



## Photoluminescence measurements of LiF TL detectors



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### HIGHLIGHTS

- ▶ Photoluminescence readouts of LiF detectors performed in a pulsed mode improved signal readability.
- ▶ First measurements of PL signal of LiF as a function of traps emptying temperature.
- ▶ Traps emptying temperature in PL effect depends on detector type and cumulated dose.

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### ABSTRACT

The properties of standard lithium fluoride (LiF) thermoluminescent (TL) detectors, which are routinely used in radiation protection systems, were investigated under light stimulation. The luminescence of different types of LiF detectors, which were irradiated with gamma rays of energy up to 300 Gy and alpha particles with a fluence up to  $5 \cdot 10^9 \text{ cm}^{-2}$ , were stimulated by a blue light and were heated up to temperature of 240 °C or 400 °C, depending on the type of detectors. The irradiated LiF detectors during the blue-light (460 nm) stimulation emit green photoluminescence (PL) with a wavelength of 530 nm. The LiF detectors showed a PL effect of much higher efficiency after they were irradiated with alpha particles than after they were irradiated with gamma rays. However, in contrast to PL, the TL readout showed a significantly lower efficiency of LiF detectors after alpha particle irradiation. These effects result from the different trap mechanisms that are responsible for TL and PL phenomena. The temperature stability of the traps responsible for the PL effect for both types of LiF detectors was studied.

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### 1. Introduction

Many researchers are interested in measuring the components of mixed radiation fields near nuclear reactors, LHC or in space. Lithium fluoride (LiF) detectors are the best known and most popular thermoluminescent (TL) dosimeters in radiation protection services. The dosimetric properties of highly sensitive LiF has been investigated for almost 40 years, and LiF detectors are still routinely applied in personal and environmental dosimetry (Bilski, 2002). LiF is also an interesting optical material that can be applied to active devices such as lasers. The luminescence of irradiated LiF detectors can be excited by heating up to 400 °C or by stimulation with a 460 nm blue light. The emission spectrum of LiF, which contains the color centers induced by radiation, consists of two broad bands at 535 nm and 640 nm. A new application of LiF luminofor occurred after the Sunna photo-luminescent film dosimeters were

developed based on the 535 nm emission band of LiF (Murphy et al., 2003). Sunna detectors in the form of films composed by a LiF powder mixed with a polymer are widely used as high-dose dosimeter in the dose measurements in food irradiation or medical sterilization.

In 2008, Oster et al. (2008) described the excitation and emission spectra of the OSL (Optically Stimulated Luminescence) and the TL of LiF detectors as functions of the irradiation dose or the fluence for beta rays as low ionization density radiation and for alpha particles as high ionization density radiation. The authors observed (Oster et al., 2008, 2010, 2011) that the TL processes and the light-induced processes are independent and not related to each other. The combined TL/OSL readout can discriminate different radiation fields due to the different efficiencies of the LiF detector to various modalities of radiation.

This work aims to investigate the photoluminescence (PL) and TL signals of LiF detectors that are irradiated with different ionizing density radiation, such as gamma rays and alpha particles, and to study the stability of the traps that are responsible for the PL effect.

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## 2. Material and methods

Standard LiF detectors, which were doped with Mg, Ti (MTS) and Mg, Cu, P (MCP) atoms, were used in the experiments. The detectors routinely used in many dosimetric systems have the shape of pellets with a diameter of 4.5 mm and a thickness of 0.9 mm.

MCP and MTS detectors were irradiated with gamma rays and alpha particles with doses in the ranges of 10–55 Gy and 10–300 Gy, respectively. The gamma rays were delivered from a  $^{137}\text{Cs}$  source (the dose rate range is  $0.016 \cdot 10^{-6}$ – $0.016$  Gy/min) and alpha particles from an  $^{241}\text{Am}$  source (the fluence range is  $0.4 \cdot 10^8$ – $50 \cdot 10^8$   $\text{cm}^{-2}$ ), which was installed in the Risoe TL/OSL reader.

Detectors were read in two steps: first, nondestructive PL, followed by the TL measurements. PL was measured using the HELIOS-2 reader, which is a portable PL reader, self-constructed in cooperation with Jan Długosz University (JDU) in Częstochowa and adapted to the measurements of PL signal of LiF detectors (Mandowski et al., 2010; Marczevska et al., 2012). The detectors were excited using a blue diode (light power 5 W), which emits light with a wavelength of 460 nm. HELIOS-2 is equipped with a Hamamatsu H8259 “photon counting” gating photomultiplier and interference filters produced by Knight Optical company, namely 460FIB12 for excitation and 532FIB25, 550FIR25, and 550FIW25 for emission.

HELIOS-2 can be used in the continuous-wave mode and the pulsed modes of stimulation. The schematic of the measurement cycle in the boxcar pulse mode is shown in Fig. 1. A PL readout cycle was performed with the following parameters: the stimulation time, using a diode, was 5000 ms ( $T_2$ ), and the readout time was 10 ms ( $T_{\text{period}}$ ).  $T_{\text{period}}$  was divided into 100 readout frames; thus, the measurement time per frame was 0.1 ms ( $t_f$ ). Due to technical limitations of the reader construction, 0.1 ms ( $t_f$ ) is the shortest time per frame that can be achieved. The cycles were repeated 100–1000 times and summed up to obtain better statistics of the results. The diode current was 6.25 mA. The TL readouts were performed at the RA'94 TL reader at a temperature range of 50–240 °C and 50–400 °C with a rate of 2 °C/s for MCP and MTS detectors, respectively.

## 3. Results

### 3.1. Boxcar pulsed mode

One of the first steps of this work was to select the optimal readout parameters for the HELIOS-2 reader. As previously mentioned, the current construction of the PL reader does not allow a  $t_f$  shorter than 0.1 ms. Fig. 1 shows the schematic of the measurement cycle, where  $T_1$  is the delay time before the pulse,  $T_2$  is the pulse duration,  $T_3$  is the delay time after the pulse,  $T$  is the duration of one cycle,  $N$  is the number of frames (boxcars) in one cycle,  $t_f$  is the frame duration, and  $R$  is the number of repetition cycles.

Fig. 2 shows the PL signal normalized to  $t_f$  for different frame durations with the same duration of one cycle. As shown in Fig. 2, a shorter  $t_f$  gives more measurement points in one cycle. The information of the dose is encoded mainly in the first measurement

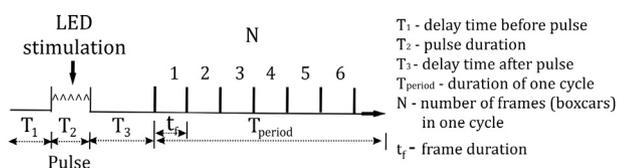


Fig. 1. Schematic of the measurement cycle of HELIOS-2 reader in the boxcar pulsed mode.

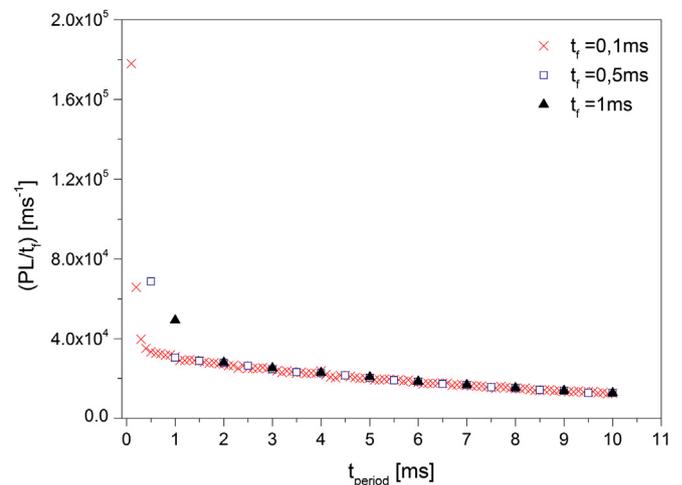


Fig. 2. PL signal normalized to  $t_f$  for different frame durations (MCP detector irradiated with  $^{137}\text{Cs}$  – 42 Gy).

point, and the remaining measurement points are close to the background. The sum of counts in the first ten frames for  $t_f = 0.1$  ms equals the number of counts in the first box for  $t_f = 1$  ms. The dose information can be easily obtained from the first measurement point for  $t_f = 0.1$  ms, and it is not necessary to extend the measurement time. The high level of background is a severe problem in PL readouts, whereas the OSL method provides a signal that is clearly separated from the background. The high level of background does not allow one to discriminate doses lower than 10 Gy, but the application of electronics with better time resolution would decrease the  $t_f$  time to microseconds, which would allow one to distinguish the PL signal better from the background.

### 3.2. PL and TL dose response of LiF to high and low ionizing density radiation

The PL and TL dose responses that were obtained for the MCP and MTS detectors irradiated with  $^{137}\text{Cs}$  and  $^{241}\text{Am}$  sources are presented in Figs. 3 and 4. The PL data represent the first measurement point (0.1 ms, as shown in Fig. 2) of the pulsed-mode readout after the background value was subtracted. The TL data represent the height of the main dosimetric peak, i.e., peak number 4 for the MCP detector and peak number 5 for the MTS detector (Bilski, 2002). Figs. 3 and 4 illustrate a large difference in the dependence of PL and TL on the ionization density of the radiation. The intensity of the PL signal after irradiation with alpha particles with fluences up to  $5 \cdot 10^9$   $\text{cm}^{-2}$  is high, but the signal after irradiation with gamma rays of up to 300 Gy is low. Meanwhile, the intensity of the TL signal after the same alpha particle irradiation is much lower than that after  $^{137}\text{Cs}$  irradiation. The measurements showed that the lowest dose level, at which the PL signal is visible above the background, is 15 Gy of gamma rays for MTS and 10 Gy for MCP or the alpha particle fluence of  $0.5 \cdot 10^8$   $\text{cm}^{-2}$  for MTS and  $0.4 \cdot 10^8$   $\text{cm}^{-2}$  for MCP. The differences between the signals after gamma rays and after alpha particles are much more visible in the TL method than in the PL method.

For the MCP detectors after irradiation with  $^{241}\text{Am}$ , the signal is 33 when the fluence is  $6.8 \cdot 10^8$   $\text{cm}^{-2}$ , but for the MTS detectors, the signal amounts to only 27 after irradiation with the same fluence. The measurements of the MCP and MTS detectors irradiated  $^{137}\text{Cs}$  with a dose of 30 Gy give 20 and 14 as the PL signal, respectively, which showed that the MCP detectors are more sensitive to alpha particles and gamma rays than the MTS detectors, and the

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