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Recycling oriented comparison of mercury distribution in new and spent fluorescent lamps and their potential risk

Julia Hobohm [a,*], Oliver Krüger [b], Srotoswini Basu [a], Kerstin Kuchta [a], Sebastian van Wasen [c], Christian Adam [e]

[a] Hamburg University of Technology, Harburger Schlossstrasse 36, 21079 Hamburg, Germany
[b] Bundesanstalt für Materialforschung und -prüfung (BAM), Unter den Eichen 87, 12205 Berlin, Germany
[c] University of Applied Sciences Berlin, Luxemburger Str. 10, 13353 Berlin, Germany
[e] Bundesanstalt für Materialforschung und -prüfung (BAM), Richard-Willstätter Str. 11, 12489 Berlin, Germany

HIGHLIGHTS

- Fluorescent lamps collection volumes in Hamburg.
- A simple method to determine Hg distribution between gaseous phase and phosphor powder.
- Differences in distribution between new and spent FL.
- Testing two digestion mixtures for elemental analysis of the phosphor powder.

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ABSTRACT

This study compares the mercury distribution in the vapor phase, the phosphor powder and the glass matrix of new and spent fluorescent lamps. The spent fluorescent lamps were obtained at the collection yards of a public waste management company in Hamburg, Germany. An innovative systematic sampling method is utilized to collect six spent and eight corresponding new, off-the-shelf fluorescent lamp samples. The efficiency of several acid digestion methods for the determination of the elemental composition was studied and elemental mass fractions of K, Na, Y, Ca, Ba, Eu, Al, Pb, Mg, Hg, and P were measured. The study also finds aqua regia to be the best reagent for acid digestion. However, no significant difference in mercury distribution was found in the different phases of the new and spent fluorescent lamps.

1. Introduction

In Germany, spent fluorescent lamps are already being collected and recycled for recovery of glass, metals and plastic. After using, the end-of-life (EOL) lamps are gathered at collection points and then transported to recycling companies. Certified recycling companies then recycle the lamps to recover materials like glass, metal, plastic etc. Secondary resource recovery, political and legal instruments drive the collection of spent lamps. However, the phosphor powder is disposed of by landfilling due to lack of recycling technologies able to recover Rare Earth Elements (REE) from the phosphor powder. This reverse supply chain can be potentially utilized to recycle REEs from these lamps.

Lightcycle Retourlogistik und Service GmbH, a non-profit organization, is responsible for collection of spent lamps from commercial and private entities in Germany (Lightcycle, 2016a). 7350 Mg of spent lamps were collected by Lightcycle in 2015 (Lightcycle, 2016b).The public waste management company of Hamburg, Stadtreinigung Hamburg, is obligated to collect spent fluorescent lamps and other types of lighting equipment from private households among other electronic waste at 12 collection

* Corresponding author.
E-mail addresses: julia.Hobohm@tuhh.de (J. Hobohm), krueger-oliver@t-online.de (O. Krüger), srotoswini.basu@gmail.com (S. Basu), kuchta@tuhh.de (K. Kuchta), Sebastian.vanwasen@web.de (S. van Wasen), christian.adam@bam.de (C. Adam).

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yards all over Hamburg, according to the collection group 4 classified in ElektroG (§9 Para. 9) or the German implementation of the Waste Electrical and Electronic Equipment (WEEE) Directive 2012/19/EU (Directive 2012/19/EU, 2012). Compact fluorescent lamps (CFLs) are exempt from RoHS, the directive on the Restriction of Hazardous Substances in electrical and electronic equipment which forbids use of hazardous substances such as mercury (Hg) in the manufacturing of electrical and electronic equipment(Directive 2002/95/EC, 2003). According to the amended RoHS (Directive on the restriction of the use of certain hazardous substances in electrical and electronic equipment), the mercury content is to be gradually lowered. The maximum Hg content allowed till 2011, 2012 and 2013 onwards was 5, 3.5 and 2.5 mg respectively (Directive 2011/65/EU, 2011). The indicative benchmark was set at 1.23 mg Hg in lamps as per implementation of the Ecodesign directive (Directive 2005/32/EC, 2005). Even though fluorescence lamps may be replaced - partly or completely - by LED in the future, they will play an important role for the time being. Thus, they have to be considered in terms of resource recovery and diminishing of health and environmental hazards.

Fluorescent lamps contain metallic Hg which upon breakage, vaporizes the Hg contained within. At room temperature, the vapor pressure of Hg is about 100 times the safe limit. Therefore, humans and animals exposed to vapor Hg can potentially exhibit symptoms of Hg poisoning. Humans exposed to broken fluorescent lamps absorb 80–97% of the inhaled mercury through lungs. Dermal exposure to elemental vaporous mercury, however, accounts only for 2.6% absorption. As absorbed elemental mercury is lipophilic, it is able to cross blood-brain-barrier, between the placenta and the foetus relatively easily (Health England, 2016).

(Nance et al., 2012) investigated and reported health risks among humans from exposure to broken compact fluorescent lamps through several scenarios. In the first scenario, mercury emissions are measured after no clean-up and with no ventilation and in the second scenario, mercury emissions are measured after clean-up with ventilation at heights of 1 and 5 feet. Different types of flooring, with or without carpet and different types of ventilation was also investigated. No significant difference in Hg levels were measured in the different types of flooring and ventilation. Acute Reference Exposure Level (REL) derived by the Office of Environmental Health Hazard Assessment (OEHHA) of the California Environmental Protection Agency is 1800 ng/m^3 for a 1-h average exposure to mercury vapor and 300 ng/m^3 level as a "safe" level for lifetime exposure. The 1-h average exposure measured by the study fell below the OEHHA 1-h REL. The OEHHA acute REL was only exceeded by averaged MAXIMUM concentration of Hg. The maximum values were found to rapidly decline below 500 ng/m^3 for both heights of 1 and 5 feet. The study also finds that mercury exposure from new fluorescent lamps pose a higher health risk than spent fluorescent lamps.

In the fluorescent lamp recycling centre in France, occupational exposure of the workers to Hg, Ba, Pb, Y and total inhalable dust was investigated. An analysis of 77 samples revealed a range of 100–86,700 ng/m^3 Hg concentration. The mean Hg concentration 15,400 ng/m^3 exceeds the acute REL values, thereby posing a great health hazard to the workers of the fluorescent lamp recycling plant (Zimmermann et al., 2014). To tackle the problem of mercury exposure among recycling plant workers and also in general public health interest, it is important to analyse the mercury distribution in spent fluorescent lamps.

A study used cold vapor atomic absorption spectrometry to analyse the mercury distribution inside eight spent fluorescent lamps from the same manufacturer by Hg extraction from the vapor phase, phosphor powder, and the contaminated glass matrix. The contaminated glass matrix needs to be washed repeatedly, in order to eliminate the Hg contaminated phosphor powder adhering to the glass matrix. Thereafter, the broken glass can be classified as non-hazardous waste. The mercury in the vapor phase was captured by negative pressure induced with a peristaltic pump. The mercury adhering to the phosphor powder and glass matrix were extracted with acid digestion. Phosphor powder contained 85.76% of the mercury whereas 13.66% was sorbed on to the glass and the vapor phase contained 0.58%. A median Hg concentration of 425.57 ± 46.5 μg/lamp was calculated (Rey-Raap and Gallardo, 2013). (Jang et al., 2005) (dos Santos et al., 2010), and (Taghipour et al., 2014) found similar HG distributions between sorbed and gaseous Hg (89 vs. 8%, 88 vs. 16% and 78.4% vs. 21.6% respectively).

The majority of mercury in fluorescent lamps is expected to be deposited onto the fluorescent powder during the course of lamp usage. This might have an important impact on the respective recycling procedure, especially in terms of work safety. Thus it is important to determine the distribution of Hg between gaseous phase and fluorescence powder and whether the usage of lamps has an influence on the distribution. Thus we tested several spent and new fluorescence lamps and determined their respective Hg amounts, both in the gas phase as well as in the fluorescence powder. Furthermore, we investigated the elemental composition of the various lamps to see possible differences with regard to lamp type and condition. We tested several acid mixtures to find the optimal digestion method.

2. Materials and methods

2.1. Characterization of spent lamps collected through a sorting analysis

The parameters of sorting are selected by comparing EOL lamps to solid waste analysis tools due to lack of WEEE analysis tools. The European Commission drafted a Methodology for the Analysis of Solid Waste (SWA-Tool) from the project "SWA-Tool, Development of a Methodological Tool to Enhance the Precision & Comparability of Solid Waste Analysis Data". The parameters like time, location and sampling unit were selected according to the recommendations suggested by this tool. The stratification of an inhomogeneous parent population is done to divide the population into smaller and more sub-populations. This may have two results – either increased accuracy in the desired result or smaller sample size (European Commission, 2004).

Historical data of collection amounts of spent lamps in 2012 and 2013 from Stadtreinigung Hamburg shows that the amount is 0.52 Mg or multiples of this amount for each collection yard. The collection frequency is however, highly variable, ranging from every month to once in three months for sparsely and densely populated areas, respectively. Hamburg collected 25 and 29 Mg of spent lamps in the year 2012 and 2013, respectively which amounts to 0.014 and 0.016 kg $_{\text{spent lamps}}$/person. The sampling unit per location is one mesh box and one post pallet, as seen in Fig. 1, in accordance to the requirements of Lightcycle Retourlogistik und Service GmbH. The mesh box can accommodate 600 to 800 pieces of lamps, lesser than 60 cm in size and other lamps of special forms and shapes. The post pallets have a capacity of 1200–1400 pieces of lamps longer than 60 cm.

2.2. Sample collection from the collection yards

The novelty of this study is the systematic comparison of spent lamps and the corresponding new lamps in terms of Hg distribution between gaseous phase and phosphor powder, which has not been reported in any previous studies. A sorting analysis was planned and conducted in 2015 in two collection yards, one mesh box and