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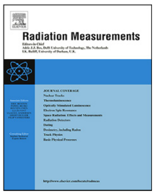

Technical report

Preliminary TL Studies of K_2GdF_5:Dy^{3+} exposed to photon and neutron radiation fields

E.C. Silva [a], N.M. Khaidukov [b], E.C. Vilela [c], L.O. Faria [d,*]

[a] Depto. de Engenharia Nuclear (DEN/UFMG-MG), Av. Antônio Carlos 6627, 31270-970 Belo Horizonte, MG, Brazil
[b] Kurnakov Institute of General and Inorganic Chemistry, RAS, Lenisnskii Prospect 31, 119991 Moscow, Russia
[c] Centro Regional de Ciências Nucleares do Nordeste, Av. Prof. Luiz Freire, 200, 50740-540 Recife, PE, Brazil
[d] Centro de Desenvolvimento da Tecnologia Nuclear, Av. Antônio Carlos 6627, C.P. 941, 30270-901 Belo Horizonte, MG, Brazil

HIGHLIGHTS

- Thermoluminescence responses of K_2GdF_5:Dy crystals exposed to X, gamma and neutron radiation fields have been investigated.
- For radiation doses ranging from 0.12 to 242 mSv, the TL output from $K_2Gd_{0.95}Dy_{0.05}F_5$ has a linear behavior and low fading.
- TL sensitivity for thermal neutrons is 18 lower than the TLD-600.
- Otherwise, for fast neutrons, K_2GdF_5:Dy crystals show TL output 5 times higher.

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ABSTRACT

Results of the investigation concerning thermoluminescence (TL) responses to X, gamma and neutron radiation fields for crystals of complex fluoride K_2GdF_5 undoped and doped with varying concentrations of Dy^{3+} ions are presented. Crystals doped with 5.0 at% Dy^{3+} have shown the most efficient TL response, with a linear response to doses for all the radiation fields. In the X rays range, the maximum TL response has been found to be 15 times more than the response for gamma. The fast and thermal neutron TL outputs were evaluated for $K_2Gd_{0.95}Y_{0.05}F_5$ and the contribution of the gamma component in the TL curve was estimated.

1. Introduction

Thermoluminescent materials such as LiF, $CaSO_4$ and CaF_2 doped with different impurities are broadly utilized in environmental monitoring, personal and clinical dosimetry (McKeever et al., 1995).

Recently, K_2YF_5 crystals singly doped with rare earth ions (RE), e.g. Ce^{3+}, Tb^{3+}, Dy^{3+} or Tm^{3+}, have been shown to be attractive TL materials for detection and discrimination of different types of radiation (Kui et al., 2006; Azorın-Nieto et al., 2007). The investigation of such materials containing high concentrations of optically active RE ions is a promising direction for the developing of novel TL phosphors by taking into account that RE ions can efficiently capture electrons and/or holes and can be simultaneously recombination and luminescent centers (Kui et al., 2006; Krumpel et al., 2008). In this context, it should be also noted that K_2YF_5 crystals singly doped with 10.0 at.% $Tb^{3+}$and 1.0 at.% Dy^{3+} have high TL sensitivity to photon radiation fields with energies in the range of X and gamma rays (Faria et al., 2004; McLean et al., 2004; Silva et al., 2007).

On the other hand, by taking into account that Tb^{3+} doped K_2GdF_5 fluorides are phosphors that show good photo-stimulated and thermally stimulated luminescence after they have been exposed to ionizing radiation (Azorın-Nieto et al., 2007; Hanh et al., 2010), one can expect that K_2GdF_5 crystals doped with Dy^{3+} ions could also show good thermoluminescent response (Krumpel et al., 2008). Also it should be noted that Gd has the highest thermal neutron cross section of any natural element, namely 49700 b and accordingly gadolinium compounds are used as neutron scintillators and converters (Ryzhikov et al., 2002, Yukihara et al., 2008).

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* Corresponding author. Tel.: +55 31 30693128; fax: +55 31 30693164.
E-mail addresses: farialo@cdtn.br, farialo@pq.cnpq.br (L.O. Faria).

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However, there are no TL detectors based on gadolinium compounds in particular. In other words, designing and testing new complex fluorides of alkali and gadolinium could give some promising direction for developing TL detectors attractive for neutron dosimetry.

Within this research, the systematic investigation concerning the thermoluminescence responses of K_2GdF_5 crystals doped with 0.2, 1.0, 5.0 and 10.0 at.% Dy^{3+} as well as undoped K_2GdF_5 crystals to photon and neutron fields have been performed from the viewpoint of possible application as TL materials for X-ray, gamma or neutron detectors as well as detectors able to separate dose components in mixed radiation fields.

2. Experimental

K_2GdF_5 single crystals doped with 0.2, 1.0, 5.0 and 10.0 at.% Dy^{3+} ions as well as undoped K_2GdF_5 crystals up to 1 cm^3 in size were grown by a direct temperature-gradient method as a result of the reaction of potassium fluoride aqueous solutions with appropriate mixtures of 99.99% pure rare earth oxides under hydrothermal conditions (Yoshimura, 1998). Polished crystalline samples with thickness of about 1 mm were utilized for the TL measurements. In addition LiF:Mg,Ti (TLD-100, TLD-600 and TLD-700) chips manufactured by Harshaw-Bicron Chemical Company were used in order to check the delivered neutron and photon doses and to obtain the relative TL sensitivities of synthesized K_2GdF_5 crystals. For evaluation of residual thermal neutrons and gamma components in the fast neutron irradiation, and residual fast neutrons and gamma component in the Thermal Neutron Irradiator, two pairs of previously calibrated TLD-600 and TLD-700, one of them covered by a thin Cd foil, were put together with the samples under investigation.

The examined samples were exposed at room temperature (RT) to gamma rays with photon energy of 662 keV from a ^{137}Cs gamma source, with delivered personal equivalent doses Hp(10) (in Sv) measured by ionization chambers calibrated at the secondary standard laboratory LCS-IRD/CNEN, which in turn is traceable to an International Atomic Energy Agency (IAEA) primary standard laboratory, as well as to X-rays with effective energies of 33.3, 41.1 and 52.5 keV. Here, the effective energy is defined as the energy of monoenergetic photons with the same value of HVT ($K_{c,\mathrm{air}}$), i.e., the half value thickness as a function of initial air collision *Kerma*, as the polyenergetic photon beam. The 41.1 and 52.5 keV energies were the W60 and W80 spectra, respectively, as defined by International Organization for Standardization (ISO) 4037-1 series (ISO 4037-1, 1996).

Neutron irradiations were performed using an Am–Be source with an emission rate of 4.46×10^8 n/s. The radiation monitor used to evaluate neutron doses at the point of interest was calibrated in terms of *Effective Dose* (*E*), formerly the effective dose equivalent. A conversion factor equal to 1.0 have been used to convert *Effective Dose* into *Hp*(10), as recommended by ICRP 60 (ICRP 60, 1991). The neutron source and the Thermal Neutron Irradiator were calibrated at Lab. Neutrons – Instituto de Radioproteção e Dosimetria (IRD/CNEN) which is the owner of the Brazilian Standard of Neutron Fluency, traceable to the Bureau International des Poids et Mesures (BIPM-France). The irradiations with thermal neutrons were performed inside of an apparatus with four irradiation channels made of aluminum-cylindrical tubes filled with paraffin. The Am–Be source was positioned outside of this apparatus. The thickness of the Aluminum walls was 5.0 mm. The distance between the neutron source and the TL samples inside the thermal irradiation channel was 17.0 cm. For fast neutron irradiation another setup was used where the samples were covered with a Cd foil and placed in a 30 cm × 30 cm × 15 cm poly(metyl methacrylate) (PMMA) phantom, with a source-sample distance of 75 cm, as recommended by ISO 10647 (ISO 10647, 1996).

The measurements of TL glow curves were performed with a Harshaw-Bicron 3500 TLD reader operating with a linear temperature profile over the range from 50 up to 300 °C in the resistive mode by using a heating rate of 10 °C/s and reading cycles of 35 s.

The photo multiplier tube (PMT) is designed to better detect photons with wavelengths ranging from 380 to 700 nm and it has an optical filter to shield photons with wavelength below 360 nm. Samples were annealed during secondary readings and the residual signal (reading 2/reading 1) was 0.01%. The samples were weighed and all data were normalized to sample mass. In this paper, TL output is defined as the amount of electric charge generated at the photomultiplier tube, which is proportional to area under the glow curve, when the light emitted by the heated sample reaches its surface, during a reading cycle.

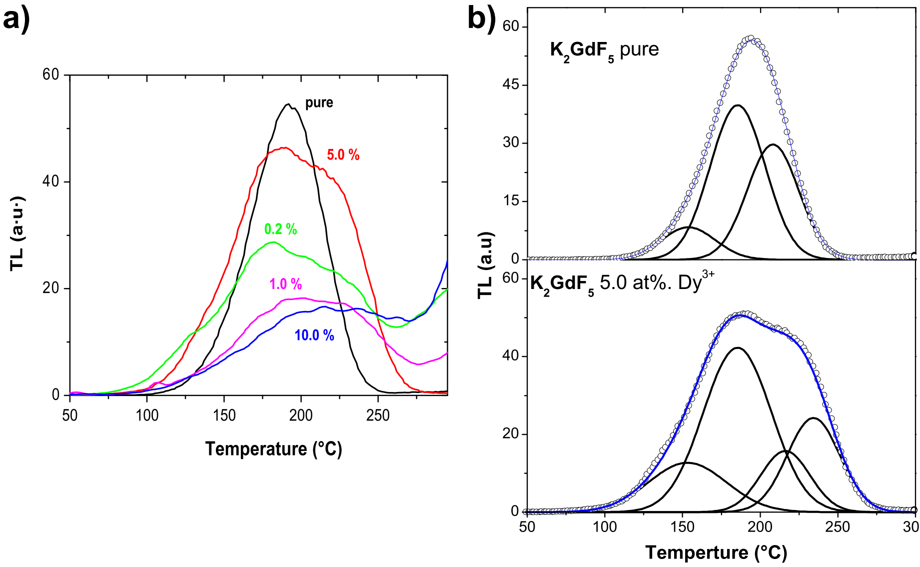

Fig. 1. (a) Glow curves for K_2GdF_5 crystals doped with 0.2, 1.0, 5.0 and 10.0 at.% Dy^{3+} ions as well as undoped K_2GdF_5, after exposing to 12.1 mSv of gamma radiation and (b) deconvolution of the TL glow curves of K_2GdF_5 and $K_2Gd_{0.95}Dy_{0.05}F_5$. In this deconvolution, the temperatures of the TL peaks are 153.1, 185.3 and 216.1 °C for both glow curves. The peak at 234.2 °C is active only for the sample doped with 5.0 at.% Dy^{3+}.