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High-dose high-temperature emission of LiF:Mg,Cu,P: Thermally and radiation induced loss & recovery of its sensitivity

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HIGHLIGHTS

► High-dose high-temperature TL emission is present in LiF:Mg,Cu,P' glow-curves.
► LiF:Mg,Cu,P can measure doses ranging from below 1 μGy to about 1 MGy.
► Thermally-induced sensitivity loss of the samples can be recovered.
► Sensitivity damage of the samples after high-dose measurements is fully reversible.
► High-dose measurements changes to the structure of the material are reversible.

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ABSTRACT

Highly sensitive LiF:Mg,Cu,P (MCP) detectors enable measurements of radiation doses from tens of nanograys up to a few kilograys, where the saturation of the signal of the main dosimetric peak occurs. Thanks to the recently observed high-dose high-temperature emission of MCP detectors heated to temperatures up to 600 °C after exposures to radiation doses ranging from 1 kGy to 1 MGy, a new method of thermoluminescent measurement of radiation doses has been recently developed at the Institute of Nuclear Physics (IFJ). This method can measure doses ranging from micrograys up to a megagray. So far, high dose measurements were performed on fresh MCP samples and each detector was used only once, because as a result of these measurements, the detectors lose their sensitivity to a large extent. In this study, a specific thermal treatment intended to fully restore the loss of MCPs TL sensitivity was sought. We have investigated several annealing procedures, applying different temperatures (from 400 °C up to 700 °C) for different periods of time (10–30 min) in argon atmosphere. In this way we were able to recover MCP sensitivity fully, allowing for reuse of the samples after high-dose irradiation and high-temperature measurement.

1. Introduction

One of the well known advantages of thermoluminescent (TL) detectors made of lithium fluoride doped with magnesium, copper and phosphorus is their very high sensitivity to ionizing radiation (Horowitz, 1993; Bos, 2001; Bilski, 2002). LiF:Mg,Cu,P (MCP) detectors enable measurements of radiation doses from tens of nanograys up to a few kilograys, when the total saturation of the signal of the so-called main dosimetric peak (at about 220 °C) occurs (McKeever et al., 1995).

Only recently, at the Institute of Nuclear Physics (IFJ), the quite unexpected properties of MCP detectors at high (Bilski et al., 2007) and ultra-high doses (Bilski et al., 2008b; Obryk et al., 2009) have been observed. Significant changes of the glow-curve shape occur for doses higher than a few kilograys and the most important finding is that a new, intense peak appears for doses above 30 kGy (see Fig. 1). This peak, denoted as peak 'B', is well separated from the rest of the glow-curve and located at temperatures exceeding 400 °C. After this discovery, comprehensive measurements of MCP detectors response to high doses of various radiation types: e.g. photon (Obryk et al., 2009), electron (Bilski et al., 2010), proton (Obryk et al., 2010) and thermal and epithermal neutron (Obryk et al., 2011b), were completed (see Table 1). All the results showed the presence of the peak 'B' in the glow-curves of detectors exposed to radiation doses higher than 30 kGy.

Although high-dose high-temperature emission was found to be present in MCP' glow-curves after irradiation with all radiation

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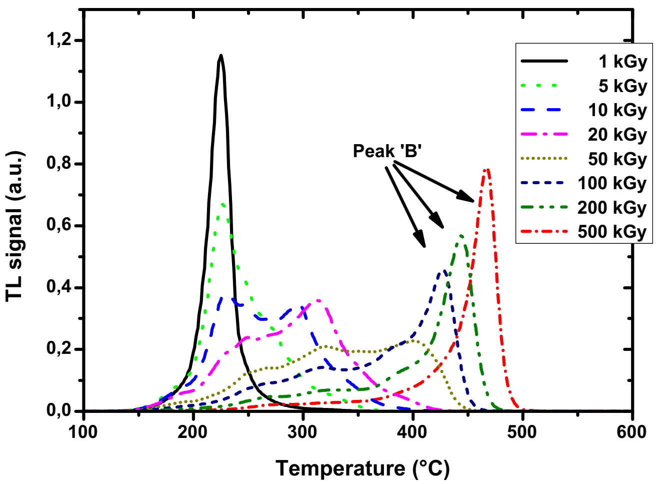

Fig. 1. MCP glow-curves resulting from gamma irradiation (^{60}Co source at KAERI) for the dose range 1 kGy–500 kGy.

qualities, also following irradiation in mixed fields, the mechanism leading to the peak ‘B’ occurrence is still under investigation. A recent study of peak ‘B’ spectra is analysed by Gieszczyk et al. 2013.

On the basis of this newly discovered behaviour of MCP at high and ultra-high doses, a method of TL measurement of radiation doses ranging from micrograys up to a megagray has been recently developed at the IFJ (Obryk et al., 2011a, 2011c; Obryk, in press). For dose measurement up to 1 kGy we exploit routine TL dosimetry which uses main dosimetric peak (peak 4) intensity, applying experimentally determined correction functions in the region of its sublinearity (Obryk et al., 2008). For doses higher than 1 kGy where significant changes of the glow-curve shape were observed the method is based on the percentage of TL signal in different temperature ranges. A parameter called the Ultra-High Temperature Ratio (UHTR) was defined in order to quantify the observed changes of the MCP glow-curve shape at very high doses and very high temperatures, which allows measuring the absorbed dose in the range from 1 kGy to 1 MGy (Bilski et al., 2010; Obryk et al., 2010). This dosimetric method was tested in a range of radiation qualities, such as gamma radiation, electron and proton beams, thermal neutron fields and high-energy mixed fields around the Super Proton Synchrotron (SPS) and the Proton Synchrotron (PS) accelerators at CERN (the European Organization for Nuclear Research) (Obryk et al., 2008, 2011c). This method allows for ultra-high dose range (at least twelve orders of magnitude) measurement with a single MCP detector. It can be used for dosimetry at high energy accelerators, thermonuclear fusion technology facilities and has great potential for accident dosimetry in particular. A number of dosimetric sets with TL detectors are currently used around the Large Hadron Collider (LHC) at CERN.

So far, high dose measurements were performed on fresh samples and each detector was used only once, because the detectors lose their sensitivity to a large extent as a result of these measurements. The main reason is the well-known feature of LiF:Mg,Cu,P detectors: sensitivity loss when heated beyond about 270 °C (Oster et al., 1993, 1996; Bilski et al., 1997; Ben-Amar et al., 1999). It was also reported by Meijvogel and Bos (1995) that high temperature readout is causing both reversible and irreversible changes of the sensitivity of the material. Also, high doses of radiation usually have some influence on various properties of the samples, among them their sensitivity (Cai et al., 1994; Bilski et al., 2008a). Following high-dose high-temperature measurements these two effects are entangled.

In this study, a specific thermal treatment intended to fully restore the loss of MCPs TL sensitivity was sought. We were encouraged by our preliminary observation that readout up to 600 °C caused smaller loss of samples' sensitivity than readout up to 400 °C, which is generally in accordance with the results of Tang et al. (2000) who showed that the change in glow-curve structure and the loss of TL sensitivity, as a consequence of annealing between 260 °C and 400 °C, can be recovered fully by annealing at 720 °C for 30 min in a nitrogen atmosphere followed by the standard anneal of 10 min at 240 °C.

Table 1
Qualities, energies and dose ranges of radiation used for tests of high-dose high-temperature emission of LiF:Mg,Cu,P detectors.

Radiation type	Radiation energy	Dose/fluence range	Reference
Gamma	1.25 MeV	1 Gy–1 MGy	Bilski et al., 2007, 2008b; Obryk et al., 2009
Electron	6 MeV, 10 MeV	5 kGy–1 MGy	Bilski et al., 2010; unpublished results
Proton	25 MeV, 24 GeV/c	1 Gy–1 MGy	Obryk et al., 2009; Obryk et al. 2010
Neutron	Thermal & epithermal	3×10^{11}–3×10^{15} n/cm^2	Obryk et al., 2011b

2. Materials and methods

Over one thousand of highly sensitive MCP detectors, made using the sintering technique were prepared for the tests. All detectors used were of typical size: 4.5 mm diameter and 0.9 mm thickness. Detectors were produced at the Department of Radiation Physics and Dosimetry of the IFJ and belonged to one production batch with TL sensitivity homogeneity below 5%.

At each stage of the experiment the standard annealing cycle was applied: for freshly produced detectors two-phase heat treatment – 260 °C for 10 min followed by 240 °C for 10 min, with fast cooling using a thick metal block after each phase, while for detectors used previously one-phase heat treatment only – 240 °C for 10 min followed by fast cooling. The individual response factor (IRF) has been also determined for each freshly produced detector using its total TL response. Irradiations have been carried out at the Secondary Standards Dosimetry Laboratory (SSDL) of the IFJ with Cs-137 source.

All readouts were performed using the MICROLAB Manual Reader-Analyzer RA'04, with a bialkali photomultiplier tube and violet filter (BG-12). Detectors were readout in argon atmosphere with a linear heating rate of 2 K s^{-1}. All high-temperature annealing were also carried out in argon atmosphere to avoid oxidizing of detectors' surface.

To quantify the observed changes of the glow-curve two methods of analysis have been applied. The first one consists of analysis of the main peak height, the main peak signal integral in the range of 200–230 °C (integral 1), integral of the ‘tail’ of the glow-curve calculated in the range of 230–275 °C (integral 2), the total TL signal (for the range 150–275 °C), and also the ratio of integral 1 to integral 2 which we have called ‘shape’ parameter. The second method applied was deconvolution of all resulted glow-curves into five peaks by the GlowFit (Puchalska and Bilski, 2006) and analysis of behaviour of individual peaks. All parameters used for sensitivity analysis are presented relative to the parameters for the reference group treated routinely, i.e. as ratio of their mean values calculated for each group of detectors treated in different way during experiments to that of reference group.