Modern trends and development in high-dose luminescent measurements

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Modern trends and development in high-dose luminescent measurements

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Abstract. Main application areas of high-dose dosimetry are described. The requirements to the materials for high-dose luminescent detectors are set. The examples of successful high-dose measurements using radiation-resistant phosphors are given. Viability of using materials with deep traps to detect intensive radiation flows is grounded. Characteristics of high-dose measurements using highly sensitive detectors TLD-500 (Al_2O_3:C) and LiF:Mg,Cu,P are discussed.

1. Introduction

The interest in high-dose measurements of ionizing radiations is increasingly growing due to development of radiation technologies. These technologies include radiation modification of the properties of various materials, medical products sterilization, disinfection of agricultural products, radiation control of defects in large machine parts and equipment [1]. All the above mentioned technologies employ intensive isotope radiation sources or charged particle accelerators providing exposure to the doses of several dozens of kGy. High-dose measurements are also used for radiation monitoring of nuclear power station (NPS) equipment and storages of spent nuclear fuel. High dose measurements for radiation resistance tests of the materials and equipment are of great importance as well. These served as the reasons for more extensive study for design and development of high-dose luminescent radiation detectors. The main advantages of luminescent dosimetry are a wide range of the measured doses, possibility to register different types of radiation (with low and high LET), high reproducibility of measurement results, automated dose readers.

The purpose of the paper is a brief analysis of the current status and modern trends in development of luminescent detectors for high-dose measurements.

2. Requirements to the materials for high-dose luminescent detectors

At present about twenty types of commercial luminescent detectors of radiation with the (10^{-6}-20) Gy measurement range are widely used for personal dosimetry and radiation monitoring of environment [2]. Still, the choice of luminescent detectors for high-dose dosimetry is limited. In the USA two types of thermoluminescent detectors (TLD): CaF_2:Mn (TLD-400) with the operating dose range (10^{-6}-10^{2}) Gy and $Li_2B_4O_7$:Mn (TLD-800) with the dose range (10^{-3}-10^{5}) Gy are used. In Russia TLD on the base of aluminophosphate (PST) and sodiumsilicate (TLD-K) glasses with the highest registered dose of (2-5) kGy are widely used [3].

A great number of storage materials are known to be promising for development of high-dose TLD as for these materials there is no tissue equivalence requirement. However, there are challenges

to their usage. One of such challenges is saturation of the thermoluminescence (TL) yield at high-dose irradiation. In our experiments we were the first to measure TL yield in a wide range of gamma-radiation doses (10 mGy - 100 kGy) of the high-dose TLD-500 (Al_2O_3:C) detectors which we developed on the base of oxygen-deficient alumina single crystals [4]. Figure 1 shows a dose response of the main dosimetric peak at Tm=480 K of this detector in the given dose range. The linear dose response of the TL yield is shown to be limited to 10 Gy. With the further dose increase, TL intensity decreases to some extent. However, its medium value is only slightly different from the maximal one with the use of the dose of seven orders. It implies that the centers responsible for the main dosimetric peak in the TLD-500 detectors are not destroyed being exposed to high doses as aluminum oxide is a radiation-resistant material. However, the use of the main TL peak for dosimetric measurements is challenging due to saturation of its height at the doses higher than 10 Gy.

One of the main causes of the TL yield saturation at high-dose irradiation is known to be a limited capacity of electron traps which can be filled with free electrons formed at the exposure [5]. As a result, saturation of the TL yield appears with a particular dose related to filling of all the traps responsible for the main dosimetric peak. Thus, the material for a high-dose detector should have a high capacity of unfilled trapping centers prior to exposure.

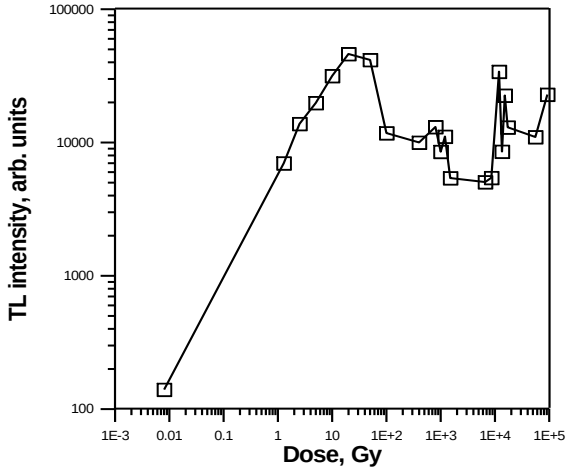

Figure 1. Dose response of TLD-500 (Al_2O_3:C) detectors at high dose γ-irradiation (Co-60).

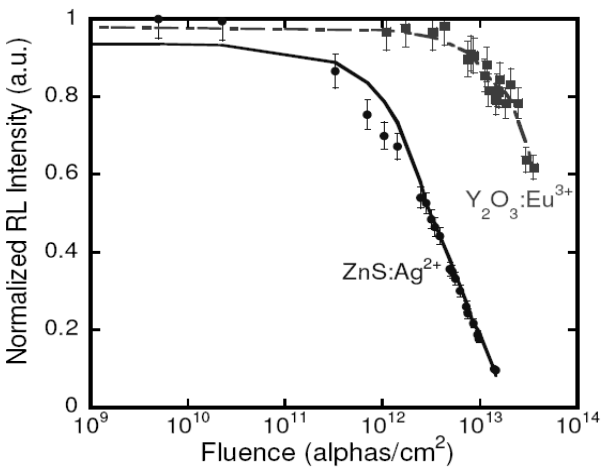

Figure 2. Normalized radioluminescence intensity as a function of alpha-particle fluence for ZnS:Ag^{2+} and Y_2O_3:Eu^{3+} [6].

Another challenge to the usage of phosphors for high dose measurements is the degradation of the luminescent centers. The degradation of the luminescent characteristics under intensive irradiation can be well seen when radioluminescence (RL) is registered. Figure 2 shows degradation of RL of the two well known phosphors exposed to α-particles [6]. Each of these phosphors has the α-particle fluence range with the constant RL intensity, then the intensity drops sharply with the fluence increase. The RL degradation kinetics proves that radiation resistance of the luminescent centers in Y_2O_3:Eu phosphors is much higher than in ZnS:Ag. Taking into account all the above mentioned, the main requirements to the material for a high-dose detector is made: the detector should be radiation-resistant and have a large capacity of the unfilled trapping centers of the charge carriers.

3. Radiation-resistant materials for high-dose dosimetry

Phosphors capable of keeping luminescent and dosimetric characteristics stable under high exposure doses belong to radiation resistance materials. Oxide dosimetric phosphors BeO and Al_2O_3 have the high radiation resistance and detectors based on the given oxides can be used for high-dose

measurements [7,8]. Recently, dosimetric characteristics of the TL detectors based on yttrium orthoaluminate crystals ($YAlO_3$) with various dopants have been studied intensively [9,10]. Radiation-resistant oxides of aluminum and yttrium contained in $YAlO_3$ allow one to make detectors capable of registering doses up to (5-10) kGy. Natural silicates such as quartz, sand, topaz, onyx, etc., are also radiation-resistant materials [11-13]. The dose response of these materials was found to change linearly in the range from 10 Gy to several kGy. As an example, Figure 3 shows a dose response of topaz pellets irradiated with gamma-rays [12]. Dependence of radiation resistance on material structure was found. In particular, it has been shown that naturally disorder multiphase compounds, low-dimensional materials (glasses, ceramics), nanostructured materials, hetero-structures have high radiation resistance. These materials are disordered, therefore additional disorder caused by structural changes under irradiation does not affect significantly their characteristics in a wide dose range.

Nanostructured phosphors are the most suitable materials for high-dose detectors. They have big capacity of the trapping centers due to the surface of the centers and high radiation resistance. A great number of researches have been made by now [14,15]. They have found the common feature shared by nanostructured phosphors of various compositions: their dose response remains linear at the doses (1.5-2) orders higher than the ones in microcrystalline analogues [8]. Figure 4 shows TL response of LiF:Mg,Cu,P detectors exposed to gamma-rays of Cs-137 [14]. It is necessary to note that the commercial polycrystalline detector provides higher sensitivity, but its TL response curve begins flattening out at the dose of 20Gy. The dose response of the nanocrystalline phosphor is linear up to 20 kGy, and it can be used for measuring high doses.

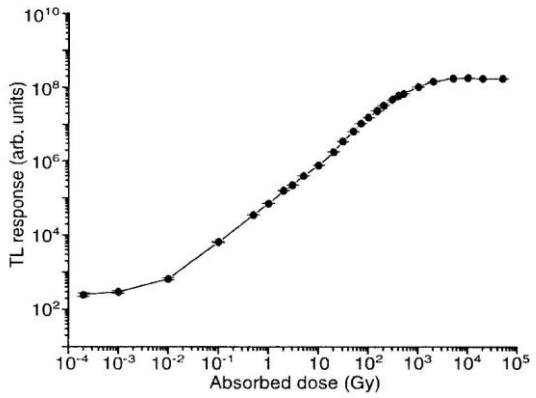

Figure 3. Dose response of topaz pellets irradiated with gamma rays (Co-60) [12].

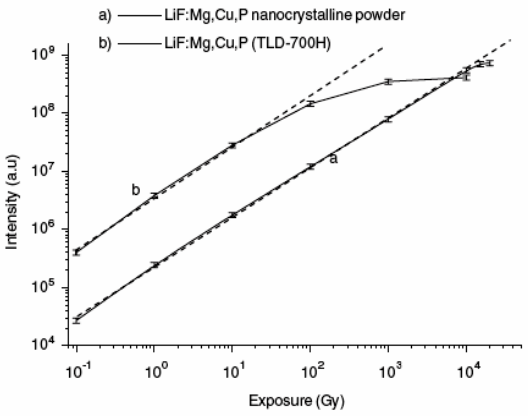

Figure 4. Comparison of dose response provided by nano- and commercial phosphor [14].

4. Use of deep traps for high-dose measurements

It is also possible to increase capacity of the traps of dosimetric phosphor using deep traps of the charge carriers. For instance, in oxygen-deficient aluminum oxide the deep traps are present both in a single crystal and in its nanostructured analogue. The energy depth of these traps is bigger than the one in the traps of the main dosimetric peak used for low-dose measurements. The nature of deep traps is not completely studied, they are considered to be formed by aggregates of the native and impurity defects [16]. Deep traps are found following high-temperature TL peaks after high-dose irradiation (Figure 5). Each of these peaks can be used for dosimetric measurements while heating the irradiated detector. For example, for the intensive TL peak at 603 K, the dose response changes linearly in the range of (1,5-80) кGy after exposure of the TLD-500 detector to an electron pulse beam from an

accelerator [8]. Measurements of the high-temperature TL peaks appearing after beta-irradiation (Sr-90) of the TLD-500 detectors showed that the dose response in the majority of them does not exceed two orders [17]. It is connected with the fact that TL of the deep traps is also saturated with a particular dose. One can consider e the following process of deep traps filling (see Figure 5). All traps in the detector material begin to be filled under exposure. However, the probability of capturing the charge carriers decreases with the increase of the trap energy depth. Therefore, the traps of the main dosimetric TL peak (A) are filled in the beginning. With the dose near its saturation the first high-temperature TL peak (B) appears and grows with the dose increase. After its saturation the TL intensity of the second high-temperature peak (C) increases with the dose growth. Similarly, deeper traps responsible for other high-temperature TL peaks are filled. The task of registering high doses consists in measuring TL yield of all deep raps.

For this purpose the phototransfer luminescence (PTTL) [18] and photo-thermostimulation luminescence (PTSL) [19] methods are used. In both methods the detector exposed to a high dose is preliminary heated to (500-550) K to empty shallow traps. To register PTTL the detector is then exposed to intensive blue light for optical emptying the traps and for transfer of the electrons delocalized from the deep traps through the conduction band to the unfilled shallow traps. The following heating of the detector allows one to measure PTTL. Its intensity is proportional to a number of electrons which filled the deep traps under high-dose exposure. The disadvantage of the PTTL method is a long measuring procedure. Moreover, not all the deep traps might be emptied under photostimulation. The PTSL method makes it possible to eliminate these disadvantages. With the simultaneous photo-thermostimulation (thermally assisted OSL), the heating enhances electron energy in the traps and the photons of lower energy are required to empty the deep traps. The electron phototransfer to the shallow traps is similar to the one described for PTTL. These traps are thermoionized and the PTSL peaks are registered. The light sum of the peaks is proportional to the dose. The results of using the PTSL method while registering high doses by means of Al_2O_3:C detectors are presented in the work [20]. The PTSL peaks are located in the temperature range convenient for registration (up to 500 K), and the dose response is linear from 10^{-2} Gy to 10^{3} Gy.

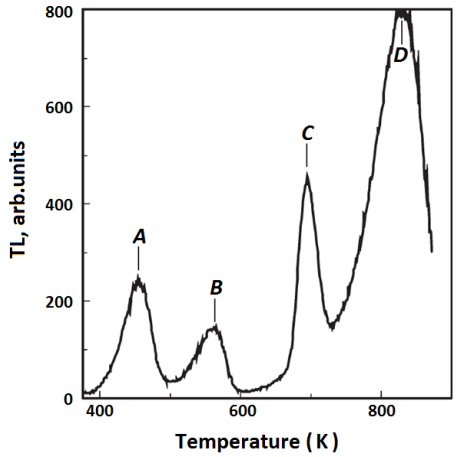

Figure 5. Glow curve of TLD-500 detector after nanosecond electron pulse irradiation (dose 75 kGy).

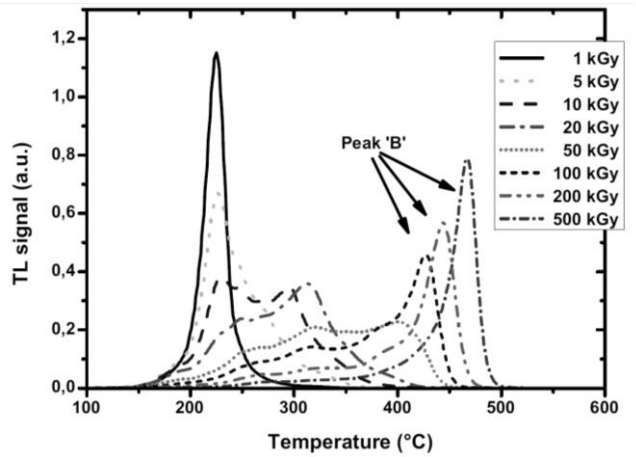

Figure 6. LiF:Mg,Cu,P glow-curves resulting from gamma irradiation (^{60}Co source) for the dose range (1–500) kGy [24].

5. The possibility of registering high doses with highly sensitive radiation detectors

At present the most sensitive to radiation detectors, i.e. detectors capable of measuring very low doses (1mkGy and lower), are TLD-500 (Al_2O_3:C) and TLD-700H (LiF:Mg,Cu,P) detectors. Due to the necessity to measure high doses, the study of the behavior of these detectors under high-dose exposure has become important. The intensity of the main dosimetric peak of the detectors under study is saturated with the (10-20) Gy dose, as it was mentioned earlier. The exposure of the LiF:Mg,Cu,P detector to high doses causes decrease of the main TL peak intensity at Tm=220 °C with the simultaneous formation of new peaks. The Tm of these peaks is shifted to the range of high temperatures with the dose growth (Figure 6). At the dose about 50 kGy a stable TL peak (peak "B") begins forming with Tm=450 °C. The dose response of this peak remains linear up to 500 kGy [21]. The TL glow spectrum acquires new bands [22] but their nature is not studied. The obtained results give evidence of formation of new electron traps and recombination centers under high-dose irradiation. In our opinion, formation of a high-temperature dosimetric peak (peak "B") under high-dose irradiation provides evidence of appearance in the LiF:Mg,Cu,P detector of deep traps connected with radiation defects and their complexes.

The behavior of the TLD-500 (Al_2O_3:C) detector under high-dose irradiation is quite different from that of the LiF:Mg,Cu,P detector [23]. Figure 7 shows that the changes of the shape and temperature position of the main dosimetric peak are slight in a wide dose range. After exposure to nanosecond pulses of an electron accelerator, the saturation of the TL yield of this peak with the dose increase is observed up to 80 kGy (Figure 7). Then, a sublinear growth of the TL yield is registered with the dose increase up to 800 kGy. The measurements of photoluminescence (PL) of the detector exposed to a high dose show that a new wide band with the maximum at 550 nm appears in the PL spectrum. The intensity growth of this band is remarkable at the exposure doses of (70-80) кGy when the TL yield starts increasing. The performed analysis has shown that an new band in the PL spectrum is a superposition of glow bands of the aggregate F_2-type centers in different charge states. These centers are additional electron traps capable of capturing free electrons, thus, increasing the TL yield in the (80-800) kGy dose range.

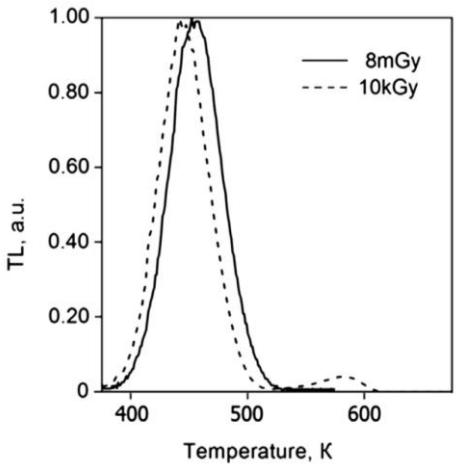

Figure 7. Normalized TL glow curves of TLD-500 detectors after low and high dose irradiation (^{60}Co).

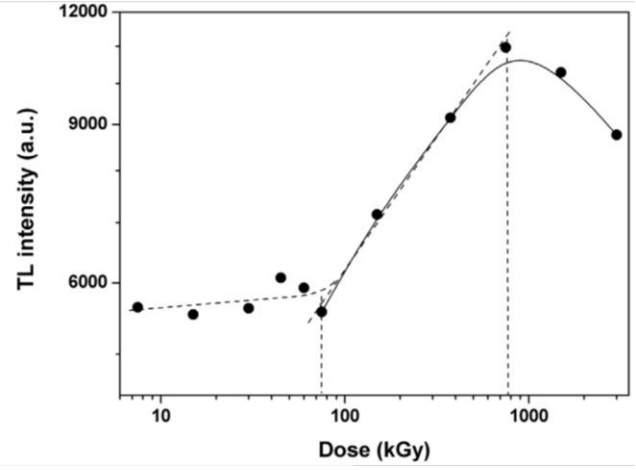

Figure 8. TL yield dependence in the main dosimetric peak at high-dose irradiation of the TLD-500 detector by an electron beam.

Thus, the possibility of registering high doses using highly sensitive detectors of radiation is shown. An increase of the traps capacity in a some dose range due to radiation disorders is a common feature.

The obtained results show the necessity of recovering the initial luminescent and dosimetric characteristics of the detectors exposed to a high dose. For his purpose, a special procedure of

annealing with experimentally defined temperature and period is required. As in the LiF crystal structural changes affecting its luminescence occur at heating above 240 °C, development of the annealing procedure of the radiation disorders in this crystal is a real challenge. There are efforts to solve this problem [24]. However, LiF:Mg,Cu,P detectors can be used only for single measurements of high doses. Whereas for radiation-resistant TLD-500 (Al_2O_3:C) detectors, appropriate annealing conditions (900°C, 1 hour) have been found. Under these conditions the initial sensitivity, luminescent and dosimetric characteristics of such detectors recover. As a result, there is a possibility of multiple repeated uses of TLD-500 detectors for high-dose measurements [23].

6. Conclusions

The results obtained allow one to make some general conclusions on high-dose TL dosimetry current state and trends:

1. At present several successful researches on luminescent and dosimetric properties of some materials suitable for high-dose detector manufacture have been carried out. A number of such materials can be expected to increase, as there is no tissue equivalence reguirement for high-dose luminescent detectors.
2. The material for a high-dose detector should be radiation resistant and have significant trapping center capacity.
3. Low-dimensional phosphors, particularly nanostructured ceramics, are promising for creating high-dose luminescent detectors. Such materials have a large amount of charge carrier traps and high radiation resistance.
4. Selection of efficient phosphors containing deep traps involved in accumulating and storing dosimetric information is required.
5. It is necessary to develop an annealing procedure for every type of high-dose detectors to restore their initial properties and to maintain reproducibility when the dose is measured.
6. Development of dosimetric devices based on luminescence registration from the deep traps of the irradiated detectors by the PTTL and PTSL (TA-OSL) methods is a vital task.

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