Selective recovery of indium from lead-smelting dust

Hikaru Sawai [a,*], Ismail M.M. Rahman [a,b,*], Yoshinori Tsukagoshi [a],
Tomoya Wakabayashi [a], Teruya Maki [c], Satoshi Mizutani [d], Hiroshi Hasegawa [c,*]

[a] Graduate School of Natural Science and Technology, Kanazawa University, Kakuma, Kanazawa 920-1192, Japan
[b] Department of Applied and Environmental Chemistry, Faculty of Science, University of Chittagong, Chittagong 4331, Bangladesh
[c] Institute of Science and Engineering, Kanazawa University, Kakuma, Kanazawa 920-1192, Japan
[d] Graduate School of Engineering, Osaka City University, Sugimoto 3-3-138, Sumiyoshi-Ku, Osaka 558-8585, Japan

HIGHLIGHTS

- A new process is proposed for recovery of indium from lead-smelting dust (LSD).
- Chelant plus acid-assisted extraction and hydroxide-precipitation are combined.
- The competing effect of LSD-matrix components (lead and zinc) is minimized.
- The extraction step with EDDS chelant reduces the requirement of acid volume.
- The multi-step separation process could selectively recover 88% of indium from LSD.

ARTICLE INFO

ABSTRACT

Non-ferrous smelting dust, especially lead-smelting dust (LSD), contains percent levels of indium and thus constitutes a novel indium resource. The main difficulty in recovering indium from LSD is the coexisting presence of lead and zinc. In this study, a unique indium separation process was designed, combining techniques that involve washing with a chelant, leaching with acid and precipitation as hydroxide. The majority of the Pb in the LSD was selectively separated during chelant-assisted washing with ethylenediaminedisuccinate (EDDS), while the residual Pb was diminished through an acid leaching treatment with a mixed solution of sulfuric acid and hydrochloric acid. The chelant washing step also ensures a decrease in the raw LSD weight at a ratio of approximately 82% due to the removal of lead and counterions such as sulfate, and the washing step also minimizes the consumption of corrosive acids in the subsequent step. Selective indium separation from LSD is further complicated by the similarity of the behavior of zinc during the acid leaching step. Therefore, hydroxide precipitation at pH 5 has been introduced as the final step, ensuring the maintenance of zinc as a soluble species in the supernatant and the selective separation of indium (~88%) as a hydroxide precipitate.

1. Introduction

The metal indium, particularly as ITO (indium-tin-oxide) thin film, is an industrially important component because ITO is necessary for building electronic devices [1]. ITO is widely utilized for manufacturing liquid crystal displays, plasma displays and solar energy cells [2], which consume approximately two-thirds of the global indium production [1]. One of the resource materials for raw indium is non-ferrous metal ore [3], which is obtained as a by-product of the smelting process of the non-ferrous metal ore [4]. Raw indium deposits are region-specific (i.e., China, Korea, and Russia) [5]. Discrepancies in demand, supply and price are therefore observed. The search for alternate sources of raw indium is vital from the point of view of resource strategy, and this search is focused mostly on the processing of indium-laden waste materials, e.g., ITO scrap [2,6–8], end-of-life liquid crystal displays [6,9–11] and etching waste [12–14].

The residue and flue dust from the smelting of non-ferrous metals, such as lead, termed lead smelting dust or LSD hereafter, also includes indium [15] and is expected to be a novel indium resource. Acid leaching is commonly employed for metal smelting from waste resources [16–18]. Indium recovery from waste material has been reported through the use of acid leaching and

hydroxide or sulfide precipitation [2,6,7,19–21]. This approach is frequently criticized both in terms of overall efficiency due to the lack of selectivity in separation and in terms of the hazardous impact on the environment. Therefore, a reduction in the use of acid or other corrosive extractants is desirable [22]. As alternative extractants for indium recovery, chelants [10,11,23], other solvents, such as carboxylic acid or phosphoric acid derivatives (e.g., di-2-ethylhexylphosphoric acid), chelating compounds (e.g., hydroxyoximes and azoles), and solvating extractants (e.g., tributyl phosphate, trioctylphosphine oxide and methyl isobutyl ketone) have been proposed [6,24]. Although alternative extractants possess unique leaching behavior (i.e., selectivity, re-usability, etc.), the leaching efficiency of these alternative extractants is not comparable with the leaching efficiency of even acids or bases.

Indium reclamation from waste resources is further complicated by the simultaneous presence of metal components having chemical similarities [25,26]. For example, tin is the major impurity in the recovery process of indium from ITO waste [2]. The issue of chemical similarities is more of a concern during the recovery of indium from LSD using acids or bases because the acid leaching process consumes a higher quantity of solvents due to the coexistence of base metals (e.g., lead, zinc, etc.) other than the indium, increasing the total operating cost.

The objective of the current work was to develop a unique technique for the quantitative reclamation of indium from LSD. The separation scheme combines chelant-assisted washing, acid leaching and hydroxide precipitation techniques, minimizing the competing effect of coexisting metals.

2. Experimental

2.1. Materials

Analytical grade reagents were used during all experiments. Ethylenediaminetetraacetate, (EDTA, Kanto Chemical, Tokyo, Japan), 3-hydroxy-2,2′-iminodisuccinate (HIDS, Nippon Shokubai, Osaka, Japan) and ethylenediaminedisuccinate (EDDS, Chelest,
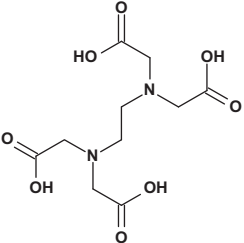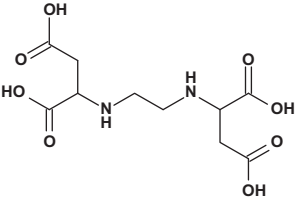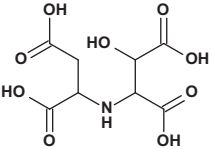
Osaka, Japan) were used as the chelating extractants (Table 1). As buffer reagents, 4-(2-hydroxyethyl)-1-piperazineethanesulfonic acid (HEPES, Nacalai Tesque, Kyoto, Japan), N-tris(hydroxymethyl)-3-aminopropanesulfonate (TAPS) and 3-cyclohexhylaminopropane sulfonate (CAPS, MP Biomedicals, Santa Ana, CA) were used. The ICP multi-element standard solution IV containing 1000 mg L^{-1} of 23 elements (Ag, Al, B, Ba, Bi, Ca, Cd, Co, Cr, Cu, Fe, Ga, In, K, Li, Mg, Mn, Na, Ni, Pb, Sr, Tl, Zn) in diluted nitric acid (Merck KgaA, Darmstadt, Germany) was used as the standard during metal analysis. Purified water (resistivity >18.2 MΩ cm) was obtained from an Arium Pro water purification system (Sartorius Stedim Biotech GmbH, Göttingen, Germany) and was used for the preparation of all reagents.

Low-density polyethylene bottles, polyethylene test tubes (Nalge Nunc, Rochester, NY), DigiTUBEs (SCP Science, Quebec, Canada), perfluoroalkoxy tubes and micropipette tips (Nichiryo, Tokyo, Japan) were used as laboratory ware. The laboratory ware was cleaned via overnight soaking in Scat 20X-PF alkaline detergent (Nacalai Tesque, Kyoto, Japan) and then in HCl (3 mol dm^{-3}), followed by rinsing with purified water after each of the previous steps.

2.2. Analytical techniques

Concentrations of metals in solution were measured by using the iCAP6300 inductively coupled plasma optical emission spectrometer (ICP-OES) (Thermo Fisher Scientific, Waltham, MA). The ICP-OES was operated under the following conditions: the radio frequency power at the torch was 1.15 kW, the plasma gas flow was 12 dm^3 min^{-1}, the auxiliary gas flow was 1 dm^3 min^{-1}, the nebulizer gas flow was 0.5 dm^3 min^{-1}, and the integration time was 30 s. The Epsilon3 X-ray fluorescence (XRF) spectrometer (PANalytical, Almelo, The Netherlands), JSM-7100F field-emission scanning electron microscope (FE-SEM) (JEOL Co., Tokyo, Japan) combined with the INCA energy dispersive X-ray (EDX) spectrometer (Oxford Instruments, Oxfordshire, UK), and MiniFlex 600 X-ray diffractometer (XRD) (Rigaku Corporation, Tokyo, Japan) were used

Table 1
Chemical structures and acid dissociation constants (pK_a) of chelants.

Chelant	Structure	pK_a	
EDTA[a], Ethylenediaminetetraacetate		pK_{a1}	2.00
		pK_{a2}	2.69
		pK_{a3}	6.13
		pK_{a4}	10.37
EDDS[a], Ethylenediaminedisuccinate		pK_{a1}	2.95
		pK_{a2}	3.86
		pK_{a3}	6.84
		pK_{a4}	10.01
HIDS[b], 3-Hydroxy-2,2′-iminodisuccinate		pK_{a1}	2.14
		pK_{a2}	3.08
		pK_{a3}	4.07
		pK_{a4}	9.61

[a] At 25 °C (μ = 0.1 M) [41].
[b] At 25 °C (μ = 0.1 M) [28,29,39].