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Impact of operating parameters of electrocoagulation-flotation on the removal of turbidity from synthetic wastewater using aluminium electrodes

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ARTICLE INFO	A B S T R A C T
Keywords: Electrochemical treatment Electrocoagulation Electroflotation Turbidity removal Aluminium electrode	This study analyzed the dominant operating parameters of electrocoagulation-flotation (ECF) that would influence the removal efficiency of turbidity from synthetic wastewater using aluminium electrodes with an interelectrode distance of 5 mm. These parameters included current density (CD), initial pH, electrolytic conductivity, coagulant dosage, operating time, initial turbidity concentration, and stirring speed. The initial turbidity concentration and electrolytic conductivity were synthesized based on typical concentrations of various resources such as drinking water, wastewater, seawater, and oil field-produced water. A novel approach has also been proposed for the evaluation of EC performance and selection of an appropriate process for the removal of sludge based on the intake's initial concentration. The experimental results revealed that the optimal conditions for removal of turbidity (90 %) were attained at CD = 0.0028 A/cm^2 , initial pH = 7.3, operating time = 5 min, and stirring speed = 500 rpm. The settling times of 5 and 15 min were also evaluated, in order to optimize the

1. Introduction

Electrocoagulation (EC) is an electrochemical technology for treatment of water and wastewater through generation of in-situ metal coagulant by electrolytic dissolving sacrificial anode materials triggered by electric current applied through the electrodes (Hakizimana et al., 2017). Generally speaking, the EC electrode material is prepared using aluminium (Al) or iron (Fe) materials (Chen, 2004; Mohammadi et al., 2017; Mahvi, 2006; Ciorba et al., 2000). The utilization of Al or Fe electrode in EC depends primarily on the solution's pH. According to the Al/Fe-pH diagram, the suitable pH ranges for formation of Al(OH)₃ is 5.5-7.5 and for Fe electrode, the alkaline pH would be suitable (Hakizimana et al., 2017). Many wastewaters have a neutral pH (e.g. food industries) in which, the Al electrode is preferably used. EC appears to be one of the most effective approaches for wastewater treatment which has attracted a great deal of attention because of its versatility, safety, selectivity, and low detrimental environmental impacts (Rajeshwar et al., 1994; Mouedhen et al., 2008).

The benefits of EC over chemical coagulation include, but not limited

to, less required chemicals, lower production of sludge, fast operating time, low environmental footprint, and simple operation. Simultaneously, the major disadvantage of EC compared to chemical coagulation (usually ferric or aluminium chloride/sulfate) is the high required conductivity of water. This fact is especially relevant for drinking water treatment, as conductivity cannot be enhanced by salts due to total dissolved solids (TDSs) limitations in drinking water (Dubrawski and Mohseni, 2013).

design of the settling tank. The results also revealed that ECF formed the flocs larger than 20 um, so that after

filtration with a filter paper of 20 µm, a removal efficiency of 98 % was attained.

Electrocoagulation-flotation is capable of reducing waste production in wastewater treatment due to vertical electrode configuration, in which, the gas bubbles that are formed on the cathode electrode would then attach to contaminants before rising to the surface, under which higher removal efficiency would be expected (Butler et al., 2011), while in horizontal EC configuration, the depth of the reactor is not enough, causing a meagre impact of gas bubbles. EC has been applied successfully to remove phenolic (Adhoum and Monser, 2004), decolorize reactive dye solutions (Daneshvar et al., 2006), clarify suspended clay solutions (Holt et al., 2006), treat textile wastewater (Khandegar and Saroha, 2013), breaking the emulsified oil (Biswas and Lazarescu,

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Abbreviations: ECF, Electrocoagulation-Flotation; DC, Direct Current; AC, Alternating Current; PR, Polarity Reversal; NTU, Nephelometric Turbidity Unit; ZPC, Zero Point of Charge; TDS, Total Dissolve Solids; EDL, Electric Double Layer.

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1991), and removal of heavy metals (Eiband, 2014) from aqueous solutions. In case of emulsion, the presence of aluminium ions and aluminium complexes destabilize the colloidal particles which would then be breaking the emulsion. This process leads to flocculation that generates bigger particles. Then, the flocs settle and form sludge at the bottom of the reactor (Singh et al., 1998).

The destabilization mechanisms in EC process, for removal of the contaminants, particle suspensions, and breaking of emulsions can be summarized into three successive steps. Firstly, the compression of diffuse double layer around the charged species by interactions of ions generated by oxidation of the sacrificial anode. Secondly, the charge neutralization of ionic species (reduction of the electrostatic interparticle repulsion) present in wastewater by counter ions produced by the electrochemical dissolution of the sacrificial anode. Thirdly, the flocs would be formed as a result of coagulation which could then create a sludge blanket that entraps and bridges colloidal particles which still exist in the aqueous medium (Thirugnanasambandham et al., 2014; Khaled et al., 2019).

In general, in designing electrocoagulation reactors, there are two electrode configurations, namely monopolar and bipolar in series or parallel circuits (Noersatyo et al., 2020; Cabrales and Machuca, 2014). The shape of the electrodes such as parallel plate (Malakootian et al., 2010), punched-hole electrodes (Khandegar and Anil K. Saroha., 2021), cylinders and tubes (Ahmed et al., 2018) may also affect the performance of the electrocoagulation which among them, parallel plate electrodes have performed in most studies (Mollah et al., 2001).

In terms of current type, direct current (DC) and alternating current (AC) are used in electrocoagulation processes, although, alternating current (AC) has rarely been evaluated for electrocoagulation and only in discontinuous operation (Vasudevan et al., 2011; Keshmirizadeh et al., 2011). Operating with the polarity reversal electrocoagulation (PR-EC) technic is also the most commonly used passivation control strategy that entails periodically switching the direction of a direct current, and is often referred to as an alternating pulsed current (Eyvaz, 2016; Mao et al., 2008; Mansoorian et al., 2014).

One of the most essentials for reducing the total cost of EC operation is to decrease the electrodes internal resistance drop (IR-drop) to enhance the current performance by improving the state of turbulence (Panizza and Cerisola, 2004). It has also a positive effect on the removal of gas bubbles (typically hydrogen) on the electrode surfaces (the passivation effect of based-gases).

Several factors have been investigated individually in parallel plate (monopolar and bipolar configuration) EC reactor design, including: current density (i) (Chen et al., 2000; Larue et al., 2003; Holt et al., 2005; Golder et al., 2007; Zhu et al., 2007; Addy, 2009; Jiang et al., 2002), coagulant dosing rate (Holt et al., 2005), pH adjustment (Bagga et al., 2008; (2008)), and stirrer effect (Vázquez et al., 2012). Nonetheless, the impacts of these factors have not thoroughly been reported on the formation of flocs and sludge behavior, through which, the selection and design of down-stream process can be optimized. Thus, the objectives of this study are to investigate the effect of ECF parameters on the formation of flocs, only with respect to nanoparticles (5-20 nm), as well as getting the knowledge of the sludge behaviour in settling tanks, in order to select an appropriate process for removal of sludge. In fact, there is rarely any report of the criteria used for the design of ECF reactors in previous respective EC studies found in the open literature (Vázquez et al., 2010; Barrera-Díaz et al., 2011). It is therefore imperative to characterize the effects of these factors of the ECF reactors on the cost reduction associated with laboratory tests (Barrera-Díaz et al., 2011). The scope of this study has been limited to laboratory scale, batch reactor operation for investigation of the effects of various operating parameters such as current density, initial pH, operating time, turbidity concentration, stirring speed, electrical conductivity on turbidity removal efficiency.

2. Material and methods

2.1. Operating conditions

An efficient EC reactor is the one that would produce the exact dosage of coagulant to assure the maximum removal efficiency. In other words, the reactor should produce sufficient metal ions from the anode electrode and hydroxide (OH⁻) from the cathode side to form coagulant. If the amount of the produced coagulant is not sufficient, then the removal efficiency would be low. In the case of excessive coagulant dosage though, the destabilization would take place leading to the waste of energy. Accordingly, to have an efficient EC reactor, the optimum number of factors that would influence the removal efficiency should be determined through experimental study. Current density (CD), initial pH, concentration, operating time, conductivity, and stirring speed are the main factors that have a significant influence on the EC treatment process which are presented in Table 1. As for the selected range of turbidity in this table, it should be pointed out that for real wastewater, (e.g. domestic wastewater), a wide range of turbidity has been reported. For instance, Mandal, 2014; reported a minimum turbidity of 39.24 NTU and a maximum of 43.00 NTU for wastewater of Phagwara, respectively. The average turbidity of 3 years was reported to be 41.10 NTU. Having said though, Ghazy et al., 2013; reported that the maximum influent turbidity of domestic wastewater was 134.5 NTU.

2.2. EC cell construction and electrode arrangement

The EC cell has been constructed in the mechanical workshop of the Technical University of Dresden in Germany (see Figs. 1 and 2). It has been designed according to the surface-area-to-volume ratio (S/V), which is a profoundly important reactor design parameter. This is the only key scale-up parameter in plant design that allows developing EC full-scale equipment from laboratory scale, keeping the same interelectrode distance when using electrode plates. (Mechelhoff et al., 2013).

The characteristics of the investigated EC are as follows:

- 1. Total volume of Reactor: 1.71 L
- 2. Volume of free space (top of the reactor): 0.19 L
- 3. Volume of electrode space (solid + liquid): 1.14 L
- 4. Volume of stirrer and primary settling space (bottom of the reactor): 0.285 L
- 5. Volume of foam collection part: 0.225 L
- 6. Volume of Al- electrodes (solid): 0.285 L
- 7. Total electrode surface: 1428 cm^2

The EC cell has been constructed from acrylglas in which, in continuous mode, the feed flows to the cell through the inlet pipe, located in the bottom and is drained from the outlet pipe. Nevertheless, in this study, the inlet and outlet pipes were closed so that, the cell would only be operated in a batch mode. The anode and cathode electrodes have also been constructed from aluminium (99.5 %) with a thickness of 2 mm, width of 85 mm and height of 120 mm with an interelectrode space of 5 mm (see Fig. 2).

Table 1

Factors and range of operating conditions.

Factors	Unit	Range
Current density	A/cm ²	0.0014, 0.0028, 0.0035, 0.0042, 0.0084
Voltage	V	2–10
Initial pH		4–12
Turbidity	NTU	10, 20, 40, 80
Operating time	min	3–11
Conductivity	ms/cm	0.34, 1, 2, 30, 150
Stirring speed	rpm	100-800
Inter-electrode distance	mm	5, 10, 20



Fig. 1. A) a schematic of the cell and electrodes arrangement, b) constructed cell and electrodes.



Fig. 2. Monopolar electrode configuration with a 5 mm inter-electrode distance, (anode: 7 electrodes, cathode: 7 electrodes). a) a schematic of electrode configuration, b) constructed electrodes.

The aluminium substance for the anode and cathode was selected from alloy EN AW-1050A due to the high purity of Al and good electrical conductivity, which contribute to the reduction of operating costs. The physical compositions of alloy EN AW-1050A are presented in Table 2.

Table 2

Parameters (at 20° C) Range of values Density (g/cm ³)	2.7
Solidification (°C)	646–657
Electrical conductivity (mS/cm)	34–36
Thermal conductivity (w/m.K)	210–220
Coeffizient (10 ⁻⁶ /K)	23.5
Modulus of elasticity (N/mm ²)	70.000

2.3. Synthetic wastewater

In total, 77 runs were performed in this work, out of which, 17 runs as calibration tests and 60 runs as main tests. Before each run, the synthetic wastewater was prepared according to the required parameters of the run number. The fluid volume for all the tests was equal to 1260 ml so that the electrodes were completely covered with the solution.

Turbidity is a major parameter in wastewater which can usually be originated from colour or suspended particles that should be removed. Additionally, some industries have only turbidity problem, such as fruit processing plants that the major problem is the high concentration of particles and solids in wastewater which increase the turbidity and should be treated. The reason for the use of synthesis of wastewater was to investigate the potential removal of turbidity by nano particles because in real wastewater there are various types of impurities that may affect the process. The colloidal particles (Nano Silicon Dioxide) were used to produce a turbidity solution with a concentration of 10, 20, 40, and 80 NTU (Nephelometric Turbidity Units). The size of these particles ranged from 5 to 15 nm. For the reduction of pH, hydrochloric acid (HCl 5 %) and for higher pH values, sodium hydroxide (NaOH 5 %) were used. For changing the electrical conductivity, a natural fine sea salt was used to investigate the effect of the turbidity removal efficiency in the wastewater conductivity (1 and 2 mS/cm), seawater conductivity (30 mS/cm) and oil field produced water conductivity (150 mS/cm).

2.4. Analytical methods and EC procedure

Turbidity was analyzed for remaining colloidal particles by a turbidity meter (model Turb 430 IR produced with Xylem in Germany), temperature, pH and conductivity were continuously monitored (before and after EC by a HQ40d Dual-Input Digital Multi-Parameter Meter manufactured by HACH, USA). Metal concentration released in the EC cell was measured to determine how much it deviates from the theoretical value (Faradaýs law) using the Hach Lange LCK 301 kit. The zeta potential has also been measured before the treatment to estimate the stability of the particles. Around 20 percent of the particles had a zeta potential of + 12 to + 15 mV and 80 percent a zeta potential of + 23 to + 25 mV. A DC power supply (PS-3010F, 30 V, 10 A) supplied DC current in galvanostatic mode. Electrodes were washed with deionized water, rinsed with 2 % HCl once surface films took place. After electrolysis, the electrodes were removed and the reactor used as a settling tank so that, the turbidity was measured in time intervals of 5 min and 20 min after the EC turned off respectively. Afterwards, the solution was filtered with a paper filter size of 20 μm then 5 $\mu m.$ The utilization of a settling tank before filtration was due to the formation of the flocs which may take a while. This should also be carried out in a separate reactor, in which, the flocs have time to become bigger. The filtration efficiency will be reduced if one filters the outflow after the EC, since some flocs are small enough to pass through the filter. The experimental setup has shown in Fig. 3 in which the EC cell was placed on a magnetic stirrer and the supplied DC power was connected to the anode and cathode electrodes at a constant operating time.

3. Results and discusion

3.1. Anodic and cathodic reactions

In EC, when an external direct current voltage is applied, then different electrochemical reactions may take place at the anode and



Fig. 3. Experimental setup for ECF process.

cathode of an electrolytic cell. EC involves generation of in-situ metal coagulant by electrolytic dissolving sacrificial anode materials (aluminium or iron) (Burns et al., 1997). The metal used in this research was aluminium and the anodic process, involves the oxidative dissolution of aluminium into aqueous solution and presented in reaction (1). In the cathode, the reductive dissociation of water would also take place, see reaction (2).

Anode;
$$Al_{(s)} \rightarrow Al_{(aq)}^{3+} + 3e^-$$
 (1)

Cathode:
$$2H_2O + 2e^- \rightarrow H_{2(g)} + 2OH_{(aq)}^-$$
 (2)

The cathodic reaction, has three important implications that can be used to improve the EC process so then higher pollutant removal efficiency would be expected: (1) the production of ample hydroxyl ions (OH⁻) which then react in bulk solution with aluminium cations to form coagulants; (2) hydrogen gas (H₂) is produced which contributes to the destabilization of colloidal particles leading to flocculation, and (3) pollutants can be floated by their adhesion onto tiny the bubbles formed by the hydrogen evolution (electro-floatation) (Poon, 1997; Casqueira et al., 2006; Howe et al., 2012).

Aluminium ions (Al^{3+}) produced by electrolytic dissolution of the anode, which act as coagulant reagent, hydrolyze and form mononuclear complexes according to the following sequence (Burns et al., 1997):

$$Al^{3+} + H_2O \rightarrow AlOH^{2+} + H^+$$
(3)

$$AIOH^{2+} + H_2O \rightarrow AI(OH)_2^+ + H^+$$
(4)

$$Al(OH)_2^+ + H_2O \rightarrow Al(OH)_3 + H^+$$
(5)

$$Al(OH)_3 + H_2O \rightarrow Al(OH)_4^- + H^+$$
(6)

3.2. Electrical double layer and particle stability

Many mineral surfaces contain surface functional groups (e.g., hydroxyl) which their charge depends on pH. Silica has hydroxyl groups on its exterior surface, and these can accept or donate protons. The pH corresponding to a surface charge of zero is defined as the zero point of charge (ZPC). Above ZPC, the surface charge will be negative (anionic), and below the ZPC the charge will be positive (cationic). The zero point of charge, as shown in Fig. 4, for silica is at pH of 2, whereas the zero point of charge for alumina is about pH of 9 (PARKS, G. A., 1967; Hakizimana et al., 2017).

The processes described above mostly lead to a negative surface charge on particles that would attract positive counter ions on the



Fig. 4. Variation in particle charge with pH. adopted from PARKS, G. A., 1967

particle's surface. Having said though, a layer of cations binds tightly to the surface of a negatively charged particle by electrostatic and adsorption forces to form a fixed adsorption layer, which is known as the Stern layer. This imbalance of electrical potential and the thermal motion will distribute ions in a second region, referred to as the diffusive layer, to achieve a more neutral total charge. The adsorbed and diffuse layers are known as the electric double layer (EDL). The electrical repulsive force resulting inside the double layer will avoid approximation of a second particle similarly charged in this region, thus creating a stable suspension (PARKS, G. A., 1967). The principal mechanism controlling particle stability is electrostatic repulsion. The reducing or eliminating the repulsive forces to aggregate the particles and form flocs is known as destabilization.

3.3. Effect of current density (CD)

In all EC processes, current density is the most important operating parameter (Addy, 2009). Current density determines the evolution of hydrogen gas (H₂) at the cathode and the coagulant dosage at the anode, governed by the Faraday's law (Lin and Chen, 1997). In this work, there were 14 electrodes in total, 7 electrodes as anode and 7 electrodes as cathode. The area of all electrodes was equal to 714 cm² (8.5 cm * 12 cm = 102 cm², \rightarrow 102 cm² * 7 = 714 cm²). It should be noted that, when calculating the current density, only the area of one electrode would be taken into account (provided that the area of the anode is equal to the area of the cathode).

After calibration, a range of currents was selected for the runs, presented in Table 3. As presented in this table, and Fig. 5, the best removal efficiency has been attained for the current density of 0.0028 A/cm² with a final turbidity 4.5 NTU (removal efficiency of 89 %). Higher removal at lower current densities shown in this work was in accordance with several studies (Holt et al., 2005; Zhu et al., 2007; Addy, 2009; Jiang et al., 2002). As reported in Table 3, at higher CDs, the removal efficiency was decreased which is in contrast with the results reported by Mansoorian et al, (Daneshvar et al., 2006), that the removal of lead and zinc increases with increase of current density. They have attributed it to the rise in absorbent coagulant produced in situ, resulting in the rapid removal of lead and zinc. The operating time of the EC process was 5 min and after removing the electrodes, the EC cell acted as a settling tank. As discussed in section 2.4, the time intervals of 5 and 20 min were considered for the settling time.

After measuring the turbidity, the solution in the settling tank was filtrated with two various filters sizes of 5 and 20 μ m to evaluate the unsettled residuals and the required filtration pore size in this process. Provided the good performance of the EC process in the production of the large flocs, the residuals in the solution were filtrated with a paper filter of 20 μ m. The effect of EC has been shown in Fig. 6. When the feed was filtrated without EC, there was no noticeable effect of filtration due to the size of the nanoparticles. When EC was applied prior to filtrated. The measured turbidity after the filtration with filter size of 20 μ m was 0.08 NTU. It should be noted that, the limitation of the turbidity for drinking water is 1 NTU (according to the drinking water standard in Germany).

Table 3

Effect of selected currents on removal efficiency (tests conditions: Initial turbidity: 40 NTU, EC time: 5 min, initial pH: 7.3, conductivity: 2 mS/cm, stirring: 500 rpm).

Curent (A)	Curent density (A/cm ²)	voltage	Final turbidity (NTU)	Removal efficiency (%)
1	0.0014	2.24	9.1	77.5
2	0.0028	2.33	4.5	89
2.5	0.0035	3.77	6	85
3	0.0042	2.04	12.9	67
6	0.0084	7.79	27	32.5



Fig. 5. The final measured turbidity based on the applied current density (tests conditions: Initial turbidity: 40 NTU, EC time: 5 min, initial pH: 7, conductivity: 2 mS/cm, stirring: 500 rpm).

3.4. Effect of initial pH

The initial pH is one of the important factors which would greatly affect the performance of EC process (Kobya et al., 2003; Can et al., 2003; Daneshvar et al., 2003; Mohammed et al., 2018). The effect of initial pH on removal of turbidity can be discussed from two perspectives. Firstly, as reported in section 3.2, the pH corresponding to a surface charge of zero is defined as the zero point of charge (ZPC). As shown in Fig. 4, ZPC for silica (applied as colloidal particles in this work) would be at pH value of 2. Secondly, depending on pH and chemical characteristics of the solution, aluminium ions may be found in different forms and phases (Zheng et al., 2017).

The effect of initial pH on the EC process was examined at different pH values, which presented in Table 4 and Fig. 7. It shows the increase of turbidity removal when pH is between 7 and 7.3 changes from 80 % (for a voltage of 2.9) to 89 % turbidity removal (for a voltage of 2.23), respectively. Similar results have been reported by other studies which reported the increase of turbidity removal in EC with an optimum pH of 7, CD of 0.01266 A/cm^2 , and a time exposure of 12 min with aluminium electrode (Mohammadi et al., 2017; Barrera-Díaz et al., 2005). Zheng et al. (Barrera-Díaz et al., 2005) reported that the optimal values of ECF time, applied current, NaCl concentration, and pH for removal of turbidity were 5 min, 0.35 A, 0.4 g/L NaCl in distilled water, and pH of 7, respectively. It is noteworthy that, the electrocoagulation can be applied as pH regulator because it increases the acidic pH and reduces the alkaline pH. This was confirmed during the experiments and the results are presented in Table 4 (comparison of the initial and final pH values).

To better understand the influence of initial pH on the removal efficiency of turbidity in this work, Figs. 8-10 present images of the solution for settling times of 5 and 15 min for the sake of comparison. As observed in Fig. 8, due to low concentration of OH^- and low negative surface charge (specially in pH = 4), the removal efficiency of turbidity is very low. Additionally, there are no significant differences between settling times of 5 and 15 min. Since in the acidic pH, the amount of Al (OH)₃ as counter ions is very low, the flocs cannot form, thus, the particles repulse each other leading to undesired suspension in the solution.

In Fig. 9, due to the release of OH^- and production of $Al(OH)_3$ in the solution, the destabilization of colloids has taken place by adsorption of positively charged ions. Then, the aggregated colloids and large flocs were formed, as observed in Fig. 9a. It should be also noted that as seen in this figure, most of the flocs were settled on the bottom of the tank but a part of the flocs were also floated on the surface. The reason for this observation has been discussed in section 3.5. It was also observed that a settling time of 5 min is not sufficient, but after 15 min, all the flocs were



Untreated wastewater Turbidity 40 NTU treated (EC + filtration) Turbidity 0.08 NTU

Filtrated wastewater (no EC)

Fig. 6. Effect of EC process in filtration.

Table 4

The initial and final pH values and removal efficiency (test conditions: Initial turbidity: 40 NTU, EC time: 5 min, CD: 0.0028 A/cm^2 , conductivity: 2 mS/cm, stirring: 500 rpm).

Initial pH	Final pH	Voltage (V)	Final turbidity (NTU)	Removal efficiency (%)
4	5.7	3.41	34.7	13.2
5	5.36	3.06	37.4	6.5
6	6.07	2.68	33.6	16
7	7.24	2.9	7.8	80.5
7.3	7.5	2.23	4.5	89
7.5	7.4	3.66	7.5	81.2
8.4	7.8	3.29	8.6	78.5
9.5	8.8	3.08	11.5	71.2
10.5	9.77	2.99	14.3	64.2
12	11.7	2.3	17.2	57



Fig. 7. The effect of initial pH on removal of turbidity (tests conditions: Initial turbidity: 40 NTU, EC time: 5 min, CD: 0.0028 A/cm², conductivity: 2 mS/cm, stirring: 500 rpm).

settled.

The well-designed EC process is the one that produces large flocs during short operation time. To achieve this goal, the reactor should be able to accelerate the reaction between Al^{3+} and OH– to form Al(OH) complex. In this work, the EC time has been reduced to 5 min with an acceptable current density while previous investigators had operated between 15 and 60 min (Mohammadi et al., 2017; Zheng et al., 2017; Comninellis and Chen, 2010).

At higher values of pH, due to the consumption of OH^- by the formed Al complex Al(OH)⁴ at this pH, the capacity of coagulant would be diminished, which leads to formation of smaller flocs (Fig. 10).

One of the novel approaches, addressed in this work, was the evaluation of EC performance by the sludge thickness (sum of settled and floated sludges). The well-operated EC produces large flocs in which, due to the higher density and porosity of flocs, the thickness of sludge increases and vice versa. This assertion can be confirmed in Figs. 8-10. In contrast to Fig. 9 in which the thicknesses of the settled and floated sludges were 45 and 10 mm respectively, in Figs. 8 and 10, not only the thickness of the sludge was lower (<20 mm), but also there was no formed sludge on the surface that indicates the impact of initial pH on the performance of EC process.

3.5. Effect of electrolytic conductivity

In order to examine the effect of chloride ion concentration on electrolysis voltage, different concentrations of sodium chloride (NaCl) based on typical concentrations of various resources were considered (Table 5):

- 1. drinking water (DW): 0.34 mS/cm
- 2. wastewater (WW): 1, 2 and 10 mS/cm
- 3. sea water (SW): 30 mS/cm
- 4. oil field-produced water: (OPW): 150 mS/cm

Table 5 and Fig. 11 show the effect of NaCl concentrations on pseudo-stationary electrolysis voltage. As NaCl dosage increases from 0 to 0.86 g/L, the cell voltage decreases rapidly from 9.47 to 3.60 V. However, an additional increase in salt did not improve the electrolysis voltage anymore. Thus, the optimum conductivity value was 2 mS/cm. This value has also been reported by Fekete et al. (Martínez et al., 2000). The decrease of electrolysis potential difference can be related to the ohmic potential drop of the solution and/or to a decrease of the anode overpotential (Mouedhen et al., 2008). The same effect of NaCl dosage on the electrolysis voltage reported with (Mouedhen et al., 2008), but for different test conditions. Huang, et al. (Bayar et al., 2011) have also investigated on the effect of chloride ions on EC to treat industrial wastewater containing Cu and Ni. In this study, the voltages 9.3, 5.2, and 3.8 have been obtained with a concentration of NaCl 1,1, and 2 (g/l) respectively. The comparison of these results shows a difference with the current study which can be explained due to higher CD (6 A/cm^2) and a concentration of metals (e.g. Cu, Ni).

3.6. Effect of coagulant dosage, electrode and energy consumption

As discussed in sections 3.1 and 3.4, the aluminium concentration and pH define possible chemical species that are present in aqueous solution. The release of metal ions in electrocoagulation follows the Faraday's law. It means that in EC, the higher current that is used and



Turbidity: 34.7 NTU, Turbidity: 37.4 NTU, Turbidity: 33.6 NTU, Settling time: 5 min, Settling time: 5 min, Settling time: 5 min,



Turbidity: 24.1 NTU, Turbidity: 25.6 NTU, Turbidity: 22.7 NTU, Settling time: 15 min, Settling time: 15 min, Settling time: 15 min,

Fig. 8. The effect of initial pH 4, 5, and 6 on removal of turbidity. a: after 5 min settling time, b: after 15 min settling time, (tests conditions: Initial turbidity: 40 NTU, EC time: 5 min, CD: 0.0028 A/cm², conductivity: 2 mS/cm, stirring: 500 rpm).



Fig. 9. The effect of initial pH 7, 8.4, and 9.5 on removal of turbidity. a: after 5 min settling time, b: after 15 min settling time, (tests conditions: Initial turbidity: 40 NTU, EC time: 5 min, CD: 0.0028 A/cm², conductivity: 2 mS/cm, stirring: 500 rpm).

h



Settling time: 5 min

Turbidity: 14.3 NTU Settling time: 5 min



Turbidity: 8 NTU Settling time: 15 min

Turbidity: 8.3 NTU Settling time: 15 min

Fig. 10. The effect of initial pH 10.5 and 12 on removal of turbidity. a: after 5 min of settling time, b: after 15 min of settling time (tests conditions: Initial turbidity: 40 NTU, EC time: 5 min, CD: 0.0028 A/cm², conductivity: 2 mS/cm, stirring: 500 rpm).

Table 5

The electrical conductivity versus initial voltage and removal efficiency (test conditions: Initial turbidity: 40 NTU, EC time: 5 min, CD: 0.0028 A/cm², initial pH: 7.3, stirring: 500 rpm).

Conductivity (mS/cm)	Water resources	Added NaCl (gr/L)	Cl ⁻ (g/ L)	Voltage (avg) (V)
0.34	DW	0	0	9.47
1	WW	0.33	0.21	5.12
2	WW	0.86	0.55	3.60
10	WW	5	3.2	2.27
30	SW	15.4	9.8	1.89
150	OPW	77.8	50	1.64

NTU: nephelometric turbidity units, CD: current density, Cl⁻: chloride.

the longer the operating time, more active coagulant would be expected. Using the Faraday's law, the metal ions, which are produced in the electrochemical process, can be calculated from the following equation (Burns et al., 1997):



Fig. 11. The effect of conductivity on initial voltage (test conditions: Initial turbidity: 40 NTU, EC time: 5 min, CD: 0.0028 A/cm², initial pH: 7.3, stirring: 500 rpm).

$$n = \frac{it}{zF} \tag{7}$$

Where i is the current (A), t is the electrolysis time (s), F is the Faraday constant (F = 96500 C/mol), and z is the charge of the cation (z= +3 for Al or 2+ for Fe) (Panizza and Cerisola, 2004; Buso et al., 2000; Cruz et al., 2019).

In this work, the aluminium concentration was measured before and after each test to determine the mass loss during each EC test. It should be noted that the measured values were lower than the calculated ones with the Faraday's law (Table 6). This has also been reported in several previous studies (Naje et al., 2017; Vepsäläinen et al., 2012; Khaled et al., 2019; Comninellis and Chen, 2010; Lima 2019). This is because the Faraday's law assumes that all supplied current would be passed through the cell and does not consider other factors that may affect metal dissolution. For instance, the actual amount of current that passes through the cell is affected by the electrode configuration and geometry (Lima 2019). The experimental mass can then be determined by the following equation:

$$m_{exp} = m_2 - m_1 \tag{8}$$

Where m_1 is the metal mass before the experiment and m_2 is after experiment. The experimental results are summarized in Table 6.

In ECF process, it is well-established that energy consumption is relatively low. (Huang et al., 2020). The cost of electrical energy (kW h/m⁻³) is calculated from (Khaled et al., 2019):

$$C = Uc^* I^* \frac{i}{V} \tag{9}$$

Where U is the voltage (V), I is the current (A), t is the time of electrolysis (h) and v is the volume (m^3) of solution. The consumption of energy in Table 7, has been calculated based on Faraday's law (eq. (7)) as follows:

 $n = \frac{it}{2F} \rightarrow it = nzF$, (1C (Electric charge) = 1A*1s), Example: calculation of energy consumption of the current 1 A: $it = \left(\frac{0.129}{27}\right)^*3^*96500 = 1383C$ charge is needed in 300 s. Average voltage= $\frac{.2.4+2.2}{2} = 2.3V$

The dissolution of 0.129 g Al requires 1383C * 2.3 V = 0.003 MJ = 0.0008 kWh electric energy for 1260 ml solution volume and 0.6 kWh/ $\rm m^3.$

3.7. Effect of current density and operating time on initial turbidity concentration

The coagulant dosage is a function of turbidity concentration. The higher that the turbidity concentration is, the larger current density and/or operating time should be expected to produce more coagulants. Nonetheless, the operation of the EC system for a long period without maintenance, the current density is suggested to be between 20 and 25 $A \cdot m^{-2}$ (Mameri et al., 1998). Large current density also increases voltage and ohmic drop between anode and cathode electrodes; which results from the ohmic resistance of the electrolyte R which can be expressed as follows:

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Table 7

Turbidity removal efficiency based on the initial concentration (tests conditions: initial pH: 7.3, stirring: 500 rpm, and conductivity: 2 mS/cm).

Initial turbidity concentration (NTU)	Current density (A/cm ²)	Operating time (min)	Final turbidity (after 5 min settling time)	Final turbidity (after 15 min settling time)
80	0.0056	6	9.7 (88 %)	1.8 (98 %)
80	0.0028	11	17.4 (78 %)	-
80	0.0028	6	22.3 (72 %)	-
40	0.0028	8	8.5 (79 %)	-
40	0.0028	5	4.5 (89 %)	4.5 (89 %)
40	0.0028	4	5.7 (86 %)	4 (90 %)
20	0.0028	6	8.9 (55 %)	5 (75 %)
20	0.0014	6		3.9 (80 %)
20	0.0014	11	9.8 (51 %)	4.5 (77 %)
20	0.0056	6	Restabilized	
20	0.0021	6	8.7 (56 %)	3.4 (83 %)
10	0.0014	6	3.74 (63 %)	2.9 (71 %)
10	0.0007	6	5.25 (48 %)	3.7 (63 %)
10	0.0014	11	6.36 (36 %)	2.6 (74 %)
10	0.0021	6	4.7 (53 %)	4.6 (54 %)
10	0.0028	3	8.6 (14 %)	6.3 (37 %)

$$R = \frac{d}{S} \frac{1}{k} \tag{10}$$

Where d denotes the inter-electrode distance, S is electrode surface area and k the water conductivity (Lin and Chen, 1997). As mentioned in section 2.3, the colloidal particles were used to produce a turbidity solution with a concentration of 10, 20, 40, and 80 NTU. According to these concentrations, a range of current densities and operating time were applied as presented in Table 7.

One of the important results which can be drawn from Figs. 9, 12, and 13, is that the settling or floating of the formed flocks would depend on the initial turbidity concentration. In Fig. 12, with an initial turbidity concentration of 80 NTU, after 5 and 15 min of settling time, the formed flocs were settled and there were not observed any floated flocs. At a lower concentration of 40 NTU (Fig. 9) though, settling and floating were observed. By further decrease of concentration from 40 to 20 and 10 NTU, the formed flocs were floated completely (Fig. 13). This could be explained by the fact that the higher the initial turbidity concentration, the more particles would be attracted by the coagulants, which would lead to higher density. It should be also noted that getting knowledge of the flocs behaviour and sludge thickness is important from the point of view of EC performance and designing an appropriate downstream process for removal of sludge (e.g. skimming).

In Fig. 12, as the initial concentration increased from 40 to 80 NTU, the removal efficiency increased when the current density was doubled from 0.0028 to 0.0056 A/cm² for a given operating time. Nonetheless as mentioned earlier, this operating condition is suitable for a short operating time. This is because in long operation, the passivation phenomena will increase the voltage that results in more energy consumption. For this reason, under such circumstances, longer operating time would be recommended.

The same results were also observed in Fig. 13, when the initial concentration decreased to 20 NTU. Nevertheless, it is worth noting that

Table 6

Theoretically calculated and experimentally measured released metal, electrode and energy consumption. (z: charge of cation, F: Faraday's constant, n: amount of dissolved metal, m1, m2: metal mass before and after the tests).

Current (A)	Time (S)	z	F (C/ mol)	n calculated (mg)	n measured (mg)	m ₁ (mg)	m ₂ (mg)	Averagevoltage (V)	Electrode consumption (g/m ³)	Energy consumption (kwh/m ³)
1	300	3	96,500	185	129	<1	129	2.3	130	0.6
2	300	3	96,500	373	261	<1	261	2.19	261	0.6
2.5	300	3	96,500	466	320	<1	320	3.56	320	2.3
3	300	3	96,500	558	390	<1	390	2.01	390	1.5
6	300	3	96,500	1127	780	<1	780	8.34	780	14.9

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CD: 0.0056 Operating time: 6 min, Settling time: 5 min,



CD· 0 0028 Settling time: 5 min,



CD-0 0028 Operating time: 11 min, Operating time: 6 min, Settling time: 5 min.



CD: 0.0028 Settling time: 15 min, Settling time: 15 min,



CD: 0.0028 Operating time: 6 min, Operating time: 11 min, Operating time: 6 min, Settling time: 15 min.

Fig. 12. The effect of current density and operating time on removal of turbidity. a: after 5 min settling time, b: after 15 min settling time (tests conditions: Initial turbidity: 80 NTU, initial pH: 7.3, conductivity: 2 mS/cm, stirring: 500 rpm).



CD: 0.0014 CD: 0.0014 CD: 0.0056 Operating time: 6 min, Operating time: 6 min, Operating time: 11 min, Operating time: 6 min, Settling time: 15 min, Settling time: 15 min, Settling time: 15 min, Settling time: 15 min,

CD: 0.0021 Operating time: 6 min, Settling time: 15 min,

Fig. 13. The effect of current density and operating time on removal of turbidity after 15 min settling time, (tests conditions: Initial turbidity: 20 NTU, initial pH: 7.3, conductivity: 2 mS/cm, stirring: 500 rpm).

the particles were restabilized when the current density increased to 0.0056 A/cm^2 . In this concentration, the higher removal efficiency was observed for a given current density of 0.0021 A/cm² and operating time of 6 min.

3.8. Effect of stirring speed

To study the effect of stirring speed, EC was performed at different stirring speeds of 0, 100, 250, 500 and 800 rpm. The results are presented in Table 8.

As seen in Table 8, the removal of turbidity was much faster for a moderate agitation speed. The turbidity removal was low without agitation; i.e. the percentage of removal did not exceed 55 % for an electrolysis time of 5 min and settling time of 5 min and 56 % for a settling time of 15 min. For a moderate agitation of 250 rpm, the elimination was much faster than 100 rpm and even further increased when the agitation was up to 500 rpm, reaching a removal percentage of 89 % after 5 and 15 min of settling time, respectively. This proves that

Table 8

The effect of stirring speed on removal efficiency (tests conditions: initial concentration: 40 NTU, current density: 0.0028 (A/cm²), EC time: 5 min, initial pH: 7.3, conductivity: 2 mS/cm).

Stirring speed (rpm)	Initial turbidity concentration (NTU)	Final turbidity (after 5 min settling time)	Final turbidity (after 15 min settling time)
0	40	18 (55 %)	17.4 (56 %)
100	40	13.3 (67 %)	10.4 (74 %)
250	40	8 (80 %)	7 (82 %)
500	40	4.5 (89 %)	4.5 (89 %)
800	40	10.7 (73 %)	4.6 (88 %)

the removal of turbidity by the coagulant aluminium hydroxide Al(OH)3 occurred very effectively for stirring speeds of 250 and 500 rpm. However, there was a slight decrease in removal efficiency after 5 min of settling time when the agitation reached 800 rpm. This could be explained by the fact that excessive agitation would result in breakage of the flocs. Similar results have also been reported by Khaled et al. (Khaled et al., 2019). Nonetheless, contradictory results have been reported by Bayar et al. (Mandal, 2014) in which the effect of stirring speed and current density on removal efficiency of wastewater for a poultry slaughterhouse by electrocoagulation has been investigated. They used the stirring speeds of 100, 150, and 250 rpm and reported the removal efficiency of 85 %, 90 %, and 75 % respectively.

4. Conclusions

Electrocoagulation-flotation was investigated as a preliminary treatment process for turbidity removal in which, the produced coagulants (Al(OH)₃) by EC, aggregated the nanoparticles in form of flocs for in-situ removal. The impact of operating parameters of electrocoagulation-flotation using aluminium electrodes (inter-electrode distance 5 mm) such as current density, initial pH, electrolytic conductivity, coagulant dosage, operating time, initial concentration, stirring speed, and passivation phenomenon were investigated. The following conclusions can be drawn from the experimental results:

• The maximum turbidity removal has been around 90 % (4.5 NTU). The test conditions included an initial concentration of 40 NTU, a current density of 0.0028 A/cm², the initial pH of 7.3, the interelectrode distance of 5 mm, stirring speed of 500 rpm, the electrolytic conductivity of 2 mS/cm, and for a given time of electrocoagulation of 5 min.

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- After filtration with a paper filter of 20 μ m, the removal efficiency was 98 % (0.8 NTU).
- The settling or floating of the formed flocks depended on the initial turbidity concentration.
- The thickness of the formed sludge (sum of settled and floated), indicates the performance of the EC process.
- The energy consumption in EC can be decreased with adding NaCl. As NaCl dosage increased from 0 to 0.86 g/L, the cell voltage decreased rapidly from 9.47 to 3.6 V. The optimum conductivity value was 2 mS/cm indicating that EC would serve as an appropriate process in wastewater treatment.
- The required settling time of 5 and 15 min have also been evaluated, in which 5 min was sufficient when the initial concentration was 40 NTU, and 15 min when the initial concentration was 80 NTU. These results can then be used for optimized design of the settling tank.

Declaration of Competing Interest

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

Data availability

Data will be made available on request.

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