

Effect of Voltage on the Treatment of Cyanide Wastewater by Three-dimensional Electrode

Di Yao, Yonghui Song*, Shan Zhang, Yuhong Tian and Xinzhe Lan

Key Laboratory of Gold and Resources of Shaanxi Province, Xi'an University of Architecture and Technology, Xi'an 710055, China

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Abstract: The effects of applied voltage on cyanide wastewater treatment by three-dimensional electrodes were primarily examined. The main electrodes were produced from self-made coal-based electrode materials, and activated carbon was used as a particle electrode to treat cyanide wastewater. Changes in the concentration of ions in wastewater were determined and analyzed through Scanning Electron Microscopy and Energy Dispersive Spectroscopy. Results show that voltage significantly affects wastewater treatment via the three-dimensional electrodes. In particular, the removal rate of the ions in wastewater increases as voltage increases. The reaction mechanism of the three-dimensional electrodes also varies at different voltages. At 1 V, the reaction mechanism of the three-dimensional electrodes involves electro-adsorption. The removal rates of CN_T , Cu, Zn, SCN^- , and CN^- in wastewater are 31.81%, 53.04%, 36.11%, 29.05%, and 29.05%, respectively. The reaction mechanism of the three-dimensional electrodes includes electro-adsorption and enrichment precipitation when voltage is increased to 2 V. The three-dimensional electrode plays a significant role in electrolytic deposition when voltage is further increased to 3 and 4 V. Cu, Zn, and other heavy metal ions are reduced on the cathode and on the particle electrode surface, and the removal rates of CNT , Cu, Zn, SCN^- , and CN^- in wastewater are 93.94%, 95.22%, 97.23%, 99.38%, and 94.93%, respectively.

Keywords: Three-dimensional electrode; Applied voltage; Cyanide wastewater; Coal-based electrode; Carbon particle electrode.

1. INTRODUCTION

Cyanide leaching is an important gold extraction method for gold mines. Cyanide is also an effective solvent for gold extraction, and other elements in ore are dissolved in a cyanide leaching solution. However, cyanide and other heavy metal ions, including copper, zinc, and iron, persist in wastewater. If these substances are discharged directly without treatment, they likely contaminate resources and harm the environment and living organisms. Cyanide-contaminated wastewater is treated with various methods, including valuable substance recovery through activated carbon adsorption, ion exchange resin adsorption, chemical precipitation, acidification, electro-dialysis and membrane treatment. In addition to these methods, several reactions, including ozone oxidation, SO_2 /air oxidation, electrolysis oxidation, pyrolysis, blowing off aeration, microbial decomposition, and natural purification method are employed to disrupt cyanide oxidation [1]. However, these methods are characterized by some disadvantages, such as incomplete treatment, secondary pollution, long treatment period, and no

heavy metal recovery. Electrochemical wastewater treatment converts directly or indirectly contaminants on electrodes under the action of an applied electric field. This method provides several advantages, such as simplicity, small equipment volume and no secondary pollution. In the field of water treatment, removing various pollutants has been widely investigated as a possible solution to address these key issues.

In electrochemical water treatment, inorganic or organic wastewater treated with two- or three-dimensional electrode systems has been extensively explored. By contrast, three-dimensional electrode systems are filled with conductive particles between two-dimensional electrodes to produce a new working electrode. The comprehensive effect on the electrochemical reaction of these particles and the main electrode surface can improve the removal efficiency of harmful ions in wastewater [2,3]. In general, a metal plate or graphite is used as anode and cathode in three-dimensional systems, while activated carbon or active coke is utilized as a particle electrode. Wu Chen [4] examined the three-dimensional electrode treatment of zinc-containing wastewater and showed that the removal rate of Zn in simulated wastewater is 95.7% at a current of 0.6 A, time of 45 min, plate spacing of 4 cm,

*To whom correspondence should be addressed: Email: syh1231@126.com

and carbon/water ratio of 10:9. Liang Li [5] treated ammonia–nitrogen wastewater with activated carbon in three-dimensional electrodes and observed that the removal rate of ammonia–nitrogen increases as current density and chloride ion concentration increase. The removal rate of ammonia nitrogen reaches 99% after 2.5 h of electrolysis. Tianlong Zheng [6] treated the secondary wastewater of a polypropylene fiber factory with a three-dimensional electrode by using activated carbon as a particle electrode and found that the respective removal rates of COD_{Cr}, NH₃-N, TOC, and UV254 are 64.5%, 60.8%, 46.4%, and 64.8% under optimum conditions. Tianlong Zheng [6] further showed that two-dimensional electrodes and activated carbon adsorption exhibit a synergistic effect. Lingyong Wei [7] used Ti/SnO₂+Sb₂O₃ as an anode, stainless steel mesh as a cathode, and different proportions of activated carbon particles and porous ceramic particles as particle electrodes to pretreat heavy oil refinery wastewater by using a three-dimensional electrode reactor. Lingyong Wei [7] indicated that the COD removal rate of the three-dimensional electrode (combined particles) is higher than that of the two-dimensional electrode (non-particle-filled). The respective removal rates of COD, total organic carbon, and toxicity units are 45.5%, 43.3%, and 67.7% at 75% activated carbon and 30 mA/cm² current for 100 min. Particle electrode is more focused on the electrochemical effect than on the adsorption effect after activated carbon undergoes adsorption and desorption. Ti/RuO₂-IrO₂ as an anode, 1Gr18Ni9Ti stainless steel as a cathode, and a pair of titanium plates as simulated particle electrodes have been applied to the work principle of filling particles in a three-dimensional electrode system [8]. The increase in the electrochemical reaction products of the particle electrode and the electrochemical products of the main plate elicited a comprehensive effect at constant voltage [8]. The energy efficiency of a three-dimensional electrode electrochemical reactor is higher than that of a two-dimensional electrode electrochemical reactor. Titanium-coated ruthenium plate as an anode, stainless steel plate as a cathode, and steel ball as a third pole have been utilized to treat Cu-containing wastewater [9]. The removal rate of copper is 82.3% at a plate spacing of 3 cm, voltage of 10 V, and electrolysis time of 45 min [9]. These results suggest that a high voltage corresponds to an enhanced treatment effect, but side reactions likely occur on filling particles at excessively high voltage. Consequently, pollutants are only partially adsorbed.

Therefore, controlling suitable voltage is a key factor in three-dimensional electrode systems. In this study, a three-dimensional electrode system composed of a coal-based electrode material and commercial activated carbon was used to treat cyanide wastewater produced by gold extraction. The influence of the applied voltage on the removal process of the ions in cyanide wastewater was discussed to provide a theoretical basis for technological applications.

2. EXPERIMENTAL

2.1. Materials

The cyanide wastewater used in the experiment was obtained from Zhong Jin Song Yuan Gold Smelting Plant. The main composition is shown in Table 1. The cathode and anode plates were mixed with low-grade pulverized coal and liquefied residue. After pyrolysis and nitric acid activation were completed, the coal-based electrode material [$\Phi 30$ mm \times 2 mm] was prepared [10]. Commercially available coconut shell activated carbon (Rui Chao) was used

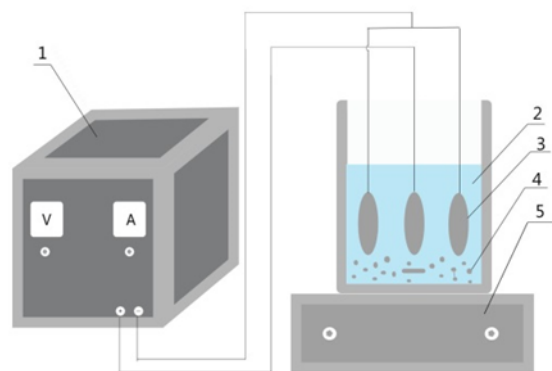


Figure 1. Experimental setup: 1 - DC power supply; 2 - cyanide waste-water; 3 - coal-based electrode; 4 - activated carbon; 5 - magnetic stirrer.

as the particle electrode.

Table 1 shows that the concentration of total cyanide, CN_T, in cyanide gold extraction wastewater is very high, while the free CN⁻ ion concentration is low. Therefore, this wastewater contains a large amount of complex ions. The concentrations of Cu and Zn in the wastewater samples are considerably larger than those of iron. Therefore, this study is focused on the reactions and removal of Cu and Zn ions.

2.2. Experimental setup

Cyanide gold extraction wastewater (50 mL) was collected and placed in a 100 mL beaker. The three coal-based electrodes were connected with the positive and negative voltages of the power supply and then connected in parallel into the solution. The plate into the water area was consistent with a plate spacing of 1 cm. Activated carbon (2 g) was added to the wastewater solution. The bottom of the solution was stirred with a magnetic stirrer at room temperature. The experiment was carried out by changing the applied voltage. During the experiment, the concentration of the ions was obtained as a sample solution at regular intervals for ion concentration analysis to calculate the removal rate of each ion. After the experiment was completed, the plate and ion electrodes were repeatedly washed with deionized water until the pH of the effluent was 7. The samples were dried and sampled for scanning electron microscopy and energy spectrum analysis. The experimental device is shown in Fig. 1.

2.3. Analysis and characterization

The concentrations of Cu, Zn, CN⁻, CN_T, and SCN⁻ in the solution were analyzed by chemical titration method. The ion removal rate was calculated according to Eq. (1):

Table 1. Various components of cyanide wastewater (mg / L)

	CN _T	CN ⁻	Cu	Zn	SCN ⁻	Fe
Cyanide wastewater	1728.60	367.20	546.10	456.20	260.10	48.17

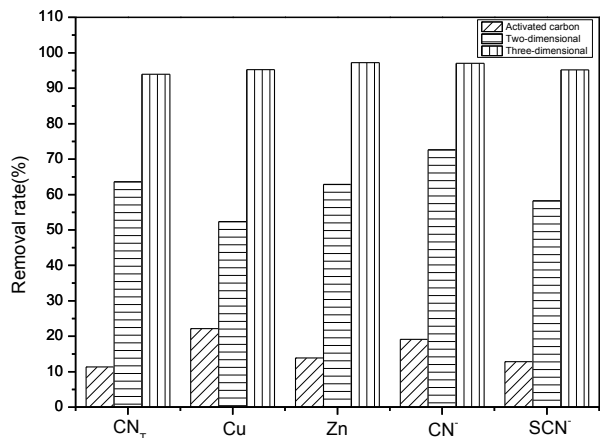


Figure 2. Two- and three-dimensional electrode and activated carbon adsorption ion removal rate

$$\varphi = \frac{C_0 - C_t}{C_0} \times 100\% \quad (1)$$

where C_0 is the initial concentration of each ion in the solution (mg/L) and C_t is the concentration of each ion in the solution after treatment (mg/L).

The surface morphology and the composition of the cathode plate, anode plate and carbon particle electrode were characterized with a JMS-6390Lv scanning electron microscope by obtaining the EDS band.

3. RESULTS AND DISCUSSION

3.1. Comparison of removals with activated carbon, two-dimensional electrodes and three-dimensional electrodes

A series of comparative experiments was performed to explain the role of activated carbon particle electrode in cyanide wastewater treatment. The experiments of the removal by the activated carbon adsorption were performed at room temperature with no external voltage, and 5 h of oscillation adsorption. The two- and three-dimensional electrodes were self-made with a coal-based electrode for anode and cathode, and the process conditions were room temperature, 4 V of applied voltage, and electrolysis for 5 h. The experimental results are shown in Fig. 2.

Fig. 2 shows that activated carbon adsorbs several major ions in cyanide gold extraction wastewater, but the adsorption rate is low. The highest adsorption rate of Cu ions is approximately 20%. The removal rate of each ion in the solution after treatment with the two-dimensional electrode was increased by a large margin. The removal rates of CN_T, Cu, Zn, CN⁻, and SCN⁻ were 63%, 50%, 60%, 70%, and 54%, respectively, and the removal efficiencies of all the ions in the solution were greatly increased after two-dimensional electrode treatment was completed. The removal efficiencies of the three-dimensional electrode were 93.9%, 95.2%,

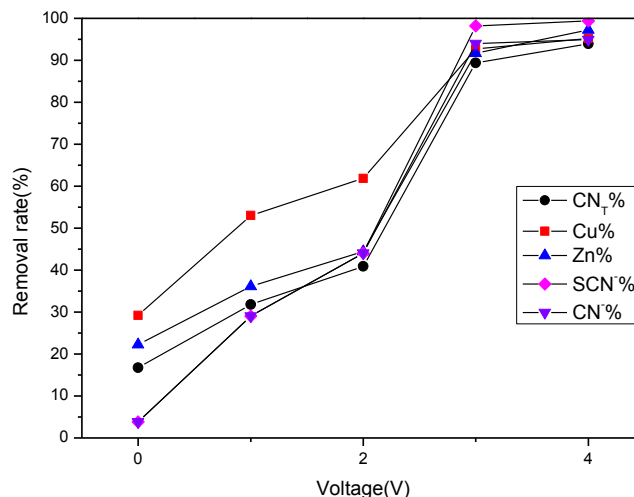


Figure 3. Variation of ion removal rates at different voltages

97.2%, 99.3%, and 94.9%, respectively. This result shows that particle electrode provides various advantages for the electrochemical treatment of cyanide wastewater; for example, good adsorption performance, electrochemical properties and catalytic properties of activated carbon particles help to increase the removal rate of each ion in wastewater [11]. At the applied voltage, activated carbon particles act as particle electrodes in the three-dimensional electrode. At a sufficiently high applied voltage, the voltage drop across the direction of the electric field exceeds the reversible potential of cathodic and anodic reactions, and positive and negative charges are induced electrostatically and become independent electrodes. Electrochemical oxidation and reduction reactions occur simultaneously at both ends and these reactions are equivalent to a number of micro-electrolysis cells observed in a previous work [12]. Thus, the treatment effect of three-dimensional electrodes is better than that of activated carbon adsorption and two-dimensional electrodes.

3.2. Applied voltage experiment

The results of three-dimensional electrode treatment of cyanide wastewater at 1, 2, 3, and 4 V are shown in Fig. 3.

Fig. 3 illustrates that the removal rate of each ion in the wastewater increases as the applied voltage increases. At 0 V, the removal rate of each ion in wastewater is less than 30%, which is mainly caused by the adsorption at the coal-based electrode material and the activated carbon particle electrode. Activated carbon and coal-based electrode materials possess a large specific surface area, large pore size distribution, and numerous surface functional groups, such as carboxyl, lactone, and phenolic hydroxyl groups, which can adsorb complex metal ions [13]. When the applied voltage was increased to 1 V, the electrode reaction did not occur and the solution system did not change. The removal rate of each ion was increased to a certain extent, but the increment was small. At this actual voltage, the cathode and anode surfaces have not reached the oxidation-reduction potential of OH⁻ and H⁺. There-

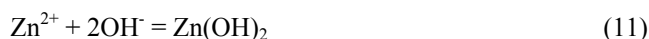
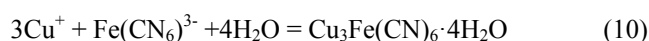
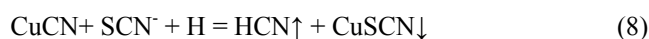
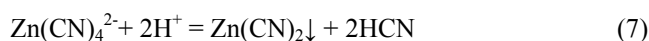
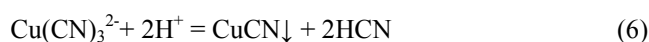
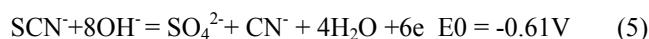
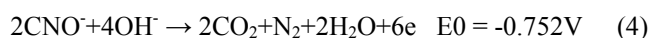
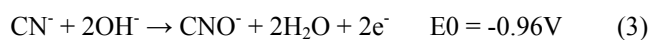
fore, the anions in the solution mainly migrated under the action of the electric field and became adsorbed on the surface of the anode and activated carbon particles. This phenomenon is a typical electric adsorption process. When the applied voltage reached 2 V, tiny bubbles emerged on the surface of the anode and cathode and a small amount of white precipitate was formed in the solution. These findings indicated that some reaction occurred at electrodes and some precipitation took place in the solution. The removal rate of each ion was slightly increased, but most of the removal rates did not exceed 50%. At this point, the actual voltage on the anode plate exceeded the decomposition voltage of water but not the decomposition voltages of the copper and zinc complex ion. The cathode plate did not appear metallic. In these conditions, the removal of ions in cyanide wastewater is mainly due to the combination of the electro-adsorption of particle electrodes and anodes and the enrichment of ions in the solution [14]. When the voltage was increased to 3 V and 4 V, the removal rates of CN^- , Cu, Zn, SCN^- , and CN^- were 93.94%, 95.22%, 97.23%, 99.38%, and 94.93%, respectively. As the applied voltage increased, the removal rates of the ions in the solution did not change much because the actual voltage applied to the anode and cathode exceeded the reduction potential of the metal complex ions, and Cu and Zn ions were deposited on the surface of the cathode. Therefore, the removal rate of heavy metal ions increased. At sufficiently high actual voltages at the anode and cathode, the repolarization degree of the particle electrode is high in the three-dimensional system, the ions in the wastewater are combined with the particle electrode, the main electrode undergo electro-adsorption, oxidation reaction occurs at one end of the particle, and reduction reaction takes place at the other end [15]. At this point, the anode oxidation reaction of CN^- may occur to produce CO_2 and N_2 and consequently produces a free cyanide solution and increases the total cyanide removal rate.

3.3. SEM-EDS analysis

The results of SEM-EDS analysis of anode plate, cathode plate, and activated carbon particle electrode after treatment of cyanide gold extraction wastewater at 1, 2, and 4 V are shown in Figs. 4, 5, and 6, respectively.

It can be observed in Fig. 4, that as the voltage increases the load on the anode plate gradually decreases with numerous white substances. Under the applied electric field at 1 V, the anode potential is smaller than OH^- and CN^- ion oxidative decomposition voltages, and the anode does not undergo an electrode reaction. The anions such as SCN^- , $\text{Fe}(\text{CN})_6^{4-}$, $\text{Fe}(\text{CN})_6^{3-}$ and $\text{Zn}(\text{CN})_4^{2-}$ migrate to the anode and adsorb on the anode surface by a pure electric adsorption process. The contents of Fe, S, Zn and other elements on the anode surface are higher than the initial values. A high ionic valence corresponds to a large electrical attraction between ions and the double layers formed at electrode surfaces. If the ionic valence is the same, the radius is small and the adsorption rate and capacity are high [14]. Therefore, the adsorption of complex ions of $\text{Fe}(\text{CN})_6^{4-}/\text{Fe}(\text{CN})_6^{3-}$ is followed by $\text{Zn}(\text{CN})_4^{2-}$, $\text{Cu}(\text{CN})_4^{2-}$, $\text{Cu}(\text{CN})_3^{2-}$, SCN^- , and CN^- . At 2 V, the anode begins to generate bubbles, and this observation indicates that the actual voltage on the anode exceeds the oxidation potential of OH^- . The gas is mainly oxygen but may also be due to oxidation of CN^- , which produces CO_2 and N_2 and oxidation of SCN^- , which requires further experiments. The anodic

reactions are together with their standard redox potentials, E° , represented by Eqs. 1–4. At the same time, large amount of white precipitate was produced in the solution near the anode, which indicated that the precipitation reaction occurred in the solution. Under the action of electric field, $\text{Cu}(\text{CN})_4^{2-}$, $\text{Cu}(\text{CN})_3^{2-}$, $\text{Zn}(\text{CN})_4^{2-}$, $\text{Fe}(\text{CN})_6^{4-}$, $\text{Fe}(\text{CN})_6^{3-}$, SCN^- , and other anions migrate toward the anode. As OH^- decomposes on the anode surface, the equilibrium of water is destroyed, increasing the local concentration of H^+ . In the meantime, $\text{Cu}(\text{CN})_4^{2-}/\text{Cu}(\text{CN})_3^{2-}/\text{Zn}(\text{CN})_4^{2-}/\text{Fe}(\text{CN})_6^{4-}/\text{Fe}(\text{CN})_6^{3-}$ and SCN^- reacted with each other to form the main white and brown precipitates, which are mainly composed of CuSCN , CuCN , ZnCN_2 , $\text{Cu}_2\text{Fe}(\text{CN})_6$, $\text{Zn}_2\text{Fe}(\text{CN})_6$, $\text{Cu}_3\text{Fe}(\text{CN})_6 \cdot 4\text{H}_2\text{O}$, and $\text{Zn}(\text{OH})_2$. The main reactions are described by Eqs. 5–10 [10,16]. Under the combined action of anodic electro deposition and enriched precipitation, the removal rate of each ion in the solution was improved. When the applied voltage increased to more than 3 V, the anode surface voltage exceeds the actual decomposition of CN^- undergoing through the $\text{CN}^- \rightarrow \text{CNO}^- \rightarrow \text{CO}_2 + \text{N}_2$ main reaction and the oxidation–decomposition reaction of SCN^- . Since the gas precipitation reaction is more intense, the anion adsorption on the anode surface is affected, and the adsorption capacity decreases [15,17]. In these conditions, the load on the anode plate is low, no precipitate is formed in the solution, and a large load is present on the cathode surface. Therefore, the cathode mainly undergoes the metal ion electro deposition reaction.



As shown in Fig. 5, at the applied voltage of 1 V the cathode plate load is low and consisted mainly from Na and K-based elements. In the electric field, Na^+ , K^+ , and other cations in the solution gradually move to the cathode and adsorb at the cathode plate surface and no electrode reaction occurs. When the voltage is 2 V, the hydrogen evolution reaction, described by Eq. (11) becomes the main cathodic reaction. Metal ion electro deposition will also occur together with Na^+ , K^+ , and other cation adsorption reactions. When the applied voltage reaches 3 V or more, the actual voltage on the cathode exceeds the precipitation voltage of $\text{Cu}(\text{CN})_4^{2-}$, $\text{Cu}(\text{CN})_3^{2-}$, $\text{Zn}(\text{CN})_4^{2-}$, $\text{Fe}(\text{CN})_6^{4-}$, and $\text{Fe}(\text{CN})_6^{3-}$. Therefore, the main cathodic reactions are Cu, Zn, and Fe electro-deposition reac-

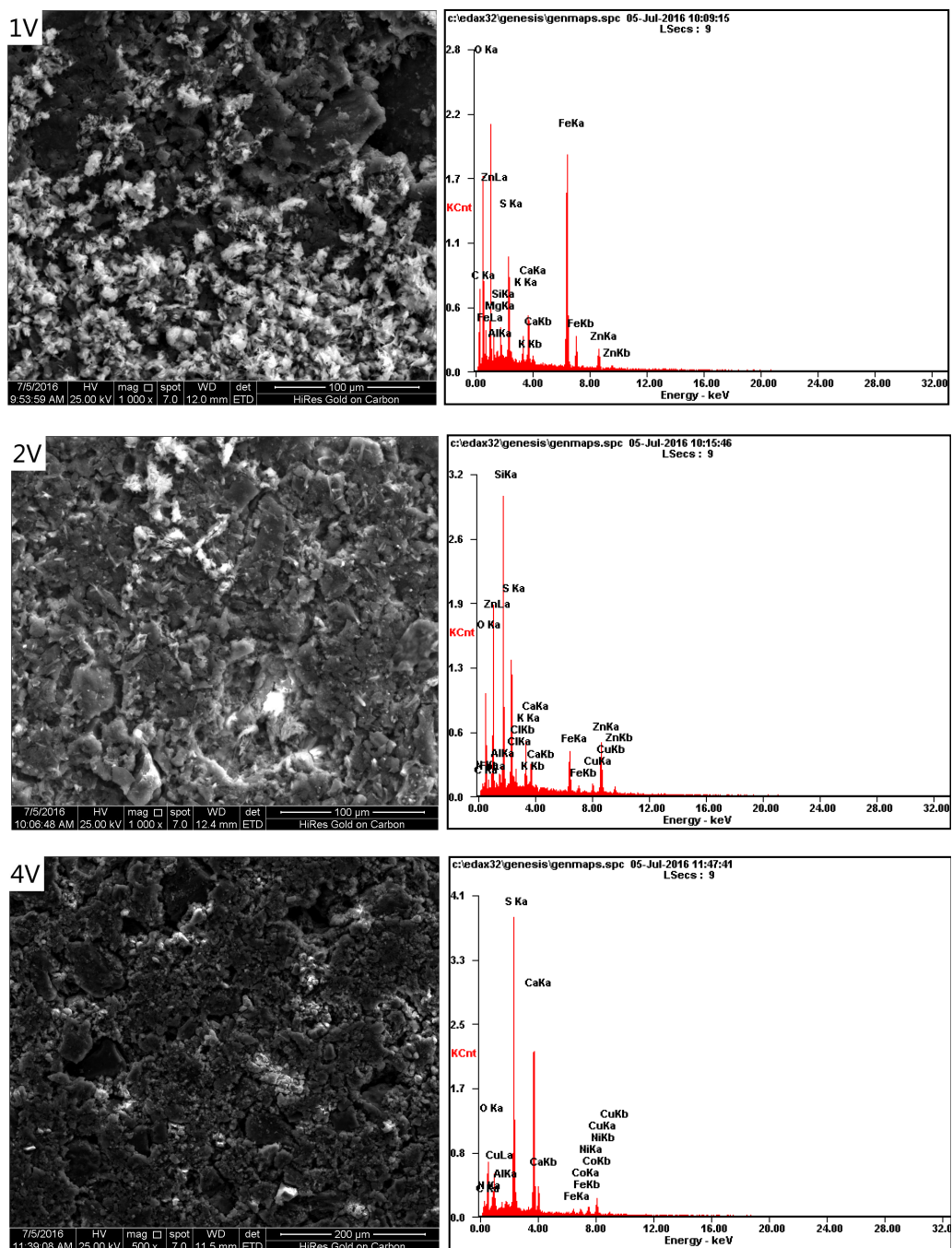


Figure 4. SEM–EDS analysis of anode plates at different voltages

tions. The surface forms a uniformly distributed metal loading layer, and the reactions are described by Eqs. 12–14.

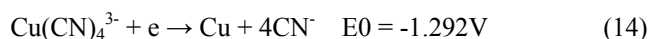
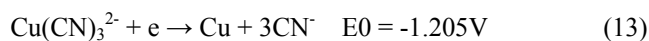
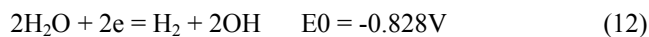


Fig. 6 shows that the particle electrode surface contains a small amount of load when the voltage is 1 V. In addition to K, S, and Ca elements, small amounts of Fe and Zn elements are also present. The activated carbon itself which has a larger surface area and pore size structure, will also be part of the wastewater adsorption of complex ions. In the case of power, the particle electrode surface will carry the charge to increase the polarity and enhance adsorption capacity [18]. When the voltage applied across the main electrode is small, the degree of repolarization of the particles is also small. In addition to the electro-adsorption at the main electrode,

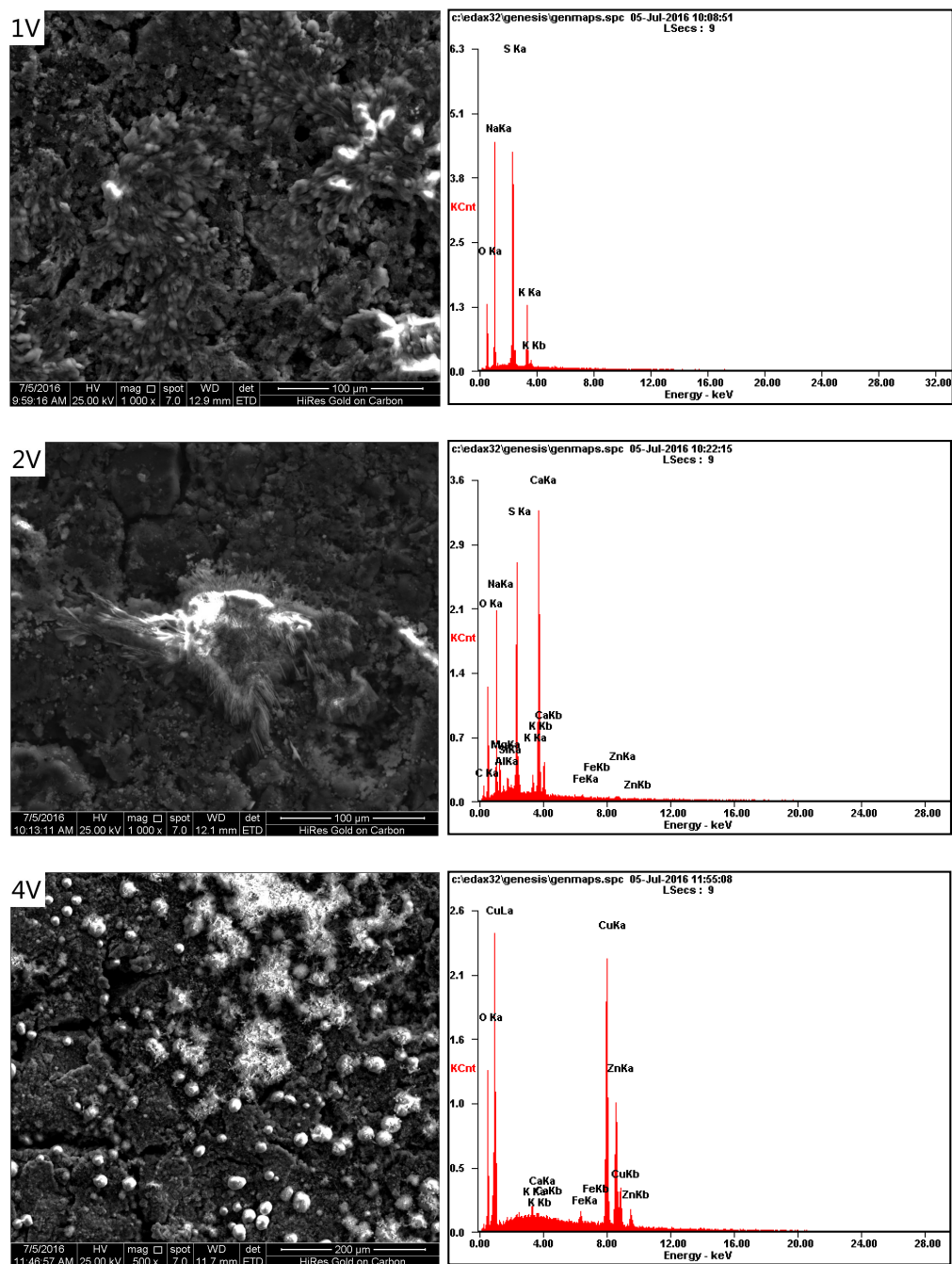


Figure 5. SEM-EDS analysis of cathode plates under different voltages

electro-adsorption of ions at the charged particle electrode may also occur. According to the literature [14], the adsorption order of metal complex on activated carbon is

$\text{Fe}(\text{CN})_6^{4-} > \text{Zn}(\text{CN})_4^{2-} > \text{Cu}(\text{CN})_3^{2-}$. Therefore, a small amount of Fe and Zn is adsorbed on the particle electrode at 1 V. When the voltage is increased to 2 V, the particles on the electrode load gradually increase, while the cathode has not yet undergone a deposition reac-

tion. The process at the anode and the particle electrode surface is mainly electro-adsorption of ions. As the voltage on the main electrode increases, the electric field intensity is enhanced, and the degree of repolarization of the particles increases. The two ends of the activated carbon yield positive and negative charges. The anions in the solution move to the anode and become adsorbed on the particle electrode. When the voltage increased to 3 and 4 V, the particle electrode repolarization degree is higher. When the cathode voltage is higher repolarization of the particle electrode is equiva-

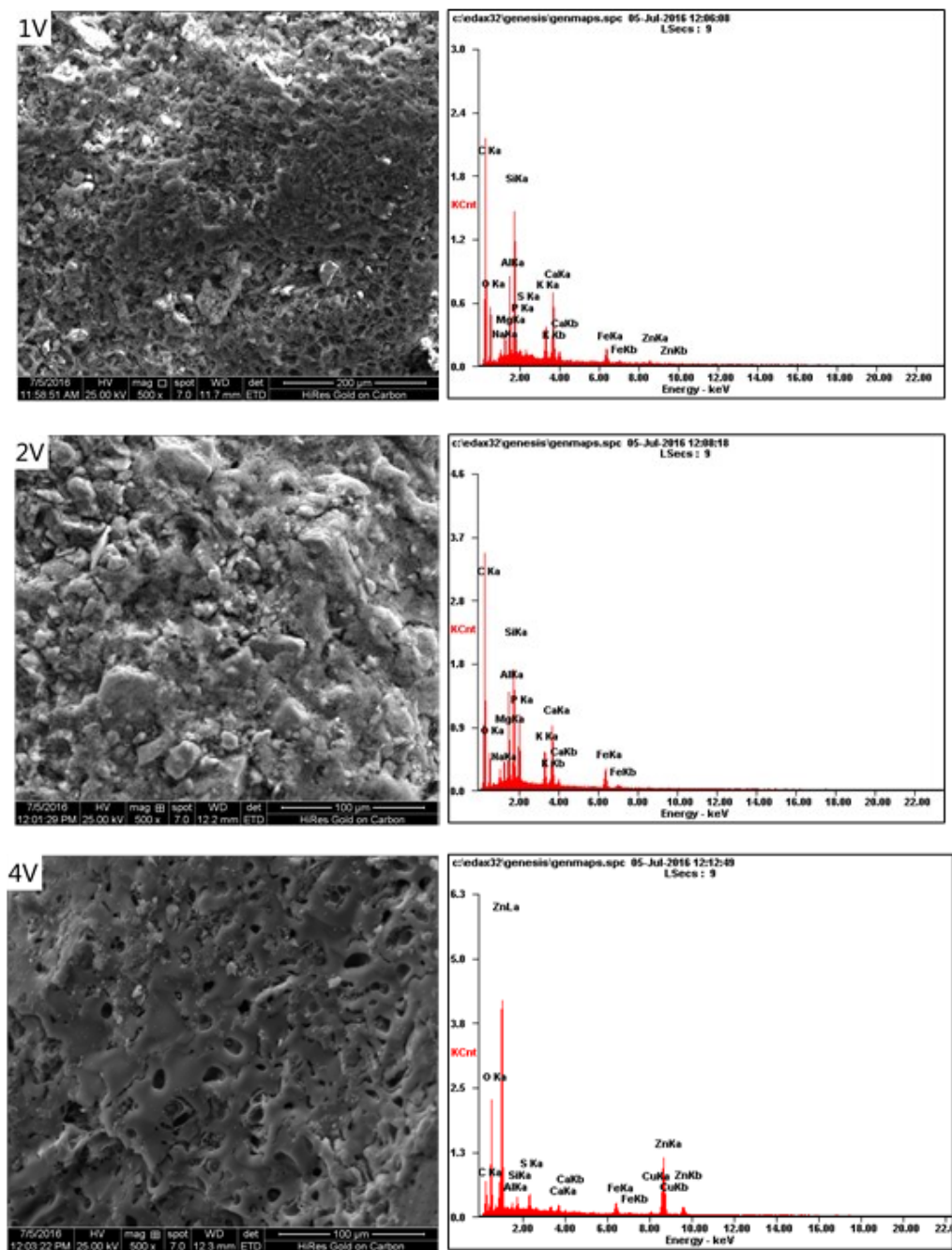


Figure 6. SEM-EDS analysis of particle electrodes at different voltages

lent to a small electrolytic cell, and adsorption and oxidation-reduction reactions occur at both ends. The electro-deposition reaction takes place on the cathode, and this finding indicates that the interaction between the electrode and the ion is shortened, and the conversion capacity is increased. Under the action of the electric field, the loading of the electric field can increase more active sites and electro-catalysis reaction site [19–23]. Therefore, the metal electrode load of the particles is high, and the removal of harmful ions in wastewater is more efficient at 4 V than at other voltages.

The removal mechanism of each ion in wastewater at different voltages is shown in Fig. 7.

4. CONCLUSION

The self-made coal-based electrode material is used as an anode and cathode electrode and the activated carbon is utilized as a particle electrode. The applied voltage influences the removal rate of the ions and the reaction mechanism. In particular, the removal rates of CN_T , Cu, Zn, SCN^- , and CN^- in the solution increase as the applied

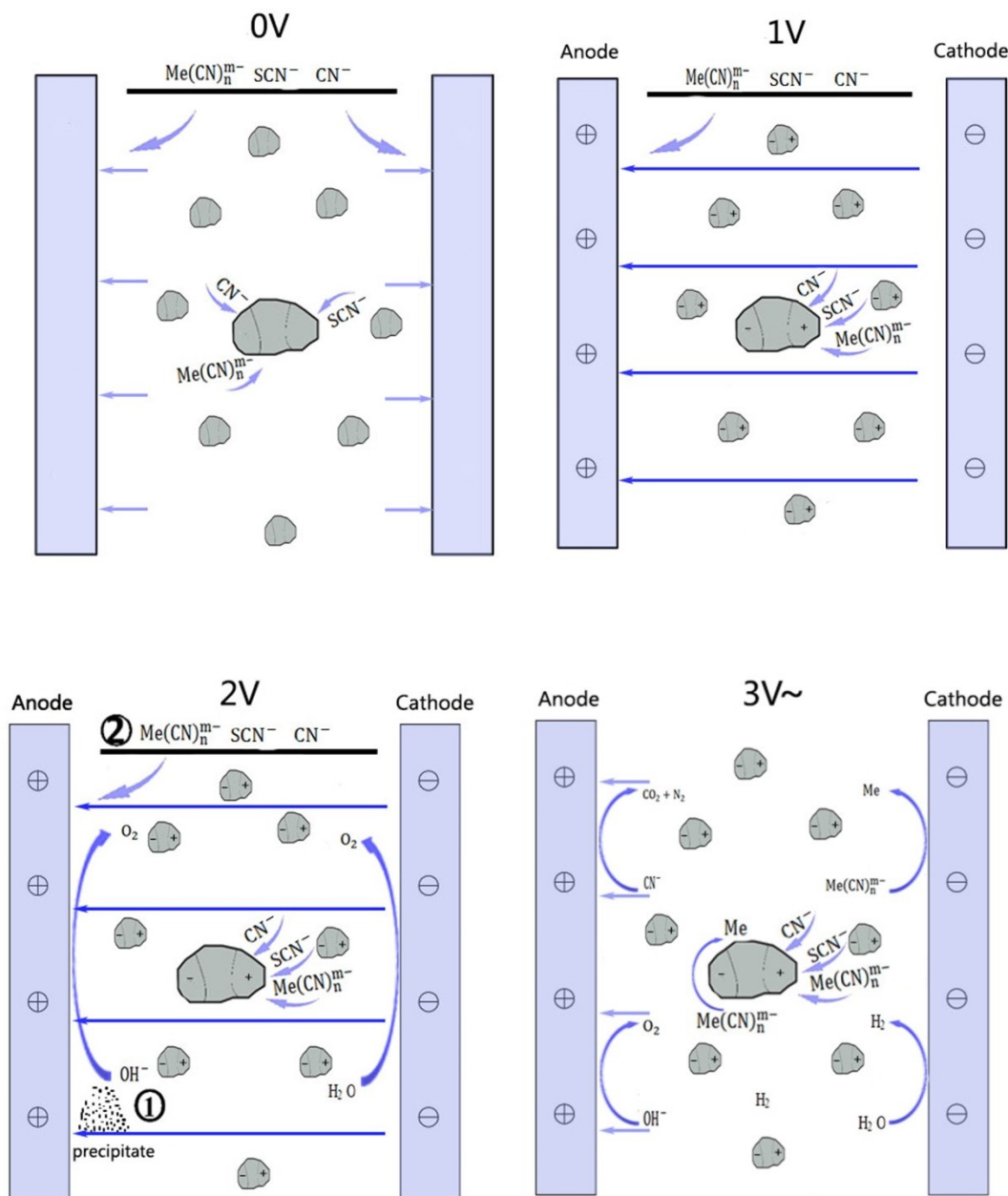


Figure 7. Schematic diagram of the mechanism under different voltages

①- CuSCN, CuCN, Zn(CN)₂, Cu₂Fe(CN)₆, Zn₂Fe(CN)₆, Cu₃Fe(CN)₆·4H₂O ②- Me-Cu, Zn, Fe

voltage increases. Under the optimum reaction conditions, the removal rates of these ions are 93.94%, 95.22%, 97.23%, 99.38%, and 94.93%, respectively. At 0 V, the adsorption capacity is the main effect, and 1 V is mainly based on electrical adsorption. In these conditions, no electrode reaction occurs on the cathode and the particle electrode surface, and the removal rate of each ion is low. At 2 V, the main reactions at particle electrode and cathode are electro-adsorption and enrichment-precipitation, while the oxidation reaction of OH⁻ takes place at the anode. As the applied voltage is further increased to more than 3 V, the electro-adsorption

of at the anode is gradually weakened, and the metal ions are mainly removed through the electro-deposition at the cathode. CN⁻ and SCN⁻ are finally converted to CO₂ and N₂ through the oxidation reaction at the anode.

5. ACKNOWLEDGMENTS

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