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SCIENTIFIC NOTE

THERMOLUMINESCENCE RESPONSE OF $K_2YF_5:Tb^{3+}$ CRYSTALS TO PHOTON RADIATION FIELDS

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This investigation has been performed to test the feasibility of using $K_2YF_5:Tb^{3+}$ crystals as thermoluminescence dosemeters (TLD). K_2YF_5 single crystals doped with 0.2, 10.0 and 50.0 at.% of trivalent optically active Tb^{3+} ions as well as K_2TbF_5 and undoped K_2YF_5 crystals have been synthesized under hydrothermal conditions. Polished crystal platelets with thickness of about 1 mm have been irradiated with X and gamma rays in order to study thermoluminescent (TL) sensitivity as well as dose and energy response in terms of the Tb^{3+} concentration in K_2YF_5. Within this concentration series, K_2YF_5 crystals doped with 10.0 at.% Tb^{3+} have been found to have maximum TL response due to a broad asymmetric TL glow peak at 269°C with good linearity of dose response and reproducibility of dose measurements. After deconvolution, the main dosimetric peak has been revealed to be composed of two individual peaks, both with linear TL response behaviour, centered at 210 and 269°C. As it has been proved, the linear TL signal coefficient for $K_2Y_{0.9}Tb_{0.1}F_5$ is almost 10 times greater than that for commercial TLD-100 (LiF:Mg,Ti), irradiated with a ^{137}Cs gamma radiation source at the same conditions. The reported results indicate that K_2YF_5 crystals doped with Tb^{3+} have potential as promising materials for radiation dosemeters.

INTRODUCTION

Till date thermoluminescence dosemeters (TLD) based on alkali fluorides and alkaline-earth fluorides, in particular, doped with rare earth ions, e.g. $CaF_2:Dy^{3+}$ (TLD-200) and $CaF_2:Tm^{3+}$ (TLD-300), are extensively utilized for dosimetry of ionizing radiation and, on the other hand, now there has been a revival of interest in developing new ultra-sensitive thermoluminescent (TL) dosimetric materials[(1)], thanks in large part to advances in environmental monitoring and radiotherapy. In this context, complex alkali yttrium fluorides can provide an exceptional possibility for developing such TL phosphors, and, in particular, the LiF–KF–YF_3 system offers a large variety of novel crystalline materials, e.g. KYF_4, K_2YF_5 and $LiKYF_5$, whose TL properties can considerably change depending on the kind of optically active rare earth ions incorporated into such hosts and their concentration in the host[(2–7)].

It should be mentioned that among the few combinations of complex alkali yttrium fluorides and optically active rare earth ions investigated so far, for the present time, K_2YF_5 fluorides doped with Tb^{3+} have the most attractive TL properties from the viewpoint of potentialities for developing TL phosphors, which look promising for application in different areas of radiation dosimetry[(2,5,7)]. Within the present work, TL and dosimetric characteristics of K_2YF_5 doped with different Tb^{3+} concentrations following X and gamma irradiation have been investigated in order to discover a $K_2Y_{1-x}Tb_xF_5$ composition having maximum sensitivity to photon radiation fields and its concrete dosimetric properties.

EXPERIMENTAL PROCEDURE

K_2YF_5 single crystals doped with 0.2, 10.0 and 50.0 at.% of trivalent Tb^{3+} ions as well as K_2TbF_5 and undoped K_2YF_5 crystals up to 1 cm^3 in size were grown by a direct temperature-gradient method as a result of the reaction of potassium fluoride aqueous solutions with appropriate mixtures of 99.99% pure rare earth oxides under hydrothermal conditions. The crystalline phases synthesized by the hydrothermal method were examined by the standard procedure of X-ray diffraction analysis, which showed that the $K_2Y_{1-x}Tb_xF_5$ solid solutions crystallized in the orthorhombic crystallographic system[(8)]. Polished crystal platelets, each with thickness of about 1 mm, were utilized for the TL measurements. In addition, unmounted LiF:Mg,Ti (TLD-100) and LiF:Mg,Cu,P (TLD-100H) chips manufactured by the Harshaw-Bicron Chemical Company were used in order to check the delivered doses and to obtain the relative TL sensitivities of synthesized K_2YF_5 crystals.

The examined samples were exposed at room temperature (RT) to gamma rays with photon energies of 662 and 1250 keV from ^{137}Cs and ^{60}Co gamma sources, respectively, as well as to X-ray beams with effective energies ranging from 25 to 100 keV, as

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defined by ISO 4037-1. The measurements of TL glow curves were performed with a Harshaw-Bicron 4500 TLD reader operating with a linear temperature profile over the range of 50–300°C in the resistive mode by using a heating rate of 10°C/s and reading cycles of 35s. Samples were annealed during secondary readings and the residual signal (reading 2/reading 1) was 0.01%. The samples were weighed and all the data were normalized to the mass.

RESULTS AND DISCUSSION

The effect of the Tb^{3+} concentration on TL response from the $K_2Y_{1-x}Tb_xF_5$ crystals has been investigated first of all in order to find compositions having maximum sensitivity to gamma radiation. The total TL outputs integrated within the 50–300°C temperature range for the $K_2Y_{1-x}Tb_xF_5$ compositions irradiated with test doses of 1 and 7 mGy are given in Table 1. It is seen from the table that the samples of K_2YF_5 containing Tb^{3+} with concentration less than 0.2 at.% have relatively low sensitivity. Also, the response of the samples having the concentration level more than 50.0 at.% Tb^{3+} is nearly similar to that of the undoped and slightly doped K_2YF_5 crystals. However, when the dopant level decreases from 50.0 to 10 at.% Tb^{3+}, the TL intensity considerably increases and K_2YF_5 doped with 10.0 at.% Tb^{3+} is the most sensitive composition to gamma radiation within this concentration series. These results show that the TL response from K_2YF_5 doped with Tb^{3+} considerably depends on the dopant level and, in particular, one can expect that subsequent synthesis and investigation of K_2YF_5 crystals containing the Tb^{3+} concentrations in the range of 0.2–20 at.% would allow discovering a composition having higher TL sensitivity than $K_2Y_{0.9}Tb_{0.1}F_5$ in principle.

The TL glow curves from a 10.0 at.% Tb^{3+} doped K_2YF_5 platelet and a TLD-100 chip exposed to a gamma radiation dose of 2 mGy are shown in Figure 1. As one can see, only one intense TL glow peak at about 269°C, showing a short tail on the low temperature side, is observed from K_2YF_5:Tb^{3+}. In other words, in contrast with TLD-100[1], the TL glow curve of K_2YF_5:Tb^{3+} have a simpler structure and the main dosimetric peak centered at the temperature higher than that for TLD-100 and the more sensitive TLD-100H[1,9] and, accordingly, there seem to be thermally and temporally more stable trapped charge carriers in irradiated $K_2Y_{0.9}Tb_{0.1}F_5$ than in those dosemeters. It should also be mentioned that in the TL glow curves from undoped K_2YF_5 and 0.2 at.% Tb^{3+} doped K_2YF_5 crystals, there is only one peak at about 160 and 110°C, respectively[2,6]. This indicates that the trap structure of K_2YF_5 drastically changes after doping with Tb^{3+} ions and the temperature positions of glow peaks considerably depend on the Tb^{3+} concentration in this host.

From Figure 1, it is also seen that the TL sensitivity of K_2YF_5:10.0 at.% Tb^{3+} is about 10 times higher than that of TLD-100. In this context, it will be noted that the Harshaw-Bicron 4500 TLD reader is equipped with a photomultiplier tube, which is optimal for measuring TLD-100 having the emission band centered at 420 nm whereas in the emission TL spectrum from K_2YF_5:10.0 at.% Tb^{3+}, the $^5D_4$-$^7F_5$ Tb^{3+} transition at 545 nm predominates[7]. In other words, the relative TL sensitivity of K_2YF_5:10.0 at.% Tb^{3+} could be higher to some extent if a TLD reader would provide higher sensitivity in the range of wavelengths at near 545 nm.

The relationship between TL response and radiation dose is illustrated in Figure 2, where the integrated TL outputs for both K_2YF_5:10.0 at.% Tb^{3+} and TLD-100 are plotted against ^{137}Cs gamma doses (D) ranging from 1 to 7 mGy. As one can see, the linearity is observed over the full range of the utilized doses and, in our case, the dose response function is fitted as $I_{TL} = K \cdot D$, where I_{TL} is the TL output intensity and K is the linear constant equal to 447.28 nC/mGy. This constant is about 7.3 times higher than the linear constant fitted for TLD-100.

Table 1. Integrated TL signals from K_2YF_5 crystals doped with Tb^{3+} ions after gamma irradiation at 1 mGy and 7 mGy doses.

Tb^{3+} concentration (%)	TL (a.u.) 1 mGy	TL (a.u.) 7 mGy
0.0	1.3	0.6
0.2	17.0	137.0
10	519.5	3234.6
20	125.5	854.7
50	41.2	360.9
100	16.8	67.3

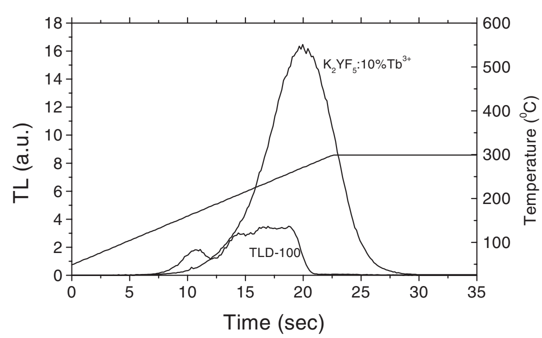

Figure 1. Thermoluminescent glow curves of 10.0 at.% K_2YF_5:Tb^{3+} and TLD-100 exposed to a ^{137}Cs gamma irradiation dose of 2.0 mGy. The reading temperature profile is plotted on the right-hand axis.

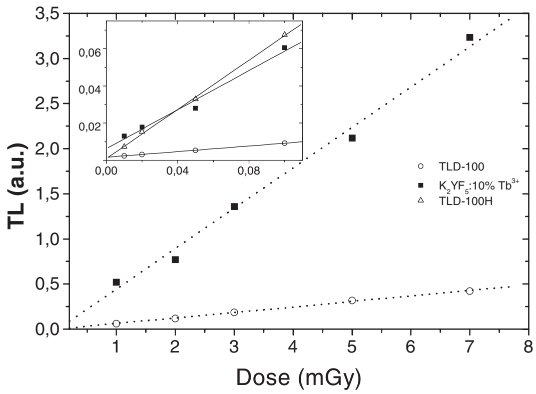

Figure 2. Dose dependences of integrated TL signals for 10.0 at.% K_2YF_5:Tb^{3+} and TLD-100 in the dose range from 1 to 7 mGy. The inset shows dose dependences for 10.0 at.% K_2YF_5:Tb^{3+}, TLD-100 and TLD-100H over the dose range from 0.01 to 0.1 mGy.

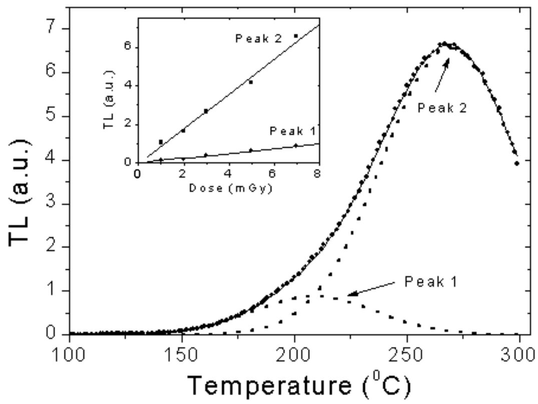

Figure 3. Deconvolution of the TL glow curve from 10.0 at.% K_2YF_5:Tb^{3+} into two components. The inset shows the linear fitting of the TL response for the deconvoluted TL peaks plotted within the gamma dose range from 1 to 7 mGy.

In order to check the response of the K_2YF_5:10.0 at.% Tb^{3+} crystal to lower doses, the data for this composition irradiated with ^{137}Cs gamma doses ranging from 0.01 to 0.1 mGy has been contrasted with the ones for commercial TLD-100 and TLD-100H chips irradiated at the same conditions. As one can see from the inset in Figure 2, the TL response of K_2YF_5:10.0 at.% Tb^{3+} is almost comparable with that of an ultra-sensitive TLD-100H, at least in this low dose range. The curve fitting has shown that the linear constant for TLD-100H is 693.4 nC/mGy, which is only 1.5 times as great as that for K_2YF_5:10.0 at.% Tb^{3+}. In this context, it should be noted that in these experiments, the TL sensitivity of TLD-100H is about 10 times higher than that for TLD-100, which does not correspond to the values reported in the literature, namely 20–50 times, and this discrepancy could be explained by the fact that the TLD-100H sensitivity considerably depends on annealing and readout conditions(1) that have not been optimized in this research.

By taking into account the fact that the shape of the glow peak from K_2YF_5:10.0 at.% Tb^{3+} is asymmetrical (Figure 1), the interpretation of this peak as the sum of several TL components has been undertaken. Figure 3 illustrates the fitting of two glow peaks to the integral glow area for this compound, namely the small one centered at 210°C and the main one at 269°C. Dose dependences for the integrated TL outputs of the individual TL glow peaks are displayed in the inset of Figure 3 and it is rather evident that both the curves show a good linearity and the 269°C peak has a sensitivity which is about seven times higher than that of the 210°C peak.

In order to determine the fading characteristics, K_2YF_5:10.0 at.% Tb^{3+} platelets have been annealed and irradiated to a 0.5 mGy dose. It has been found that for a 6-month storage period at room temperature, fading is less than 10%. Within this work, photon energy response for K_2YF_5:10.0 at.% Tb^{3+} have also been investigated and the results are given in Table 2 by taking into account that the integrated TL outputs following X-ray radiation are normalized to that measured with ^{60}Co gamma rays (1250 keV). The relative photon energy response for K_2YF_5:10.0 at.% Tb^{3+} is 12.6 at 30 keV, which is comparable with those for TLD-200 (CaF_2:Dy) and TLD-400 (CaF_2:Mn).

Table 2. Relative photon energy response of K_2YF_5:10 at.% Tb^{3+}.

E_{eff} (keV)*	25	30	40	44	85	100	662	1250
Relative intensity	12.25	12.6	10.71	8.54	4.55	3.15	1.0	1.0

*E_{eff} (for X rays only) is the energy of the monoenergetic X rays with the same Half Value Layer, as defined by ISO 4037-1.

Finally, it should be noted that the presented results concerning dosimetric properties of K_2YF_5:Tb^{3+} have been obtained without using any special thermal treatment and radiation sensitization processes. Obviously, following investigation of these processes from the viewpoint of their effect on dosimetric properties would allow increasing the TL sensitivity of phosphors based on K_2YF_5 doped with Tb^{3+} as well as co-doped with some other rare earth ions.

CONCLUSION

K_2YF_5 single crystals doped with different concentrations of Tb^{3+} ions have been grown under hydrothermal conditions. The TL properties of these crystals have been investigated and the 269°C glow peak induced by photon radiation fields in $K_2Y_{0.9}Tb_{0.1}F_5$ has been discovered to be promising for dosimetry in the dose range from 10^{-5} to 10^{-2} Gy. In particular, $K_2Y_{0.9}Tb_{0.1}F_5$ shows a TL sensitivity which is about 10 times higher than that of a TLD-100 detector.

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