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Effects of Gamma and Beta Radiations to Dosimeters Fabricated from K_2YF_5 and K_2GdF_5

Ha Xuan Vinh, Doan Phan Thao Tien, Nguyen Chi Thang

Nhatrang Institute of Technology Research and Application - VAST

02 Hung Vuong, Nhatrang, Khanhhoa

Email: vinhhx@gmail.com

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Abstract: K_2YF_5 and K_2GdF_5 doped with rare earth can be used as thermoluminescent (TL) dosimeters for gamma, beta radiations. In this study, the K_2YF_5 and K_2GdF_5 doped with Tb, Pr, Sm, and Dy with different concentrations were synthesized by solid state reaction method. These double fluoride dosimeters were irradiated with different radiations, namely beta and gamma. The study results showed that in general, the TL intensity of K_2GdF_5 is higher than that of K_2YF_5. The K_2GdF_5 crystals doped with Tb^{3+}, Pr^{3+} have very high TL sensitivities. But the sensitivities of Sm^{3+}, Dy^{3+} doped- are very low. The TL glow curve of K_2YF_5:Tb consists of three peaks at temperatures 132^oC, 207^oC and 240^oC, and its intensities are approximately. The TL glow curve of K_2GdF_5:Tb has dosimetric peak at the temperature 196^oC (heating rate 2^oC/s), the temperature of this peak suitable for dosimetry application. The TL sensitivity of K_2GdF_5:Tb is higher than that of TLD-100 and TLD-900 dosimeters for the gamma and beta radiation. The dosimeters K_2GdF_5:Tb has high sensitivity and linearity for gamma, beta radiations. In addition, the thermal fading effect of TL intensity was very low. The study results showed that these materials can be used in nuclear radiation dosimeters.

Keyword: *K_2YF_5:Tb, K_2GdF_5:Tb, thermoluminescence (TL).*

I. INTRODUCTION

Fluoride materials have a number of interesting properties from the viewpoint of both basic research and technological applications. These materials pose some striking physical properties of fluoride compounds, such as high transparency in range of wavelength from ultraviolet to infrared region. Besides it exhibit high radiation stability in compare to other halogen materials. The materials based on double potassium fluoride have been widely studied during past years and exhibit efficient luminescent when Ce^{3+}, Pr^{3+}, Sm^{3+}, Tm^{3+}, Yb^{3+}, Er^{3+} ions were doped [1-3].

Thermoluminescence (TL) is a form of luminescence of the material, which previously absorbed energy from electromagnetic radiation or other ionizing radiation. After then, they are re-emitted as light when heated. The phenomenon is distinct from that of black body radiation. Thermoluminescence dosimeters have the advantages of high sensitivity, wide measurable dose range and others. In the field of dosimetry, the materials with TL sensitivity higher than the old TL materials such as $Li_2B_4O_7$, $CaSO_4$:Dy and LiF:Mg,Cu,P are being actively investigated [4]. Some recent studies show that K_2YF_5 and K_2GdF_5 crystals doped rare earth ions have very good thermoluminescent properties, especially the shape of TL glow-curve depends on the characteristics of nuclear radiation [5]. A recent study [6] reported that the shape of TL

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glow curve of K_2YF_5 doped Tb ions depends on the kind of radiation such as: alpha, beta, gamma, neutron and X-ray. Almost papers were mainly studied on the kinetic of the materials, the available application for dosimeter did not focused [7, 10].

In this study, the TL properties and dose response of Tb^{3+}, Pr^{3+}, Sm^{3+} and Dy^{3+} doped-K_2YF_5 and K_2GdF_5 were investigated under point of view in dosimeter materials. The samples were irradiated by gamma, beta radiation fields. The shape of the TL glow curves and effect of radiative sources have been studied. For the application, the important factors of TL materials were analyzed and compared to TLD-100, TLD-900 dosimeters.

II. EXPERIMENTAL PROCEDURE

K_2YF_5 and K_2GdF_5 crystals, with dopant concentration of 2 mol % of Tb, Pr, Sm, and Dy ions, were synthesized by solid state reaction method [8]. The raw compounds are KF, YF_3 , GdF_3, TbF_3, PrF_3, SmF_3 and DyF_3 in 99.99% pure powder form of Aldrich Chemistry Company. The synthesized samples are in powder form, divided into the fractions of 20 mg and sealed in plastic capsules for irradiation.

The samples were irradiated with doses in range from 0.5 to 8 Gy using ^{60}Co gamma source. Besides, the effect and dose response for beta irradiation on the materials was estimated using $^{90}Sr/^{90}Y$ at different doses (section III.3). The TL glow curve measurements were performed with a Harshaw-Bicron 3500 TLD reader. The curve shapes and peak intensities were analyzed and processed by the Winrem program of Harshaw reader. The TL intensities are calculated in current (unit is nA) of the optical detector and processed by Winrem control software.

The weight of a sample in each measurement is 20 mg. The TL measurements were set up same conditions, such as the range temperature from 50ºC to 400ºC, heating rate of 2 ºC/s.

In order to study the TL response and sensitivity of the materials, the samples were irradiated with different doses from low to high. The TL glow curves of each type material will be evaluated, analyzed the characteristics. Furthermore, the TL characteristics of the K_2YF_5, K_2GdF_5 crystals were compared with that of TLD-100 and TLD-900 dosimeters to evaluating the TL sensitivities.

III. RESULTS AND DISCUSSION

A. Effect of dopant ion on the glow curve

The results of TL measurements of the samples were shown in Fig. 1. It is also seen that the TL glow curves of K_2GdF_5:Tb and K_2GdF_5:Pr have simple sharps and consist of two peaks at about 200ºC and 300ºC. However, the intensity of the first peak is higher than that of the second peak. The dosimetric peak is often selected in range from 160ºC to 230ºC in order to evade the black body effect. Therefore, the dosimetric peaks were selected at 192ºC and 187ºC for K_2GdF_5:Tb (Fig. 1a), K_2GdF_5:Pr (Fig 1b), respectively.

The TL glow curve of K_2GdF_5:Sm consists of main peak at the range of high temperature 294ºC (Fig. 1c). Similarly, the glow curve of K_2GdF_5:Dy (Fig. 1d) have also peak at 291ºC and a other at 149ºC. Furthermore, the TL intensities of K_2GdF_5 doped with Sm^{3+} or Dy^{3+} ions are too low. The peak in the range of low temperature (< 160ºC) is often quenched by thermal fading effect. Therefore, these materials cannot be used for application in dosimetry.

The TL glow curves of Tb^{3+} -doped K_2YF_5 have complex shapes, with structure 3 peaks at temperature 132ºC, 207ºC and 240ºC (Fig 1e), which have a same TL intensity. Similarly, the glow curve of K_2YF_5:Pr consists of 3 peaks, with highest TL intensity at 148ºC and lower TL intensity at 208ºC and 297ºC. With such glow curve sharps, Tb^{3+} or Pr^{3+} -doped K_2YF_5 cannot be used for application in dosimetry of the gamma.

So in this study, only Tb^{3+}, Pr^{3+} -doped K_2GdF_5 have the most remarkable glow curves. The sharp of the TL glow curve is simple, and the main peak temperature locates at about 200°C.

The K_2GdF_5Tb samples were synthesized at three different times, their thermoluminescence properties were exactly alike. During the period of a year, the research results showed that the stability of the samples was very good.

B. Study the response to gamma radiation doses.

To investigate the linear relation of TL intensities and gamma doses, the samples were irradiated by ^{60}Co radiation source with doses from 0.5 - 8 Gy, the responses dose of K_2GdF_5 doped others different ions are listed in Table I.

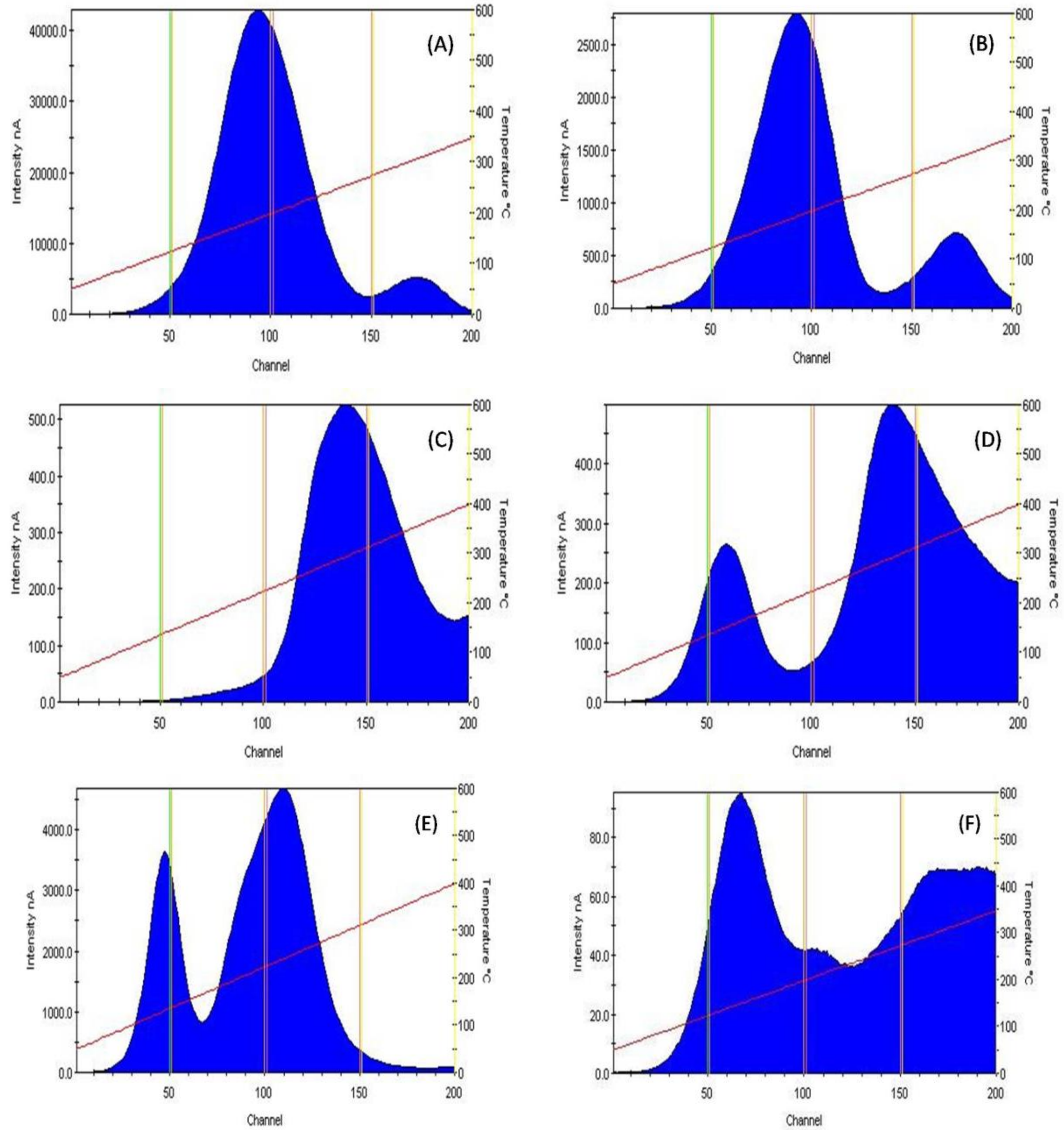

Fig. 1: The glow curve Tb^{3+}(A), $Pr3^{+}$(B), Sm^{3+} (C), Dy^{3+} (D) -doped K_2GdF_5; and Tb^{3+} (E), Pr^{3+} (F) doped-K_2YF_5 after gamma irradiation.

In this study, K_2YF_5 and K_2GdF_5 crystals doped with concentration of 2 mol % of Tb, Pr, Sm, and Dy ions. The results show that the role of doped ions is very important for TL sensitivity. The maximum intensity of Tb^{3+}-doped K_2GdF_5 doped with Tb ions is about 7 times higher than that of Pr^{3+}-doped K_2GdF_5. In case of Sm^{3+}, Dy^{3+}-doped K_2GdF_5, the TL intensities are lower than that of Tb^{3+}-doped K_2GdF_5 about 100 times. Thus the role of Tb^{3+} ions in the double fluoride materials is very noticeable, that is consistent with some recent studies on this material. In comparison with Tb^{3+}-doped K_2GdF_5, the TL intensity of Tb^{3+}-doped K_2YF_5 is very lower (10 times).

Table I. The TL intensities of main peak (nA) of materials irradiated by gamma doses

Dose(Gy)	Intensity peaks (nA)					
	K_2YF_5:Tb	K_2GdF_5:Tb	K_2GdF_5:Pr	K_2GdF_5:Sm	K_2GdF_5:Dy	TLD100
0.5	656	4722	879	106	177	1101
1	781	6393	1911	215	229	1577
2	1785	16995	3017	275	464	3018
4	2985	34036	6015	301	837	7243
6	4863	48528	7852	357	1026	12135
8	7637	69312	10181	459	1264	16549

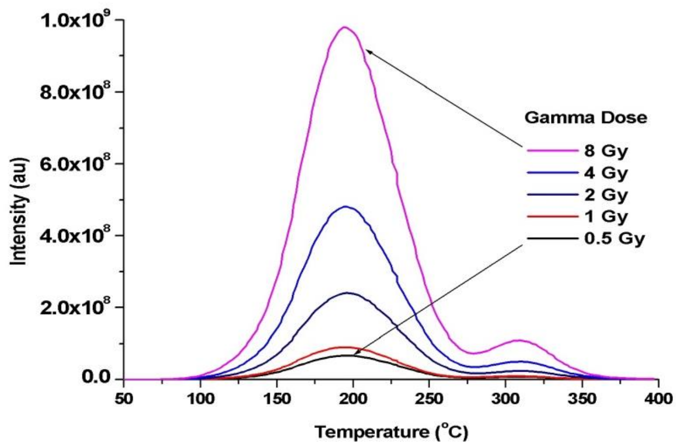

Fig. 2: The response dose of Tb^{3+}-doped K_2GdF_5

Figure 2 shows the effect of irradiated dose on the TL intensity of Tb^{3+}-doped K_2GdF_5. The shape of glow curves of Tb^{3+}-doped K_2GdF_5 did not depended on the irradiated doses of gamma source.

For dosimeters, one of the most important requirements of thermoluminescense material is linear dose response in the measuring range. The relationship between TL intensity and radiative dose is illustrated in Fig. 2. The results showed that the calibration curve could be used for calculating doses in the samples.

When irradiated with the low doses, the thermoluminescence response of K_2GdF_5:Tb was better than that of CaSO4:Dy (TLD900). With the same irradiated dose of 120 µGy, the thermoluminescence intensity of K_2GdF_5:Tb was about five times higher than that of CaSO4:Dy. Therefore, the K_2GdF_5:Tb material could be used in personal dosimetry.

C. The study of the TL response of K_2GdF_5:Tb to beta radiation.

K_2GdF_5 doped with Tb^{3+} ions with concentrations of 2% and 5% were irradiated by beta source $^{90}Sr/^{90}Y$. The samples were irradiated with doses from 0.25 Gy to 4 Gy. The thermoluminescent glow curves were measured in range temperature from 50°C to 400°C with the heating rate is 2°C/s.

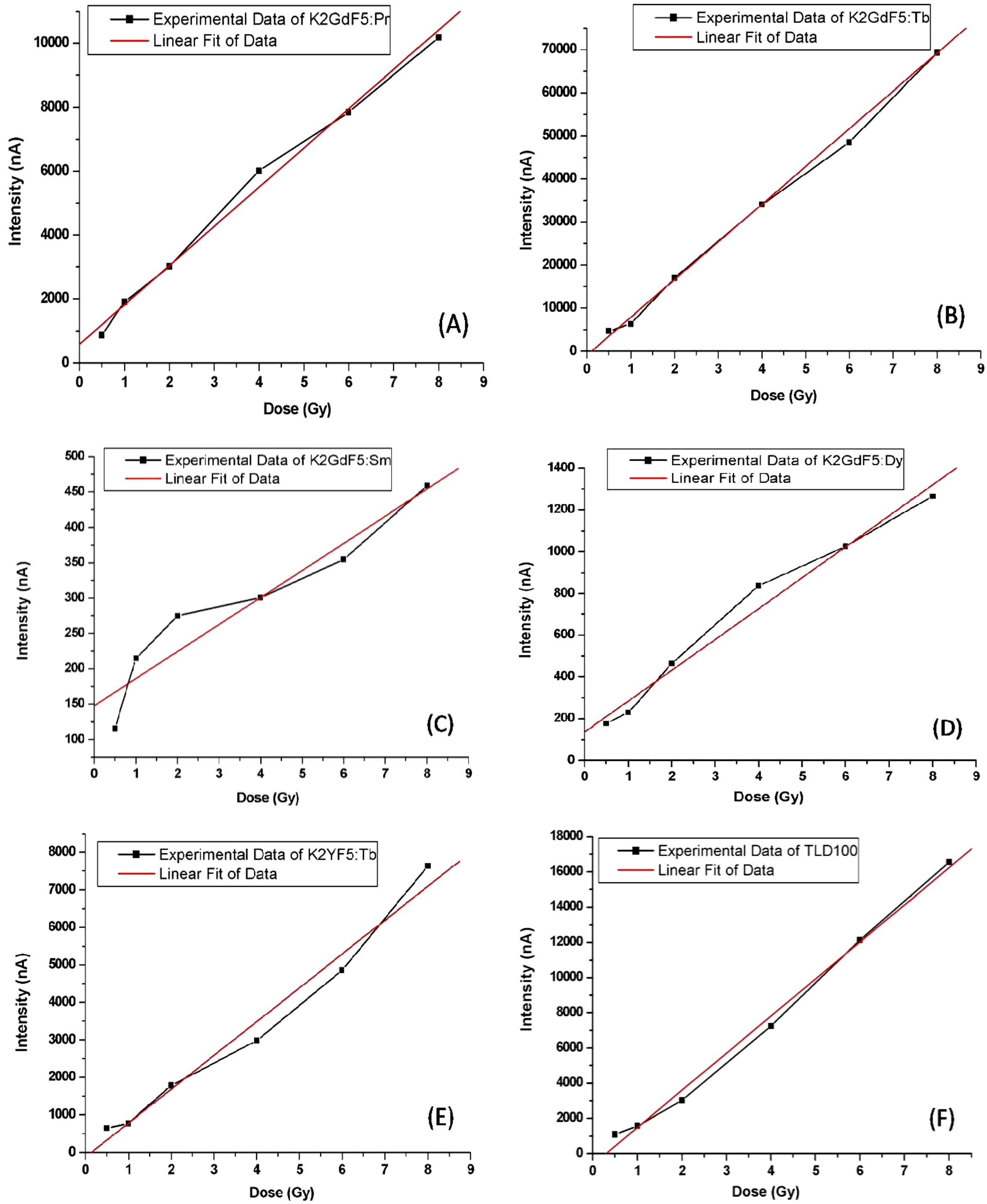

Fig. 3. The relationship between TL intensities and gamma radiation doses of Pr^{3+} (a), Tb^{3+} (b), Sm^{3+} (c), Dy^{3+} (d)-doped K_2GdF_5, Tb^{3+}-doped K_2YF_5 (e), and TLD-100.

The change of Tb^{3+} ion concentration from 2% to 5% did not affected to the sensitivity of the materials. The results from XRD data (not show here) of these samples didn't show any difference. The reason originates from the low concentration of Tb^{3+} ions, which was out of the detection limit of XRD method. The response beta dose of Tb^{3+}-doped K_2GdF_5, TLD-900 and TLD-100 are presented in Fig. 4.

The TL sensitivities of K_2GdF_5:Tb (2% Tb^{3+}) are similar for both radiative source (gamma and beta). This sensitivity is same order to the popular TLD dosimeters.

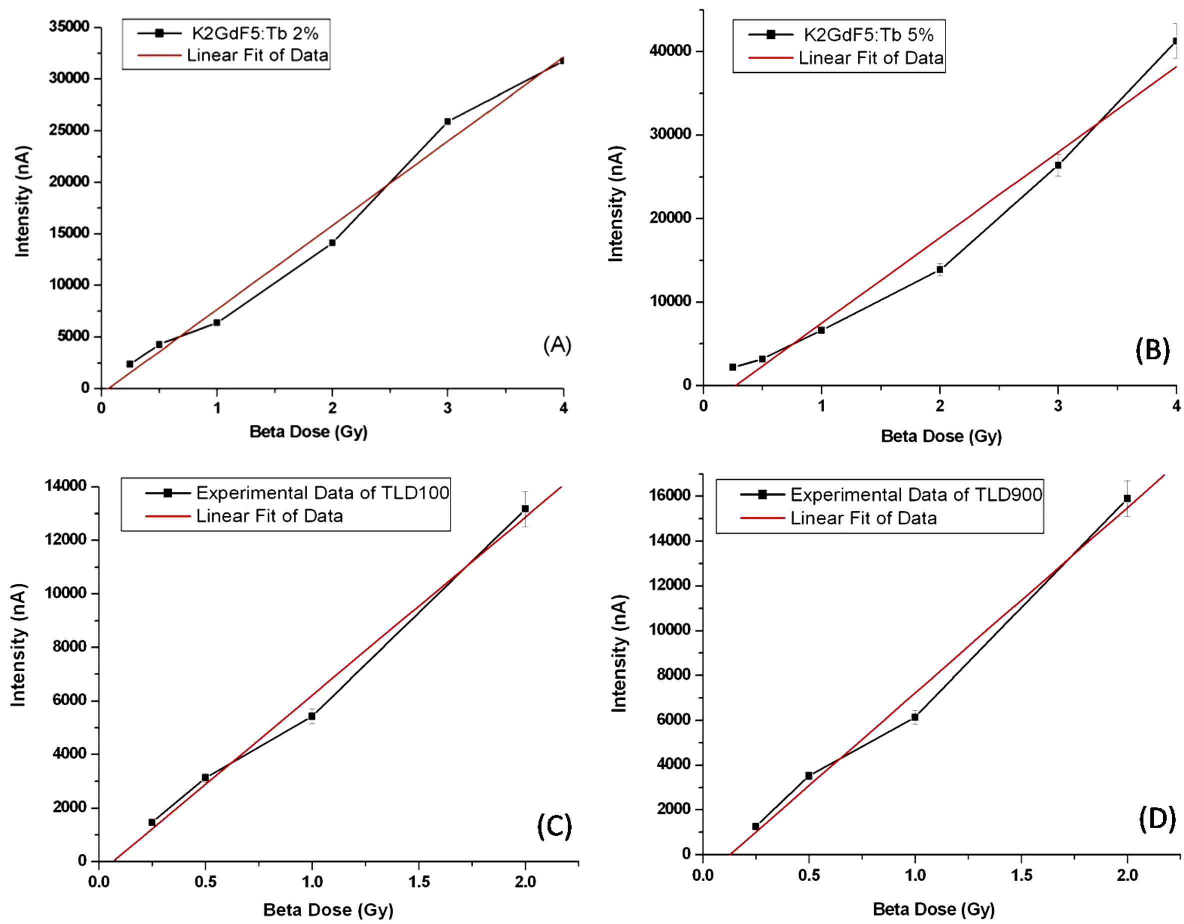

Fig. 4. The response beta dose of K_2GdF_5 doped with 2% (a), 5% (b) of Tb^{3+} ions, TLD-900 (c), and TLD-100 (d).

D. The thermal fading effect.

The TL intensity has been reduced over time at room temperature; this effect is called thermal fading. To determine the fading effect of the irradiated samples, the TL intensities were measured for a given periods of time after irradiation.

The low fading is an important factor of thermoluminescent materials in dosimetry. Materials having low fading are used to measure the cumulative doses for a long time. To achieve accuracy for dosimetry, the fading effect must be added to the measured values.

The fading effect is related to the purity of the crystals. For materials with high purity, electron traps in restricted area have simple configuration, such that the thermoluminescence will be able to satisfy "one trap" model. In restricted area, the adjacent energy levels of electron traps are cause of fast decay of electron concentration in trap, since that the thermal fading effect will be increase.

The samples of K_2YF_5:Tb and K_2YF_5:Tb were exposed by 2 Gy gamma dose for investigating the thermal fading effect. After exposing, the samples were storaged in a black box to prevent optical effects and at a temperature of about 30 ^oC. The results were listed in table II. The fading decay of K_2YF_5:Tb 2% are 5.4%, 7.4% after 15, 25 days of irradiation, and the fading of K_2GdF_5:Tb 2% are 5, 7.7, 24 % after 16, 21, 51 days. These values were rather low, indicating that the materials had been synthesized with high purity. Therefore, this material can be applied as dosimeters to measure accumulated radiation dose.

Table II. Study results of thermal fading effect

Period time after irradiation (day)	TL Intensity of main peak (nA)	Decay (%)
K_2YF_5:Tb 2% mol		
1	14540	
15	13756	5.4
25	13451	7.4
K_2GdF_5:Tb 2% mol		
2	23892	
16	22676	5
21	22055	7.7
51	18124	24

IV. CONCLUSION

This investigation was performed with the purpose to find the material having good TL properties for dosimetry. The results confirmed that Tb^{3+}-doped K_2GdF_5 consists of suitable thermoluminescence properties, namely the linear of response dose, low thermal fading, high sensitivity for gamma and beta irradiation. The TL sensitivity of Tb^{3+}-doped K_2GdF_5 is higher than that of well known dosimeters TLD-100 and TLD-900 in gamma dosimetry. This new material can be applied in nuclear radiation dosimetry.

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