Investigation of Spodumene TLD Response to Monoenergetic Neutron Fields


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INTRODUCTION

The application of ionizing radiations in different fields requires the use of equipment capable of detecting their presence or quantifying them. Detectors of the most different shapes, composition and characteristics are required to cover the full range of application of ionizing radiations. At present, thermoluminescent detectors (TLD) are the most widely employed for individual gamma monitoring. Thermoluminescence (TL) is a process in which heated insulator or semiconductor emits the light in correlation with previous absorption of energy from ionizing radiation [1]. Several materials that present intense TL are used as TLD to measure radiation doses for manifold applications [2], including neutron dosimetry, where nowadays a sufficiently reliable method, covering the whole range of neutron energies, does not exist [3].

For neutron dosimetry applications, elements with large neutron cross section (as 6Li and 10B for thermal neutrons [4]) are added to the basic components of TLD. Generally, because the neutron cross section depends markedly on the neutron energy, there are different detectors for thermal neutrons and fast neutrons. The 10B(n,α)7Li reaction is most often used for detection of thermal neutrons, while the reaction 6Li(n,α)H is also used in the neutron albedo detectors.

In this work, a series of preliminary measurements, investigating the neutron response of spodumene TLD crystals (LiAlSi2O6), is described. As far as we know, TL response of any type of spodumene TLD to neutrons radiation has never been reported.

These crystals emit intense light with a wavelength around 600 nm, when irradiated with high doses from X and gamma rays, or beta particles from 90Sr-90Y source, with the main TL peak in a temperature range suitable for dosimetric applications [5-8]. The feasibility to use boron as a spodumene dopant has been also investigated.

MATERIALS AND METHODS

The TL neutron response of β-spodumene pellets, agglutinated with Teflon, was considered. Synthetic β-devitrification method [9]: undoped and doped with controlled concentration of boron (0.5 wt.%) with natural isotopic abundances (19.9% of 10B and 80.1% of 11B). A set of 20 mg pellets was obtained, with 0.2 mm thickness and 6.0 mm diameter, following the procedure described in a previous paper [5]. For their sintering, they were exposed to thermal treatments of 300 °C for 30 min and 400 °C for 1.5 h in a resistive oven (EDG-1800 type).

Spodumene crystals were irradiated by neutrons generated by a proton beam, incident on a thin (700 μg/cm2) LiF target (threshold energy, Eth=1.88 MeV). Two proton energies of 1935 keV and 2900 keV were selected to obtain two quasi-monoenergetic neutron beams of 144 keV and 1200 keV, respectively. The neutron energy and fluence produced via 7Li(p,n) reaction were calculated at the irradiation positions with the DROSG-2000 code [10], by taking into account also neutron production from the 0.430 MeV excited level of 9Be, for the latter proton energy. Protons were generated by the CN Van de Graaf accelerator of the LNL-INFN, whose working current ranged from 100 to 200 nA during the experimental campaign.

The crystals were placed on a thin PMMA plate (3 mm thick) centered on the beam axis at a distance of 10 cm from the target holder. The Plexiglas holder plane was perpendicular with respect to the impinging proton beam. Nine dosimeters were contemporary irradiated without overlapping; one of the dosimeters was coaxial with the beam, while the maximum displacement between beam axis and external detectors was 2 cm. Within this distance, the detectors were uniformly irradiated, given maximum neutron emission cone half-angles of 90° for both 1935 keV and 2900 keV proton energies [11].

In order to characterize the neutron dose field, LB 6411 neutron dose rate detector by Berthold was used. The average current of the proton beam was measured with a Faraday cup connected to a charge integrator. Average dose rates of 8.16 ± 0.01 mSv/h and 0.31 ± 0.01 mSv/h were measured at the irradiation position for 1935 keV and 2900 keV, respectively. Photon contribution is mainly due to nuclear reactions of incident protons with the 7Li and 19F in the target, but the related dose was negligible [12, 13], considering the spodumene detectors relatively low photon sensitivity [7].

After exposure, TL signal was immediately acquired by means of a portable reader (Harshaw, mod. 200C). A linear readout profile was adopted from 25 to 29 °C, with a heating rate of 10 °C/s.

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EXPERIMENTAL RESULTS

The TL neutron response was investigated for boron loaded and unloaded spodumene crystals. Table 1 shows the results in terms of charge collected by the TLD reader, before and after irradiations with 144 keV and 1200 keV neutrons for a 5 mSv absorbed dose. Only the boron loaded samples produced a TL signal higher than background, while a similar phenomenon was not reported for the undoped spodumene detectors.

The behavior of undoped detectors was expected, because their response to neutrons is only referable to their natural lithium content. However, $^6\text{Li}$ (which is the neutron sensitive isotope) natural abundance is 7.5%, which is not enough to improve spodumene response to neutrons.

The energy and dose dependence of the response was also investigated. As $^{10}\text{B}$ neutron cross section increases with neutron energy decreasing, a higher sensitivity is expected for lower energies (as already shown in table 1). This was verified by irradiating boron loaded spodumene with 144 keV and 1200 keV neutrons for different doses (figure 1). While boron loaded detectors response seems not to be affected by increasing dose at 1200 keV neutron energy; they showed a higher response with increasing doses if irradiated with 144 keV neutrons. Reported error bars in figure 1 are mainly due to batch variability. The energy and dose dependence of the response has been loaded with boron and preliminary results of its response to neutrons have been presented in this work.

To increase the neutron sensitivity, future experiments will be performed adding $^{10}\text{B}$ as a dopant instead of natural boron and also examining the possibility to add $^6\text{Li}$ to the crystal composition.

Studies on newer detector configurations will be also initiated, to include shallow converting layers suitable to improve boron loaded spodumene sensitivity to higher energy neutrons.

CONCLUSIONS

Spodumene has been proven to be a promising material in gamma ionizing radiation detection, via its TL properties. In order to achieve a better sensitivity to neutrons, spodumene has been loaded with boron and preliminary results of its response to neutrons have been presented in this work.

A fairly clear response of boron loaded detector to 144 keV neutrons, increasing with delivered dose, has been found. A similar response has not been shown by both unloaded spodumene samples and by boron loaded spodumene at 1200 keV energy.

To increase the neutron sensitivity, future experiments will be performed adding $^{10}\text{B}$ as a dopant instead of natural boron and also examining the possibility to add $^6\text{Li}$ to the crystal composition.

Studies on newer detector configurations will be also initiated, to include shallow converting layers suitable to improve boron loaded spodumene sensitivity to higher energy neutrons.

Table 1. Detector responses (charge) of boron loaded and unloaded spodumene crystal before and after 5 mSv irradiations at 144 and 1200 keV.

<table>
<thead>
<tr>
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<th>Boron doped spodumene</th>
<th>Undoped spodumene</th>
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<tbody>
<tr>
<td>Background</td>
<td>(2.0 ± 0.8) x 10^{-2}</td>
<td>(15.6 ± 7.3) x 10^{-2}</td>
</tr>
<tr>
<td>144 keV irradiation</td>
<td>(11.2 ± 2.2) x 10^{-2}</td>
<td>(11.1 ± 4.4) x 10^{-2}</td>
</tr>
<tr>
<td>1200 keV irradiation</td>
<td>(5.4 ± 1.7) x 10^{-2}</td>
<td>(8.9 ± 4.9) x 10^{-2}</td>
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Fig. 1. Boron loaded spodumene TL detected signal ($±$ 1 s.d.) as a function of neutron dose for irradiation energies of 144 and 1200 keV.

REFERENCES