



Removal of mercury bonded in residual glass from spent fluorescent lamps

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ABSTRACT

The current technologies available for recycling fluorescent lamps do not completely remove the phosphor powder attached to the surface of the glass. Consequently, the glass contains the mercury diffused through the glass matrix and the mercury deposited in the phosphor powder that has not been removed during treatment at the recycling plant. A low-cost process, with just one stage, which can be used to remove the layer of phosphor powder attached to the surface of the glass and its mercury was studied. Several stirring tests were performed with different extraction mixtures, different liquid–solid ratios, and different agitation times. The value of the initial mercury concentration of the residual glass was 2.37 ± 0.50 µg/g. The maximum extraction percentage was 68.38%, obtained by stirring for 24 h with a liquid–solid ratio of 10 and using an extraction solution with 5% of an acid mixture prepared with HCl and HNO₃ at a ratio of 3:1 by volume. On an industrial scale the contact time could be reduced to 8 h without significantly lowering the percentage of mercury extracted. In fact, 64% of the mercury was extracted.

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1. Introduction

Mercury is one of the most toxic elements on Earth, yet it is essential to achieve the generation of ultraviolet radiation in a fluorescent lamp (Sobral et al., 2006). This heavy metal, both in its excited state and in its ionic state, is very reactive and hence it interacts with the different components of the lamp and produces strong bonds (Hildenbrand et al., 2000; Doughty et al., 1995; Thaler et al., 1995; Dang et al., 2002). During lamp usage, metallic mercury reacts with the phosphor powder to form more soluble compounds of mercury (Lee et al., 2009). In fact, when a lamp becomes waste, most of the mercury is in its divalent state (Dunmire et al., 2003) and, furthermore, its components (especially the phosphor powder (Rey-Raap and Gallardo, 2012)) are contaminated by mercury (Durão et al., 2008).

In view of this situation, the European Commission, under Directive 2002/95/EC on waste electrical and electronic equipment (WEEE), states as one of its objectives the reduction of hazardous components in WEEE, in which fluorescent lamps are included (in the case of compact fluorescent lamps (CFLs), the concentration of mercury should not exceed 5 mg per lamp), in order to improve the effectiveness of environmental protection. According to this directive, Member States shall ensure that producers recover a minimum of 70% by average weight per appliance. As a result, it is

necessary to use sophisticated recycling technologies that remove mercury from the components of the lamps (Chang et al., 2007).

The drawback of such technologies is that they do not completely remove the phosphor powder attached to the surface of the glass. Consequently, the glass contains the mercury diffused through the glass matrix and the mercury deposited in the phosphor powder that has not been removed during the plant treatment. This is not a serious problem for countries that have mercury lamp production where the use of this type of glass is feasible. Nevertheless, in countries where there is no lamp production this fact represents an environmental problem to be considered, since if the residual glass did not contain mercury, it could be used in the cement sector and in the ceramics industry, especially in the manufacture of frits and glass mosaic (Chen et al., 2002; Corinaldesi et al., 2005; Caligaris et al., 2011). These sectors use high temperature processes where mercury is easily released and is dragged along with the process air emissions. To improve the possibilities of recovery, in particular for the residual glass that is the major component of the lamps, it is necessary to improve the treatment process or to dispose of an additional treatment that can be applied to the residual glass in order to reduce its mercury concentration.

Much research has been conducted on removal of mercury from flue gases using sorbents, catalysts, photocatalysts, and direct ultraviolet irradiation (Granite et al., 2000; Granite and Pennline, 2002; Granite et al., 2008; Presto and Granite, 2008), whereas less research has been conducted on the removal of mercury from fluorescent lamp waste glass. In 1992 Cogar proposed a wet treatment which, after several washing steps, managed to remove the phosphor powder attached to the surface of the glass and to recover

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the mercury by an ion exchange process. Pogrebnaja et al. (1998) used an alkaline solution to obtain a complex mercury compound which was later submitted to a heat treatment. Sobral et al. (2006) studied the possibility of treating phosphor powder from fluorescent lamps by an electroleaching process that removed 99% of the mercury. Jang et al. (2005) applied a rotary shaking process using different mixtures of acids as extraction solutions, which removed 36% of the mercury. Bussi et al. (2010) combined the extraction solution of sodium hypochlorite with a photocatalytic process in order to reduce the amount of mercury dissolved by using titanium oxide as a catalyst and citric acid as a reducing agent, with this method efficiencies of over 95% were achieved. The advantages of these methods are that they can operate under normal pressure and temperature conditions and in continuous mode, they have a low cost and they can be applied on both large and small scales. But the drawback is that they require more than one stage.

Accordingly, the aim of this research was to study a low-cost process with just one stage by which to remove the layer of phosphor powder attached to the surface of the glass and therefore the mercury deposited in it, while at the same time removing the mercury diffused through the glass matrix. Stirring was utilized to remove the mercury from the residual glass. Several tests were performed with different extraction mixtures, different liquid–solid (L/S) ratios and different agitation times. This made it possible to determine which process features are the optimal to obtain a higher rate of extraction of mercury. The solubility of mercury of the two species (excited and ionic state), is not exactly the same and, therefore, the extraction solutions used in this study should be selected taking into account the solubility of divalent mercury. According to Capri (1997), divalent mercury is more stable in water than elemental mercury and it reacts easily with chlorides to form salts that are soluble in water, such as HgCl_2 . This means that the solubility of divalent mercury increases as pH decreases (Bilinski et al., 1980).

2. Experimentation

2.1. Methodology

The methodology used to carry out this study is shown in Fig. 1. The first step was to perform appropriate quartering to obtain representative results. After quartering, 28 samples were obtained and each of one was further divided into two parts: the first one was used to determine the initial concentration of mercury in the residual glass, and the second one was submitted to stirring with different extraction mixtures, diverse L/S ratios and various contact times. After each stirring process, the solid (glass) was separated from the liquid (extraction solution with suspended phosphor powder). The glass was dried at room temperature for 24 h wrapped in laboratory bench paper, and the pH of the extraction solution was measured. This solution was stored at 4 °C until one hour before the analysis in order to be at room temperature when testing time began. The mercury concentration was determined by a mercury analyzer for both liquid and solid samples.

2.2. Sample preparation

For this study residual glass from a fluorescent lamp treatment plant was used. At the treatment plant the lamps are first broken in a crushing unit, where the different materials are separated while, by injecting compressed air, some of the phosphor powder attached to the surface of the glass is removed. Therefore, the residual glass used in the study contained particles, 70% of which were smaller than 5 mm, and some phosphor powder, which could not be removed in the treatment plant, adhered to its surface.

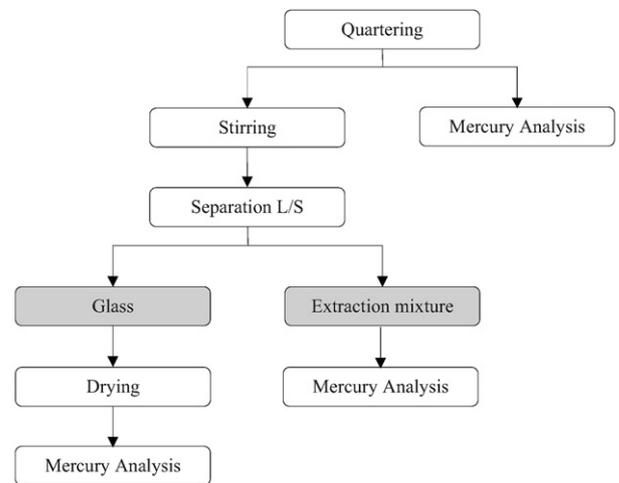


Fig. 1. Working methodology.

After quartering, a total of 28 samples were obtained, each weighing 50 g. These samples were then used to analyze both the initial concentration of mercury and the final concentration of mercury after its corresponding test.

2.3. Stirring processes

The stirring processes were carried out in an SBS/ABT-6 shaker using polypropylene bottles with a volume of 1000 ml. A total of 28 tests were performed, each of which varied the following aspects, as shown in Table 1: acidity of the extraction solution, L/S ratio and time of contact between phases. Each test was performed in triplicate.

2.3.1. Extraction solutions

In this study two extraction solutions were used: ultrapure water and an acid solution. The selection of the acid solution was made taking into account the results of two different research studies. The first one, conducted by Jang et al. (2005), indicates that the use of a mixture of HCl and HNO_3 favors the extraction of mercury over the use of just one of them and that the maximum extraction of mercury is reached when a 5% acid solution is used. The second one, conducted by Fernández-Martínez and Rucandio (2005), indicates that HCl is able to extract more mercury than HNO_3 . Hence, the acid solution used in this research contains 5% of an acid mixture that was prepared with HCl and HNO_3 at a ratio of 3:1 by volume. In each test the samples were placed inside the shaker bottles with a stirring speed of 10 rpm for 24 ± 0.5 h, with an L/S ratio of 10. The pH was

Table 1
Stirring process conditions.

Test no.	pH	L/S	t (hours)	Test no.	pH	L/S	t (hours)
1	Neutral	10	1	15	Acid	10	6
2	Neutral	2	1	16	Acid	2	6
3	Acid	10	1	17	Neutral	10	8
4	Acid	2	1	18	Neutral	2	8
5	Neutral	10	2	19	Acid	10	8
6	Neutral	2	2	20	Acid	2	8
7	Acid	10	2	21	Neutral	10	16
8	Acid	2	2	22	Neutral	2	16
9	Neutral	10	4	23	Acid	10	16
10	Neutral	2	4	24	Acid	2	16
11	Acid	10	4	25	Neutral	10	24
12	Acid	2	4	26	Neutral	2	24
13	Neutral	10	6	27	Acid	10	24
14	Neutral	2	6	28	Acid	2	24

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