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A study on the removal of sulfate and nitrate from the wet scrubber wastewater using electrocoagulation

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ABSTRACT

Wet scrubber is widely used to remove pollutants from a furnace flue gas or from other gas streams. However, high cost for the treatment or exchange of contaminated washed water is one of the problems. Electrocoagulation (EC) is an electrochemical method for treating polluted water, whereby sacrificial anodes dissolve to produce active coagulant precursors in the solution. This technology can be used for the removal of color, anionic contaminants, and colloidal particles. The goal of the present study was evaluation of EC process for treatment of wet scrubber wastewater and optimization of the process. In this study, the influence of electrode material and electrode distance on removal efficiency (%) of nitrate, sulfate, and total suspended solids (TSS) was investigated with synthetic wastewater. Using an Al electrode, 60.6% of nitrate, 50.0% of sulfate, and 96.8% of TSS were removed by EC treatment within 30 min. In case of Fe electrode, 69.7% of nitrate, 75.0% of sulfate, and 98.8% of TSS were removed. The treatment efficiency for real wet scrubber wastewater was evaluated with Fe electrode. After 180 min of EC treatment, 50% of color, 40% of nitrate, 40% of sulfate, and 95% of TSS were removed, respectively. All these results give clear evidence that EC process can effectively reduce the TSS, nitrate, and sulfate ions from wet scrubber wastewater.

Keywords: Electrochemical treatment; Electrocoagulation; Wet scrubber wastewater; Sacrificial electrode

1. Introduction

The electrochemical treatment is considered as one of the advanced oxidation processes, potentially a powerful method of pollution control, offering high removal efficiencies in compact reactors with simple equipment for control and operation of the process. The treatment process would be relatively non-specific, that is, applicable to a variety of contaminants but capable of preventing the production of unwanted

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side-products. In the recent years, there has been a growing interest in the treatment of industrial effluents by electrochemical methods as an alternative to traditional biological treatments [1].

During the last few years, the electrochemical methods have been developed and used as alternative options for the remediation of water and wastewaters mainly due to their advantages, e.g. environmental compatibility, versatility, high energy efficiency, amenability of automation and safety, and cost effectiveness. Electrochemical methods include electrocoagulation (EC), electrooxidation, and electroreduction. EC is a water treatment process whereby an electric current is applied across metal plates to remove various contaminants from water. Heavy metals (ions) and colloids (organics and inorganics) are primarily held in solution by electrical charges and particle size [2]. In view of the advantages of EC, relative small EC reactors could be implemented for localized wastewater treatment plants [3]. EC plants are able to treat water from different sources i.e. pond water to domestic wastewater [4]. In addition, the formed EC flocs are acid resistant and more stable than those formed by chemical coagulation. Gas bubbles generated at electrode also make the separation of EC flocs easier than chemically coagulated flocs. During EC treatment, several chemical and physical reactions occur at the surfaces and the interfaces [5]. Another advantage of this process is that it is easier to treat wastewaters having low temperature and low turbidity than other process [6,7].

Under electrical current, aluminum and iron anodes dissolve and Al^{3+} and Fe^{2+} ions are formed [7,8]. At the same time, hydrogen gas and hydroxide ions are released on the cathode [8]. The electrophoretic motion carries the hydroxide anions towards the anode, where they form ion-pairs with metal cations. Ion-pair formation generates polymeric iron and aluminum hydroxides i.e. coagulation agents [6].

The mechanisms of removal of ions by EC will be explained with two specific examples involving aluminum and iron, since these two metals have been extensively used to clarify wastewater [9–13].

The electrolytic dissolution of the aluminum anode produces the cationic monomeric species, such as Al^{3+} and $Al(OH)_2^+$ at low pH, which at appropriate pH values are transformed initially into $Al(OH)_3$ and finally polymerized to $Al_n(OH)_{3n}$ according to the following reactions:

Anode:

$$Al_{(s)} \rightarrow Al^{3+} + 3e^- \quad (1)$$

Cathode:

$$3H_2O + 3e^- \rightarrow \left(\frac{3}{2}\right)H_2 + 3OH^- \quad (2)$$

Overall:

$$Al^{3+}_{(aq)} + 3H_2O \rightarrow Al(OH)_3 \downarrow_{(s)} + 3H^+ \quad (3)$$

Aluminum hydroxide is polymerized as follows,

$$nAl(OH)_3 \downarrow_{(s)} \rightarrow Al_n(OH)_{3n} \downarrow_{(s)} \quad (4)$$

These gelatinous-charged hydroxo cationic complexes can effectively remove pollutants by adsorption to produce charge neutralization, and by enmeshment in a precipitate [7].

Upon oxidation in an electrolytic system, Iron produces $Fe(OH)_n$, where n is 2 or 3. Two mechanisms have been proposed for the production of $Fe(OH)_n$ [14–16].

(1) Mechanism 1

Anode:

$$4Fe_{(s)} \rightarrow 4Fe^{2+}_{(aq)} + 8e^- \quad (5)$$

$$4Fe^{2+}_{(aq)} + 10H_2O + O_{2(g)} \rightarrow 4Fe(OH)_{3(s)} + 8H^+_{(aq)} \quad (6)$$

Cathode:

$$8H^+_{(aq)} + 8e^- \rightarrow 4H_{2(g)} \quad (7)$$

Overall:

$$4Fe_{(s)} + 10H_2O_{(l)} + O_{2(g)} \rightarrow 4Fe(OH)_{3(s)} + 4H_{2(g)} \quad (8)$$

(2) Mechanism 2

Anode:

$$Fe_{(s)} \rightarrow Fe^{2+}_{(aq)} + 2e^- \quad (9)$$

$$Fe^{2+}_{(aq)} + 2OH^- \rightarrow Fe(OH)_{2(s)} \quad (10)$$

Cathode:

$$2H_2O + 2e^- \rightarrow H_2 + 2OH^- \tag{11}$$

Overall:

$$Fe_{(s)} + 2H_2O_{(l)} \rightarrow Fe(OH)_{2(s)} + H_{2(g)} \tag{12}$$

The emissions from ships, which are engaged in international trade in the seas contain much hazardous substances, such as SOx and NOx, which are generated through fuel combustion process to operate the vessels. The prevention of the vessel source pollution is formed by IMO and UNCLOS. As the air pollution prevention is formulated by the Kyoto Protocol, IMO adopted the "Regulations for the Prevention of Air Pollution from Ships" as Annex VI of MARPOL. The regulation of air pollution from vessel engines will take effect on 2016.

Wet scrubber is widely used to remove pollutants from a furnace flue gas or from other gas streams. Wet scrubber system can be an alternative method to satisfy the IMO's regulation. On the other hand, high cost for the treatment or exchange of contaminated washed water is one of the problems. In the wet scrubber system, the SOx and NOx in the flue gas are dissolved into the scrubbing liquid, and converted into sulfate and nitrate ions, this being so, wet scrubber wastewater has very high level of sulfate and nitrate.

Electrochemical coagulation was investigated to treat nonbiodegradable wastewater, such as leachate [13], bilgewater [17], mechanical polishing wastewater [18], and mine drainage [9]. Ilhan et al. [13] investigated the treatment of leachate by EC process. About 56% of COD was removed using Al electrode and 35% of COD was removed using Fe electrode with 30 min of treatment in the batch process [13]. Asselin et al. [17] studied organics removal in bilgewater by EC process. They reported that 1.5 A of electric current and 60 min of treatment time showed best performances. Under these conditions, 93.0 and 95.6% of biochemical oxygen demand and O&G (oil and grease) was removed, respectively. There was no previous study about treatment of wet scrubber wastewater with EC process.

The main aim of this study was to investigate the treatment performance of a wet scrubber wastewater by EC treatment. The influence of electrode material and distance on removal efficiency (%) of nitrate, sulfate, and total suspended solids (TSS) was investigated. Al and Fe electrodes were used to evaluate the removal efficiency (%) of pollutant. The removal efficiency (%) of pollutants was investigated as a function of time at the different electrode distances.

2. Materials and methods

2.1. Experimental method

The volume of wastewater used in EC was 2L. Tests were conducted using a DC power supply (Unicorn tech., UDP-1501) in constant current mode. The EC reactor consists of two electrode plates which have 55 cm^2 of surface area. Aluminum and iron plates were tested in this study. The electrode distance was adjusted using the slits above the EC reactor. Fig. 1 shows the schematic diagram of the EC system used in this study.

In the lab-scale test, synthetic wastewater containing TSS, nitrate, and sulfate was tested to evaluate removal efficiency and optimize the operation parameter. The synthetic wastewater used in this study was adjusted to 250 mg/l of TSS, 96 mg/l of sulfate, and 301.1 mg/l of nitrate. 421.2 g of sodium sulfate (Na_2SO_4) and sodium nitrate ($NaNO_3$) were obtained from Sigma-Aldrich (USA), kaolin was obtained from Samchun Chemicals (Korea). 173.1 mg of Na_2SO_4, 421.2 mg of $NaNO_3$, and 250 mg of Kaolin were dissolved in distilled water.

Removal of pollutants by EC treatment was also tested with real wet scrubber wastewater. Wet scrubber wastewater was collected from the wet scrubber system which was used for treatment of SOx and NOx in flue gas generated from 100-horsepower vessel engine. The composition of wet scrubber wastewater is highly variable and depends on the type of flue gas and scrubber process condition used. The main characteristics of wet scrubber wastewater used in this study are presented in Table 1.

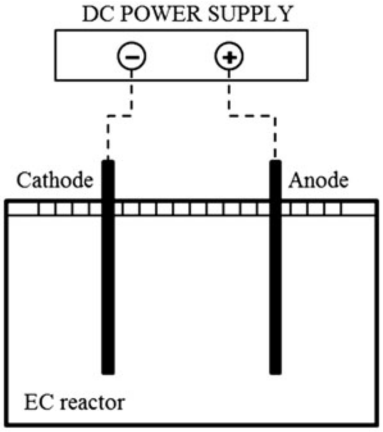

Fig. 1. Schematic diagram of the EC system for wet scrubber wastewater treatment.

Table 1
Characteristics of wet scrubber wastewater

Parameter	Content
pH	10.08
Color (CU)	455
TSS (mg/l)	10,790
Sulfate (mg/l)	2,650
Nitrate (mg/l)	4,295.7

2.2. Analytical method

Generally, wet scrubber wastewater contains high concentration of TSS, color, nitrate, and sulfate. Because of that, the electrochemical processes can be evaluated by removal efficiency (%) of inorganic pollutants. In this study, removal efficiency (%) of TSS, color, nitrate, and sulfate were tested in the lab-scale test.

Experimental analysis was made according to the following method: TSS was determined by gravimetric method (EPA method 160.2). Sulfate ions were determined by turbidimetric method (EPA method 9038), while nitrate ions were determined by chromotropic acid method (Hach method 10020). Absorbances of the samples were measured by DR5000 spectrophotometer (Hach, wavelength range 340–900 nm). Color was analyzed with 2100AN color meter (Hach, 455 nm). All the experiments were performed in the batch mode at room temperature, and the pH levels in the reactors were not controlled.

3. Results and discussion

3.1. Removal of TSS in the lab-scale EC treatment system

Fig. 2 shows the removal efficiency (%) of TSS compared with time when Al and Fe electrodes were used. Above 95% of TSS was removed within 20 min in both the cases. The TSS removal efficiency (%) increased rapidly over the first 5 min of the treatment and then increased slightly between 5 and 20 min followed by a steady state until the end of experiment. There was no significant difference in electrode distance.

In Muruganananthan's study [19], colloidal size solids are coagulated within 600 s and the size of the coagulum remains almost constant thereafter. Mean diameter of the solids increased to 34 μm with iron as anode and 61 μm with the use of aluminum as anode. This clearly suggests the coagulation of colloidal solids was due to the presence of Al^{3+} and Fe^{3+} ions released from the anode. These ions are generally preferred in the coagulation process because of their multivalent character and low solubility ($k_s = 10^{-32.7}$) of their hydroxides. Al or Fe ions dissolved from anode form a range of coagulant species that destabilize and aggregate the suspended solid particles and precipitates. Thus the suspended solids were simultaneously coagulated and floated effectively in the presence of soluble anodes [19].

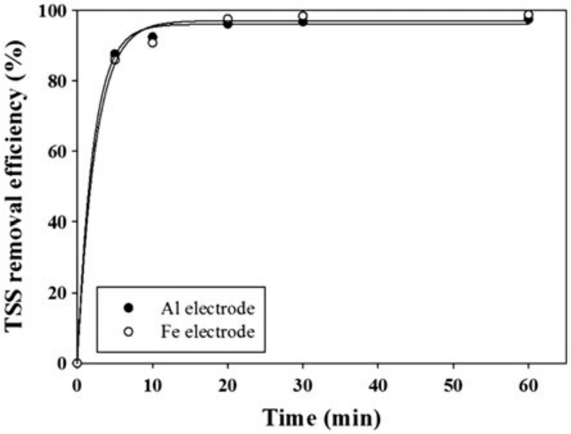

Fig. 2. Effect of electrode substance on the removal efficiency (%) of TSS with time in EC treatment (current density: 180 A/m^2, initial concentration: 250 mg/l, electrode distance: 5 cm, the electrode used in each treatment is given in parentheses).

3.2. Removal of nitrate in the lab-scale EC treatment system

Fig. 3 shows the effect of electrode distance in EC treatment system when using Al and Fe electrodes. In both the cases, 5 cm of electrode distance shows best performance, and it means EC treatment was more efficient under shorter distance between anode and cathode. This is due to the fact that the movement of ions get slower by increasing the electrode distance. This leads to a decrease in the electrostatic attraction, and results in the less formation of flocs needed to coagulate of the pollutant [20].

When using Al electrode, at 5 cm of electrode distance, the maximum removal efficiency (%) was 77.9%, and it took approximately 40 min to achieve equilibrium. On the other hand, about 85.9% of nitrate was removed when Fe electrode was used at the same condition, and it took 20 min to achieve equilibrium. Fe electrode is more effective in removing nitrate ions from the wastewater. The reduction of NO_3^- to N_2 gas is possible during the EC process and nitrate removal can occur with precipitation of metal hydroxide [21,22]. According to the research of Ghanim and Ajjam [23], The Fe^{2+}cation hydrolyzes in water to form hydroxo-amorphous polymeric complexes (flocs) with the adsorbed nitrates and can be precipitated from water Eq. (13).

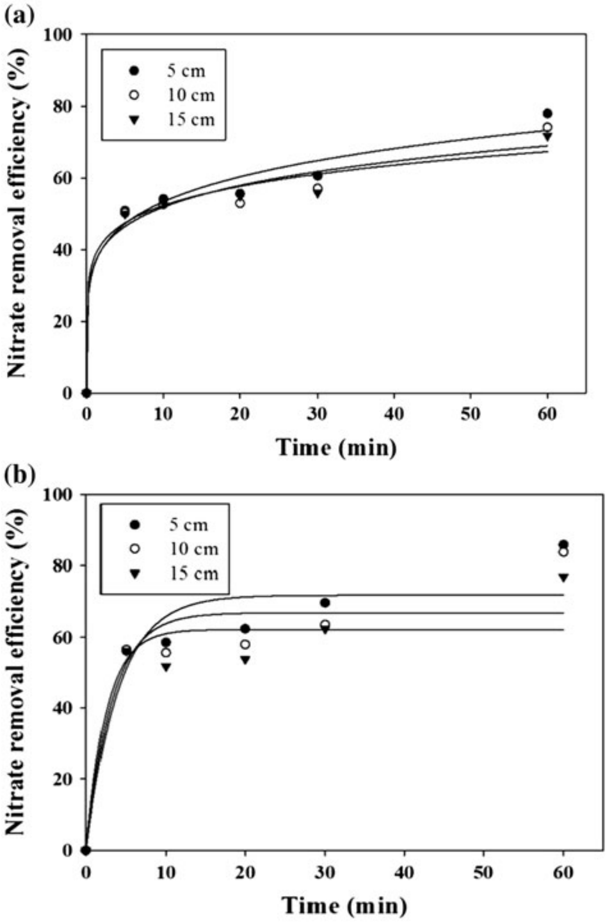

Fig. 3. Effect of electrode distance on the removal efficiency (%) of nitrate with time in EC treatment; (a) Al electrode and (b) Fe electrode (current density: 180 A/m^2, initial concentration: 301.1 mg/l, the electrode distance set in each treatment is given in parentheses).

$$ {}_n\mathrm{Fe(OH)}_{3(s)} + \mathrm{NO}_{3(aq)}^{-} \rightarrow [\mathrm{Fe}_n(\mathrm{OH})_{3n} \cdot \mathrm{NO}_3^{-}]_{(s)} \qquad (13) $$

3.3. Removal of sulfate in the lab-scale EC treatment system

The effect of electrode distance on the removal of sulfate is shown in Fig. 4(a) and (b). Fig. 4(a) and (b) represent the results that were obtained during the EC treatment using Al and Fe electrodes. When using Al electrode, 77.1, 66.7, and 64.6% of sulfate ions were removed at 5, 10, and 15 cm of electrode distance, respectively. On the other hand, Fe electrode shows 87.5, 85.4, and 83.3% of sulfate removal efficiency (%) at the same conditions. When using Fe electrode, the maximum removal efficiency (%) was about 87.5% at 5 cm of electrode distance and reaches equilibrium in 60 min. Similar result was reported by Drouiche et al. [18], maximum sulfate removed was about 80% in the study.

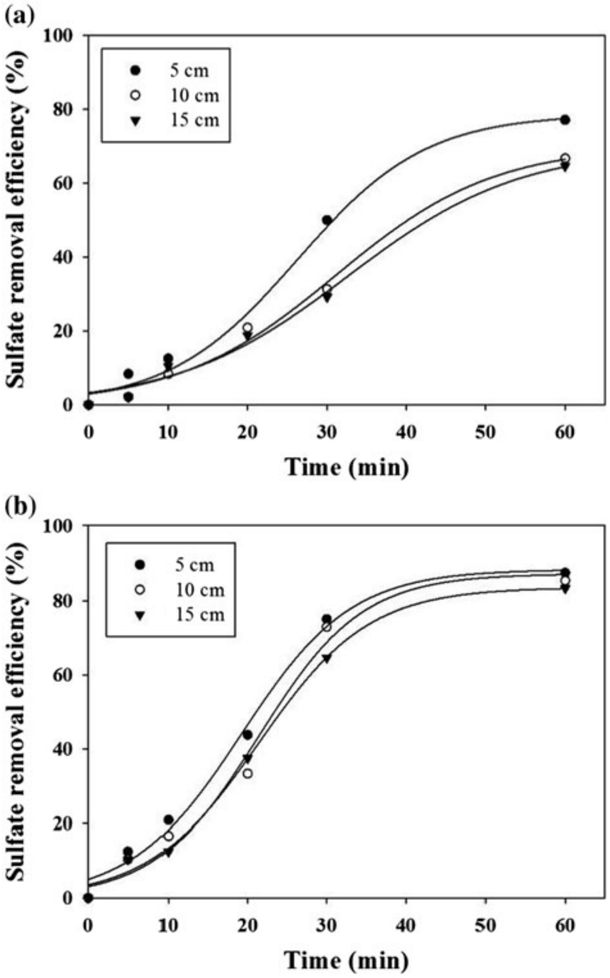

Fig. 4. Effect of electrode distance on the removal efficiency (%) of sulfate with time in EC treatment; (a) Al electrode and (b) Fe electrode (current density: 180 A/m^2, initial concentration: 96 mg/l, the electrode distance set in each treatment is given in parentheses).

As shown in Fig. 4(a) the shorter distance between anode and cathode resulted in decrease in time required to achieve equilibrium concentration. Similar to the previous case, shorter electrode distance was more efficient to remove sulfate ions. The more sulfate ions were removed in the condition of shorter electrode distance.

3.4. Comparison of removal efficiency (%) of Al and Fe electrodes

It was found that the time taken to reach equilibrium for removal of nitrate was much less than sulfate in both the electrodes i.e. 10 min for nitrate and 50 min for sulfate (Fig. 5). It can be explained by the fact that reactivity of nitrate is higher than sulfate. One possible explanation for this observation is the competition among anions that were to be adsorbed onto the growing metal hydroxide precipitates [24], this effect has been previously studied in several investigations [12,25–27].

The comparison of pollutants removal efficiencies (%) by electrode substance is presented in Fig. 6. After 30 min of treatment, 60.6% of nitrate, 50.0% of sulfate, and 96.8% of TSS were removed by EC treatment using Al electrode. In case of Fe electrode, 69.7% of nitrate, 75.0% of sulfate, and 98.8% of TSS were removed. The TSS removal efficiencies (%) were outstanding in both Al and Fe electrodes. Except for the case of TSS, the EC treatment using Fe electrode was more efficient in reduction of pollutants. It followed same characteristics as reported by Mahajan et al. [28] and Katal and Pahlavanzadeh [29]. In the studies, Fe electrode showed higher removal efficiency than Al electrode. It can be explained by the difference of z-potential of flocs generated from each electrode materials. In the study of Lacasa et al. [24], during the EC treatment, z-potential was increased for iron, and was maintained at approximately zero for Al. The anionic pollutants can be absorbed onto the surface of the iron hydroxide, explaining the resulting positive charge.

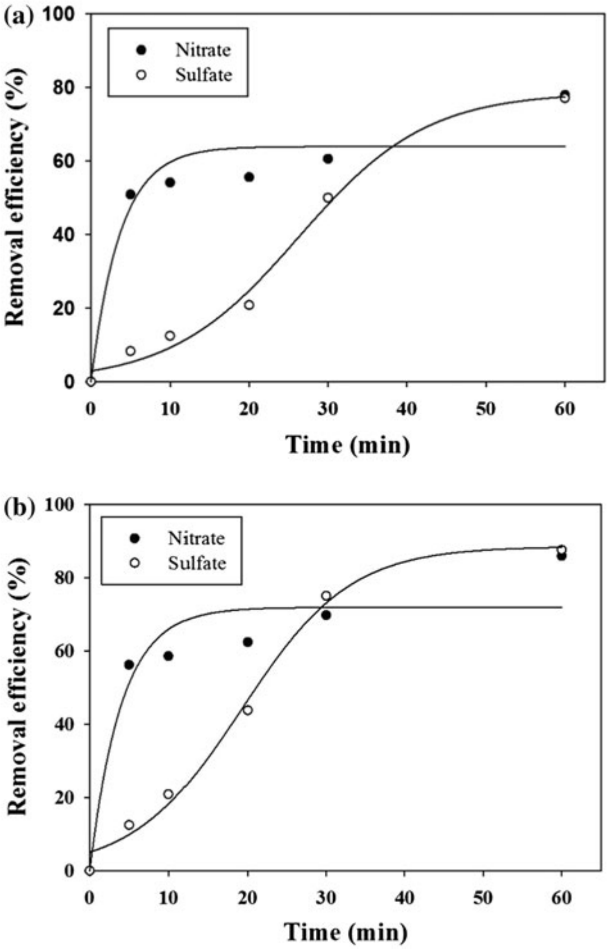

Fig. 5. Removal efficiency (%) of nitrate and sulfate with time in EC treatment; (a) Al electrode and (b) Fe electrode (current density: 180 A/m^2, initial concentration: 301.1 mg/l of nitrate, 96 mg/l of sulfate, electrode distance: 5 cm).

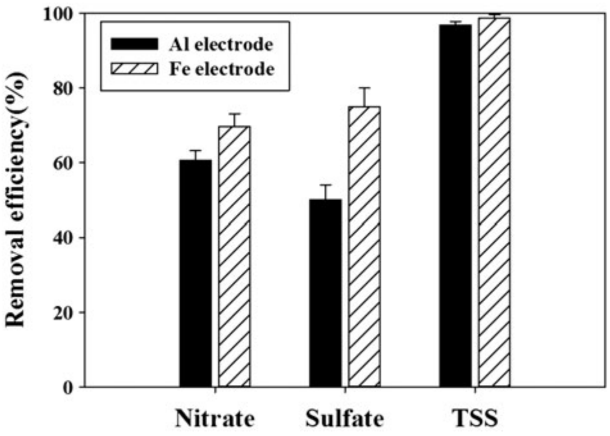

Fig. 6. Removal efficiency (%) of nitrate, sulfate, and TSS in EC treatment using Al and Fe electrodes (current density: 180 A/m^2, electrode distance: 5 cm, EC treatment time: 30 min, initial concentration: 301.1 mg/l of nitrate, 96 mg/l of sulfate, and 250 mg/l of TSS).

3.5. Application for removal of pollutants from real wet scrubber wastewater

Pollutants removal by EC was also studied with real wet scrubber wastewater. The removal

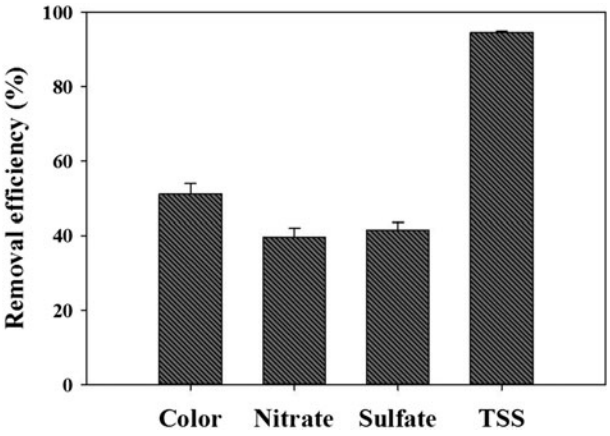

Fig. 7. Removal efficiency (%) of color, nitrate, sulfate, and TSS in EC system for wet scrubber wastewater treatment; (current density: 180 A/m^2, electrode distance: 5 cm, EC treatment time: 180 min).

efficiency (%) of color, nitrate, sulfate, and TSS were tested using Fe electrode (Fig. 7). The removal efficiency (%) of color was 51.2%. Nitrate and sulfate were reduced to 39.7 and 41.5%, respectively, after 180 min of EC treatment, and 94.5% of TSS was removed by EC treatment.

As reported by the previous studies, the real wastewater with high concentration of pollutants needs longer treatment time. Kumar and Goel [30] studied the EC treatment on synthetic wastewater, they reported that the removal efficiency (%) reached equilibrium after 180 min. In the study by Drouiche et al. [18], 300 min of treatment time was needed to treat chemical-mechanical polishing (CMP) wastewater.

4. Conclusions

In this study, the influence of electrode material and distance on removal efficiency (%) of nitrate, sulfate, and TSS was investigated. Using an Al electrode, 60.6% of nitrate, 50.0% of sulfate, and 96.8% of TSS was removed by EC treatment within 30 min. In case of Fe electrode, 69.7% of nitrate, 75.0% of sulfate, and 98.8% of TSS were removed. The more effective removal capacity was achieved when Fe electrode was used. It was higher than Al electrode about 9% for nitrate and 25% for sulfate. Pollutants removal by EC was also studied with wet scrubber wastewater. After 180 min treatment, 50% of color, 40% of nitrate, 40% of sulfate, and 95% of TSS were removed by EC treatment system.

Further research is needed to yield a more complete understanding of the mechanism of the EC process. And parameters for EC treatment of wet scrubber wastewater, such as the effects of pH, temperature, alkalinity, and pressure should be considered.

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