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Separation and recovery of glass, plastic and indium from spent LCD panels

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ABSTRACT

The present paper deals with physico-mechanical pre-treatments for dismantling of spent liquid crystal displays (LCDs) and further recovery of valuable fractions like plastic, glass and indium. After a wide experimental campaign, two processes were designed, tested and optimized. In the wet process, 20%, 15% and 40% by weight of the feeding panels are recovered as plastic, glass and indium concentrate, respectively. Instead, in the dry process, only two fractions were separated: around 11% and 85% by weight are recovered as plastic and glass/indium mixture. Indium, that concentrated in the −212 µm fraction, was completely dissolved by sulphuric acid leaching (0.75 mol L^{-1} H_2SO_4 solution, 80 °C, 10% vol H_2O_2, pulp density 10%wt/vol, leaching time 3 h). 100% of indium can be extracted from the pregnant solution with 5%wt/vol Amberlite™ resin, at room temperature and pH 3 in 24 h. Indium was thus re-extracted from the resin by means of a 2 mol L^{-1} H_2SO_4 solution, at room temperature and S/L of 40% wt/vol.

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1. Introduction

Nowadays liquid crystal displays (LCDs) are widely used in TVs, laptops, desktops and any other device coupled with a screen. LCD panels belong to waste electronic and electric equipment (WEEE) that contain a wide range of compounds, some of which are toxic or hazardous whereas others like base and precious metals are valuable and can be recovered. Recovery of scarce resources like the critical raw materials (CRMs) was pointed out in many European policies: some are focused on waste treatment (Directive 2008/98/EC; Directive 2012/19/EU), whereas others address promotion of cleaner and more environmentally-friendly productions (Ecodesign Directive 2009/125/EC and Ecolabel Regulation 66/2010) (Ardente et al., 2014). CRMs at EU level are the following: antimony, beryllium, cobalt, fluorspar, gallium, germanium, graphite, indium, magnesium, niobium, PGMs, rare earths, tantalum, tungsten. The aim of the study carried out by the European Commission was to work out a detailed analysis of the CRMs and the relevant risks and issues of their supply chains, and forward looking trends and forecasts for supply and demand, also giving some guidelines and suggestion on how to establish and improve recycling schemes (European Commission, 2014).

In 2012, 9051 kt of EEE were put on the global market, of which 3474 kt were collected once spent. 2601 kt were recovered, i.e. recycled and/or used for energy recovery worldwide. The main producers of indium in the period 2010–2012 were China (58% of the total world production), Japan (10%), South Korea (10%) and Canada (10%). In 2012, the indium imports of EU came mainly from China (24%), Hong Kong (19%), Canada (13%) and Japan (11%) (EUROSTAT, 2012). Indium has a sustainability index equal to 0.82, whereas the end-of-life recycling input rate is 0%. The sustainability index is a measure of the difficulty in substituting the material, scored and weighted across all applications. Values are between 0 and 1, with 1 being the least substitutable. The recycling input, instead, take into account the proportion of metal and metal products that are produced from end-of-life scrap and other metal-bearing low grade residues in end-of-life scrap worldwide. As previously mentioned, indium plays a crucial role in our electronic devices, so that it is important to develop and improve new recycling methods. In this perspective, it would be very interesting to estimate the impact of recycled indium that could be reasonably recovered from WEEE. A deep study on the potential of indium recycled from LCDs in China during the period from 2015 to 2030 was performed by Wang et al. (2015). This study highlighted that 350 tonnes of indium will be required in manufacture of LCDs in China by 2035, but recycled indium will only account for 48% of the total indium demand. An extensive study on how to enhance

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the resource efficiency of energy using products, with a special focus to LCDs, was carried out by Ardente and Mathieux (2014). The proposed method is divided into five steps: reusability, recyclability, recoverability, recycled and use of hazardous substances. Peeters et al. (2013) concluded that recycling of TVs by mechanical treatments achieves recovery of less than 10% of the precious metals, while the manual dismantling of such waste increases the recovery of more than 90% of metals. Regarding printed circuit boards (PCBs), automatic pre-treatments like gravity, magnetic, electrostatic, density and pneumatic separation are usually used to recover metals, but they are characterized by heavy metal loss up to 35% (Birloaga et al., 2013). Our group is one of the first research groups that investigated a full recycling process in order to recover all the valuable fractions from LCD panels. That work started in 2009 in the ambit of the 7th FP of the European Union (ID FP7-SME-2008-1 HydroWEEE (2009–2012) and continuing within the second HydroWEEE-DEMO 308549 project (2012–2016).

1.1. Thermal pre-treatments

Thermal treatments, especially pyrolysis, were applied as pre-treatment of LCD panels in order to remove organics prior to leaching of indium. Pyrolysis of polarizing films at 570 °C under nitrogen flow produced 15.4%wt gas, 70.5%wt oil and 14.1%wt char, the last one containing ITO that can undergo leaching stage. (Wang et al., 2013; Wang and Xu, 2014a, 2016). In the work by Lu et al. (2012), the pyrolysis temperature was set at 577 °C. The indium fraction was thus concentrated in the solid residue, from which it can be recovered by acid leaching. Removal of the organic fraction and further recovery of indium were also the goals of the paper written by Li et al. (2009). The LCD polarizing film was separated by a thermal shock, hence liquid crystals between glass substrates were removed by ultrasonic cleaning. Indium can thus be recovered from glass by leaching. Thanks to these pre-treatments, 85% wt of indium can be extracted from the ITO glass. Indium was then dissolved by a leaching with diluted aqua regia at 60 °C for 30 min, leading to a recovery of 92%wt. Vacuum carbon-reduction is an alternative way to reclaim indium from waste LCDs, avoiding hydrometallurgical treatments. The overall recovery rate of indium was equal to 90%wt (He et al., 2014).

1.2. Mechano-chemical pre-treatments

Hasegawa et al. (2013) recovered ITO by aminopolycarboxylate chelants (APCs) in combination with mechano-chemical pre-treatments. Crushed LCD panels were milled from 0.5 to 10 h, hence five chelants were tested to extract ITO, both during and after the grinding process. Mechanical activation was also used by Lee et al. (2013). Glass was crushed to micron size particles by using high energy ball grinding. The best indium extraction yield was 86%.

1.3. Extraction and recovery by hydrometallurgy

Solvent extraction is a typical technique used in hydrometallurgy. Indium(III) can be effectively extracted by Cyanex 923 dissolved in toluene from HCl, H_2SO_4 and HNO_3 solutions. Indium can be stripped off selectively from the organic phase with a final recovery of 95% and 99% purity (Gupta et al., 2004). Leaching of crushed LCD glass was investigated by using HCl, HNO_3 and H_2SO_4. Indium was extracted from pregnant solutions with DEHPA, TBP, Cyanex 272 and Cyanex 923. More than 99% of indium was recovered with a purity of 90% (Yang et al., 2013, 2016). Swain et al. (2016) investigated the beneficiation of H_2O_2 as oxidant during leaching of indium, with a good enhancement of the extraction yield.

Recovery of indium and polarizing film from waste LCDs was also investigated by Fontana et al. (2015). Thermal and chemical treatments with liquid nitrogen and some solvents were applied; indium-containing powder was leached with different strong acids and indium was preliminary recovered by solvent extraction with polyethylene glycol-ammonium sulphate-water system. Kato et al. (2013) carried out liquid-liquid extraction of indium from a hydrochloric acid synthetic solution that simulated the extraction of indium and other metals such as Fe, Al, Ca, Sn, Si, Sr from dismantled LCDs. Zonyl FSA dissolved in acetone was used as extractant. The extraction of indium in the sedimented phase was greater than 96%. Adsorption of indium onto resins was also tested in the past. For instance, Yuan et al. (2010) studied various parameters such as solution pH, ionic strength, contact time, indium ion concentration and flow rate were studied in the adsorption of indium (III) onto coated solvent impregnated resins. The adsorption capacity of impregnated resins did not change significantly after ten sorption-desorption cycles. Macroporous resin HZ830 impregnated with trialkylphosphate Cyanex 923 was used to adsorb indium from hydrochloric acid solution; the adsorption efficiency of indium was greater than 90% (Wei et al., 2016). Rocchetti et al. (2015) focused on maximization of indium extraction in leaching of LCD panels, after crushing and grinding pre-treatments. H_2SO_4 concentration, leaching temperature, time and number of steps in the cross-current leaching were investigated. Increase of number of stages increases indium concentration up to 115 mg L^{-1} in the sixth stage, but contemporaneously the indium extraction yield decreases. Savvilotidou et al. (2015) studied the leaching behaviour of In, As and Sb from milled LCDs. It was found that hydrochloric and nitric acid solutions were the most effective, although recovery of In was rather low (60%). Cementation by zinc powder is another way that can be used in recovery of indium from pregnant solutions. Almost 99.8% of indium contained in the leaching solution can be recovered in optimal conditions (Rocchetti et al., 2016). Indium is also recovered from LCD screens of discarded cell phones, for instance by leaching and precipitation with NH_4OH (Silveira et al., 2015).

Indium can be extracted from LCD glass by sub-critical water. NaOH, KOH, Na_2CO_3, diethyl amine, $Ca(OH)_2$ and NH_3 were used as alkaline reagents: the recovery yield was 99% at 160 °C for colour filter glass (Yoshida et al., 2015). Chloride volatilization was studied by Park et al. (2009), who recovered indium from LCD panels by using PVC as chlorination agent. The study focused on recovery of In from pure In_2O_3 and spent LCD powder. Recovery of indium from In_2O_3 increased with an increasing molar Cl/In ratio in N_2 and air atmospheres.

1.4. Recycling of LCD glass

Recycling of glass from dismantled LCD panels can also be carried out directly, for instance as raw material for production of glass-ceramic tiles. After crushing, grinding and sieving at 16 mesh, the glass powder was heated up at 800–950 °C for 6 h. The characterization of the end-products confirmed the full recyclability of such waste glass (Lin et al., 2009). Kim et al. (2016) also used waste glass from LCD panels to manufacture ceramic tiles. Overall results were good, since properties such as water absorption and thermal expansion coefficient were positively affected by LCD waste glass replacement at 1100 °C. Spent LCD and cathode ray tube (CRT) glasses can be used as partial or full aggregate replacement in cementitious materials. Some properties are enhanced and others are defected with the inclusion of such recycled glasses (Rashad, 2015). LCD glass is also recycled in partial replacement for cement powder in mortar production. Wang (2011) investigated 10, 20, 30,

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