarrow M_{(s)}$	$M_{(s)} \rightarrow M^{n+}_{(aq)} + n\,e^-$

the removal of emerging contaminants (Moussa *et al.*, 2017). Recent studies have demonstrated that the integration of external aeration into the EC system markedly enhances the removal efficiency of COD and other pollutants (Kumar, Nidheesh and Suresh Kumar, 2018; Akansha *et al.*, 2020; Khalifa *et al.*, 2020; Syam Babu, Nidheesh and Suresh Kumar, 2021; Trinh *et al.*, 2021; Nidheesh and Gökkuş, 2023). Although this enhancement is primarily attributed to the increase in the dissolved oxygen required for the occurrence of the oxidation-reduction reactions represented in Table 1, fewer studies have investigated the effect that aeration-induced stirring may cause at high aeration rates. In this work, we evaluate the effect of implementing an external high-rate aeration system into an EC reactor. The results of this evaluation are compared with that of a traditional EC treatment in a lab-scale reactor. The specific goals of this study are to (i) determine the most effective electrode materials for the EC-treatment of pharmaceutical and synthetic textile effluents in a non-aerated reactor, (ii) assess the effects of high-rate aeration in the residence time, and removals of COD, turbidity, and colour, and (iii) identify the optimal residence time for maximum removals of turbidity, colour, and COD upon implementation of aeration.

METHODS

Wastewater Characteristics

The synthetic textile effluent was prepared before each experiment with a concentration of 0.1 g/L blue dye to emulate the dosages used by local textile industries, with an initial COD concentration of 600 ± 29 mg/L. The pharmaceutical effluent was collected from untreated wastewater generated by a company within the sector before mixing with domestic wastewater. For this sample, the initial COD was greater than 10000 mg/L in all essays. All samples were stored in the absence of light at 10 ± 5 °C pending EC treatment, with no more than 2 days in storage. Following EC treatment, samples were allowed to settle in a light-isolated location for 6 h. No chemicals were added to preserve the water during this settling time. After the sedimentation period, samples were filtered with a fiberglass filter (1 µm porosity). The initial characteristics of the samples are listed in Table 2. Given the diverse range of medications produced by the company from which the pharmaceutical effluent was sourced, information on specific contaminants was not available.

Analytics

Prior to each experiment, the iron electrodes were subjected to a stripping process adapted from ASTM B 633 standard (ASTM, 2001), using 0.1 M hydrochloric acid as a washing solution. The pH and conductivity of the effluents were measured with a Horiba multiparameter, model Laqua-PC1100. COD was determined according to the closed reflux method (American Public Health Association, 2018). Turbidity was measured with a Lamotte turbidimeter, model 2020 we/wi. The intensity of colour was related to the absorbance at 450 nm. Water spectra were measured with a Cole-Parmer spectrophotometer. All parameters were measured in triplicate.
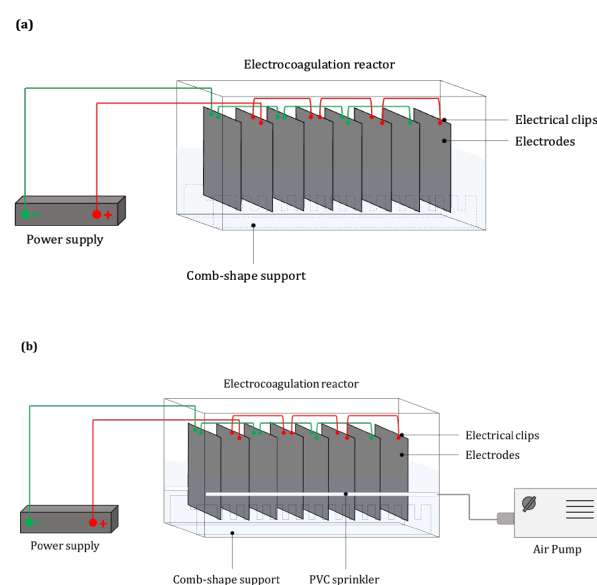

Figure 1. *Schematics of the EC lab-scale reactors without aeration (a) and with external aeration (b)*

EC System

Two batch reactors were used for the EC treatment, one without aeration (Figure 1a) and another with external aeration (Figure 1b). The non-aerated reactor, adapted from Baiza *et al.*, 2016, was built in acrylic. This reactor had a maximum volume of 1.5 L and was equipped with a drainage system for extracting treated samples. Construction of the aerated reactor involved scaling up the non-aerated reactor to achieve a 5 L-volume for incorporating the aeration system. A PVC sprinkler connected to an external aerator supplied high-rate aeration at 6-8 L/min. This aeration rate produced intense stirring in the EC reactor. As schematized in Figure 1, comb-shaped supports were placed in both reactors to maintain a fixed distance of 3 mm between the electrodes during the EC treatment. A direct current power supply with a variable voltage of 0-12 V was used for the electrical system. The current was measured with a clamp meter (Amprobe, ACDC-54NAV). The dimensions of the electrodes were 9×15 cm in the non-aerated reactor and 12×18 cm in the

Table 2. *Characteristics of the wastewater samples (N.A.: Not applicable).*

Sample	COD (mg/L)	Conductivity (mS/cm)	pH	Turbidity (NTU)	Absorbance (A.U.)
Pharmaceutical effluent	11900 ± 700	1.96 ± 0.01	7.50 ± 0.10	0.98 ± 0.03	N.A.
Textile synthetic effluent	600 ± 30	1.31 ± 0.03	8.52 ± 0.10	N.A.	1.08

aerated reactor. In both cases, a total of 8 electrodes were used for the EC treatment. As shown in Figure 1, the connection between electrodes was made in a parallel monopolar configuration and electrical clips were used to complete the electrical system of the reactor.

Experimental procedure

An initial volume of 1.5 L was utilized in all the experiments in the non-aerated reactor. The power source was set at the minimum current to reduce electrical consumption. The maximum duration of the treatment was contingent upon the initial current, facilitating the formation of at least two insoluble layers: one phase of solids, and the other phase of treated water. If the conductivity of the effluent produced excessively high currents, the residence time was reduced, with the power source's capacity serving as the operational limit. A maximum residence time of 60 min was established, with sample extractions at various intervals to assess COD and turbidity in the pharmaceutical effluent, or colour in the textile effluent. These extractions allowed the identification of the optimal residence time. All evaluations were conducted with iron (Fe) and aluminium (Al) electrodes, the most widely used electrode materials (Tezcan and Oduncu, 2014; Zhang, Lin and Hu, 2016). The combinations of electrode material and current densities are listed in Table 3 for each water type. These tests aimed to identify the optimal electrode material and maximum residence time for the highest COD, turbidity, or colour removals. Initial current densities depended on the initial conductivity of the effluent and the electrode material.

Table 3. *Electrode materials and initial current densities used in the non-aerated EC reactor.*

Type of water	Electrode material (cathode/anode)	Initial current density (A/m²)
Pharmaceutical effluent	Fe/Fe	103.9
	Al/Al	104.4
	Al/Fe	115.6
Synthetic textile effluent	Fe/Fe	63.8
	Al/Al	64.9
	Al/Fe	81.7

Upon determining the optimum electrode material and the time for maximum removals in the non-aerated reactor, the operating conditions were replicated in the aerated reactor, maintaining the current density as a scaling factor. To find the optimal residence time in the aerated reactor, samples were extracted at different treatment intervals (10 min for the pharmaceutical effluent and 5 min for the synthetic textile effluent) to determine whether the presence of high-rate aeration improved process efficiency, expressed in terms of removal capacity or the time at which maximum COD, turbidity, and colour removals were achieved.

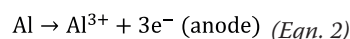The removal efficiency (%Removal) of colour and turbidity was calculated with Eqn. 1, where N_o is the initial value of the target parameter, and N is the value of the parameter at the moment of the analysis. This expression was not utilized for evaluating the removal of COD. For this parameter, the removal efficiency was assessed in terms of the COD reduction (mg/L) over treatment periods.

$$\%\text{Removal} = \left(\frac{N_o - N}{N_o}\right) \cdot 100 \quad \textit{(Eqn. 1)}$$

RESULTS

Determination of the Optimum Electrode Material in the Non-Aerated Reactor

The electrode combinations Al/Fe, Al/Al, and Fe/Fe were used in the EC treatment of both waters. Maximum residence times of 60 and 30 min were used for the pharmaceutical and synthetic effluents, respectively. In the pharmaceutical effluent, these configurations of electrode materials generated reductions of 56%, 78%, and 70% from the initial turbidity. For COD, the Al/Fe combination produced a removal of 0.9%; the Fe/Fe combination produced no removal of the contaminant load; and the Al/Al electrodes achieved a 24% COD removal. During the treatment of the textile effluent, all the combinations generated 100% colour removal. For COD, the Al/Al combination generated a 53% removal of COD, while the Al/Fe and Fe/Fe combinations generated no removals of the contaminant load. Therefore, the combination Al/Al was defined as the optimum electrode material for the treatment of both water types. Aluminium was also found as optimum electrode material for the treatment of other pharmaceutical and textile effluents (Chaabane *et al.*, 2013; Khandegar and Saroha, 2013; Nariyan, Aghababaei and Sillanpää, 2017; Bener *et al.*, 2019). The reaction mechanisms taking place at the aluminium electrodes are expressed by Eqns. 2 and 3.

$$Al \rightarrow Al^{3+} + 3e^- \text{ (anode)} \quad \textit{(Eqn. 2)}$$

$$2H_2O + 2e^- \rightarrow H_2 + 2OH^- \text{ (cathode)} \quad \textit{(Eqn. 3)}$$

As the anode releases Al^{3+}, this species undergoes subsequent hydrolysis reactions that yield various monomeric subproducts, including , $Al(OH)^{2+}$, $Al(OH)_2^+$ and $Al(OH)_3$ (Mouedhen *et al.*, 2008). Dimeric, trimeric, and polynuclear hydrolysis products may form as well, including $Al_2(OH)_2^{4+}$, $Al_3(OH)_4^{5+}$, $Al_6(OH)_{15}^{3+}$, $Al_7(OH)_{17}^{4+}$, $Al_8(OH)_{20}^{4+}$, $Al_{13}O_4(OH)_{24}^{7+}$ and $Al_{13}(OH)_{34}^{5+}$ (Larry D, Joseph F and Barrow L, 1982; Gerasko *et al.*, 1993; Bayramoglu, 2004; Kobya *et al.*, 2006; Mouedhen *et al.*, 2008). These cationic products potentially react with OH^- and transform into amorphous $Al(OH)_3$ (Larry D, Joseph F and Barrow L, 1982; Bayramoglu, 2004; Kobya *et al.*, 2006). Therefore, the reduction of the pollutant load in the pharmaceutical and synthetic textile effluent is likely to occur by the neutralization of negatively charged colloids and the incorporation of impurities in the $Al(OH)_3$ precipitate, a process called "sweep flocculation" (Mouedhen *et al.*, 2008).

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Upon determining the optimum electrode material and the time for maximum removals in the non-aerated reactor, the operating conditions were replicated in the aerated reactor, maintaining the current density as a scaling factor. To find the optimal residence time in the aerated reactor, samples were extracted at different treatment intervals (10 min for the pharmaceutical effluent and 5 min for the synthetic textile effluent) to determine whether the presence of high-rate aeration improved process efficiency, expressed in terms of removal capacity or the time at which maximum COD, turbidity, and colour removals were achieved.

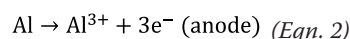The removal efficiency (%Removal) of colour and turbidity was calculated with Eqn. 1, where N_o is the initial value of the target parameter, and N is the value of the parameter at the moment of the analysis. This expression was not utilized for evaluating the removal of COD. For this parameter, the removal efficiency was assessed in terms of the COD reduction (mg/L) over treatment periods.

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Effect on Turbidity and Colour

As shown in Table 4, 100% turbidity removal in the pharmaceutical effluent was achieved after 10 min and this level remained constant during the evaluation period. Upon implementation of aeration, this trend remained unaltered. A similar trend was observed in the synthetic textile effluent. In this case, a maximum colour removal of 100% was achieved after 5 min of treatment without aeration. When aeration was implemented, the peak removal value attained this maximum after 50 min of treatment. Despite this longer time frame, the colour removal exhibited a steady increase throughout the EC treatment, with a remarkable removal of 99.5% at 5 min which is comparable with the maximum removal obtained in the EC treatment without aeration. Overall, these results show that both turbidity and colour removal achieved their peak values at the initial stages of the treatment of the pharmaceutical and textile effluents, irrespective of the presence or absence of aeration. Consequently, these parameters were not established as part of the criteria for determining the residence time of the process.

Table 4. *%Removal of turbidity and colour in the pharmaceutical and synthetic textile wastewater effluents.*

Treatment time (min)	%Removal of turbidity in the pharmaceutical effluent		Treatment time (min)	%Removal of colour in the synthetic textile effluent	
	Without aeration	With aeration		Without aeration	With aeration
0	0	0	0	0	0
10	100	100	5	100	99.5
20	100	100	10	100	99.7
30	100	100	15	100	99.8
40	100	100	20	100	99.8
50	100	100	25	100	100.0
60	100	100	30	100	100.0

Effect on COD

Figure 2a shows the COD concentration over time for the pharmaceutical effluent. As observed, the COD concentration in the water depicted a decreasing trend during the first 40 min of treatment in the reactor without aeration, with a minor increment in the 10 min of treatment. The highest removal was 30% and occurred at 40 min. Upon implementation of the high-rate aeration, a maximum COD removal of 56% was achieved at 10 min of treatment. Beyond this optimal time, the COD level increased due to the stirring caused by the high aeration rate. After 50 min of treatment, the final COD concentration was comparable to that obtained without aeration.

The COD removal in the treatment without aeration of the synthetic textile effluent (Figure 2b) exhibited a predominant decreasing trend, with a maximum removal of 80% at 25 min. When the aeration was implemented, the COD levels followed an oscillatory trend, with removal peaks of 20% at 5 min and 51% at 15 min. After 15 min, significant removals were not achieved, and the COD concentration increased even above its initial value. This increment may be associated with the presence of the dissolved aluminium species described by Larry D, Joseph F and Barrow L, 1982; Gerasko *et al.*, 1993; Bayramoglu, 2004; Kobya *et al.*, 2006; Mouedhen *et al.*, 2008.

The optimal time was defined as that yielding the highest removals of COD. Given that turbidity and colour were completely removed in the first minutes of treatment for both types of water, the optimal residence time corresponded to the peaks of 30% and 80% for the pharmaceutical effluent and synthetic water in the reactor without aeration, with values of 40 min and 25 min in each case. The residence time was also defined as one of the most sensitive variables due to the oscillations in COD removal upon prolonged operation and overmixing. This oscillatory trend is associated with the destruction of flocs generated by extended treatment periods (Abdelwahab, Amin and El-Ashtoukhy, 2009; Trinh *et al.*, 2021).
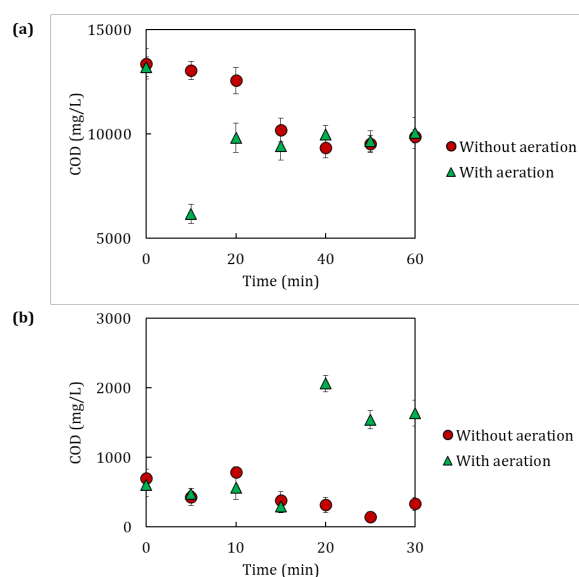

Figure 2. *COD removal in the EC treatment of the pharmaceutical (a) and synthetic textile (b) wastewater effluents.*

The implementation of aeration led to a significant improvement in treatment time for both waters, resulting in reductions of up to 80% of the residence time for achieving maximum removals. This behaviour is caused because the presence of air enhances the movement of sludge in the reactor and the removal of contaminants by adsorption and sweeping phenomena (Abdelwahab, Amin and El-Ashtoukhy, 2009; Kumar, Nidheesh and Suresh Kumar, 2018; Akansha *et al.*, 2020). As result, the treatment time for pharmaceutical effluent was reduced from 40 min to 10 min, while the maximum COD removal percentage increased from 30% to 56%. Despite this improvement in the pharmaceutical ef-

fluent, the maximum COD removal of the synthetic effluent decreased from 80% to 51%, and the treatment time decreased from 25 min to 15 min. In previous studies, this decline in removal efficiency has been attributed to overmixing. High levels of aeration may induce excessive mixing, leading to the disintegration of the sludge and the subsequent re-solubilization of contaminants. The implications of these findings are significant in an industrial context, as they underscore the necessity to fine-tune the aeration flow rate. Moreover, these results demonstrate that intense agitation may potentially undermine the benefits of electrocoagulation compared to other treatment methods.

CONCLUSIONS

In this study, EC was used to treat wastewater from a pharmaceutical industry and a synthetic effluent was prepared according to the dosages used by industries in the textile sector. For the pharmaceutical effluent, the Al/Al combination with a current density of 104.4 A/m^2 produced maximum removals of 30% COD and 100% turbidity after 40 min of treatment. Incorporation of aeration reduced the treatment time to 10 min and increased COD removal to 56%, maintaining total turbidity reduction. Therefore, optimal conditions for treating this effluent include the use of aluminium electrodes, a current density of 104.4 A/m^2, and a residence time of 10 min implementing high-rate aeration in the reactor. For the synthetic textile effluent, the evaluation of the same combinations of electrode materials showed that aluminium electrodes and 64.9 A/m^2 current density produced the optimum results, with an 80% reduction in COD load and 100% colour elimination at 25 min of treatment. After implementing aeration, the optimal treatment time was reduced to 15 min, but maximum COD removal dropped to 51%. Overall, these results show that the improvement of the process should be conducted based on a target parameter. For instance, if the goal is to reduce residence time in the batch reactor, the introduction of aeration would be beneficial, albeit at the expense of COD removal, and vice versa. Given the substantial benefits that EC presents over alternative methods, it remains crucial to optimize the integration of aeration. As demonstrated in this study, including external high-rate aeration into the EC system may enhance certain parameters but markedly diminish the overall efficiency of the process.

ACKNOWLEDGMENTS

DECLARATION OF COMPETING INTERESTS

The authors declare no known competing financial interests or personal relationships that could have appeared to influence the results reported in this paper.

Index of Abbreviations
A.U.:   Absorbance units
BOD: Biological Oxygen Demand
COD: Chemical Oxygen Demand
EC: Electrocoagulation
NTU: Nephelometric Turbidity Unit
TOC: Total Organic Carbon
TSS: Total Suspended Solids

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