Nucl. Sci. and Tech., Vol.14, No. 1 (2024), pp. 01-06

Nuclear Science and Technology

Journal homepage: http://jnst.vn/index.php/nst

Thermoluminescence characteristics of in-house synthesized K_2GdF_5:Tb powder for photon and neutron

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Abstract: This work presents an in-house synthesis process of K_2GdF_5:Tb thermoluminescence (TL) powder by using the solid-state reaction method. The K_2GdF_5:Tb powder TL glow curves (called as TL spectra), responded to photons (i.e., X-ray beams, gammas) and neutrons, have been investigated to optimize the setting parameters of the TL reader. Batch homogeneity of the in-house synthesized K_2GdF_5:Tb powder has been investigated to study the feasibility in photon and neutron personal dosimetry. The batch homogeneity of the in-house synthesized K_2GdF_5:Tb powder was investigated as less than 27.0%, which satisfied the requirement of the IEC and the ISO criteria (maximum of 30.0% is acceptable).

Keywords: *TL spectra, heating rate, variation coefficient, beam qualities.*

I. INTRODUCTION

Recently, there has been a continual interest in the development of new thermoluminescence (TL) materials that can be utilized as radiation dosimeters for various applications in industrial, scientific, and medical areas. Whereas, TL-based dosimeters can be popularly used for neutron space dosimetry and/or for extracting the gamma and neutron contributions in the mixed radiation field (e.g., such as in the neutron capture therapy applications). It is well-known that two gadolinium isotopes (i.e.,^{155}Gd and ^{157}Gd) are very sensitive to neutrons (with thermal neutron absorption cross-sections as high as about 61,000 barns and 255,000 barns, respectively) but they are not be utilized as neutron TL dosimeters (even, some gadolinium-based materials are proposed as scintillators or imaging screen phosphors for neutron detection). Some rare-earth element (RE) ion-doped fluoride compounds have been investigated to be used as effectively promising TL phosphor materials for neutron detection.

There have been some existing publications concerning on the synthesis of the RE ion (i.e., Dy^{3+}, Tb^{3+})-doped complex fluorides (e.g., K_2GdF_5:Tb) by the hydrothermal method [1-5] and/or by the solid-state reaction method [6-9]. Then, the characteristics of those TL glow curves (called as TL spectra, the distribution of TL intensity as a function of elapsed time as well as heating temperature), responded to different radiations (i.e., to alphas, betas, and X-ray beams [5] and to betas, gammas, and neutrons [6-9]), have been also investigated. However, those publications did not mention on the batch homogeneity (an important characteristics of TL materials for radiation dosimeters) of synthesized TL materials.

https://doi.org/10.53747/nst.v14i1.447

Received 28 December 2023, accepted 06 June 2024

In this work, the K_2GdF_5:Tb powder was synthesized using the solid-state reaction method. The TL characteristics (i.e., the TL spectrum and the batch homogeneity) of the synthesized K_2GdF_5:Tb powder, responded to photons and neutrons, has been investigated and compared with the IEC and ISO requirements [10-14]. This work has been performed to understand the feasibility of the synthesized K_2GdF_5:Tb powder utility in photon and neutron personal dosimetry.

II. MATERIAL AND METHOD

A. Sample of synthesized K_2GdF_5: Tb thermoluminescence powder

Several batches of K_2GdF_5:Tb powder have been synthesized at the Dalat Nuclear Research Institute (DNRI) by using the solid-state reaction method, more details can be found in a previous work [15]. As being experienced, it takes about 09 days to synthesize a batch of K_2GdF_5:Tb powder with a total mass of about 5.0 g (dependent on the mass ratio between KF, GdF_3, and TbF_3 in the pre-synthesis mixed chemical compounds). In this work, 03 random batches of synthesized K_2GdF_5:Tb powder were selected to investigate the TL characteristics after being irradiated to photons and neutrons. In each batch, the synthesized K_2GdF_5:Tb powder of 20 mg was sampled in numerical black capsules.

B. Investigation of synthesized K_2GdF_5: Tb thermoluminescence characteristics

There were many different groups of TL capsule samples were used for 02 purposes: (i) to investigate the TL spectrum and (ii) to investigate the TL batch homogeneity of the synthesized K_2GdF_5:Tb powder. All TL capsule samples were respectively irradiated to ambient dose equivalents of 10.0 mSv - for the purpose (i) and 3.0 mSv - for the purpose (ii) by 04 different radiation beam qualities (i.e., the ISO 4037 narrow spectrum X-ray beams of the nominal maximum energy of 100 keV; gammas from ^{137}Cs and ^{60}Co sources; as well as neutrons from ^{241}Am-Be source, moderated by a 10 cm thick polyethylene block between samples and the source). The irradiation geometries are illustrated in Fig.1.

Fig. 1. Irradiation geometries with: (a) RF-200EGM2 X-ray generator; (b) ^{137}Cs source; (c) ^{60}Co source; and (d) ^{241}Am-Be source

To optimize the setting parameters of TL reader, all TL capsule samples (after being irradiated to different radiation beam qualities) were read out by the Rexon UL-320 TL reader (with different setting parameters of the TL reader). The main setting parameters are as: the heating temparature range; temparature heating rate (possible values of 2, 5, 10, 15, and 20^{o}C/s); heating type (linear or non-linear); measuring cycle (total measuring steps in a TL reading process). Many TL capsule samples were read out with different setting parameters to find out the optimal TL reading process. The TL reading process is considered as acceptable since its setting parameters can lead to obtain a stable integrated TL intensity as well as a clearly distinguished TL spectrum (i.e., could distinguish different energy peaks). Then, those setting parameters of the TL reader are chosen.

To investigate the TL batch homogeneity of the synthesized K_2GdF_5:Tb powder, all TL capsule samples (after being irradiated to different radiation beam qualities) were separated into different groups (each of at least 10 TL capsule samples). Those TL capsule samples were read out using the Rexon UL-320 TL reader (with chosen optimal setting parameters). As results of the reading process, the output TL intensity counts (N_i) were available, the variation coefficient (V) and the TL batch homogeneity (H) - for different radiation beam qualities, can be then calculated as Eq. (1) and (2), respectively [10-14]. Where, D is the standard deviation of N_i values; $N_{i,max}$ and $N_{i,min}$ are the maximum and minimum values of N_i, respectively.

$$V = \frac{D}{\bar{N}} \tag{1}$$

$$H = \frac{N_{i,max} - N_{i,min}}{N_{i,min}} \tag{2}$$

III. RESULT AND DISCUSSION

A. Thermoluminescence spectrum of synthesized K_2GdF_5: Tb powder

Fig. 2 shows the TL spectra of synthesized K_2GdF_5:Tb powder, after being irradiated to 04 different radiation beam qualities. Ones can figure out that: there are a TL peak (at 160^{o}C/s appeared in each TL spectrum (a-d) of Fig.2. This is owned to the thermal fading effect of the synthesized K_2GdF_5:Tb powder, thus this peak should not be used for the dosimetry purpose. In other hand, there are 02 other possible dosimetric peaks appeared in the TL spectra (i.e., at 210^{o}C/s and at 290^{o}C/s they are quite clearly distinguished from each other), which are due to the trap depths of the K_2GdF_5:Tb powder. From Fig.2(d) and others (i.e., Fig.2 a,b,c), one can extract the contribution portions from gammas and neutrons from each other.

It has been found that the optimal setting parameters of the TL reader are as follows: the total measuring time for a reading process is 50 s with 05 following steps: (i) 8 s for pre-heating to 160^{o}C/s (ii) 7 s for heating TL material at a constant temperature of 160^{o}C/s; (iii) 16 s for heating the TL sample from 160^{o}C/s to 320^{o}C/s with the temperature heating rate of 10^{o}C/s (this was the main step of the TL reading process); (iv) keeping the temperature at 320^{o}C/s for next 11 s; and (v) 8 s for reader cooling to room temperature. The green lines in Fig.2 depict the “temperature-time” profile of the optimal TL reading process (applied for the Rexon UL-320 TL reader, in this work).

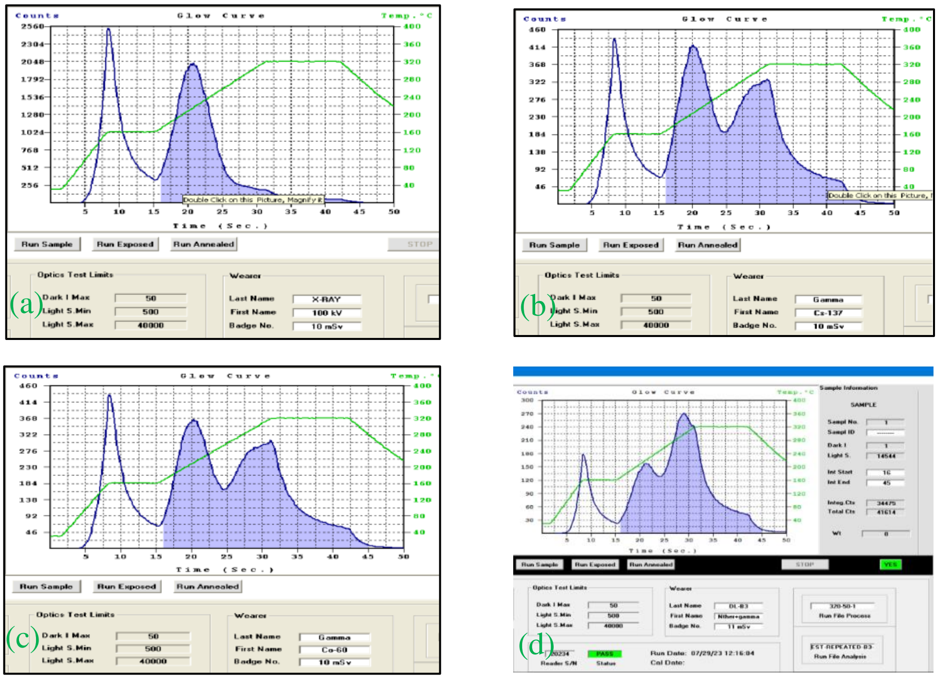

Fig. 2. Thermoluminescence spectra of in-house synthesized K_2GdF_5:Tb powder, after being irradiated to: (a) X-ray beams; (b) gammas from ^{137}Cs source; (c) gammas from ^{60}Co source; and (d) neutrons from ^{241}Am-Be source

B. Batch homogeneity of synthesized K_2GdF_5: Tb powder

Table I shows the values of N_i; $\overline{N}$; D; V; $N_{i,max}$; $N_{i,min}$; and H of 03 different in-house synthesized K_2GdF_5:Tb powder batches for 03 different radiation beam qualities. Ones can find out that: the maximum value of V was as about 7.1% and the maximum value of H was as 27 % (this is satisfied IEC/ISO requirements, maximum 30% is acceptable [10-14]).

Table I. The thermoluminescence counts (N_i), average counts ($\overline{N}$) and their standard deviations (D); variation coefficients (V), maximum ($N_{i,max}$), and minimum ($N_{i,min}$) of N_i; and the homogeneity coefficients (H) of 03 different batches of in-house synthesized K_2GdF_5:Tb powder after being irradiated to 03 different radiation beam qualities

Beam quality	***N_i (counts)***			***Beam quality***	***N_i (counts)***		
	Batch 1	***Batch 2***	***Batch 3***		***Batch 1***	***Batch 2***	***Batch 3***
X-ray	46,824	42,514	49,249	^{137}Cs	3,610	3,507	3,708
	44,770	40,957	43,105		3,625	3,626	3,247
	49,838	44,747	47,660		3,091	4,071	3,661
	41,085	44,246	46,390		3,233	4,295	3,400
	43,503	49,404	45,445		3,064	3,914	3,846
	47,490	43,294	42,714		3,259	4,136	3,463
	45,877	46,254	45,795		3,373	4,047	3,353
	45,572	44,253	44,266		3,699	3,626	3,671
	48,239	47,913	45,361		3,179	4,171	3,305
	41,789	42,210	41,749		3,218	4,295	3,948

$\bar{N}$	45,499	44,579	45,173	$\bar{N}$	3,335	3,969	3,560
D	2,645	2,492	2,183	*D*	219	274	228
V (%)	5.8	5.6	4.8	*V (%)*	6.6	6.9	6.4
$N_{i,max}$	49,838	49,404	49,249	$N_{i,max}$	3,699	4,295	3,948
$N_{i,min}$	41,085	40,957	41,749	$N_{i,min}$	3,064	3,507	3,247
H (%)	21.3	20.6	18.0	*H (%)*	20.7	22.5	21.6
^{60}Co	4,127	3,514	3,856	^{241}Am − Be	2,483	2,464	2,249
	3,759	3,840	3,327		2,266	2,236	2,366
	3,591	4,056	3,619		2,646	2,530	2,236
	3,665	3,725	3,905		2,242	2,114	2,539
	3,915	4,183	3,772		2,446	2,110	2,525
	4,222	3,501	3,858		2,612	2,447	2,408
	3,957	3,703	4,016		2,105	2,335	2,381
	3,751	3,374	3,828		2,551	2,607	2,259
	3,885	3,875	3,686		2,545	2,523	2,493
	3,524	3,972	3,959		2,284	2,274	2,004
$\bar{N}$	3,840	3,774	3,783	$\bar{N}$	2,418	2,364	2,346
D	214	247	189	*D*	173	168	156
V (%)	5.6	6.5	5.0	*V (%)*	7.1	7.1	6.6
$N_{i,max}$	4,222	4,183	4,016	$N_{i,max}$	2,646	2,607	2,539
$N_{i,min}$	3,524	3,374	3,327	$N_{i,min}$	2,105	2,110	2,004
H (%)	19.8	24.0	20.7	*H (%)*	25.7	23.6	26.7

IV. CONCLUSION

Several batches of the K_2GdF_5:Tb thermoluminescence (TL) powder have been synthesized at the Dalat Nuclear Research Institute by using the solid-state reaction method (about 09 days is needed for a synthesis process with the total mass of 5.0 g for each batch). The TL characteristics (i.e., the TL spectrum and the batch homogeneity) of the synthesized K_2GdF_5:Tb powder has been investigated after being irradiated to 04 different radiation beam qualities of photons and neutrons. The TL reader setting parameters were chosen based on the investigation of TL spectra as well as TL reading "temperature-time" profiles. The batch homogeneity was investigated for 03 randomly selected batches of the in-house synthesized K_2GdF_5:Tb powder as less than 27.0% which satisfied IEC/ISO requirements as maximum acceptable of 30.0%. The in-house synthesized K_2GdF_5:Tb powder can be utilized in ionizing radiation personal dosimetry.

ACKNOWLEDGMENT

The authors would like to thank the Ministry of Science and Technology for providing financial support for the research project No. ĐTCB.03/22/VNCHN during 2022-2023 fiscal year period.

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