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Preliminary study on development and characterization of high sensitivity $LiAlO_2$ optically stimulated luminescence material

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ABSTRACT

Preliminary results of an attempt to prepare $LiAlO_2$ material with high optically stimulated luminescence (OSL) sensitivity are reported. $LiAlO_2$ was prepared by melting a stoichiometric mixture of Li_2CO_3 and Al_2O_3 powders in a RF heating furnace. CW-OSL signal as recorded on Risoe TL/OSL reader using blue (470 nm) stimulation was found to be up to 16 times of that of Al_2O_3:C. The promising characteristics of $LiAlO_2$, open up a possibility of an improved material not only for passive dosimetry of mixed fields of neutron and gamma rays but also for online measurements and dose mapping/imaging applications.

1. Introduction

Optically stimulated luminescence (OSL) is finding applications in a wide variety of radiation dosimetry requirements due to its optical nature of readout (Yukihara and McKeever, 2011; Pradhan et al., 2008). In view of unavailability of a better material, Al_2O_3:C continues to remain the material of choice in spite of its non-tissue equivalence, reduced sensitivity to high LET radiation and insensitivity to neutrons. It is realized that the progress on the development of OSL materials has been very limited and due attention needs to be paid to accept a challenge of developing improved OSL materials. High OSL sensitivity of Al_2O_3:C is attributed to the creation of a large oxygen vacancies during the preparation at high temperatures leading to production of radiative recombination pathways and a large numbers of F and F+ centres (McKeever, 2011). From the success story of Al_2O_3:C, an evident direction for development of an efficient OSL material could be a choice of oxide (to withstand high temperature heat treatments) with effective atomic number lower than Al_2O_3:C and inclusion of an element like Li to provide a choice for mixed field dosimetry of neutron and gamma rays as in the case of TLD for the use of LiF:Mg, Ti, LiF:Mg, Cu, P and LiF:Mg, Cu, Si, each with ^{6}Li and ^{7}Li enriched LiF to form a pair of neutron sensitive and neutron insensitive dosimeters (Cherestes et al., 2010; Kralik et al., 2010; Lee et al., 2008). One of the obvious choices has been $LiAlO_2$ with effective atomic number of 10.7 (lower than 11.3 of Al_2O_3:C) indicating that its photon energy dependence should be lower than that of Al_2O_3:C. Although, some attempts with partial success have been made to develop $LiAlO_2$:Mn, $LiAlO_2$:Ce and $LiAlO_2$:Tb (Mittani et al., 2008; Dhabekar et al., 2008 and Teng et al., 2010), a need for a fresh attempt by drawing a similarity from Al_2O_3:C was recognized. The present study reports the first results of a successful attempt.

2. Material and methods

Li_2CO_3 and Al_2O_3 powders were taken in stoichiometric ratio and were mixed thoroughly. The mixture was heated up to 1200 °C in a RF heated furnace in Ir and graphite crucibles. This temperature was maintained for 1 h to allow the synthetic reaction for formation of $LiAlO_2$. The temperature was further raised to melt the product. For this, the crucibles were cladded by ZrO_2 felt and bubbles to obtain the required insulation. High purity N_2 gas was always flown and maintained during the heating to protect the crucibles after creating a partial vacuum for evacuating oxygen in the heating zone. A seed of $LiAlO_2$ was used to dip in the center of the melt surface. The melt with the seed was cooled by using a preset profile down to room temperature. The aim of using a graphite crucible was to have a reducing atmosphere for diffusion of C for carbon doping. The reactivity of graphite with Li compound resulted in a polycrystalline and opaque material as compared to transparent product in the Ir crucible. The polycrystalline materials were crushed in a mortar and a pestle to obtain phosphor grains in the

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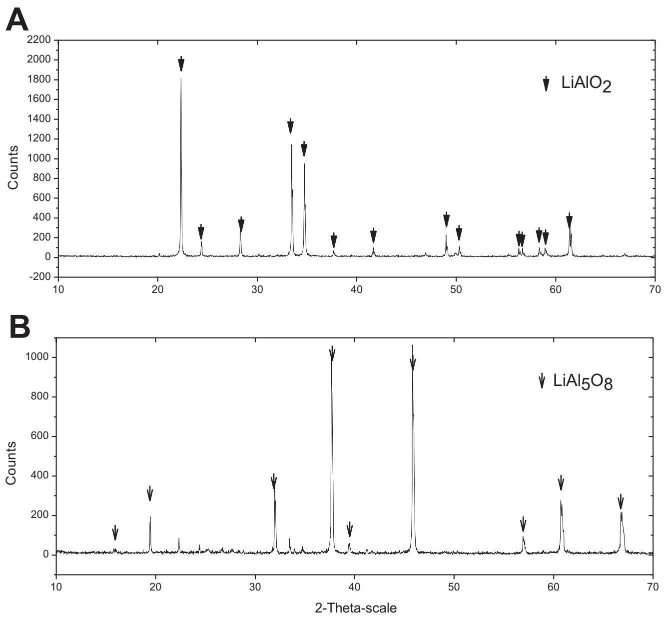

Fig. 1. Results of X-ray diffraction analysis of Ir crucible melt (dominated by $LiAlO_2$ formation, Fig. 1A) and graphite crucible melt (dominated by $LiAl_5O_8$, Fig. 1B) with all other parameters remaining the same.

range from 100 to 300 microns. A high-resolution X-ray diffraction equipment (D-5000 Siemens) was used for the analysis. Additional heat treatments to the phosphors were given in a separate muffle furnace. All irradiations and OSL readouts were carried out at room temperatures in a dark room lit by dim red light. OSL and TL readouts were made on a Risoe reader (TL/OSL-DA-20), equipped with blue (470 nm) LEDs for optical stimulation and a bialkali EMI 9235QA PMT for recording OSL. This OSL reader (under routine use for dating studies using quartz) has a combined filter of Hoya U-340 (transmission window 270–380 nm) and Schott BG-39 (transmission window 330–620 nm) in front of the PM tube to ensure minimal interference from the stimulating light. The reader can accommodate up to 48 samples and has an attached ^{90}Sr–^{90}Y beta irradiator to provide a dose rate of 90 mGy/s. For comparison, Al_2O_3:C powder procured from Landauer Inc., USA was used.

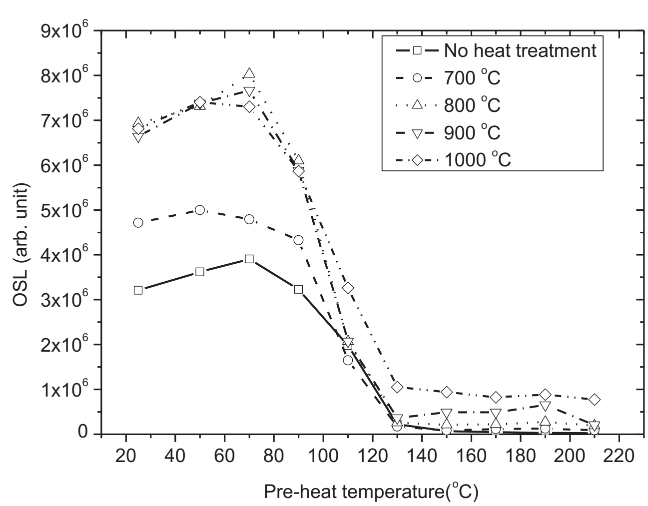

Fig. 2. Effect of heat treatment for 30 min before irradiation and pre-heat before readout (in the OSL reader) on OSL sensitivity of $LiAlO_2$. The OSL sensitivity is represented by an integration of CW-OSL signal for the first 10 s during readout for 40 s.

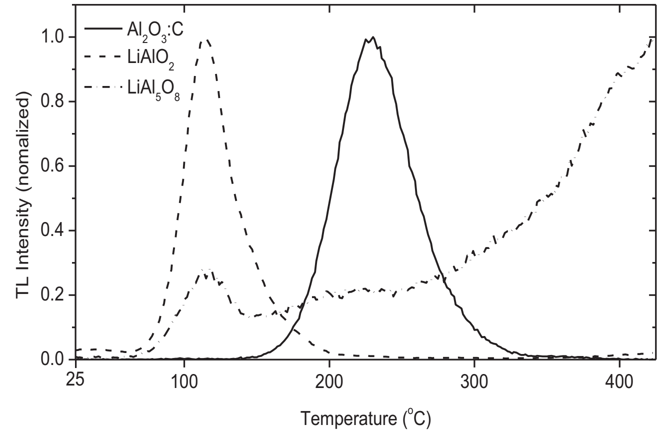

Fig. 3. TL glow curves of Al_2O_3:C, $LiAlO_2$ and $LiAl_5O_8$ irradiated to 900 mGy beta rays from ^{90}Sr–^{90}Y and recorded on Risoe reader (heating rate 5 °C/s).

3. Results and discussion

Fig. 1 shows the results of X-ray diffraction analysis. Ir crucible melt gave a product dominated by $LiAlO_2$ formation whereas graphite crucible melt resulted in $LiAl_5O_8$ dominated product. Preparation of the samples in graphite crucible changed the composition to $LiAl_5O_8$ due to the reaction of Li compounds with graphite. The powdered samples from the Ir crucible melt and the graphite crucible melt will henceforth be referred as $LiAlO_2$ and $LiAl_5O_8$, respectively. $LiAlO_2$ samples were subjected to additional heat treatments for 30 min in air at temperatures ranging from 700 to 1000 °C to enhance the ion vacancies leading to probable enhancement of production of F and F+ centres as suggested by Teng et al. (2010) by drawing a similarity from Al_2O_3:C. The effect of heat treatment (before irradiation) on OSL sensitivity of $LiAlO_2$ samples is shown in Fig. 2. For this, OSL sensitivity is presented by an integration of CW-OSL signal for the first 10 s from readout for 40 s. It can be seen that a heat treatment at 800 °C for 30 min in air appears optimum. Fig. 2 also shows the effect of pre-heat treatment of 10 s given in the readout system prior to OSL readout. The OSL sensitivity was found not to reduce significantly with increasing pre-heat temperatures up to 80 °C, above which it rapidly deceased but stabilized at a highly reduced level beyond 120 °C. This indicates that ambient temperatures below 70 °C may not have significant influence on the OSL signal. Normalized TL glow curves of Al_2O_3:C, $LiAlO_2$ and $LiAl_5O_8$ recorded by using a heating rate of 5 °C/s in the Risoe reader, are shown in Fig. 3. $LiAlO_2$ exhibited a glow peak at 95 °C indicating some correlation with the decrease in OSL signal above 80 °C. $LiAl_5O_8$ had peaks at higher temperatures up to 400 °C in addition to 95 °C peak but its poor OSL sensitivity (Table 1) makes it no more attractive.

The OSL sensitivities of $LiAlO_2$ and $LiAl_5O_8$ are compared with Al_2O_3:C in Table 1 for different types of reporting of OSL readout

Table 1
The OSL sensitivities of $LiAlO_2$, $LiAl_5O_8$ and Al_2O_3:C (all samples were bleached by 90% of the total LED power for 200 s prior to beta ^{90}Sr–^{90}Y irradiation of 90 mGy) for different types of reporting of OSL readout. OSL readout of un-irradiated samples with or without bleaching was about the same and was more than three orders of magnitude lower than that of the samples irradiated to 90 mGy.

Type of reporting of OSL readout	Al_2O_3:C	$LiAlO_2$[a]	$LiAlO_2$[b]	$LiAl_5O_8$
Initial intensity	1.00	7.7	15.8	0.04
Integral for 1 s	1.00	4.6	8.5	0.03
Integral for 10 s	1.00	2.3	4.3	0.02

[a] No thermal treatment before irradiation.
[b] Thermally treated at 800 °C for 30 min in air before irradiation.