

Electrocoagulation Treatment of Wet Flue-Gas Desulfurization Wastewater Using Iron-Based Electrodes: Influence of Operating Parameters and Optimization

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The aim of this study was to evaluate the performance of electrocoagulation (EC) in treating wastewater from wet flue-gas desulfurization (WFGD). The effects of initial pH, electrolysis time, applied voltage and inter-electrode distance on turbidity and on the removal of heavy metals were examined. The experimental results revealed that the initial pH of 9.0 was optimal for the treatment. There were decreases in the turbidity (99.2%), Cu²⁺ (99.3%), Fe²⁺ (99.9%), Ni²⁺ (98.9%) and Zn²⁺ (97.8%) with an electrolysis time of 40 min at an applied voltage of 20 V and an inter-electrode distance of 4 cm in the presence of Fe-Fe electrodes. By comparison, EC required a lower initial pH than chemical coagulation (FeSO₄) to achieve an improved removal efficiency for heavy metals and turbidity. The percentage of Fe_b species, zeta potential and floc size in the EC process correlated well to the removal of heavy metals and turbidity. The process was also determined to be effective for a reduction in conductivity (75.6% removal).

Keywords: Electrocoagulation, wastewater of WFGD, heavy metals, turbidity

1. INTRODUCTION

Flue gas desulfurization (FGD) is used in many fossil fuel power plants to reduce emissions of sulfur dioxide. Among various FGD technologies, wet flue-gas desulfurization (WFGD) is the most popular [1, 2]. According to the Development Report on China Desulfurization and Denitration Industries in 2017, 91% of all FGD projects in China have adopted this technology. Moreover, WFGD accounts for an average of 85% of all FGD projects worldwide [3]. Depending on the resources of the power plant and the characteristics of the by-products, WFGD effluents contain a very complex matrix

of heavy metals, which do not transform easily into harmless forms and large amounts of suspended particulate matter, Cl^- and SO_4^{2-} [4]. The concentrations of these substances in wastewater may be higher than acceptable standard levels. WFGD wastewater is typically treated by chemical precipitation methods in which $\text{Ca}(\text{OH})_2$ or NaOH are added to increase the pH of the wastewater, causing heavy metals to precipitate as hydroxides [5, 6]. This addition of chemicals makes the process less attractive than other processes since it increases the net dissolved constituents of the wastewater, which may make it impossible to reuse in other applications. Compared with chemical processes, EC appears to be a cleaner and more efficient technology for separating these pollutants from their aqueous phase, removing pollutants by means of two processes: electrocoagulation and electroflotation [7]. In large-scale and industrial EC processes, the addition of a chloride salt is an effective and widely used method for depassivation of electrodes [8]. In this study, the initial chloride concentration was approximately 23000 mg/L (Table 1), which was sufficient to improve the conductivity of the solution. Some authors [9] have reported a noticeable affinity of sulfate species for aluminum to form complexes, passivating the anodic surface, but sulfate has no complexation affinity with iron, and it does not inhibit oxidation of iron anodes. For this reason, given concentrations of sulfate approximately 3800 mg/L, we chose iron as both anode and cathode. The purpose of the present study was to investigate the efficacy of EC for the removal of heavy metals and turbidity from an actual WFGD wastewater. The effects of operating parameters such as pH, electrolysis time, applied voltage and electrode distance were tested and optimized.

2. EXPERIMENTAL

WFGD wastewater was collected from a 600 MW coal-fired power plant in Jiangsu province, China. The main wastewater characteristics are shown in Table 1. According to the Discharge Standard of Wastewater from Limestone-gypsum Flue Gas Desulfurization System in Fossil Fuel Power Plants in China (DL/T 997-2006), the concentrations of Zn^{2+} and Ni^{2+} both exceed standard limitations by 67 % and 32 %, respectively. The concentrations of Fe^{2+} and Cu^{2+} are also high.

Table 1. Measured values of main parameters in raw WFGD wastewater

Parameters	Measured values
pH	6.92
Turbidity	488 NTU
Conductivity	14.33 mS cm^{-1}
Zn^{2+}	3.34 mg L^{-1}
Ni^{2+}	1.32 mg L^{-1}
Cu^{2+}	0.57 mg L^{-1}
Fe^{2+}	4.11 mg L^{-1}
Hg^{2+}	2.09 μg L^{-1}
$\text{As}^{3+}+\text{As}^{5+}$	20.01 μg L^{-1}

Cl ⁻	22962 mg L ⁻¹
F ⁻	-
SO ₄ ²⁻	3728 mg L ⁻¹
SO ₃ ²⁻	439 mg L ⁻¹
Ca ²⁺	4380 mg L ⁻¹
Mg ²⁺	1493 mg L ⁻¹

The EC experiments were performed in a $\phi 15 \times 20$ cm cylindrical glass reactor. A pair of $15 \text{ cm} \times 4 \text{ cm} \times 0.3 \text{ cm}$ iron sheets were employed as anode and cathode. Before the EC experiments, all electrode sheets were soaked in dilute HCl for 12 h to remove oxides on their surfaces and then were polished using abrasive paper. To optimize experimental conditions, lime was used to adjust the pH of the wastewater. The distance between the anode and cathode was varied from 1 cm and 6 cm, the voltage of EC was tested from 5 V to 25 V and was provided by a constant voltage DC power supply (RXN-305D, China), and the electrolysis time was varied from 10 to 80 min. After the EC experiments, followed by 30 min standing, The wastewater was resampled. The concentrations of Zn²⁺, Ni²⁺, Cu²⁺ and Fe²⁺ were determined by inductively coupled plasma spectrometry (ICP-OES, Varian 720-ES, USA) device. The removal efficiency of heavy metals was calculated according to Equation (1):

$$\eta = \frac{C_1 - C_2}{C_1} \times 100\% \quad (1)$$

where C_1 and C_2 were heavy metals concentrations in wastewater before and after the EC experiment. The turbidity and conductivity of samples were analysed using a turbidity meter (WZS-185, INESA) and a conductivity meter (DDS-307, INESA). The samples' pH values were continuously monitored by a pH instrument (FHS-29A, INESA). Prior to zeta potential analysis, the samples were filtered through a $0.45 \mu\text{m}$ filter. Approximately 10 mL of filtered sample was taken to measure the zeta potential on a Zetasizer 3000 HSa (Malvern Instruments, UK) at room temperature. The ferron method was used to determine Fe floc speciation. According to this method, Fe floc species can be generally divided into three types: (1) Fe_a, monomeric species of Fe (III) are the species that react with ferron in the first 30 s, determined by the 600 nm visible light absorbance at 30 s; (2) Fe_b, polymeric species of Fe (III) that react with ferron between 30 s and 180 min, determined by the 600 nm visible light absorbance at 180 min; and (3) Fe_c, species of Fe (III) that fail to react with ferron or react very slowly. Fe_c is calculated by subtracting Fe_a and Fe_b from the known total Fe concentration [10]. The size and fractal dimension analysis in the EC process were determined using a Mastersizer 2000 (MalvernMalvernInstrument, UK). In this study, floc sizes were chiefly expressed using the weight-average floc diameter (D_w) and described by Equation (2) [11].

$$D_w = \sum \left(\frac{N_i \times D_i^2}{N_i \times D_i} \right) \quad (2)$$

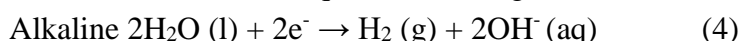
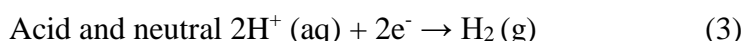
N_i =number of flocs having diameter D_i .

3. RESULTS AND DISCUSSION

3.1 Effect of initial pH

In wastewater treatment, pH has an important influence on the chemical coagulation (CC) process and on the EC process, which relies on existing forms of heavy metals in aqueous solution and on newly formed hydroxides from the coagulant and the sacrificial anode. In this study, the existing forms of heavy metals in solution were simulated using Visual MINTEQ version 3.1 and the results are shown in Figure 1. In the concentration range studied, and when the pH was greater than 10.0, Cu^{2+} , Fe^{2+} , Ni^{2+} and Zn^{2+} in aqueous solution were all presented mainly as hydroxide precipitates. For this reason, we compared the effects of initial pH on CC and EC. We first optimized the treatment of WFGD wastewater by CC. Figure 2 shows the effect of initial pH on the reduction of heavy metals, turbidity and conductivity by CC. The turbidity slightly and continuously decreased until the pH reached 10.0 and then exhibited a large increase. The removal efficiency for Cu^{2+} , Fe^{2+} , Ni^{2+} and Zn^{2+} gradually increased at higher pH values with an optimum pH range of 10.0–11.0 for all. It could be seen that pH had an important influence on CC. In contrast, Figure 3 shows the effect of initial pH on heavy metals, turbidity and conductivity removal by EC. Turbidity decreased with higher initial pH values from 7.0 to 9.0 and then stabilized. For Cu^{2+} removal, the optimum pH range was 8.0–11.0. For Fe^{2+} removal, the optimum pH range was 8.0–10.0. Zn^{2+} removal reached a maximum when pH was higher than 8.0. pH had a small influence on Ni^{2+} removal in the 7.0–12.0 range. The removal efficiency of Cu^{2+} , Fe^{2+} , Ni^{2+} and Zn^{2+} reached almost 95% when the initial pH value exceeded 8.0 and optimum removal occurred at 9.0. EC required a lower pH than CC, which might be related to the continuous production of hydroxyl ions (OH^-) and hydrogen gas (H_2) by reduction reactions on the cathode according to Equation (3) and (4). The pH of solution near the cathode gradually increased and the generation of H_2 bubble contributed to the movement of suspended substances. A large amount of free Fe^{2+} (Equation (5)) was generated in situ by electrochemical dissolution from the sacrificial anode. The generated Fe^{2+} immediately underwent hydrolysis to produce corresponding hydrates, hydroxides, polyhydroxides and polyhydroxymetallic complexes [12–14], which were responsible for trapping colloidal particles and heavy metals as they precipitated from the wastewater. Other studies reported that ferrous ions, which are known to be highly soluble and a poor coagulant, began to be oxidized to ferric ions if the pH was above 5.0, while complete oxidation only occurred at pH of approximately 8.0–9.0 [15–17]. Since the EC process continuously produces OH^- , it requires a lower initial pH than CC to maintain a better pH range.

Cathode:



Anode:



To further explore the influence of pH on EC performance, the distribution of Fe species, floc size and zeta potential were investigated. As shown in Figure 4 (a), high pH accelerates the hydrolysis

reaction of Fe^{2+} . The proportion of Fe_a species, which are monomeric species with low molecular weights, significantly decreased with increasing pH before $pH < 9$ and then slightly increased. Fe_b , referring to oligomers and polymers, reached a maximum proportion of 51.4% when pH was 9.0. Fe_c , refers to colloidal hydroxides, which noticeably increased with increasing pH. The largest floc size and the most negative zeta potential were found at the highest concentration of Fe_b at pH 9.0 as shown in Figure 4 (b). The greater the proportion of Fe_b flocs, the better the effect of the iron coagulant treatment. As in a previous report [18], Fe_b floc is regarded as the most active species responsible for coagulation. Generally, Fe_a species are considered to capture pollutants quickly, mainly by charge neutralization. The destabilized particles were absorbed by the products of metal hydrolysis, leading to incompact flocs. While Fe_b removed contaminants by bridging, adsorption and charge neutralization because of their larger molecular weight and lower positive charge [19]. By contrast with Fe_a floc, the hydrolysis process of Fe_b was much slower because they were more stable [20, 21], thus, the destabilized particles were aggregate, bridged and entrapped by the hydroxide precipitate, to form more compact flocs. This caused the higher removal efficiency for heavy metals and turbidity. The difference in hydrolysis products and floc formation might be the main reason for the difference in fractal dimension.

For both EC and CC, pH has almost no influence on lowering the conductivity of the solution, although the removal efficiency of conductivity by EC was much higher than that of CC. This means EC can remove more salts.

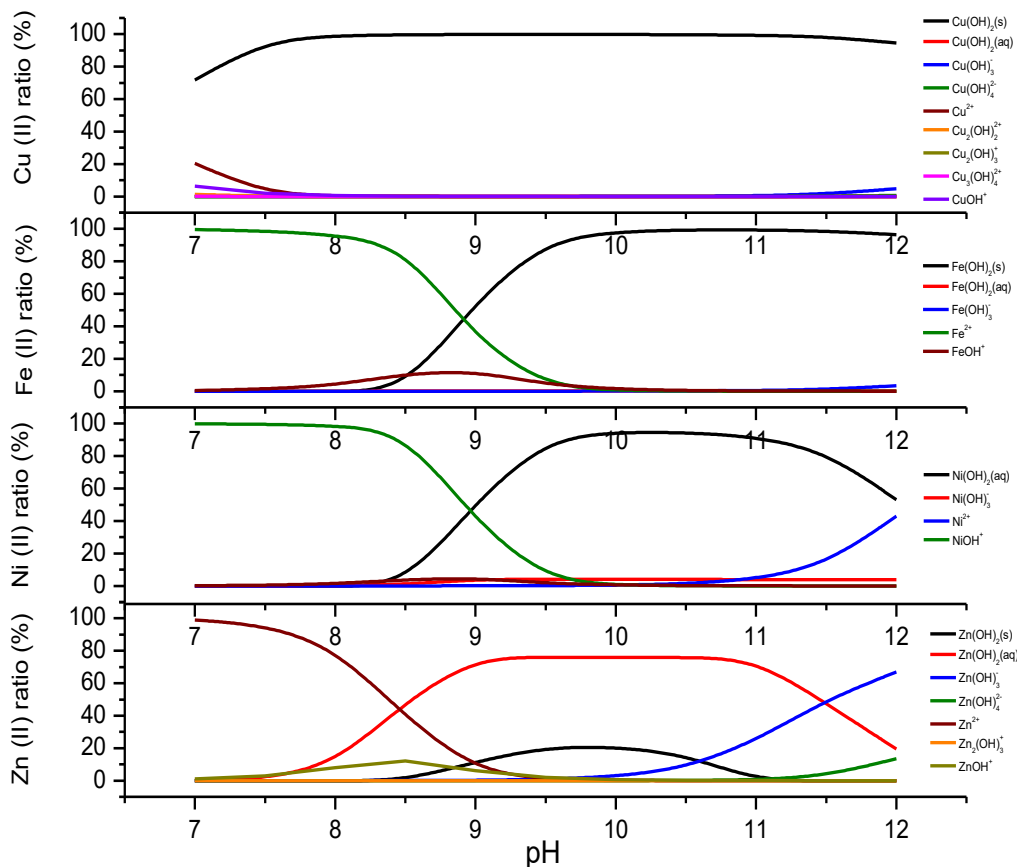


Figure 1. Distribution diagrams of copper, iron, nickel and zinc species in solutions simulated by Visual MINTEQ version 3.1.

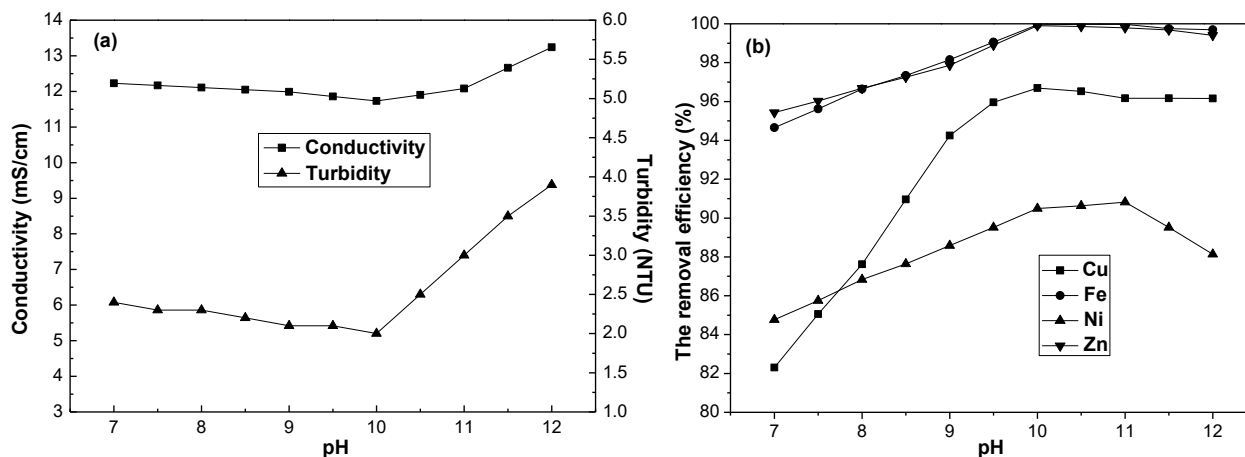


Figure 2. Effect of initial pH (a) on conductivity and turbidity and (b) on metals removal by CC: $[FeSO_4]=25$ mg/L, strong stirring intensity=200 rpm, rapid stirring time=10 min, slow stirring time=20 min and then standing 30 min.

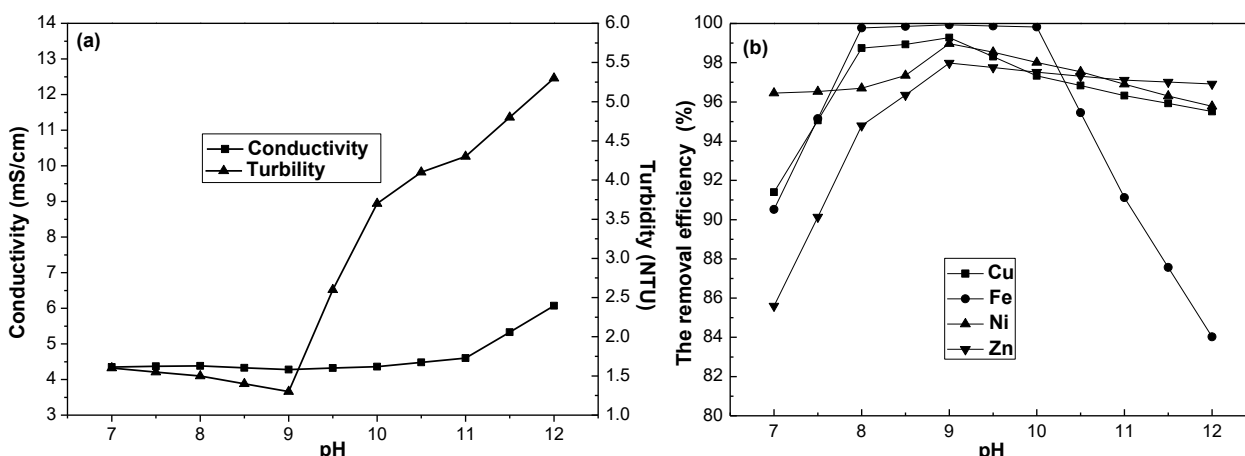


Figure 3. Effect of initial pH (a) on conductivity and turbidity and (b) on metal removal by EC: Fe--Fe, applied voltage=20 V, electrolysis time=40 min, distance between electrodes=6 cm

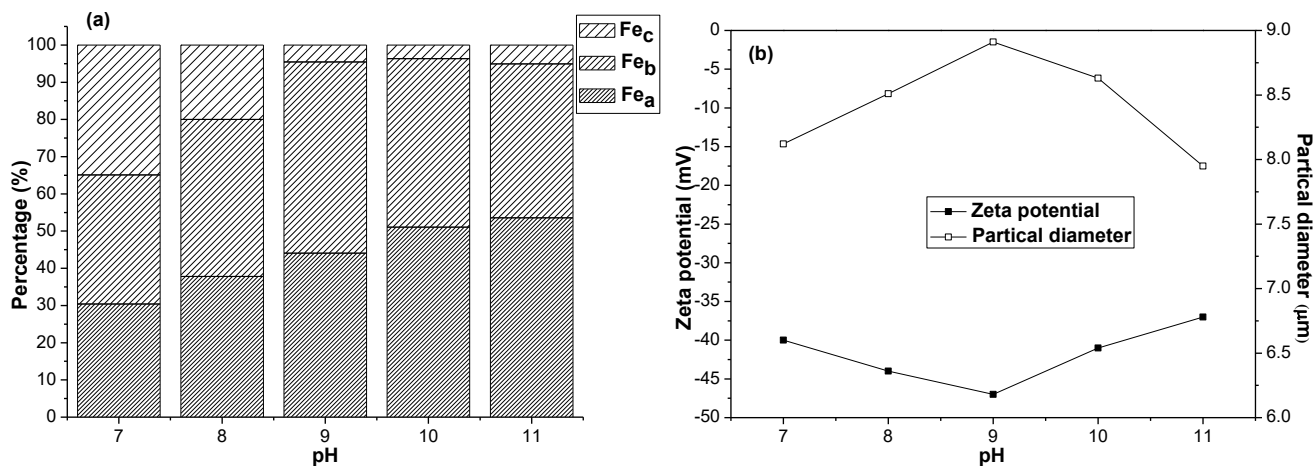


Figure 4. Effect of initial pH (a) on percentage of Fe_b and (b) on zeta potential and particle diameter by EC: Fe--Fe, applied voltage=20 V, electrolysis time=40 min, distance between electrodes=6 cm

3.2 Effect of electrolysis time

The electrolysis time was varied from 10 to 80 min. The results are reported in Figure 5 (a) and (b). From Figure 5 (a), it can be seen that turbidity gradually decreased with increases in electrolysis time until the time exceeded 40 min, after which turbidity increased. This finding might be due to the prolongation of electrolysis time leading to production of too much flocculant to affect turbidity. From Figure 5 (b), the results suggested that the removal efficiency for heavy metals increased up to an electrolysis time of 40 min. Further increases in electrolysis time did not improve the removal of the metals by EC. Similar observations were found in the study of the removal of hardness by Brahmi et al. [22], in removal of arsenic by Kumar et al. [23], and in removal of chromium by Bazrafshan et al. [24].

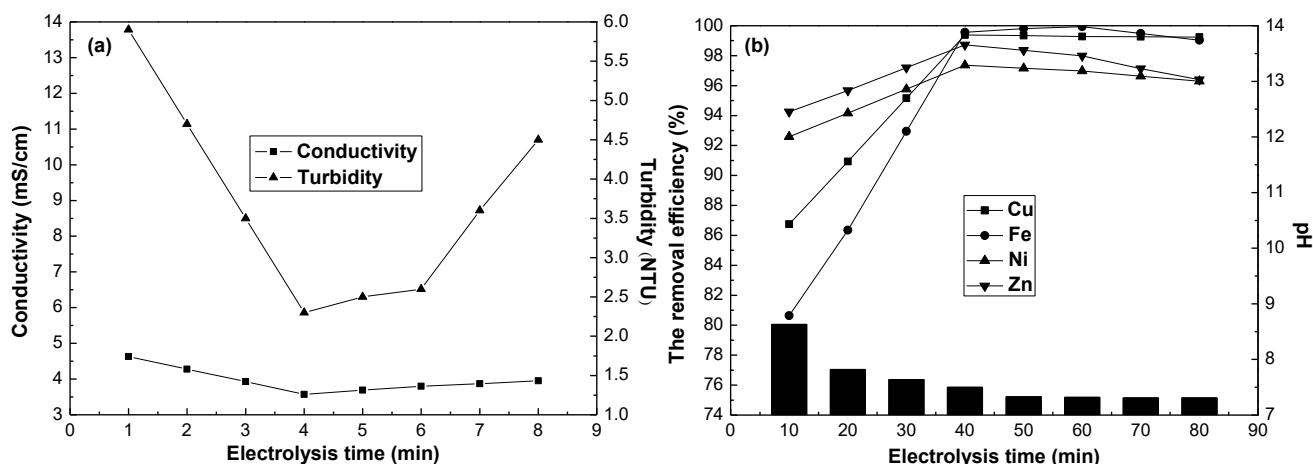


Figure 5. Effect of electrolysis time (a) on conductivity and turbidity and (b) on metals removal and pH change by EC: Fe--Fe, applied voltage=20 V, initial pH= 9.0, distance between electrodes=6 cm.

Moreover, as some reports [25, 26] have demonstrated, the use of soluble anodes changes the solution's pH during EC. Some studies concluded that pH increases as electrolysis time progresses [27, 28], while some hold the opposite opinion [22]. Escobar et al. [29] even declared that the pH increased when the initial pH was acidic and decreased when the initial pH was alkaline, meaning that the EC could stabilize the pH. In this study, with increases in electrolysis time, the pH value of the solution decreased continuously and then tended towards stability at 7.33. As previously reported [30], this finding may be explained that the observed decrease in pH was due to OH⁻ consumption in the formation of CaCO₃, Ca(OH)₂, Mg(OH)₂, and MgCO₃ or to neutralize the pH of the solution, and the stable phase of pH represents an equilibrium between the generation and consumption of OH⁻. To verify the influence of the extension of the electrolysis time on floc composition, zeta potential, and particle diameter, we extended the electrolysis time to 480 min. We found that the percentage of Fe_b floc has a small increase with electrolysis time as shown in Figure 6 (a), and electrolysis time promoted floc growth and smoothly increased the absolute value of zeta potential (Figure 6 (b)).

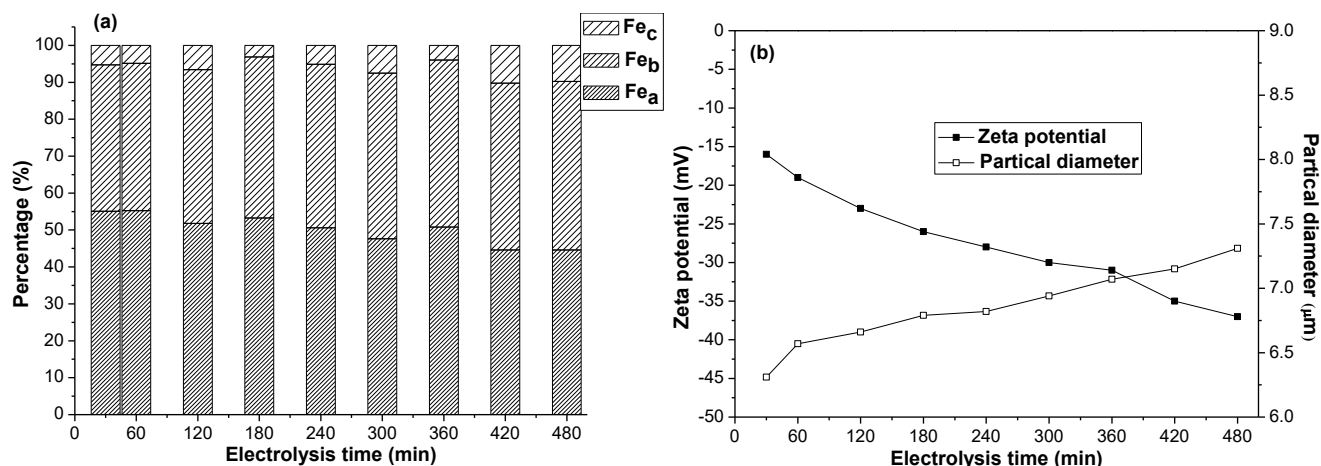


Figure 6. Effect of electrolysis time (a) on percentage of Fe_b and (b) on zeta potential and particle diameter by EC: Fe--Fe, applied voltage=20 V, initial pH=9.0, distance between electrodes=6 cm

3.3 Effect of applied voltage

The applied voltage was responsible for the production ratio of Fe²⁺, H₂ bubbles, the size and species of flocs during the EC process. With increasing applied voltage, the dissolution of the sacrificial anode accelerated, leading to increasing concentrations of hydrates, hydroxides, polyhydroxides and polyhydroxymetallic complexes, which can strongly adsorb or easily co-precipitate with contaminants. Simultaneously, the generation rate of H₂ bubbles increased and the size of bubbles decreased, which could improve the coagulation performance and mass transfer in the EC process, increasing the removal efficiency. However, the results (Figure 7 (a) and (b)) of this study showed that a turning point occurred in both turbidity reduction and heavy metals removal when the applied voltage was higher than 20 V. The best treatment results are achieved at an applied voltage of 20 V with pH stabilized at 7.35. The amount of Fe_b floc (Figure 8 (a)), which is regarded as the most active species, was the highest among Fe species, and from Figure 8 (b), it is obvious that the absolute value of zeta potential and the floc size also reached maximum levels at an applied voltage of 20 V. These observations resulted in identification of an applied voltage of 20 V as the ideal applied voltage. When too high a voltage was used, there was increased chance of electrode polarization and passivation, leading to the decline of electrical efficiency as some studies reported [31, 32]. Furthermore, increases in applied voltage may result in uneconomical wasting of electrical energy through heating of the water.

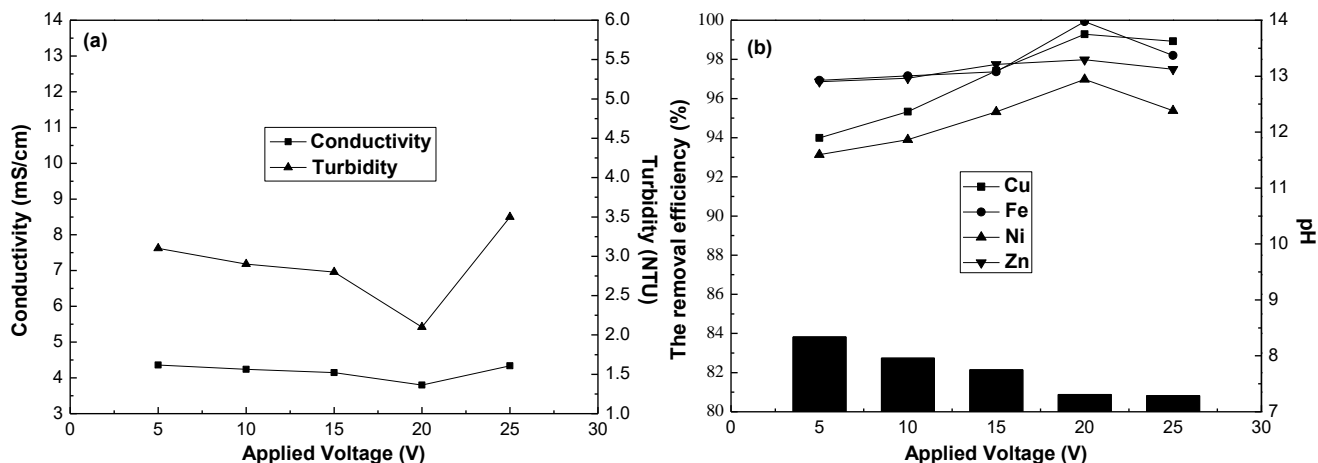


Figure 7. Effect of applied voltage (a) on conductivity and turbidity and (b) on metals removal and pH change by EC: Fe-Fe, electrolysis time= 40 min, initial pH= 9.0, distance of electrodes=6 cm

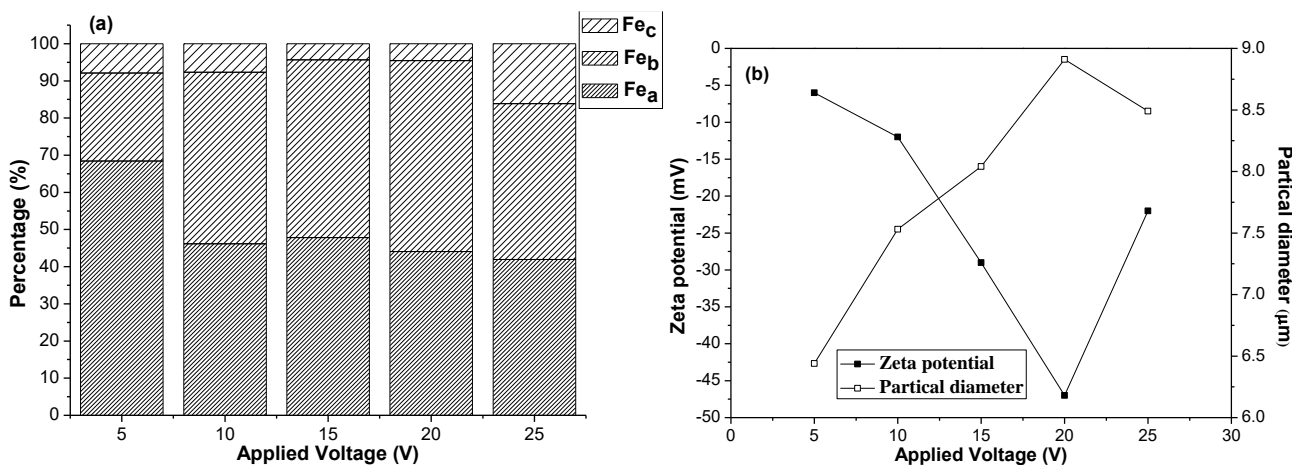


Figure 8. Effect of applied voltage (a) on percentage of Fe_b and (b) on zeta potential and particle diameter by EC: Fe-Fe, electrolysis time= 40 min, initial pH= 9.0, distance of electrodes=6 cm

3.4 Effect of inter-electrode distance

Electrode distance is also an important factor to be investigated, since it can affect turbulence, mass transfer and energy consumption. In this study, the gap between the electrodes was varied from 1 to 6 cm under fixed operating conditions, viz., initial pH 9.0, electrolysis time 40 min, and applied voltage 20 V. The results showed that the changes in removal efficiency of the four heavy metals and in conductivity were minor, but the reduction in turbidity first increased then decreased as the electrode gap was moved from 1 to 6 cm. The maximum removal of turbidity and of Cu²⁺, Fe²⁺, Ni²⁺, and Zn²⁺, especially Ni²⁺, occurred at an inter-electrode distance of 4 cm (Figure 9). Similar observation has been reported by Sahu et al. [33] for the reduction of COD, which increased and then decreased with electrode distances from 10 mm to 25 mm and reached the maximum at 20 mm. These results were different from some previous reports. Nandi and Patel [34] observed that there was maximum dye removal with short inter-electrode distances. Sharma and Chopra [35] found that the percentage removal of NO₃⁻ and SO₄²⁻ increased with decreases in electrode distance from 2.5 to 0.5 cm. According to

previous reports [31, 36], the gap distance between the electrodes will have two opposite effects. On the one hand, as the inter-electrode distance decreases, the electrical resistance (IR drop) decreases, the current density will be higher at the same applied voltage, and the energy consumption of the EC process will be reduced; on the other hand, as the inter-electrode distance decreases, there is a low mixing of the fluid between electrodes, resulting in an insufficient increase in the concentration polarization layer on the electrode surface. In this study, the optimum inter-electrode distance was 4 cm. The pH of the solution is stable at approximately 7.3 regardless of distance. Figure 10 shows that the percentage of Fe_b floc, the absolute zeta potential, and the particle diameter all were at a maximum when the inter-electrode distance was 4 cm. This result once again proved the correspondence between these parameters and the removal effect.

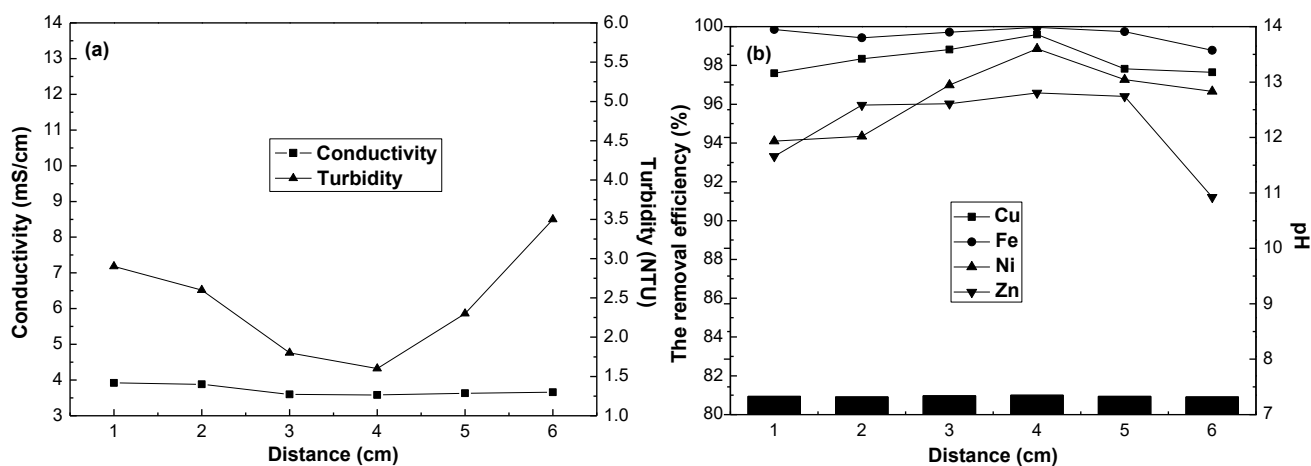


Figure 9. Effect of inner-electrode distance (a) on conductivity and turbidity and (b) on metals removal and pH change: Fe-Fe, applied voltage=20 V, electrolysis time= 40 min, initial pH= 9

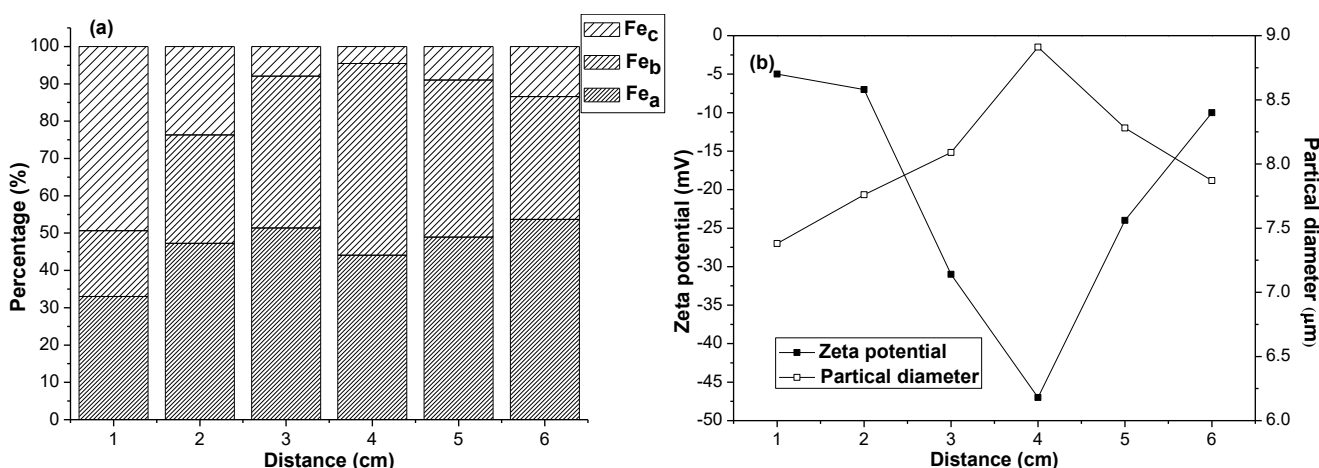


Figure 10. Effect of inner-electrode distance (a) on percentage of Fe_b and (b) on zeta potential and particle diameter: Fe-Fe, applied voltage=20 V, electrolysis time= 40 min, initial pH= 9

4. CONCLUSIONS

(1) For the efficient treatment of WFGD wastewater to reduce the turbidity, conductivity and heavy metals efficiently, the optimal conditions are: an initial pH of 9.0, an electrolysis time of 40 min, an applied voltage of 20 V and an inter-electrode distance of 4 cm.

(2) EC requires a lower initial pH than CC to achieve a better removal efficiency of heavy metals and turbidity. With continuous electrocoagulation, the pH will drop and stabilize at approximately 7.35.

(3) pH has a significant influence on Fe species distribution, the largest amount of Fe_a species occurred at pH of 7.0. With increases in the initial pH, the content of Fe_c species increased.

(4) Fe_b species fraction in the EC process correlated well to the the removal of heavy metals and turbidity. The zeta potential and particle diameter also correlated well with heavy metals and turbidity removal.

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