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PRELIMINARY RESULTS ON TL AND OSL ALUMINUM OXIDE DOSIMETERS DEVELOPED AT IPEN

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ABSTRACT

The aluminum oxide composes the modern TL and OSL radiation dosimeters. TL and OSL phenomena are related to chemical elements in the crystalline structure of α-Al_2O_3. The aim of this work was to develop materials based on aluminum oxide for use in TL and OSL dosimetry. The studies included the dosimetric properties of alumina samples obtained by electrofusion, adsorption and coprecipitation. Electrofused alumina commercially available as abrasive particles was used to produce the pellets by glass sintering. Adsorption and coprecipitation were the methods used to insert metal ions to alumina. The best results were achieved with electrofused alumina and Tm^{3+} doped Al_2O_3 pellets. The electrofused alumina-glass (EAG) pellets show TL and OSL signals and the TL curve has two peaks. Its minimum detectable radiation dose is 7.2 mGy and the linearity of TL response as function of dose is up to about 800 mGy. The α-Al_2O_3:Tm pellets produced by sintering at 1550 $^{\circ}$C presented a meaningful TL glow curve so that it is worth studying their properties and viability of use in dosimetry.

1. INTRODUCTION

The aluminum oxide aroused interest as radiation dosimeter since the announcement of anion-defective α-Al_2O_3:C crystals as a highly sensitive thermoluminescence (TL) material to detect ionizing radiation by Akselrod *et al.* [1]. Thereafter it was found that the TL signal of α-Al_2O_3:C was susceptible to light-induced fading and this material presented optically stimulated luminescence (OSL). α-Al_2O_3:C became a relevant material for OSL dosimetry applications [2].

The TL and OSL materials have defects in the crystal structure that can trap the excited electrons during irradiation. When the trapped electrons are released by thermal or optical stimulation and returning to the ground state, luminescence can occur. These materials are useful for radiation measurements if the luminous intensity or the light quantity can be correlated to radiation dose [2, 3].

Electrofused alumina is an impure aluminum oxide available as an abrasive commercial product. By the electrofusion process, the alumina is molten in an electric-arc furnace where heavy electric current is applied through the carbon rods. After the fusion, the melted alumina is left to cool. Once solidified, the alumina is broken, crushed and milled to the required grain

size. The composition depends on the raw material and on the additives used to modify the alumina properties [4].

Adsorption and coprecipitation were methods employed to insert metal ions into the structure of aluminum hydroxide or aluminum oxide. The adsorption method is based on the fact that aluminum hydroxide and activated alumina have high capacity to adsorb ions and molecules from aqueous solutions. These materials are composed by very fine particles and have a high specific surface area. The adsorbent capacity is attributed to the hydroxyl groups which present amphoteric behavior [5].

In the coprecipitation method, the aluminum hydroxide is simultaneously precipitated with the doping ion. This method was employed to obtain Hungarian Al_2O_3:Mg,Y dosimeter [3, 6] and it is suitable when the metal activator forms hydroxide with a small solubility product (K_{ps}). Some transition elements and lanthanides can be used as activators. This method is also adopted to synthesize other materials [7-9].

When aluminum hydroxide is heated it loses the mass and its characteristics are changed. The mass loss proceeds from the elimination of water molecules and hydroxyl groups. Several metastable forms of alumina are produced depending on the raw material and the temperature. Greek letters are used to identify these different forms. The stable crystalline α-Al_2O_3 occurs at around 1200 ºC [10].

This work aimed to study and develop materials based on aluminum oxide for use in TL and OSL dosimetry.

The developing procedures and measurements were performed at the Instituto de Pesquisas Energéticas e Nucleares - IPEN-CNEN/SP, at different Centers: Materials Science and Technology - CCTM, Radiation Technology - CTR and Radiation Metrology – GMR.

2. MATERIALS AND METHODS

The electrofused alumina-glass (EAG) pellets were produced by means of glass sintering. The white electrofused alumina supplied by Elfusa Geral de Eletrofusão Ltda. was sieved and grains with sizes from 75 to 180 μm were selected. A clean soda-lime glass bottle was crushed and grinded to powder with particles smaller than 75 μm. Both of the powders were mixed in the 1:1 ratio by weight, then shaped in the form of 6 mm diameter pellets and sintered at 732 $^{\circ}$C in a microwave muffle furnace (MAS-7000 ^{TM} model, CEM Corporation).

The raw materials used as adsorbents in the adsorption method were the commercially available activated alumina (Merck S.A.) or aluminum hydroxide (Vetec Química Fina Ltda.). Cu^{2+} and Mn^{2+} solutions were prepared by dissolving the respective salts in water in order to reach the dopant concentration of 0.1 mol % in Al_2O_3 matrix. The alumina powder was introduced to the solution, stirred and dried by heat. The doped powder or the cold pressed pellets were heated at temperatures higher than 1200 ºC and TL was measured.

In order to insert the dopant ion to alumina by the coprecipitation method a blend solution composed of thulium ion and aluminum chloride salt ($AlCl_3.6H_2O$ – Vetec Química Fina Ltda.) was prepared and reacted with the ammonium hydroxide solution in excess. The gelatinous hydroxide was separated from secondary products by filtering and washed. The

doped powder was dried, calcined and pressed. The cold pressed pellets were sintered at 1550 °C in the Lindberg Blue furnace at the CCTM laboratory.

The scheme of the doping process by coprecipitation is shown in Fig.1. A^{n+} represents the metal ion activator.

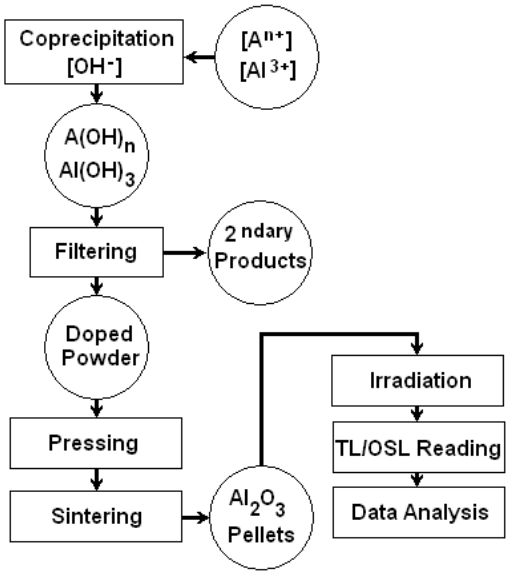

Figure 1. Scheme of the doping process by coprecipitation to obtain Al_2O_3 pellets.

For checking the thermoluminescence, the alumina pellets were irradiated in a pneumatic TLD dosimeter irradiator (Mark IV-G model, J. L. Shepherd and Associates) with ^{137}Cs sealed radiation source. Four sets of electrofused alumina pellets were submitted to 1, 2, 5 and 10 Gy in a Panoramic type irradiator with ^{60}Co radiation source located at CTR. Other pellet samples were irradiated from 100 to 800 mGy at GMR, using another ^{60}Co gamma source.

TL measurements were performed at the GMR laboratory using two Harshaw TL readers 2000 model, one connected to a computer via data acquisition card. The complementary TL and OSL measures were obtained by means of the RISØ TL/OSL reader, TL/OSL-DA-20 model.

3. RESULTS

The electrofused alumina and thulium doped alumina pellets presented the best results in this study. The characteristics of the two types of pellets are summarized in Table 1. EAG refers to the electrofused alumina pellets obtained by glass sintering and α-Al_2O_3:Tm is the thulium doped alumina pellets.

Table 1. Characteristics of EAG and α-Al_2O_3:Tm pellets

Characteristics	EAG	α-Al_2O_3:Tm
Composition	Glass and Al_2O_3	Al_2O_3
Weight (g)	0.051 ± 0.001	0.049 ± 0.001
Diameter (mm)	5.55 ± 0.05	4.20 ± 0.05
Thickness (mm)	1.10 ± 0.05	1.20 ± 0.05
Sintering temperature (ºC)	732	1550
Sensitivity to ^{60}Co γ-rays (nC.mGy^{-1})	0.76	---
Minimum dose detectable (mGy)	7.2	---
Peak temperature (ºC)	175 and 290	208
OSL signal	yes	yes

The pellets obtained by doping aluminum oxide or aluminum hydroxide with Cu^{+2} and Mn^{+2} ions were submitted to irradiation however they did not present expected TL signal. The results were not presented in this paper.

The sample doped with Tm^{3+} by the coprecipitation method presented meaningful TL response. The typical TL glow curve of α-Al_2O_3:Tm pellets presents the dosimetric peak at 208 °C as was shown in Fig. 2. This curve was obtained at 5 °C s^{-1} after β irradiation with RISØ TL/OSL reader.

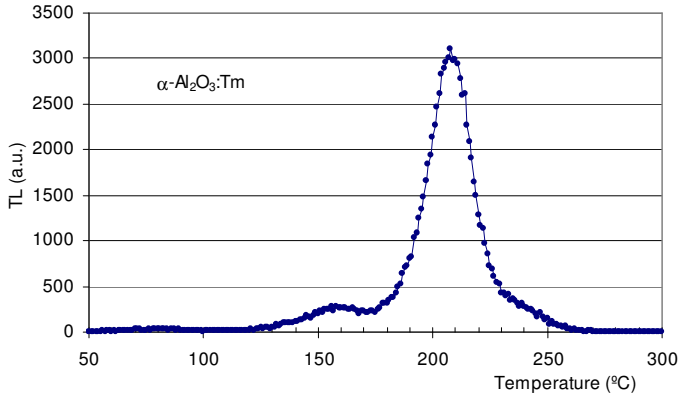

Figure 2. TL glow curve of α-Al_2O_3:Tm pellets.

The TL glow curve of electrofused alumina powder presents two peaks, one at around 130 ºC and the second at 230 ºC, at 10 ºC min^{-1} heating rate. When the powder was transformed into pellets by glass sintering, the peaks were shifted to a higher temperature. The first peak at 175 ºC subjected to thermal fading can be excluded by post-irradiation heat treatment (HT) (Fig.3).The second dosimetric peak appears at about 290 °C.

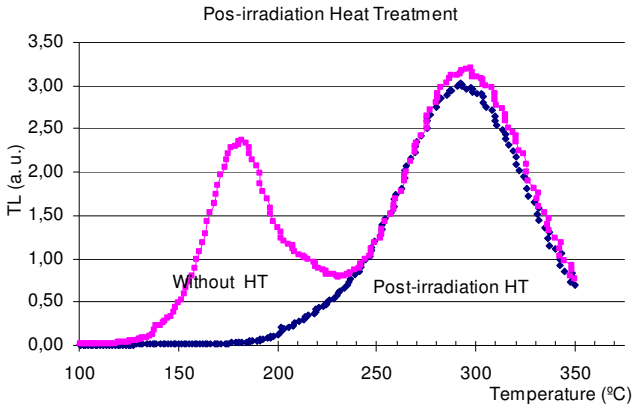

Figure 3. Effect of post-irradiation heat treatment to exclude the first peak.

The TL response in function of ^{60}Co gamma radiation dose of electrofused alumina pellets is shown in Fig. 4. The linearity is observed till around 800 mGy. The TL readings of pellets irradiated with 1, 2, 5 and 10 Gy are outside this range and have a supralinear tendency. These data were obtained with Harshaw TLD reader 2000 model, integrating the TL readings from 50 to 350 °C at 10 °C.s^{-1}.

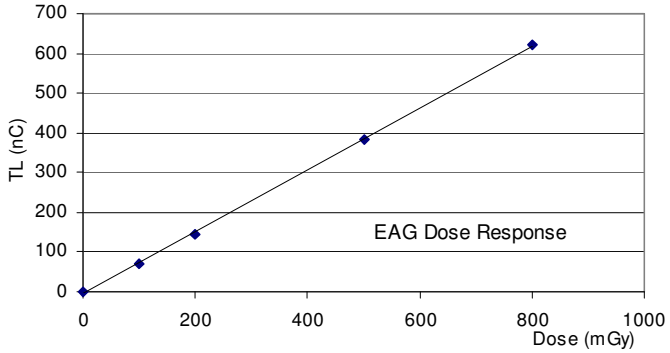

Figure 4. TL response in function of ^{60}Co gamma radiation dose of electrofused alumina pellets.

4. DISCUSSION AND CONCLUSIONS

The electrofused alumina was sintered with soda-lime glass at around 732 ºC which is much lower than the temperature for alumina sintering. Glass provides a transparent medium for that the light emitted by TL or OSL is more effectively detected by a photomultiplier tube. Despite the low softening point [10], the glass allows to be heated to temperatures higher than that permissible temperature for plastic polymers.

The EAG pellets are promising dosimeters but other characteristics as thermal fading, light sensitivity and OSL signal should be analysed.

The Cu^{2+} and Mn^{2+} doped pellets obtained by means of adsorption method did not show expected TL signals. The causes of the poor TL signal were probably due to the chemical form of the dopant and pH of the solution used in adsorption.

The TL glow curve of α-Al_2O_3:Tm pellets prepared by coprecipitation and sintering has shown similar behavior of the alumina obtained by combustion synthesis reported by Barros *et al*. [11]. Furthermore considering that the dosimetric peak of α-Al_2O_3:Tm pellets is predominant at around 200 ^{o}C, these findings point to continuing studies of its properties and the viability of use in dosimetry.

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