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Variations in heavy metals and turbidity in wastewater from wet flue gas desulfurization by electrocoagulation

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ABSTRACT

Wastewater containing various contaminants, including total suspended solids (TSS), heavy metals, and salts, is produced from the wet flue gas desulfurization (WFGD) process. Electrocoagulation (EC) was compared to traditional chemical coagulation methods and applied as a novel technology to treat WFGD wastewater in this study. Based on three different combinations of electrodes (Al-Al, Fe-Al and Fe-Fe), the dependence of the heavy metal (Zn and Ni) contents and the turbidity on the electrolyzer voltage and electrode gap was investigated. The results show that optimal removal efficiencies of 82.56% and 52.37% for Zn and Ni, respectively, can be achieved at a voltage of 20 V and an electrode gap of 2 cm, while the turbidity sharply decreases from 488 NTU to 8.3 NTU. The application of Fe as an anode can enhance pollutant removal and electrochemical stability in WFGD wastewater.

Keywords: Wastewater; Wet flue gas desulfurization (WFGD); Electrocoagulation (EC); Heavy metals

1. Introduction

For flue gas purification, many coal-fired boilers or sintering machines are equipped with flue gas desulfurization (FGD) technology. Among various FGD technologies, wet FGD (WFGD) technology is preferred due to its higher purification efficiency, stable operation and lower cost [1]. From a mass balance perspective, wastewater is inevitably produced from the WFGD process and must be discharged to maintain a stable level of chloride ions (e.g., 12000–20000 mg/kg) [2,3]. Generally, WFGD wastewater discharge has the following properties: (1) weak acidity, (2) high concentrations of total suspended solids (TSS) and Cl^-, (3) high hardness due to calcium-based desulfurizers and (4) various heavy metals and toxic metalloid elements. Depending on the WFGD process, operating conditions, and fuel quality, the substances in WFGD wastewater vary greatly and are of great concern to human health. At present, the commercial methods for WFGD wastewater treatment include the mixing of ash and wastewater, chemical coagulation, and flue duct evaporation, among others. However, there are still many challenges for these forms of WFGD wastewater treatment, as it is difficult to balance performance and cost. With increasingly stringent environmental requirements, the application of chemical coagulation (CC) is limited due to the use of complex facilities and the higher operation and maintenance costs [4,5]. In recent years, flue duct evaporation seems to be promising as an emerging technology, as near-zero or zero emissions of wastewater and lower operating costs can be realized [6,7]. At the same time, the effect of spraying wastewater into flue gas on the moisture and temperature of the flue gas was studied and is believed to cause damage to devices downstream [8,9].

Among a variety of technologies for industrial wastewater treatment, electrocoagulation (EC) is an increasingly competitive option, in which Al^{3+} or Fe^{3+} floc produced from the sacrificial anode plays the same role as in CC.

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Furthermore, these flocs exhibit better characteristics than those from CC because of the decreased amount of bound water, easier filtration, and greater resistance to shear [10,11]. Meanwhile, H_2 gas generated at the cathode promotes flotation. As a result, the EC performance is improved, and the costs decrease without the addition of extra reagents and facilities. EC has been successfully applied to treat oil emulsion-containing wastewater [12], mining wastewater and dye wastewater [13–17]. EC has significant advantage over other technologies for the heavy metals removal from wastewater due to its simple operation, less sludge production and lower costs. Therefore, EC has become an attractive technology to treat industrial wastewater containing different levels of heavy metals in recent two decades, and influence factors, mechanism as well as products have been intensively investigated [18,19]. As mentioned, WFGD wastewater usually contains suspended substances, heavy metals, and other components that can be removed effectively by means of EC, without adding chemical flocculants and avoiding re-contamination. To the best of our knowledge, there are few literature references to WFGD wastewater treatment using EC. In this paper, we attempt to investigate the effect of specific EC parameters, e.g., anode materials, multiple dilutions of wastewater, EC electrolyzer voltage and electrode gap, on heavy metal content, turbidity and conductivity, and the feasibility of WFGD wastewater treatment by EC is explored.

2. Material and methods

The EC experiments were performed in a φ15 × 20 cm cylindrical glass reactor, and a 15 cm × 4 cm aluminium (or iron) sheet with a thickness of 0.3 cm was employed as the anode. An aluminum (or iron) sheet with the same shape was used as the cathode. All electrode sheets were soaked in dilute HCl acid for 12 h to remove surface oxides and then polished with abrasive paper prior to the EC experiments. The combinations of anode-cathode in this study were Al-Al, Fe-Fe and Fe-Al, where the gap between each pair was fixed at 2 cm, 4 cm, 6 cm and 8 cm. The electrodes were connected to a DC power supply under constant voltage or current (RXN-305D, China).

WFGD wastewater was collected from a 600 MW coal-fired power plant in China that is equipped with a limestone-gypsum scrubber. Five 1.6 L wastewater samples with different dilution ratios of 0 (i.e., raw sample), 1:2, 1:4, 1:6, and 1:8 were sequentially adopted for the EC experiments, during which the 1.6 L wastewater was agitated mechanically at 200 rpm in the reactor vessel. Each experiment duration was 60 min, which allowed time for sufficient EC before 50 ml of wastewater was sampled for further measurement. After these samples were allowed to stand for 30 min, the supernatant was measured for heavy metal content, turbidity and conductivity. Fig. 1 gives the schematic diagram of the EC system.

Sample pH values were continuously monitored by a pH instrument (FHS-29A, INESA). The concentrations of Zn and Ni as typical heavy metals in wastewater were determined by inductively coupled plasma spectrometry (ICP-OES, Varian 720-ES). The turbidity and conductivity of the samples were analysed by a turbidity metre (WZS-185, INESA) and a conductivity metre (DDS-307, INESA), respectively. The removal efficiency of heavy metals or turbidity was calculated according to Eq. (1):

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

where C_1 and C_2 are the heavy metal concentrations or turbidity values in the supernatant before and after the EC experiments, respectively. All experiments were performed at 20–25°C.

3. Results and discussion

3.1. Raw wastewater quality

As an extremely complex matrix, the constituents of wastewater discharged from WFGD are heavily dependent on the quality of the fuel and supplemental water,

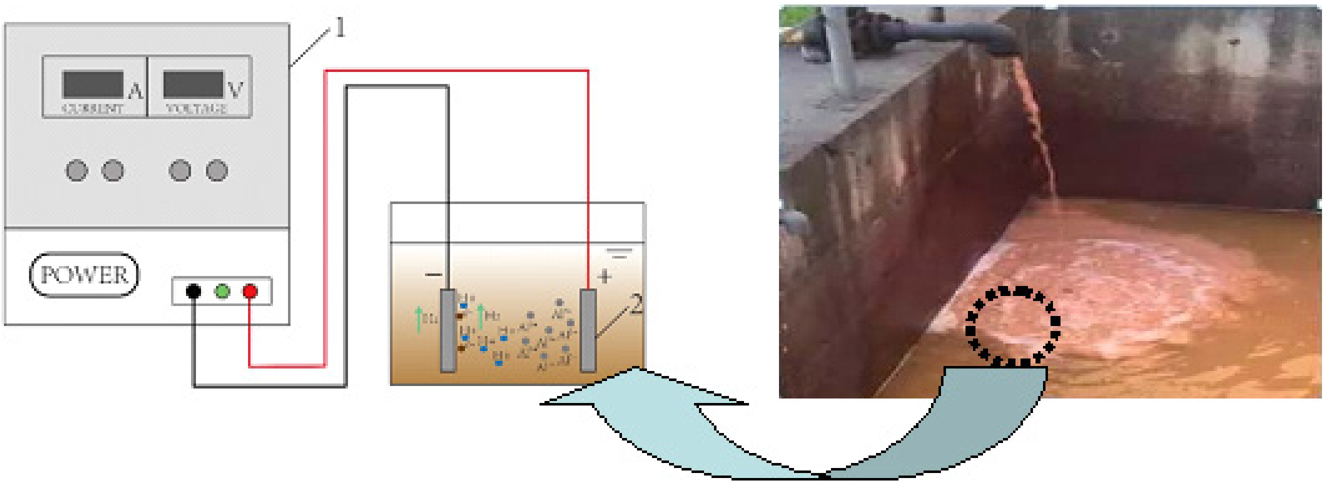

Fig. 1. Schematic diagram of the EC system for WFGD wastewater. 1, DC power and 2, anode (Al, Fe).

scrubber type, additive use and other operating conditions. The information for the collected WFGD wastewater is shown in Table 1. The raw wastewater shows extremely weak acidity with pH = 6.92. The Zn and Ni concentrations in the raw wastewater are 3.34 mg L^{-1} and 1.32 mg L^{-1}, respectively, which exceed the limits for heavy metals in the Chinese standard for discharged wastewater from limestone-gypsum FGD systems in fossil-fuel power plants (DL/T 997-2006). Additionally, the high concentrations of Cl^- and TSS and the high conductivity in the samples endow WFGD wastewater with unique salinity and corrosivity properties, causing difficulty in the treatment of the wastewater.

3.2. Effect of the electrolyzer voltage on variations in heavy metals and turbidity

EC experiments were conducted in raw wastewater at an initial pH = 6.9, where two Al sheets with a gap of 2 cm were adopted as electrodes. The electrolyzer voltage was set from 5 V to 25 V with a regular interval of 5 V. Fig. 2 shows the dependence of the heavy metal (Ni and Zn) content and the turbidity on the electrolyzer voltage. A significant drop in turbidity with increasing electrolyzer voltage is observed as the voltage approached 20 V; however, the turbidity increases at a higher voltage of 25 V. The removal efficiency of heavy metals, such as Zn and Ni, gradually increases with elevated voltage. At 5 V, the removal efficiencies of Zn and Ni are very low, with values of 21.1% and 12.4%, respectively. The optimal removal efficiency for Zn and Ni is obtained at approximately 20 V, which is consistent with the turbidity drop as mentioned above. As the driving force to push EC forward, the electrolyzer voltage is one of the key parameters. Vakil believed that a higher voltage facilitated the generation of more destabilized particles, which coagulated and flocculated over time, therefore improving the turbidity [20]. The voltage applied to the EC system depends on the nature of the solution (i.e., conductivity and pH) since the gap between electrodes was kept constant (i.e., 2 cm). The variations in pH shown in Fig. 3 indicate that after 60 min, the pH in the EC experiments increased slightly from the initial pH of 6.92. The formation of monomeric species (such as $Al(OH)_3$ and $Al(OH)_4^-$) and the buffering capacity are responsible for the pH variations [21,22]. With increasing voltage, the pH slightly declines from 7.74 to 5.99, while the conductivity increases. The lowest pH value obtained at 20 V is also ascribed to the more rapid consumption rate of OH^- relative to the OH^- production rate at the cathode, in accordance with the results of the turbidity removal efficiency. However, a voltage above 20 V may cause anode passivation and result in a negative influence on EC. In our cases, a decline of Zn and Ni removal efficiency takes place at voltage above 25V. It can

Table 1
Main parameters of raw WFGD wastewater

Parameters	Measured values	Standard values (DL/T 997-2006)
t	51°C	–
pH	6.92	6–9
Turbidity	488 NTU	–
Conductivity	14.33 mS cm^{-1}	–
Zn	3.34 mg L^{-1}	2.0 mg L^{-1}
Ni	1.32 mg L^{-1}	1.0 mg L^{-1}
Cu	0.57 mg L^{-1}	–
Fe	4.11 mg L^{-1}	–
Hg	2.09 μg L^{-1}	50 μg L^{-1}
As	20.01 μg L^{-1}	500 μg L^{-1}
Cl^-	22962 ppm	–
F^-	–	30 mg L^{-1}
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}	–

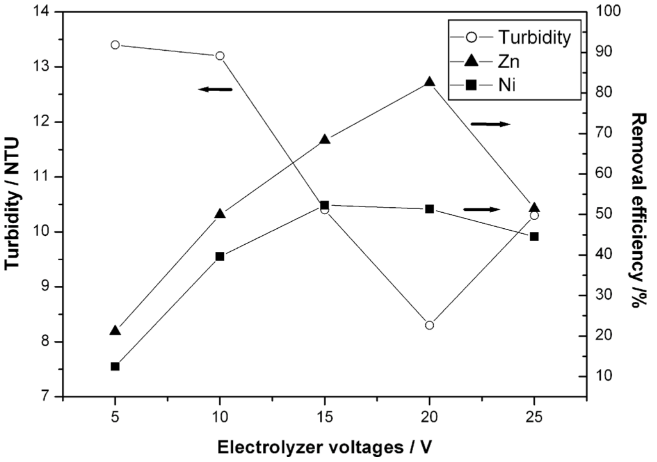

Fig. 2. Effect of electrolyzer voltage on the removal of turbidity, Zn and Ni: Al-Al, duration = 60 min, electrode gap= 2 cm, initial pH = 6.92.

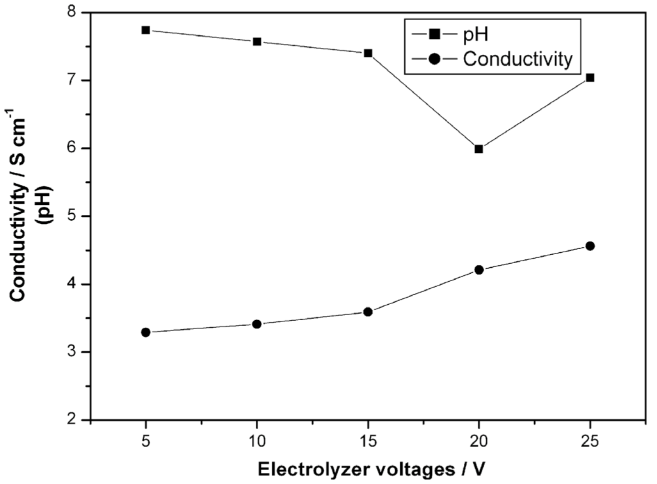

Fig. 3. pH and conductivity evolution at different voltages: Al-Al, duration = 60 min, electrode gap = 2 cm, initial pH = 6.92.

be attributed to the electrode passivation in WFGD wastewater, and the passivation occurring on the surface of electrode inhibits the electrode dissolution and therefore limits the electrocoagulation process.

3.3. Effect of the electrode gap on variations in heavy metals and turbidity

The electrode gap is another important variable in the design of an EC reactor and the associated costs. A moderate conductivity was measured for raw FGD wastewater, and the electrode gap in our study was tested over a wide range from 2 cm to 6 cm [23]. Fig. 4 shows the dependence of the heavy metal (Ni and Zn) content and the turbidity on the electrode gap. With a greater electrode gap, an increase in turbidity and a decreased Zn (or Ni) removal efficiency are observed. The evolution of pH and conductivity at different electrode gaps is given in Fig. 5. Except for the lower pH value obtained at 2 cm, few distinct differences in pH values are found for the electrode gaps investigated. In contrast, the conductivity decreases with larger electrode gaps. Since the electrical resistance between electrodes depends on the solution between them, the electrical resistance increases distinctly with larger gaps in a moderately conductivity solution [24]. As a result, the ions produced between electrodes move more slowly, and the voltage required for the optimal current increases. This explains why the sharp decline in the heavy metal removal efficiency at larger gaps occurs in Fig. 4.

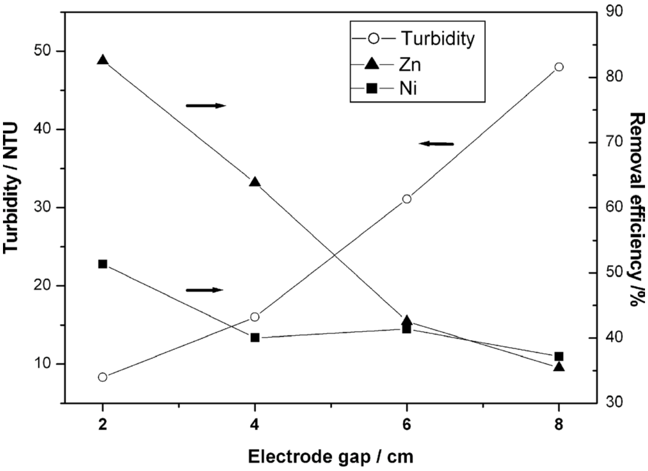

Fig. 4. Effect of electrode gap on the removal of turbidity, Zn and Ni: Al-Al, duration = 60 min, voltage = 20 V, initial pH = 6.92.

3.4. Effect of electrode materials on variations in heavy metals and turbidity

Three kinds of electrode pairs, Al-Al, Fe-Al and Fe-Fe, were used for the removal of turbidity and heavy metals. The synchronized variations in turbidity and heavy metal removal at different voltages are presented in Fig. 6. When the turbidity drops with voltage, the Zn or Ni removal efficiencies increase correspondingly. The flocs formed during EC contribute to the turbidity drop and to the adsorption of heavy metals as described in the EC mechanism, regardless of the electrode species [25]. The use of Fe as the anode, either in an electrode pair of Fe-Al or Fe-Fe, shows better performance than that of the Al-Al pair. Of course, it is admitted that the removal efficiency for WFGD wastewater only by electrocoagulation still requires to be enhanced, since maximum removal efficiency in case of Ni is not high enough. Some advanced technology, e.g., ozone-assisted or ultrasonic-assisted can be combined with traditional electrocoagulation process, in order to achieve higher removal efficiency in effluent with complicated composition [18,26].

Meanwhile, the optimized voltage shifts lower. That is, with the Fe anode, the optimal performance achieved is 15 V, which is lower than the optimized voltage (e.g., 20 V) for the Al anode. The optimal voltages for EC become more distinct between Fe and Al anodes. Further, once the voltage applied exceeds 20 V, the EC performance for pollutant removal becomes weaker in the case of Al, while no performance drop is found in the case of Fe. According to the half reactions for different electrode combinations taking place

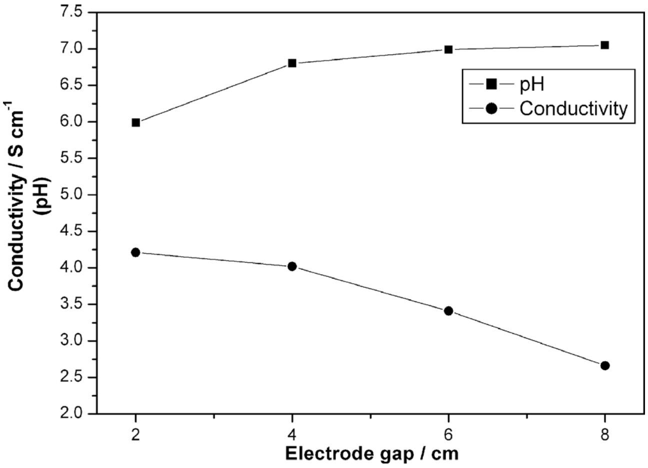

Fig. 5 pH and conductivity evolution at different electrode gaps: Al-Al, duration = 60 min, voltage = 20 V, initial pH = 6.92.

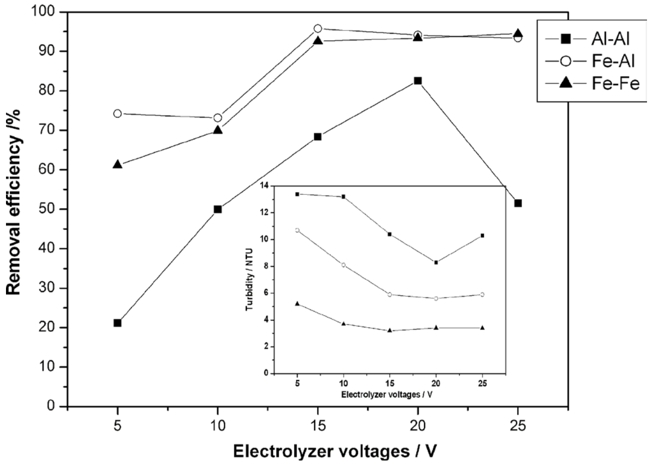

Fig. 6. Effect of electrode material on turbidity and Zn removal: duration = 60 min, electrode gap = 2 cm, initial pH = 6.92.

in the anode of the EC cell, the corresponding standard electrode potentials can be expressed as follows:

$$\text{H}_2\text{O reduction: } 2H_2O \rightarrow O_{2(g)} + 4H^{+}_{(aq)} + 4e^{-} \quad E^{o} = 1.229 \text{ V} \tag{2}$$

$$\text{Al reduction: } Al(s) \rightarrow Al^{3+}_{(aq)} + 3e^{-} \quad E^{o} = -1.662 \text{ V} \tag{3}$$

$$\text{Fe reduction: } Fe(s) \rightarrow Fe^{2+}_{(aq)} + 3e^{-} \quad E^{o} = -0.447 \text{ V} \tag{4}$$

In view of the half-reaction standard potentials, Al or Fe dissolution occurring at the anode yields a significantly lower potential than that of water reduction at the anode; therefore, Al or Fe dissolution will ultimately dominate [27]. Table 2 demonstrates the weight loss of the Al and Fe electrodes during EC. The weight loss of Al is always larger than that of Fe in terms of current. The result also indicates that Fe^{3+} or Fe^{2+} generated from the electrode per unit current plays a more significant role as a flocculant than does Al^{3+}. The dissolved Al^{3+} or Fe^{3+} is present in the hydrated cation form of $Al(H_2O)_6^{3+}$ or $Fe(H_2O)_6^{3+}$, respectively, which immediately hydrolyse in sequence into monomeric (or oligomer), polymeric and amorphous hydroxyl complexes [28]. Different flocculants have different mechanisms for the removal of pollutants. In case of sacrificial electrode Fe, the formed ferrous hydroxyl complexes can decrease or even eliminate the ξ potential of colloids in an aqueous solution. As a result, the agglomeration of colloids takes place through neutralization, bridge and sweep mechanisms, and a $Fe(OH)_3$ gel with a high degree of polymerization is finally produced. This $Fe(OH)_3$ gel has a strong adsorption and flocculation capacity for the hydrated cations, e.g., Cr, Zn, and Ni. It is noted that different from Al dissolution, the sequent reduction of divalent Fe into trivalent Fe takes place, which further enhances the electrocoagulation performance. In addition, when Al is used as sacrificial electrode, its environmental impact thereupon has to be considered. Compared to iron ion, there is more rigid limitation on residual aluminum in drinking water in USEPA guidelines. (The maximum concentrations are 0.05–0.2 mg/L for aluminum and 0.3 mg/L for iron, respectively). (https://www.epa.gov/dwregdev/drinking-water-regulations-and-contaminants).

However, the higher oxidation potential (–0.447 V) of Fe than of Al (–1.662 V) contributes to the difference in the observed electrochemical stability. The Al anode becomes passive when the voltage increases to 25 V, and a higher voltage is required to obtain a comparable removal performance [29]. However, no decline of Zn and Ni removal efficiency are observed when Fe anode was applied, and it suggests that no passivation happens in case of Fe anode. A mass of sulfate anion present in WFGD wastewater promotes the passivation of anode Al due to the formation of sulfate-Al complex [30]. From the perspective of energy saving and costs reduction, Fe applied as anode is also more favorable than Al, due to less energy demand and cheaper Fe plate than Al plate [31].

Table 2
Weight loss of the Al and Fe electrodes

Current /A	1	2	3	4	5
Fe loss/g	0.52	0.52	0.55	0.52	0.53
Al loss/g	0.68	2.12	2.07	2.84	3.69

Variations in heavy metal removal and turbidity with different electrode gaps are given in Fig. 7. A near-linear decrease in Zn removal efficiency indicates the significant role of the electrode gap distance. The decrease is attributed to the increase in ohmic loss between electrodes in certain concentrations of electrolyte solution. The negative effect of larger electrode gaps on removal happens regardless of the electrode materials. Fig. 8 shows the comparison of EC tests run at intervals of 12 min with the three electrode combinations. The amount of flocs produced in the Al-Al pair increases with reaction time, while that of flocs in the Fe-containing pair is relatively stable. However, the colour in Fe-Fe or Fe-Al solutions turns brown, which is distinct from that in the Al-Al pair, demonstrating that Fe dissolution into trivalent Fe happens after an EC run.

4. Conclusion

As one of the by-products from coal-fired power plants equipped with a WFGD process, WFGD wastewater has become a concern due to its unique properties of high salinity, corrosivity, heavy metal contents, etc. EC technology was first applied for the treatment of typical WFGD wastewater samples to remove the pollutants (e.g., turbidity and heavy metals) measured in the sample. An optimal electrolyzer voltage of 20 V is required for the removal of pollutants in this EC experiment. Removal efficiencies of 82.56% and 52.37% for Zn and Ni, respectively, can be achieved, and the turbidity sharply decreases from 488 NTU to 8.3 NTU. An electrolyzer voltage higher than 20 V adversely influences the pollutant removal. A lower electrical resistance can be obtained with a smaller electrode gap (e.g., 2 cm) and can reduce the electrolyzer

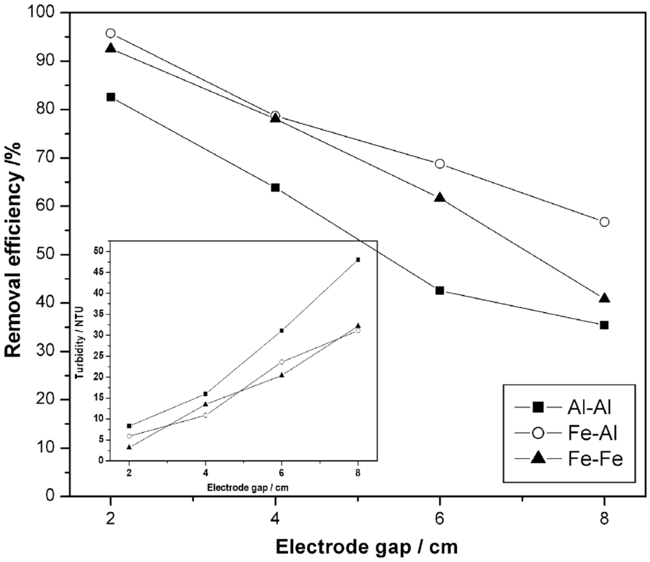

Fig. 7. Effect of electrode material on turbidity and Zn removal: duration = 60 min, voltage = 20 V, initial pH = 6.92.

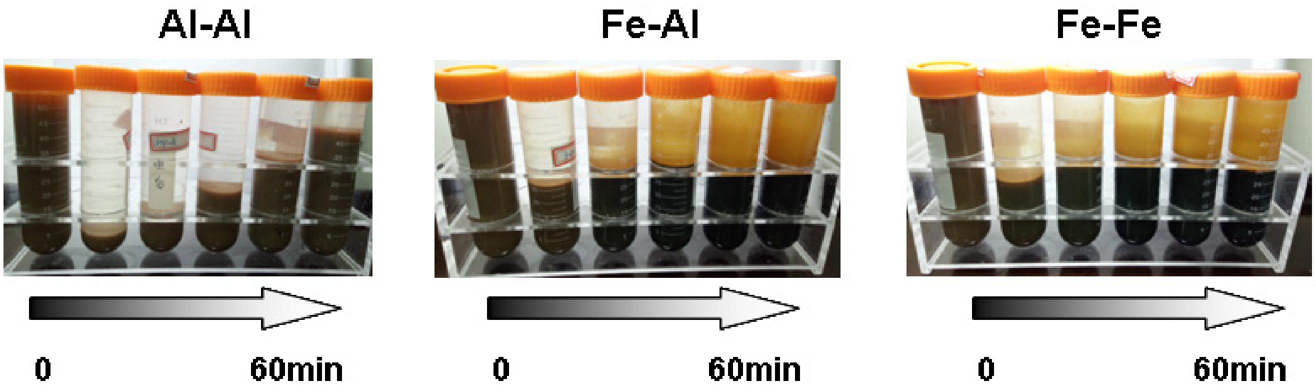

Fig. 8. Effect of EC experiments with three kinds of electrode pairs.

voltage required for EC. The application of Fe as an anode in WFGD wastewater electrocoagulation treatment significantly improves the removal of turbidity and heavy metals due to the higher oxidation potential of Fe and its electrochemical stability.

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