



Investigations regarding the wet decontamination of fluorescent lamp waste using iodine in potassium iodide solutions



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ARTICLE INFO

Article history:

Received 22 May 2014

Accepted 24 October 2014

Available online 28 November 2014

Keywords:

Florescent lamp waste

Mercury

Iodine

Decontamination

Leaching

Waste treatment

ABSTRACT

With the rising popularity of fluorescent lighting, simple and efficient methods for the decontamination of discarded lamps are needed. Due to their mercury content end-of-life fluorescent lamps are classified as hazardous waste, requiring special treatment for disposal. A simple wet-based decontamination process is required, especially for streams where thermal desorption, a commonly used but energy demanding method, cannot be applied. In this study the potential of a wet-based process using iodine in potassium iodide solution was studied for the recovery of mercury from fluorescent lamp waste. The influence of the leaching agent's concentration and solid/liquid ratio on the decontamination efficiency was investigated. The leaching behaviour of mercury was studied over time, as well as its recovery from the obtained leachates by means of anion exchange, reduction, and solvent extraction. Dissolution of more than 90% of the contained mercury was achieved using 0.025/0.05 M I₂/KI solution at 21 °C for two hours. The efficiency of the process increased with an increase in leachant concentration. 97.3 ± 0.6% of the mercury contained was dissolved at 21 °C, in two hours, using a 0.25/0.5 M I₂/KI solution and a solid to liquid ratio of 10% w/v. Iodine and mercury can be efficiently removed from the leachates using Dowex 1X8 anion exchange resin or reducing agents such as sodium hydrosulphite, allowing the disposal of the obtained solution as non-hazardous industrial wastewater. The extractant CyMe₄BTBP showed good removal of mercury, with an extraction efficiency of 97.5 ± 0.7% being achieved in a single stage. Better removal of mercury was achieved in a single stage using the extractants Cyanex 302 and Cyanex 923 in kerosene, respectively.

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1. Introduction and background

Mercury is known to be a toxic element for living organisms (Clifton II, 2007). Despite this, due to its physical and chemical properties, mercury is an important constituent in various applications, fluorescent lamps being a representative example. These types of lamps rely on the excitation of mercury vapours to produce UV photons that are converted to visible light by phosphors containing rare earth elements (REEs) (NEMA, 2005; Ronda et al., 1998). Every year large amounts of fluorescent lamps come onto, and are taken off the market. The number of compact fluorescent lamps (CFLs) sold in the US and Europe in 2007 was approximately 397 and 288 million, respectively (McKeown and Swire, 2009; Travis, 2011; US Department of Commerce, 2007). Because mercury is a vital constituent in all fluorescent lamps (Jang et al., 2005; Raposo et al., 2003), such end-of-life products are classified

as hazardous waste and require proper collection, treatment and disposal.

Although the quantity of mercury used in the lighting industry has decreased significantly in recent decades (NEMA, 2005), products containing amounts above the 5 mg/CFL limit set by the European Union (The European Parliament and the Council of the European Union, 2003) are still common. Analysis of 15 different CFL types revealed total mercury contents of between 1.6 and 27 mg/lamp, with 40% of the investigated samples failing to pass the requirements set by European legislation (dos Santos et al., 2010).

Studies showed that during the use of a fluorescent lamp, most of the mercury is absorbed, mainly in the phosphor powders and, but also in the aluminium end caps and glass (Dang et al., 1999; Doughty et al., 1995; Raposo et al., 2003; Thaler et al., 1995). This makes the recovery of macro fractions and phosphor constituents (e.g. the REEs) much harder to achieve. Special treatment is needed in order to ensure that the recycled components are not contaminated. Studies using thermo-desorption atomic absorption spectrometry (Raposo et al., 2003) have revealed the presence of both

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metallic (Hg^0) and ionic mercury species (Hg^{1+} and Hg^{2+}) in the phosphors powder matrix. The predominant species found were Hg^0 and Hg^{1+} , with some exceptions.

The processing of end-of-life fluorescent lamps for the recovery of some of the contained macro-fractions (e.g. aluminium end caps, electronics, or glass) is common. Crushing of lamps is often used to reduce the volume of the waste and to assist in the separation of these fractions. The presence of mercury, however, poses environmental and health risks, as some of these fractions (notably the phosphors fraction) are contaminated. Decontamination can be achieved via thermal treatment (distillation) or by chemical leaching. Stabilization is a possibility but has the disadvantage of leaving mercury in the stream (Duraó et al., 2008). Electroleaching (Sobral et al., 2004) and heterogeneous photocatalysis (Bussi et al., 2010) have also been suggested for the treatment of the waste. These methods show high mercury recovery efficiencies: 99.5% and 99.9%, respectively. Thermal desorption has been the subject of several studies (Chang et al., 2010, 2009; Duraó et al., 2008; Jang et al., 2005), and it has been concluded that temperatures as high as 800 °C are required in order to liberate all of the mercury absorbed in the glass. The previous results favour heating versus acid leaching as the decontamination method for the glass residues. The latter has been reported to be less efficient: up to 36% mercury recovery (Jang et al., 2005). Nevertheless, the mercury absorbed in the glass accounts for a small amount compared to the mercury absorbed in the phosphors (13.66% vs. 85.76%) and, by removing all of the powder attached to the glass surface, it is possible to reduce the mercury concentration in the glass below the limits set by legislation (Rey-Raap and Gallardo, 2012). Strong oxidants, such as hypochlorite and nitric acid solutions, have been found to be efficient for decontamination of phosphor powders; removal of more than 99% of absorbed mercury has been achieved (Sobral et al., 2004).

Despite these results, the decontamination of certain fluorescent lamp waste streams raises some issues, especially if the recovery of the REEs contained in the phosphor powders is desired. Fluorescent lamps are acknowledged to be important urban mining sources of REEs – specifically yttrium, whose recovery has been the target of several studies (De Michelis et al., 2011; Innocenzi et al., 2013).

Thermo-desorption is an energy intensive process that requires access to special equipment. This method can prove troublesome in a process where large batches of fluorescent lamps are crushed together and separation of constituents is carried out using wet sieving procedures – a treatment employed by several European lamp processors (Binnemans et al., 2013). Moreover, a stabilization step meant to contain elemental mercury during crushing (e.g. hypochlorite treatment) may lead to additional problems, and limit the applicability/efficiency of some of the previously mentioned processes. The moisture in the waste, and the higher desorption temperatures needed by mercury compounds in higher oxidation states versus metallic mercury (Raposo et al., 2003), may require additional optimization or other decontamination methods. A wet decontamination process employing nitric acid will interfere with the subsequent recovery of the REEs contained in the phosphor powders. Some of the REEs will be leached together with mercury, complicating their recovery and leading to additional secondary waste streams. A simpler decontamination process that uses common chemical reagents, and does not require complicated or expensive equipment, is desired for treating waste fractions obtained via wet-sieving techniques.

In this study the hydrometallurgical removal of mercury from real fluorescent lamp waste fractions, obtained via a wet-based treatment of spent appliances, is addressed. Decontamination is desired to further process the stream for the recovery of the contained REEs. Because of this, emphasis was placed on a selective

leaching process for mercury. The potential of using iodine in potassium iodide solutions (hereafter denoted as I_2/KI) was investigated. This leaching agent has previously been used with good results for the removal of mercury from soil and other contaminated materials (Cox et al., 1996; Ebadian, 2001). The leaching of fluorescent lamp waste using I_2/KI solutions of various concentrations and at different solid/liquid ratios was studied. The results were compared with previously published results that have been obtained by treating the same type of material with ammonium chloride, hydrochloric, nitric and acetic acid solutions (Tunsu et al., 2014a). Investigations regarding other possibilities for the recovery of mercury from the iodine based leachates, aside from those already documented (Foust, 1993; Mattigod et al., 2009), were carried out. Three processes were studied: anion exchange using Dowex 1X8 resin, reduction using sodium hydrosulphite, and solvent extraction using 6,6'-bis(5,5,8,8-tetramethyl-5,6,7,8-tetrahydro-benzo-[1,2,4-triazin-3-yl]-[2,2']bipyridine (CyMe₄BTBP), trialkylphosphine oxides (Cyanex 923) and bis(2,4,4-trimethylpentyl) monothiophosphinic acid (Cyanex 302).

2. Experimental

2.1. Samples and chemicals

The waste was collected from a facility specializing in the treatment of spent lighting appliances. A wet process involving the crushing of end-of-life fluorescent lamps, followed by chemical stabilization of elemental mercury was tested by the recycler. Macro-fractions such as aluminium end caps, glass, plastics and electronic boards were separated and recovered in the process. The remains, a wet mix of phosphors powder, glass fragments, metallic and plastic parts, and other impurities, constitute the waste that was sampled. The material was previously characterized and investigated in order to assess its recycling potential, and the main findings are reported elsewhere (Tunsu et al., 2014a).

I_2/KI solutions of various concentrations (0.005/0.01 M, 0.025/0.05 M, 0.05/0.1 M and 0.25/0.5 M) were prepared by dissolving solid iodine (99.8%, Sigma Aldrich) and potassium iodide (>99.5%, Merck) in pure water (MilliQ, Millipore, >18 MΩ/cm). The aqua regia used in the experiments was prepared directly prior to its use by mixing three parts hydrochloric acid solution (37%, puriss, Sigma Aldrich) and one part nitric acid solution (64–66%, puriss, Sigma Aldrich). Dowex 1X8 resin (chloride form, Fluka) and sodium hydrosulphite (>82%, Sigma Aldrich) were used for the removal of mercury from the iodine-based leachates. The CyMe₄BTBP extractant used in the solvent extraction experiments was synthesized in-house. The other organic solvents were used as supplied by the manufacturer, with no additional treatment: nitrobenzene (>99%, Fluka), bis(2,4,4-trimethylpentyl) monothiophosphinic acid (Cyanex 302, Cytec), Cyanex 923 (93%, Cytec), and kerosene with low aromatic content (solvent 70, Statoil).

Three replicate tests were performed for each experiment in order to estimate the errors. All aliquots were filtered using polypropylene filters (0.45 μm, VWR). The investigated elements were quantified using Inductively Coupled Plasma-Optical Emission Spectroscopy (ICP-OES) (iCAP 6500, Thermo Fischer).

2.2. Mercury content of the investigated waste

An estimation of the average soluble mercury content in the waste was carried out using aqua regia; a reagent mentioned in various EPA methods for analysing mercury in solids/sediments. Waste samples were dried at 21 °C under normal laboratory conditions. Approx. 1 g dry waste was treated with 10 ml aqua regia at

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