Analysis and development of optical methods to re-use optical stimulated luminescent dosimeters

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Abstract. Optically Stimulated Luminescence (OSL) is a worldwide used technique in ionizing radiation dosimetry applications, such as personal and environmental monitoring. At the CDTN-CNEN facilities is produced a ceramic detector of $\alpha$-$\text{Al}_2\text{O}_3$:C, which is used in the routine environmental monitoring and for research purposes. In order to reuse these detectors, it was developed, at Luminescent Dosimetry Laboratory an optical bleaching device, which can operate with green and blue LED illumination modes. The present work describes the use of such device to bleach the optically accessible traps, taking the detectors to its background signal levels. Reference dosimeters $\alpha$-$\text{Al}_2\text{O}_3$:C commercial and magnesium tetraborate were used also for comparison. As a general procedure, before any irradiation, the used detectors were optically bleached by 36 hours. For the best efficiency of bleaching, after irradiation, the ceramic and commercial aluminum oxide detectors were optically bleached under blue LED light for 3 and 6 hours, respectively. For tetraborate detectors, the required time of optical bleaching was 2 hours, under blue LED illumination.

Keywords: Optically stimulated luminescence, aluminum oxide, optical bleaching.

1. Introduction

The Optically Stimulated Luminescence (OSL) is a well-established technique in personal and medical dosimetry [1, 2]. Recently, some Optically Stimulated Luminescent Dosimeters (OSLD) has been produced in film format and is shown to be a viable solution in 2D dose mapping for quality assurance in radiotherapy and radiodiagnostic [3]. The OSLD used for 2D dosimetry has a high spatial and temporal resolution, which is required when it goes towards conformal radiotherapy [4]. Usually, the OSLD is produced as a pellet and its reduced size (3~6 mm) is ideal for in vivo dosimetry, to verify the entrance skin dose in patients during radiotherapy and diagnostic approaches. Also, the OSLD can be attached to optical fibers to evaluate dose rates in real-time [5]. Currently, there are two OSL materials commercially available for dosimetry, which are the $\alpha$-$\text{Al}_2\text{O}_3$:C (e.g., Urals Polytechnical Institute) and the BeO (Brush Ceramic Products, Materion Co.), nevertheless different materials had been suggested and studied for this practice [6, 7].

The main advantage of OSLD concerning its precursor technique, the thermoluminescent dosimetry (TLD), lies in the fact that the optical nature of the readings allows a controlled stimulation. Therefore, the signal from the detector can be partially depleted with the possibility of re-reading (dose re-evaluation). Moreover, the optical nature of the technique avoids some common effects seen in thermoluminescent dosimetry, such as temperature gradient in the sample due to poor thermal contact...
between the pellet and the heating element. Some OSLD are made with polymeric resins (e.g., polytetrafluorochydrolylene, PTFE) and, therefore, it is not possible to subject them to any thermal reading. Another disadvantage currently reported in the TLD is the high temperature of the dosimeters heat treatment, which can cause sensitization in the samples and, consequently, reduce their reproducibility [8]. To reset the OSL dosimeters, the conventional thermal treatment can be replaced by optical bleaching (OB), which has similar efficacy, is a less aggressive way and has a low energy consumption compared to thermal treatment [9]. The OB consists of exposing the detector to a certain range of light wavelengths, which can vary from UV to IR in a certain time, to release the remaining charges from the traps. In order to increase the effectiveness of the OB, it is recommended that the wavelength of the chosen light be the same as the stimulation light used during the OSL reading. At CDTN–CNEN facilities, the alumina ceramic detectors (α-Al₂O₃) are produced by the sol-gel method and are used for area monitoring [2]. Since these detectors have thermally and optically stimulated luminescence properties, an optical bleaching device (OBD) was developed at the Luminescent Dosimetry Laboratory (LDL/CDTN) to make the optic treatment of the TL and OSL residual signals with the purpose of demonstrating the effectiveness of OBD in the bleaching of α-Al₂O₃ ceramic detectors produced in CDTN and to restore their responses to background levels for reuse in routine. In addition to the α-Al₂O₃ detectors (CDTN-CNEN), a ceramic tetraborate detector (MgB₄O₇:Ce, Li) was also tested at the OBD. The MgB₄O₇:Ce, Li is considered potentially important OSL detector and it has been applied in different fields in radiation dosimetry [9, 10].

2. Materials and methods
The OBD is composed of three pairs of symmetrical LED plates. The first pair is composed of blue high intensity LEDs (470 nm), the second of green high intensity LEDs (525 nm) and the last one is composed by white high intensity LEDs (figure 1). As the emission width of white LEDs covers the entire visible spectrum (wide peak until 534 nm), the present work focused on testing the OB with blue and green LEDs, that has narrow peaks. The gap between each plate pair was designed to insert a PMMA transparent tray in which the dosimeters are placed for the OB. The device is blocked from external ambient light with a protective casing. The tests were carried out using the polycrystalline ceramic α-Al₂O₃ detectors produced at CDTN, TLD-500 detectors, and the polycrystalline ceramic MgB₄O₇:Ce, Li produced via solid-state synthesis at Federal University of Sergipe (UFS) [9-11]. During the tests, the detectors were irradiated with two different doses (12.5 mGy and 25 mGy). Following each irradiation the samples were bleached with blue or green light by different times until the samples reach the background levels. Finally, the OSL emission of the samples was checked after OB. Both, the irradiations and OSL readings were done at RISO TL/OSL - DA-200 reader system equipped with blue LED clusters (emission centered at 470 nm), and a Hoya U-340 filter (7.5 mm thickness, transmission between 290 and 390 nm, Hoya Corp. Tokyo, Japan) was placed in front of the photomultiplier (PMT) to block the stimulation light to reach the PMT. The irradiations were performed with a 90Sr/90Y betas-ray source, coupled to the reader, delivering 12.5 mGy/min to the sample position. All the measurements were performed in continuous wave mode (CW-OSL).
The tests were performed with 10 $\alpha$-Al$_2$O$_3$ ceramic dosimeters, nine commercial single crystal TLD-500 detectors, and nine polycrystalline MgB$_4$O$_7$:Ce, Li. For the sake of simplicity, from this point forward the $\alpha$-Al$_2$O$_3$ detectors will be referred as ceramic (CDTN) and single crystal (TLD-500). The ceramic and single crystal detectors were irradiated with 12.5 mGy and 25 mGy. After each irradiation, the samples were exposed to OB with blue light with times varying from 1 minute to 36 hours. Finally, after each OB, the OSL was measured to evaluate its efficacy. The MgB$_4$O$_7$:Ce, Li pellets were irradiated with absorbed dose of 500 mGy and submitted to OB with blue light for 60 and 120 min. The data analysis was done through measurements of residual OSL signal after each OB. For the purpose of this work, the residual OSL value is defined according to equation 1.

$$\text{OSL} = \frac{\text{OSL}_{\text{ref}} - \text{OSL}_{\text{BG}}}{\text{OSL}_{\text{ref}} - \text{OSL}_{\text{BG}}}$$

where,

- OSL$_{\text{ref}}$ is the OSL signal without OB;
- OSL$_{\text{res}}$ is the OSL signal after OB
- OSL$_{\text{BG}}$ is the OSL signal of the samples without irradiation.

3. Results

In figure 2 is shown the average OSL counts and the respective standard deviation of non-irradiated detectors after an OB with sunlight, for two hours, and after OB by 36 hours in the OBD with blue light. This experiment aims to reach the minimal signal, which is the residual OSL signal and considered here, as zero reference. The choice was made also because the OSL signal of non-irradiated samples optically bleached by 36 hours is very close to the counts of the reader instrumental noise, represented in Figure 2 as a dotted line. The instrumental noise here is the OSL readings without any sample. This result shows the efficacy of the OBD with blue light in bleaching the OSL emission centers for both alumina detectors types and due to the higher sensitivity of single crystal $\alpha$-Al$_2$O$_3$:C, the non-irradiated samples still presented some OSL signal due to absorption of natural radiation [11] and so the OB reduced the OSL signal in 60 %. For the ceramic, the OB reduced the signal in 18 %. 
Figure 2. OSL signal of non-irradiated samples and after OB by sunlight (2 hours) and OB by 36 hours with blue light at OBD. The dotted line is the instrumental noise level of the reader.

In figure 3 is seen that OSL signal of the ceramic samples reached the background level after an OB of 15 hours with green light or 6 hours with blue light. For the single crystal samples, figure 4, an OB of 15 hours with green light was not enough to take the signal to the background level. In fact, it is seen a slight increase in the signal, leading us to believe that the green light promotes charge phototransference in this material. Indeed, the results showed that the OB of 6 hours with blue light is effective for the single crystal samples and the lower efficiency presented by the OB green light is possibly due to its lower energy, compared to blue emission.

Figure 3. The OB of ceramic detector with blue and green light the samples were previously irradiated with 25mGy.

Figure 4. Single crystal detector with blue and green light the samples were previously irradiated with 25mGy.

In figure 5 is presented the residual OSL achieved through equation 1, for both alumina detector types, optically bleached with blue light at different times. Note that an OB by 3 hours reduces the OSL signal in 90%, however, for single crystal samples, figure 6, an OB by 6 hours is recommended to take the samples to background level. The higher standard deviation showed in the OSL residual measurements were observed for the OB below 10 min and reached maximum value of 30%, for the
first 5 minutes of OB. Above 10 min of OB, the relative standard deviation decreased considerably, as an example, the relative standard deviation was 5 % for OB of the samples at 3 hours and 6 hours, for the ceramic and single crystal, respectively.

In figure 7 is shown the results for MgB$_4$O$_7$ samples. For this test, the dose was increased up to 500 mGy, once that the material has lower sensitivity compared to the $\alpha$-Al$_2$O$_3$:C [8, 9]. In the figure is seen the OSL exponential decay curve of samples exposed to 500 mGy of beta radiation ($^{90}$Sr/$^{90}$Y), afterward the samples were optically bleached by 60 and 120 minutes with blue light. According to the results, the OB by 120 minutes take the OSL signal within the standard deviation of the OSL measurements of non-irradiated samples and so we can affirm the effectiveness of the optical treatment for the tetraborate case. All the perceptual standard deviation in this measurement was below 15 %.

**Figure 5.** Residual OSL from ceramics after OB (blue light) for different times and for two absorbed dose values.  
**Figure 6.** Residual OSL from single crystal after OB (blue light) for different times and for two absorbed dose values.  
**Figure 7.** OSL decay of polycrystalline MgB$_4$O$_7$:Ce, Li exposed to 500 mGy at beta ($^{90}$Sr/$^{90}$Y) radiation source.
4. Conclusion
The purpose of this work was to determine the effectiveness of OBD operating with blue and green light for bleaching the OSL signals in different solid-state radiation detectors instead using common thermal treatments up to 900° C for alumina detector, for example. In a general way, for the types of detectors studies, OB with green light needs at least the double of time to bleach the samples (single crystal and ceramic) with the same efficacy in comparison to the blue light. In addition, the green light can possibly promote phototransference of charges among traps. It is also recommended that prior to any kind of irradiation, the Al2O3:C (ceramic or single crystal) should be bleached by 36 hours using blue light, in order to achieve the lowest signal for the samples. This is a very important step to perform in dosimeters, once that some parameters in dosimetry, such as lowest detectable dose, is related to the lowest signal that a non-irradiated sample can reach. Furthermore, the best operation conditions were determined as OB by 3 hours for the ceramic alumina detectors, OB by 6 hours for TLD-500 detectors although was observed that after 3 hours, the bleaching efficacy is around 90 %. For polycrystalline MgB4O7:Ce, Li produced at dosimetry group from UFS and it was possible to reach the lower level of signal after 2 hours of bleaching. Previous results recommended an OB about 60 minutes with blue light to bleach these samples. However, in the present study, the required OB period was twice as long to achieve the same result. Although we are dealing with the same composition, the batches sensitivity may differ, which is directly related to the amount of traps available [11].

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