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Recovery of nickel and water from wastewater with electrochemical combination process

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ABSTRACT

Owing to its precious value and high toxicity, nickel recovery from wastewater is a worthwhile work. However, the concentration of Ni^{2+} varies from tens to thousands of mg/L due to different wastewater sources, no process can simultaneously meet the requirements of metal recovery and water reuse, both for high concentration wastewater (HCW, $\geqslant$500 mg/L) and low concentration wastewater (LCW, <500 mg/L). In this paper, the feasibility of nickel recovery and water reuse was investigated using electrochemical combination processes: electrolysis (EL), electrodialysis (ED), and electrodeionization (EDI). First, nickel containing wastewater were separately treated by EL, ED and EDI, effects of there operating parameters, voltage, initial Ni^{2+} concentration, pH and water flux on the nickel recovery and water quality were investigated and optimized. Then, an EL–ED–EDI combination process was developed for nickel containing wastewater treatment. The results showed that almost 99.8% of nickel could be recovered with the purity of 93.9% and almost 100% of water could be reused (Ni^{2+} concentration was less 1 mg/L) by the combination process, with the energy consumption of EL $\approx$ 25.7 kW h/kg, ED $\approx$ 0.5 kW h/m^3 and EDI $\approx$ 0.2 kW h/m^3.

1. Introduction

Due to its high corrosion resistance, toughness, and super alloys, nickel is considered as a strategic metal, widely used in various industries such as electroplating, nonferrous metal, mineral processing, paint formulation, stainless steel, batteries manufacturing, and forging [1–4]. Accordingly, a large number of wastewater containing Ni^{2+} produced from these industries, the concentration of Ni^{2+} varied from tens to thousands of mg/L due to different wastewater sources [5,6]. Ni^{2+} is a kind of non-biodegradable toxic heavy metal ion present in wastewater. Trace amounts of Ni^{2+} are beneficial to human organism as an activator for some enzyme systems, but higher concentrations of Ni^{2+} may cause different types of diseases such as lung cancer, renal edema, skin dermatitis, gastrointestinal disorder, headache, dizziness, chest pain, dry cough, rapid respiration, cyanosis, and extreme weakness [7–12]. In order to protect the environment and human health, the maximum level of Ni^{2+} discharged is 1 mg/L which was established by the Environmental Protection Agency of China [13]. Therefore, it is necessary to develop some effective and inexpensive methods to remove or recover nickel from water. Additionally, in recent years, the prices of non-ferrous metals have risen drastically due to the increased global demand, especially nickel, the price is more than US$16/kg in China. So, from the viewpoint of environmental protection and resource saving, nickel recovering and wastewater reusing is strongly expected, and an effluent-free technology should be developed.

Chemical precipitation is the most common method for removal of dissolved metals from wastewater. However, it produces precipitation sludge which may lead to secondary pollution [14]. Ion exchange and adsorption are also conventional methods of Ni^{2+} removal, but limited by the capacity of exchange or adsorption, they can only used for low concentration wastewater treatment, and how to deal with the saturated adsorbents and ion exchange resins are still annoying problems. Recently, some researches used electrochemical methods such as EL, ED and EDI in heavy metal wastewater treatment, for their capabilities of heavy metal recovery as well as water reuse [15–22].

Heavy metal can be recovered by EL in one stage without the necessity of sludge disposal and regeneration of saturated ion exchanger resin. It is reported that over thirty kinds of heavy metals can be recovered from aqueous solutions by EL, including nickel [23]. But, the main problem of EL is the high energy consumption in treating low concentration wastewater and the effluent usually cannot meet

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the discharge standard [24]. ED is one of the most recent technologies that applied to the separate plating chemicals from rinse water. Meanwhile the ED for acidic aqueous solution that contains nickel ions has been studied by several workers. Advantages of ED are that the low concentration of heavy metals can be concentrated and the other part of effluent water can be diluted for reuse. However, it does not fit for high concentration due to the restriction of membrane pollution [25]. EDI is a hybrid method that includes ED and ion exchange, in which the purifying compartments and sometimes the concentrating compartments of the electrodialysis stack are filled with ion-exchange resins. An important application for EDI is the production of pure water. Now, some studies have been done on the application of EDI process treating heavy metal wastewater [19–22].

In our previous study [18], copper wastewater can be recycled successfully with the combination process of EL–ED. Comparing with copper, the recovery of nickel is more difficult and challenging, because the electro-deposition of nickel on cathode is difficult due to the hydrogen evolution reaction, and the recovery efficiency is low [25]. Based on the previous experience of copper containing wastewater treatment, this time, a new technology (EDI) was introduced into our system for nickel containing wastewater treatment, coupled with EL and ED technologies. The aim is to become the heavy metal wastewater into resources.

For the purpose of nickel recovery and water reuse, our work was divided into two parts. In the first part, EL, ED and EDL were separately used for the experiments of nickel wastewater treatment, to investigate their proper range of nickel concentration and optimize their operating parameters. In the second part, EL, ED and EDL were integrated into a complete water treatment system, to investigate the feasibility of nickel recovery as well as water reuse, both for high and low concentration wastewater. Additionally, the purity of recovered nickel, the quality of reused water, and the consumed energy were also analyzed.

2. Materials and methods

2.1. Materials

Nickel-containing wastewater were prepared by diluting $NiSO_4 \cdot 6H_2O$ (purity 99.0%) with tap water (the conductivity of tap water ≈300 μs/cm). 0.1 M NaOH and H_2SO_4 were used for pH adjustment in order to approach the actual wastewater. Nickel concentration was monitored by atomic absorption spectrometry (Solaar M6, Thermo Elemental, USA). Power supplies (PS-303D, Hongkong Longwei Ltd. China) were used to provide stable voltage. A pH meter and a conductivity meter (PHSJ-4A, DDSJ-308A, Shanghai Leici Ltd., China) were used for the determination of pH and conductivity. The morphology and elements distribution of recovered copper were analyzed by SEM/EDS (XL-30, Philips) and Stereo Microscope (SMZ660, Nikon). All chemicals used here were analytical grade.

2.2. Setups

As shown in Fig. 1, the EL system mainly consists of three parts: EL unit, power supply and water supply. The EL unit (3.7 L) contained four anode plates and three cathode plates with an area of $21 \times 14\ cm^2$, and the distance between each electrode plates is 1 cm. The anode plates made from titanium grid covered with iridium and ruthenium oxides, and the cathode plates made from graphite. Cathodes are all smooth plates, but anodes were punched dozens of holes on plates which can increase the turbulence within the unit and thus greatly improve current efficiency. Moreover, an air pump was installed at the bottom of the electrolysis unit to keep the water always in turbulence state.

The ED system mainly consists of three parts: ED unit, power supply and water supply, as shown in Fig. 2a. The ED unit consists of 110 membrane pairs packed in two stacks, and the membrane active surface is $26 \times 15\ cm^2$. The cation exchange membrane CEM (YLM001) and the anion exchange membrane AEM (YLM201) were purchased from the Shanghai Chemistry Plant (China).

Similar with the ED system, the EDI system consists of EDI unit, power supply and water supply, as shown in Fig. 2b. The EDI unit consists of 13 diluted chambers and 13 concentrated chambers, one cathode chamber and one anode chamber, with the active surface $28 \times 14\ cm^2$. All diluted and concentrated chambers were filled with mixed cation resin (D113) and anion resin (D103) with the volume ratio of 1:1.5.

2.3. Methods

Taguchi method was used for design the experiments of EL and ED, which is an efficient and systematic approach to optimize designs for performance, quality, and cost, especially when the number of factors is high [26]. Nickel recovery (R_n), water recovery (R_w), energy consumption of EL (E_{el}), energy consumption of ED (E_{ed}), energy consumption of EDI (E_{edi}) and separation efficiency (η_s) were calculated as Eqs. (1)–(6):

$$R_n = \frac{C_{in} - C_{out}}{C_{in}} \times 100\% \quad (1)$$

$$R_W = \frac{Q_7}{Q_1 + Q_2} \times 100\% \quad (2)$$

$$E_{el} = \frac{\sum U \cdot I \cdot t}{(C_{in} - C_{out}) \cdot Q_{in} \cdot t} \quad (3)$$

$$E_{ed} = \frac{\sum U \cdot I \cdot t}{1000 \cdot (Q_4 + Q_5) \cdot t} \quad (4)$$

$$E_{edi} = \frac{\sum U \cdot I \cdot t}{1000 \cdot (Q_6 + Q_7) \cdot t} \quad (5)$$

$$\eta_s = \frac{C_{ie} - C_{de}}{C_{ie}} \times 100\% \quad (6)$$

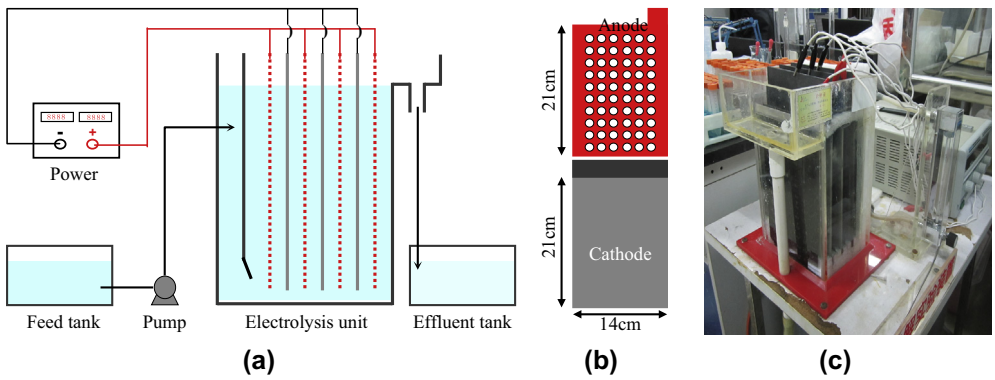

Fig. 1. Schematic of (a) EL system, (b) graphite cathode and Ti/Ir, Re anode, and (c) real photo.