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Dependence of thermoluminescence glow curve of alumina ceramic radiation detectors with the heating rate

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ABSTRACT

Anion defective single-crystal detectors of aluminum oxide α-Al_2O_3 grown by the Czochralski method are widely used for personal and area monitoring. A methodology for the production of polycrystalline ceramic detectors α-Al_2O_3 based on the sol-gel method was developed in our research group. The dependence of the thermoluminescent response with the heating rate was studied in the range from 0.1 K/s to 10 K/s for both, ceramic produced by our research group and commercial single crystal detectors (TLD-500 like). The studies were performed at a RISØ TL/OSL DA-20 reader - DTU Nutech. All irradiations were done employing a $^{90}Sr/^{90}Y$ beta source facility installed at the reader. In order to study the influence of the heating rate on the luminescent response the ceramic and single-crystal alumina detectors were irradiated with an absorbed dose about 12.5 mGy. In a comparison between the two types of alumina detectors, the single-crystal showed a

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response five times greater than the ceramic response with a linear heating rate of 0.1 K/s. Nevertheless, the response of both of them is almost the same at a heating rate of 7.7 K/s, which is very interesting for routine applications. It is important to note that while the response of the single-crystal decay almost exponentially with the heating rate, which is in accordance with specialized literature, the response of the ceramic detector decays almost linearly in the studied range.

Keywords: **Dosimetry, Thermoluminescence, Heating Rate, Ceramic Detector.**

1.

1. INTRODUCTION

The radiation dosimetry is essential for radiation protection purposes in evaluating absorbed doses related to practices involving ionizing radiation. The evaluation of the absorbed dose is important in several areas that use ionizing radiations for research and development in the nuclear and related areas. For this purpose, different types of dosimeters can be used, considering the advantages and disadvantages of each type. Some of the most commonly used dosimeter types are ionization chambers, dosimetric films, thermal and optical stimulation luminescence (TL and OSL), and semiconductor dosimeters.

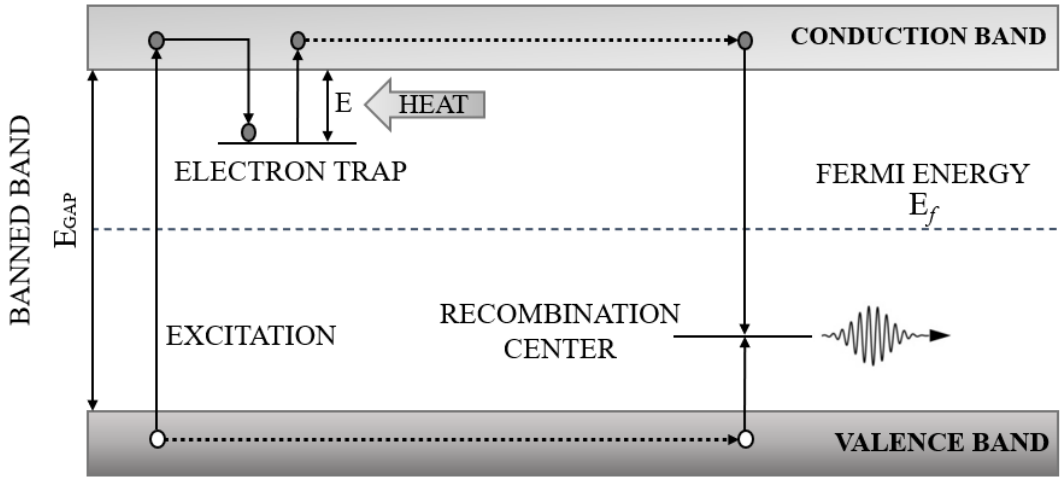
Luminescent detectors based on luminescence effects stimulated by thermal and optical processes can be described by models based on the energy band theory for electron energy levels in solids. Certain materials when heated after being exposed to ionizing radiation fields may show luminescence effects. These materials are generally insulating crystals, in which the valence band is filled, and the conduction band is empty, both separated by a broad energy gap known as forbidden band. A two levels model called the One Trapping One Recombination (OTOR) model provides a simple description of these phenomena [1].

According to this model, when the electrons in the valence band of a TL/OSL material absorb energy from the ionizing radiation field they are excited and may eventually, after thermalization, occupy energy levels as free electrons on the conducting band leaving a complementary charge carrier or hole at the valence band. After the relaxation time characteristic of the conduction band of

this material, the electron undergoes relaxation processes through which is possible to return to the valence band.

This is true for an idealized crystal, however, real crystals present defects in their crystalline structure such as anionic and cationic vacancies and the presence of intrinsic, extrinsic, or substitutional ions. The existence of such defects can induce energy levels in the forbidden band and such levels may act as traps for charge carriers or recombination centers. This charge configuration is metastable. When the TL material is conveniently heated, the trapped electrons can absorb enough energy to return to the conduction band, allowing the subsequent recombination of electron-hole pairs at luminescent centers. In this model, a trap acts as a luminescent center when the probability of recombination is greater than the probability of confinement of charge carriers [2]. Figure 1 represents this simplified model of the thermoluminescence mechanism.

Figure 1: *Simplified model of the thermoluminescence mechanism. During the excitation process, pairs electron-hole are produced. Electrons excited for the conduction band decay spontaneously and can be captured by an electron trap and symmetrically holes can be trapped in holes traps, remaining in a metastable configuration. Thermal stimulation with energy greater or equals to E releases electrons from the trap to the conduction band. The electrons in the conduction band decay spontaneously and become available to recombine with holes at a hole trap which acts as a luminescent center. The energy interval E is called trap depth or activation energy and is E_f is the Fermi energy.*



Source: Elaborated by the authors.

According to this model, when the incident energy from the radiation field is greater than the energy of the bandgap (E_{GAP}), electron excitation occurs from the valence band to the conduction band, producing thus electron-hole pairs [2-4]. The electrons excited for the conduction band can be trapped in electron traps after spontaneous relaxation and the holes can be trapped in hole traps. After a convenient stimulation, trapped electrons can return to the conduction band and, eventually, decay to a hole trap, which at this moment acts as a recombination center [1-3,5-7].

According to Bos [2], the probability of releasing an electron from a trap follows the Arrhenius equation:

$$p = s \exp\left(-\frac{E}{kT}\right) \quad (1)$$

where *p* is the probability per unit time, *s* is the frequency factor which represents the attempt of an electron escaping from a trap. The frequency factor *s* is about 10^{12}-10^{14} s^{-1}. The activation energy between the trap and the conduction band is represented by *E*, as shown in Fig. 1, and k is Boltzmann's constant, typically, in eV.

The intensity TL can be written as :

$$I(t) = -\frac{dn}{dt} = sn \exp\left(-\frac{E}{kT}\right) \quad (2)$$

where *n* is the number of trapped electrons.

In this model, if the temperature is kept constant, then the probability is also constant and the intensity is given by the following equation:

$$I = I_0 \exp(-tp) \quad (3)$$

However, if the temperature varies linearly in time, *p* is no longer a constant, and the equation for the luminescent intensity is given by:

$$I(t) = -\frac{dn}{dt} = sn_0 \exp\left[-\frac{E}{kT(t)}\right] \exp\left[-s\int_0^t \exp\left(-\frac{E}{kT(t')}dT'\right)\right] \quad (4)$$

where n_0 is the total number of trapped electrons in time $t = 0$. As the temperature increases, the intensity rises until a maximum value and, after that, decreases. The intensity takes the form of a peak which is called the first-order glow peak [3].

Usually, TL is observed raising the temperature linearly with the time, according to the following equation:

$$T = T_0 + \beta t \quad (5)$$

where β is the heating rate and T_0 is the temperature at time $t = 0$.

In this case, the intensity is given as a function of the heating rate according to the following equation:

$$I(t) = -\frac{1}{\beta}\frac{dn}{dt} = \frac{s}{\beta} n_0 \exp\left[-\frac{E}{kT(t)}\right] \exp\left[-\frac{s}{\beta}\int_0^t \exp\left(-\frac{E}{kT(t')}dT'\right)\right] \quad (6)$$

This equation is known as Randall-Wilkins's first-order single-peak expression [2].

Like thermoluminescence, the optically stimulated luminescence phenomenon also occurs due to the existence of defects in the crystalline structure. In this case, the material, after being exposed to ionizing radiation, is stimulated by a convenient external light source. Then, the trapped electrons can absorb energy enough to return to the conduction band, allowing the recombination of electron-hole pairs at luminescent centers [8].

The present study aims to compare and evaluate the thermoluminescent response of single-crystal poly-crystal ceramic aluminum oxide detectors when subjected to readings at different heating rates.

2. MATERIALS AND METHODS

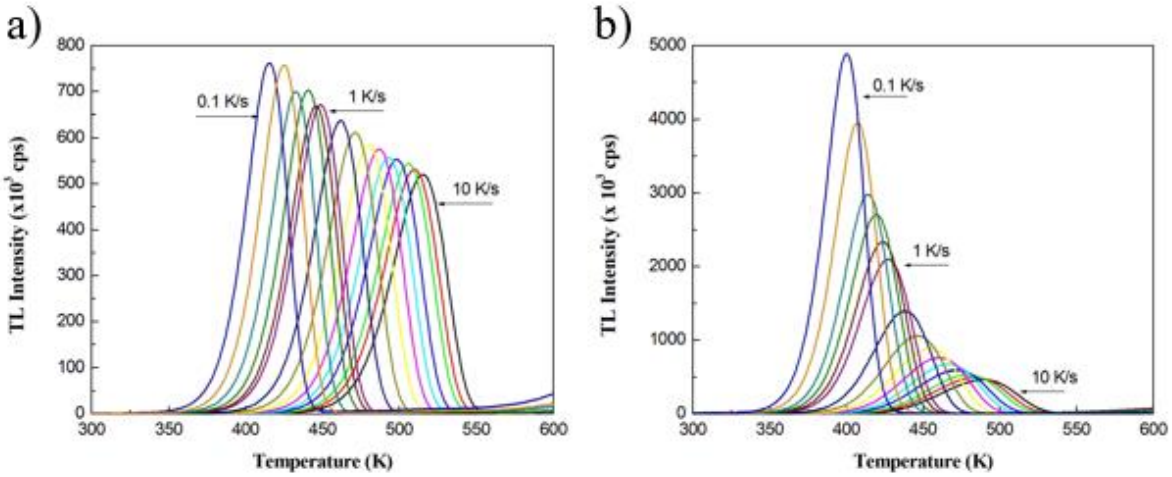
The dosimetric studies were performed using a RISØ TL/OSL DA-20 reader – DTU, produced by the National Laboratory Denmark. The reader is installed at the Luminescent Dosimetry Laboratory – CDTN. The reader is equipped with a carousel of 48 positions and a $^{90}Sr/^{90}Y$ beta source [9]. The ceramic samples were chosen from the same production batch. Five samples were selected by the similarity of their TL responses. The same procedure was made for commercial single-crystal alumina detectors, however, only four detectors were used.

After that, both, the ceramic and single-crystal alumina detectors were annealed at 648 K. After that, they were irradiated at RISØ TL/OSL with 12.5 mGy of beta radiation. The TL response was measured at different heating rates, ranging from 10 K/s to 0.1 K/s. From 10 K/s to 1 K/s the heating rate was decreased in steps of 1 K/s and from 1 K/s to 0.1 K/s the heating rate was decreased in steps of 0.2 K/s.

3. RESULTS AND DISCUSSION

The glow curve for the ceramic detectors for increasing heating rates is plotted in Figure 2 a). Note that the glow peak shifts to higher temperatures with the increase of the heating rate. In the same way, the height of the glow peak decreases with the increase in the heating rate. The result for single-crystal detectors is presented in Figure 2 b). Similarly to the glow curve of the ceramic detector, the single-crystal detector features a shift of the glow peak to higher temperatures with the increase of the heating rate, however, the height of the glow peak decreases with the increase of the heating rate faster when compared to the first case.

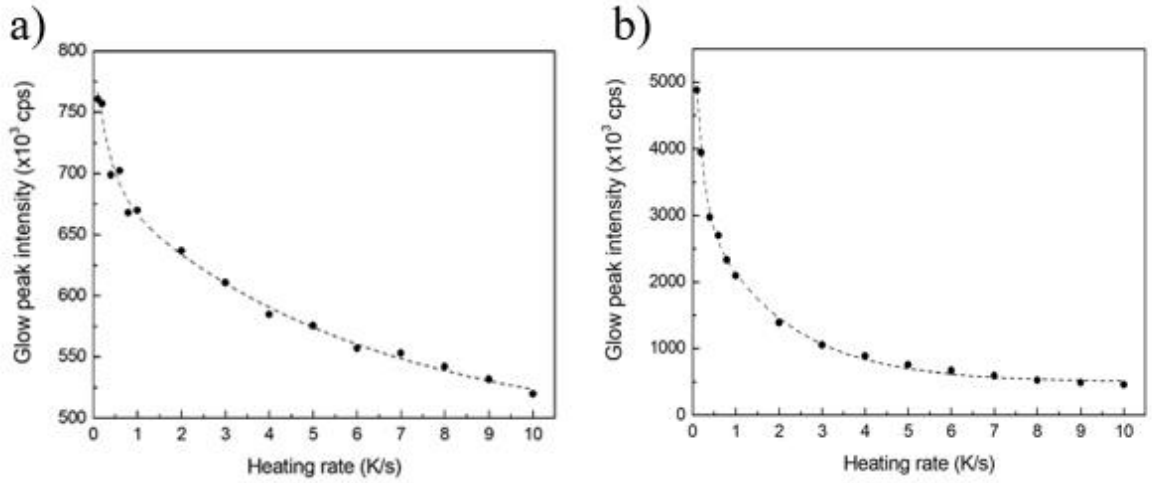
Figure 2: *Dependence of the glow curves with the heating rate for ceramic (a) and single-crystal (b) detectors.*



Source: Elaborated by the authors.

In Figure 3 the peak intensities are plotted as a function of the heating rate, for both: single-crystal (a) and ceramic (b) detectors. Both curves decay exponentially - in fact, a second-order exponential. It is important to note that, although the reduction of the amplitude with the increase of the heating rate is more pronounced for the single-crystal detector, the functional form of such behavior is the same in both cases.

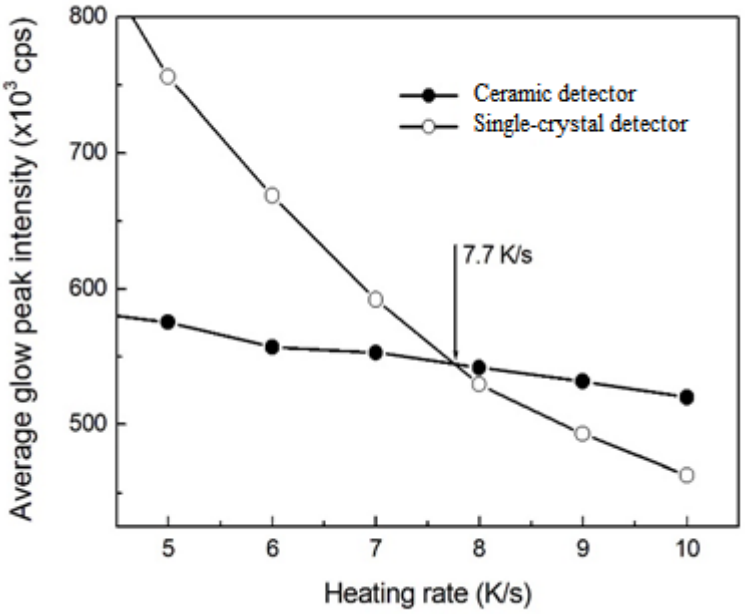
Figure 3: *Dependence of the glow peak intensities as a function of the heating rate for ceramic (a) and single-crystal (b) detectors.*



Source: Elaborated by the authors.

In Figure 4 is presented a superposition of both decays in order to determine the interception point. The interception point was determined graphically at a heating rate about 7.7 K/s.

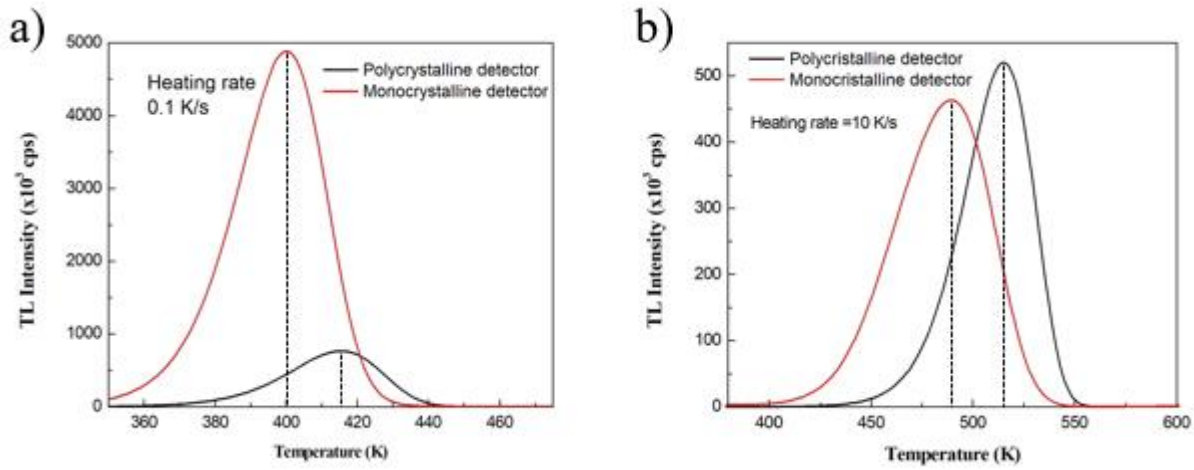
Figure 4: *Interception point of the curves of the average glow peak intensity of single-crystal and ceramic detectors with heating rate.*



Source: Elaborated by the authors.

In Figure 5 it is possible to observe a comparison between ceramic and single-crystal detectors glow curves obtained at different heating rates. At left (Figure 5a), the glow curves obtained at 0.1 K/s present a huge difference. The amplitude of the TL signal is about six times greater for the single-crystal detector than the amplitude for the ceramic detector. At right (Figure 5b), at 10 K/s an inversion occurs and the amplitude of the TL signal for single-crystal detector corresponds to 90% of the amplitude of the ceramic detector glow peak.

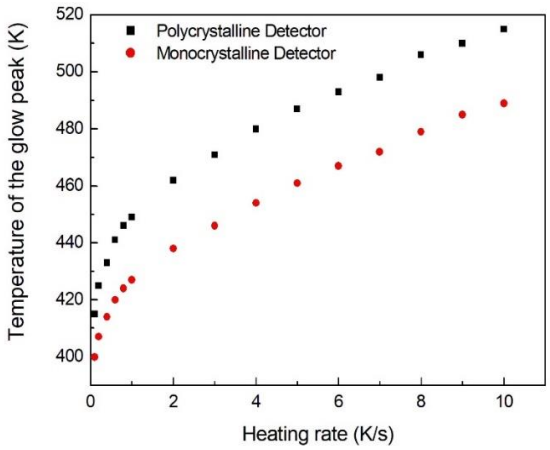
Figure 5: *Comparison between the amplitude of glow curves of the ceramic and the single-crystal detector at heating rates of 0.1 K/s (a) and 10 K/s (b).*



Source: Elaborated by the authors.

Finally, in Figure 6 is presented the shift of the glow peak with the heating rate, for both types of studied detectors. Although the shift of the glow peak with the heating rate follows the same behavior, the temperature range is different what suggests that the required energy to release electrons from the traps is larger.

Figure 6: *Shift of the glow peak with the heating rate for ceramic and single-crystal detectors.*



Source: Elaborated by the authors.

It is interesting to note that the shift of the glow peak to higher temperatures with the increase in the heating rate presented by both detectors is consistent with the need for a larger amount of energy to release the electrons trapped in deeper traps.

Another important observation is the heating rate 7.7 K/s, in which the amplitudes of the thermoluminescent signal of both detectors are, essentially, the same. For ceramic detector, this behavior is especially interesting for applications in which the use of high heating rates is required or desirable.

4. **CONCLUSION**

In this work were studied two different types of aluminum oxide solid-state detectors in its alpha phase. One of them is a commercial single-crystal detector and the other is a polycrystalline ceramic developed and produced at CDTN – Centro de Desenvolvimento da Tecnologia Nuclear, through the sol-gel method. The dependence of the thermoluminescent signal as a function of the temperature at different heating rates was studied.

Like the commercial detector, the ceramic detector exhibited a shift of the glow peak to higher temperatures with the increase of the heating rate. This fact is consistent with the need for a greater amount of energy for the release of trapped electrons. Besides, the amplitude of the glow peak of the ceramic detector decreases with the increase of the heating rate, but with lower decay constants when compared to the behavior of the single-crystal detector.

Although the amplitude of the thermoluminescent signal of the single-crystal detector is larger than the thermoluminescent signal amplitude of the ceramic detector at low heating rates, such a situation is reversed at high heating rates. There is a particularly interesting rate at 7.7 K/s in which the thermoluminescent signal amplitudes of both types of detectors are practically the same. This

behavior of the ceramic detector is especially interesting for applications in which the use of high heating rates is required.

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