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Improving the work environment in the fluorescent lamp recycling sector by optimizing mercury elimination

Marie-Thérèse Lecler [a,*], François Zimmermann [a], Eric Silvente [a], Alain Masson [a], Yves Morèle [a], Aurélie Remy [b], Alain Chollot [a]

[a] Department of Process Engineering, Institut National de Recherche et de Sécurité, rue du Morvan, 54519 Vandœuvre, France
[b] Department of Toxicology and Biometrology, Institut National de Recherche et de Sécurité, rue du Morvan, 54519 Vandœuvre, France

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ABSTRACT

One of the main issues in the fluorescent lamp recycling sector is the mercury contamination of output fractions and occupational exposure associated with recycling operations. The aim of this study is to carry out effective mercury mass balance determinations and improve mercury recovery by finding the optimal levels for the recycling process parameters. These optimizations will allow upstream mercury emissions to be reduced, which will help to avoid mercury exposure among WEEE recycling workers.

Firstly, the distribution of mercury was assessed in new and spent lamps. For new fluorescent tubes, the mean percentage of mercury in the solid phase is lower in new fluorescent tubes (19.5% with 5.5% in glass, 9.7% in end caps and 4.3% in luminescent powder) than in spent tubes (33.3% with 8.3% in glass, 12.9% in end caps and 12.1% in luminescent powder).

The parametric study also shows that the finer the grains of glass, the higher the concentration of mercury (1.2 μg Hg/g for glass size particle >1000 μm and 152.0 μg Hg/g for glass size particle <100 μm); the crushing time required for the optimal removal of mercury from spent tubes is 24 h; on average 71% of the mercury is desorbed at a temperature of 400 °C. The effects of air flow rate, rotation speed and number of balls could not be determined due to wide variations in the results. It is recommended that recycling companies employ processes combining as heating and mixing techniques for the recovery of mercury from lamps in order to both (i) remove as much of the mercury as possible in vapor form and (ii) avoid adsorption of the mercury at new sites created during the crushing process.

Contents

* Corresponding author.
E-mail address: marie-therese.lecler@inrs.fr (M.-T. Lecler).

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1. Introduction

European RoHS Directive, 2011/65/EU and Directive 2012/19/EU on WEEE define the framework for the management of waste electrical and electronic equipment (WEEE) in the European Union. The WEEE Directive is dedicated to the management chain for WEEE, with a view to promoting recycling and reuse rather than sending to landfill. The RoHS Directive focuses on restricting the use of hazardous substances (lead, mercury, cadmium, etc.) in electrical and electronic equipment (EEE).

For example, the use of mercury is still allowed in lamps (maximum authorized content: 2.5 mg per lamp). Indeed, due to its specific physical and chemical properties, such as its liquid form at room temperature and its low vapor pressure, mercury remains an essential material in the production of lamps. Mercury is still required to generate ultraviolet radiation in fluorescent lamps, which are able to produce visible light by making use of the ultraviolet excitation of luminescent powder. The mercury is introduced in the lamps either in elemental form or as an amalgam, and the amount of mercury in the lamps can vary with the type of lamp and company (Chang et al., 2007, 2009).

The toxicity of mercury depends strongly on its chemical form (Nance et al., 2012). In fluorescent lamps, mercury can be present in solid, liquid and gaseous forms, has oxidation states of 0 to 2, and can interact with the constituents of lamps via physisorption, chemisorption, amalgamation and oxidoreduction reactions. Mercury can be emitted both instantaneously (as mercury vapor) and/or time-delayed (the kinetics of emission being a function of the vapor pressure of adsorbed and oxidized forms) (Aucott et al., 2003). The operating conditions and usage time of the lamp can also affect the distribution and speciation of mercury in the different constituents.

In France, ESR (Eco-systèmes Récylum) is the eco-organization in charge of the collection, transportation and processing of spent lamps in order to make use of their glass, metal and plastic fractions. The luminescent powders are not currently recycled for economic reasons; they are sent to landfill. For the last four years, the quantity of spent lamps collected in Fance is relatively stable, at around 4700 tons, or 47,000,000 units of tubes and lamps per year. This must be seen in relation to the entire flux of WEEE, which has increased from 455,000 to 667,000 tons in the same period (ADEME, 2016).

Two main principles of treatment processes for fluorescent lamps are applied in recycling facilities. The first is the "end cut process", which can be only used on fluorescent tubes as the inside of the tube is flushed to remove luminescent powder before the glass and metallic parts are crushed. The second is generally based on shredding, followed by the separation of the glass, phosphor powder and the end caps. For example, well-known processes for treating mercury lamps have been designed and commercialized by MRT System (Mercury Recovery Technology) (Chang et al., 2007).

Due to the toxic effect of mercury, processing fluorescent lamps may cause problems in terms of the environment and health in the workplace. Zhang et al. (2016) estimated the fate of mercury in fluorescent lamps in mainland China, and determined that 27.51 and 11.79 tons of mercury were emitted into the atmosphere and the land respectively. Different exposure scenarios have demonstrated the release of mercury and the consequent risks to human health when compact fluorescent lamps are broken (Lucas and Emery, 2006; Nance et al., 2012). In 2014, an exhaustive occupational exposure study of the lamp recycling sector in France showed that all the stages and processes involved in lamp recycling were affected by high levels of mercury emissions (Zimmermann et al., 2014). The crushed glass, end caps and luminescent powders polluted with mercury are major sources of Hg in the work environment of the employees concerned. In addition to the health in the workplace aspect, recovering mercury from the spent lamps would make it possible to reduce the quantity of waste by allowing the re-use of the glass, plastic and metal fractions and reducing potential environmental risks (Raposo and Roeser, 2001; Durão et al., 2008; Coskun and Civelekoglu, 2014).

Industrial processes aimed at recovering mercury, or at least at decontaminating the mercury in the end fractions (glass, phosphor powder, end caps), are based on wet or dry technologies. Mercury in spent lamps is recovered using various processes that convert the mercury into less toxic compounds or by recovering all the mercury in its pure form (Rey-Raap and Gallardo, 2013). The thermal (pyrometallurgical) process has been very widely developed and applied (Massacci et al., 2000; Fujiwara and Fujinami, 2004, 2005; Durão et al., 2008; Chang et al. 2009). Massacci et al. (2000) removed and recovered all the mercury from the waste without any pre-treatment by thermal desorption at 500–700 °C. Park and Rhee (2016) examined the mercury content of phosphor powders from spent fluorescent lamps using thermal techniques; the optimal conditions were determined to be 400 °C for a retorting time of 6 h. An alternative to the thermal desorption is chemical (hydrometallurgical) leaching (Fábrega et al., 2005; Jang et al., 2005; Tunsu et al., 2014, 2015). Coskun and Civelekoglu (2015) investigated the hydrometallurgical extraction of mercury with oxidative leaching followed by its cementation using three different reducing agents. Sobral et al. (2006) studied the possibility of treating luminescent powders from spent lamps by an electroleaching process that removed 99% of the mercury. Al-Ghouti et al. (2016) showed, firstly, that microwave-assisted leaching combined with classical acid-leaching doubled the amount of mercury leached, and secondly, the potential for detoxifying the leached mercury using bacterial strains. Ozgur et al. (2016) combined oxidative leaching with the electrowinning process to recover 81% of the mercury. Other processes such as stabilization/-solidification were developed by Piao and Bishop, 2006; Randall and Chattopadhyay, 2009; Zhang et al., 2009; Wang et al., 2012. Piao and Bishop (2006) added sulfide and ferric ions to solidify the mercury-containing waste to inhibit mercury release after solidification. These methods have long been used to stabilize hazardous wastes. They are highly effective, but the mercury cannot be re-used. Bussi et al. (2010) combined the extraction solution of sodium hypochlorite with a photocatalytic process. The amount of mercury dissolved is reduced by using titanium oxide as a catalyst and citric acid as a reducing agent. This technique makes it possible to recover more than 95% of the mercury. 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. However, the drawback is that they require more than one stage. Of all these methods, thermal processing is the most widely used process in recycling spent lamps.

The aim of the present study is therefore to reduce the exposure levels of employees at lamp processing plants by eliminating the mercury which might be readily desorbed at an ambient tempera-

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