

Elimination of natural organic matter by electrocoagulation using bipolar and monopolar arrangements of iron and aluminum electrodes

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Received: 2 February 2017 / Revised: 17 June 2017 / Accepted: 11 July 2017
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Abstract The aim of this research was to evaluate the efficiency of electrocoagulation (EC) for the removal of natural organic matter (NOM) by using iron (Fe) and aluminum (Al) electrodes. The effects of several operational parameters such as initial pH (3–10), time of electrolysis (5–30 min), initial concentration of organic matter (10–50 mg NOM/L), current density (0.25–1.25 mA/cm²), type of electrode material ($n = 4$, 2 sides \times 11 cm \times 10 cm, wall thickness = 2 mm, distance between each electrode = 5 mm), and type of connection of electrodes (bipolar and monopolar configurations) were explored for the removal of NOM from synthetic humic acid solution in

a 2 L laboratory-scale EC cells ($A_s/V = 0.110 \text{ cm}^{-1}$). The optimum conditions for the process were identified as pH = 3 and 7, electrolysis time = 20 and 10 min for Fe and Al electrodes, respectively. Using both electrodes at current density = 0.25 mA/cm² and initial concentration of organic matter = 50 mg/L, a NOM removal efficiency of almost 100% could be achieved in the bipolar mode. Based on the optimum conditions, specific reactor electrical energy consumptions were 14.90 kWh/kg Al (or 0.092 kWh/m³) and 2.88 kWh/kg Fe (or 0.11 kWh/m³). Specific electrode consumptions were obtained to be 0.0062 and 0.0382 kg/m³, and operating costs of the EC system were preliminary estimated at 0.057 and 0.119 \$/m³ for Al and Fe electrodes, respectively.

Editorial responsibility: Abhishek RoyChowdhury.

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Keywords Natural organic matter · Electrocoagulation · Aluminum electrode · Iron electrode

Introduction

Natural organic matter (NOM) is found as a mixture of organic compounds in all water environments, especially surface water. In such waters, NOM contains both hydrophobic and hydrophilic compounds. Aromatic hydrocarbons form an integral part of the hydrophobic component of NOM. High aliphatic carbons and nitrogenous compounds like hydrocarbons, proteins, sugars, and amino acids form an integral part of the hydrophilic NOM (Leenheer 2004). Hydrophobic acids are a major part of the NOM fraction (more than half the total dissolved organic carbon (DOC)) in water (Matilainen et al. 2011), and the major components of these acids include humic (C₁₈₇H₁₈₆O₈₉N₉S₁) and fulvic (C₁₄H₁₂O₈) acids. The presence of NOM makes drinking water become undesirable with a characteristic unpleasant

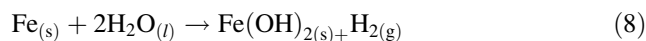
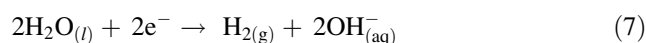
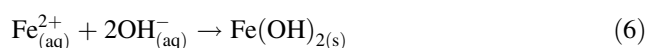
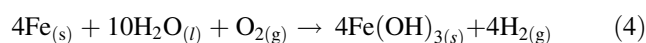
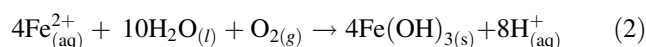
color, odor, and taste (Vosoughi Niri et al. 2015). During the chlorination process, they can also react with chlorine to produce hazardous disinfection by-products such as halo-methanes (i.e., CFCl_3 (trichlorofluoromethane), CF_2Cl_2 (dichlorodifluoromethane), CF_3Cl (chlorotrifluoromethane), and CF_3Br (bromotrifluoromethane)). Moreover, they negatively affect the adsorption process, lessen membrane efficiency, and trigger biological regrowth in distribution systems (Yildiz et al. 2007; Matilainen et al. 2011).

Various reactions including adsorption, chemical coagulation and sedimentation, oxidation, ion exchange and filtration (involving several membranes) have been employed in the removal of NOM from aqueous solutions (Metcalf and Eddy 2004; Ali and Gupta 2006; Ali 2010, 2012, 2014; Ali et al. 2012a). Electrocoagulation (EC) has been recognized as an environmentally benign method in terms of operational cost when compared with other treatment methods (Matilainen et al. 2006; Ali et al. 2011, 2012b, c, 2013, 2014). Some advantages of this method are that simple equipment is required, reaction time is shorter, and capital and operating costs are low. Small sludge volumes are produced by EC, and this is due to the low water content of the sludge (Askari et al. 2014), so that dehydration easily occurs in the sludge. Additionally, less space for installation is needed in this process, no adjustment of the chemical properties of wastewater is required, and the cost of equipment, operation, and maintenance is also low. The possibility that secondary pollution will occur is reduced because there is no addition of chemicals in this process (Yildiz et al. 2007; Boudjema et al. 2014). Considering the above-mentioned advantages of the EC process, great efforts have been done by a number of researchers for the removal various contaminants by using this promising technology (Yetilmezsoy et al. 2009; Zongo et al. 2009a, b; Lemlikchi et al. 2012).

EC can be described as a process that involves the synthesis of a metal hydroxide (i.e., $\text{Al}(\text{OH})_3$ (aluminum hydroxide), $\text{Fe}(\text{OH})_2$ (iron(II) hydroxide)) in water or wastewater by dissolving electrons of iron (Fe) or aluminum (Al) soluble anodes (Aleboyeh et al. 2008). Adsorption, charge neutralization, precipitation, and trapping in the hydroxide (OH^-) precipitates are some examples of the mechanisms that hydroxides and metal cations (Al^{3+} , Fe^{2+} , Fe^{3+} , Ti^{2+} , Ag^+ , Au^{3+} , etc.) use to remove contaminants. Electroflotation, EC, and precipitation all occur simultaneously during the electrochemical process (Chen 2004). Within the EC reactor or cell, the EC method can be easily and reliably performed for generation of coagulants in situ by electrochemical dissolution of Fe or Al electrodes into electrolyte solution to destabilize pollutants.

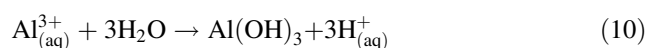
When Fe is used as the electrode material, Fe-based oxidation reactions produce iron hydroxide, $\text{Fe}(\text{OH})_n$, where $n = 2$ or 3. Two mechanisms have been proposed for the production of $\text{Fe}(\text{OH})_n$ (Yetilmezsoy et al. 2009):

(a) The first mechanism for the production of $\text{Fe}(\text{OH})_3$ consists of Eqs. (1) and (2) for anode, Eq. (3) for cathode, and Eq. (4) for the overall reaction, and (b) the second mechanism for the production of $\text{Fe}(\text{OH})_2$ consists of Eqs. (5) and (6) for anode, Eq. (7) for cathode, and Eq. (8) for the overall reaction. The following equations are the summarized reactions of the EC process using the Fe electrodes (Yetilmezsoy et al. 2009):



Other products of the EC process involving the use of Fe electrodes are $\text{Fe}(\text{H}_2\text{O})_6^{3+}$, $\text{Fe}(\text{H}_2\text{O})_5\text{OH}^{2+}$, $\text{Fe}(\text{H}_2\text{O})_4(\text{OH})_2^+$, $\text{Fe}_2(\text{H}_2\text{O})_8(\text{OH})_2^{4+}$, $\text{Fe}_2(\text{H}_2\text{O})_6(\text{OH})_4^{2+}$ (Lemlikchi et al. 2012).

Oxidation of Al ions occurs at the anode. The Al ions are later change to polymeric species or flocs of $\text{Al}(\text{OH})_3$. When Al is used as the electrode, the electrolytic dissolution of the Al anode produces the cationic monomeric species such as Al^{3+} and $\text{Al}(\text{OH})_2^+$ at acidic conditions. At appropriate pH values, they are transformed initially into $\text{Al}(\text{OH})_3$ and finally polymerized to $\text{Al}_n(\text{OH})_{3n}$ according to the following reactions (Yetilmezsoy et al. 2009; Drouiche et al. 2011):



Depending on the pH of the aqueous medium, some other products of the Al anode reactions, such as $\text{Al}(\text{OH})_2^+$, $\text{Al}_3(\text{OH})_4^{5+}$, $\text{Al}_6(\text{OH})_{15}^{3+}$, $\text{Al}_7(\text{OH})_{17}^{4+}$, $\text{Al}_{13}\text{O}_4(\text{OH})_{24}^{7+}$, $\text{Al}(\text{OH})_2^{2+}$, $\text{Al}_2(\text{OH})_2^{4+}$, and $\text{Al}(\text{OH})_4^-$, may be formed in the system (Boudjema et al. 2014). Additionally, various forms of charged multimeric hydroxo Al^{3+} species may also be present under appropriate conditions. These gelatinous charged hydroxo cationic complexes can effectively remove pollutants by adsorption process (Yetilmezsoy et al. 2009).

There are two types of connection modes (i.e., bipolar and monopolar configurations) in the EC reactor. In bipolar

connections, the electrodes with no electrical connections were positioned such that the two electrodes were aligned parallel with each other (Lemlikchi et al. 2012). There was division of current between the connections involving all anodes and cathodes (Emamjomeh and Sivakumar 2009). To the best of the authors' knowledge, although a number of EC configurations (Dubrawski and Mohseni 2013; Askari et al. 2014; Mohora et al. 2014; Chellam and Sari 2016) have been conducted in recent years regarding the treatment of NOM from aqueous solutions, there is still an important gap in the relevant literature concerning the investigation of the efficiency and particularly the economics of this process using bipolar and monopolar arrangements of Fe and Al electrodes. Filling this gap is a prerequisite for the effective planning of project infrastructure for large-scale EC applications and for developing an up-to-date database on this subject.

Based on the foregoing facts, the specific objectives of this study were: (1) to study and the effects of current density, initial concentration, initial pH, electrolysis time, and bipolar and monopolar electrode configurations on the efficiency of the present EC process; (2) to investigate the optimal experimental conditions for removal of NOM from a synthetic matrix solution in a batch reactor using aluminum and iron electrodes; and (3) to estimate energy consumption and operating cost of the present system at the optimum conditions.

Materials and methods

Electrocoagulation setup

Figure 1a, b shows the laboratory-scale cylindrical shape glass EC cells (2-L beakers) for bipolar and monopolar electrode configurations, respectively. In EC tests, two common use types of sacrificial electrodes (Al and Fe) were studied to select the most appropriate material for the present application, since they are cheap and their production is very simple (Yetilmezsoy et al. 2009). For comparative purpose, preliminary EC tests were carried out with both materials used as sacrificial anode under the same conditions. Both Al or Fe cathodes and anodes were made from rectangular-shaped plates with the dimensions of 110 mm × 100 mm × 2 mm (width, height, and wall thickness, respectively). The active surface area (A_s) for each electrode that was submersed into the synthetic humic acid solution was 220 cm² (2 sides × 11 cm × 10 cm). There were four electrodes during the electrolysis, and the distance between each electrode was 5 mm.

The ratio between the active surface area ($A_s = 220$ cm²) and the volume of solution ($V = 2$ L = 2000 cm³) in the EC reactor (A_s/V) as crude measure of the potential for

delivering Al coagulant and electrochemical bubbles to ECF system was 0.110 cm⁻¹, which was consistent with the A_s/V value (0.118 cm⁻¹) reported by Mohora et al. (2014). In this study, we examined both monopolar and bipolar electrode connection modes in which the volume of treated water was 1250 mL. Moreover, the power was supplied by a digital Lodestar Leda LP6003D (0–60 V, 0–3A adjustable) device used as the DC source.

Experimental protocol

An electrolysis time of 20 min was considered to examine the performance of the EC process for each run, and samples were taken at 5-min intervals from the drain tube section in the electrocoagulator. The electrode plates were washed thoroughly with 5% HCl (Sigma-Aldrich, Germany), tap water, and then distilled water after each use. The samples were magnetically stirred during the EC treatment at the speed of 100 rpm (≈ 10.47 rad/s). The reactor-specific energy consumption was calculated in terms of kWh/m³ calculated using Eq. (1) as follows (Mohora et al. 2014):

$$C_{\text{energy}} = (U \times I \times t_{\text{EC}}) / (V) \quad (12)$$

where C_{energy} is the consumed electrical energy (kWh/m³, where 1 W = 1 A × 1 V), U is the applied cell potential (V), I is the operating current (A), t_{EC} is the treatment or reaction time (h), and V is the volume of the solution (m³).

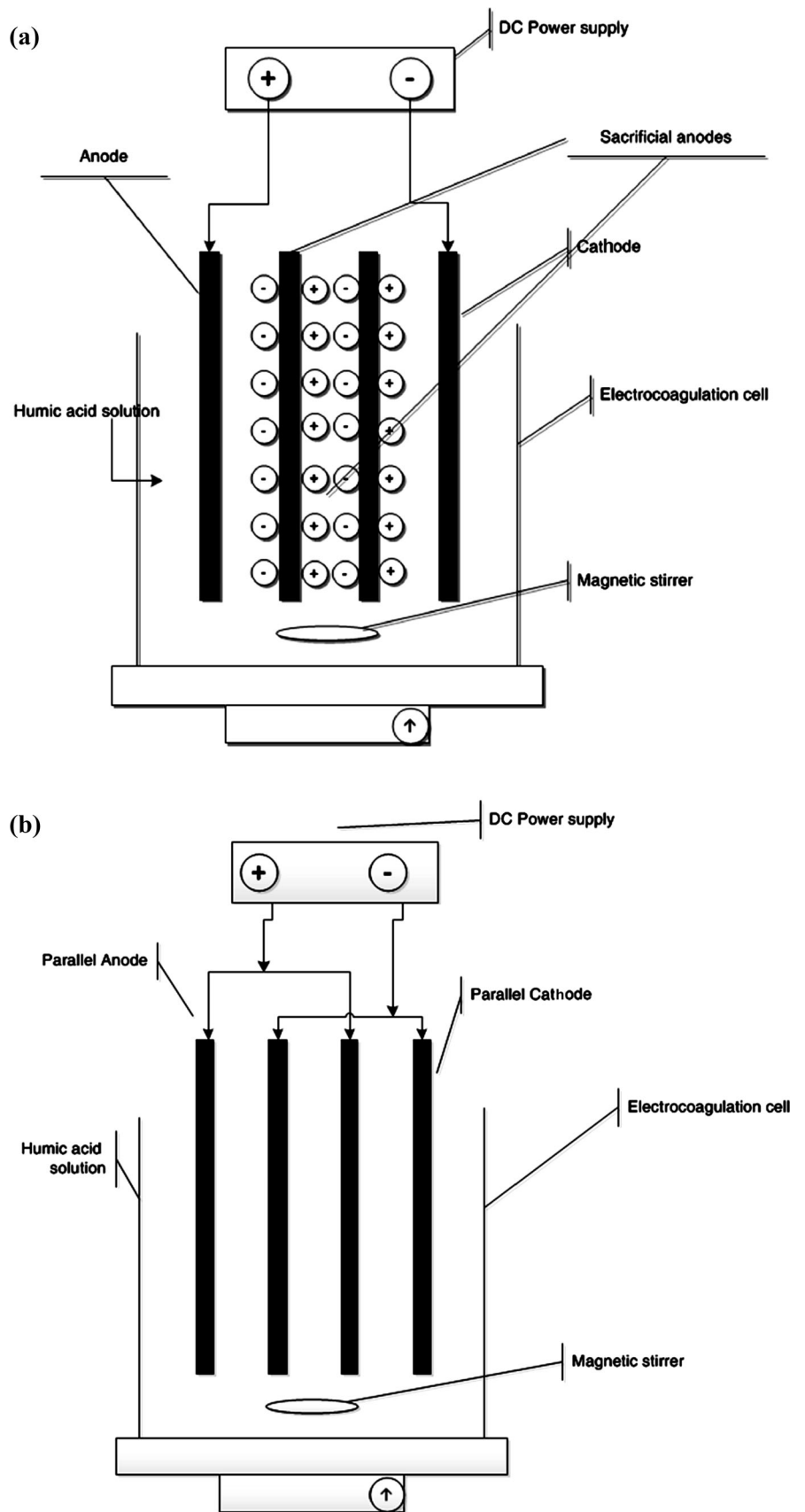
Solution chemistry

The humic acid (humic acid, sodium salt 50–60%) was obtained from Across[®] Systems GmbH (Karlsbad, Karlsruhe, Baden-Württemberg, Germany) and used as a representative compound for NOM-impacted synthetic solution. The amount of humic acid as sodium salt (50–60%) was 905 mg to produce 1 L of solution (500 mg/L) by diluting the prepared stock solution of humic acid. The stock solution of the humic acid was prepared in alkaline conditions to prevent precipitation, and the initial pH of this solution was 11 ± 0.09 . During the EC experiments, the pH of samples was adjusted by the gradual addition of 0.1 N HCl (hydrochloric acid, 36% w/w aq. soln.) solution. All experiments were conducted at a room temperature of 25 ± 2 °C.

Analytical techniques

A TOC analyzer was used to measure the NOM of the samples (Shimadzu, Tokyo, Japan). A turbidimeter was used to measure turbidity (2100 AN, HACH, Loveland, CO, USA). Electrical conductivity was determined by a conductivity meter (Sension 5, HACH), and the pH was

Fig. 1 Bipolar (a) and monopolar (b) electrode connections in the electrocoagulation reactor



measured with a pH meter (827 Metrohm, Herisau, Switzerland). The weights of chemicals to be added to synthetic media were quantified with an electronic precision analytical balance (Secura 313-1S milligram). After filtration, a UV/VIS spectrophotometer (Lambda 25, PerkinElmer, Waltham, MA, USA) was used to measure the absorbance of samples at a 254 nm wavelength.

Results and discussion

Effects of current density

In several electrochemical processes, the most important determinants for controlling reaction rate in the reactor are current density and electrolysis time. Apart from the determination of coagulant dosages, current density also determines the mixing rate in EC (Chou et al. 2010). Thus, investigations regarding the effect of current density on the removal of pollutants were conducted. Figure 2 shows the effect of current density on NOM removal efficiency. A gradual increase in the current density from 0.25 to 1.25 mA/cm² led to an increase in NOM removal efficiency for Fe electrodes, but no further change occurred with higher densities, when Al electrodes were used. This could be ascribed to the reactions between organic matter with Fe and Al ions present in the solution to form insoluble products.

The effect of current density on the removal efficiency of NOM and the determination of optimum current density was considered at the average electrolysis time of 20 min. Polymeric species of Fe can neutralize the surface charge of organic molecules in the electrocoagulator. It therefore implies that these species neutralize the electrostatic charges on the surface of the NOM molecules, thus reducing the electrostatic interparticle repulsion, which increases the van der Waals force of attraction, and finally removes NOM from the aqueous solution (Chou et al. 2010). Consequently, 0.25 mA/cm² was chosen as the optimum current density for the present case. Moreover,

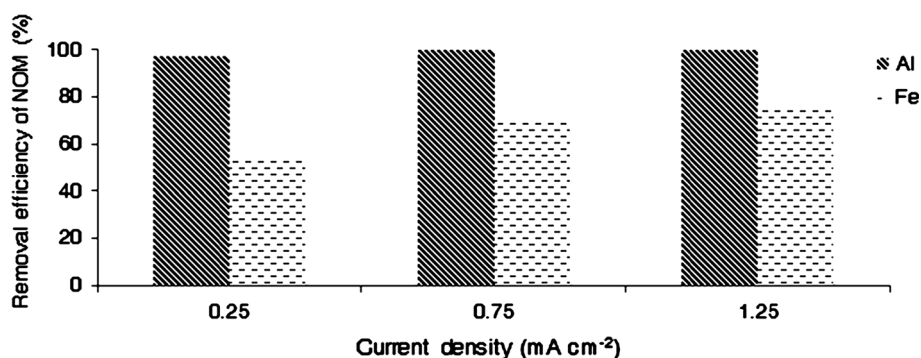
charge loading was plotted against the voltage between electrodes (Fig. 3). When the charge loading increased to more than 4 Faradays/m³, there was a slightly increase in voltage between the electrodes. Therefore, it is important to keep the optimum charge loading at the end of the sharp increase stage (Zongo et al. 2009a, b).

Effect of electrolysis time

Figure 4 illustrates the effect of electrolysis time on the removal of NOM at 0.25 mA/cm². The removal efficiency of NOM increased with the increase in the electrolysis time. The expected electrolysis times for a removal efficiency of more than 50% for the Al and Fe electrodes were 10 and 20 min, respectively. It was observed that the removal efficiency of NOM was rapid in the beginning and then reached almost a constant level after 20 min. To summarize, longer electrolysis times (>20 min) unnecessarily prolonged the process to obtain similar result. Consequently, taking into account the cost of energy consumed in the EC process, an electrolysis time of 20 min was considered to be optimal for further batch experiments. Yetilmesoy et al. (2009) reported similar results for the decolorization and chemical oxygen demand (COD) reduction of anaerobically pretreated poultry manure wastewater by the EC process.

The core component of the present treatment facility is the electrode assembly. Therefore, it is important to select quality materials. The most common electrode materials for EC are Al and Fe (Yetilmesoy et al. 2009). They are affordable in terms of price, readily available, and have proved effective in functionality. Thus, in this study, these two electrodes were tested under the similar experimental conditions. According to the results obtained for both electrodes with the same current density (0.25 mA/cm²) and operating time ($t_{EC} = 20$ min for Fe and $t_{EC} = 10$ min for Al), Al electrodes had a higher removal efficiency than the Fe electrodes in the removal of NOM from the synthetic wastewater. In addition, the treated sample with Al electrodes was very clear and stable, whereas the one

Fig. 2 Effect of current density on the removal efficiency of NOM (initial NOM concentration = 50 mg/L, pH = 7, agitation speed = 100 rpm)



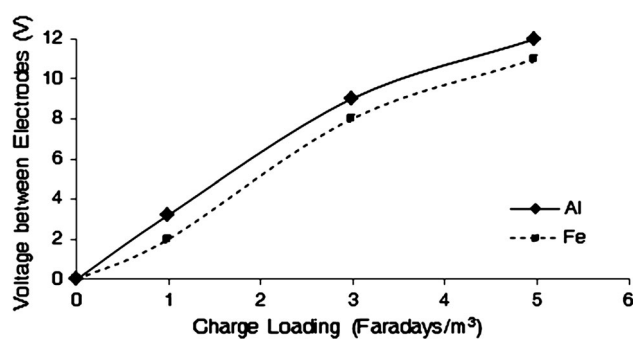


Fig. 3 Variation of voltage between electrodes as a function of charge loading for different current density, pH = 7

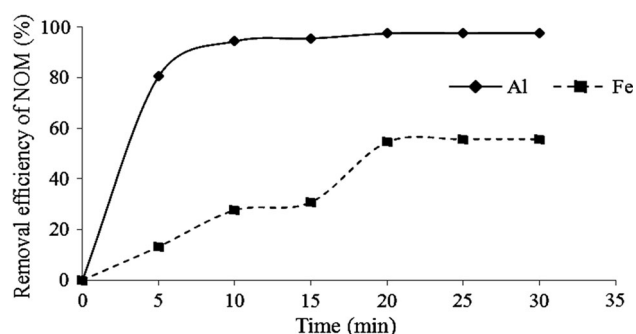


Fig. 4 Effect of electrolysis time on the removal efficiency of NOM at the current density of 0.25 mA/cm² (NOM = 50 mg/L, pH = 7, agitation speed = 100 rpm)

treated with Fe electrodes was initially greenish. In a study on EC-based treatment of paper mill wastewater, Zodi et al. (2011) performed an evaluation on the performance of EC for the removal of arsenic and non-biodegradable organic pollutants and revealed that the COD (Chemical Oxygen Demand) abatement obtained with Al electrodes was higher than that of Fe electrodes.

Effect of initial pH

Chen (2004) demonstrated that the initial pH has a notably influence on the performance of the EC process. In this study, according to the previous literature (Askari et al. 2014), an initial pH range of 3–10 was studied to determine NOM removal efficiency. As seen from the results presented in Fig. 5, for an initial NOM concentration of 50 mg/L, the removal efficiency value was almost 100% at pH = 7, whereas the removal values were about 84% at the pH values of 3 and 10 for Al electrodes at a constant treatment time of 20 min. Therefore, in order to attain the highest NOM removal efficiency, the optimum pH was chosen as 7 for Al electrodes. For Al electrodes, a slow decrease in NOM removal efficiency in acidic (pH = 3 < 7.0) or alkaline (pH = 10 > 7.0) medium can be ascribed to the fact that acidic or alkaline medium

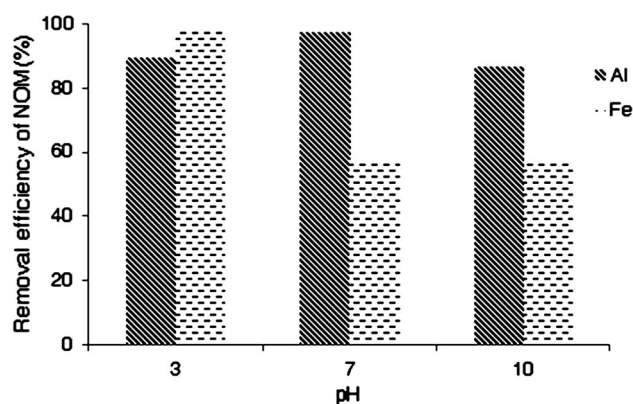


Fig. 5 Effect of pH on NOM removal efficiency as a function of initial concentration of 50 mg/L and current density of 0.25 mA/cm²

affects the mechanism of electrocoagulation and leads to the formation of different chemical compounds in the aqueous solution. Therefore, it is noted that the initial pH is one of the major factors that influences NOM removal in the EC treatment process (Yetilmezsoy et al. 2009). The pH of the solution also affects the solubility and aluminum hydroxides formation, such that at pH = 7, Al(OH)_{3(aq)}, Al(OH)₂⁺, AlO⁺, Al(OH)₄⁻, and Al(OH)_{3(s)} are the species available in the solution (Chou et al. 2010). The adsorption of NOM can occur on the surface of hydroxides of Al because of their insoluble nature (Gheraout et al. 2009). Moreover, the decrease in pH of the alkaline solution could be due to hydroxide precipitates formation (i.e., Al(OH)₄⁻) with other cations (Yildiz et al. 2007).

When Fe electrodes were used, and the initial NOM concentration was 50 mg/L at pH = 3, NOM removal efficiency was almost 100%. Observations revealed that the highest NOM removal for Fe electrodes was achieved in acidic medium. The dominant species in the Fe system was Fe(OH)₄⁻ at pH = 10, a soluble form of iron hydroxide (İrdemez et al. 2006). As a result, it can be concluded that EC treatment of NOM may also be conducted at the optimum pH of 3, using Fe electrodes. Final pH in the acidic solution was increased after electrocoagulation, but that of the alkaline solution was decreased. As also seen from Eq. (3) (consumption of H_(aq)⁺ ions by the first mechanism) or Eq. (7) (formation of OH_(aq)⁻ ions by the second mechanism), evolution of hydrogen gas at the cathode could be the cause of the increase in pH in acidic conditions (Chen 2004).

Figure 6 shows the electrode consumption per kg of NOM removed in relation to the initial pH. For instance, in alkaline medium (pH > 7.0), Fe had a consumption value of about 0.01 kg electrode/kg NOM, and this was nearly 25% lower than that of Al for the same pH range. This could probably be as a result of the differences in the mechanisms of NOM removal for the two sacrificial



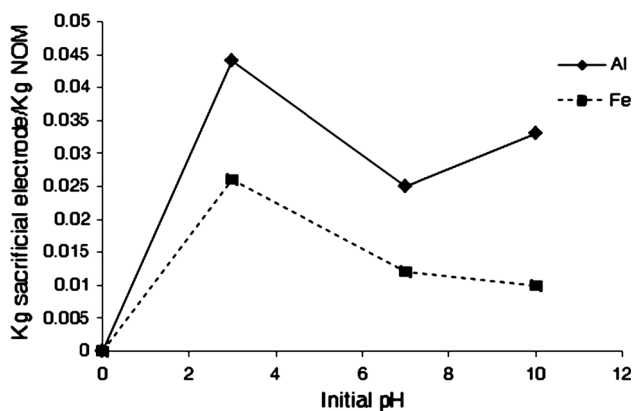


Fig. 6 Effects of initial pH on electrode consumption (initial NOM concentration = 50 mg/L, conductivity = 1141 and 1000 $\mu\text{S}/\text{cm}$ for Al and Fe electrodes, respectively)

electrodes. The mechanism of NOM removal using Al electrodes is EC, while the NOM removal by Fe electrodes was as a result of the collective effects of EC and electro-oxidation (Kobyta et al. 2003).

Effect of initial concentrations

The effect of the initial concentrations of NOM on the removal efficiency was investigated within the range of 10–50 mg NOM/L. The results are depicted in Fig. 7. Previous studies concluded that an increase in the initial concentration of NOM led to a decrease in its removal efficiency, but with low NOM concentrations, its removal efficiency was volatile (Kashefialasl et al. 2006; Yildiz et al. 2007; Dubrawski et al. 2013). Figure 7 shows that the removal efficiency of NOM was almost 100% for an initial concentration of 50 mg/L, when the Fe electrodes were used. An NOM removal efficiency of 85% was recorded for the two initial NOM concentrations of 10 and 25 mg/L.

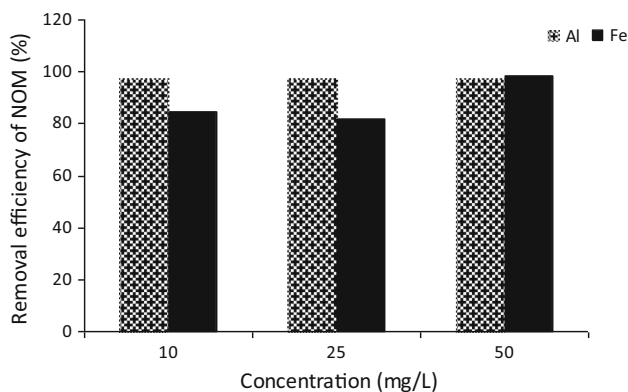


Fig. 7 Effect of initial concentration on the removal efficiency of NOM at optimum conditions (pH = 7 for Al electrode, pH = 3 for Fe electrode, current density = 0.25 mA/cm², and the hydrogen ion concentration was kept constant)

It should be also noted that Fe²⁺ produced by EC forms a complex with NOM (especially at the low concentration), and this prevents the conversion of Fe²⁺ to Fe³⁺ to generate Fe(OH)₃ precipitate. Thus, NOM mediates the oxidation of Fe²⁺ (Bagga et al. 2008). The NOM concentrations have also reduced during the treatment process. The dilution theory vividly explains this phenomenon as the diffusion of a layer of ions in the vicinity of the electrodes in dilute solutions decreases the reaction rate of EC. However, the diffusion rate of metal ions on the surface of electrodes is unaffected in highly concentrated solutions (Gharibi et al. 2012).

Effect of bipolar and monopolar electrode configurations

Figure 8 shows the NOM removal efficiencies of two electrodes (bipolar and monopolar) connections under the optimum conditions of pH and electrolysis time with Al and Fe electrodes. For bipolar connections, the NOM removal efficiencies at the initial concentration of 50 mg/L were found to be higher than those obtained for monopolar ones. The present results were consistent with the findings of other research conducted for the removal organics from

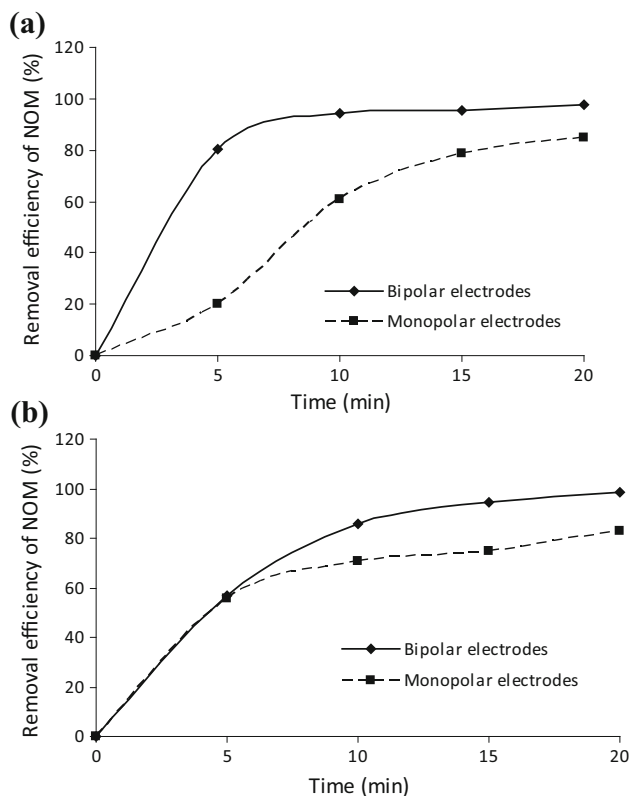


Fig. 8 Effect of bipolar and monopolar electrode connections on NOM removal efficiency at the initial concentration of 50 mg/L and optimum conditions: **a** Al electrode and **b** Fe electrode

oily bilgewater by electrocoagulation process (Asselin et al. 2008). Similarly, Jiang et al. (2002) evaluated the performance of an EC flotation process for water treatment and reported that the bipolar connection was more efficient in the removal of organic compounds than a monopolar connection.

Operating costs and electrical energy consumption

One of the most important factors that affect the applicability of the EC process is the operating cost including cost of electrode material, cost of electrical energy consumed, and cost of chemicals used for pH adjustment per volume of treated water. The amount of electrochemically formed Al or Fe ions, which depends on the operating time (t_{EC}) and the applied current (I), was estimated by using the following expression (Mohora et al. 2014):

$$C_{\text{electrode}} = (I \cdot t_{EC} \cdot M_{w, \text{Al or Fe}}) / (z \cdot F \cdot V) \quad (13)$$

where $C_{\text{electrode}}$ is the theoretical amount of Al or Fe ions produced (kg Al or Fe electrode/m³ treated humic acid solution) by current I (A) passed for a duration of operating time t_{EC} (s), z is the number of electrons involved in the oxidation/reduction reaction (i.e., $z_{\text{Al}} = 3$ or $z_{\text{Fe}} = 2$), F is Faraday's constant (96,485 C/mol, where 1 Faraday = 96,485 coulombs (C), and 1 C = 1 A × 1 s), $M_{w, \text{Al or Fe}}$ is the molecular weight of Al or Fe (i.e., 0.02698 kg/mol for Al or 0.05585 kg/mol for Fe), and V is the volume of humic acid solution treated (m³) for t_{EC} .

Based on both Eqs. (12) and (13), the following equation was used to determine the operating cost (OC) in \$/m³ (Mohora et al. 2014):

$$\text{OC} = a \cdot C_{\text{energy}} + b \cdot C_{\text{electrode}} + c \cdot C_{\text{chemical}} \quad (14)$$

where C_{energy} is the electrical energy consumption (kWh/m³) expressed in Eq. (12), $C_{\text{electrode}}$ is the electrode consumption (kg Al/m³ or kg Fe/m³) defined in Eq. (13), and C_{chemical} is the chemical consumption for pH adjustment per volume of treated water (kg HCl/m³). Moreover, unit prices for a and b are given for Asian market, November 2016 as follows: (1) a is electrical energy price (0.122 \$/kWh); (2) b is Al or Fe electrode material price for wall thickness of 2 mm (i.e., 3 \$/kg Al or 0.625 \$/kg Fe); and (3) c is hydrochloric acid price (0.25 \$/kg HCl, 36%). According to the optimum conditions, specific reactor electrical energy consumptions were determined as 0.092 kWh/m³ (or 14.90 kWh/kg Al) and 0.11 kWh/m³ (or 2.88 kWh/kg Fe) for Al and Fe electrodes, respectively.

Considering the process-related inputs and optimal conditions ($V_{\text{sample}} = 2$ L, $I = 0.25$ mA/cm², distance between electrodes = 0.5 cm, $t_{EC} = 10$ min for Al and 20 min for Fe electrodes, initial pH = 7 for Al and 3 for Fe electrodes, $z_{\text{Al}} = 3$ for Al and $z_{\text{Fe}} = 2$ for Fe electrodes,

$U = 5$ V for Al and 3 V for Fe electrodes, $M_w = 0.02698$ kg/mol for Al and 0.05585 kg/mol for Fe electrodes, and $b = 3$ \$/kg for Al and 0.625 \$/kg for Fe electrodes), the electrode consumptions ($C_{\text{electrode}}$) were obtained to be 0.0062 and 0.0382 kg/m³ for Al and Fe electrodes, respectively.

Taking into account both HCl volumes (1 cc for Al and 3 cc for Fe, where 1 cc = 20 drops = 1 mL) consumed to decrease pH from its initial value (pH of the stock humic acid solution = 11 ± 0.09) to the optimum values (pH = 3 for Fe and pH = 7 for Al) and other chemical-related data ($c = 0.25$ \$/kg for HCl (Shijiazhuang Xinlongwei Chemical Co., Ltd., 2016), $M_w = 36.461$ kg/mol for 0.1 N HCl, normality of HCl = 0.1 N, purity of HCl = 36%, density of HCl = 1.18 g/mL), the chemical costs were calculated as 0.0273 and 0.082 \$/m³ for Al and Fe electrodes, respectively.

Electrical energy consumption was determined at the optimum time of 20 and 10 min for Fe and Al electrodes, respectively. For the present case, the specific reactor electrical energy consumptions (C_{energy}) according to Eq. (12) were calculated as 0.092 and 0.11 kWh/m³ for Al and Fe electrodes, respectively. Comparison of electrical energy consumption for both bipolar and monopolar modes at the optimum conditions of pH and reaction time showed that higher energy consumption was observed in the bipolar mode for both Al and Fe electrodes. Increased energy consumption in the bipolar connection could be a result of the potential difference between the electrodes. A decrease in the conductivity of the solution increased the cell voltage, and this was as a result of the enhanced consumption of solution energy (Gharibi et al. 2012). Finally, using Eq. (14), total operating costs of the present EC treatment system were preliminary estimated to be 0.057 and 0.119 \$/m³ for Al and Fe electrodes, respectively.

Findings of the present study clearly revealed that the EC technology could be successfully implemented an advanced treatment technique for removal of residual NOM-based contaminants (such as hydrophobic and hydrophilic compounds) from aqueous solutions. Looking at the outcome of the research, further investigations may be needed to investigate the economics of the EC process for highly NOM-impacted real waters. Since the present work describes the results of our preliminary efforts on the optimization of a laboratory-scale EC process conducted with two types of sacrificial electrodes, this has been left for future studies.

Conclusion

The applicability of EC was demonstrated to be a viable and economical technique for the removal of NOM from aqueous solutions. The optimum pH for Al and Fe

electrodes was determined as 7 and 3, respectively. The removal of NOM could be rapidly and cost-effectively achieved at 0.25 mA/cm² within 20 min of electrolysis time. In case of use of Al and Fe electrodes, the removal NOM efficiency was almost 100% for the initial concentration of 50 mg/L under the optimum conditions. The Al–Al arrangement was observed to have a higher performance in comparison with Fe–Fe arrangement, and the bipolar connection was found to be better than the monopolar one. The economics of the studied process indicated that the EC technique could be cost-effectively applied to eliminate the hazardous impacts of hydrophobic and hydrophilic compounds on the aquatic ecosystem.

Acknowledgements The authors would like to express their profound gratitude to the Department of Environmental Health Engineering, School of Public Health of the Tehran University of Medical Sciences, for their unmeasurable support.

Compliance with ethical standards

Conflict of interest There is no conflict of interest declared by the authors.

References

- Aleboyeh A, Daneshvar N, Kasiri MB (2008) Optimization of CI acid red 14 azo dye removal by electrocoagulation batch process with response surface methodology. *Chem Eng Process* 47:827–832
- Ali I (2010) The quest for active carbon adsorbent substitutes: inexpensive adsorbents for toxic metal ions removal from wastewater. *Sep Purif Rev* 39:95–171
- Ali I (2012) New generation adsorbents for water treatment. *Chem Rev* 112:5073–5091
- Ali I (2014) Water treatment by adsorption columns: evaluation at ground level. *Sep Purif Rev* 43:175–205
- Ali I, Gupta VK (2006) Advances in water treatment by adsorption technology. *Nat Protoc* 1:2661–2667
- Ali I, Khan TA, Asim M (2011) Removal of arsenic from water by electrocoagulation and electro dialysis techniques. *Sep Purif Rev* 40:25–42
- Ali I, Asim M, Khan TA (2012a) Low cost adsorbents for the removal of organic pollutants from wastewater. *J Environ Manag* 113:170–183
- Ali I, Gupta VK, Khan TA, Asim M (2012b) Removal of arsenate from aqueous solution by electro-coagulation method using Al–Fe electrodes. *Int J Electrochem Sci* 7:1898–1907
- Ali I, Khan TA, Asim M (2012c) Removal of arsenate from groundwater by electrocoagulation method. *Environ Sci Pollut Res* 19:1668–1676
- Ali I, Asim M, Khan TA (2013) Arsenite removal from water by electro-coagulation on zinc–zinc and copper–copper electrodes. *Int J Environ Sci Technol* 10:377–384
- Ali I, Al-Othman ZA, Alwarthan A, Asim M, Khan TA (2014) Removal of arsenic species from water by batch and column operations on bagasse fly ash. *Environ Sci Pollut Res* 21(5):3218–3229
- Askari M, Alimohammadi M, Dehghani MH, Emamjomeh MM, Nazmara Sh (2014) Removal of natural organic matter from aqueous solutions by electrocoagulation. *J Adv Environ Health Res* 2:91–100
- Asselin M, Drogui P, Brar SK, Benmoussa H, Blais J-F (2008) Organics removal in oily bilgewater by electrocoagulation process. *J Hazard Mater* 151:446–455
- Bagga A, Chellam Sh, Clifford DA (2008) Evaluation of iron chemical coagulation and electrocoagulation pretreatment for surface water microfiltration. *J Membr Sci* 309:82–93
- Boudjema N, Drouiche N, Abdi N, Grib H, Lounici H, Pauss A, Mameri N (2014) Treatment of Oued El Harrach River water by electrocoagulation noting the effect of the electric field on microorganisms. *J Taiwan Inst Chem Eng* 45:1564–1570
- Chellam S, Sari MA (2016) Aluminum electrocoagulation as pretreatment during microfiltration of surface water containing NOM: a review of fouling, NOM, DBP, and virus control. *J Hazard Mater* 304:490–501
- Chen G (2004) Electrochemical technologies in wastewater treatment. *Sep Purif Technol* 38:11–41
- Chou W-L, Wang C-T, Hsu C-W, Huang K-Y, Liu T-C (2010) Removal of total organic carbon from aqueous solution containing polyvinyl alcohol by electrocoagulation technology. *Desalination* 259:103–110
- Drouiche N, Aoudj S, Lounici H, Mahmoudi H, Ghaffour N, Goosen MFA (2011) Development of an empirical model for fluoride removal from photovoltaic wastewater by electrocoagulation process. *Desalin Water Treat* 29:96–102
- Dubrawski KL, Mohseni M (2013) Standardizing electrocoagulation reactor design: iron electrodes for NOM removal. *Chemosphere* 91:55–60
- Dubrawski KL, Fauvel M, Mohseni M (2013) Metal type and natural organic matter source for direct filtration electrocoagulation of drinking water. *J Hazard Mater* 244:135–141
- Emamjomeh MM, Sivakumar M (2009) Review of pollutants removed by electrocoagulation and electrocoagulation/flotation processes. *J Environ Manag* 90:1663–1679
- Gharibi H, Sowlat MH, Mahvi AH, Keshavarz M, Safari MH, Lotfi S, Bahram Abadi M, Alijanzadeh A (2012) Performance evaluation of a bipolar electrolysis/electrocoagulation (EL/EC) reactor to enhance the sludge dewaterability. *Chemosphere* 90:1487–1494
- Ghernaout D, Ghernaout B, Saiba A, Boucherit A, Kellil A (2009) Removal of humic acids by continuous electromagnetic treatment followed by electrocoagulation in batch using aluminum electrodes. *Desalination* 239:295–308
- İrdemez Ş, Demircioğlu N, Yıldız YS, Bingül Z (2006) The effects of current density and phosphate concentration on phosphate removal from wastewater by electrocoagulation using aluminum and iron plate electrodes. *Sep Purif Technol* 52:218–223
- Jiang J-Q, Graham N, André C, Kelsal GH, Brandon N (2002) Laboratory study of electro-coagulation–flotation for water treatment. *Water Res* 36:4064–4078
- Kashefialasl M, Khosravi M, Marandi R, Seyyedi K (2006) Treatment of dye solution containing colored index Acid Yellow 36 by electrocoagulation using iron electrodes. *Int J Environ Sci Technol* 2:365–371
- Koby M, Can OT, Bayramoglu M (2003) Treatment of textile wastewaters by electrocoagulation using iron and aluminum electrodes. *J Hazard Mater* 100:163–178
- Leenheer JA (2004) Comprehensive assessment of precursors, diagnosis, and reactivity to water treatment of dissolved and colloidal organic matter. *Water Sci Technol* 4:1–9
- Lemlikchi W, Khaldi S, Mecherrri MO, Lounici H, Drouiche N (2012) Degradation of disperse red 167 azo dye by bipolar electrocoagulation. *Sep Purif Technol* 47:1682–1688
- Matilainen A, Vieno N, Tuhkanen T (2006) Efficiency of the activated carbon filtration in the natural organic matter removal. *Environ Int* 32:324–331



- Matilainen A, Gjessing ET, Lahtinen T, Hed L, Bhatnagar A, Sillanpää M (2011) An overview of the methods used in the characterisation of natural organic matter (NOM) in relation to drinking water treatment. *Chemosphere* 83:1431–1442
- Metcalf and Eddy (2004) *Wastewater engineering treatment and reuse*, 4th edn. McGraw-Hill Inc, New York, p 13
- Mohora E, Rončević S, Agbaba J, Tubić A, Mitić M, Klašnja M, Dalmacija B (2014) Removal of arsenic from groundwater rich in natural organic matter (NOM) by continuous electrocoagulation/flocculation (ECF). *Sep Purif Technol* 136:150–156
- Vosoughi Niri M, Mahvi AH, Alimohammadi M, Shirmardi M, Golastanifar H, Mohammadi MJ, Naeimabadi A, Khishdost M (2015) Removal of natural organic matter (NOM) from aqueous solution by NaCl and surfactant-modified clinoptilolite. *J Water Health* 13:394–405
- Yetilmezsoy K, Ilhan F, Sapci-Zengin Z, Sakar S, Gonullu MT (2009) Decolorization and COD reduction of UASB pretreated poultry manure wastewater by electrocoagulation process: a post-treatment study. *J Hazard Mater* 162:120–132
- Yildiz YŞ, Koparal AS, İrdemez Ş, Keskinler B (2007) Electrocoagulation of synthetically prepared waters containing high concentration of NOM using iron cast electrodes. *J Hazard Mater* 139:373–380
- Zodi S, Louvet J-N, Michon C, Potier O, Pons M-N, Lapicque F, Leclerc J-P (2011) Electrocoagulation as a tertiary treatment for paper mill wastewater: removal of non-biodegradable organic pollution and arsenic. *Sep Purif Technol* 81:62–68
- Zongo I, Leclerc J-P, Maïga HA, Wéthé J, Lapicque F (2009a) Removal of hexavalent chromium from industrial wastewater by electrocoagulation: a comprehensive comparison of aluminum and iron electrodes. *Sep Purif Technol* 66:159–166
- Zongo I, Maïga AH, Wéthé J, Valentin G, Leclerc J-P, Paternotte G, Lapicque F (2009b) Electrocoagulation for the treatment of textile wastewaters with Al or Fe electrodes: compared variations of COD levels, turbidity and absorbance. *J Hazard Mater* 169:70–76

