Thermoluminescent response of CaSO₄: Eu,Ag detectors in ⁹⁰Sr/⁹⁰Y beta radiation beam

Ivón Oramas Polo ^{1,2}, Danilo Oliveira Junot¹, Patricia Nicolucci², Linda Caldas¹

¹ Instituto de Pesquisas Energéticas e Nucleares, Comissão Nacional de Energia Nuclear (IPEN-CNEN/SP). Av. Prof. Lineu Prestes, 2242 - 05508-000, São Paulo, SP, Brazil.

² Departamento de Física. Faculdade de Filosofia, Ciências e Letras. Ribeirão Preto. Universidade de São Paulo. Av. Bandei rantes, 3900, 14040-901, Ribeirão Preto, SP, Brazil.

ivonoramas67@gmail.com, dan.junot@gmail.com, nicol@usp.br, lcaldas@ipen.br

Abstract

The results of the thermoluminescent (TL) response of the CaSO₄:Eu,Ag detectors in the 90Sr/90Y beams of the Beta Secondary Standard 2 system of the Calibration Laboratory of IPEN (LCI/IPEN) are presented. The thermoluminescent glow curves were obtained from doses between 30 mGy and 500 mGy. The detectors showed a good reproducibility of the thermoluminescent response. The calculated calibration factor was $(1.166 \pm 0.024) \times 10^{-3}$ mGy/a.u. and the factor determined by linear fitting was $(1.120 \pm 0.014) \times 10^{-3}$ mGy/a.u., showing a difference of only 3.9%. The lower limit of detection was (4.96 ± 0.06) mGy. The detectors presented an appropriate sensitivity for ⁹⁰Sr/⁹⁰Y beta radiation. Pre-liminary results showed suitable dosimetric characteristics for the establishment of a transfer system for beta radiation dosimetry of ⁹⁰Sr/⁹⁰Y beams.

Key words: thermoluminescent dosemeters; response functions; calcium sulfates; strontium 90; yttrium 90; europium; sensitivity.

Respuesta termoluminiscente de detectores de CaSO₄: Eu, Ag en un haz de radiación beta de ⁹⁰Sr/⁹⁰Y

Resumen

Son presentados los resultados de la respuesta termoluminiscente de los detectores de CaSO₄:Eu,Ag en haces de ⁹⁰Sr/⁹⁰Y del sistema Beta Secondary Standard 2 del Laboratorio de Calibración del Instituto de Pesquisas Energéticas y Nucleares (LCI/IPEN). Fueron obtenidas las curvas de emisión luminiscente con dosis entre 30 mGy y 500 mGy. Los detectores mostraron una buena reproducibilidad de la respuesta termoluminiscente. El factor de calibración calculado fue (1.166 \pm 0.024) x10⁻³ mGy/a.u. y el factor determinado mediante el ajuste lineal fue (1.120 \pm 0.014) x10⁻³ mGy/a.u., mostrando una diferencia de solamente 3.9%. El límite inferior de detección fue (4.96 \pm 0.06) mGy. Los detectores presentaron una sensibilidad apropiada para la radiación beta de 90Sr/90Y. Los resultados para dosimetría de radiación beta en haces de ⁹⁰Sr/⁹⁰Y.

Palabras clave: partículas beta; dosímetros termoluminiscentes; funciones de respuesta; sulfatos de cálcio; estroncio 90; itrio 90, europio, sensibilidade.

Introduction

In the nuclear energy industry, in many situations, the skin is subjected to low energy beta radiation. Therefore, the dose may be limited [1]. The extrapolation chamber is the primary instrument for measurements in beta radiation beams [2]. These measurements should be taken under laboratory reference conditions of temperature, pressure and humidity. In addition, the extrapolation chamber must be handled with extreme care, because it is very heavy and its entrance window is very delicate. Thermoluminescence may be an alternative method for dosimetry to constitute a beta-radiation transfer system. Thermoluminescence has already been used in applications of beta radiation [1-6]. This kind of radiation has a low penetration power; therefore, the choice of the dosimeter material is very important [1, 7-10]. The dosimeter response depends on the energy range of the beta radiation. However, dosimeters intended to measure doses of this radiation should be as thin as possible compared to the ranges of all the electron energies of interest [6, 11].

For the calibration of beta radiation fields, thin thermoluminescent (TL) dosimeters of materials with low atomic number, such as LiF, $Li_2B_4O_3$, $Mg_2B_4O_7$, Al_2O_3 , among others, may be used [12]. Some types of fine detectors have been prepared with TL high-sensitivity phosphors such as $Mg_2B_4O_7$:Dy, $CaSO_4$:Dy, AI_2O_3 :C and LiF:Mg,Cu,P. For beta radiation, the high sensitivity of the phosphor is very important. In the case of low energy beta radiation, for example ²⁰⁴TI (Emax = 0.77 MeV), the TL sensitivity decreases with increasing dosemeter thickness much faster as compared to that of high-energy beta rays, for example ³²P (Emax = 1.71 MeV) [1].

 $CaSO_4$ doped with Rare Earths (RE) has been widely studied in some works as a TL material [1,13-14]. Calcium sulfate doped with dysprosium (CaSO_4:Dy) is a material already well studied in beta radiation beams [1,4, 6, 10, 15]. Calcium sulfate doped with europium (CaSO_4:Eu) presents a suitable TL response [16]. The addition of silver to CaSO_4:Eu allows the increase of the TL intensity [17-18].

The objective of the present work was to perform a TL response analysis of $CaSO_4$:Eu,Ag detectors in ⁹⁰Sr/⁹⁰Y beams, for the establishment of a transfer system or alternative/complementary method for beta radiation dosimetry.

Materials and methods

For the dosimetric characterization of the detectors, the ⁹⁰Sr/⁹⁰Y source of the Beta Secondary Standard BSS2 of the Calibration Laboratory (LCI) of the IPEN/ CNEN in Brazil was used. The main characteristics of this source are the following: 460 MBq of nominal activity; 0.8 MeV of average beta energy and 10,483 days of half-life. The calibration date was 11/19/2004. The calibration distance was 11 cm without the use of the beam flattening filter [19].

The dosimetric system consists of the detectors, the TL reader, the thermal treatment system and auxiliary materials that allow performing the luminescent dosimetry.

The CaSO₄:Eu:Ag detectors were produced by stages in the Laboratory of Medical Physics (LFM) of the Department of Physics (DFI) of the Federal University of Sergipe (UFS) and in the Laboratory of Dosimetric Materials (LMD) of the Radiation Metrology Center (CMR) of IPEN. The crystals of CaSO₄:Eu:Ag were produced by a route based on the mixture of Calcium Carbonate (CaCO₃), Sulfuric Acid (H₂SO₄), Europium Oxide (Eu₂O₃) and silver metal particles Ag0. The dopants were incorporated in the proportion of 0.1 mol %. For the production of the pellets, powdered Polytetrafluoroethylene (Teflon) was added in the proportion of 1:1 for the mass of the phosphor and the mass of Teflon. The detectors have 6 mm in diameter, 1 mm in thickness and 40 mg in mass [17].

For the TL measurements, the RISÖ TL/OSL-DA20 system was used. The system allows up to 48 samples to be individually heated at any temperature up to 700 °C. The measurements were performed in a vacuum chamber. The emitted luminescence was measured by a light detection system, composed of a photomultiplier valve and suitable detection filters. TL measurements were

performed under N2 atmosphere. The luminescence was detected by a bialkali EMI 9235QB photomultiplier tube (PMT) which has a maximum detection efficiency between 200 nm and 400 nm. The 7 mm band pass filter Hoya U-340 (transmission band 250–390 nm Full width at half maximum (FWHM)) was utilized [20]. The heating rate was 10°C/s, and the maximum temperature was 350°C. Figure 1 shows the RISÖ TL/OSL-DA20 measuring system of LCI/IPEN.



Figure 1. RISÖ TL/OSL-DA20 measuring system of the Calibration Laboratory IPEN/ CNEN/SP.

The thermal treatment was performed in a furnace manufactured by the Institute of Radioprotection and Dosimetry (IRD), CNEN, in Rio de Janeiro. The samples were thermally treated at 400 °C for 1h under air atmosphere.

Figure 2 shows the holder for irradiation of the detectors and the support for performing the thermal treatment. The holder for the irradiation is made of polymethylmethacrylate (PMMA), and it has the following dimensions: 110 mm in width and length, and 18 mm in depth. It allows the irradiation of 25 detectors. The holder cover is a 0.015 mm Hostaphan sheet. The support for the heat treatment is a circular aluminum plate.



Figure 2. Circular aluminum plate for thermal treatment (left). PMMA holder and cover for irradiation of detectors (right).

Results and discussion

For the evaluation of the response reproducibility of the detectors, thirty detectors were used. They were irradiated with a dose of 1 Gy of the RISÖ system ⁹⁰Sr/⁹⁰Y source. The TL response was considered as the integral under the whole glow curve. After irradiation, the

TL reading, the thermal treatment and the reading of the background were performed. This procedure was repeated for 5 cycles. Twelve detectors of this batch were chosen for the study of TL response. Table 1 shows the mean values of the TL response, the standard deviation and the coefficient of variation (C.V.) of the chosen detectors.

Table 1	. Reproducibility	of CaSO ₄ :Eu:Ag	TL detectors
---------	-------------------	-----------------------------	--------------

Detector number	Mean values of the TL response (a.u.)	Standard devia- tion (a.u.)	Coefficient of variation (%)
1	1.38 x10 ⁶	2.08 x10 ⁴	1.5
2	1.42 x10 ⁶	1.13 x10 ⁴	0.8
3	1.43 x10 ⁶	1.83 x10 ⁴	1.3
4	1.38 x10 ⁶	1.31 x10⁴	0.9
5	1.45 x10 ⁶	2.08 x10 ⁴	1.4
6	1.42 x10 ⁶	1.88 x10 ⁴	1.3
7	1.37 x10 ⁶	1.92 x10 ⁴	1.4
8	1.39 x10 ⁶	2.58 x10 ⁴	1.8
9	1.40 x10 ⁶	2.97 x10⁴	2.1
10	1.35 x10 ⁶	2.33 x10 ⁴	1.7
11	1.38 x10 <u>6</u>	2.04 x10 ⁴	1.5
12	1.42 x10 ⁶	1.29 x10 ⁴	0.9

In order to consider that the detectors have a good response reproducibility, the recommended coefficient of variation should be less than 5% [21]. The results of Table 1 are in agreement with these recommendations.

Figure 3 shows the TL glow curves of $CaSO_4$:Eu:Ag detectors. Detector 1 was chosen arbitrarily for the representation. The dose range considered was 30 mGy up to 500 mGy.



Figure 3. TL glow curves of the CaSO₄:Eu:Ag detectors in BSS2 90 Sr/ 90 Y radiation beam, for doses of 30 mGy up to 500 mGy.

To obtain the TL response curves as a function of the absorbed dose of the $CaSO_4$:Eu:Ag detectors, they were irradiated in the BSS2 ⁹⁰Sr/⁹0Y radiation beam with doses of 30 mGy up to 500 mGy. The dose range for the study was established, considering the irradiation times of the BSS2 system [19]. Figure 4 shows the dose-response curve of the CaSO₄:Eu:Ag detectors in the range of the aforementioned doses.



Figure 4. TL dose-response curve of CaSO4:Eu:Ag detectors in BSS2 90Sr/90Y radiation beam. The maximum uncertainty was 2.6%.

The curve was fitted linearly by a computational program, and the R2 correlation coefficient was 0.99828. The detectors showed a linear behavior in the tested dose range. The calibration factor was determined by linear fitting, and it was determined by Equation 1:

$$F_c = 1/c \tag{1}$$

where c is the slope of the fitted line.

The calibration factor can also be determined as the ratio between the absorbed dose and the mean value of the TL measurements at each point of the calibration curve [21].

The determined calibration factor was $(1.166 \pm 0.024) \times 10^{-3}$ mGy/a.u. and the factor determined by linear fit was $(1.120 \pm 0.014) \times 10^{-3}$ mGy/a.u. The calibration factors obtained by both methods are in good agreement. The difference between them was only 3.9%.

The lower detection limit (LDL) is the minimum dose that can be detected by luminescent material [20]. The LDL is important in low dose measurements where the dosimeter signal is almost equal to the background signal. The LDL was determined by Equation 2:

$$LDL = 3^* \sigma_{BKG}^* F_c \tag{2}$$

where $\sigma_{\mbox{\tiny BKG}}$ is the standard deviation of the zero dose reading [21].

Table 2 shows the values of the material TL sensitivity for each detector. For this test, a dose of 50 mGy was chosen.

4 9 10 10 10 10 10 10 10 10 10 10 10 10 10					
Detector number	TL sensivity (Counts/mGy.g)	Detector number	TL sensivity (Counts/mGy.g)		
1	$(2.10 \pm 0.03) \ x10^4$	7	$(2.07 \pm 0.03) \ x10^4$		
2	$(2.22 \pm 0.03) \text{ x}10^4$	8	$(1.80 \pm 0.03) \ x10^4$		
3	$(1.99 \pm 0.03) \times 10^4$	9	$(2.02 \pm 0.03) \ x10^4$		
4	$(1.99 \pm 0.03) \times 10^4$	10	$(2.01 \pm 0.03) \times 10^4$		
5	$(2.15 \pm 0.03) \text{ x}10^4$	11	$(1.92 \pm 0.03) \ x10^4$		
6	$(2.15 \pm 0.03) \times 10^4$	12	$(1.84 \pm 0.03) \times 10^4$		

Table 2. Intrinsic sensitivity of CaSO₄:Eu:Ag detectors for TL response.

The detectors show an appropriate sensitivity for ⁹⁰Sr/⁹⁰Y beta radiation, suggesting a potential use of these detectors for beta dosimetry with also the other BSS2 radiation sources (⁸⁵Kr and ¹⁴⁷Pm).

Conclusions

The analysis of the TL response of the CaSO₄:Eu:Ag detectors in 90 Sr/ 90 Y radiation beams was performed. The TL glow curves for doses between 30 mGy and 500 mGy were obtained. The reproducibility of the response, the calibration factor, the lower limit of detection and the intrinsic sensitivity of the detectors were determined.

The preliminary results of the tests carried out show suitable dosimetric characteristics for the establishment of a transfer system or alternative/ complementary method for dosimetry of beta radiation.

Acknowledgements

The authors thank the partial financial support of the Brazilian funding agencies CNPq (Process: 142297/2015-1 fellowship of I. O. Polo, and 301335/2016-8), and CA-PES (Project: 554/2018).

References

- LAKSHMANAN A. Development and application of solid forms of CaSO4:Dy thermoluminescent dosemeters in radiation protection dosimetry — A review. Radiat. Prot. Dosim. 2018; 181(2): 57-99.
- [2]. BÖHM J. The national primary standard of the PTB for realizing the unit of the absorbed dose rate to tissue for beta radiation. Braunschweig, Germany: Physikalisch-Technische Bundesanstalt, PTB-Dos-13. 1986.
- [3]. CALDAS LVE. Some calibration and dosimetry methods for beta radiation. Ph.D. Thesis. Physics Institute/São Paulo University (In Portuguese). 1980.
- [4]. CONTENO G, MALISAN MR, PADOVANI R. Response of thermoluminescence dosimeters to beta radiation and skin dose measurement. Phys. Med. Biol. 1984; 29: 661-678.
- [5]. CROSS WG. Empirical expressions for beta ray point source dose distributions. Radiat. Prot. Dosim. 1997; 69: 85-96.
- [6]. KUMAR M, RAKESH RB, SNEHA C, RATNA P, et. al. Beta response of CaSO4:Dy based thermoluminescent dosimeter badge and its angular dependence studies for personnel monitoring applications. Radiat. Prot. Environ. 2016; 39: 132-137.

- [7]. INTERNATIONAL ATOMIC ENERGY AGENCY (IAEA). Calibration of Photon and Beta Ray Sources used in Brachytherapy. IAEA-TECDOC 1274. Vienna: IAEA, 2002.
- [8]. International Commission on Radiation Units and Measurements (ICRU). Dosimetry of beta rays and low-energy photons for brachytherapy with sealed sources. J ICRU. 2004; 4(2).
- OLIVEIRA ML, CALDAS LVE. Performance of thin CaSO₄:Dy pellets for calibration of a ⁹⁰Sr+⁹⁰Y source. Nucl. Instr. Meth. Phys. Res. 2007; 580: 293-295.
- [10]. ANTONIO PL, OLIVEIRA ML, CALDAS LVE. Thin CaSO₄: Dythermoluminescent dosimeters for calibration of ⁹⁰Sr+⁹⁰Y applicators. Appl. Radiat. Isot. 2012; 70: 790-793.
- [11]. KUMAR M, DHABEKAR B, MENON S, BAKSHI S, et. al. Beta response of LiMgPO₄:Tb,B based OSL discs for personnel monitoring applications. Radiat. Prot. Dosim. 2013: 155(4): 410-417.
- [12]. International Commission on Radiation Units and Measurements (ICRU). Dosimetry of External Beta Rays for Radiation Protection. ICRU Report 56. Bethesda: ICRU, 1997.
- [13]. YAMASHITA T, NADA N, ONISHI H, KITAMURA S. Calcium sulphate phosphor activated by thulium or dysprosium for thermoluminescence dosimetry. Health Phys. 1971; 21: 295-300.
- [14]. KÁSA I, CHOBOLA R, MELL P, SZAKÁCS S, et. al. Preparation and investigation of thermoluminescence properties of CaSO₄:Tm,Cu. Radiat. Prot. Dosim. 2007; 123: 32-35.
- [15]. CAMPOS LL, LIMA MF. Thermoluminescent CaSO₄:Dy teflon pellets for beta radiation detection. Radiat. Prot. Dosim. 1987; 18 (2): 95-97.
- [16]. DENG W, GOLDYS EM. Plasmonic approach to enhanced fluorescence for applications in biotechnology and the life sciences. Langmuir 2012; 28 (27):10152-10163.
- [17] JUNOT DO, VASCONCELOS DF, CHAGAS MAP, et. al. Silver addition in CaSO₄:Eu, TL and TSEE properties. Radiat. Meas. 2011; 46 (12): 1500-1502.
- [18]. JUNOT DO, COUTO DOS SANTOS MA, ANTONIO PL, CAL-DAS LVE, et. al. Feasibility study of CaSO₄:Eu, CaSO₄:Eu,Ag and CaSO₄:Eu,Ag(NP) as thermoluminescent dosimeters. Radiat. Meas. 2014; 71: 99-103.
- [19]. BSS2. Beta secondary standard 2. Operation manual. Isotrak. QSA Global GmbH. 2005.
- [20]. DTU Nutech. Guide to "The Risö TL/OSL Reader". Denmark: DTU Nutech. 2015.
- [21]. FURETTA C. Questions and answers on thermoluminescence (TL) and optically stimulated luminescence (OSL). London: World Scientific Publishing, 2008.

Recibido: 22 de abril de 2019 Aceptado: 14 de octubre de 2020