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New OSL detectors based on $LiMgPO_4$ crystals grown by micro pulling down method. Dosimetric properties vs. growth parameters

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HIGHLIGHTS

- Crystals of $LiMgPO_4$ compound have been grown by micro pulling down technique.
- Growth parameters and conditions influence the crystals dosimetric properties.
- The higher the growth rate, the lower the fading (i.e. higher residual OSL signal).
- For higher growth rates, the OSL signal along the growth axis is more nonuniform.
- Response nonlinearity does not exceed 10%, regardless of the growth parameters.

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ABSTRACT

Rod-shape crystals of the $LiMgPO_4$ compound were grown by micro pulling down technique under the different growth conditions. Influence of the different growth rates, thermal setups and gaseous atmospheres on the crystals dosimetric properties was investigated. Samples were irradiated with $^{90}Sr/^{90}Y$ β particles and optically stimulated luminescence spectra were measured with the automatic Risø TL/OSL-DA20 reader. The sensitivity level, repeatability, dose–response dependence and short-time fading were compared for all grown crystals. It was found that the crystal grown from the iridium crucible was about three times more sensitive to radiation as compared to the crystal grown from the graphite crucible. Also the radio-sensitivity measured for the crystals grown from the graphite crucible was higher in case of higher growth rates. It was also shown that the residual OSL signal measured one and two weeks after the irradiation was higher for the crystals grown with higher growth rates. There was also no correlation observed between the growth conditions and the level of dose–response nonlinearity. Over the studied dose range the response nonlinearity of the studied samples fluctuated around over a dozen percent, regardless of the applied growth parameters. The obtained results tend to suggest that $LiMgPO_4$ crystals may be considered as promising dosimeters in different fields of research.

1. Introduction

Increasing number of applications of ionizing radiation (e.g. in medicine, radiotherapy, industry, science) stimulates the need of development of new measurement techniques. There is a need for new materials to produce simple and cheap dosimeters, which will be able to measure progressively lower doses of radiation over a broad energetic range. The special attention should be paid for the young domain of optically stimulated luminescence (OSL), which recently becomes more and more widely applied measurement technique. However, the number of available OSL materials is quite limited. In practice, there are only two available OSL materials, Al_2O_3:C (Viamonte et al., 2008) and BeO (Sommer et al., 2008), which are effectively utilized. This stimulates efforts for seeking new dosimetric materials usually in the form of complex oxides crystals.

A relatively new method of single crystal growth is micro pulling down (MPD). This method has been mostly developed since 1992 (Fukuda and Chani, 2007). The MPD is an example of the crystal growth from the melt, similarly as the better known Czochralski method. In case of the MPD, however, there is no need for crystal rotation and the crystal is pulled downward through the micro channel made in the bottom of the crucible. The growth process is supported by capillarity and gravity forces, which

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constitute the main driving forces in MPD technique. The crystal growth process begins by touching the bottom of the crucible by appropriately oriented seed. Then, the solid/liquid interface is formed under the crucible, what is supported by the properly established temperature gradient. Such a system configuration causes that the as-grown crystals diameters may be ranged approximately from 0.1 to 10 mm. The MPD technique is also well suited for material research. This method allows for obtaining single crystals of various chemical compounds in relatively short period of time (in the order of several hours). It also requires a small amount of the starting material. Therefore, this method is dedicated for searching the materials of specified properties, for optimization of growth parameters and conditions etc.

In recent years, the papers indicating high OSL sensitivity of $LiMgPO_4$ (LMP) compound have been published (Kumar et al., 2013; Menon et al., 2012; Minqiang et al., 2013). These papers show that the LMP is well suitable for personal dosimetry applications (Kumar et al., 2013), dosimetry of high radiation doses (Minqiang et al., 2013), as well as for real time dosimetry (Menon et al., 2012). The possibility of neutron measurements also constitutes a potential field of application for the LMP compound, because of the content of lithium, which has a high cross section for thermal neutron capture. Within this work an attempt was undertaken to obtain OSL detectors based on $LiMgPO_4$ compound crystals grown by the MPD technique at the different growth parameters. The detailed analysis of the studied growth parameters and their influence on the as-grown crystals OSL properties are discussed.

2. Materials and methods

2.1. Powder preparation

In this study the raw $LiMgPO_4$ powder was prepared by solid state reaction between:

$LiOH \cdot H_2O$, $Mg(NO_3)_2 \cdot 6H_2O$ and $NH_4H_2PO_4$

in air. Several annealing cycles at temperatures from 200 to 750 °C were included to the preparation procedure. Then the material was ground and sieved to achieve the powder of grain size below 212 μm.

2.2. Crystal growth setup and growth conditions

$LiMgPO_4$ crystals were grown by micro pulling down technique, using dedicated equipment available at the Institute of Nuclear Physics Polish Academy of Sciences in Krakow, Poland. This device consists of inductive furnace, 20 kW middle-frequency generator and dedicated closed circuit cooling system. Inside of the furnace temperatures above 2000 °C can be easily reached. Raw LMP material (in an amount of about 1 g) was loaded into a crucible and heated to the melting temperature. The relatively low melting point of the LMP compound, which is about 1025 °C, makes it possible to use both the iridium and graphite crucibles (due to poor excitation features, it is difficult to use graphite crucibles for materials of high melting points). The graphite after-heater and two layers of alumina ceramic thermal isolation were also applied in order to assure appropriate temperature gradient within the growth area. The growth processes were performed at different pulling rates as well as in the presence of different inert gas atmosphere. The applied growth parameters and thermal setup configurations for all crystals studied within this work are shown in Table 1. The obtained rod-shape crystals had around 3 mm diameter and up to 60 mm length (see Fig. 1). The crystals were cut into smaller samples. Individual samples had around 3 mm diameter and 3–5 mm length.

2.3. Irradiation and readout conditions

All measurements presented in this work were performed using automatic Risø-TL/OSL-DA20 reader. The reader is equipped with beta radiation source ($^{90}Sr/^{90}Y$), which irradiates the samples with the dose rate of about 67.4 mGy/s and maximum beta particles energy of 2.27 MeV. The continuous wave (CW) OSL signal was measured with the bialkali photomultiplier tube (EMI9235QB), which has maximum detection efficiency between 200 and 400 nm. For sample stimulation, blue LEDs (470 nm) arranged in 4 clusters each containing seven individual LEDs were utilized. The total power from 28 LEDs is 80 mW/cm^2 at the sample position (Bøtter-Jensen et al., 2003). Detailed specification of the reader and its performance was recently described by Bilski et al., 2014 and Wróbel et al. 2015. All measurements were performed at room temperature over the stimulation time period of 600 s.

3. Results and discussion

The OSL measurements for the obtained $LiMgPO_4$ crystals were performed using the automatic Risø reader. The measured signal was additionally filtered by the Hoya U-340 optical filter, which has maximum transmittance value of about 0.8 for the wavelength range of 300–350 nm. The measured OSL glow curves for all studied LMP samples are shown in Fig. 2 (for comparison, the glow curve measured for commercially available Al_2O_3:C detector is also presented). These curves were measured for the samples irradiated with the dose of 0.5 Gy. It is visible that the sample, which was grown from the iridium crucible exhibits the highest sensitivity to radiation. For a given growth rate, this sample shows more than 3 times higher response as compared to the other analyzed samples. These results can be found in Fig. 3. It should be noted that all samples were fully bleached before the measurements. This difference may be related to reducing atmosphere, caused by the presence of graphite, as well as by addition of the hydrogen to the growth atmosphere in case of samples 275 and 276. The general trend observed for the samples grown from graphite crucible is that the higher the growth rate, the higher sensitivity to radiation. This probably results from the fact that for the higher crystallization rates, more structure defects are created. Because the luminescence occurs on structure defects, higher luminescent signal for more defected crystals is expected. However, this was not observed for samples grown in reducing hydrogen atmosphere, for which the response is constant within calculated uncertainties, regardless of the applied growth rate.

In the second step, the repeatability of the studied samples was checked, since response repeatability for reproducible exposure conditions is highly required for radiation detectors. For this purpose the series of consecutive measurements were performed. Repeatability investigations were carried out in the following sequence: bleaching (1 h treatment under the GL-FG 115 lamp, at 455 nm), low dose irradiation (0.2 Gy, $^{90}Sr/^{90}Y$ beta source), readout (600 s CW-OSL measurements). The obtained results are presented in Fig. 4. It should be stressed that the length of each bar shown in Fig. 4 indicates the minimum and maximum deviations calculated for particular sample originating from the same LMP crystal. Overlapping bars for the growth rates of 1.0 and 2.0 mm/min have been slightly shifted to improve readability of the graph. It is clearly visible that increasing growth rate causes that the OSL signal measured along the crystal growth axis becomes nonuniform. Therefore, standard deviations calculated for several samples originating from the same LMP crystal are higher. It seems that higher crystallization rate not only produces more structure defects (which generate higher luminescent signal, see Fig. 3), but also makes the OSL-related structure defects distribution strongly