

Treatment of wastewaters from food Aromsa and ingredients production by electrocoagulation (EC) treatment aided by mucilage of *Opuntia ficus-indica*

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Abstract. The electrocoagulation (EC) treatment process was aided by the cladode mucilage of *Opuntia ficus-indica* (OFI) to improve chemical oxygen demand (COD) removal efficiency (%) of wastewater from a plant producing flavoring and food additives. The studied variables included initial pH, current density, types of electrodes (Fe and Al), mucilage concentration, and charge loading. The EC treatment using Al electrodes, assisted by an optimum concentration of 200 mg L⁻¹ OFI mucilage, a current density of 60 A/m², and an initial pH equal to 3, improved the COD removal efficiency (69.77%) at an EC treatment time of 40 min. The EC operating costs, energy, and electrode consumptions values were equal to 3.809 US \$/m³, 0.810 kWh/kg COD removed, and 0.7898 kg/m³, respectively. However, no improvement was noted when Fe electrodes were used with or without adding OFI mucilage (under conditions of pH 6, current density of 50 A/m², and EC time of 120 min), and COD removal efficiency (%) was equal to 41.5%. The operational costs, energy and electrode consumptions were equal to 4.2201 US \$/m³, 2.8 kWh/kg COD removal and 3.0964 kg/m³, respectively. The OFI mucilage could be used as a promising bio-coagulant with EC treatment using Al electrodes, to treat the plant producing flavoring and food additives wastewater.

Keywords: *Electrocoagulation process, Food production, COD, Opuntia ficus indica, mucilage.*

Introduction

The rapid urbanization and industrialization generate a large amount of polluted water every year, leading to the problem of wastewater improper disposal into the environment. In this respect, natural resources have become interesting alternatives for conventional wastewater treatment processes that can generate a secondary pollution, once they are a sustainable option, offering economic and financial benefits.

Thus, green technology based on the exploitation of inexpensive adsorbents made from industrial by-products, farming wastes, or natural resources are currently being developed (Zhang *et al.*, 2006; Abid *et al.*, 2009; Yin, 2010; Asrafuzzaman *et al.*, 2011; Torres *et al.*, 2012; Adjeroud-Abdellatif *et al.*, 2020).

Some studies are developed about the use of OFI mucilage as an eco-friendly adsorbent to eliminate heavy metals as copper, arsenic, and dyes from water (Abid *et al.*, 2009; Yin, 2010; Fox, 2011; Adjeroud *et al.*, 2015; Vecino *et al.*, 2016; Adjeroud *et al.*, 2018) and also for turbidity removal in association with electrocoagulation-electroflotation (EC-EF) process (Adjeroud *et al.*, 2015; Adjeroud *et al.*, 2018; Djerroud *et al.*, 2018; Adjeroud-Abdellatif *et al.*, 2020).

The mucilage, as a complex carbohydrate polymer, is made of a long polymeric chain of up to 30,000 monosaccharides (Amin *et al.*, 1970; McGarvie and Parolis, 1981). The polymers that make up mucilage have a high molecular weight of 15.3 to 15.7 × 10⁶ g mol⁻¹ and an extremely ramified structure comprising residues of xylose, rhamnose, galactose, arabinose, and galacturonic acid (Amin *et al.*, 1970; McGarvie and Parolis, 1979; Felkai-Haddache *et al.*, 2015). Some studies showed the potential of this carbohydrate as a source of industrial hydrocolloids, as an emulsifier, as a thickener for food, as an adhesive for lime, as a water purifier, and as a foodstuff (Sáenz *et al.*, 2004; Wiese *et al.*, 2004; Alila and Boufi, 2009; Adjeroud *et al.*, 2015; Adjeroud *et al.*, 2018).

The most usual method for extracting mucilage from OFI cladode is the maceration under heating or at room temperature of the crushed cladodes in distilled water (Sepúlveda *et al.*, 2007; Adjeroud-Abdellatif *et al.*, 2020). However, this is an inefficient extraction method, which, in addition, can be expensive. Felkai-Haddache *et al.* (2015) found that microwave-assisted extraction (MAE) was efficient in comparison to the conventional method for extracting mucilage from OFI cladodes, allowing a reduction of extraction time and rising extraction quality by saving mucilage physicochemical and rheological qualities. The MAE is an alternative process for extracting substances from materials, because it allows saving solvent amounts, extraction time, and energy feeding (Dahmoune *et al.*, 2015; Felkai-Haddache *et al.*, 2015).

The EC treatment has several advantages, including the simplicity of the equipment required, low operating costs, reduced treatment time, low sludge production and most importantly the decreasing of the amount of chemical coagulants used in conventional methods (Pouet and Grasmick, 1995; Chen *et al.*, 2000; Attour *et al.*, 2014). Some studies reported the effectiveness of this method to treat various wastewaters discharges (Pouet and Grasmick, 1995; Mollah *et al.*, 2001; Pichler *et al.*, 2012; Attour *et al.*, 2014; Omwene and Kobya, 2018).

The purposes of the study are to (i) investigate the impact of OFI mucilage, as a bio-coagulant aid in association with EC aluminum and iron electrodes, (ii) to scrutinize the effect of EC operational parameters (pH and current density) on COD removal efficiency, (iii) and to analyze the obtained treatment sludge at optimum conditions. In addition, the operating cost, charge loading, energy and electrodes consumption were calculated at different process conditions.

Material and Methods

Wastewater characteristics

The wastewater was recovered from Aromsa facility producing flavoring and food additives (Gebze-Kocaeli, Turkey) in June 2017. The initial values of the wastewater pH, COD and conductivity were 12.15, 6000 mg L⁻¹, and 9.40 mS cm⁻¹, respectively. The obtained wastewater was saved at 4 °C until being used.

Mucilage extraction

The OFI cladodes were picked from Aokas region (Bejaia) in January 2017, at the East North of Algeria. The average weight of the cladodes was 985 ± 18 g, and the physical dimensions were 408, 170, and 15 mm, for length, width, and thickness, respectively. The cladode cuticle was peeled after cleaning and washing the cladodes with distilled water. Then mucilage was extracted using microwave-assisted extraction (MAE) according to the protocol of Felkai-Haddache *et al.* (2016). A volume of crushed cladode was diluted in distilled water with a mass to liquid ratio of 1:1. The mixture solution was irradiated for 3.5 min in a domestic microwave (2.45 GHz, Samsung Model NN-S674MF, Kuala Lumpur, Malaysia) at 700 W. The obtained filtrate was precipitated in one volume of distilled water with 3 volume of ethanol 95% at ratio of 1:3. The mucilage was recovered and rinsed three times with ethanol (75%, v/v). After freezing at -20 °C, the mucilage was conserved as a powder after using a lyophilizer (ALPHA 1-2 LD plus, Germany) at -55 °C during 12 h. For EC experiments assisted with OFI mucilage, the mucilage was dissolved by gentle mixing (10-15 min) in 1 M NaOH, prior to each run.

Electrocoagulation device

The samples of Aromsa wastewaters were treated in a Plexiglas reactor of 1 L ($12 \times 9.5 \times 8.5$ cm dimension). Two electrode materials, aluminum and iron were used, with size of $5.0 \times 7.5 \times 0.3$ cm. In the reactor, a range of four electrodes was arranged vertically and separated by a distance of 13 mm, with a total operative electrode area of 225 cm^2 (Figure 1). A continuous stirring (200 rpm) within the reactor was kept for each experiment. The electrodes were powered by a digital dc source (Model 6675A of Agilent, 120 V, 18 A) and operated with a galvanostatic system. During EC treatment, wastewater samples were collected at fixed intervals, filtered through a microscopic membrane ($0.45 \mu\text{m}$) and then analyzed. Prior to each experiment, electrodes were rinsed by HCl solution (35%), aqueous hexamethylenetetramine solution (2.8%) and dried to reduce electrode passivation (Can *et al.*, 2003).

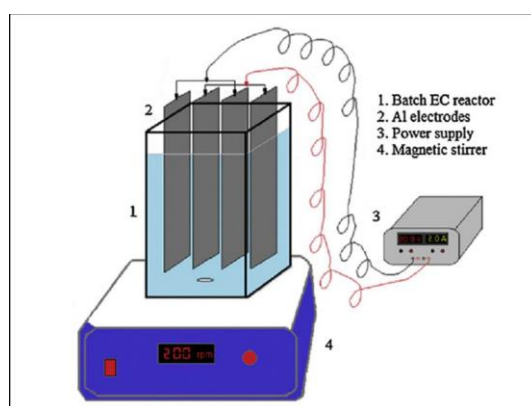


Figure 1. The experimental EC batch process.

Analytical procedure

The evolution of COD removal efficiency (%) was monitored with an analysis was performed on the samples as defined in the standard methods (Apha, 2005). The COD measurements were taken with a COD reactor, which is measured according to standard methods (Apha, 2005) on the basis of organic compounds oxidization to carbon dioxide with a strong oxidizing agent under acidic conditions. The results were directly read on a spectrophotometer (DR/2000, Hach, USA).

The initial pH of the producing flavoring and food additives wastewater was measured by pH meter (Mettler Toledo, S220 Seven Compact). The conductivity values were determined using a conductivity meter (Mettler Toledo, Seven Go Pro). The desired pH was obtained by adding H_2SO_4

(0.50 N) or NaOH (0.50 N) solutions to the effluent. The initial pH was set at 3, 4, 6, and 8, and after each run, the pH was measured. The tested current densities were 20, 30, 50, and 60 A m⁻², and the EC time was set at 120 min. All trials were repeated two times, and the experimental error was calculated less than 2%. After each experiment, the produced sludge was recovered and characterized by X-ray diffraction (XRD; Rigaku 2000 D/max with CuK α -radiation, $\lambda = 0.154$ nm at 40 kV and 40 mA) and Fourier transform infrared spectroscopy (FTIR, Bio Rad FTS 175C spectrophotometer).

Calculation for the EC process

The charge loading (q (C)), electrode and electrical energy consumptions ($C_{\text{electrode}}$, kg /m³ and C_{energy} kWh m⁻³) were calculated using Equations (1-3) as follows:

$$q \text{ (C)} = i \times t_{EC} \quad (1)$$

$$C_{\text{energy}} \text{ (kWh m}^{-3}\text{)} = \frac{i \times t_{EC} \times U}{V} \quad (2)$$

$$C_{\text{electrode}} \text{ (kg m}^{-3}\text{)} = \frac{i \times t_{EC} \times M_w}{z \times F \times V} \quad (3)$$

where i : applied current(A), t_{ec} : EC time (hour and second for the energy and electrode consumption, respectively), V : volume of wastewater treated (m³), U : cell voltage (V), M_w : molecular mass of Al and Fe electrodes ($M_{w,Fe} = 55.85$ g/mol and $M_{w,Al} = 26.98$ g mol⁻¹), Z : number of Al and Fe electrons transferred ($z_{Fe} = 2$ and $z_{Al} = 3$), and F : Faraday's constant (96,487 C mol⁻¹).

The operating costs (OC \$ m⁻³) in the EC treatment are principally the electrical energy and electrodes costs. The OC was calculated through Eq. (4):

$$\text{OC (\$ m}^{-3}\text{)} = \alpha \times C_{\text{energy}} + \beta \times C_{\text{electrode}} \quad (4)$$

where C_{energy} : energy consumption (kWh m⁻³), $C_{\text{electrode}}$: electrode consumption (kg m⁻³). α and β are the electrical energy cost and the electrode unit, respectively. According to the Turkish Market, the price of the electrical energy (α) in august 2017 is 0.120 \$ kWh⁻¹ and the electrodes price (β) is equal to 0.952 \$ kg⁻¹ and 4.10 \$ kg⁻¹ for Fe and Al electrodes, respectively.

Results and discussion

Effect of potential hydrogen on EC treatment performance

The initial pH is a significant parameter that strongly impact the EC performance (Chen *et al.*, 2000; Shalaby *et al.*, 2014; Kobya and Demirbas, 2015), because conductivity, anodic dissolution and zeta potential are affected by this variable (Belkacem *et al.*, 2008; Moussa *et al.*, 2016). However, during EC treatment, the final solution pH changed depending on the initial pH, and it is not evident to determine clearly the relation (Mansoorian *et al.*, 2014; Gatsios *et al.*, 2015).

The impact of initial pH on the COD removal efficiency (%) of food flavoring and additives wastewater, was tested using two different electrode materials, Al, and Fe. The trials were monitored by adjusting the effluent pH to 3, 4, 6 and 8 using an acid solution or sodium hydroxide, and the remaining experimental conditions were determined as initial COD value of 6400 mg L⁻¹, current density of 30 A m⁻², and EC time of 120 min. The figure 2 illustrates the effect of initial pH on COD removal efficiency (%) using aluminum (Figure 2a), and iron (Figure 2b) electrodes. The COD removal efficiency (%) changed according to the solution initial pH. The necessary time to remove COD was set at 120 min corresponding to 4579 C of charge loading (Eq. (1)). The COD

was removed to 4112 mg L⁻¹ (35.47%) at initial pH 3 and 4096 mg L⁻¹ (36.16%) for iron and aluminum electrodes, respectively. The COD was removed to 5004 mg L⁻¹ (21.47%) at initial pH of 4 and 4576 mg L⁻¹ (28.63%) for Fe and Al electrodes, respectively. At initial pH of 6, the COD was removed to 3936 mg L⁻¹ (38.23%) and 4240 mg L⁻¹ (33.90%) for Fe and Al electrodes, respectively. In the initial pH of 8, the COD was removed to 3620 mg L⁻¹ (43.19%) and 4320 mg L⁻¹ (32.64%) for both electrodes Fe and Al, respectively. In each initial tested pH 3, 4, 6 and 8, after each treatment (t_{EC}= 120 min), final pH values were equal to 9.47, 9.74, 9.32 and 9.76 for Al electrodes, respectively, whereas with Fe electrodes final pH values were equal to 5.67, 5.94, 11.2 and 11.52, respectively.

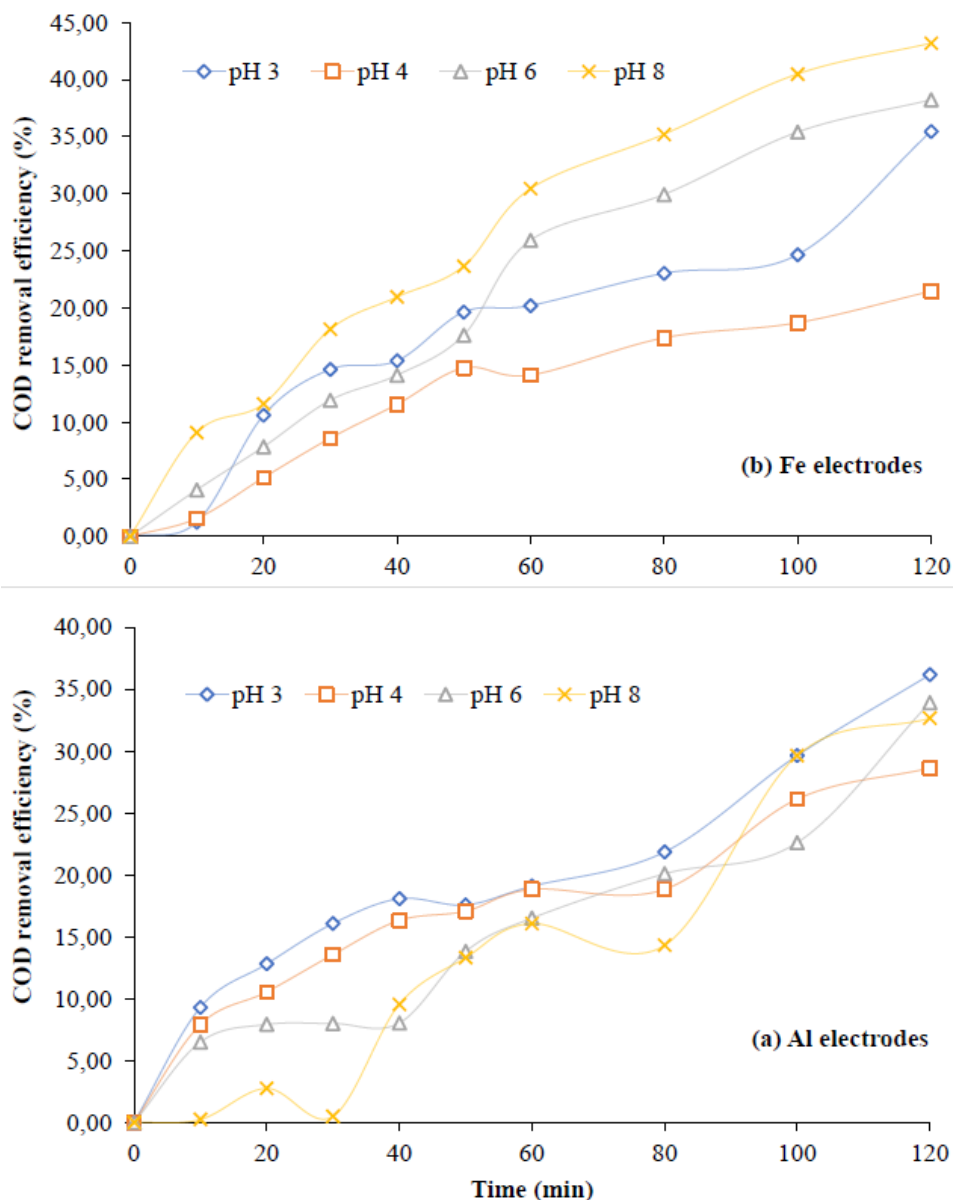


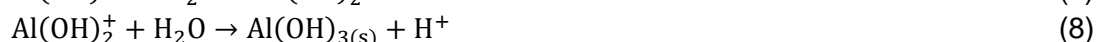
Figure 2. The effect of pH on COD removal efficiency (%) in the EC treatment using (a) Al electrodes and (b) Fe electrodes, at fixed parameters $j = 30 \text{ A/m}^2$, $t = 120 \text{ min}$, $v = 850 \text{ mL}$.

The COD removal efficiency (%) in EC treatment was influenced by initial pH and type of electrodes (Fe and Al). The final pH rises with EC time for both electrodes was also observed. The electrodes dissolution increases with the decrease of the pH solution for EC treatment with Al and Fe electrodes. In acidic aqueous media, the trivalent ion Al³⁺ is in the hydrated form Al(H₂O)₆³⁺ (simply

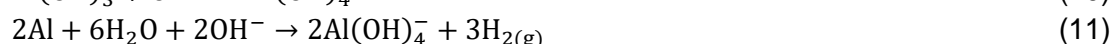
written as Al^{3+}). The Al^{3+} ions generated from the anodes interact with water, attracting OH^- ions to form cationic, anionic, or neutral complexes according to the following reaction:



The hydroxyl ions are released from water electrolysis or from the addition of a basic solution to the effluent. Considering only the mononuclear species, Al^{3+} reactions are as follow:



The cathodes and anodes generate ions of OH^- and Al^{3+} respectively for EC treatment with Al electrode, and subsequently form various aluminum species. In the $pH < 3.5$, the predominant species in solution are the free Al^{3+} form of aluminum and beyond this pH, the dominants species are the aluminum hydroxyl forms, monomeric and polymeric ones $Al_x(OH)_y(H_2O)^{(3xn-y)+}$ (such as $Al(OH)^{2+}$, $Al(OH)_2^+$, $Al_2(OH)_2^{4+}$, $Al_6(OH)_{15}^{3+}$, $Al_7(OH)_{17}^{4+}$, $Al_8(OH)_{20}^{4+}$, $Al_{13}(OH)_{34}^{5+}$). According to extensive precipitation reactions, aluminum hydroxide, $Al(OH)_3(s)$, eventually forms as an insoluble chemical with an amorphous structure (Rebhun and Lurie, 1993; Can *et al.*, 2003). In the pH ranging from 4 to 7, cationic complexes of $(Al(OH))$ formed, such as $Al_8(OH)_{20}^{4+}$, these are efficient flocculants because these forms scavenge effectively the impurities through their adsorption mechanism, producing charge neutralization and entrapment into precipitates (Rebhun and Lurie, 1993). However at $pH > 9.5$, the soluble anionic ions $Al(OH)_4^-$ are the dominant affecting directly the contaminants present in the effluent (Georgantas and Grigoropoulou, 2007). Also, when the pH increase, the cathode can be altered by OH^- (hydroxyl ions) species produced during $H_2(g)$ evolution (Picard *et al.*, 2000; Mouedhen *et al.*, 2008).

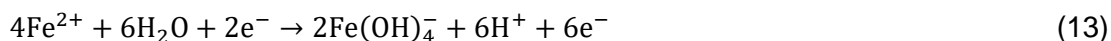


In the EC treatment with Fe electrodes has been shown that at pH values between 5 and 8, there is an increase in abatement rate because at neutral pH, the reduced reaction at the cathode generate the hydrogen bubbles that are thinner, smaller and facilitate the combination of tiny weakened colloidal particles (Kabdaşlı *et al.*, 2012). The EC treatment at $pH < 4$ can be likened to an accelerated corrosion process. In fact, at an acidic pH and in an oxygenated medium, Fe^{2+} ions oxidize to Fe^{3+} ions, according to the reaction:



Subsequently, numerous complexes ($(Fe(OH)_2, Fe(OH)_2^+, Fe(OH)_3)$ rising from Fe^{3+} ions act as coagulants and participate in the adsorption and destabilization of the colloidal elements (Pykhteev *et al.*, 1999; Moreno-Casillas *et al.*, 2007). The EC treatment with Fe electrodes generates ferric ions, which according to the electrochemical reaction, may complexify and generate monomeric and polymeric iron hydroxyl complexes such as $Fe(OH)^{2+}$, $Fe(OH)_2^+$, $Fe_2(OH)_2^{4+}$, $Fe(H_2O)_5(OH)^{2+}$, $Fe(H_2O)_4(OH)_2^+$, $Fe_2(H_2O)_8(OH)_2^{4+}$, $Fe_2(H_2O)_6(OH)_4^{4+}$ depending on the pH of solution, the final obtained compound was $Fe(OH)_{3(s)}$. In the pH above 10, $Fe(OH)_4^-$ was the major Fe hydroxyl complexes within the solution (Pykhteev *et al.*, 1999). The ferrous and ferric ions

(Fe²⁺ and Fe³⁺) can be formed under acidic conditions (pH < 6.5) during EC treatment (Moussa *et al.*, 2016). In addition, Fe²⁺ and Fe³⁺ ions, produced by dissolution, have the ability to associate with hydroxyl ions and precipitate in the form of Fe(OH)_{2(s)} and Fe(OH)_{3(s)} species, that participate to remove the organic pollutants by co-precipitation and adsorption phenomena. The predominance of the anionic species Fe(OH)₄⁻ under alkaline conditions causes a drop in the pH of the water and a decrease in the performance of the process according to Eq. (13) (Aoudjehane *et al.*, 2010; Vepsäläinen, 2012):



In the pH > 6.5, the insoluble ferric hydroxyl complex Fe(OH)_{3(s)} was the prominent species formed. Thus, the optimal pH required for an effective EC process should be close to neutral (Pykhiteev *et al.*, 1999; Chen *et al.*, 2000; Shalaby *et al.*, 2014). However, Moussa *et al.* (2016) concluded that the optimal experimental pH for EC process with Fe electrodes was ranging from 5 < pH < 9 in order to promote the oxidation of ferrous ions.

The results showed that Al electrodes are less efficient than Fe electrodes on COD removal efficiency (%), which means that the Al and Fe ions interact differently with the particles of pollutants especially on COD removal. These results are similar to those suggested by Kobya *et al.* (2003) on the EC treatment of industrial textile wastewater using Al and Fe electrodes. The use of Al electrodes shows that the COD removal at initial pH less than 6 was of 98% and this value drops when initial pH > 6. However, when using Fe electrodes, the COD abatement was effective at initial pH < 7 with 98% rate and drops at initial pH > 10 into 87%. Kobya *et al.* (2009) studied the EC treatment with Fe electrodes to treat a textile wastewater and found that the COD abatement efficiency attained 78% under condition of pH 6.9 and current density of 10 mA cm⁻². Furthermore, for both electrode types, the electrodes consumptions during EC treatment according to Faraday's law (Eq. (3)) for initial pH 3, 4, 6 and 8 at treatment time of 120 min, were found equal to 0.8008 kg m⁻³ for theoretical (C_{electrodes, theo}) and 0.7680, 0.8933, 1.0683 and 1.0246 kg m⁻³ for experimental (C_{electrodes, exp}) values for Al electrodes, respectively; whereas, for the Fe electrodes the consumptions were of 1.6556 kg m⁻³ for the theoretical (C_{electrodes, theo}) and experimental (C_{electrodes, exp}) values were of 1.7366, 2.1953, 1.7005 and 1.5836 kg m⁻³, respectively. Therefore, current efficiency values (CE) at initial pH 3, 4, 6 and 8 (calculated according to Eq. (14)) were of 96, 111, 133 and 116%, respectively for Al electrodes, 105, 133, 103 and 96% for Fe electrodes.

The COD abatement shows same pattern for both Al and Fe materials. The Fe presents a high effectiveness at alkaline and neutral medium, especially between pH 6 and 8, while for Al materials, they are effective in acidic medium at initial pH 3. Although their synthesis is favored at pH varying from 3 to 7, the hydroxyl complexes of aluminum Al(OH)_{3(s)} and iron Fe(OH)_{3(s)} are insoluble in this range. Additionally, their production promotes the development of cationic hydroxide solutions (iron and aluminum oxide surfaces) made of oxide, which can adsorb the colloidal particles enhancing thereby the removal efficiency. The most particles are negatively charged the most are readily adsorbed by aluminum hydroxyl complexes (Pulkka *et al.*, 2014). Based on these conclusions, initial pH of further wastewater treatment was set at pH 6 and 3 for Fe and Al electrodes, respectively.

Effect of current density on EC process performance

The current density parameter is a significant parameter and the most important variable in electrocoagulation treatments, strongly affecting the yield of these proceedings (Can *et al.*, 2003; Attour *et al.*, 2014; Kobya and Demirbas, 2015; Attour *et al.*, 2016). The bubble formation rate

increases at a high current density, with small size that improve turbidity and pollutants elimination of treated wastewater (Merzouk *et al.*, 2009; Adamovic *et al.*, 2016).

The figure 3 illustrates current densities effects on COD removal efficiency (%) from the food flavoring and food additives wastewater using Fe and Al electrodes. The four current densities (20, 30, 50 and 60 A m⁻²) were investigated at constant conditions initial pH 3 and pH 6 for EC using Al and Fe electrodes, respectively and at set time $t_{EC} = 120$ min.

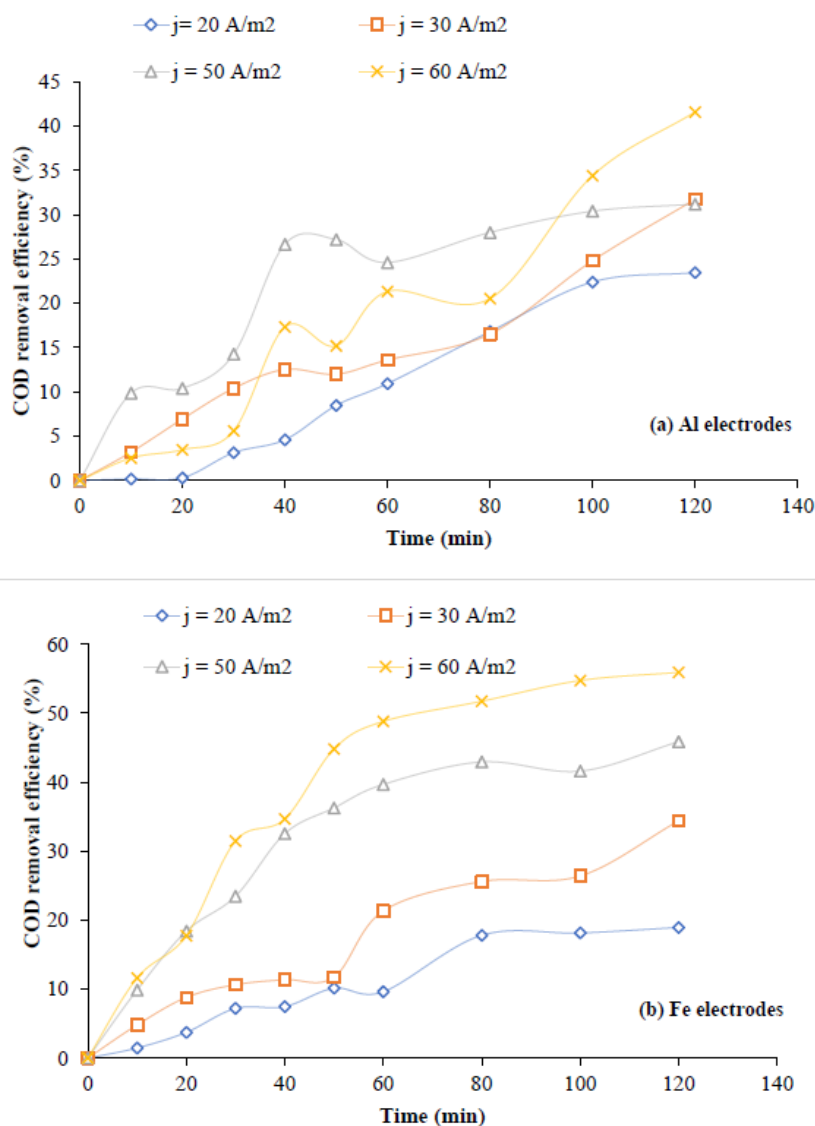


Figure 3. The effect of current density (j) on COD removal efficiency (%) at fixed conditions (a) Al electrodes at pH 3, $t_{EC} = 120$ min, $v = 850$ mL and (b) Fe electrodes at pH 6, $t = 120$ min, $v = 850$ mL.

In figure 3, COD removal efficiency (%) increases with a rise in current density for both aluminum and iron electrodes and when Al electrodes were used, the set treatment time of 120 min (corresponding to 9144 C of charge loading Eq (1)) was required to reduce effectively initial COD from 6000 to 4592 mg L⁻¹ (23.47% COD removal efficiency) at 20 A m⁻², then to 3504. In figure 3, COD removal efficiency (%) increases with a rise in current density for both aluminum and iron electrodes (41.60%) at 60 A m⁻²; whereas, for Fe electrodes, the corresponding removal values (in mg L⁻¹) were 4864 (18.93%), 3936 (34.4%), 3248 (45.87%), and 2645 (55.52%), for current density values of 20, 30, 50 and 60 A m⁻², respectively. The EC time required for EC treatment using Fe

electrodes was more effective (55.52%) in comparison to Al electrodes (41.60%) at 60 A m⁻². As current density and EC treatment time increase, the amounts of dissolved iron and aluminum increase too. According to these results, the dissolution of Al electrodes is less important than Fe electrodes.

The results are consistent with Faraday's law (Eq. (3)) at current density values of 20, 30, 50 and 60 A m⁻², and at set time of $t_{EC}=120$ min, the theoretical Al electrodes consumption ($C_{electrodes,theo}$) were calculated as 0.5339, 0.8008, 1.3347 and 1.5992 kg m⁻³, and experimental Al electrodes consumption ($C_{electrodes,exp}$) were calculated as 1.2878, 0.76800, 1.5751 and 2.0781 kg m⁻³, respectively. The Fe electrodes consumption for the theoretical ($C_{electrodes,theo}$) and experimental ($C_{electrodes,exp}$) values were 1.1044, 1.6566, 2.7610 and 3.3080 kg m⁻³ and 1.1922, 1.7005, 2.9147 and 3.3812 kg m⁻³, respectively. Theoretically, in EC treatment, 28 g of iron and 9 g of aluminum are dissolved at anodes whenever one Faraday of charge passes over the circuit. According to this, the mass of iron electrode dissolved is about twice the mass of aluminum electrode. According to this result, the theoretical electrodes consumption values were less than those of the experimental consumption values. The experimental value can rise or fall depending on the wastewater characteristics, operational conditions and chemical corrosion which caused holes that led practically on the electrode surface (Chen *et al.*, 2000; Mansouri *et al.*, 2011; Kobya and Demirbas, 2015).

Therefore, the current efficiency values (CE) at 20, 30, 50 and 60 A m⁻² (calculated according to Eq. (14)) were equal to 241, 96, 118 and 130% for Al electrodes, and 18, 102, 105 and 102% for Fe electrodes, respectively. Some EC studies reported that current efficiencies close to 100% were taken when iron electrodes were used (Zongo *et al.*, 2009; Chafi *et al.*, 2011) and in average of 100 to more than 300% for aluminum electrodes (Zongo *et al.*, 2009; Mansouri *et al.*, 2011; Omwene and Kobya, 2018).

$$CE(\%) = (C_{electrode,exp}/C_{electrode,theo}) \times 100 \quad (14)$$

Mollah *et al.* (2004) explain that energy consumption rises simultaneously with current density, which means the release of excessive oxygen and high generation. This is considered to be a negative effect for the materials (corrosion of the cathodes especially the Fe electrode) and for the electrical energy consumption since it rises the cost of the process. That's why, the current density should be limited to reduce the costs of the treatment, and the optimal current densities were fixed at 60 and 50 A m⁻² for Al and Fe electrodes, respectively.

Effect of *Opuntia ficus-indica* mucilage concentration on EC process performance

The orientation to the bio-coagulant/flocculants uses become an alternative for wastewaters treatment due to their coagulation and flocculation abilities (Miller *et al.*, 2008; Freitas *et al.*, 2018; Adjeroud-Abdellatif *et al.*, 2020). Under the optimal conditions for Al (a) electrode: $j=60$ A m⁻², pH 3 and $t_{EC}=120$ min, for Fe (b) electrode $j=50$ A m⁻², pH 6 and $t_{EC}=120$ min and initial. The figure 4 illustrates the progress of the effect of mucilage concentrations of 100, 150, 200 and 300 mg L⁻¹ on COD removal efficiency (%) in EC proceeding.

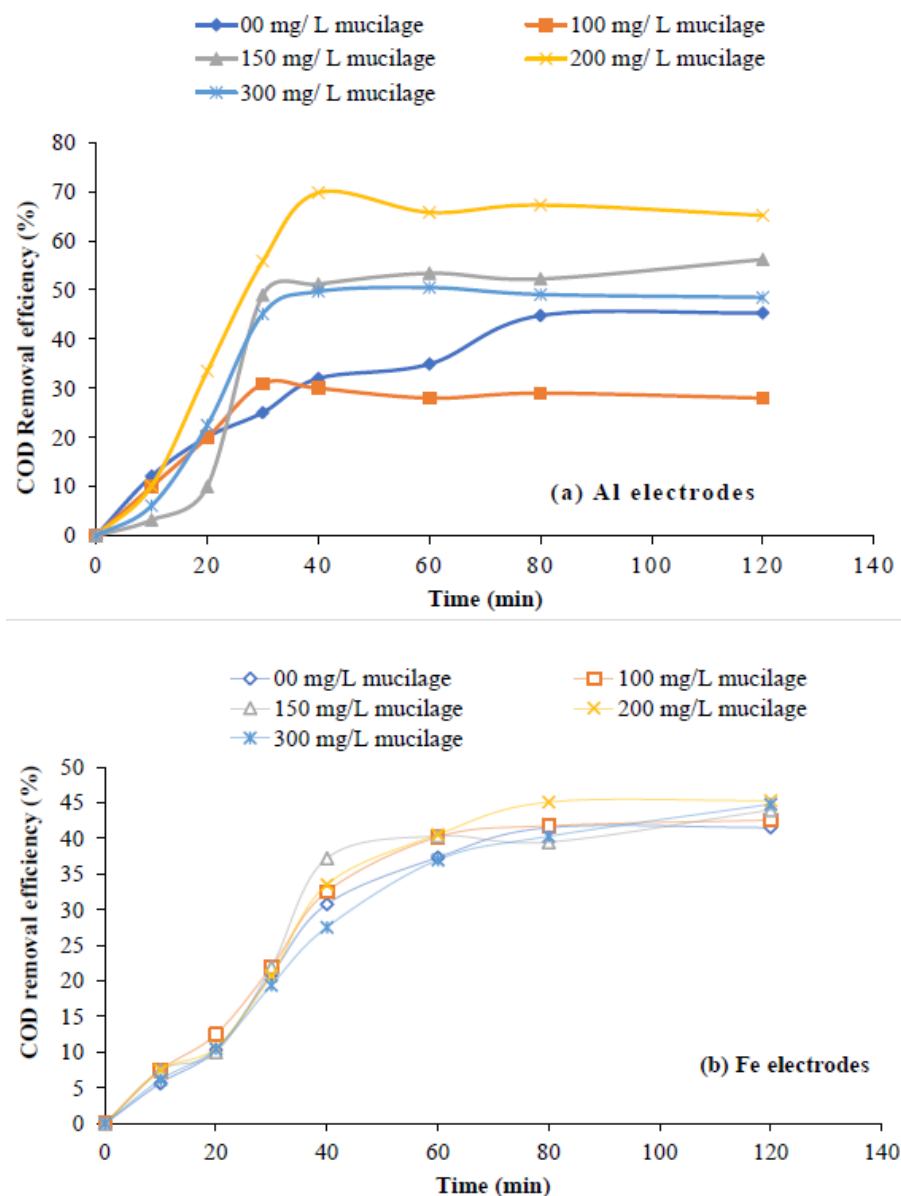


Figure 4. The effect of the mucilage concentrations on COD removal efficiency from the food Aromsa wastewater at constant conditions, (a) electrodes: initial pH 3, $j = 60 \text{ A m}^{-2}$, and $t_{EC} = 120 \text{ min}$ for Al electrodes, (b) initial pH 6, $j = 50 \text{ A m}^{-2}$, $t_{EC} = 120 \text{ min}$ for Fe electrodes.

For all tested mucilage concentrations, a progressive evolution of the COD removal abatement (%) according to time is observed in figure 4, which shows that COD removal efficiency (%) increases with the time for both electrodes. For Al electrode, the treatment stabilizes after 40 min corresponding to charge loading of 3084 C from Eq. (1), whereas for Fe electrodes the stabilization appears after 80 min of treatment, thus the required time was 120 min corresponding to 7632 C of charge loading Eq. (1).

Concerning the adding of mucilage concentrations at 00, 100, 150, 200 and 300 mg L^{-1} , the COD removal efficiency (%) after 40 min of treatment were equal to (in mg L^{-1}) 4303 (35.77%), 4690 (30%), 3270 (51.19%), 2025 (69.77%) and 3369 mg L^{-1} (49.71%), respectively when Al electrodes are used. For Fe electrodes, the corresponding COD removal efficiency (%) at EC time of 120 min were (in mg L^{-1}) 3625 (41.5%), 3849 (42.55%), 3755 (43.95%), 3666 (45.28%) and 3699 mg L^{-1} (44.79%), respectively.

The COD removal was enhanced when 200 mg L⁻¹ of mucilage was added using Al electrodes (69.77%), whereas no obvious improvement was noted for COD removal when varying mucilage concentrations using Fe electrodes. The OFI mucilage was able to enhance COD removal efficiency by more than 24.47% using Al electrodes at initial pH 3, current density of 60 A m⁻², and at 40 min of EC time. The results showed that Al electrodes have a more impressive performance compared with Fe electrodes towards mucilage addition.

The Al and Fe electrodes consumptions were calculated according to (Eq (3)) when 00, 100, 150, 200 and 300 mg L⁻¹ of mucilage concentrations were added at fixed time of 40 and 120 min for Al and Fe electrodes, respectively. They were found to be equal to 0.5330 kg m⁻³ for theoretical values ($C_{\text{electrodes, theo}}$), 0.6927, 1.2430, 0.9727, 0.7898 and 0.7705 kg m⁻³ for experimental values ($C_{\text{electrodes, exp}}$), respectively, when using Al electrodes. Whereas, for Fe electrodes consumption the theoretical ($C_{\text{electrodes, theo}}$) and experimental ($C_{\text{electrodes, exp}}$) values were 1.3347 kg m⁻³ and 1.4091, 3.1125, 2.4358, 3.0967 and 1.9293 kg m⁻³, respectively. Therefore, the current efficiency values (CE) (calculated according to Eq. (4)) at 00, 100, 150, 200 and 300 mg L⁻¹ of mucilage concentrations were 130, 233, 182, 148 and 144%, respectively for Al electrodes, and 105, 233, 182, 232 and 144% for Fe electrodes, respectively.

Majdoub *et al.* (2001) reported that the coagulation capacity of OFI mucilage is due to the presence of the polysaccharide stocked in OFI cladodes, which have the ability of water retention. According to Trachtenberg and Mayer (1981), the neutral sugars are composed of galactose (40.1%), arabinose (24.6%), xylose (22.2%), rhamnose (13.1%). Majdoub *et al.* (2001) described that galacturonic acid was present at 10.7 to 19.5%. Miller *et al.* (2008) studied the turbidity removal experiments by addition of the above-mentioned components to turbid water separately and in combination to deduce their coagulations abilities and found that the galacturonic acid by more than 50% may be the main coagulation inducing agent that reduces turbidity.

The addition of OFI mucilage as a coagulant to treat a kaolin turbid solution, Yin (2010) and Miller *et al.* (2008) supposed that the adsorption of colloids and bridging through the galacturonic acid is likely the predominant mechanism of particle destabilization. However, it was noted that the flocs generated when the OFI mucilage was added were whitish, denser, bulkier compared to those obtained without adding mucilage. This phenomenon may be explained by the release of Al⁺³ ions from oxidation reaction of the electrode at the anode, the Al⁺ interact with OH⁻ of the galacturonic acid present in the OFI mucilage to form the hydroxyl polymers, this finally complex have a significant ability to improve the coagulation subsequently to remove pollutants from aqueous solutions.

EC process operating charge, energy, and electrode consumptions

The electrical energy consumption is one of the most significant factors in the electrochemical treatment, primarily because it is linked to running costs (Bayramoglu *et al.*, 2006; Kobya *et al.*, 2009; Ghanbari *et al.*, 2014). According to the previous studies, the addition of OFI mucilage as a bio-coagulant to the EC treatment enhances copper removal and turbidity removal and decreases energy consumption (Adjeroud *et al.*, 2018; Djerroud *et al.*, 2018).

The calculated C_{energy} per COD removal efficiency (%) on the EC treatment using both electrodes Fe and Al electrodes is shown in figure 5.

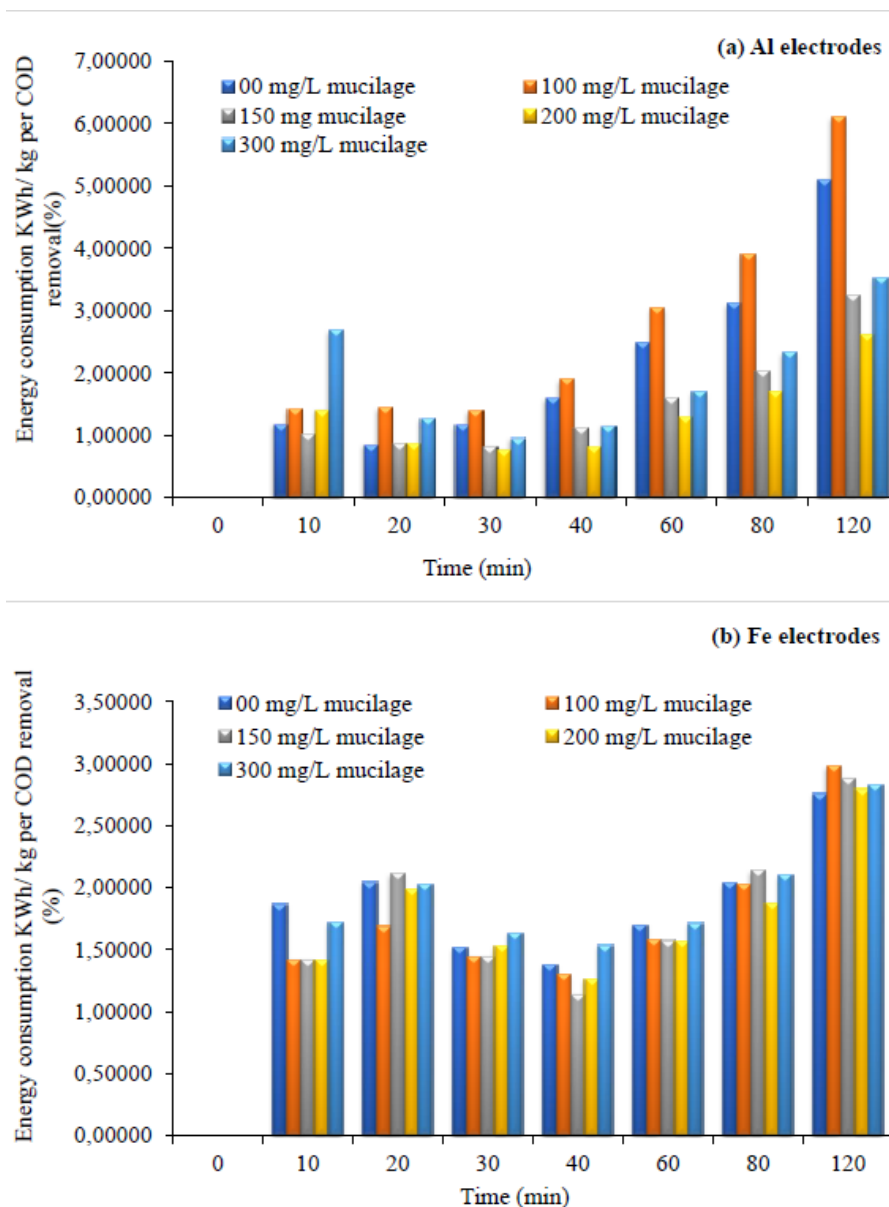


Figure 5. The evolution of the energy consumption (kWh kg^{-1} per COD removal (%)) according to EC time and mucilage concentrations: (a) conditions of Al electrode: initial pH 3, $t_{EC}= 40$ min and $j= 60 \text{ A m}^{-2}$, (b) conditions for Fe electrode: initial pH 6, $t_{EC}= 120$ min and $j= 50 \text{ A m}^{-2}$.

The determined energy consumption per COD removal efficiency (%), (C_{energy}) was calculated according to (Eq. 2). The figure 5 shows for Al electrode at fixed time of 40 min the energy consumption was estimated to be 1.59, 1.89, 1.11, 0.81 and 1.14 kWh kg^{-1} per COD removal efficiency (%) when 00, 100, 150, 200 and 300 mg L^{-1} of OFI mucilage were added, respectively. Whereas for the same concentrations of mucilage the C_{energy} values were 2.75, 2.97, 2.87, 2.8 and 2.82 kWh kg^{-1} per COD removal (%), respectively for EC treatment with Fe electrodes. The addition of 200 mg L^{-1} led to decrease the C_{energy} to 0.81 kWh kg^{-1} per COD removal (%) after 40 min of treatment for EC treatment with Al electrodes.

In the basis of these results, OFI has a beneficial effect to improve the COD abatement efficiency and reduce the energy consumption, especially for Al electrodes. That may be explained by the relationship between hydrogen and aluminum (correlated with their poor Faradic yields) released with energy consumption hence the effect of mucilage on EC treatment.

The energy consumption is correlated with current density; when current density increases the quantity of generated sludge and the energy consumption increased too, since the anode oxidation reaction accelerates and the released Al_3^+ cations rise too. Consequently, the number of $\text{Al}(\text{OH})_3$ compounds also increases. (Chen *et al.*, 2000; Mouedhen *et al.*, 2008; Merzouk *et al.*, 2009). In addition, operating cost increased with rising current density and treatment time (Kobyas *et al.*, 2006; Ghanbari *et al.*, 2014).

The operating costs for Al electrode at 00 and 200 mg L^{-1} of mucilage concentration, were calculated according to Eq. (4) as 3.41 and 3.80 $\text{\$ m}^{-3}$ ($C_{\text{energy}} = 4.76 \text{ kWh m}^{-3}$, $C_{\text{electrode exp.}} = 0.6927$ and 0.7898 kg m^{-3} , respectively) at optimum condition parameters (initial pH 3, $j = 60 \text{ A m}^{-2}$ and $t_{\text{EC}} = 40 \text{ min}$). At the same mucilage concentrations, operational costs for Fe electrodes were calculated as 2.6134 and 4.2201 $\text{\$ m}^{-3}$, respectively ($C_{\text{energy}} = 10.6 \text{ kWh m}^{-3}$, $C_{\text{electrode exp.}} = 1.4091$ and 3.0964 kg m^{-3} , respectively) at the optimum condition parameters (initial pH 6, $j = 50 \text{ A m}^{-2}$ and $t_{\text{EC}} = 120 \text{ min}$). From an industrial standpoint, energy consumption and consequently the running costs of EC process should be decreased to make it possible to implement efficient processes. Although it has been overlooked for some time, the economic aspect of EC performance is now one of the key concerns for veritable application in the industrial sector.

Analysis of generated treatment sludge from EC process

For Fe electrodes, the dried precipitate was red-brown due to the presence of the iron-pollutant and hydroxyl iron-pollutant in the precipitate formed (Wang *et al.*, 2016). The same observation was obtained for the dried sludge when OFI mucilage was added, this could imply that the mucilage does not contribute to the pollutant adsorption mechanism of iron, thus in EC process with or without OFI mucilage addition, the formed precipitates could consist of iron-pollutants and -hydroxyl iron-pollutants.

In the case of Al electrodes, the dried sludge was whitish grey; however, the gotten flocs are bulky within the sludge when the OFI mucilage was added in comparison with smaller flocs in the absence of mucilage. This can be due to the fact that OFI mucilage could engender double precipitation mechanism reaction (pollutant-Al-mucilage-OH and pollutant- $\text{Al}(\text{OH})_3$ -mucilage-OH), whereas in the absence of mucilage the precipitate formed consisted of pollutant-Al and pollutant- $\text{Al}(\text{OH})_3$ (Heidmann and Calmano, 2008; Wang *et al.*, 2016; Adjeroud *et al.*, 2018).

According to Adjeroud *et al.* (2018), the scanning electron microscope analysis (SEM) observation of the lyophilized mucilage extract from the OFI cladodes show an extremely porous structure which confer the capacity to store water and moisture. Three-dimensional network structures are characteristic of adsorbents (Ren *et al.*, 2013). The adsorbents are water insoluble (hence the need to dissolve the mucilage in NaOH). Basically, adsorption is the fixation of the elements on porous material's at the internal cavities or trapped at the carbon nanotubes (Malainine *et al.*, 2001).

The figure 6 (a) show the Fourier transform infrared spectroscopy (FTIR) spectrum of sludge when using the Fe and Al electrodes, in the presence or not of OFI mucilage for both processes. The wavelength range of $3300\text{-}3400 \text{ cm}^{-1}$ is due to hydroxyl (-OH) (Li *et al.*, 2007; Fox *et al.*, 2012), and (-CO) (Singh and Bothara, 2014) groups in the precipitates for forth obtained sludges (Kamnev *et al.*, 1998). The peak at 1636.1 cm^{-1} was ascribed to alkenes (C=C ring) (Nharingo and Moyo, 2016) and to galacturonic acid (Lefsih *et al.*, 2016). The strong absorption peak seen for sludge obtained with Al electrode with or without OFI mucilage, which is a band at about 1126.4 cm^{-1} , was related to -C-O- vibration mode (Li *et al.*, 2007; Fox *et al.*, 2012). According to Jadhav and Mahajan (2014), the band at 1658 cm^{-1} is assigned to proteins. Nharingo and Moyo (2016) study showed

that OFI mucilage comprises functional groups and according to FTIR spectra, it may contribute to adsorption of solution contaminants. In the FTIR analysis of dried cactus, carboxylic acid groups present in the bio-sorbent were defined as responsible for the binding of cationic metals (Pb, Cd) (Barka *et al.*, 2013), also by FTIR analysis, Adjeroud-Abdellatif *et al.* (2020) found that OFI mucilage is made of an extensive range of functional groups as hydroxyl, carboxyl, ketones, phosphate, aldehydes, sulphate groups, and others.

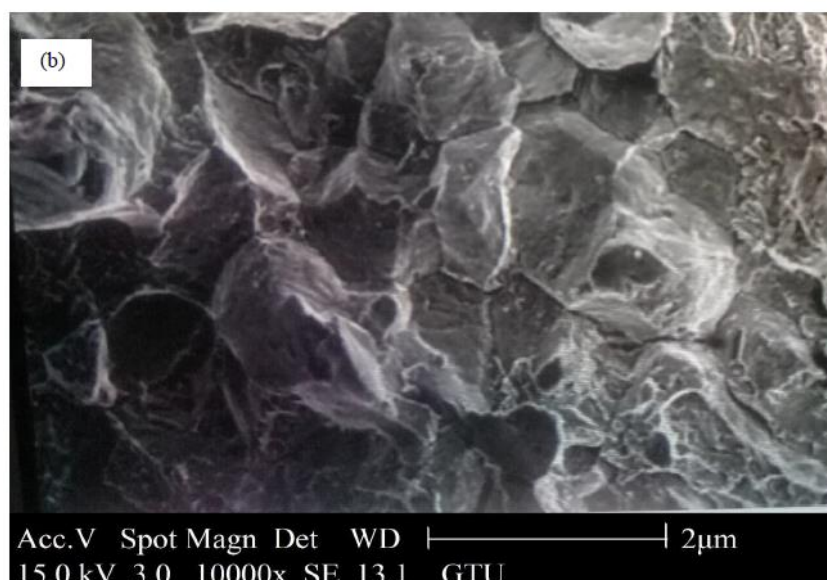
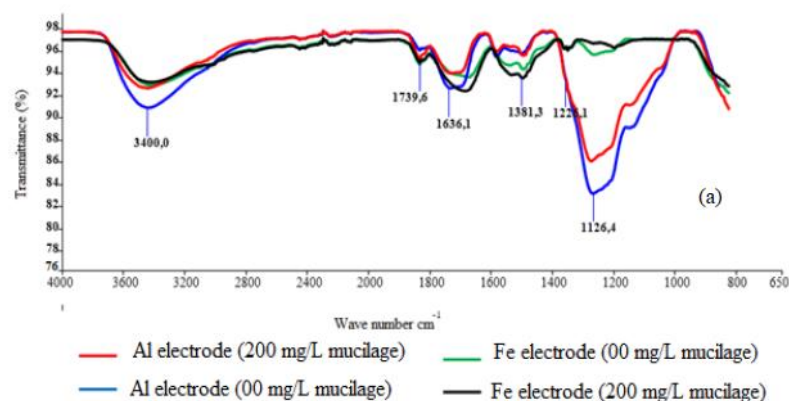


Figure 6 (a). The Fourier transform infrared spectroscopy (FTIR) spectra of sludge for both Al and Fe electrode, (b) scanning electron microscope analysis (SEM) images of the electro-generated sludge of EC treatment using Al electrodes and assisted with OFI mucilage.

The figure 7 (a-b) depicts the X-ray diffraction profile of Al and Fe electro-generated sludges. The results showed that X-ray scheme for Al electro-generated sludge produced a high productivity signal. Conversely, for Fe electrodes no quantitative description for the electro-generated sludge structure and any distinct peaks have been mentioned, which can be explained by the weak adjustment of the iron sludge generated when Fe electrodes were used (Yang *et al.*, 2006). The Al electrodes X-ray diffraction results could reflect the obtained efficiency when Al electrodes were used in EC process assisted with OFI mucilage.

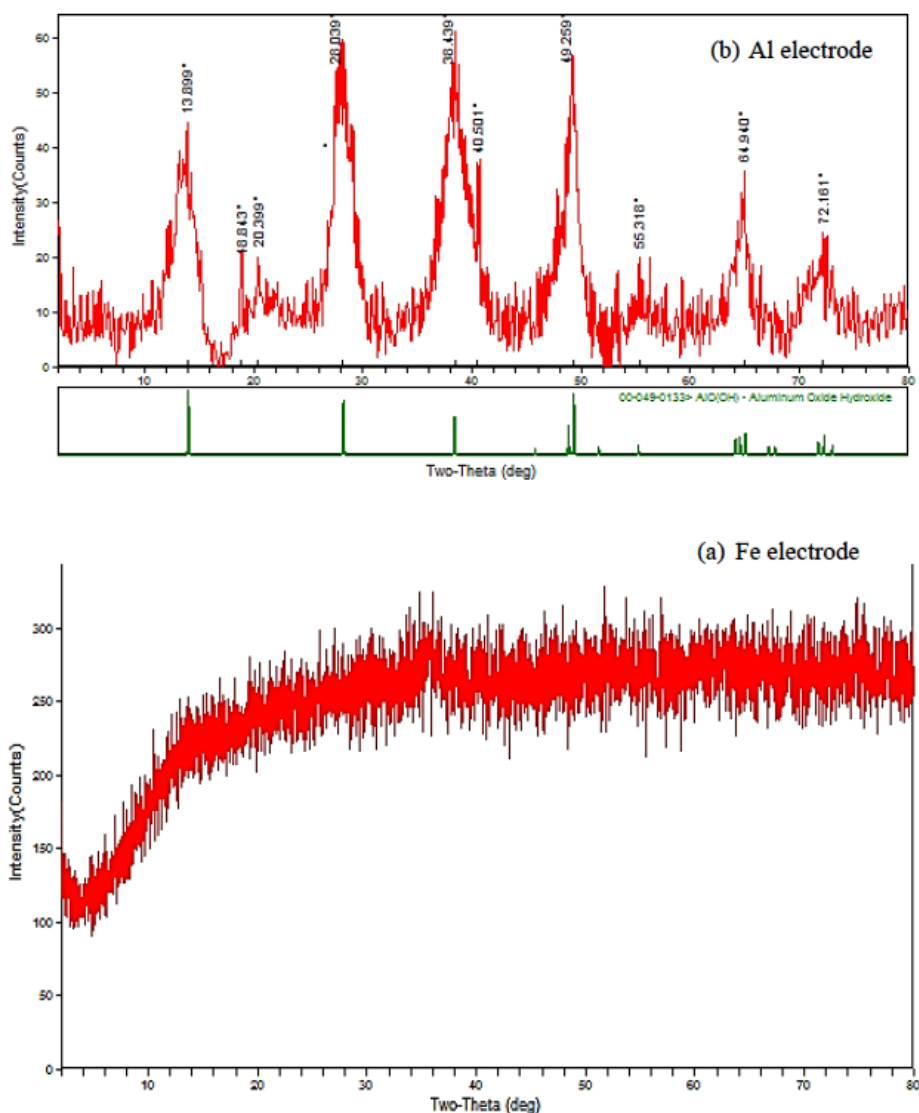


Figure 7. The X-ray diffraction result of generated EC sludge: (a) Al electrodes and (b) Fe electrodes.

Conclusions

The EC treatment using Al electrodes assisted with 200 mg L^{-1} OFI mucilage (current density of 60 A m^{-2} , initial pH of 3), improved the COD removal efficiency (69.77%) after 40 min of EC treatment. This study allows the valorization of OFI plant, which is largely available and accessible, and easy to grow from one hand, and from the other hand OFI responds to the actual growing demand of environmentally/friendly and renewable alternative resources. In addition, in order to be consistent with environmental concerns, is very interesting to reduce the use of chemical adjuvants and replace them by natural coagulants to enhance EC performances, which is already recognized as a very effective treatment process. The OFI cladode mucilage as a natural coagulant proves to be an excellent eco-friendly and economical candidate alternative for the improvement of EC effectiveness. The results showed an improvement of COD removal efficiency, treatment time, and a reduction in specific energy and operating costs. Thus, OFI mucilage addition improves the lucrative practicability of EC process.

Ethics Statement

Not applicable.

Consent for Publication

Not applicable.

Availability of Supporting Data

All data generated or analyzed during this study are included in this article.

Competing Interests

The authors declare that they have no competing interests.

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Author Contributions

Conceptualization, L.G.C. and S.V.G.; methodology, L.G.C., D.G.R. and S.V.G.; formal analysis, L.G.C., D.G.R. and S.V.G.; investigation, L.G.C., D.G.R. and S.V.G.; resources, S.V.G.; data curation, L.G.C.; writing—original draft preparation, L.G.C., D.G.R. and S.V.G.; writing—review and editing, L.G.C., D.G.R. and S.V.G.; project administration, S.V.G.; funding acquisition, S.V.G.

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