Removal of Gaseous Elemental Mercury in a Diffusion Electrochemical Reactor Based on a Three-Dimensional Electrode

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ABSTRACT: A novel three-dimensional electrochemical reactor with nickel foam and carbon paper used as the anode and stainless steel mesh used as the cathode was studied in this research. Oxidation mercury removal is performed in a self-made diffusion reactor. The influence of the electrolysis voltage, pH, gas flow, and other factors on mercury removal is discussed, as well as the mechanism of anodization mercury removal is explored. The experimental results show that nickel foam has a significant effect on the removal of Hg⁰, and 80–85% removal can be achieved under optimal conditions. Meanwhile, nickel foam has stable performance at high temperatures (60 °C) and in strong alkaline electrolytes, which also play an effective role in anodized oxidation. Although carbon paper is more stable than nickel foam and less affected by experimental factors, it is sensitive to reaction temperature and can only work in the neutral electrolyte at low temperatures. In contrast, electrochemical catalytic oxidation technology using the nickel foam is more promising for Hg⁰ removal.

1. INTRODUCTION

Mercury has received global attention due to its volatility, high toxicity, durability, easy migration, and bioaccumulation.¹ ² A large number of emission inventory measurements reveal that natural, anthropogenic, and re-emission mercury are the three major sources of mercury emission. Coal combustion is considered as one of the main anthropogenic atmosphere mercury emission sources. Therefore, mercury emission control in coal-fired power plants has become a research hotspot.

In flue gas, it is generally believed that there are three forms of mercury species, i.e., particulate mercury (Hg⁰), oxidized mercury (Hg²⁺ or Hg⁴⁺), and gaseous elemental mercury (Hg⁰), in which the main form of elemental mercury (Hg⁰) during the fire enter the atmosphere with the flue gas and cause pollution to the atmosphere, water, and soil environment. It can also enter the body through the digestive, skin, or respiratory tract to cause damage to the health of the organism.³

Recently, there has been a growing interest in how to control mercury pollution. Hg⁰ can be captured by the existing equipment electrostatic precipitator or bag filter, and Hg²⁺ can be absorbed by the wet flue gas desulfurization equipment of power plants owing to its strong water solubility.⁷–¹⁰ However, Hg⁰ is difficult to be captured by the existing pollutant control equipment due to its low water solubility and strong volatility.¹¹,¹² Therefore, Hg⁰ emitted from coal-fired flue gas is one of the main man-made pollution sources, and the removal technology urgently needs to be optimized. At present, the application of adsorption is increasing, scholars mainly focus on the study of modified adsorbents, such as halogen-loaded activated carbon.¹⁶–²¹ The adsorption technology is a mature mercury capture technology, yet the high price of mercury adsorbent limits the application of mercury adsorption technology. Catalysis oxidation technology has been proven to be an effective Hg⁰ oxidation method that can transfer insoluble Hg⁰ into soluble Hg²⁺, which can be subsequently removed by a wet flue gas desulfurization process.²² The research tended to focus on exploring the oxidation methods for Hg⁰. However, the catalysis activity of catalysts is seriously suppressed by NH₃, SO₂, and SO₃.²³–²⁵ The electrochemical oxidation process mainly relies on electron transfer without secondary pollution and can be carried out at normal temperature and pressure without an external catalyst. It includes two modes of oxidation: indirect oxidation of the cathode and direct oxidation of the anode. The indirect oxidation of the cathode generates H₂O₂ in situ by the two-electron process of oxygen reduction, and H₂O₂ can oxidize the contaminants. The direct oxidation of the anode is one of the main man-made pollution sources, and the removal technology urgently needs to be optimized. At
previous research\textsuperscript{30} found that the gas diffusion electrode (GDE)\textsuperscript{28,31} could overcome the problem of low H\textsubscript{2}O\textsubscript{2} yield of the traditional cathode electrode and oxidize Hg\textsuperscript{0} to achieve the flue gas demercuration. Direct oxidation of the anode directly oxidizes Hg\textsuperscript{0} by reaction with the electrode. To improve the efficiency of anodization, it is necessary to choose a suitable anode material, enhance the contact time, and increase the contact area between the gas flow and the electrode. Studies have shown that Ni, Cu, etc. have good reactivity,\textsuperscript{32} among which the nickel foam is the electrode or electrode substrate commonly used due to its three-dimensional network structure, good electrical conductivity, high porosity, large specific surface area, and good electrode performance. Nickel foam is often used as a catalyst carrier,\textsuperscript{33} and its special network structure can make water or gas pass smoothly. Also, the carbon paper also has a similar porous structure. Produced during the papermaking production, the carbon paper can fully exert its conductive protection function and is often used as a diffusion electrode of a proton-exchange membrane fuel cell. The large electroactive surface areas and unique porous structure of nickel foam\textsuperscript{34−36} and carbon paper\textsuperscript{37,38} are ideal for enlarging the electrochemical active site as well as enhancing mass transfer. These two materials enable a large amount of gas to diffuse into the electrodes’ interior and surface and communicate with the electrolyte at the same, so the gas—liquid—solid phase is fully contacted to achieve an effective electron transport.

In this work, nickel foam and carbon paper were applied for Hg\textsuperscript{0} removal from simulated flue gas at a diffusion electrochemical reactor below 80 °C. The nickel foam and carbon paper are characterized by cyclic voltammetry (CV), inductively coupled plasma emission spectrometer (ICP), and the mercury detector. Different addition concentrations were used to explore the effect on the mercury oxidation efficiency and select the optimum addition concentration. The effects of the electrolysis voltage, pH, gas flow, and other factors on the Hg\textsuperscript{0} removal performance are investigated. The mercury electrochemical oxidation on nickel foam mechanism was investigated via combining the experimental data and theoretical simulation calculation. Therefore, we choose the nickel foam electrode as the anode and the stainless steel mesh as the cathode in the self-designed reactor to study the electrochemical oxidation of Hg\textsuperscript{0}.

2. RESULTS AND DISCUSSION

2.1. Electrochemical Characterization. The cyclic voltammetry (CV) is an electrochemical analysis method widely used to study the properties, mechanism, and kinetic parameters of electrode processes. CV is used to test the electrochemical performance of nickel foam electrode and carbon paper electrode. To avoid the effect of the oxygen evolution reaction (OER), the scanning electrochemical window is usually controlled below the oxygen evolution potential. The electric double-layer capacitance is also an important factor affecting the electrochemical surface-active area of the electrode, whose relationship with the charging current can be expressed by eq 1\textsuperscript{39}

\[ i_{\text{charging}} = C_{\text{double-layer}} V \]  

The nickel foam electrode (15 × 15 mm\textsuperscript{2}), Pt electrode, saturated calomel electrode, and Na\textsubscript{2}SO\textsubscript{4} solution (0.5 M) were used as the working electrode, auxiliary electrode, reference electrode, and electrolyte solution, respectively. The cyclic voltammetry curve at different scan rates (0.01−1 V/s) between −0.4 and 0.6 V is shown in Figure 1a. The CV curve of the carbon paper electrode (40 × 40 mm\textsuperscript{2}) is shown in Figure 1c.

As shown in Figure 1a,c, the cyclic voltammetry curves are close to a rectangle and the coverage area is large, indicating that the nickel foam and carbon paper have good electrochemical activities. Meanwhile, as the scan rate increases, the response current also increases. Due to the porous nature of
the two electrodes and the predominant diffusion control during the reaction, the response current is proportional to the mass transfer rate.

The current of nickel foam anode is relatively stable between −0.1 and 0.1 V (vs RHE), so the average value of this current is selected for the electric double-layer capacitance calculation (Figure 1b). For carbon paper anode, the average current of 0−0.2 V (vs RHE) is selected (Figure 1d). The electric double-layer capacitance of the carbon paper is calculated to be $4 \times 10^{-8}$ F, similar to that of the titanium plate ($3.5 \times 10^{-8}$ F). Also, the capacitance of nickel foam is $2.6 \times 10^{-6}$ F, which is 74.2 times that of the titanium plate, and the electrochemical performance is improved.

2.2. Direct Oxidation of Hg$^0$ with Different Anodes.

The reaction are carried out under following conditions of pure air flow rate (100 mL/min), water bath temperature (50 °C), electrolyte solution Na$_2$SO$_4$ (0.5 M, 80 mL), pH (6), voltage (4 V), and reaction at room temperature (25−30 °C). After optimization, the optimal conditions should be selected for subsequent experiments.

2.2.1. Effects of Electrolytic Voltage on Hg$^0$ Removal. The magnitude of voltage affects the ability of electron transfer and generation of strong oxidation groups, which determines the capability of the system to remove Hg$^0$. Figure 2a shows the effect on Hg$^0$ removal at 3, 4, and 5 V. In the first 10 min of the electrolytic reaction, the removal of Hg$^0$ increases rapidly but becomes stable within 10−30 min at 4 and 5 V. However, the removal efficiency fluctuates after 40 min at 3 V. It is indicated that the lower voltage, the lower reaction rate, and the longer time are required to achieve a balance. Meanwhile, under the three selected voltages, the anode undergoes a water decomposition reaction to generate oxygen, which promotes the indirect oxidation of the cathode, but it has a little impact on the difference in the results. The final efficiencies are 60, 72, and 72%, respectively, which show the lower effect of voltage on the ability of nickel foam to oxidize Hg$^0$ when the voltage is over 4 V. The consumption increases with voltage under the same efficiency, so 4 V is selected in subsequent experiments.

As shown in Figure 3a, under the condition of 3−5 V, the removal of Hg$^0$ can reach more than 75%, especially at 3 V (>85%). The concentration of Hg$^0$ is low, and the removal difference under unequal voltages is less than 10%. Therefore, it can be considered that the voltage has little effect on the removal with a carbon paper electrode. According to the test result, 3 V was chosen for further studies.

2.2.2. Effects of Flue Gas Flow on Hg$^0$ Removal. The effect of flue gas flow is manifested in two ways: one is the influence of the residence time of mercury on the three-phase interface and the other is the mass transfer effect. Electrolysis is carried
out for 40 min at different air flow rates (25, 50, and 100 mL/min), achieving the removal of 53, 71, and 86%, respectively (Figure 2b). As the gas flow rate increases, the removal of Hg⁰ and the reaction rate also increase. At 100 mL/min, the efficiency stabilizes after 40 min, while the other two air flow rates were the opposite. The result shows that the oxidation rate increases as the gas flow increases. The possible causes are that more oxygen in the solution accumulates with the increase of gas flow, thereby accelerating the direct oxidation rate. However, the smaller the gas flow, the longer the gas stays at the interface and it is possible to bring out some adsorbed Hg⁰, not increase but decrease on Hg⁰ removal in the early reaction time at 25 and 50 mL/min. In addition, the rise of the air flow can improve the mass transfer, accelerating the generation of strong oxidizing groups by indirect oxidation into the solution, which will oxidize part of Hg⁰ in the upper layer solution, thereby improving the removal. As a result, a gas flow of 100 mL/min was selected for subsequent experiments.

Using carbon paper as the anode, the flue gas flow has a significant effect on the reaction rate but limited effect on the removal of Hg⁰. As shown in Figure 3b, at 25 mL/min, the final equilibrium is not reached after 40 min; at 100 mL/min, equilibrium is reached in 15 min. However, as the gas flow rate increases, the carbon papers easily caused cracks so that the removal decreases after 25 min. The effect of flow on carbon paper is similar to that of nickel foam, which increases the direct oxidation rate and enhances indirect oxidation.

2.2.3. Effects of Initial Mercury Concentrations on Hg⁰ Removal. In the chemical reaction, the increasing concentration of reactants will exacerbate the intermolecular collision and increase the reaction rate. However, in the electrochemical oxidation system, the number of activating molecules is limited, and it is necessary to study the effects of initial mercury concentration on the removal.

We adjusted the temperature of the water-bath to control the initial concentration (490−1380 μg/m³). As shown in Figure 2c, mercury removal is significantly increased from 72 to 85% as the initial concentration of Hg⁰ is increased from 490 to 1000 μg/m³ but the oxidation rate all tends to become stable after 10 min of electrolysis. The removal efficiency drops into 75% as the concentration of Hg⁰ increased to 1380 μg/m³. However, the total amount of Hg⁰ removed is 1030 μg/m³, higher than 1000 μg/m³. There are two reasons for the analysis. First, the increase in the initial concentration of mercury adds the Hg⁰ passing through the electrode within the same time and the same area and promotes the direct oxidation efficiency with sufficient oxygen. Second, higher initial concentration of mercury produces more intermediates (such as Hg⁺), which compete for strong oxidizing groups, limiting the system’s ability to oxidize Hg⁰.
For carbon paper, as the initial concentration increases from 200 to 800 μg/m³, the removal is almost constant (Figure 3c), so it can be considered that a good removal effect can be achieved regardless of the initial concentration of Hg⁰.

2.2.4. Effects of Electrolyte Concentration on Hg⁰ Removal. In the electrochemical reaction, the electrolyte concentration is an important factor affecting the reaction result. The 0.05 M, 0.1 M, and 0.5 M Na₂SO₄ (aq.) solutions were used as electrolytes for 40 min electrolysis, and the results are shown in Figure 2d. The increase of Na₂SO₄ concentration is beneficial to Hg⁰ removal, which reached 56–85% at the end. The best removal was achieved for the 0.5 M electrolyte but fails to equilibrate during the 40 min electrolysis. The possible reasons to explain the phenomenon are as follows.¹⁵,⁴⁴,⁴⁵ (1) increase in electrolyte concentration accelerates the electron transfer and improves the conductivity and mass transfer of the solution, thereby improving the removal, and (2) related studies have found that SO₄²⁻ of the electrolyte can become SO₄⁻ with active radicals. Although the oxidation ability of SO₄⁻ is less than that of active radicals, it is sufficient to oxidize Hg⁰. Hence, with the increase of electrolyte concentration, the concentration of sulfate ion increases and improves the removal.

When carbon paper is used as the anode, the removal of Hg⁰ rises as the concentration of the electrolyte increases. In the three concentrations of the experiment (0.05 M, 0.1 M, and 0.5 M), the removal is 87% under 0.5 M Na₂SO₄ and more than 75% under the other two solutions. As shown in Figure 3d, the removal and reaction rate are close at different electrolyte concentrations, and the reaction effect is relatively stable.

2.2.5. Effects of pH on Hg⁰ Removal. Take 80 mL of 0.5 M Na₂SO₄ (aq.) as an electrolyte and adjust the pH to 3, 6, 9, and 11 using 1 M NaOH (aq.) and 1 M H₂SO₄ (aq.). After 40 min of electrolysis, although the removal reaches 80% at pH 6 and 11 (shown in Figure 2e), the total mercury removal at pH 11 is higher than that at pH 6 (960 and 800 μg/m³, respectively). Therefore, when the electrolyte is alkaline, the mercury removal effect is better than that under other pH conditions. However, the removal is only 61% at pH 3 and 67% at pH 9 (shown in Figure 2e). This phenomenon can be explained as follows. The alkaline solution promotes the direct oxidation reaction of the anode. In addition, the concentration of OH⁻ in electrolyte increases with the elevated pH, thus the anode semireaction of water decomposition becomes more intense and more oxygen is produced on the anode. Oxygen is further reduced to H₂O₂, and active radicals at the cathode promote the oxidation reaction.⁴¹,⁴⁶,⁴⁷ The H₂O₂ is extremely unstable to decompose into strong oxidizing groups under alkaline conditions but is relatively stable under acidic conditions.⁴⁰,⁴³ It can be easily known that direct and indirect oxidations synergistically increase the removal under alkaline conditions, while acidic conditions limit the oxidizing ability and decrease the removal.

As shown in Figure 3e, the removal of Hg⁰ corresponding to pH of 3, 5, 7, 9, and 11 are 78, 87, 87, 72, and 68%, respectively, and is best at pH 7. With the enhancement in acidity or alkalinity of the electrolyte, the removal of Hg⁰ drops significantly. The reason for this phenomenon could be that H₂O₂ is relatively stable under acidic conditions and the indirect electrooxidation effect is weakened. Under alkaline conditions, the electrolytic properties of carbon paper are degraded and the direct electrooxidation effect is weakened.

2.2.6. Effects of Reaction Temperature on Hg⁰ Removal. Since the actual temperature of the coal-fired flue gas is higher than room temperature, we study whether the temperature has an effect on the electrochemical system. The heating belt was wound around the reactor, and the solution was heated to 30, 45, and 60 °C by adjusting the voltage. The results are shown in Figure 2f. As the reaction temperature increases, the Hg⁰ removal decreases slightly, and the change can be almost ignored. A comparison of the total amount of mercury removal at three temperatures, about 750–820 μg/m³, also indicates that the temperature has little effect on the reaction system. This phenomenon is different from that described in related literatures.⁸³ In theory, the solubility of O₂ decreases with the increased temperature, weakening indirect oxidation and thus reducing the removal. According to this difference, it can be explained that this reaction is dominated by direct oxidation.

For carbon paper, the removal of Hg⁰ is 87% at the temperature of 25 °C but is only 30–40% when the temperature reaches 50–80 °C (Figure 3f). The higher the temperature, the worse the removal effect. It can be said that carbon paper is not suitable for use at high temperatures. To be applied to the actual flue gas, the flue gas must be cooled first to achieve a better removal.

According to the above analysis of influence factors, the optimum reaction conditions for Hg⁰ removal using nickel foam are electrolysis voltage of 4 V, gas flow of 100 mL/min, water bath temperature of 50 °C (initial mercury concentration of about 1000–1100 μg/m³), 80 mL of 0.5 M Na₂SO₄ (aq.) as an electrolyte, pH of 11, the reaction temperature of 30 °C. Under these experimental conditions, the removal of Hg⁰ can reach 80–85% after 40 min of electrolysis.

Unlike nickel foam, carbon paper is mainly affected by the reaction temperature and flue gas flow, and other factors have almost negligible effects. Carbon paper can only achieve a high removal rate under conditions of low temperature, neutral electrolyte, and suitable flow. The practical application of carbon paper is limited, considering the temperature and the gas flow of the actual flue gas. Conversely, the nickel foam can achieve 85% removal efficiency at low voltages in neutral or strong alkaline electrolytes with high flow and has practical application value.

2.3. Reaction Mechanism of Hg⁰ Oxidation. Under optimal conditions, the initial Hg⁰ concentration is 800 μg/m³ and the Hg⁰ concentration at the end is 230 μg/m³. After the electrolysis is complete, the electrolyte is taken to determine the mercury content by ICP. The result showed that the mercury content is less than 0.05 mg/L.

Assuming that Hg⁰ in the solution is oxidized to form Hg⁰⁺ or HgCl⁺ instead of forming an alloy with the metal, the theoretical concentration of mercury ions is 0.03 mg/L (eq 2). The theoretical concentration is consistent with the ICP results. It can be inferred that the assumption is true and the reaction mechanism of anodized Hg⁰ can be analyzed. Hg⁰ (g) diffuses to the gas—liquid—solid three-phase interface with the carrier gas and loses electrons on the nickel foam (anode) to form Hg²⁺ or HgCl⁺ (as shown in Figure 4). In the electrolyte, the oxidized mercury will further to form precipitates. Meanwhile, during the study, it is found that green floculent precipitates exist in the solution after the electrode has reacted many times. Also, the precipitate is analyzed as Ni(OH)₂ via dimethylglyoxime spectrophotometric detection. It can be inferred that some of the atoms in the nickel foam dissolved as the reaction proceeded, and metal ions further react with OH⁻.
in the solution to form a precipitate. The main reaction formula is as follows (eqs 3−7).

\[
\frac{(800 \times 10^{-5} \text{mg/L} - 230 \times 10^{-5} \text{mg/L}) \times 40 \text{ min} \times 100 \text{ mL/min}}{80 \text{ mL}} = 0.03 \text{ mg/L}
\]

(2)

Anode:

\[
2\text{Hg}^0 + \text{SO}_4^{2-} \leftrightarrow \text{Hg}_2\text{SO}_4 + 2e^-
\]

(3)

\[
\text{Hg}^{2+} \leftrightarrow 2\text{Hg}^{2+} + 2e^-
\]

(4)

\[
4\text{OH}^- \leftrightarrow \text{O}_2 + 2\text{H}_2\text{O} + 4e^-
\]

(5)

\[
\text{Ni} + 2\text{OH}^- \leftrightarrow \text{Ni(OH)}_2 + 2e^-
\]

(6)

Cathode:

\[
2\text{H}_2\text{O} + 2e^- \leftrightarrow \text{H}_2 \uparrow + 2\text{OH}^-
\]

(7)

3. CONCLUSIONS

The diffusion electrochemical reactor is prepared using nickel foam as an anode and stainless steel mesh as a cathode and applied to the removal of elemental mercury (Hg\(^0\)) from simulated flue gas. The Hg\(^0\) capture performance can be efficiently enhanced at low temperatures. The Hg\(^0\) removal performance of the nickel foam is better than those of the carbon paper. We investigate the effects of voltage, gas flow, initial Hg\(^0\) concentration, pH, and reaction temperature on Hg\(^0\) removal and determined that the optimal conditions for the reaction were 4 V, gas flow of 100 mL/min (initial mercury concentration of about 1000–1100 \(\mu\)g/m\(^3\)), 80 mL of 0.5 M Na\(_2\)SO\(_4\) (aq.) as an electrolyte, pH of 11, and reaction temperature of 30 °C. The removal performance of Hg\(^0\) could reach 80–85%. In addition, the reaction mechanism is also explored. Carbon paper is also applied as an anode to the electrochemical system, but it is found that the removal of 87% is achieved only in low-temperature and neutral electrolytes with a certain gas flow. Compared with nickel foam, carbon paper has more application restrictions.

4. EXPERIMENTAL SECTION

4.1. Experimental Apparatus and Procedures. The self-made reactor is shown in Figure 5. Figure 5b is a stainless steel mesh (200 mesh, Shanghai Xin’an Metal Screen Co., Ltd., China) as a cathode and placed in the upper part of the reactor; Figure 5c is a nickel foam as an anode and placed in the lower part of the reactor; between them is a plexiglass board (10 mm thick); the electrolyte solution is sodium sulfate; and the pH of the solution was adjusted with 1 M sodium hydroxide and 1 M sulfuric acid. All reagents were of analytical grade.

4.2. Analytical Methods. The experimental procedure\(^{30,49}\) (Figure 5) consists of three parts: simulated flue gas generating device; electrochemical reaction device; and elemental mercury detection system. The air is fully mixed with the mercury
vapor, which is generated by mercury permeation tube (QMG-6-6, Suzhou Qing’an Instrument Co., Ltd., China) in the U-shape tube to form simulated flue gas. Then, the simulated flue gas enters the electrochemical reaction system through the lower part of the self-made reactor and stays at the gas–liquid–solid three-phase interface for a period of time to be reacted by turning on the power source. After the reaction, the gases pass through the electrode and enter the mercury detector (SG-921, Jiangsu Jiangfen Electroanalytical Instrument Co., Ltd., China) from the upper outlet of the reactor to detect the concentration of Hg\textsubscript{0} in real-time. The experiment also set up an exhaust gas absorption bottle to treat the remaining gas. The concentration of mercury in the solution is monitored by ICP (inductively coupled plasma emission spectrometer).

According to the principle of cold atomic absorption spectroscopy, mercury vapor strongly absorbs the characteristic line at the wavelengths at 253.7 nm so that the concentration of Hg\textsubscript{0} in the gas can be measured.

Mercury removal is calculated using eq 8, where \( C_i \) is the outlet Hg\textsubscript{0} at a certain time and \( C_{\text{initial}} \) is the initial concentration.

\[
\eta(\%) = \frac{C_{\text{initial}} - C_i}{C_{\text{initial}}} \times 100
\] (8)

The concentration of mercury ions in the electrolyte after electrolysis was measured via ICP technology.

The electrochemical performances of the electrodes were calculated and characterized using the cyclic voltammetry curve.

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**Notes**

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