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On LiF:Mg,Cu,P and LiF:Mg,Ti phosphors high & ultra-high dose features

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HIGHLIGHTS

- TL peak 'B' occurs for LiF:Mg,Cu,P after high doses of all radiation types.
- Highly irradiated LiF:Mg,Ti glow-curve shape depends on the radiation quality.
- High dose features of both phosphors are significantly different in many aspects.
- Associated PL/TL high-dose measurements are possible using LiF:Mg,Cu,P.
- Dopants role is crucial for high-dose features of lithium fluoride based phosphors.

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ABSTRACT

LiF:Mg,Ti and LiF:Mg,Cu,P are well known thermoluminescence (TL) dosimetry materials since many years. A few years ago their properties seemed well known and it was widely believed that they are not suitable for the measurement of doses above the saturation level of the TL signal, which for both materials occur at about 1 kGy. The high-dose high-temperature TL emission of LiF:Mg,Cu,P observed at the IFJ in 2006, which above 30 kGy takes the form of the so-called TL peak 'B', opened the way to use this material for measuring the dose in the high and ultra-high range, in particular for the monitoring of ionizing radiation around the essential electronic elements of high-energy accelerators, also fission and fusion facilities, as well as for emergency dosimetry. This discovery initiated studies of high and ultra-high dose characteristics of both these phosphors, which turned out to be significantly different in many aspects. These studies not only strive to refine the method for measuring high doses based on the observed phenomenon, but also, and perhaps above all, bring us closer to understanding its origin and essence. This manuscript aims to review existing research data on the high and ultra-high dose features of both LiF based phosphors.

1. Introduction

A big challenge of dosimetry nowadays is the growing demand for high-level dosimetry materials and methods due to intensive development of radiation technologies (among them materials testing, sterilization and processing) and nuclear installations (high-energy accelerators for research, e.g. Large Hadron Collider – LHC, and hadron therapy of cancer, also fission and fusion power facilities, e.g. International Thermonuclear Experimental Reactor – ITER) including accident dosimetry at these facilities (Bilski et al., 2007a). In connection with these growing needs the development of high-level dosimetry methods occurred and still is progressing (Schönbacher et al., 2009). Their number is very large, some are more universal, but most is useful in some special applications (Benny and Bhatt, 2002; Göksu et al., 1989; McLaughlin, 1996; Teixeira and Caldas, 2012; Wieser and Regulla, 1989). Among the most popular passive systems usable at high doses are polymer-alanine dosemeters, radio-photoluminescence glass detectors, optical absorption dosemeters (e.g. LiF single crystals, dyed polymeric foils), liquid chemical dosemeters (e.g. Fricke dosemeter) and hydrogen pressure detectors. In addition calorimetry and ionisation chambers are still used; however, new developed active systems are based on semiconductor p-FET or MOS-FET

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detectors (field effect transistors based on silicon dioxide), PIN diodes (p–n junction with built-in intrinsic undoped layer), also optically stimulated luminescence and diamond based detectors.

It is worth mentioning that none of the known high-level dosimetry materials and methods include both the dose range typical for radiation protection (miligrays) and high-dose range (kilograys). Surprisingly, to meet this challenge, came forth the discovery of unexpected properties of highly sensitive LiF:Mg,Cu,P phosphor at high doses and high temperatures (Bilski et al., 2008b; Obryk et al., 2009; Obryk, 2010). It was very fortuitous and has enabled the development of a method of measurement in the dose range of twelve orders of magnitude with a single thermoluminescent (TL) detector (Obryk et al., 2011a; Obryk, 2010, 2013). Recently observed features of this highly sensitive TL material inspired further research on high and ultra-high dose characteristics of both lithium fluoride based phosphors routinely used for TL dosimetry: LiF:Mg,Cu,P and LiF:Mg,Ti.

2. Materials and current status of research

2.1. LiF based phosphors general features

Lithium fluoride based phosphors have long been well known luminescent dosimetric materials (e.g. McKeever et al., 1995; Vij, 1993). LiF:Mg,Ti (MTS, equivalent to TLD-100) is used for the production of thermoluminescent detectors widely applied in dosimetry of ionizing radiation from the 1960s by the dosimetry services worldwide (e.g. Stadtmann et al., 2011; Obryk et al., 2011c). LiF:Mg,Cu,P (MCP, equivalent to TLD100H) was introduced in 1980s (Nakajima et al., 1978), and due to its high sensitivity is now equally widely used in TL dosimetry, especially for environmental measurements (e.g. Budzanowski et al., 2004; Ilgner et al., 2010). Main dopant concentration (related to trapping centres), magnesium for both materials, is ten times higher for MCPs (about 0.2 weight %) than for MTS, while the role of luminescence activator is played by titanium for MTS (10–15 ppm) and by phosphorus for MCP (1–4 weight %); in addition copper (0.02–0.05 weight %) for MCP plays a role still not completely clear, also the oxygen impurities seem to be important for MCP (Bilski, 2002; Chen and Stoebe, 1998, 2002). The glow-curve structure for both materials is quite similar, the main dosimetric peak (peak 5 for MTS while peak 4 for MCP) occurs at a temperature of about 220°C, preceded by a few minor peaks. The main peak's activation energy and frequency factor are similar for both materials ($E > 2$ eV; $s > 10^{20}$ s^{-1}), which implies similarity of TL processes related to both main glow peaks of these phosphors (Bilski, 2002; Horowitz, 1993). Both phosphors emit light in the short-wavelength part of the spectrum (McKeever et al., 1995), but emission bands differ, being in the range 420–460 nm for MTS while ca. 380 nm for MCP, which TL spectral characteristic have been thoroughly investigated by McKeever in 1991.

The most apparent difference between both phosphors is in their sensitivity to radiation: approximately thirty times higher for LiF:Mg,Cu,P than for LiF:Mg,Ti for gamma radiation (Horowitz, 1993; McKeever et al., 1995; Bos, 2001; Bilski, 2002); taking into account the high MTS internal background (i.e. TL signal of unexposed detector) the ratio grows to the level of about hundred. The detection threshold of MCP detectors is below 1 μGy while for MTS in the range of 20–50 μGy. Linearity range for both materials end up at a few Gy, while the upper limit of a useful dose range (manifested by a decrease of dose sensitivity to an unacceptable value), i.e. saturation dose is about 1 kGy. Another important difference between the dosimetric properties of these phosphors is in their dose response. MTS features the well-known linear-supralinear response, while MCP dose response is linear-sublinear. The sublinear dose response of MCP has some further consequences. It is generally accepted that this is responsible for the much lower TL efficiency with which heavy charged particles and high-LET particles are detected by MCP (Bilski, 2006). Dosimetry with LiF based phosphors outside the linearity limit (using experimentally determined non-linearity correction functions) was so far possible only up to 1 kGy, which is the level of saturation of the TL main peak for both materials. The high-dose high-temperature TL emission of MCP recently observed at the IFJ enabled dose measurement of up to 1 MGy (Obryk et al., 2011a), which is impossible with MTS. High-dose features of both phosphors have been determined experimentally since the discovery of TL peak 'B' of MCPs and most of them seem to be significantly different.

2.2. High-dose experiments

Until now TL emission glow-curves of highly irradiated LiF:Mg,Cu,P and LiF:Mg,Ti detectors have been studied with radiation qualities of a relatively broad LET range: gammas (^{60}Co), electrons (6 and 10 MeV), protons (25 MeV and 24 GeV/c), also after exposures to high thermal neutron fluences (up to 3×10^{15} n/cm^2), alpha particles (in the range 10^7–10^{11} particles/cm^2), low energy heavy ions (10^7–10^9 particles/cm^2) and in high energy mixed field which consisted of charged hadrons, muons, neutrons as well as photons and electrons with energy spectrum extending from fractions of eV to several hundreds of GeV (up to 10^{15} HEH/cm^2). TL emission spectra of highly irradiated detectors of both materials, their photoluminescence (PL), optically stimulated luminescence (OSL) and the preliminary characterization of their electron paramagnetic resonance (EPR) signals after high and ultra-high doses have been also studied. In addition thermally- and radiation-induced sensitivity loss and recovery of detectors have been investigated. The data on high-dose experiments with LiF based detectors are summarized in Table 1 together with references to their results, which are described in detail in the next section.

3. Characteristics of highly irradiated LiF:Mg,Cu,P and LiF:Mg,Ti phosphors

The basic difference between the typical TL low-dose glow-curve of LiF:Mg,Cu,P, structure of which doesn't change up to 1 kGy,

Table 1
Qualities, energies and dose ranges of radiation used for tests of high-dose high-temperature emission of LiF based detectors so far.

Radiation type	Radiation energy	Dose/Fluence range	Reference
Gamma	1.25 MeV	1 Gy–1.2 MGy	Bilski et al., 2007b, 2008b; Obryk et al., 2009; Obryk, 2010; Khoury et al., 2011; Gieszczyk et al., 2013b; 2013c
Electron	6 MeV, 10 MeV	5 kGy–1 MGy	Bilski et al., 2010; Obryk, 2010; Mrozik et al., 2014
Proton	25 MeV, 24 GeV/c	1 Gy–1 MGy	Obryk et al., 2009, 2010; Obryk, 2010
Neutron	Thermal & epithermal	3×10^{11}–3×10^{15} n/cm^2	Obryk, 2010; Obryk et al., 2011b
Alpha-particle	5.5 MeV	1×10^7–1×10^{11} α/cm^2	Olko et al., 2011; Gieszczyk et al., 2012
Low energy heavy ion	5.0–9.3 MeV/n	10^5–10^9 particles/cm^2	Gieszczyk et al., 2013a; Gieszczyk et al., 2014
Mixed field	>20 MeV, HEH	Up to 10^{15} HEH/cm^2	Obryk et al., 2008; Obryk, 2010; Mala et al., 2014