Radiation Measurements xxx (2017) 1–6

Contents lists available at ScienceDirect

Radiation Measurements

journal homepage: www.elsevier.com/locate/radmeas

Effect of dopants' concentration on high-dose high-temperature thermoluminescence of LiF:Mg,Cu,P detectors: Mg and Cu influence

Barbara Obryk [a, *], Mariusz Kłosowski [a], Patrycja Baran [b, d], Krzysztof Hodyr [c]

[a] Institute of Nuclear Physics Polish Academy of Sciences (IFJ), Ul. Radzikowskiego 152, 31-342 Kraków, Poland
[b] AGH University of Science and Technology, Al. Mickiewicza 30, 30-059 Kraków, Poland
[c] Institute of Applied Radiation Chemistry, Lodz University of Technology, Ul. Wróblewskiego 15, 90-924 Łódź, Poland
[d] ARC Centre of Excellence in Advanced Molecular Imaging, School of Physics, The University of Melbourne, Parkville 3010, Australia

HIGHLIGHTS

- Variation of dopants' concentration is influencing the shape of glow-curve.
- Magnesium presence is crucial for occurrence of the TL peak 'B'.
- Mg influences the shape of the glow curve and high-temperature peaks intensity.
- Cu influences high-temperature peaks intensity mainly.

ARTICLE INFO

Article history:
Received 29 September 2016
Received in revised form
14 February 2017
Accepted 7 March 2017
Available online xxx

Keywords:
Thermoluminescence
Lithium fluoride
High-dose
TL peak 'B'

ABSTRACT

In search of origin and mechanism of the high-dose high-temperature TL emission with presence of the peak 'B' about thirty batches of LiF:Mg,Cu,P detectors with different dopants' concentration were produced and comprehensive studies of their high-dose TL features have been performed. Within each step of the research, these characteristics were compared with the high-dose TL properties of the typical MCP-N detectors' batch highly exposed simultaneously with the detectors from batches of varying dopants concentration. We determined the effect of concentration of specific dopants on the TL peak 'B'. It was found that both magnesium and copper are necessary to obtain high-dose TL signal, however, the important for high-dose TL features seems to be also concentration of phosphorus. In the framework of this study we present results of magnesium and copper concentration changes to the TL signal after high-doses of electron radiation.

1. Introduction

The high-dose properties of the LiF:Mg,Cu,P phosphor, observed at the IFJ in 2006, manifesting as high temperature TL emission with maximum at about 400°C (denoted as the TL peak 'B') (Bilski et al., 2008, 2010; Obryk et al., 2009, 2010; 2011a), allow for the dose calibration with this phosphor in the kilogray range [Obryk et al., 2011b; Obryk, 2013). Since LiF:Mg,Ti phosphor high-dose features are very limited (Bilski et al., 2010, Khoury et al., 2011), therefore it is believed that the dopants play the lead role in this phenomenon (Obryk et al., 2014, Remy et al., 2016). Until now, studies of dopants concentration influence on TL emission were conducted in detail for the LiF based phosphors in the mGy dose range, where TL glow-curve is a typical one without the high-temperature TL peak 'B' presence (e.g. Bos et al., 1996; Bilski et al., 1996, 1997; Chen and Stoebe, 1998, 2002; Lee et al., 2008; Patil and Moharil, 1995; Shoushan, 1988; Tang et al., 2008).

In a typical LiF:Mg,Cu,P detector (Nakajima et al., 1978), there are three types of dopant: magnesium, copper and phosphorous. The most important dopant responsible for the formation of trapping centres is magnesium. It was observed that even small changes in the concentration of magnesium produce a significant change in the shape of the glow-curve and the intensity of the individual peaks for low dose range (e.g. Bilski et al., 1997). In the case of zero magnesium concentration practically only peak 4 (with maximum intensity at about 220°C) is observed. It is also noted that the area under the peak at the right side of the glow-curve increases with increasing magnesium concentration reaching a

* Corresponding author.
E-mail address: barbara.obryk@ifj.edu.pl (B. Obryk).

http://dx.doi.org/10.1016/j.radmeas.2017.03.003
1350-4487/

maximum for a certain concentration of magnesium. However, the value of this maximum is related to the concentration of copper in the detector. In addition, it was observed that the proportion of the low-dose higher temperature peaks to the total TL signal rises nearly in proportion to the concentration of magnesium in the detector (Bilski et al., 1996).

The role of copper as activator, despite a significant effect on the TL signal, even at low doses is not entirely clear. The concentration of copper has a strong influence on all the peaks of the glow-curve, both peak 4 and higher temperature peaks for low doses. It was observed that the response of the TL peak 4 increases with the increase of copper concentration to a value of 0.02 mol%, and then decreases at higher concentrations (Bos et al., 1996). For detectors with zero copper content it appears that the higher temperature peaks reached the maximum, and increase in concentration reduces the amount of these peaks for low doses. It is also noted that the same level of higher temperature peaks are obtained with much lower concentration of copper, when the content of magnesium is reduced accordingly. Therefore, the effect of copper appears to be dependent on the magnesium content, copper can inhibit the effect of magnesium excess in the detector (Bilski et al., 1997).

Phosphorus is necessary to achieve high intensities of the peak 4 for low doses. This means that the intensity of the peak 4 depends on both the magnesium and phosphorus. With increasing concentration of phosphorus the amplitude of the signal increases to a threshold value. Above this threshold there is no correlation between the phosphorus content and the intensity of the peak 4 at low doses (Bilski et al., 1996).

All mentioned above concern low-dose range studies where the high-temperature peak 'B' is not present. A systematic investigation of the high-dose (kGy range) TL properties of LiF:Mg,Cu,P samples with different concentration of all activators has been performed at the IFJ recently. The effect of varying content of one of three dopants sequentially, namely magnesium, copper and phosphorus, while maintaining the typical concentration of the other two dopants in each case, have been studied. About thirty batches of sintered chips with dopants varied over the following ranges: 0–0.4 mol% Mg, 0–0.1 mol% Cu, 0–1.875 mol% P, were produced and comprehensive studies of their high-dose TL features in the different radiation fields (Co-60 gammas, 6 MeV electrons, 23 GeV protons, and reactor neutrons) have been performed. In this way, the impact of the type of dopants and their concentration on the occurrence and characteristics of the high-dose high-temperature TL peak 'B' has been verified. Comprehensive results of these studies will be presented in upcoming articles. In this work, being the first one of a series planned, we present results of magnesium and copper concentration changes to the TL signal after high-doses of electron radiation.

2. Materials and methods

2.1. Materials and synthesis

All detectors' batches have been produced at the IFJ in Kraków using sintering method. First lithium fluoride was synthesized in chemical processes between LiCl (AlfaAesar 99% reagent grade) and HF >48% (Sigma-Aldrich, puriss p. a.). Obtained LiF powder was thoroughly mixed with activators of the following forms: $MgCl_2$ solution (obtained from magnesium oxide POCH S.A. pure p.a. with fuming hydrochloric acid for trace analysis by Sigma-Aldrich), $CuCl_2$ solution (obtained from cooper oxide POCH S.A. pure p.a. with fuming hydrochloric acid for trace analysis by Sigma-Aldrich) and H_3PO_4 (POCH S.A> pure p.a.). The dopants were added in concentration related to LiF listed in Table 1. The pellets of 4.5 mm diameter and 0.9 mm thickness have been formed mechanically from each powder type. The last stage was sintering of pellets at a temperature between 600 and 700°C in gas atmosphere and platinum containers for a certain period of time, as due to this stage LiF:Mg,Cu,P phosphor gains higher sensitivity. Two step pre-irradiation annealing of all samples has been applied as routinely for LiF:Mg,Cu,P detectors, i.e. 260°C for 10 min followed by immediate cooling and then 240°C for 10 min finalized with immediate cooling also.

Table 1
The concentration of individual dopants for the studied detectors' batches.

Sample's mark	Dopants' concentration		
	Mg^{2+} [%mol]	Cu^{2+} [%mol]	PO_4^{3-} [%mol]
Ref	0.2	0.05	0.625
0M	0	0.05	0.625
0.5M	0.1	0.05	0.625
2M	0.4	0.05	0.625
0C	0.2	0	0.625
0.5C	0.2	0.025	0.625
2C	0.2	0.1	0.625

2.2. Irradiation and measurements

Detectors were exposed using an electron linear accelerator (Linac) at the Laboratory of Linear Electron Accelerator at the Institute of Applied Radiation Chemistry of the Lodz University of Technology, Lodz, Poland. Detectors have been exposed with 6 MeV electron beam with dose rate of 6.1 kGy/min. The distance from the output window of the accelerator was 160 cm. In order to analyse the high-dose changes in the glow-curve for different types of detectors ten steps of doses have been selected (i.e. 1, 5, 10, 20, 30, 50, 100, 200, 500, 1000 kGy) and applied. Dosimetry was performed during the irradiation by the radiochromic foils.

The readout were made using Harshaw 3500 manual TL reader. The reading conditions for all detectors were the same: temperature range from 100°C to 550°C, a linear heating rate of 2°C/s, high-dose filter applied (CVI Melles Griot Laser Optics) which task was to reduce the signal reaching the photomultiplier in order to avoid its' malfunction due to not sufficient dynamic of its response.

3. Results and discussion

For each production batch the low-dose sensitivity has been evaluated. For this purpose several detectors from each production batch have been selected and irradiated with a dose of 1 mGy, using Cs-137 gamma source. In addition, reference LiF:Mg,Cu,P detectors were exposed simultaneously. On the basis of average TL signal values for detectors of each batch and for the reference detectors the sensitivity was determined as the ratio of these two values. From the viewpoint of low-doses the batch with sensitivity closest to the reference detectors is the one with copper concentration reduced 50% to the standard one. Other batches are not showing sufficient low dose sensitivity in comparison to the reference MCP-N phosphor, as its value vary from 37.6% for 0C batch to 71.0% for 2C (only 0M batch low dose sensitivity is not acceptable being 0.2% only).

The impact of the type of dopants and their concentration on the occurrence and characteristics of the high-dose TL signal has been verified. Fig. 1 presents the TL glow-curves for the batches of detectors listed in Table 1 after exposures to selected doses (i.e. 1, 10, 100, 1000 kGy). At a dose of 1 kGy minimum intensity in relation to the reference detectors' glow-curve exhibit 0C, 0M and 0.5C batches. TL peak intensity for the other detectors is slightly higher