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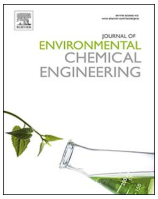

Leaching of nickel from a secondary source by sulfuric acid

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ABSTRACT

In the present study, leaching of tailing containing 29.6 wt.% Ni in sulfuric acid was investigated. Tailing was characterized by X-ray powder diffraction (XRD), X-ray fluorescence (XRF) and inductively coupled plasma (ICP-OES) methods. Rinsing process for the separation of tungsten from tailing was conducted in distilled water at 80 °C for 240 min. After the filtration of solid residue from tungsten solution by vacuum filtration, batch leaching tests were carried out on solid residue at atmospheric pressure in stirred acid solution. The effects of sulfuric acid concentration (1–4 mol), pulp density (1/10–1/20 gmL^{-1}), particle size (−600 +420 to −180 +125 mm) and temperature (25–80 °C) on the nickel dissolution were studied. The results showed that leaching of about 99% of nickel content of the washed tailing could be achieved using 4.0 mol sulfuric acid, pulp density of 1/20 gmL^{-1} and particle size of −180 +250 mm at 80 °C after 240 min. Kinetic analysis of the leaching data properly followed the shrinking core model, and the dissolution rate of nickel was found to be controlled by diffusion through the product layer for 25 and 40 °C and chemical reaction for 80 °C. Leaching rate of nickel at 60 °C at the first stage (0–60 min) was controlled by diffusion through the product; then, at the second stage, it was controlled by the chemical reaction (60–240 min).

1. Introduction

Nickel is one of the important and strategic metals because it has many applications in petrochemical, metallurgical, electroplating and other industries. It is harmful for several animal species, micro-organisms and plants. Large utilization of nickel products leads to environmental pollution [1,2]. There are not many significant mineral resources of nickel. Therefore, it is essential to consider the secondary sources of nickel coming from various industry wastes. It exists in various sources such as spent batteries [3,4], catalysts [5], wastewater [6–8] and electronic wastes [9,10].

Two different routes can be employed for nickel recovery from wastes: (i) leaching of wastes and subsequently recovering Ni from solution (hydrometallurgical process); (ii) thermally treating wastes (pyrometallurgical process) [11]. Pyrometallurgical processes are not a suitable method for nickel recovery from spent battery because rare earth elements that exist in a spent battery will be lost in the slag [12]. Moreover, pyrometallurgical separation of Ni from Co is not feasible [13].

Bartolozzi et al. [14] described a hydrometallurgical process for the recovery of nickel and cadmium from Ni-Cd spent batteries. After the primary treatment (grinding, drying, mechanical separation), leaching of powder obtained from primary treatment was conducted with H_2SO_4 and H_2O_2 at pH = 0.3 for 60 min at 60 °C. As a result, 98% nickel was recovered in this process. Nogueira and Margarido [15] investigated the leaching behavior of electrode materials of spent nickel–cadmium batteries in sulfuric acid media. The first chemical step of their process was sulfuric acid leaching. Hydroxide phases of Ni, Cd and Co were readily soluble at low acid concentration (e.g. pH ≈ 1) for 30 min at room temperature. They reported that electrode metallic nickel leached completely at high temperature (95 °C) and mild acid concentration (2.5 mol H_2SO_4) after 4 h. Kim et al. [16] investigated the recovery of nickel, copper, tin and lead from waste multi-layer ceramic capacitors (MLCC) by hydrometallurgical process. They reported that the extraction of nickel was 97% with 1 mol HNO_3 at 90 °C for 90 min and 5 gL^{-1} pulp densities.

It is reported that sulfuric acid can be used for the leaching of Ni containing wastes and nickel sulfate is commercially important due to its usage in electroplating [17]. Some studies have demonstrated that sulfuric acid is more efficient than hydrochloric acid for nickel recovery from spent catalyst [18]. Also, 99% of nickel is recovered when using acid concentration of 50%, particle size of below 500 mm, pulp density of 1/12 and time of 5 h at 100 °C [11]. Sahu et al. [19] studied the recovery of nickel from a spent catalyst by H_2SO_4 as a leaching agent

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Table 1
Chemical analysis of the raw material.

component	Na	Mg	Al	Si	Ca	Ti	Mn	Fe	Co	Ni	Cu	W	Ta
Amount (wt%)	1.89	0.15	0.04	0.65	1.8	0.05	0.03	3.25	0.79	29.6	9.76	8.45	0.60

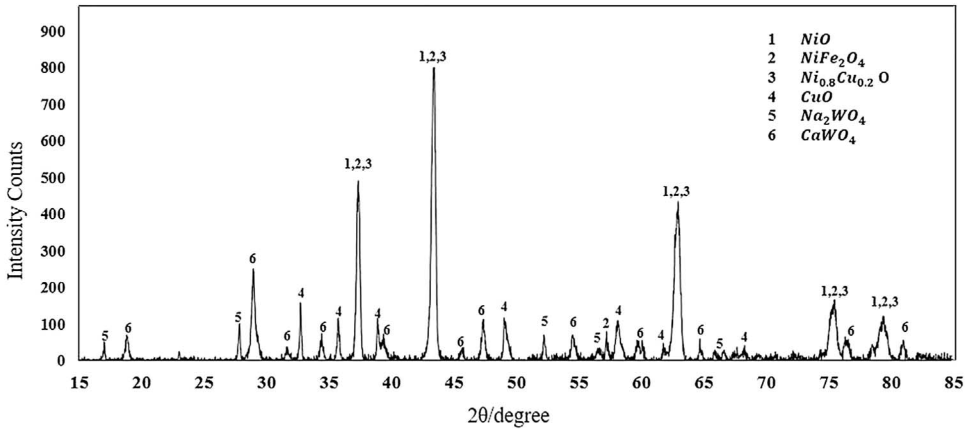

Fig. 1. XRD analysis of the raw material.

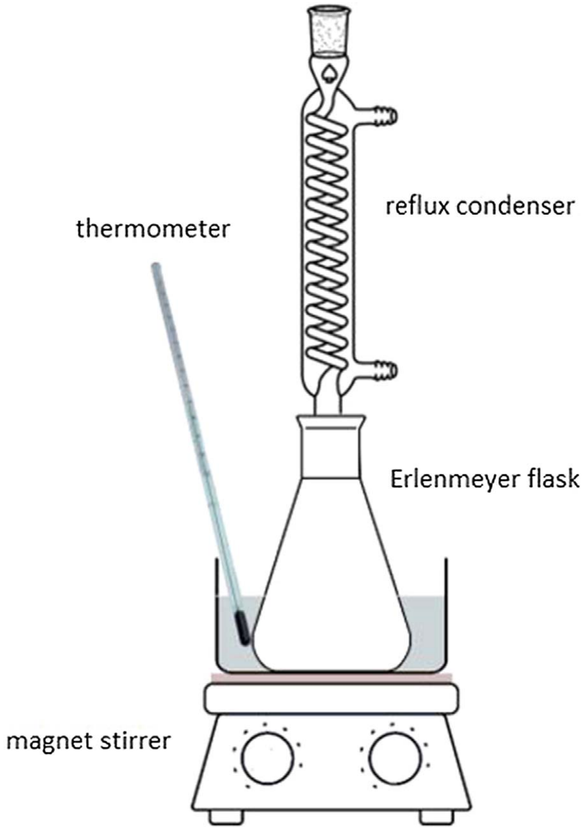

Fig. 2. Apparatus used for rinsing and leaching processes.

and showed that the dissolution of nickel was a diffusion controlled process. An increase in temperature and sulfuric acid concentration improved nickel recovery. Marafi and Stanislaus [20] showed that the leaching efficiency of all the valuable metals present in the spent catalyst (Mo, V and Ni) can be improved using ultrasonic agitation in a short time at relatively low temperature.

Kinetic studies on the leaching process of zinc plant residue containing nickel (Ni filter cake) were performed by Gharabaghi et al. [21]. They indicated that diffusion through the product layer was the rate-controlling step during dissolution. Safarzadeh et al. [22] studied the dissolution kinetics of cadmium of a Cd-Ni filter cake and reported that leaching of cadmium in sulfuric acid at 25–55 °C and time of less than 5 min followed a shrinking core model and diffusion through layer products was proposed to be the reaction rate controlling mechanism.

Nazemi et al. [23] introduced different approaches to evaluate kinetics of leaching processes based on shrinking core model. Less calculations were required in their process and the contribution of each of the steps in controlling the process can be predicted. Simultaneous action of steps which act in series in their method were combined together as follows:

$$t = \tau_F X + \tau_P\left(1 - 3(1-X)^{2/3} + 2(1-X)\right) + \tau_R\left(1 - (1-X)^{1/3}\right) \tag{1}$$

$$\tau_F = \frac{\rho_s R_0}{3k_l C_{Ab}}$$

$$\tau_P = \frac{\rho_s {R_0}^2}{6D_e C_{Ab}}$$

$$\tau_R = \frac{\rho_s R_0}{k_s C_{Ab}}$$

where X is fraction of reacted nickel, ρ_s is density (kg m^{-3}), R_0 is initial radius of particle (m), k_l is mass transfer coefficient of the liquid film (m^3liquid/(m^2surface s)), k_s is reaction rate constant at the particle surface (s^{-1}), C_{Ab} is concentration of sulfuric acid (kgm^{-3}), D_e is effective diffusion coefficient in porous structures (m^3fluid/(msolid s)), τ_F, τ_P and τ_R are time for complete dissolution by liquid film, product diffusion and chemical reaction control (min), respectively. t (in Eq. (1)) and t_i (in Eq. (2)) are time of reaction and start of second stage (min), respectively. The contribution of each resistance can be obtained by placing the experimental data in the above formula and calculating the coefficients. These coefficients are calculated by a multi-linear regression analysis using the least square technique. Without considering boundary conditions, coefficients could get negative, which is not acceptable by definition. As a result, the above equations are rewritten by applying boundary conditions as follows:

$$\varphi = \sum_i \left[\left(\tau_F X + \tau_P\left(1 - 3(1-X)^{2/3} + 2(1-X)\right) + \tau_R\left(1 - (1-X)^{1/3}\right)\right) - t_i\right]^2 \tag{2}$$

Min φ subject to τ_F, τ_P and $\tau_R > 0$.