Applied Radiation and Isotopes xxx (xxxx) xxx–xxx

Contents lists available at ScienceDirect

Applied Radiation and Isotopes

journal homepage: www.elsevier.com/locate/apradiso

Dosimetric properties of α-Al_2O_3: Tm + PTFE phosphor

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HIGHLIGHTS

- The synthesis of alpha alumina phosphor was obtained by combustion of solutions method.
- The dosimetric characteristics of α-Al2O3: Tm + PTFE was studied.
- At 2 mol% concentration of Tm in the α-Al2O3:Tm matrix was highly sensitive to radiation.
- The kinetic parameters were determined by deconvolution of glow curve.
- This newly developed α-Al2O3: Tm + PTFE phosphor it is promising for medical physics radiation dosimetry.

ARTICLE INFO

Keywords:
Solid state detectors
Thermoluminescence
Dosimetry

ABSTRACT

A new α-Al_2O_3 doped with Tm^{3+} ions was obtained by combustion method; doped α-Al_2O_3 in the powder form was mixed with polytetrafluoroethylene resin (PTFE) to obtain dosimeters in pellet form, in order to be used as radiation dosimeter. The glow curve, linearity, lower detection limit, repeatability and fading, were studied. The kinetic parameters were determined by deconvolution method. Morphological characteristics were studied by low vacuum scanning electron microscopy and X-ray diffraction. Results showed that α-Al_2O_3 + PTFE obtained is a promising material to radiation dosimetry.

1. Introduction

Aluminum oxide (Al_2O_3) is considered one of the most important ceramic materials due to its multiple applications; it is used in the structural, electrical, optical and nuclear technologies. In the nuclear field, it is used as an inorganic ion exchange for the elimination of radioisotopes and the fixation of very high concentration of radioactive waste. However, due to its covalent nature, the pure material requires very high temperature (> 1973 K) to be obtained (Khalil et al., 2000).

α-Al_2O_3 doped with C, is one of the most recognized materials for ionizing radiation dosimetry (Azorín et al., 1993). And, it is commonly used as thermally stimulated light emission or optically stimulated emission dosimetric material (McKeever and Moscovitch, 2003). The importance of this material is that it can be used in conventional dosimetry, as well as for retrospective dosimetry, in cases of radiological accident [Holston et al. (2015)]. Some authors have obtained this material by using different preparation methods such as: melting of solids (Chen et al., 2013), laser ablation (Villarreal-Barajas et al., 2001), spray pyrolysis (Lopez-Estrada, 1994), evaporation (Azorín et al., 2002) and sol-gel (Rivera et al., 2010). In this paper, we propose a new method for obtaining α-Al_2O_3 phosphor based on solution combustion method (Kingsley et al., 1990).

2. Materials and methods

2.1. Synthesis

The parameter called "elemental stoichiometric coefficient" $\emptyset_e$ reflects the relationship between the intra molecular "fuel" and "oxidizer". Whereas $\emptyset_e = 1$ for stoichiometrically balanced compositions, they may differ substantially at other compositions. The deviations become more apparent when the fuel contains the "oxidizer" and the oxidizer contains "fuel" elements [Bakhman, 1968].

Calculation of $\emptyset_e$:

$$\emptyset_e = \frac{o_x}{r}$$

where o_x and r are integers representing the total composition of the oxidizing and the reducing elements respectively in the mixture. A mixture is fuel rich if $\emptyset_e < 1$, fuel lean if $\emptyset_e > 1$, and stoichiometrically

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https://doi.org/10.1016/j.apradiso.2018.02.012
Received 27 November 2017; Received in revised form 10 February 2018; Accepted 12 February 2018

balanced at $Ø_e = 1$. To calculate th_e specific formulas of the oxidizer and the fuel components are first calculated (Jain et al., 1981).

To prepare Al_2O_3, taking into account the stoichiometric ratio, we used $Al(NO_3)_3$ (oxidizer) and $CO(NH_2)_2$, (fuel) in a ratio 1:2.5, in this case $Ø_e = 0.686 < 1$, so the mixture is fuel rich. The dopant was incorporated in different concentrations; 0.1, 0.5 1.5 and 2 mol%. After eliminating the water presented in the mixture, it was submitted to an ignition process, which is carried out in an oven preheated at 773 K. The reaction began instantly producing an incandescent flame; then the product was foam-shaped at the end of the reaction. In order to homogenize the alpha phase of alumina, the material obtained was submitted to a sintering process in the temperature range between 1073 K and 1373 K, at different periods.

2.2. Characterization of the samples

X-ray powder diffraction (XRD) was conducted using a D8 Discover Bruker diffractometer with monochromatic Cu-Kα radiation (α = 1.5418 Å). The XRD pattern was obtained with a step size of 0.03077 in 2-theta, with a 2-theta range from 10° to 80°. Microstructural characterization was performed in a Jeol JSM-5900LV low vacuum scanning electron microscope (LVSEM) equipped with an Oxford energy dispersive X-ray spectroscope (EDS).

To facilitate the handling, the new TL material was formed in pellets mixing the TL powder with PTFE resin powder in the ratio 2:3. The discs obtained had average mass of 23 ± 2 mg, a diameter of 5 mm, and a thickness of 0.6 mm. After sintering, the pellets were irradiated with ^{60}Co gamma radiation by performing their dosimetric characterization. In addition, the kinetic parameters were determined by glow curve deconvolution method. TL readouts were made in a TLD System Harshaw model 4000 digitizing the TL signal by means of an interface-CR232. The readout parameters were the following: preheating temperature 323 K, heating rate 10 K/s, maximum temperature 673 K and acquisition time 30 s. All readouts were made in nitrogen atmosphere to avoid any spurious TL signals.

2.3. Thermal annealing

In order to eliminate the spurious TL signal generated during manipulation, due to previous irradiations or reutilization of α-Al_2O_3:Tm powder or pellets, an annealing thermal treatment was established. The optimal thermal annealing consisted in heating at 623 K, for 30 min. This procedure was considered as standard treatment for annealing of Al_2O_3:Tm.

2.4. Lower detection limit (LDL)

It is understood as the detection threshold at the minimum dose that can be measured with a dosimeter. To carry out this test, ten α-Al_2O_3:Tm +PTFE pellets were submitted to the standard thermal annealing and allowed to cool to room temperature and then proceeded to obtain the background reading (zero dose) and estimate the dose. The expression $tSD = LDL$ was used to evaluate LDL. Where t is the t-student for $n–1$ (n = number of dosimeters used in the test) degrees of freedom for a 95% confidence level, and SD is the standard deviation expressed as dose units (IEC, 2012).

2.5. Repeatability of measurements

Repeatability means, ideally, that the same TL response must always be obtained by irradiating the same dosimeter at the same dose for a certain number of times. The coefficient of variation (CV) of the readouts obtained in each of the cycles of thermal annealing, irradiation and readout of an optimal TL material for environmental and personal dosimetry, should not exceed ± 5% (Furetta, 2003). Therefore, the expression $100(SD/m) = CV$ was used to evaluate the CV. Here m is the average of the readouts and SD is the standard deviation.

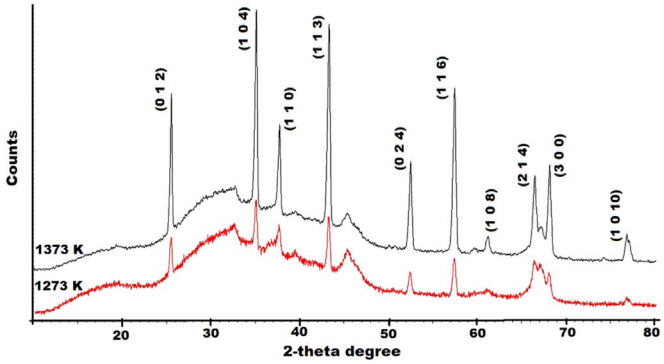

Fig. 1. X-ray diffractogram of Al_2O_3:Tm sintered at (a) 1273 K, (b) 1373 K.

2.6. TL response as function of radiation dose

For this test, the α-Al_2O_3:Tm + PTFE (2 mol%) dosimeters were submitted to the standard thermal annealing, and after that, were irradiated with ^{60}Co gamma radiation at different doses, in the range between 0.001 and 100 Gy. All the readings were taken after 24 h with constant nitrogen flow to avoid spurious signals.

2.7. Stability of TL signal (Fading)

This test consists in verifying the stability of the TL information, after the dosimeters have been irradiated at a known dose. In this test, a batch of α-Al_2O_3:Tm + PTFE (2 mol%) submitted to the standard thermal annealing, were allowed to cool to room temperature and irradiated with ^{60}Co gamma radiation at a dose of 10 Gy making reading after 24 h since irradiation at different time intervals along 45 days (IEC, 2012).

2.8. Kinetic parameters by deconvolution

The deconvolution technique is based on the sequential quadratic programing glow curve deconvolution (SQPGCD) developed at National Institute of Nuclear Research-Mexico (Lopez-Estrada, 1994). The deconvolution is based on the general Eq. (1):

$$I(T) = sn_0 \exp\left(-\frac{E}{kT}\right)\left[1 + \frac{s(b-1)}{\beta}\int_{T_0}^{T} \exp\left(-\frac{E}{kT'}\right)dT'\right]^{-\frac{b}{b-1}} \quad (1)$$

where:

where n_0 is the initial concentration of the trapped charges, s is the frequency factor (s^{-1}), b is the kinetics order, ranging from 1 to 2, β is the heating rate (K/s). E is the trap depth (eV), k is the Boltzmann's constant ($8.6{\cdot}10^{13}$ eV/K) and T is the absolute temperature (K). The Eq. (1), could be rewritten as follows (Chen, 1984):

$$I(T) = \frac{I_m \exp\left(\frac{E}{kT_m} - \frac{E}{kT}\right)}{\left[1 + \left(1-\frac{1}{b}\right)\left(\frac{E}{kT_m^2}\right)\int_{T_0}^{T} \exp\left(\frac{E}{kT_m} - \frac{E}{kT'}\right)dT'\right]^{\frac{b}{(b-1)}}} \quad (2)$$

From the last equation it is possible to obtain the expression (3), to input data for the deconvolution in function of the temperature of the peak at the maximum (T_M), and the TL intensity at the maximum of the peak (I_M) (Lopez-Estrada, 1994; González et al., 2013).

$$I(T) = \frac{I_m \exp(W(T - T_m))}{\left[\frac{1}{b} + \left(1-\frac{1}{b}\right)\exp(W(T - T_m))\right]^{\frac{b}{(b-1)}}} \quad (3)$$

where: $W = \frac{E}{kT_m^2}$