Hydrometallurgy 137 (2013) 148–155

Contents lists available at SciVerse ScienceDirect

Hydrometallurgy

journal homepage: www.elsevier.com/locate/hydromet

Separation and concentration of indium from a liquid crystal display via homogeneous liquid–liquid extraction

Takeshi Kato [a], Shukuro Igarashi [a,*], Yoshiyuki Ishiwatari [b], Makoto Furukawa [c], Hitoshi Yamaguchi [d]

[a] Department of Biomolecular Functional Engineering, Faculty of Engineering, Ibaraki University, 4-12-1 Nakanarusawa, Hitachi, Ibaraki 316–8511, Japan
[b] Industrial Technology Institute of Ibaraki Prefecture, 3781–1 Nagaoka, Ibaraki-machi, Higashi Ibaraki-gun, Ibaraki 311–3195, Japan
[c] PerkinElmer Japan Co., Ltd., Technical Center 4F, Yokohama Business Park, 134 Godo-cho, Hodogaya-ku, Yokohama, Kanagawa 240–0005, Japan
[d] Research Center for Strategic Materials, National Institute for Materials Science, 1-2-1 Sengen, Tsukuba, Ibaraki, 305–0047, Japan

ARTICLE INFO

Article history:
Received 27 August 2012
Received in revised form 29 May 2013
Accepted 1 June 2013
Available online 7 June 2013

Keywords:
Homogeneous liquid–liquid extraction
HoLLE
Indium
Liquid crystal display
Recycling

ABSTRACT

Separation and concentration of indium from a liquid crystal display (LCD) was conducted using homogeneous liquid–liquid extraction (HoLLE) in order to completely recycle the LCD. First, using 2.40 M hydrochloric acid, 1.52 mg of indium was leached from the LCD of a mobile phone while keeping the amount of leached foreign metals, such as iron and aluminum, as low as possible. The ability to recycle the LCD glass is also expected because arsenic and antimony, which are used as antifoamers for the glass, were separated in the leaching residue. Next, on the basis of the metal leaching results for a mobile phone LCD, HoLLE with Zonyl FSA ($CF_3(CF_2)_nCH_2CH_2SCH_2CH_2COOH$, $n = 6–8$, Du Pont Co.) was conducted via the formation of metal-1,10-phenanthroline chelates on a solution formulated to simulate the leachate from a mobile phone LCD. Using this technique, more than 96.7% of the indium was extracted from the simulated leaching solution into the sedimented liquid phase. After phase separation, the volume ratio (Va/Vs) of the aqueous phase (Va) and the sedimented liquid phase (Vs) was 438 (46 ml → 0.105 ml). In addition, the sedimented liquid phase was dropped on a filter and evaluated using X-ray fluorescence analysis. After determination of the elements concentrated into the sedimented liquid phase, the mass concentration of indium in the sedimented liquid phase was estimated to be 10.4 wt%. This result indicated that the mass concentration ratio was 405 because the mass concentration of indium in the mobile phone LCD was 0.0257 wt% on the basis of being determined by heating and leaching with aqua regia.

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1. Introduction

The demand for liquid crystal displays (LCDs) has been increasing worldwide because they can save electricity and resources. In addition to a marked increase in the production quantity of LCDs, it is expected that the production of larger LCDs will also increase. Because of these circumstances, the available supply of indium, which is a key material in the production of LCDs, has been largely consumed (Park et al., 2009). During 2011, U. S. imported for consumption of unwrought indium metal and indium powders totaled 146 tons, a 24% increase from the 117 tons imported in 2010 (Tolcin, 2013). Moreover, the estimated world refinery production of indium has increased in recent years (583 tons in 2009, 662 tons in 2011) (Tolcin, 2013).

However, to meet such a large continuous demand for indium, a constant supply of this substance is still required. Recycling of indium is considered to be one solution. The separation and recovery of indium from the indium tin oxide (ITO) used in the production of LCDs and other devices has been achieved using various methods, after following the leaching of the indium with hydrochloric acid or sulfuric acid (Inoue et al., 2008; Li et al., 2011), such as the sulfide method (Li et al., 2011), the hydroxide method (Yamada, 1997), the displacement deposition method (Hsieh et al., 2009), the solvent extraction method (Inoue et al., 2008; Kang et al., 2011; Koleini et al., 2010; Virolainen et al., 2011a), and the electrowinning method (Chou and Huang, 2009; Kang et al., 2011). Moreover, separated and recovered indium has been refined using electrolytic refining method (Alfantazi and Moskalyk, 2003). However, indium recycling methods have only been applied to unused ITO targets (Hsieh et al., 2009; Li et al., 2011). Methods by which indium can be recycled from used or defective products have not yet been developed (Endo et al., 2007). Although the some trials have been completed, there is still room for improvement before such a method will be practical for industrial usage (Inoue et al., 2008; Virolainen et al., 2011b). The treatment of LCD glass affects the ability to recycle indium from these products, and moreover, the process is also very expensive (Endo et al., 2007). Therefore, a method for easily treating LCD glass and efficiently and cost-effectively recycling indium is required.

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* Corresponding author. Tel.: +81 294 38 5059; fax: +81 294 38 5078.
E-mail address: igarashi@hcs.ibaraki.ac.jp (S. Igarashi).

0304-386X/$ – see front matter © 2013 Elsevier B.V. All rights reserved.
http://dx.doi.org/10.1016/j.hydromet.2013.06.004

The concentrations of rare metals in industrial products are low. Therefore, it is necessary to both separate and concentrate the rare metals in order to recover and recycle them from the industrial products. The separation and concentration of rare metals for recycling has been accomplished via pyrometallurgical and hydrometallurgical treatments (Cui and Zhang, 2008; Georgi-Maschler et al., 2012). Hydrometallurgical treatment has typically been achieved using solvent extraction. Moreover, solvent extraction is a useful metallurgical process for indium. In the general solvent extraction method, there is an interface between the aqueous phase and the water-immiscible organic solvent phase. Thus, mechanical shaking is required. The shaking increases the surface area of the contact interface and causes the solute to move through the interface. However, while solvent extraction has been established as a general process, it has some problems, such as the complexity of the process. In homogeneous liquid-liquid extraction (HoLLE) (Igarashi and Yotsuyanagi, 1992), the initial condition before phase separation is a homogeneous solution; that is, there is no interface between the aqueous and organic solvent phases. In other words, and, the surface area of the interface is infinitely large. Therefore, no vigorous mechanical shaking is necessary. The procedure is simple and requires only the addition of a reagent. Using HoLLE, which involves the addition of acids and so on to a solution with rare metal chelates, it is possible to concentrate the chelates into a sedimented liquid phase of small volume. HoLLE can thus be used to achieve the 100–100,000 fold concentration in several minutes down to micro-volume(μL) levels (Takagai and Igarashi, 2002). Therefore, rare metals can be quickly and efficiently concentrated.

The examples of the concentration of rare metals using HoLLE are as follows: HoLLE–inductively coupled plasma optical emission spectrometry (ICP-OES) of lanthanoid ions with di(2-ethylhexyl)ester in strongly acidic aqueous media (Fuchimukai et al., 2006), HoLLE–ICP-OES of metal-1,10-phenanthroline chelates in weakly acidic aqueous media (Yamaguchi et al., 2005), and HoLLE of uranium(VI) from an aqueous acetate solution (Takahashi et al., 1999). These examples suggest that HoLLE has the potential to be effective for the concentration of rare metals from used industrial products. However, there have been no reports regarding the application of HoLLE in a practical recycling system for rare metals. We believe that construction of a new, progressive, rare metal recycling system based on a combination of HoLLE and urban mining (Nanjo, 1987), from which the good use of resources has not yet been achieved, is possible. Therefore, we examined the separation and concentration of a rare metal (indium) using HoLLE.

2. Experimental

2.1. Reagents and materials

An LCD of a mobile phone produced by DoCoMo and received from Re-tem Corporation (Tokyo, Japan) was used as a raw material in this study. Aqueous solutions of metal ions were prepared by diluting 1000 mg l^{-1} standard solutions obtained from Wako Pure Chemical Industries, Ltd. (Osaka, Japan). The used metal salts were as follows: included In (in HNO_3), $Al(NO_3)_3$, $Fe(NO_3)_3$, $CaCO_3$, Sn (in HCl), Na_2SiO_3, $SrCO_3$. A solution of Zonyl FSA ($CF_3(CF_2)_nCH_2CH_2SCH_2CH_2CO_2H$, $n = 6–8$, DuPont, Tokyo, Japan) was prepared by diluting the pure substance with an equivalent amount of distilled water. The complexing agent 1,10-phenanthrolinium chloride monohydrate, nitric acid, hydrochloric acid, ammonia (to adjust pH), and other basic chemicals were obtained from Wako Pure Chemical Industries, Ltd. (Osaka, Japan). All of the reagents used in this study were of analytical grade.

2.2. Apparatus

The following instruments were utilized in this study: an M-12 pH meter manufactured by Horiba (Kyoto, Japan), a 7780 centrifugal separator manufactured by Kubota (Tokyo, Japan), an XGT-5000WR X-ray fluorescence spectrometer manufactured by Horiba (Kyoto, Japan), and an ICP ULTIMA 2 ICP-OES manufactured by Horiba (Kyoto, Japan).

2.3. Methods

2.3.1. Determination of the indium content in a mobile phone LCD

The LCD of a mobile phone was divided into four parts, and all of the parts were added to a 300 ml conical beaker. Aqua regia (40 ml) was added to the conical beaker, and the mixture was heated at 230 °C for 150 min on a sand bath. After heating, the solution was allowed to cool for several minutes and then filtered by gravity through filter paper using cellulose. The filtrate was adjusted to 100 ml by adding distilled water and then analyzed using ICP-OES.

2.3.2. Leaching

The LCD of a mobile phone was cut into 2–3 mm pieces, each of which was placed into a 100 ml beaker. Hydrochloric acid (60 ml; 3.20, 2.40, or 1.60 M) was added to each of the beakers, and the mixtures were stirred at 1000 rpm for 4 h. After stirring, each of the solutions was filtered and separated into a leaching solution and a leaching residue. The leaching solutions were adjusted to 100 ml with distilled water and then analyzed using ICP-OES.

2.3.3. HoLLE of a pure indium solution

A 1000 mg l^{-1} indium standard solution was added to a 50 ml centrifuge tube such that 1 mg of indium was placed in the tube. A 0.01 M 1,10-phenanthroline solution (3 ml) was then added to the centrifuge tube, and the solution was adjusted to 30 ml with distilled water. The pH of the solution was then adjusted to 2.0 with 5 M nitric acid and 13.4 M ammonia. Next, 50 v/v% Zonyl FSA (1 ml) and acetone (10 ml) were added, and the solution was centrifuged at 2500 rpm for 30 min.

2.3.4. HoLLE of a simulated leaching solution from a mobile phone LCD

A simulated leaching solution containing seven metals was adjusted to 20 ml in a 50-ml centrifuge tube such that the metals were leached with 2.40 M hydrochloric acid. The concentrations of seven main metals in the simulated leaching solution are shown in Table 1. Next, a 0.01 M 1,10-phenanthroline solution (4 ml) was added to the centrifuge tube, and the solution was adjusted to 30 ml with distilled water. The pH of the solution was then adjusted to 2.0 with 5 M hydrochloric acid and 13.4 M ammonia, and 50 v/v% Zonyl FSA (1 ml) and acetone (10 ml) were added, and the solution was centrifuged at 2,500 rpm for 30 min. The phases were separated, and the aqueous phase was heated at 100 °C for 30 min in order to remove acetone prior to ICP-OES analysis. Hydrochloric acid (5 M, 5 ml) was then added to the water phase, and the volume was adjusted to 50 ml with distilled water. This solution was analyzed using ICP-OES.

Table 1
Metal concentrations in the simulated HCl leaching solution of mobile phone LCD screen.

Element	Concentration (mg l^{-1})
In	15.2
Al	5.82
Fe	3.77
Ca	2.19
Sn	1.34
Si	1.17
Sr	0.72

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