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# Recovery of indium from liquid crystal displays of discarded mobile phones using solvent extraction



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#### ABSTRACT

One of the greatest challenges in the development of hydrometallurgical processes for the recovery of indium from secondary sources is the fact that the leaching step generally results in very dilute solutions. Therefore, this diluted indium solution must be concentrated before being sent to a subsequent step such as recovery of the metal by electrowinning. The aim of the present work was to apply solvent extraction using bis(2-ethylhexyl) phosphate (D2EHPA) in order to maximize the indium concentration in the solution. Firstly, the extraction and stripping steps were investigated using a synthetic solution. After definition of the best conditions using a synthetic solution, a real solution obtained from the leaching of discarded LCDs was employed.

#### 1. Introduction

The amount of waste electrical and electronic equipment (WEEE) has increased significantly over the past few years, as a result of the technology revolution, and constitutes a new environmental challenge (Tuncuk et al., 2012; Maris et al., 2015; Islam et al., 2016; Charles et al., 2017). Among the various types of WEEE are the liquid crystal displays (LCDs) that are widely used in computers, notebooks, mobile phones, and television sets (Wang and Xu, 2016; Ferella et al., 2017). The typical average lifespans are between three and five years for notebooks and less than two years for smartphones (Li et al., 2009, 2015).

LCD screens usually contain conductive electrodes made of tindoped indium oxide (ITO), which are placed between two glass panels. ITO is a mixture of indium [III] oxide  $(In_2O_3)$  and tin [IV] oxide  $(SnO_2)$ , with 80–90 wt% of  $In_2O_3$  and 10–20 wt% of  $SnO_2$  (Silveira et al., 2015; Fontana et al., 2015). More than 65% of the total amount of indium extracted worldwide is employed in the manufacture of ITO films for LCDs (Dodson et al., 2012; Zhang et al., 2015).

Indium is a rare and strategic metal, and is mainly obtained as a byproduct of zinc and lead production (Li et al., 2015; Zhang et al., 2015). The average content of indium in zinc ores varies from less than 1–100 ppm, while in LCD screens it is present at approximately 250 ppm (Yang et al., 2013). According to Takahashi et al. (2009), LCD screens may contain indium at up to 1400 g/ton. Therefore, discarded LCD screens are a potential resource for indium recovery, considering the high indium content, compared to the minerals (Dodbiba et al.,

#### 2012; Yang et al., 2016).

The main methods used for the recovery of indium from wastes are hydrometallurgical and pyrometallurgical processes (Chou et al., 2016). The pyrometallurgical route includes a pyrolysis process that is employed to recover the organic matter from discarded LCD panels. The products generated are oils, gases, and glass containing ITO (Wang and Xu, 2016). The hydrometallurgical route includes leaching, cementation, and solvent extraction (Dodbiba et al., 2012; Rocchetti et al., 2016).

However, there are some difficulties in the development of recycling technologies for indium recovery. In the case of indium recovery from secondary sources, leaching of wastes containing ITO results in solutions with very low indium concentrations (< 50 mg/L) (Grimes et al., 2017).

Although indium is found associated with tin, lead, copper, and iron, it is usually obtained as a byproduct of the zinc smelting process (Worrel and Reuter, 2014). Alfanzati and Moskalyk (2003) presented various commercial recovery processes for obtaining indium from zinc waste. In these processes, the zinc waste undergoes different purification and concentration steps in order to recover the metal. In one case, solvent extraction was used to concentrate the indium in solution from around 2 g/L to 20–30 g/L, with aluminum plates being immersed in the solution in order to remove the indium by cementation.

In efforts to combat the depletion of natural resources, much attention has been given to extracting indium from discarded LCDs, as can be observed in the available literature. However, to the best of our

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knowledge, there have been no studies of the use of solvent extraction to obtain a concentrated indium solution. Although several studies have reported the solvent extraction of indium using D2EHPA, these have mainly concerned definition of the optimal conditions for separation and recovery of indium.

Therefore, the aim of this work was to develop an efficient method for concentrating indium solutions obtained from the leaching of LCDs from discarded mobile phones. Obtaining a concentrated indium solution is fundamental for making it possible to recover indium directly in its metallic form, for example by means of electrowinning.

Firstly, a synthetic solution of indium sulfate  $[In_2(SO_4)_3]$  was used to determine the best conditions for the extraction and stripping of indium. A real solution obtained from the leaching of discarded LCDs was then treated under the best conditions determined for the synthetic solution. For the solvent extraction, the following variables were studied: contact time, pH, D2EHPA concentration, and aqueous to organic phase ratio (A:O). For the stripping, the following variables were studied: A:O phase ratio, HCl concentration, and contact time.

#### 2. Experimental

#### 2.1. Reagents and instruments

The following reagents were used in this study:  $In_2(SO4)_3 xH_2O$  (99.99%, Sigma-Aldrich),  $H_2SO_4$  (98%, Sigma-Aldrich), HCl (37%, Sigma-Aldrich), NaOH (97%, Sigma-Aldrich), bis(2-ethylhexyl) phosphate (D2EHPA, 97%, Sigma-Aldrich), and commercial grade kerosene (Nacional).

D2EHPA is a highly popular reagent and is widely used in practical applications because it has good selectivity for indium, over other metal ions (Li et al., 2015). The following general expression describes the extraction of indium (III) from sulfuric acid solutions by D2EHPA (Sato and Sato, 1992):

 ${}_{m}In^{3+}_{(aq)} + (2 m+1)(HR)_{2(org)} \leftrightarrow In_{m}R_{2(2m+1)}H_{m+2(org)} + 3mH^{+}_{(aq)};$ 

#### where $m \ge 1$ .

The LCD screens were processed in a ball mill (Model MA-500, Marconi). The metal concentrations were determined by atomic absorption spectrometry (Model 240FS, Agilent). All measurements were performed in triplicate. The pH was measured with a pH meter (Model ITMPA-210, Instrutemp). A mechanical stirrer (Edutec) was used in the extraction and stripping experiments.

#### 2.2. Methods

Firstly, a synthetic indium-containing solution was used to determine the best conditions for the extraction and stripping. A summary of the experimental conditions used in the solvent extraction and stripping experiments is shown in Table 1. The initial conditions used in step 1 were based on a previous study (Li et al., 2015). After definition of the best conditions, a real indium-containing solution was obtained from the leaching of a batch of LCDs that were manually removed from discarded mobile phones. The LCD panels (without polymers) were comminuted in a ball mill for 4 h at 60 rpm. After milling, all the resulting material was passed through a 150  $\mu$ m sieve.

In the LCD leaching step, the following conditions were used: solid/liquid ratio of 1:20, 1.0 M  $H_2SO_4$ , 90 °C, 1 h, and stirring at 500 rpm. Further details about this step are available in a previous publication (Silveira et al., 2015).

#### 2.2.1. Definition of the ideal conditions for solvent extraction

The concentration of indium in the synthetic solutions was 33.74 mg/L. This concentration was chosen in order to simulate the concentration typically obtained from the leaching of LCD panels (Silveira et al., 2015; Rocchetti et al., 2015).

In all the solvent extraction experiments, the aqueous and organic phases were placed in a 500 mL three-neck volumetric flask and were mixed using a mechanical stirrer at 800 rpm (at 25 °C). To avoid kerosene volatilization, temperatures higher than 25 °C were not used. Afterwards, the two phases were separated using a funnel. The conditions evaluated for the extraction were the contact time, pH, and A:O phase ratio (v:v), as indicated in steps 1, 2, 3, and 4 Table 1. The pH was adjusted using 0.5 M  $H_2SO_4$  and 0.5 M NaOH.

#### 2.2.2. Definition of the ideal conditions for stripping

The stripping of indium from the loaded organic phase was carried out using HCl. The organic and aqueous phases were placed in a 100 mL beaker and stirred at 800 rpm (at 25 °C). A funnel was used to separate the two phases. The conditions evaluated for the stripping were the HCl concentration and the A:O phase ratio, as indicated in steps 5 and 6 Table 1.

#### 2.2.3. Maximization of the indium concentration

In order to maximize the indium concentration in the stripped solution, an initial evaluation was made using different D2EHPA concentrations and A:O phase ratios, as shown in steps 7 and 8 Table 1. The ideal times for the extraction and stripping steps were also determined, as shown in steps 9 and 10 Table 1.

### 2.2.4. Application of the best conditions using the solution obtained from leaching of the LCD panels

In this step, the best conditions for extraction and stripping, as determined using the synthetic solution, were applied to the solution obtained from the leaching of the LCD panels. The conditions employed are shown in steps 11 and 12 Table 1.

#### 3. Results and discussion

#### 3.1. Definition of the ideal conditions for solvent extraction

#### 3.1.1. Effect of contact time

The extraction efficiency increased with the contact time (Fig. 1), with the maximum extraction percentage (99.78%) being achieved in 3 min and remaining practically unchanged (99.67%) after 6 min. Therefore, a contact time of 3 min was defined as the ideal for use in the next tests.

#### 3.1.2. Effect of pH

The effect of pH on indium extraction is illustrated in Fig. 2. The results showed that the indium extraction increased as the pH increased up to pH 1.0. At pH 0.0, the extraction efficiency was less than 50%. However, at pH 0.5, the efficiency reached 99.78% and then remained constant until pH 1. This behavior was because at very low pH, the high H<sup>+</sup> concentration hindered the cation exchange reaction of  $In^{3+}$  with D2EHPA (Li et al., 2015). At pH higher than 1.5, the extraction efficiency decreased, possibly due to hydrolysis of the indium ions in this pH range (Li et al., 2015). Therefore, under the conditions tested, the ideal pH for indium extraction was in the range 0.5–1.0.

Experiments were not performed above pH 3.0, due to the risk of indium precipitation. According to the elemental speciation diagram obtained using Hydra/Medusa chemical equilibrium database and plotting software (www.kth.se/en/che/medusa/downloads, 2017), precipitation of indium occurs at pH values greater than 3. Therefore, the ideal pH for the extraction of indium from an aqueous solution was defined as 0.5.

#### 3.1.3. Effect of phase ratio

Fig. 3 shows the influence of the A:O phase ratio on the indium extraction efficiency. It can be seen that the efficiency decreased as the A:O phase ratio increased. High extraction efficiencies exceeding 99% were achieved at phase ratios of 3:1, 6:1, and 9:1.

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