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Separation and recovery of gold from waste LED using ion exchange method

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ABSTRACT

The separation and recovery of gold from waste LED using ion exchange method was investigated, employing commercial polyamine type anion exchanger, Diaion WA21J. Leaching of gold from LED terminal was successfully carried out with aqua regia at 80 °C. Separation of gold from the leaching solution was then carried out by column adsorption with WA21J. The selective adsorption of gold was achieved, while adsorption of the co-existing metals in the leaching solution was suppressed. Quantitative elution of gold from WA21J was achieved with diluted thiourea solution. Elementary gold was then obtained from the eluent by simultaneous precipitation and reduction of gold with sodium borohydride with 100% purity.

1. Introduction

Gold is currently used as a raw material in numerous industrial applications, due to its specific physical properties of electrochemical potential, and electro and thermal resistivity. The industrial demand of gold is expected to increase in the fields of pollution and emission control, chemical processing, and fuel cell applications (Corti et al., 2004, 2005). With the increasing demand, separation and recovery of gold from waste materials become an important issue from the view point of resource sustainability.

Generally, metals in waste materials are separated and recovered by solvent extraction and/or ion exchange in the hydrometallurgical process. There are several reports concerning the separation of gold by solvent extraction (Ohara et al., 1996; Narita et al., 2006; Pan and Gu, 2012) and by ion exchange (Iglesias et al., 1999; Matsubara et al., 2000; Sánchez et al., 2000; Gomes et al., 2001; Al-Merey et al., 2003; Hidalgo et al., 2006; Ertan and Gülfen, 2009; Erim et al., 2013). Comparing these separation methods, ion exchange has an advantage as an environmentally friendly process, since solvent extraction requires large amounts of organic solvent to dissolve both extractant and extracted metals, which leads to loss of the organic solvent (Kabay et al., 2003). The ion exchange method is also more effective for applying to the diluted metal solution leached from waste, rather than the solvent extraction method, since large amounts of organic solvent cause insufficient extractability of diluted metal ions (Saha et al., 2000). In the case of ion exchange for gold, anion exchange resins, based on amine group or quaternary ammonium salt group, are mainly used, since most gold exists as anionic species.

There are several reports concerning the separation and recovery of gold from waste materials. One of the waste materials applied for the separation and recovery of gold is printed circuit boards (PCBs). Adsorption process with Amberlite XAD-7HP, acrylic polymeric adsorbent, followed by leaching of the PCBs (Kim et al., 2011), adsorption process with biosorbent from persimmon residue (Fan et al., 2014), and electrochemical oxidation process for enrichment of gold (Fogarasi et al., 2014) have been reported. Adsorptive separation of gold from waste rinse water of the semiconductor manufacturing industry (Nguyen et al., 2010) and from wastewater of gold-plating (Soleimani and Kaghazchi, 2008) has also been reported. Chip-on-board LED (COB-LED) is also one of the attractive materials for the recovery of gold, since the COB-LED contains gold as wire. According to our literature survey, however, no hydrometallurgical separation and recovery process of gold from waste COB-LED was reported.

In the present work, the separation and recovery of gold from waste COB-LED was investigated using the ion exchange method. Commercial anion exchange resin, Diaion WA21J, having a polyamine group was used as an adsorbent. Leaching behavior of gold and co-existing components from LED terminal was first investigated by using aqua regia. Adsorption properties of gold and co-existing metals in the leaching solution were also investigated by both the batchwise and column adsorption systems. Precipitation and reduction of gold were then carried out by adding sodium borohydride to the eluent from the column operation. Recovery yield of the gold from the leaching solution was finally evaluated.

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2. Experimental

2.1. Reagents

Commercial ion exchange resin, WA21J, was kindly supplied by Nippon Rensui Co., Ltd. (Tokyo, Japan). Gold(III) chloride was supplied by Alfa Aesar, and all other inorganic reagents were supplied by Wako Pure Chemical Industries, Ltd. (Osaka, Japan), as analytical-grade reagents.

2.2. Leaching of waste LED

Actual waste COB-LED samples (Panasonic) were used in the present work. The waste LED terminal was first taken off from the lamp. The LED terminal (0.5 g) was then treated with aqua regia ([aqua regia] = 12.0 mol/L, ORP = 972 mV) (20 mL) in an autoclave at 80 °C and 1 atm for 24 h. The resultant suspension was then filtered, and the metal concentrations were determined by an inductively coupled plasma atomic emission spectrometer (ICP-AES, Shimadzu ICPS-7000 ver. 2). The concentration of aqua regia was defined by hydrogen ion concentration, which was determined by neutral titration with 1.0 mol/L NaOH solution. The oxidation–reduction potential (ORP) was measured by ORP meter (Lutron YK-23RP) using an Ag/AgCl electrode as a reference electrode.

2.3. Adsorption metals with WA21J

Gold single aqueous feed solution was prepared by dissolving appropriate amounts of gold(III) chloride to aqua regia solution for fundamental studies. Diluted actual leaching solution was also used for the practical study.

Batchwise adsorption was carried out by adding 20 mg of WA21J to 20 mL aqueous solution, and the mixture was shaken at 25 °C for 24 h. For examining the effect of aqua regia concentration on the adsorption, gold single solution in aqua regia was used as the aqueous solution. In this case, the concentration of gold was set at 50 mg/L, and the concentration of aqua regia was varied from 0.6 to 6.6 mol/L, and the ORP of the solution was measured by ORP meter. For examining the effect of coexisting components on the adsorption, the actual leaching solution after 10-time dilution ([aqua regia] = 1.2 mol/L, ORP = 702 mV) was used as feed solution. The adsorption isotherm of gold was investigated by using aqueous solution containing 50–850 mg/L of gold in 1.2 mol/L aqua regia. After shaking for 24 h, the suspended solution was filtered, and the metal concentrations were determined by ICP-AES. The amount of metal ion adsorbed, q (mmol/g), is defined by Eq. (1):

$$q = \frac{([\mathrm{M}]_{\mathrm{feed}} - [\mathrm{M}]) \cdot L}{w} \tag{1}$$

where $[\mathrm{M}]_{\mathrm{feed}}$ and [M] represent the initial and equilibrium concentrations of the metal ion in the aqueous phase (mmol/L), and L and w are the volume of aqueous solution (L) and weight of WA21J (g), respectively.

The column operation using WA21J was investigated. WA21J (weight: 0.6 g, wet volume 0.6 mL) was packed into a column tube (10 cm in length and 8 mm inner diameter), sandwiched between quartz wool. The aqueous feed solution, containing 53 mg/L of gold in 1.2 mol/L aqua regia, or actual leaching solution was then fed upward through the column using a dual-plunger pump (Flom KP-11) at 0.15 mL/min (SV = 15 h^{-1}). The effluent was collected with a fraction collector (EYELA DC-1500). After breakthrough, 0.01 mol/L hydrochloric acid was fed into the column to wash out the aqueous feed solution remaining in the column, and then the gold adsorbed was then eluted with 0.1 mol/L thiourea solution. The metal concentrations collected by the fraction collector were determined by ICP-AES. The bed volume of the effluent was calculated by Eq. (2):

Table 1
Leaching amount of metals.

Element	Fe	Ca	Ag	Au	Mn	Zn
Leaching amount (mg/g)	12.8	6.77	>1.96	2.17	0.0584	0.596

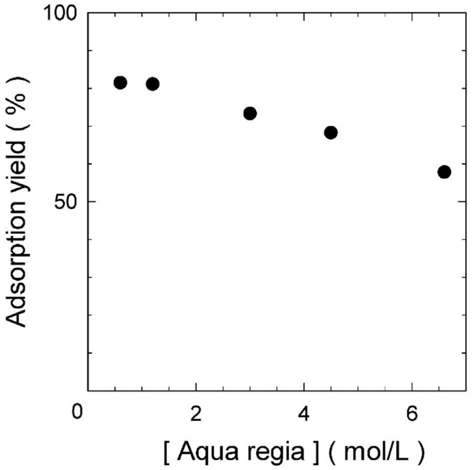

Fig. 1. Effect of aqua regia concentration on the adsorption yield of gold in single metal system; $[\mathrm{Au}]_{\mathrm{feed}}$ = 50 mg/L.

$$\text{Bed Volume} = \frac{v \cdot t}{V} \tag{2}$$

where v is flow rate of solution (mL/min), t is the time of feed solution application (min), and V is the wet volume of adsorbent in the column (mL).

2.4. Precipitation and reduction of gold

Gold in the eluent from the column operation was precipitated and then reduced by sodium borohydride ($NaBH_4$). Eluent solution containing 145 mg/L of gold was collected by the column experiment. $NaBH_4$ (0.01 g − 0.1 g) was then added to 20 mL of the collected eluent, and the mixture was stirred at room temperature for 1 h with magnetic stirrer. The resultant solution was then filtered, and pH was measured with a pH meter (Horiba F-23). The metal concentrations in the filtrate were determined by ICP-AES. The precipitates were dried in vacuo at room

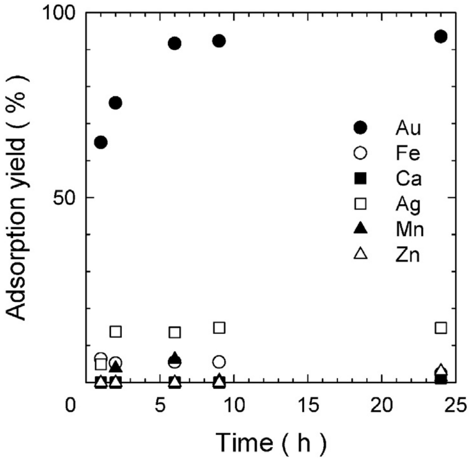

Fig. 2. Time course variation of adsorption yield of gold, iron, calcium, silver, manganese, and zinc with WA21J. $[\mathrm{Au}]_{\mathrm{feed}}$ = 6.30 mg/L, $[\mathrm{Fe}]_{\mathrm{feed}}$ = 35.3 mg/L, $[\mathrm{Ca}]_{\mathrm{feed}}$ = 18.0 mg/L, $[\mathrm{Ag}]_{\mathrm{feed}}$ = 4.87 mg/L, $[\mathrm{Mn}]_{\mathrm{feed}}$ = 0.206 mg/L, $[\mathrm{Zn}]_{\mathrm{feed}}$ = 1.45 mg/L, and $[\text{aqua regia}]_{\mathrm{feed}}$ = 1.2 mol/L.