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Photoluminescence of gamma-, proton- and alpha-irradiated LiF detectors*

Research Article

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Abstract: Lithium fluoride (LiF), one of the most pervasive alkali halides in optical device research, is routinely used in optical data storage and radiation protection. LiF crystals may contain different aggregate defects produced by several types of ionizing radiation, with the number of defects being proportional to the cumulative radiation dose. Stimulation of irradiated LiF detectors by heating or with blue light causes thermoluminescence (TL) or photoluminescence (PL), respectively. We developed a new PL reader equipped with a blue light-emitting diode for stimulation and a Hamamatsu photomultiplier for registering green emissions, dedicated to examining LiF detectors as well as more broadly investigating TL/PL emission from standard LiF detectors irradiated with gamma rays, 60 MeV protons and alpha particles. The results confirmed very high efficiency PL signal from alpha-irradiated LiF detectors corresponding to their low efficiency after gamma irradiation, and vice versa for TL readout. Combining the TL and PL readouts permits us to discriminate between how different kinds of radiation affect efficiency in LiF detectors.

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Keywords: LiF • photoluminescence • thermoluminescence • dosemeters

1. Introduction

Thermoluminescence (TL) and Optically Stimulated Luminescence (OSL) are very well known passive methods for measuring radiation dose, and lithium fluoride (LiF) detectors are the most popular detectors routinely used in TL dosimetry. LiF is also considered a promising optical material due to its specific physical and optical properties [1, 2]. In its solid form, LiF is hard and almost insoluble in water. It has a short cation-anion distance, Li^+ and F^- ions possess the smallest radius among the alkali and halide ions, respectively. LiF can posses color centers (CCs) generated during exposure to ionization radiation. Ionising radiation can simultaneously generate various kinds of primary and aggregate color centers. The simplest CC, called an F center, is formed by an electron trapped in an anion vacancy. In the structure of LiF two centers play the most significant role: F_2 and F_3^+. The

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F_2 center occurs when one F center is aggregated to another F center, and F_3^+ is an ionized F_3 center consisting of two electrons bound to three anion vacancies. A typical absorption spectrum of a LiF crystal after irradiation has its first intense band centered at 250 nm due to F centers, and an M band centered at 442nm, which is composed of F_2 and F_3^+ bands [3–8]. Excitation with 442 nm light causes luminescent emission peaks at several different wavelengths. Of these, two are the most important for dosimetric applications: namely those at 535 nm and 645 nm [9]. The M-center population grows almost linearly with increasing radiation dose, so M-luminescence can be used in radiation dosimetry. On this basis, the Sunna 535 nm photo-luminescent film dosimeters [9], consisting of LiF and a polymer, were developed which are widely used as high-dose dosimeters for food irradiation or medical sterilization. In this case, we could measure the emission of 535 nm green light, and block the 645 nm band using a filter.

Oster et al. have recently reported [10, 11] the presence of OSL emission in LiF irradiated with high-ionization density radiation (the effect should is really photoluminescence rather than OSL, as blue light stimulation induces green light emission, as opposed to OSL situations in which the emission is at a wavelength shorter than the stimulation light). Oster et al. presented the stimulation and emission spectra of PL, as well as TL, both studied as a function of irradiation dose/fluency for both beta and alpha particles. The TL processes and light-induced effects are connected with different types of traps, and are therefore are independent [10, 11]. This conclusion offers a possibility to discriminate between components of a radiation field by combining the TL and PL readouts, and exploiting different efficiency of both processes in LiF detector to various modalities of radiation.

The aim of our work was to realize this idea by developing a special, dedicated device — a small PL reader optimized to PL measurements of LiF and to study thermal efficiency — and PL of LiF detectors irradiated with radiation of various modalities.

2. Materials and methods

The standard LiF detectors activated with Mg and Ti (LiF:Mg,Ti) were used as the tested material [12]. The detectors were manufactured at the IFJ PAN in Kraków as pellets, 4.5 mm in diameter and 0.9 mm thick. All detectors were annealed at 400°C for 30 minutes before irradiation. The pellets were gamma-irradiated with a Cs-137 source with doses ranging from 5 to 225 Gy. For doses up to 100 Gy, we used the 60 MeV proton beam from the AIC-144 cyclotron at the IFJ PAN. Alpha irradiations were achieved by exposing the LiF detectors to an isotopic Am-241 source at a fluence rate of 1.7×10^4 s^{-1} cm^{-2} alpha particles, for periods ranging from a few hours up to 14 days, to accumulate fluences up to 10^{10} cm^{-2}.

The TL measurements were done with a standard TL reader equipped with a bialkali photomultiplier at temperatures ranging from 40°C up to 400°C, with the temperature increasing at 2°C/sec.

The spectral photoluminescence (PL) measurements were conducted at the Jan Długosz University (AJD) in Czestochowa. These measurements were carried out in an optical cryostat. The samples were stimulated using a 75 W xenon arc lamp installed in a high-intensity illumination system (PTI). We monitored luminescence using an Acton SP150 spectrograph connected to the LN/CCD-1024E camera (Princeton Instruments, spectral range 190–1080 nm). The CCD camera is cooled with liquid nitrogen to reduce the dark current. The samples during PL measurements were excited by blue light (433 nm) in air and room temperature. To reduce stray light, the Knight Optical filter 460FIB12 was used for stimulation, and the filters 550FIR25 (Knight Optical) and GG495 (Schott) were used for detecting PL emission.

3. Experiments and discussion

Testing the effects of different efficiency of LiF detectors for the various kinds the radiation had to be done with a TL reader and an optical reader adapted to the stimulation of LiF luminophore. Since TL readers are easily available as standard equipment in our laboratory, we concentrated on constructing a special reader dedicated to our LiF investigations. As is now well known ([2, 9]), LiF stimulated blue light produces a green emission. Oster's results [10, 11], and also our own preliminary measurements, indicated that the 535 nm emission band is sensitive to alpha particles, while 645 nm band does not distinguish between radiation types and represents mostly a non-radiation induced signal.

Our first goal in optimizing the measurement conditions was therefore to choose proper filtering. We tested several normal and interference optical filters for both the stimulation and emission wavelength controls, using spectroscopic equipment at the AJD in Częstochowa. These tests were performed using a batch of LiF detectors irradiated at IFJ PAN in Kraków with gamma rays, protons and alpha particles. We selected the most appropriate filters, and estimated the optimal stimulation wavelength and emission spectrum range during preliminary tests. Fig. 1 presents the results of stimulating LiF with

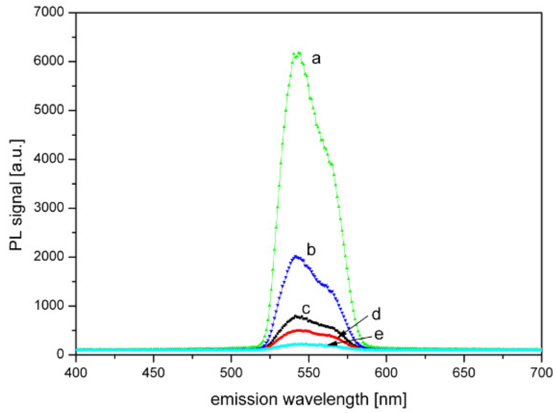

Figure 1. Spectrally resolved PL signal for LiF detectors irradiated with alpha particles (a — 1.2×10^{10} cm^{-2}, b — 1.7×10^{9} cm^{-2}) and gamma rays (c — 180Gy, d — 5Gy) during stimulation with a blue light (433 nm) with an application of two filters 550FIR25 (Knight Optical) and GG495 (Schott) at the emission. Curve e represents the background level.

blue light (433 nm), after irradiation with 5 Gy and 180 Gy of gamma rays, and with alpha particles to a fluence of 1.2×10^{10} cm^{-2} and 1.7×10^{9} cm^{-2}. Applying optical filters limited the spectrum to the short range 520–580 nm and cut off the long wavelength peak at 645 nm. On the basis of these results, we constructed a new PL reader called HELIOS-2 (to distinguish it fromHELIOS-1 also designed at AJD, which was optimized for work with Al_2O_3 detectors [13]). HELIOS-2 consists of a blue diode, two interference filters produced by Knight Optical company, namely 460FIB12 for the excitation, and 532FIB25 for emission, together with the Schott OG515 filter and the Hamamatsu H8259 photon-counting head. HELIOS-2 is a compact, portable PL reader with very user-friendly software, similar in construction to standard TL readers. Relative transmittance characteristics of optical filters, which were used for measuring PL emission and the characteristics of other filters used in PL reader are presented in Fig. 2

We used HELIOS-2 to examine LiF:Mg,Ti detectors irradiated with protons, alpha particles and gamma rays. Fig. 3 presents the PL signals of LiF detectors after irradiation with gamma rays (dose of 58 Gy) and alpha particles with a fluence of 1.9×10^{10} cm^{-2}. The PL signal after gamma irradiation is only 2% higher than the background, and 40% higher for the alpha-irradiated detectors. These measures confirm the high efficiency of PL effect for detectors exposed to radiation with a high ionization density. We observe also that the luminescence intensity does not change during stimulation, as is typical for PL (contrary to OSL, in which the number of trapped charge carriers

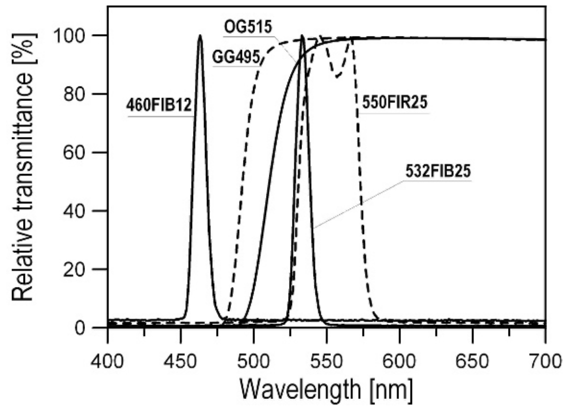

Figure 2. Relative transmittance characteristics of optical filters. Dashed lines represent 550FIR25 and GG495 filters, which were used for measuring PL emission (Fig. 1). The other filters, represented by solid lines, were used in PL reader: 460FIB12 for excitation and 532FIB25 together with Schott OG515 for emission.

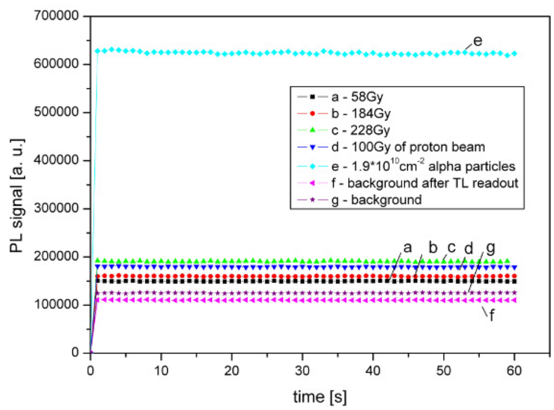

Figure 3. PL signal for LiF detectors irradiated with 58Gy, 184Gy and 228Gy of Cs-137 gamma rays, alpha particles with a fluence of 1.9×10^{10} cm^{-2} of Am-241 source and 100Gy of 60MeV protons beam.

decreases during stimulation).

The TL glow curves of the same detectors up to 400°C are presented in Fig. 4. All of the TL curves have typical shapes, but the main peak amplitude is several times higher for the gamma-irradiated detector than after alpha irradiation, contrary to PL. The PL reading must take place before the TL reading, because the heating process bleaches the PL effect, whereas the PL reading has no effect on the TL signal. Due to the different trap origins connected with the PL and TL effects, the combined readouts performed even on one detector can give some information about the radiation field.

To describe ability of detectors to discriminate between

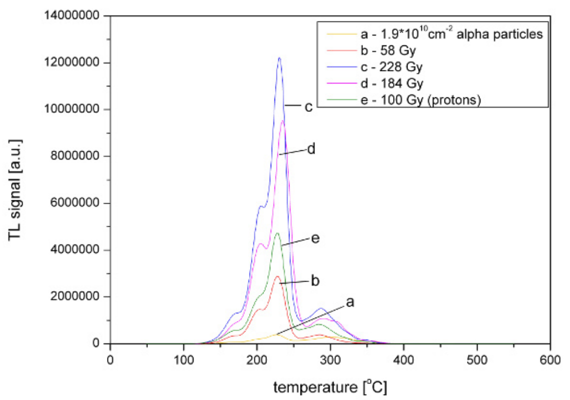

Figure 4. TL signal for LiF detectors irradiated with 58Gy, 184Gy and 228Gy of Cs-137 gamma rays, alpha particles with a fluence of 1.9×10^{10} cm^{-2} of Am-241 source and 100Gy of 60MeV protons beam.

radiation of high- and low-ionization density, Oster has defined a coefficient R [10]:

$$R = \left(\frac{_\beta I_{TL}}{_\alpha I_{TL}}\right) : \left(\frac{_\beta I_{PL}}{_\alpha I_{PL}}\right) = 500\,, \qquad (1)$$

where $_\beta I_{TL}$ – amplitude of TL peak after low-ionization density β, γ irradiation. $_\alpha I_{TL}$ – amplitude of TL peak after α particle irradiation. $_\beta I_{PL}$ – PL signal after low-ionization density β, γ irradiation. $_\alpha I_{PL}$ – PL signal after α particle irradiation.

Oster et al. reported R=500 [10], while we measured R=250, half of Oster's value. The coefficient R cannot, however, be treated as an absolute parameter, for it may depend on radiation type, dose, equipment and measurement conditions. Among all factors the most problematic one seems to be the high background during PL readouts, combined with a low gamma signal. The signals measured after relatively high gamma doses 50–225 Gy) are only a few percentage points higher than the background. The PL background level and its stability are therefore determining factors for this method's detection limit. In our case, the background level was estimated as a mean value from ten readouts performed in a reader with an annealed (but not irradiated) detector. We are continuing to work on minimizing the background problem, by exploiting time separation between radiation and non-radiation induced signals.

4. Conclusions

Our results confirmed the high efficiency of LiF photoluminescence after alpha particle irradiation, and very low efficiency of LiF photoluminescence irradiated with gamma rays or protons. Thermoluminesence has the opposite efficiency; high for exposure with gamma-rays, and low for alpha particles.

The newly developed dedicated PL reader, optimized for stimulation with blue light and measurement of green light emissions, is useful for combined PL/TL measurements aimed at discriminating between radiation types in mixed-radiation fields.

The largest constraint on our method's detection limit is the high background signal. Our further investigation will focus on decreasing the background, including checking time patterns of radiation and non-radiation induced signals and exploiting possible time separations between them.

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