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Thermoluminescent and photoluminescent studies on the phosphor material from waste compact fluorescent lamps
Estudios de termoluminiscencia y fotoluminiscencia del material fosforescente recuperado de lámparas compactas ahorradoras de desecho

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Abstract

The aim of this work was to investigate the thermoluminescent properties of the recovered phosphor powder from compact fluorescent lamps (energy-saving bulbs) when it is exposed to UV radiation of 254 and 365 nm. This study was complemented with the photoluminescent characterization of this material. Thermoluminescent results showed that UV radiation of 254 nm induce high intensity glow curves characterized by two width peaks located at 162 and 292 °C, while UV radiation of 365 nm produce weak TL signals. The photoluminescence results indicate that the emission spectra are composed by three major bands at 451, 512 and 611 nm corresponding to transitions of Ce^{3+}, $Tb^{3+}$and Eu^{3+}, responsible for the white light produced by these light bulbs.

Keywords: Compact fluorescent lamps; phosphorous material; thermoluminescence; photoluminescence, material recycling.

Resumen

El objetivo de este trabajo fue investigar las propiedades termoluminiscentes del polvo fosforescente recuperado de lámparas fluorescentes compactas (“Focos ahorradores”) cuando se expone a radiación UV de 254 y 365 nm. Este estudio fue complementado caracterizando la respuesta fotoluminiscente de este material. Los resultados TL mostraron que la radiación UV de 254 nm induce curvas de brillo intensas caracterizadas por dos picos localizados en 162 y 292 °C, en tanto que la radiación UV de 365 nm produce señales TL de baja intensidad. Los resultados de fotoluminiscencia indican que los espectros de emisión están compuestos principalmente por tres bandas en 451, 512 y 611 nm, asociadas con transiciones de Ce^{3+}, Tb^{3+} y Eu^{3+}, responsables de la luz blanca producida por estos focos.

Palabras Clave: Lámparas compactas fluorescentes, material fosforescente, termoluminiscencia, fotoluminiscencia, reciclaje.

1. Introduction

“Solid waste” is the term now used to describe non-liquid waste materials coming from domestic, trade, commercial, industrial, agriculture, mining activities and the public services (Bhumika and Kuity, 2021). Solid wastes include vegetables, paper, glass, plastics, wood, foods, electronic components, lamps, radioactive and hazardous wastes. Unfortunately, with the population growth, the volume of the solid wastes produced has increased worryingly due to the high level of pollution that they produce on the ground and air.

Several programs to decrease the wastes production have been implemented in all the world. Most of them are based in simple concepts, for example, waste reduction can be done by reducing the waste generation as well as recycling and reusing some used materials. In particular, recycling of the solid wastes helps in reducing the production of new products and raw materials; additionally, also helps to save energy because this process requires less energy (Bhumika and Kuity, 2021).

One of the solid wastes produced in almost all sectors of the society, comes from the compact fluorescent lamps (CFL) used for lighting. It is worth mentioned that in the last decades, the use of CFLs has increased around the world because

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fluorescent lamps are more efficient converting electricity into light and can substitute incandescent light bulbs with the consequent save of electricity. A CFL consists of a glass tube containing mercury vapor at low pressure and an inert gas like argon or krypton. The interior of the glass tube is coated with a phosphor powder, which fluoresces when is exposed to ultraviolet light. The physical basis is similar to other fluorescent lamps: electrons that are bound to mercury atoms are promoted to excited states, afterwards they return to their ground states emitting ultraviolet radiation which is converted into visible light when it strikes the fluorescent coating powder (Tanushevski. and Rendevski, 2016), (Lokendra, et.al., 2013).

According to the chemical and crystallographic studies reported by (Trinidad Juarez, et.al., 2019), the phosphor powder contain yttrium (Y), barium (Ba), cerium (Ce), europium (Eu) and terbium (Tb) in different proportions; these chemical elements are part of the three crystalline phases: $Y_{1.95}Eu_{0.05}O_3$ (Europium Yttrium Oxide), $MgCe_{0.67}Tb_{0.33}Al_{11}O_{19}$ (Magnesium Cerium Terbium Aluminum Oxide), $Ba_{0.9}MgEu_{0.1}Al_{10}O_{17}$ (Barium Magnesium Aluminum Europium Oxide). It is worth mentioning that these materials have excellent optical properties, however, they are thrown away when the compact fluorescent lamps stop working.

The aim of this work was to investigate the photoluminescent and thermoluminescent properties of the recovered phosphor powder from a CFL when it is exposed to UV radiation of 254 and 365 nm of wavelength. It should be mentioned that thermoluminescence is the thermally stimulated emission of light from a semiconductor following the absorption of energy from any type of ionizing and no-ionizing radiation, such as γ-rays, β^- particles, X-rays and UV light, and it is related to the release of electrons from the electron traps in the irradiated material. The amount of light is proportional to the radiation dose received by the material (Villaseñor Cerón, et.al., 2019).

2. Experimental

The phosphor material was collected from a commercial CFL using a spatula. It consists of a white fine powder as is shown in figure 1a. For the thermoluminescent and photoluminescent analysis, the phosphor material was pressed to obtain pellets of 5 mm in diameter and 1 mm in thickness (figure 1b).

Photoluminescence (PL) measurements were performed at room temperature placing the sample at 60° respect to the excitation light beam. PL spectra were obtained using a FluoroMax 4 spectrofluorometer (Horiba Jobin Yvon) equipped with a 150 W Xenon lamp as excitation source. Emission spectra were acquired using 285 and 394 nm as excitation wavelength, whereas excitation spectra were obtained from 300 to 750 nm keeping the emission at 541 and 611 nm.

For TL analysis, UV irradiation was conducted using a Hg low pressure lamp with emission at 254 and 365 nm wavelength. TL readings were made using a Harshaw model 4000 TL Analyzer; the glow curves were recorded 24 hours after irradiation, from 60 to 360 °C at a heating rate of 5 °C/s. The UV irradiation and TL reading were made under dark conditions.

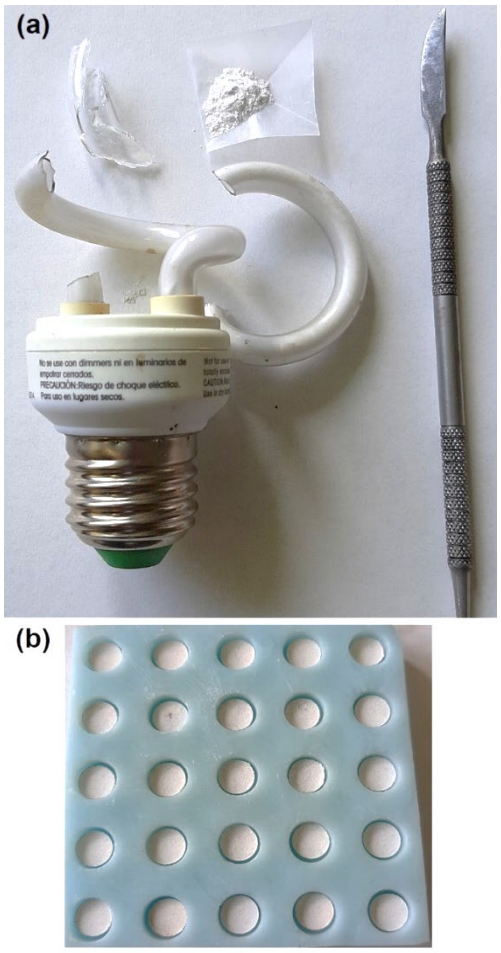

Figure 1. (a) Recovered phosphor powder from a CFL. (b) Pellets of phosphor material.

3. Results

The Photoluminescent emission spectrum of the phosphor material using an excitation wavelength of 394 nm is shown in figure 2. The PL spectrum shows the presence of very intense emissions peaking at 451, 512 and 611 nm and four emissions of lower intensity at 587, 593, 599 and 630 nm. The broad bands at 451 and 512 nm are attributed to the transition of Ce^{3+} and Tb^{3+} whereas the emission line at 611 nm corresponds to Eu^{3+}. These emissions correspond to the blue, green and red colors which combined produce white light, which is the light that the CFL produce for illumination.

Figure 3 shows the PL emission spectrum of the phosphor material excited with 285 nm in order to determine in more detail the emission in the blue and green regions. This spectrum shows a broad band with its maximum close to 450 nm attributed to the localized 5d Ce^{3+} transition. The two intense emission peaks at 486 and 541 nm are associated with forbidden 5D4 → 7F6 and 5D4 → 7F5 transitions of Tb^{3+} ions (Viorel, et.al. 2021).

Figure 4 shows the TL response, as a function of the irradiation time of the phosphor material subjected to UV irradiation of 365 nm. Very wide glow curves with low

intensity are observed. Figure 5 shows a logarithmic increase of the TL intensity as the exposure time increases; this behavior indicates a fast TL-saturation level of the irradiated powder. The low TL signal and nonlinear relationship between TL intensity and irradiation time limit the use of this phosphor material as detector of UV radiation of 365 nm.

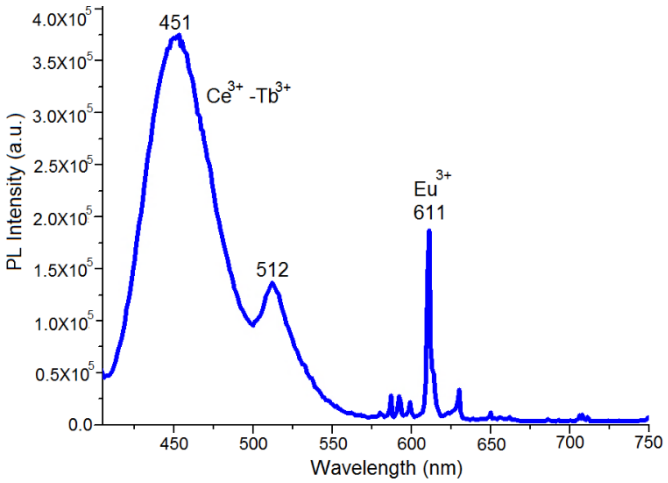

Figure 2. Photoluminescent spectrum of the phosphor material excited with 394 nm wavelength.

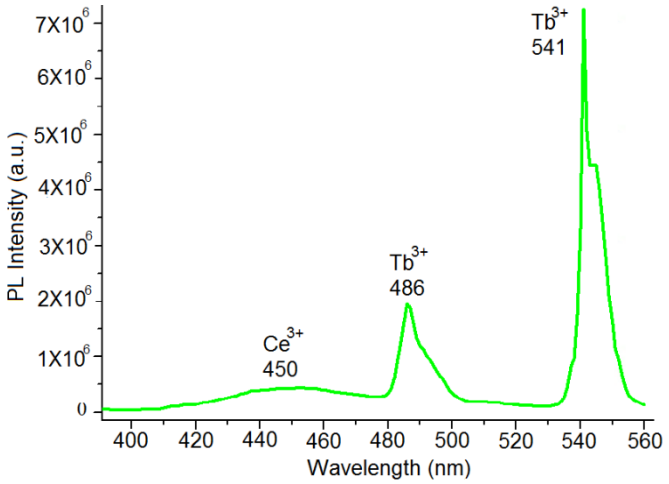

Figure 3. Photoluminescent spectrum of the phosphor material excited with 285 nm wavelength.

Figure 6 shows the TL glow curves obtained after UV irradiation at 254 nm for different times (5, 10, 20, 30, 40, 50 and 60s). In this case the TL glow curves show two wide bands peaking close to 162 and 292 °C. As was mentioned before, the phosphor material is composed by a mixture of $Y_{1.95}Eu_{0.05}O_3$, $MgCe_{0.67}Tb_{0.33}Al_{11}O_{19}$ and $Ba_{0.9}MgEu_{0.1}Al_{10}O_{17}$. To our best knowledge only few works have reported results on thermoluminescent studies of mixtures of these crystalline phases. Nguyen Thi and coworkers reported one prominent peak located at 167 °C when Y_2O_3:Eu^{3+} is exposed to UV radiation (Thi, et.al, 2018); therefore, the TL peak at 162 °C could be associated with the $Y_{1.95}Eu_{0.05}O_3$ crystalline phase. (Nagpal, et.al. 1988) reported the luminescence studies on lamp phosphors; however, the chemical characteristics of the analyzed material and the experimental conditions were different and therefore, a comparison with our results is not possible.

Figure 7 shows the TL response as a function of the irradiation time (5, 10, 20, 30, 40, 50 and 60s). A lineal TL response from 5 to 40 sec is clearly observed. The increase in the TL intensity occurs because with an increase in irradiation time, a higher number of trapping centers responsible for the TL peaks are filled. During TL reading, the heating release charge carriers from the trapping centers and the successive recombination of the charge carriers with their counterparts at the recombination centers increases the TL intensity; it should be mentioned that the linear relation between TL intensity and time irradiation is an important property of TL materials. For higher exposure times, the saturation of the TL response is observed.

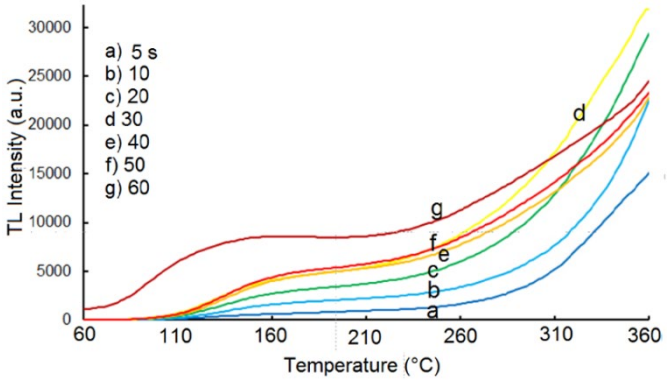

Figure 4. Thermoluminescent glow curves of the phosphor material exposed to UV radiation of 365 nm at different times (seconds).

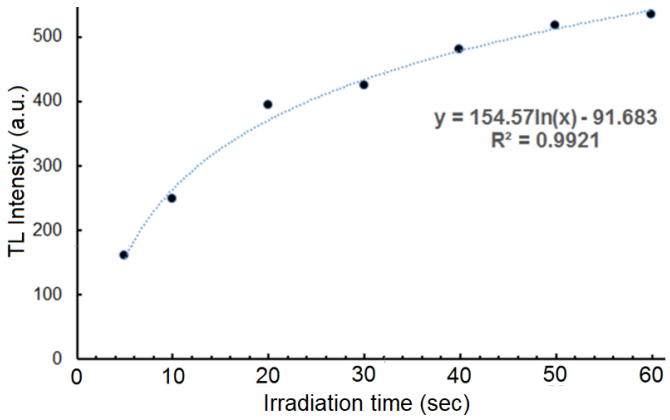

Figure 5. TL response induced by UV radiation of 365 nm as a function of the irradiation time.

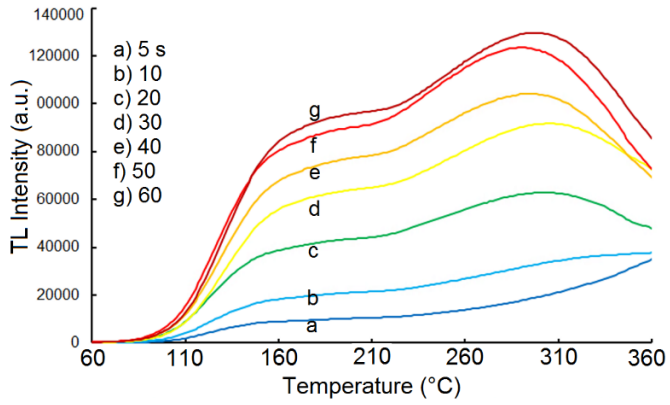

Figure 6. Thermoluminescent glow curves of the phosphor material exposed to UV radiation of 254 nm at different times (seconds)

Figure 8 shows the fading of the phosphor material exposed to UV radiation of 254 nm stored in dark conditions for several days. A fading of around 27% during the first 24 hours after irradiation is observed. This is attributed to the presence of shallow electronic traps. After three days of storage time, a fading of 14% is observed. For longer storage times, from 5 to 10 days, a fading of 10% is reached. These results indicated that this phosphor material stores the information due to the UV irradiation. On the other side, the lineal relationship between TL intensity and exposure time, suggest that this phosphor material could be used as dosimeter of UV radiation of 254 nm.

It is worth mentioning that the use of UV radiation has increased in recent decades in scientific activities (optical or catalytic properties of novel materials, for example) and industrial applications (sterilization of foods, curing, photocatalysis, etc). Therefore, the developments of systems to measure with precision and accuracy the quantity of UV radiation used in specific applications are well received. The results reported in this work can contribute to this field.

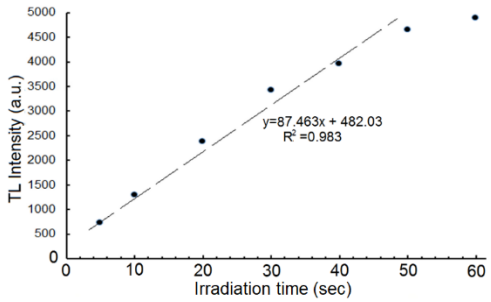

Figure 7. TL response induced by UV radiation of 254 nm as a function of the irradiation time.

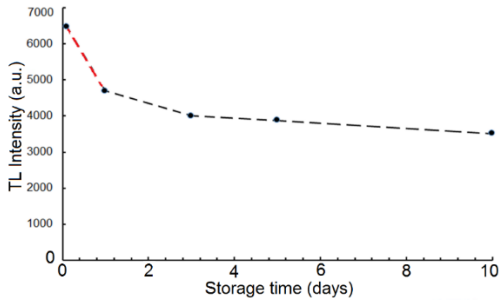

Figure 8: Fading of phosphorous material.

4. Conclusions

The TL characteristics of the investigated phosphor material recovered from discarded CFL indicate that it could be a very good candidate for dosimetry of UV radiation of 254 nm. Additionally, waste fluorescent lamps become an option of cheaper and direct sources of rare earths which can be reused in lighting and optical applications.

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