Research Paper

Electrooxidation of nickel-ammonia complexes and simultaneous electrodeposition recovery of nickel from practical nickel-electroplating rinse wastewater

Wei Guan^{a,b}, Shichao Tian^c, Di Cao^a, Yutong Chen^a, Xu Zhao^{a,d,*}

^a Key Laboratory of Drinking Water Science and Technology, Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences, Beijing 100085, PR China

^b Chongqing Key Laboratory of Environmental Materials & Remediation Technologies, Chongqing University of Arts and Sciences, Chongqing 402160, PR China

^c Environmental Science and Technology Laboratory, Tsinghua-Berkeley Shenzhen Institute, Shenzhen 518055, PR China

^d University of Chinese Academy of Sciences, No. 19(A) Yuquan Road, Shijingshan District, Beijing 100049, PR China

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ABSTRACT

In this experiment, a combined electrooxidation-electrodeposition (EO-ED) system using RuO₂/Ti anode and stainless steel cathode was employed to treat nickel-ammonia complexes wastewater, which simultaneously achieved the decomplexation of nickel-ammonia complexes and recovery of nickel metal. Effects of current density, pH value and reaction temperature on electrochemical performance were analyzed. When the current density was 32 mA/cm², pH was 9.0 and the temperature was 60 °C, the nickel recovery percentage was 99% and the ammonia removal efficiency was 70%. Combined with the SEM-EDX, XRD and XPS analysis for the cathode surface, it was confirmed that the generation of deposition on the cathode surface was zero-valent nickel. Firstly, Nickel-ammonia complexes were destroyed by electrooxidation process, generating Ni²⁺ ions and free NH₃. Then, Ni²⁺ ions were reduced onto the cathode and free NH₃ were further oxidized. Stability of the system for nickel-ammonia complexes treatment was examined by repeated experiments for fifteen successive cycles. The electrochemical system exhibited a satisfactory stability for nickel recovery and ammonia removal, achieving nickel recovery percentage of 85–95% and ammonia removal efficiency of 65–70%, respectively.

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1. Introduction

The nickel-ammonia complexes in wastewater has attracted the attention due to its large discharge and high toxicity to the ecosystem, which mainly comes from industrial process such as electroplating industry, hard alloy, high efficiency catalysts, coated materials and multilayer ceramic capacitors [1,2]. Ammonia and nickel ions (Ni²⁺) can form metal complexes, which is difficult to be removed by traditional chemical precipitation [3,4]. Advanced oxidation processes have been largely investigated for treating refractory organics and organic complexes. Applications of photocatalysis [5], sonochemical process [6] and H₂O₂/ultraviolet (UV) process [7] in treatment of metal-binding complexes have been

studied. Therefore, a reliable and cost-effective method for treating nickel-ammonia complexes is desired.

Previous studies on the removal or recovery of nickel ions included adsorption [8], ion-exchange [9], electrodialysis [10] and electrodeposition [11]. These methods have high removal efficiencies for nickel ions in wastewater. However, these methods are limited for ammonia removal. Biotreatment, electrooxidation and UV/chlorine methods have been used for ammonia removal, but the recovery efficiency of nickel ions is limited using these methods [12–14]. Some electrochemical techniques such as electrocoagulation, electrodeionization and electrodeposition [15–17] are usually used for the recovery of dissolved metal ions from industrial effluent streams. However, these methods are limited for treating metal complexes [18,19].

Although some researches on nickel recovery and ammonia removal have been done, the treatment of nickel-ammonia complexes and simultaneous recovery of nickel from wastewater in pilot or full-scale studies is rarely reported [20–22]. The environmental, technological and economic methods need to be

* Corresponding author at: P.O. Box 2871, 18 Shuangqing Road, Haidian District, Beijing, 10085, PR China.

E-mail address: zhaoxu@rcees.ac.cn (X. Zhao).

addressed with additional work, such as the optimum design of the electrochemical cells/reactors and the determination of optimal operating conditions [23,24]. Electrolysis has been considered as a promising technology for ammonia removal due to its high robustness, versatility and amenability to automation [25,26]. However, oxidation of nickel-ammonia complexes at the anode and the recovery of nickel ions at the cathode have not been achieved in practical nickel-electroplating rinse wastewater treatment.

Herein, decomplexation of nickel-ammonia complexes and simultaneous recovery of nickel could be achieved by the combination of electro-oxidation and electro-deposition. In this experiment, effects of current density, pH value and reaction temperature were analyzed. The cathode surface was analyzed by SEM-EDX, XRD and XPS techniques. The mechanism of reaction for treating nickel-ammonia complexes was explored.

2. Experimental

2.1. Materials

Sodium perchlorate, sodium hydroxide, nitric acid and sulfuric acid (98% w/w) were purchased from Sinopharm Chemical Reagent Co., Ltd. Deionized water was used for preparation and dilution of all solutions. RuO₂/Ti anode and stainless steel cathode were purchased from Beijing Henglitai Corporation. The RuO₂/Ti electrode used in this system was called dimensionally stable anodes (DSA), which had more advanced performance (catalytic activity, stability and low cost) than those of conventional electrodes (metal, carbon, etc) [27]. The target wastewater was the practical nickel-electroplating rinse wastewater. The initial concentration of nickel ions was 2156 ± 50 (mg/L) and ammonia concentration was 9680 ± 50 (mg/L). The initial pH of the solution was 8.8 ± 0.2.

2.2. Experiment procedures

The experiments were performed in a glass reactor with 250 mL and the sodium perchlorate acted as an electrolyte. The electrochemical system included a direct-current (DC) power supply, an electrolytic cell, a stainless steel cathode, a RuO₂/Ti mesh anode and a magnetic stirring apparatus. The electrochemical experiments were carried out at a constant current density, which was controlled by a DC power supply (DH1718E-6, Dahua Electronic, Co., Ltd., Beijing, China). The RuO₂/Ti electrode (70 mm × 40 mm × 0.5 mm) was directly connected to the positive electrode of the DC power supply and the stainless steel electrode with the same area was directly connected to the negative electrode of the DC power supply. The reaction solution was stirred magnetically at a speed of 450 rpm and heated by magnetic stirring apparatus. The power of the magnetic stirring apparatus was 15 W (OS20-Pro, Dragon Laboratory Instruments Limited, Beijing, China). Effects of current density (8, 16, 24, 32 and 40 mA/cm²), pH values (3.0, 5.0, 7.0, 9.0 and 11.0) and reaction temperature (20, 40, 60 and 80 °C) on the treatment of nickel-ammonia complexes were investigated. The pH was adjusted by H₂SO₄ (0.2 M) or NaOH solution (0.2 M).

2.3. Analysis

Concentration of Ni²⁺ ions was measured using a 710 series inductively coupled plasma optical emission spectrometer (ICP-OES, Agilent Technology, U.S.A.). Concentration of ammonia was detected according to the standard methods and pH value was measured with a pH meter [28]. The deposition on the cathode was analyzed by scanning electron microscope (SEM, model 6700F,

Tokyo, Japan) and energy dispersive X-ray analysis (EDX) (JEOL, Japan). X-ray powder diffraction (XRD) patterns were obtained with an X'Pert PRO Powder diffractometer machine (PANalytical Co.) by using Ni-filtered Cu K_α radiation from 10° to 90° [29]. X-ray photoelectron spectra (XPS) was used to analyze the surface variation of the electrode using a PHI Quantera SXM (PHI-5300/ESCA, ULVAC-PHI, INC) [30].

3. Results and discussion

3.1. Effect of current density on electrochemical performance

The performance of electrochemical system for Ni recovery and ammonia removal was investigated. The equations about recovery percentage of Ni ions (RP) and removal efficiency of ammonia (RE) were defined using the following Eqs. (1)–(2) [31] :

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

Where C₀ is the concentration of nickel ions at the initial time, mg/L; C_t is the concentration of nickel ions at a given time, mg/L.

$$RE = \frac{C_0 - C_t}{C_0} \times 100\% \quad (2)$$

Where C₀ is the concentration of ammonia at the initial time, mg/L; C_t is the concentration of ammonia at a given time, mg/L.

The Effect of current density on nickel ions recovery and ammonia removal was investigated. As shown in Fig. 1(a), corresponding to the current density of 8 mA/cm², 16 mA/cm², 24 mA/cm², 32 mA/cm² and 40 mA/cm², the recovery percentage of nickel ions are 29.14%, 77.83%, 90.12%, 98.26% and 99.03% after a 180 min reaction. The recovery percentage of nickel ions is increased with increasing current density. The recovery rate of nickel ions is fast and the kinetics process is current density control in the first 2 h (Figure SM-1). In addition, the increasing current density enhances the removal efficiency of ammonia (Fig. 1(b)).

The electrode potentials were measured as 1.0 V, 2.6 V, 3.8 V, 4.2 V and 6.0 V for the applied current densities of 8 mA/cm², 16 mA/cm², 24 mA/cm², 32 mA/cm² and 40 mA/cm², respectively. The increased current density would improve electrode potential in the system, so the oxidation ability of the anode was enhanced. In addition, the nickel ions were reduced onto the cathode via Eq. (3).

But when the current density increased from 32 to 40 mA/cm², the recovery percentage of nickel ions remained nearly unchanged. The kinetics process was changed from current control to mass transport control. When the current density was 32 mA/cm², the recovery percentage of nickel ions was 98%. In the actual wastewater treatment, both of the recovery efficiency of metals and low energy consumption were taken into account. Therefore, the optimal current density for this system was 32 mA/cm².

3.2. Effect of pH value on the electrochemical performance

Recovery efficiency of nickel ions and removal efficiency of ammonia at various pH values were analyzed. As shown in Fig. 2(a), the recovery efficiency of nickel ions is increased with increasing pH value. Comparing with recovery efficiency of nickel ions at different pH conditions, after a 180 min reaction, the recovery efficiency of nickel at pH = 9.0 and pH = 11.0 is the same. But insoluble hydroxides (such as Ni(OH)₂) at the cathode surfaces are easily formed at high pH condition (pH > 10.0), which influences the purity of nickel recovery. However, in the

Fig. 1. Effect of current density on electrochemical performance: (a) Recovery percentage of nickel ions and (b) Removal efficiency of ammonia (Initial concentration of nickel ions, 2156 mg/L; pH, 9.0; reaction temperature, 40 °C).

electrochemical system, no isolation of the reactor was provided and nickel carbonate could be deposited on the electrode in alkaline media. The pH value played an important role on the morphology of the deposition. As shown in Fig. 2(b), the removal efficiency of ammonia is higher in alkaline condition. According to the report, the oxidation of ammonia required high pH values (pH=9.0) [32]. The ammonia in water exists in two forms: ammonium ions and free ammonia. The proportion of free ammonia is increased with increasing pH via the Eq. (4).

The removal efficiency of ammonia rapidly increased from pH=9.0 to pH=11.0 (Figure SM-2). At higher pH condition, ammonia was transformed to ammonia gas and vitalized to air, which required further treatment. In addition, the initial pH of the solution was 8.8 ± 0.2 , without addition of H_2SO_4 or NaOH solution to adjust the pH. Therefore, the optimal pH for this system was 9.0.

3.3. Effect of reaction temperature on the electrochemical performance

Recovery efficiency of nickel ions and removal efficiency of ammonia in various temperatures were analyzed. As shown in Fig. 3(a), the reaction temperature has an obvious effect on the recovery of nickel ions. When the temperature is higher than 60 °C, the recovery efficiency of nickel ions increases in the initial 90 min, and remains unchanged with the extension of reaction time. When the temperature is lower than 40 °C, the recovery percentage of nickel ions is increased continuously with the extended reaction time. A different trend is observed in the EO process of ammonia (Fig. 3(b)). The removal efficiency of ammonia is obviously changed during the last 90 min. These interesting results imply that nickel-ammonia complexes are destroyed by EO process in the initial 90 min, and Ni^{2+} ions are reduced onto the cathode at the same time. Then, free NH_3 generated from the destruction of nickel-ammonium complexes is further oxidized by the EO process. The removal efficiency of ammonia is increased quite quickly from

Fig. 2. Effect of pH value on the electrochemical performance: (a) Recovery percentage of nickel ions and (b) Removal efficiency of ammonia (Initial concentration of nickel ions, 2156 mg/L; current density, 32 mA/cm²; reaction temperature, 40 °C).

Fig. 3. Effect of reaction temperature on the electrochemical performance: (a) Recovery percentage of nickel ions and (b) Removal efficiency of ammonia (Initial concentration of nickel ions, 2156 mg/L; current density, 32 mA/cm²; pH, 9.0).

Fig. 4. Morphology and element distribution analysis of the cathode electrode: (a) optical camera, (b) SEM image and (c) EDX (Initial concentration of nickel ions, 2156 mg/L; current density, 32 mA/cm²; pH, 9.0; reaction temperature, 60 °C).

60 °C to 80 °C (Figure SM-3). In addition, considering the energy consumption, the optimum temperature for treating nickel-ammonia complexes was 60 °C.

3.4. Surface variation of the cathode

The deposition on the surface of stainless steel cathode was investigated by SEM-EDX, XRD and XPS. As shown in Fig. 4(a, b), there is some deposition on the cathode. In the process of nickel-ammonia complexes oxidation, nickel-ammonia is destroyed and nickel ions are reduced onto the cathode. Meantime, EDX spectra of the deposition on the cathode after a 180 min reaction are shown in Fig. 4(c). EDX spectra results declare that the major elements of the deposition are Ni and O. Therefore, the results indicate the deposition on the cathode contains Ni element.

The structural properties of the deposition on the cathode were further investigated by XRD in the range of 10–90°. As shown in Fig. 5, the pronounced diffraction peaks in the XRD pattern at 2θ of 44.80°, 52.75° and 77.52° are corresponded to (110), (200) and (220) Bragg's reflections of monoclinic Ni phase in accordance with the standard [33]. Furthermore, the intensity of the diffraction peaks is increased as the reaction is prolonged. The result indicates that the zero-valent nickel is gradually crystallized during the ED process. X-ray photoelectron spectroscopy (XPS) was used to analyze the chemical nature and oxidation state of the

electrodeposited film on the cathode. As shown in Fig. 6, the XPS spectra of the Ni2p core level binding energy shows that the major Ni2p_{3/2} appears at 856.15 eV and 861.50 eV. In addition, the major Ni2p_{1/2} peaks centers at 874.26 eV. The results indicate the deposition on the cathode is assigned to zero-valent nickel. Thus, it

Fig. 5. XRD patterns of deposit on the cathode in the electrodeposition process (Initial concentration of nickel ions, 2156 mg/L; current density, 32 mA/cm²; pH, 9.0; reaction temperature, 60 °C).

Fig. 6. X-ray photoelectron spectra (XPS) analysis of the deposit on the cathode in the electrodeposition process (Initial concentration of nickel ions, 2156 mg/L; current density, 32 mA/cm²; pH, 9.0; reaction temperature, 60 °C).

is concluded that the nickel ions are reduced onto the cathode by the electrochemical process [34].

3.5. A proposed electrooxidation pathway of nickel-ammonia complexes

In the electrochemical system, the nickel-ammonia complexes were destroyed by anode oxidation in the initial 90 min, generating Ni²⁺ ions and free NH₃. Then Ni²⁺ ions were reduced on the cathode and the free NH₃ were further oxidized by the generated HOCl. The mechanism of ammonia oxidation had been widely studied [35,36]. The ammonia was decomposed via direct (non-mediated) electro-oxidation and indirect oxidation by active radicals such as hydroxyl radicals and hypochlorite ion [37]. In the electrochemical system, the Cl₂ was generated in situ by Cl⁻ ions losing two electrons on the anode and then converted to hypochlorous acid in alkaline condition. The ammonia could be completely oxidized by hypochlorous acid and most of the ammonia was converted into N₂. The reactions result from the indirect oxidation by HOCl as shown in the following Eqs. (5)–(10).

During the oxidation process of ammonia, the catalytic activity of the anode decreased due to the adsorbed NH_{ads} on the surface of the anode. The formation mechanism of NH_{ads} and NH_{2(ads)} could be described by the following Eqs. (11)–(14) [38].

Therefore, the stability of this electrochemical system for destroying nickel-ammonia complexes should be analyzed.

3.6. Stability of the electrochemical system and energy consumption

Stability of the system for destroying nickel-ammonia complexes was studied by repeating the experiment for fifteen successive cycles. Before each cycle, the anode and cathode were washed with dilute nitric acid and then washed by deionized water. As shown in Fig. 7, the recovery percentage of Ni and removal efficiency of ammonia remains nearly constant for fifteen cycles. Nickel-ammonia complexes are oxidized by the anode and then nickel ions are reduced to zero-valent nickel. Thus, the system exhibits high stability for ammonia removal and nickel recovery.

Energy consumption is an estimate for the degradation of 1 g contaminant in an electrochemical system. During the oxidation process of nickel-ammonia complexes, the main energy consumption includes DC power and the magnetic stirring apparatus. It is calculated according to the following equation [39]:

$$W(\text{kWh/g}) = \frac{(U * I + P) * t}{\Delta X * 1000} \quad (15)$$

Where U is the voltage (V), I is the current intensity (A), t is the reaction time in h, P is the power of magnetic stirring apparatus (15 W), ΔX is the recovery quality of Ni ions and the unit is milligramma (mg). According to Table 1, under the optimal conditions for nickel recovery and ammonia removal, the energy consumption is 0.11 × 10⁻³ kWh/g.

Fig. 7. Variation of nickel concentration and ammonia concentration for the repeated experiments for fifteen successive cycles (Initial concentration of Ni²⁺ ions, 2156 mg/L; current density, 32 mA/cm²; pH, 9.0; temperature, 60 °C).

Table 1

The energy consumption for recovery of 1 g Ni under different current densities.

Current density (mA/cm ²)	Voltage (V)	Current intensity (A)	Energy (kWh/g)
8	1.0,	0.225	0.29×10^{-3}
16	2.6	0.450	0.16×10^{-3}
24	3.8	0.675	0.14×10^{-3}
32	4.2	0.900	0.10×10^{-3}
40	6.0	1.125	0.12×10^{-3}

4. Conclusions

In this experiment, nickel-ammonia complexes were totally removed by a combination of electrooxidation and electrodeposition processes. Following the satisfactory results obtained from the experiment, the higher current density enhanced the oxidation ability of the anode. In addition, the pH and temperature influenced the existence form of ammonia in water. Therefore, when the current density was 32 mA/cm², pH was 9.0 and temperature was 60 °C, it was highly efficient for electrooxidation of nickel-ammonia complexes and electrodeposition recovery of nickel from wastewater. The mechanism of Ni recovery and ammonia removal was proposed. The nickel-ammonia complexes were destroyed by anode oxidation in the initial 90 min, generating Ni²⁺ ions and free NH₃. Then Ni²⁺ ions were reduced onto the cathode and the free NH₃ was further oxidized by the generated HOCl. After the repeated experiments for fifteen successive cycles, the system exhibited high stability for ammonia removal and nickel recovery.

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Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at <http://dx.doi.org/10.1016/j.electacta.2017.06.121>.

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