



A multistep leaching of nickel-based superalloy scrap for selective dissolution of its constituent metals in hydrochloric acid solutions

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ABSTRACT

A selective two-step leaching for nickel and rhenium from PWA 1484 nickel-based superalloy scrap in hydrochloric acid solutions was investigated. The scrap was easily comminuted under 150 μm by a pyro-metallurgical pretreatment at 1500 $^{\circ}\text{C}$, viz. the formation of Al_3Ni intermetallic compound with aluminum. At the first leaching step at 90 $^{\circ}\text{C}$ and 40 g/L solid-to-liquid(S/L) ratio in 4.0 mol/L hydrochloric acid solution, over 95% of nickel was dissolved in 60 min with aluminum, cobalt, and chromium, whereas 97.6% of rhenium was remained with tantalum in the residue. The leaching percentage of rhenium was increased with the S/L ratio from 2.4% at 40 g/L S/L ratio to 25% at 200 g/L S/L ratio. The second leaching step for rhenium from the residue was carried out using the electrogenerated chlorine as an oxidant at 80 $^{\circ}\text{C}$, 0.43 mmol/min chlorine supply, and 10 g/L S/L ratio in 4.0 mol/L HCl solution. Over 99% of rhenium was leached in 20 min, leaving tantalum in the residue.

1. Introduction

Nickel-based superalloys have been commonly used for high temperature applications, e.g., turbine blade material for aircraft jet engines and gas turbines of stationary power plant, because of their excellent mechanical strength and oxidation resistance at high temperature (Srivastava et al., 2014). In particular, adding rhenium (Re) can improve their properties. PWA 1484 used in this study is the second-generation alloy containing 3%(w/w) Re, which was developed by Pratt & Whitney (Cetel and Duhl, 1992). The material cost of 3%(w/w) Re is equivalent to 60% of the total. Therefore, even though the superalloy scrap contains many valuable metals, the recovery of Re from the scrap is the most important.

The tough chemical and physical properties of superalloys make recycling difficult and complex. Numerous processes have been proposed to recycle valuable metals in the superalloy scrap (Hundley and Davis, 1991; Srivastava et al., 2014). The processes fall into two categories: pyrometallurgical, and hydrometallurgical processes. The hydrometallurgical processes can be selective and produce pure metals, whereas the pyrometallurgical processes are not selective for pure metals. An electrochemical process containing pyrometallurgical pretreatment, electrodissolution, and electrodeposition in double-membrane electrolytic cell has also been tried (Redden and Greaves, 1992). The superalloy scrap can also be recycled by sequential pyrometallurgical and hydrometallurgical processes. The Bureau of Mines

reported two techniques for recycling bulk superalloy scrap (Atkinson and Nicks, 1986). The first technique includes melting the scrap with aluminum (Al) to form intermetallic compounds that are brittle and easily crushed. The crushed materials from the melts react rapidly with acid solutions to dissolve the metals. The second technique includes dissolving superalloy scrap in molten zinc, followed by distillation of the zinc. A friable residue containing all of the metals present in the original superalloy can be easily crushed, and readily leached with mineral acids. Most of the valuable base metals in superalloy scrap, except rhenium, can be easily leached by acids. It has been reported that rhenium is dissolved in chlorine (Cl_2) water, hydrogen peroxide, and hot concentrated oxidizing acids, such as nitric acid and sulfuric acid, whereas it is insoluble in bases and non-oxidizing acids (Schweitzer and Pesterfield, 2010).

In this study, we propose a multistep leaching process for the selective leaching of metals of interest from PWA 1484 nickel-based superalloy scrap. In particular, the target metals are nickel (Ni) and rhenium; nickel is the most abundant metal of PWA 1484, whereas rhenium is the most expensive element. For the selective leaching of valuable metals from the PWA 1484 superalloy scrap, the intermetallic compound formation with Al was adopted as a pretreatment for easy comminution, and then a hydrometallurgical step was followed to selectively leach the metals. Without the pretreatment for easy grinding under micrometer size, the superalloy scrap over millimeter size practically did not react with hydrochloric acid because of its tough

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chemical resistive property. The selective leaching consists of two steps: an acid leaching, followed by an electrogenerated chlorine leaching. The first acid leaching can effectively dissolve base metals, such as nickel, aluminum, cobalt, and chromium, while rhenium is remained in the residue. The subsequent electrogenerated chlorine leaching of the residue can dissolve Re, and generates the final residue that contains totally insoluble metal. It is necessary to use Cl_2 as an oxidant involving expensive Cl_2 -resistant equipments: electrochemical cell for Cl_2 generation, chlorine gas transport tubing & valve system, closed leaching cell, and innocuous treatment system of unconsumed Cl_2 gas. Comparing with a one-step leaching using Cl_2 that can dissolve Ni and Re simultaneously, the proposed multistep leaching has a merit that can minimize the scale of Cl_2 -resistant equipment and chlorine leaching operation.

The effects of experimental variables, such as acid concentration, solution temperature, S/L ratio, and Cl_2 feeding rate, were investigated for developing an optimum condition for selective Ni and Re leaching, with higher Ni and lower Re dissolution at the first leaching, and higher Re at the second leaching step.

2. Experimental

Fig. 1 shows a schematic of the selective leaching system, which comprises two parts: a leaching reactor, and an electrochemical cell. Depending on the volume of the leaching solution used, a double-walled pyrex glass cell of 0.5 or 1.0 L capacity was used as the leaching reactor. The electrochemical cell connected to the leaching reactor with polytetrafluoroethylene (PTFE) tube was operated only at the Re leaching step for Cl_2 electrogeneration. The electrochemical cell was separated by a 28 cm^2 anion exchange membrane (Neosepta, AMX); anodic and cathodic compartments were filled with 300 mL of 1.0 mol/L hydrochloric acid (HCl) solution in which chlorine is generated, and 600 mL of 35% HCl solution, respectively. The leaching was operated as a closed system, and the solution temperature was controlled within $\pm 0.1 \text{ }^\circ\text{C}$ using a heating bath. A mechanical seal stirrer was used to agitate the leaching solution. Unconsumed chlorine from the leaching reactor was trashed via potassium iodide solution for harmless treatment.

Fig. 2 shows an experimental flow of the selective leaching of Ni and

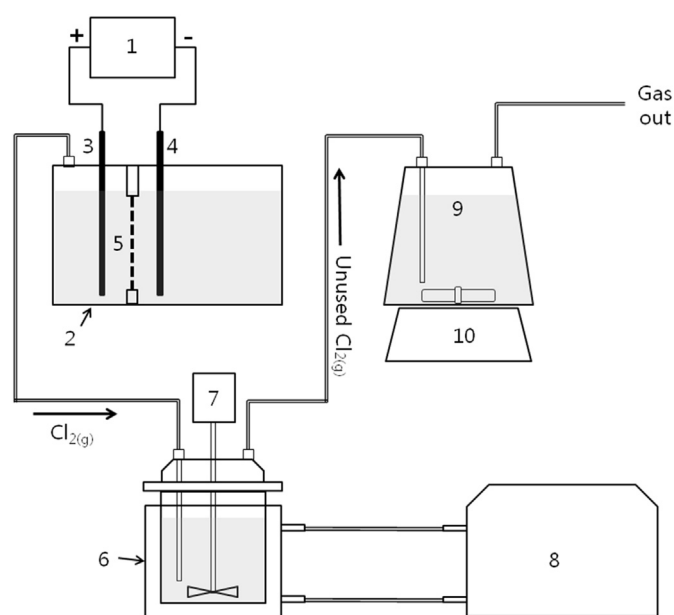


Fig. 1. A diagram of the experimental apparatus (1: power supply; 2: electrochemical cell; 3 & 4: graphite electrode; 5: anion exchange membrane; 6: glass leaching reactor; 7: stirring motor; 8: heating bath circulator; 9: glass bottle; and 10: magnetic stirrer).

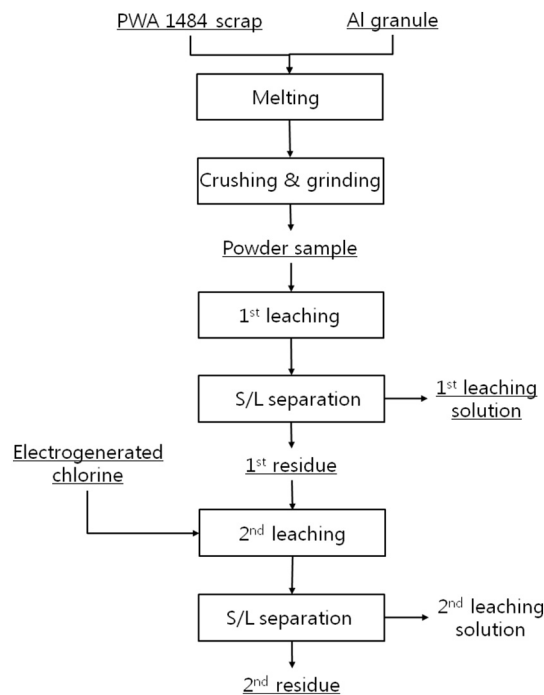


Fig. 2. Experimental flow chart.

Re from PWA 1484 superalloy scrap. It consists of three steps: a pre-treatment for easy comminution, an acid leaching of base metals for Ni dissolution and Re enrichment in the residue, and an electrogenerated chlorine leaching of Re from the acid leaching residue.

The superalloy scrap was cut into pieces, and mixed with Al granule (Junsei, 8–10 mm, chemical pure) in a 1:1 weight ratio, and then melted at $1250 \text{ }^\circ\text{C}$ in an electric induction furnace. The air-cooled melt was crushed and ground into powder for leaching experiment by mortar. The powder sample was analyzed by X-ray diffractometry (XRD, Rigaku D/Max 2200). The leaching of the powder sample ($< 150 \mu\text{m}$) was carried out in the reactor with hydrochloric acid solution by agitation at 300 rpm. After a filtration of the first leaching solution, the obtained leaching residue was dried at $90 \text{ }^\circ\text{C}$ for 24 h and used as a feeding sample at the second leaching step. At the second leaching step, the electrogenerated chlorine was sparged into the newly prepared HCl solution contained in the leaching reactor. The amount of Cl_2 dose was controlled by the applied current. A theoretical current efficiency of 100% was considered for Cl_2 generation, and the current density of 16.2 mA/cm^2 (1.0 A) corresponds to 0.31 mmol/min Cl_2 generation rate (Kim et al., 2016). On the other hand, a leaching of Re metal powder (SIGMA-ALDRICH, $< 150 \mu\text{m}$, $\geq 99.9\%$) was performed in the reactor with 400 mL solution to verify the effectiveness of electrogenerated Cl_2 as an oxidant acting on Re. The leaching process was operated as a closed system. At a certain time interval, 5 mL of the solution was taken using a syringe with $0.45 \mu\text{m}$ pore size filter. The solution samples were diluted and analyzed using an inductively coupled plasma spectrophotometer (ICP, Jobin Yvon JY 38 plus). The leaching percentage of each metal was calculated using Eq. (1).

Leaching percentage (%)

$$= \frac{\text{Dissolved amount of element metal in leaching solution (g)}}{\text{Amount of element metal in leaching sample (g)}} \times 100 \quad (1)$$

Reagent grade hydrochloric acid (Junsei Chemical Co., Ltd.) was used for the Cl_2 electrogeneration and leaching. All solutions were prepared using deionized water ($18 \text{ M}\Omega \text{ cm}$). The iodometric titration method (Aieta et al., 1984; Skoog et al., 1992) was used to determine the concentration of aqueous chlorine in HCl solutions.

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