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## Impact of selected parameters on extraction of indium from LCD screens

### Wpływ wybranych parametrów na proces ługowania indu z ekranów LCD

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#### **Abstract**

Due to the minute availability of indium and its crucial importance to the world economy, it is necessary to find alternative sources of this metal. As a large proportion of indium production is consumed for the purpose of LCD screen manufacturing, it seems reasonable to investigate their recycling leading to the recovery of this metal. The present work investigates the impact of time, temperature, and the concentration of sulfuric acid on the effectiveness of indium extraction from milled LCD panel glass scrap originating from portable computers manufactured between 2005 and 2012. The conclusion of our research defines the optimal conditions for extraction.

**Keywords:** indium, ITO, LCD panels, recycling

#### **Streszczenie**

Ze względu na niewielkie zasoby indu i jego kluczowe znaczenie dla gospodarki światowej konieczne jest poszukiwanie alternatywnych zasobów tego metalu. Ponieważ ind wykorzystywany jest głównie w produkcji ekranów w technologii LCD, racjonalny wydaje się ich recykling w kierunku odzysku tego pierwiastka. W niniejszej pracy zbadano wpływ czasu, temperatury oraz stężenia kwasu siarkowego na efektywność ługowania indu ze zmielonej frakcji szklanej paneli LCD pochodzących z komputerów przenośnych z lat 2005–2012. W wyniku badań określono optymalne warunki ługowania.

**Słowa kluczowe:** ind, ITO, panele LCD, recykling

## 1. Introduction

One of the most-important applications of indium in the world economy is the manufacture of ITO (Indium-Tin-Oxide). It is estimated that this process is responsible for more than 80% of the world's indium demand [1, 2]. ITO is a mixture of two oxides:  $\text{In}_2\text{O}_3$ , and  $\text{SnO}$ , with mass proportions of 90% and 10% [3], respectively, whereas the content of indium is around 74%. The most-wanted property of ITO is the connection of two particular

features – its low electric resistance (ca.  $10^{-3} \Omega\text{cm}$ ) [4] and good transmission of the visible spectrum of light (up to 75%). Thus, it is not surprising that one of the most-popular applications of ITO is the creation of optically transparent and electrically conductive layers. These layers are commonly used, mainly in the construction of all types of LCD (Liquid Crystal Display) and TFT (Thin Film Transistor) flat screens [5] as well as photovoltaic panels [4, 6].

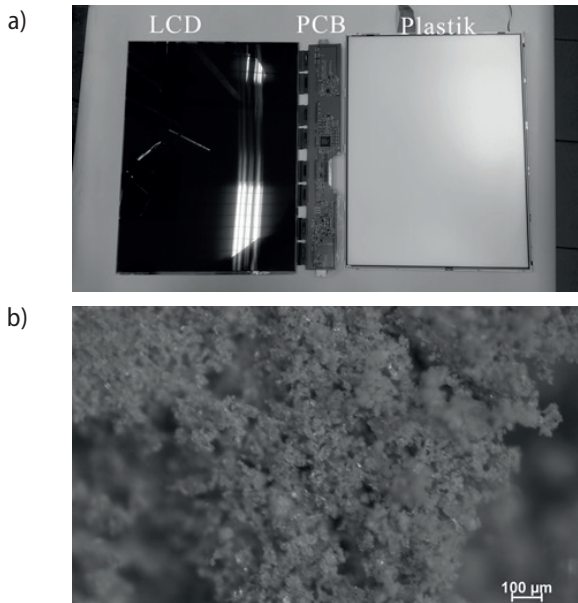
With technological development, we can observe a growing demand for devices that include LCD displays. These devices include portable computers, computer screens, television sets, and mobile phones/smartphones. Considering the scarce world indium resources and their very significant scattering in Earth's crust [7] (as well as the short service life of the aforementioned devices into account), used LCD screens can be utilized as an alternative source of indium [8].

Currently, the vast majority of the proposed indium recycling methods are based on acidic or alkaline extraction from initially processed LCD screens. This initial processing of an LCD panel usually includes its selective dismantling, crushing, and milling [9]. The removal of the organic phase from the adhesives that bind the respective glass elements of an LCD panel together may form another stage in the initial processing [10]. Research on acidic extraction most-frequently involves the use of hydrochloric acid [11, 12], nitric acid (V) [11, 13], sulfuric acid (VI) [14, 15], and mixtures of these acids (or with the addition of oxidizers) [14]. The present paper presents the impact of temperature, time, and concentration of sulfuric acid on the indium extraction process for used LCD panels.

## 2. Experiment

In our research, we utilized 12- to 17-inch LCD screens from portable computers manufactured between 2005 and 2012. The investigated LCD panels were manually removed from the computer cases and divided into glass scrap, plastics, and PCB. After separation from the remaining elements, the glass scrap was cut into pieces sized ca.  $5 \times 5$  cm and then milled in a rod mill. In the case of this mode of LCD panel preparation, we achieved a powder with a grain size of less than 0.5 mm. No method for the initial removal of organic scrap was utilized during the preparation of the LCD panels for extraction. Figure 1 presents the dismantled LCD panel with PCB connections and plastic scrap (Fig. 1a) and the already milled scrap prior to extraction (Fig. 1b).

The extraction process was conducted in a 2 M solution of sulfuric acid with a solid to liquid phase ratio of 0.1 g/ml. During the process, the solution was intensively stirred with the use of a mechanical stirrer with a rotating speed of 300 rpm and kept at a constant temperature. Due to the intensive agitation and high degree of fragmentation of the solid phase, the sample for determining the indium concentration was only taken at the end of each experiment. For our experiments, we utilized analytical-grade reagents and deionized water.



*Fig. 1. Dismantle LCD panel: a) product fractions of LCD: LCD panel, PCB, plastic, and PTFE-isolating layer; b) LCD panel after grinding*

The determination of the elements in the extract was performed with the use of an MP-AES (Atomic Emission Spectroscopy), Agilent 4200 (Australia). The determination of the main elements in the glass scrap was performed with the use of a WD-XRF (Wavelength Dispersive X-ray Fluorescence) of Rigaku Primini (Japan) apparatus.

### **3. Results and discussion**

#### **3.1. Characteristics of chemical composition of used LCD panels**

Experiments on recycling selected types of consumer goods frequently encounter the problem of ascertaining the chemical composition of the processed material. This is the result of the differences in manufacturing processes among different manufacturers. For this reason, we obtained large differences while comparing different research results. In the case of processing LCD panels, an additional difficulty comes from the presentation method for the content of the respective elements in the processed screens, as depending on the publication, this is quoted for the glass scrap alone or the screen as a whole – together with the frame, electrical connections, and PCB. Table 1 presents the main components of the glass scrap of the LCD panels subject to our research. The analysis was performed with the use of the WD-XRF technique, assuming that all elements were in the form of oxides.

Table 1. Main components of glass scrap of LCD panels, obtained by WD-XRF technique

| Composition | SiO <sub>2</sub> | Al <sub>2</sub> O <sub>3</sub> | CaO  | SrO  | MgO  | Fe <sub>2</sub> O <sub>3</sub> | As <sub>2</sub> O <sub>3</sub> | P <sub>2</sub> O <sub>5</sub> |
|-------------|------------------|--------------------------------|------|------|------|--------------------------------|--------------------------------|-------------------------------|
| Mass [%]    | 61.95            | 17.98                          | 8.13 | 6.05 | 2.72 | 1.75                           | 0.48                           | 0.44                          |

We also performed a trace analysis of In and Sn content through mineralization of the test sample in 4 M H<sub>2</sub>SO<sub>4</sub> and a subsequent analysis of the resulting solution with the MP-AES. The determined contents of In and Sn were 300 ± 25 and 100 ± 20 ppm, respectively, which corresponds to results obtained in other works where the indium content of the processed panels was within a range of 50 ppm to 600 ppm [1, 15, 16].

### 3.2. Impact of temperature and extraction time

The impact of temperature and time on the efficiency of In, Sn, Fe, and Al extraction from milled LCD panels was investigated for 2 M H<sub>2</sub>SO<sub>4</sub> in temperatures ranging from 30°C to 70°C and processing times from 15 minutes to 24 hours. The research results are presented in Figure 2.

What is particularly clear is the strong impact of temperature on the extraction efficiency of In and Fe during the first minutes of the process. In the research conditions for a temperature of 70°C, the extraction degree of In exceeds 70% and 45% for Fe after only 15 minutes. Whereas at 30°C, the extraction degree after 15 minutes was just 25% for In and 21% for Fe. The extraction process at 70°C allowed us to achieve a nearly 100% extraction rate for indium after 8 hours, with solution concentration levels in the range of 35 mg/l, whereas the maximum indium gain at 30°C after 24 hours was in the range of just 80% and concentrations below 30 mg/l. When we compare the aforementioned results with those obtained by Rocchetti *et al.* [1] (where an indium extraction rate of nearly 100% was achieved after 20 minutes of extraction at 80°C), we see how significant the temperature is for the extraction process. The leaching efficiency is influenced far less by temperature for Fe and Sn. After 24 hours, the leaching degree of Fe ranged between 50–55% (depending on the temperature used) and 55–65% in the case of Sn. A strong influence of temperature on the final leaching degree is only visible in the case of aluminum, where at 30°C, only 3% of this element was transferred to the solution; an increase of temperature to 70°C raises the final leaching efficiency to 12%, and the concentration of Al<sup>3+</sup> ions in the resulting solution exceeds 1g/l. We can adopt 8 hours as the time required for achieving concentrations of In, Sn, and Fe ions for the whole investigated temperature range; a further extension of leaching times does not cause a significant increase in In concentrations and causes only a slight increase in Al<sup>3+</sup> ion levels, which can be considered as undesirable in this solution. When indium extraction is considered, extraction at 70°C can be finished just after 2 hours, with leaching efficiency near 90% for that period of time.

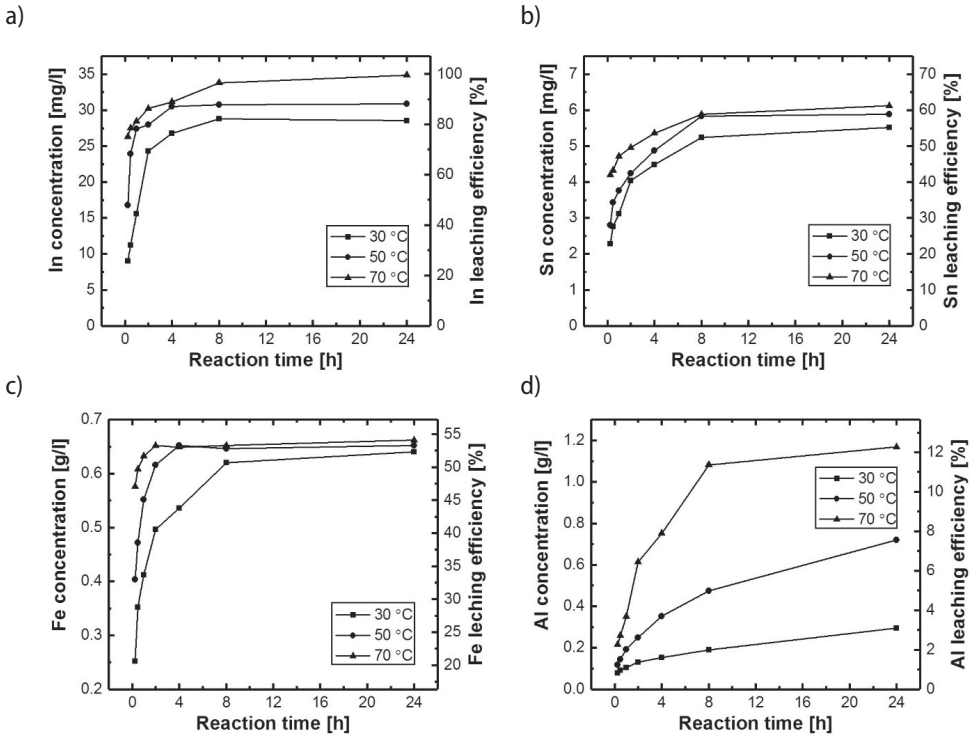


Fig. 2. Effect of temperature on: a) indium leaching; b) tin leaching; c) iron leaching; d) aluminum leaching

### 3.3. Influence of acid concentration on extraction efficiency

In order to investigate the influence of sulfuric acid concentration on In, Sn, Fe, and Al extraction efficiency for milled LCD panels, we conducted an experiment with  $H_2SO_4$  concentrations ranging from 0.01 M to 2 M. The experiment was conducted at a constant temperature of 70°C for 2 hours, and the ratio of the solid to liquid phase was 0.1 g/ml. The change in In, Sn, Fe, and Al ion concentrations in the solutions as well as the leaching efficiency are presented in Figure 3. Extraction of indium under the aforementioned conditions ensures an extraction efficiency of greater than 80%, provided that the  $H_2SO_4$  concentration is equal to or higher than 0.5 M. For  $H_2SO_4$  concentration ranges of 0.1 M to 2.0 M, the extraction efficiency of Fe is rather constant. In the case of Sn and Al, a significant decrease in extraction efficiency was observed for  $H_2SO_4$  concentrations below 1.0 M. Therefore, it seems beneficial from the viewpoint of indium leaching to lower the  $H_2SO_4$  concentration in the solution to approximately 0.5 M, as this lowers the Sn and Al ion content in the solution.

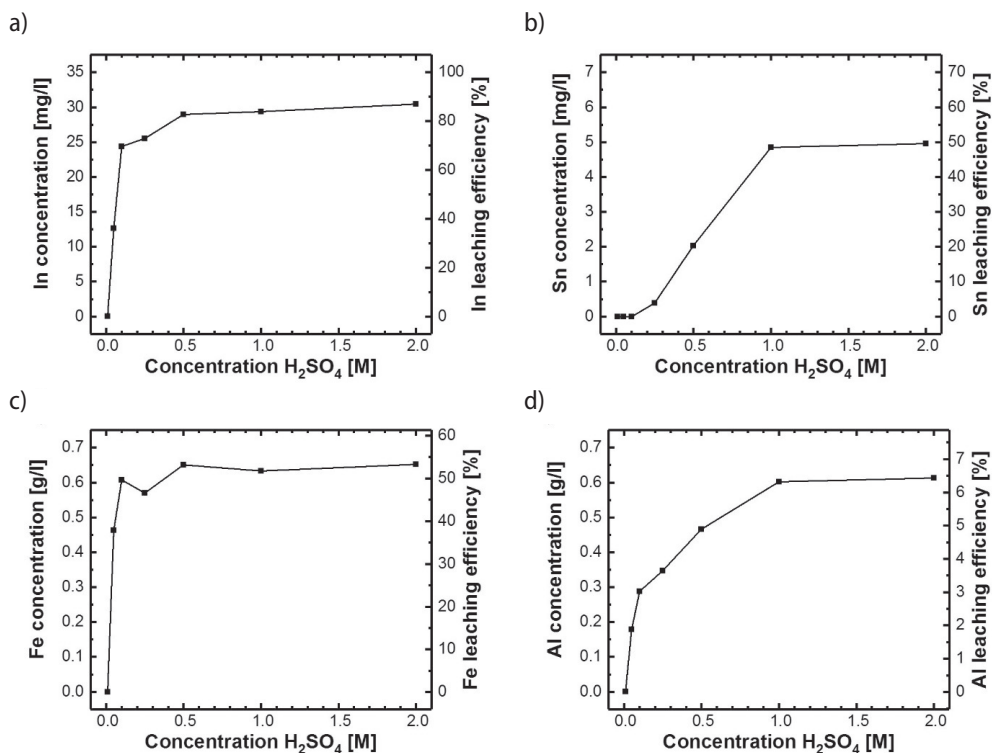


Fig. 3. Determination of concentration and leaching efficiency for In (a), Sn (b), Fe (c), and Al (d) with respect to  $H_2SO_4$  concentration

## 4. Conclusions

- The efficiency of indium leaching from milled LCD panels in a 2 M solution of sulfuric acid at  $70^\circ C$  is near 100% after 8 hours of the process, and the indium concentration in the solution is approximately 35 mg/l.
- Within the investigated temperature ranges, the concentrations of indium, iron, and tin are stabilized in the solution after 8 hours of leaching.
- Increases in extraction temperature beneficially influences the indium-leaching degree and the rate of the process. An increase in process temperature from  $30^\circ C$  to  $70^\circ C$  allows us to reach similar results in 2 hours instead of 24.
- Lowering the  $H_2SO_4$  concentration to 0.5 M does not cause a significant decrease in the efficiency of In extraction but significantly lowers the concentration of the undesirable Sn and Al ions in the solution.
- Extensions of extraction time to times in excess of 8 hours only causes an increase in  $Al^{3+}$  ion concentration in the solution for each of the investigated temperatures; this should be considered detrimental.

## Acknowledgements

The financial support from the Polish Ministry of Science and Higher Education Contract No. 15.11.180. 912 is gratefully acknowledged.

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