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Sensitivity changes of LiF:Mg,Ti and LiF:Mg,Cu,P TL detectors after proton exposures

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HIGHLIGHTS

- Sensitivity changes of TLDs were studied after proton and gamma doses up to 30 Gy.
- Sensitivity loss of LiF:Mg,Ti and LiF:Mg,Cu,P is different for protons and γ-rays.
- A prolonged annealing time partly restores the sensitivity.

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ABSTRACT

Two most common types of LiF-based thermoluminescent (TL) detectors LiF:Mg,Ti (natural Li: MTS-N) and LiF:Mg,Cu,P (natural Li: MCP-N) have been studied for sensitivity loss after 60 MeV proton and gamma rays (^{60}Co) doses ranging between 0.5 and 30 Gy. The sensitivity (main peak area) of LiF:Mg,Ti was found to be reduced by almost 10% after the highest proton dose of 30 Gy with respect to the lowest dose of 0.5 Gy. The sensitivity loss was noticeable even after 5 Gy of protons. After gamma-ray exposures the sensitivity loss of LiF:Mg,Ti was smaller than after protons. LiF:Mg,Cu,P detectors show opposite effect: gamma-rays cause a stronger sensitivity loss than protons. The high-temperatures peaks of LiF:Mg,Ti decreased much more significantly than the main peak (by 40%). The decreased sensitivity may be partly (depending on the dose radiation type) recovered by application the extended annealing procedures: 400 °C/20 h for MTS-N and 10 min at 270 °C followed by 1 h at 240 °C for MCP-N. The observed changes of sensitivity are of importance for dosimetry in proton radiotherapy.

1. Introduction

Thermoluminescence detectors (TLDs) based on LiF:Mg,Ti and LiF:Mg,Cu,P are often used in dosimetry of heavy charge particle radiation e.g. in proton radiotherapy beams or space dosimetry. It is well known that ionization density has significant influence on several properties of TLDs, like relative efficiency or dose response (Geiss et al., 1998; Horowitz and Olko, 2004; Berger and Hajek, 2008). Within the past few years we realized a systematic investigations on the relative TL efficiency dependence on proton dose and energy (Sądel et al., 2013; Bilski et al., 2014). In the proton radiotherapy the doses to be measured are rather high – a typical dose fraction in eye tumor therapy is at the level of 15 Gy. At this dose range the radiation damage effects (sensitivity loss) in TLDs might be already observed, as shown by Bilski et al. (2008) for gamma-ray exposures. For higher doses this problem was studied several times (Jain et al., 1975; Piesch et al., 1976). There is however no experimental data available on the possible radiation damage after exposures to protons with doses at the radiotherapy level. In medical applications the requirements for dose measurement accuracy are very high, so even small changes of TLD sensitivity should be taken into account. Therefore, the goal of this work was to study this problem for two types of commonly used LiF-based detectors: LiF:Mg,Ti, LiF:Mg,Cu,P (both with natural Li) and exploiting proton and gamma irradiations for doses ranging from 0.5 to 30 Gy.

2. Materials and methods

The proton irradiations were carried out at the Proton Eye Radiotherapy Facility IFJ PAN in Krakow (Swakon et al., 2010). The protons were accelerated using AIC-144 isochronous cyclotron, operating at the nominal energy of 60 MeV, which was designed

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and constructed at the IFJ. A uniform lateral dose distribution was achieved by passive scattering with a single tantalum foil. The proton beam range was controlled with a PMMA range shifter. During irradiation, proton dosimetry was carried out with a PTW 23,343 Markus ionization chambers coupled with a PMMA moderator of variable thickness. Measurements of the dose-depth distribution were performed with a resolution better than 0.1 mm. The initial energy at the position of TLD exposures was 58.7 MeV. Range of such protons in LiF is greater than TLD thickness (0.9 mm) and according to the SRIM program by Ziegler et al. (2010), such protons passing through 0.9 mm LiF lose 2.0 MeV. This energy loss results in an increase of the average dose by about 3% (Sądel et al., 2015). Taking this into account, the average dose was corrected by a factor 1.03. The proton dose values were 0.5, 1, 2, 5, 7, 10, 13, 16, 20, 23, 26 and 30 Gy.

The gamma exposures of TLDs were performed using the ^{60}Co gamma source (Theratron 780E) at the IFJ PAN. The applied doses were identical with those used in proton exposures. Dosimetry for both protons and γ-rays was performed according to TRS 398 Code of Practice (TRS No. 398, IAEA, Vienna., 2000).

The experiments were realized using standard LiF:Mg,Ti (MTS-N) and LiF:Mg,Cu,P (MCP-N) thermoluminescent detectors in form of sintered pellets with dimensions 4.5 $\times$ 0.9 mm and density 2.5 g cm^{-3} manufactured at the IFJ Krakow. The standard annealing procedures consisted of 1 h at 400 °C for MTS-N and 10 min at 240 °C for MCP-N. Before experiments, to minimize and correct any spread of sensitivity between TLDs pellets of one group, individual response factors for each detector were determined.

The whole experimental procedure is presented in Fig. 1. After the first standard annealing the detectors were divided into two equal parts: one underwent irradiation with protons, another with gamma-rays. Both groups were irradiated in the same time and read out together, to avoid any possible fading effects. The read out system was a manual Harshaw Series 3500. TL glow-curves were registered by heating up to 350 °C for.

MTS-N and 275 °C for MCP-N at a heating rate of 5 °C s^{-1} under nitrogen flow.

After this first measurement, which mainly aimed to evaluation of TLD dose response (the results are partly presented in Bilski et al. (2014)), the detectors were again annealed in standard conditions, then irradiated with the test dose of 0.5 Gy γ-rays and read out. This allowed to evaluate changes in TL characteristics in dependence on dose and radiation type. As such changes were observed, in an attempt to remove these radiation damage effects, extended annealing cycles were applied to the same detectors: 400 °C/20 h for MTS-N and 10 min at 270 °C followed by 1 h at 240 °C for MCP-N. This annealing was again followed by irradiation with the test dose and readouts in the standard condition. The time of annealing was chosen based on available experimental data concerning this subject (Piesch et al., 1976; Bilski et al., 2008). According to that such extended annealing time should not cause any thermally induced sensitivity loss.

Glow-curves were quantified by the standard methods of glow-curve analysis, i.e. integration of a glow-curve over the main dosimetric peak area (from 100 °C to 240 °C in case of MCP-N and from 100 °C to 248 °C in case of MTS-N, with main peak position set at 220 °C). Additionally, for MTS-N detectors the high temperature area (HT area) was investigated. The HT part of TL glow-curves was calculated for the temperature range between 248 and 310 °C according to approach of Berger et al. (2006).

3. Results and discussion

Fig. 2 shows glow-curves shape for MTS-N detectors measured after the test γ-ray dose of 0.5 Gy. There is no significant changes in the shape of the main peak area between detectors after pre-doses of protons and γ-rays within the studied dose range.

On the other hand, an effect of the less pronounced high-temperature peak following higher doses is clearly visible (Fig. 2: panels A2, B2). Especially after the highest pre-dose of 30 Gy, the HT amplitude is reduced by more than half with comparison to the pre-dose of 1 Gy.

After extending annealing time, the shape of LiF:Mg,Ti glow-curve is changed. The HT peaks after different pre-doses became approximately equalized, but on the reduced level comparing with the initial value. In case of LiF:Mg,Cu,P (MCP-N) detectors, no differences between glow-curve shapes after various pre-doses was observed.

Fig. 3 present the signals (main peak area) for the investigated MTS-N and MCP-N detectors, normalized to the value obtained for the lowest pre-dose of 0.5 Gy. The vertical error bars represent the statistical uncertainties. Fig. 4 shows analogous data obtained after the prolonged annealing procedures.

It is apparent that radiation causes a sensitivity loss in all studied cases: both for protons and γ-rays and both for MTS-N and MCP-N. The strongest decrease is observed for MTS-N irradiated with protons – the effect is measurable even at 2 Gy and for 30 Gy it is close to 10%. Gamma irradiations causes smaller decrease in these detectors. It is somewhat surprising that MCP-N detectors show an opposite tendency: γ-rays induce stronger sensitivity loss than protons. In general, MCP-N were found to be more radiation resistant than MTS-N.

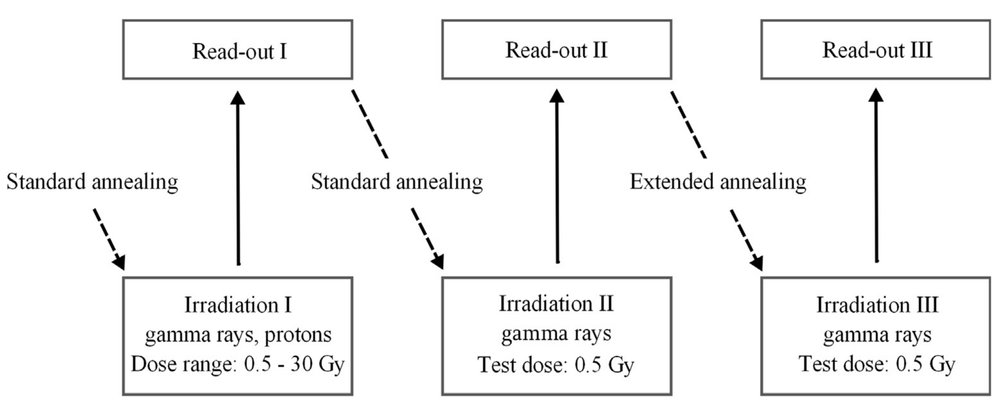

Fig. 1. Diagram illustrating the applied experimental procedure.