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Thermo-transferred thermoluminescence (TTTL) in potassium-yttrium double fluoride doped with terbium

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This paper presents results of studying the thermo-transferred thermoluminescence (TTTL) phenomenon in potassium-yttrium double fluoride doped with terbium (K_2YF_5:Tb) at different impurity concentrations (0.8%, 0.95% and 0.99%) Previously to study the TTTL phenomenon, structural characterization and chemical composition of the materials were determined. The structural studies were conducted using a scanning electron microscope (SEM); meanwhile, chemical composition was analyzed using energy dispersive X-ray spectroscopy (EDS). Thermoluminescence (TL) kinetics was studied irradiating the samples with ^{137}Cs gamma rays as well as with ^{90}Sr/^{90}Y beta rays, analyzing the glow curves by the deconvolution method for obtaining the kinetic parameters.

Keywords: Thermoluminescence; rare-earth ions; doping.

En este trabajo se presentan resultados de estudiar el fenómeno de termoluminiscencia termo-transferida (TLTT) en el fluoruro doble de potasio e itrio dopado con terbio (K_2YF_5: Tb) a tres concentraciones diferentes de la impureza (0.8%, 0.95% y 0.99%). Previamente al estudio del fenómeno de TLTT, se determinaron la caracterización estructural y la composición química del material. Los estudios estructurales fueron realizados usando un microscopio electrónico de barrido (SEM); mientras que, la composición química fue analizada mediante espectroscopía de energía dispersa (EDS). La cinética de la termoluminiscencia (TL) fue estudiada irradiando las muestras con rayos gamma ^{137}Cs así como con partículas beta de ^{90}Sr/^{90}Y, analizando las curvas TL por el método de deconvolución para obtener los parámetros cinéticos.

Descriptores: Termoluminiscencia; iones de tierras raras.

PACS: 78.60.Kn; 76.30.Kg; 74.62.Dh

1. Introduction

The thermo-transferred thermoluminescence (TTTL) phenomenon was observed by us, for the first time, during the characterization of the thermoluminescent (TL) properties of K_2YF_5:Tb, after submitting this phosphor to a thermal treatment at a fixed temperature known as isothermal decay (ID) [1,2]. This phenomenon consists in the migration of trapped electrons from low energy meta-stable levels to meta-stable states located at higher energies during the heating of the material at a constant temperature higher than room temperature. The phenomenon is evident due that the material does not show a complete natural fading when is stored at a temperature higher than room temperature, but when it is read after a storing time presents a glow curve characteristic of an irradiated material. Isothermal decay of K_2YF_5:Tb showed an anomalous fading which can be associated to the isothermal-transfer phenomenon which consists in transferring the shallow trapped electrons to deeper traps We have named this phenomenon as thermo-transferred thermoluminescence (TTTL).

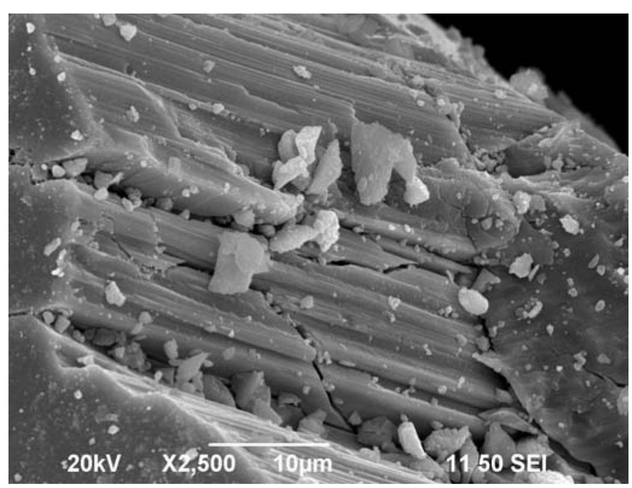

FIGURE 1. K_2YF_5:Tb micrograph obtained by TEM.

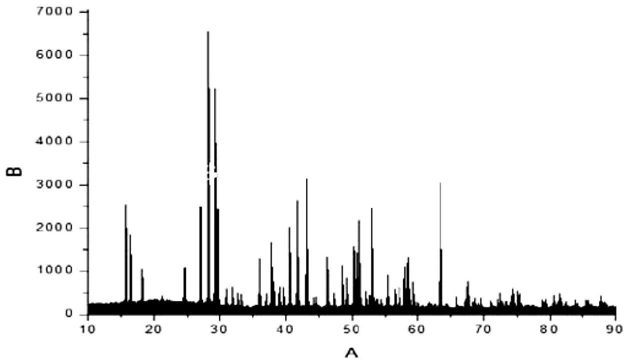

FIGURE 2. X ray diffraction spectrum of K_2YF_5:Tb.

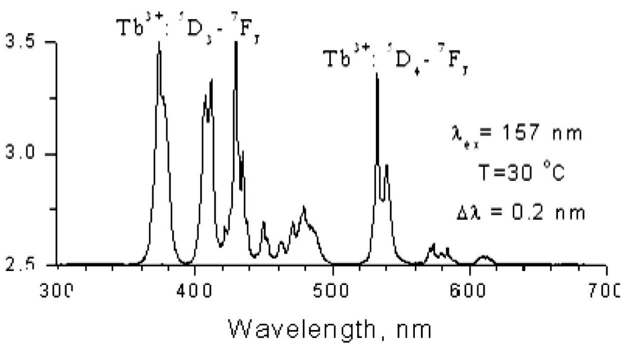

FIGURE 3. Photoluminescent emission spectrum of K_2YF_5:Tb.

In other hand, deconvolution is a general method that allows through a computer-aided analysis the representation of a complex curve as the sum of more elementary components. This method can be applied also to multi-peak glow curves, and shows the advantage in providing a simultaneous evaluation of peak parameters without any further thermal treatment. Besides this consideration, it allows the optimization of some fundamental characteristics of a TL system.

The development of the deconvolution method has become more and more refined. The first work dealt with the numerical analysis of the Randall-Wilkins model [3] using the following expression for the TL intensity as a function of temperature:

$$I(T) = I_m \exp\left(\frac{E}{kT_m} - \frac{E}{kT}\right) \times \exp\left(-\frac{E}{kT_m^2}\int_{T_m}^{T} \exp\left(\frac{E}{kT_m} - \frac{E}{kT'}dT'\right)\right)$$

The integral in this equation has not an analytical solution but it can be approximated by the expression proposed by Bos *et al.* [4] and by Horowitz and Yossian [5].

$$\int_0^T \exp\left(-\frac{E}{kT'}\right) dT' \approx \frac{E}{k}\int_x^\infty x'^{-2} \times \exp(-x')dx' = \frac{E}{k}\frac{1}{x}E_2(x)$$

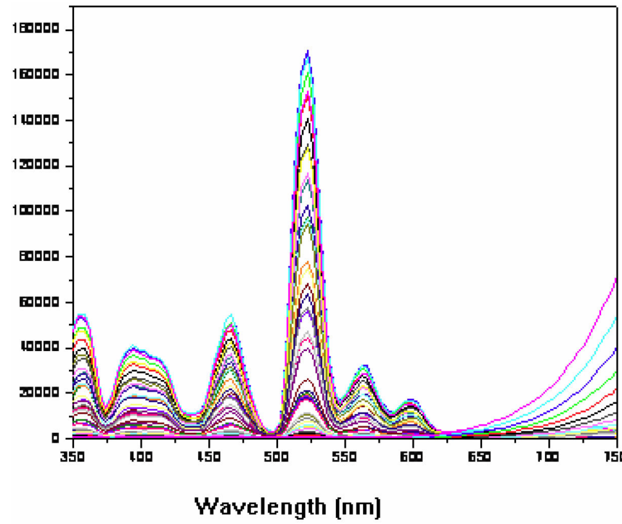

FIGURE 4. TL emission spectrum of K_2YF_5:Tb.

Where: $x = E/kT$, $E_2(x) = \alpha(x)\exp(-x)$, and:

$$\alpha(x) = 1 - \frac{a_0 + a_1x + a_2x^2 + a_3x^3 + x^4}{b_0 + b_1x + b_2x^2 + b_3x^3 + x^4}$$

Then, the Randall-Wilkins equation is reduced to:

$$I(T) = I_m \exp\left(\frac{E}{kT_m} - \frac{E}{kT}\right) \times \exp\left(\frac{E}{kT_m}\left(\alpha\left(\frac{E}{kT_m}\right) - \frac{T}{T_m}\exp\left(\frac{E}{kT_m} - \frac{E}{kT}\right)\alpha\left(\frac{E}{kT}\right)\right)\right)$$

which can be solved using the Levenberg-Marquard method for a non linear function.

2. Experimental methods

Previously to irradiation, structural characterization and chemical composition of the samples of K_2YF_5:Tb (0.8, 0.95 and 0.99%) were determined. Morphology of the samples was observed by TEM (Model JEM-100 CX, JEOL, Tokyo, Japan). The chemical composition and crystalline phases were identified by X-ray diffraction, Siemmens D-500 diffractometer, using CuKα radiation with intervals of 0.03°, using integration time of 0.3 s for each point.

To investigate the thermo-transferred thermoluminescence (TTTL), the samples were irradiated with ^{137}Cs gamma rays at an absorbed dose of 50 mGy and with ^{90}Sr/^{90}Y beta particles at an absorbed dose of 300 mGy. After irradiation, samples were submitted to isothermal annealing, storing them at two fixed temperatures: T_1=120°C and T_2=15°C) obtaining their respective glow curves at different time intervals.

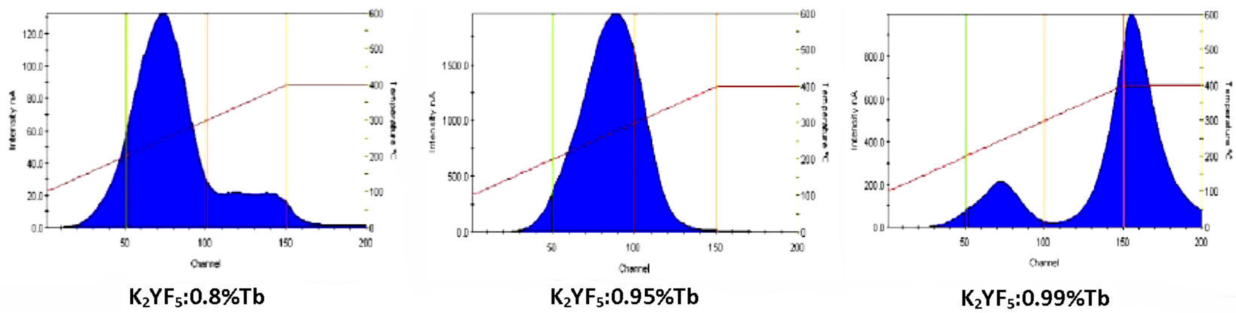

FIGURE 5. Glow curves of K_2YF_5:Tb at three different percentages of dopant immediately after irradiation and before isothermal decay treatment.

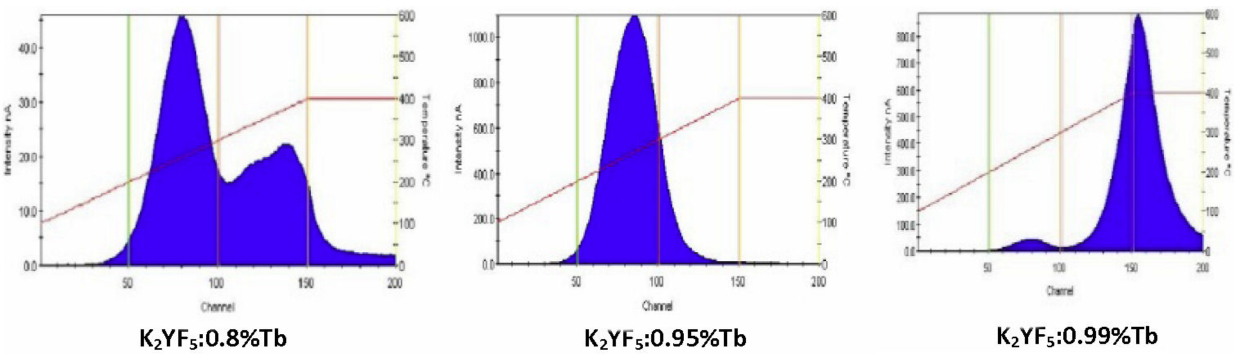

FIGURE 6. Glow curves of irradiated K_2YF_5:Tb at three different percentages of dopant after 40 minutes of isothermal decay at 150°C.

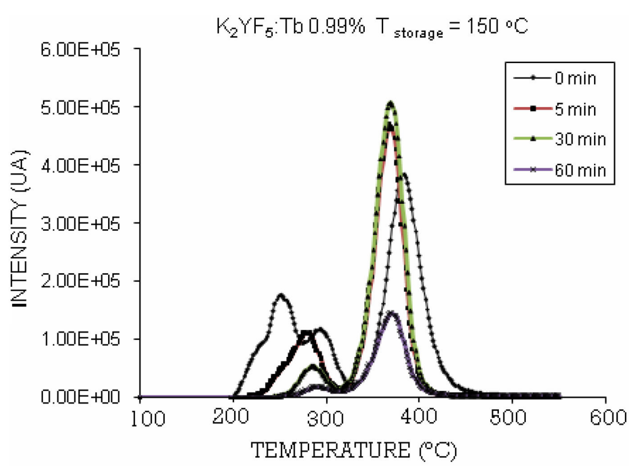

FIGURE 7. Glow curves of K_2YF_5:Tb (0.99% Tb) submitted to an isothermal decay at 150°C during different elapsed time intervals.

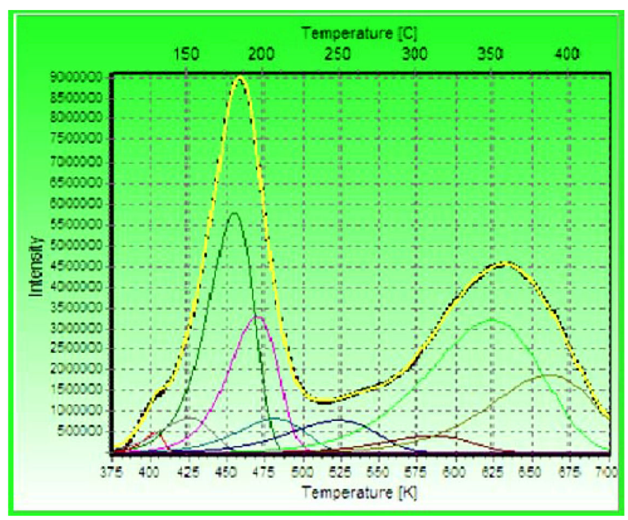

FIGURE 8. Deconvoluted glow curve of K_2YF_5:Tb.

TL readings were performed using a TL analyser Harshaw model 3500, integrating from 100°C to 500°C during 40 s at a heating rate of 10°C/s. Special running, integrating the signal during 200 s at a heating rate of 2°C/s, were performed in the TL reader in order to determining the kinetic parameters by means of the deconvolution method. All TL readings were made in Nitrogen atmosphere to avoid spurious signals. In order to know the quality of the light emitted, the TL emission spectrum was also obtained.

Kinetics parameters were determined using the glow curve deconvolution method as implemented in the GlowFit V1.1 software [6]. This method allows the determination of the activation energy and peak temperature of the isolated peaks in the TL glow curve.

3. Results

The surface morphology of K_2YF_5:Tb powder is presented in Fig. 1. In this micrograph it is possible to observe a crystalline material. The crystalline structure observed by TEM is accordant with that of the sample examined by XRD which

TABLE I. Activation energy values of K2YF5:Tb obtained by deconvolution method

Peak	Tm (°C)	E (eV)
1	127	0.70 ± 0.03
2	150	0.92 ± 0.02
3	178	0.98 ± 0.08
4	195	1.11 ± 0.05
5	205	1.22 ± 0.03
6	250	1.40 ± 0.07
7	310	1.45 ± 0.10
8	350	1.77 ± 0.11
9	380	2.17 ± 0.13
10	385	-

spectrum is shown in Fig 2. This figure shows the X-ray diffraction patterns of K_2YF_5:Tb Diffraction patterns suggested an orthorhombic structure of the analyzed powder samples.

The emission spectrum as well as the excitation spectrum of the studied material matched well with those characteristic of the terbium ions used as dopant. This fact is an indication that the terbium ions are the emission centers for all the peaks in the glow curve of K_2YF_5:Tb previously irradiated with beta particles or gamma radiation.

As an example, Fig. 3 shows the emission spectrum of K_2YF_5:Tb (0.99% Tb^{3+}) irradiated with ^{90}Sr/^{90}Y beta particles. It can be observed that this spectrum coincide with the emission spectrum characteristic of the Tm^{3+}ion, in which the transitions from $^5D_3$ and $^5D_4$ to ^7F_J states are observed.

TL emission spectrum is shown in Fig. 4. In this spectrum it can be observed an intense peak at 525 nm and other less intense peaks at 400, 470, 540, and 600 nm.

Figure 5 shows the glow curves obtained for samples of K_2YF_5:Tb doped at three different percentages of terbium, immediately after irradiation with beta particles at an absorbed dose of 300 mGy, and before submitting them to an isothermal decay treatment. This glow curve exhibits two peaks at about 260°C and 380°C respectively. In Fig. 6 it can be observed that after storing the irradiated samples at a temperature of 150°C during 40 minutes, the first peak (260°C) in their glow curves decreases meanwhile the second one (380°C) increases its intensity .

In Fig. 7 it can be observed more clearly the behavior of the glow curve of K_2YF_5:Tb (0.99% Tb) submitted to an isothermal decay at 150°C during different elapsed time intervals. It is observed that as the first peak decreases the second one increases.

Figure 8 shows the deconvolution of the glow curve of K_2YF_5:Tb (0.99% Tb) irradiated with beta particles made using the Glowfit v 1.1 software which discomposes the glow curve into ten individual peaks at 127, 150, 178, 195, 205, 250, 310, 350, 380 and 400°C. Values of activation energies obtained for K_2YF_5:Tb by deconvolution of the glow curve are shown in Table I.

4. Conclusions

Results obtained confirm that thermo-transferred thermoluminescence (TTTL) is occurring in K_2YF_5:Tb. However, the fact that TTTL has been observed persistently makes necessary further research to identify the mechanism causing it as well as the fact if it is only inherent to this material or could be presented by other materials.

The deconvoluted glow curve fits well to the experimental glow curve of K_2YF_5:Tb (FOM = 0.95) and the activation energy values obtained agree well with those obtained in previous work by other methods [2].

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