

Effect of Storage on the TL Properties of Glow Curve of Synthesis: Dy, Tm and Dy/Tm Doped CaSO₄

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Authors' contributions

This work was carried out in collaboration between all authors. Author MD prepared samples and processed the measured data for fresh samples. Authors FK and DEA designed the study. Author FK processed the measured data for stored samples. She performed the statistical analysis, wrote the protocol and wrote the first draft of the manuscript. Authors managed the analyses of study and managed the literature searches. All authors read and approved the final manuscript.

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ABSTRACT

Aims: CaSO₄ doped with one or two rare earth elements dysprosium (Dy) and thallium (Tm) was studied using thermoluminescence (TL) technique with different annealing temperatures and different concentrations.

Study Design: In the present paper, stored and fresh sample synthesis effects on the kinetic parameters of dosimetric peaks were investigated.

Place and Duration of Study: Department of Physics (Atomic Physics Lab's, The University of Jordan), between July 2013 to August 2016.

Methodology: The preliminary studies TL properties of synthesis doped and co-doped samples with different concentrations and annealing temperatures, actual samples experiment and an

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analysis of results. Samples saved three years to study the effect of storage on the TL properties of samples. The experimental samples were divided into three groups.

Results: This paper gives the account of the development of a new and sensitive phosphor $\text{CaSO}_4:\text{Dy,Tm}$ and its characterization. The standard production procedure based on the re-crystallization method was used to prepare $\text{CaSO}_4:\text{Dy, Tm}$. The TL-studies were carried out by exposing it with 1rad of beta radiation (^{90}Sr - ^{90}Y -source). The theoretical studies to determine the number of peaks and kinetic parameters related to the TL glow peaks in $\text{CaSO}_4:\text{Dy,Tm}$ was performed using the computerized glow curve de-convolution (CGCD) method. Experiments were performed to determine optimum concentration of the dopants Dy and thallium Tm in the host CaSO_4 so that maximum sensitivity of the phosphor may be achieved. The optimum dopant concentration turned out to be 0.2 wt%. As there were two dopants Dy and Tm their relative ratio were varied the concentration of total dopant (Dy and Tm). General-order kinetics formula with a two and three energy levels of traps was adopted as the fitting function.

Conclusion: The results indicate that both stored and fresh samples have pronounced influence on the kinetic parameters. In the storage times experiments, change in traps distribution due to diffusion at room temperature may be the main reason causing the variations in activation energies. In the different sources experiments, different spatial distributions of trapped charge carries due to the different photoelectric effect of different photon energies may influence the binding energies. Other factors such as trapped charge conversion from one type of trap to another, competing non-radiation defect interaction and different photon energies may be used for interpreting the drift phenomenon of activation energies. Temperature dependence of frequency factor, variations of temperature lag and fluctuation and shift of the distribution frequency factor can be used to interpret the drift phenomenon of frequency factor. Kinetic order variation during the heating stage can also be used to interpret the drift phenomenon of kinetic order.

Keywords: CaSO4:Dy; Tm thermoluminescence; dosimetry; TL emission.

1. INTRODUCTION

Thermoluminescence (TL) and luminescence are well established as a sensitivity technique for recording changes in the defect concentration of insulators and is widely used for radiation dosimetry. One of the key dosimetric materials is calcium sulfate doped with rare earth (RE) ions dysprosium (Dy) or thulium (Tm) is an important thermoluminescence dosimetry (TLD) material due to its high sensitivity, large dynamic range, thermal stability and ease of preparation. It is extensively used in environmental monitoring of radiation since its qualities include a very high sensitivity, which makes it superior to many other dosimeters in this field [1,2]. Several papers have been published on the TL of [3-11] a single dopant RE sensitizer of either Dy or Tm and inter-comparisons of studies of different systems introduce uncertainties due to experimental errors of thermal contact, but very few have been devoted to the TL of co-doped $\text{CaSO}_4:\text{Dy/Tm}$. For example CaSO_4 samples co-doped with Tm and Dy [12-14] show a systematic temperature difference between the dosimetric peaks of the two dopants, which cannot be explained by a simple model of isolated trapping and recombination centers. The explanation given for

these examples is that the trapping and recombination processes occur within a large defect complex. A further test of this suggestion is synthesis here with new samples, namely $\text{CaSO}_4:\text{Dy}; \text{CaSO}_4:\text{Tm}$ and $\text{CaSO}_4:\text{Dy/Tm}$ in order with different concentrations to study the effect of co-doped on dosimetric peak compared with single-doped material and standard. By recording the emission spectra, and using dopants with characteristic emission, it is further possible to search for any temperature different between the peaks of the two dopants, as was observed between the glow peaks near 230 to 240°C for Dy/Tm [12-14]. [15-19] have found two peaks at 140 and 200°C using a heating rate $\beta = 9.7^\circ\text{C}\cdot\text{s}^{-1}$. Comparison between various authors indicates the presence of some common peaks e.g., about 120°C and 220°C, independent of the heating rate used. In this work of principle these new materials could be used in certain applications where radiation exposure needs to be measured at high temperatures. The samples were subjected to a series of treatments including, different thermal cycles, different concentrations of co-doped and excitation by β -particle and comparison with other and a standard material.

2. EXPERIMENTAL AND MEASUREMENTS

The samples were synthesized by the Yamashita method [20-21]. Sixteen sets of samples in three groups were synthesized for this work, including single doped with either Dy or Tm and fourteen other sets of samples co-doped with both Dy and Tm in different concentrations.

Group A: CaSO₄: Dy powder samples were prepared by dissolving the desired impurity Dy (30 mg) in the form of Dy₂O₃ in 50 ml of concentrated sulfuric acid (H₂SO₄) and 7 gm of calcium sulfate (CaSO₄·2H₂O) added. The solution is evaporated by using a temperature controlled electric heater at 350°C in a well closed system to avoid the escape of the acid vapors to the atmosphere.

Group B: And by same method prepared samples CaSO₄: Tm with concentration (0.2 wt%).

Group C: also for samples co-doped both Dy and Tm in a fixed concentration of Dy (0.1 wt%) with different concentrations of Tm (0.01, 0.05, 0.1, 0.2, 0.5, 0.8 and 1 wt%) and a fixed concentration of Tm (0.1 wt%) with different concentrations of Dy (0.01, 0.05, 0.1, 0.2, 0.5, 0.8 and 1 wt%).

Finally, the powder was sieved using a mesh of particle size less than $\leq 75 \mu\text{m}$. A separated CaSO₄ sample was also studied for reference purposes. Samples used in the measurements were shaped out in the form of circular discs with dimensions 13 mm diameter and thickness about 1.3 mm., each of mass 350 mg, and pressed under a 5.0 ton pressure for reading after synthesized them directly. Before the beta exposure the samples were kept in porcelain crucibles, annealed in the range of 200 up to 700°C for 1 h duration, then cooled naturally to room temperature (RT). Samples were irradiated, at RT with a calibrated ⁹⁰Sr-⁹⁰Y-source by β -particles using VINTEN Model 623 automatic dosimeter irradiator of nominal activity 1 mCi. The temperature variation in the range from ambient to 673 K was 0.5K. (Dose = 2.87 $\mu\text{Gy/s}$) irradiator. This test dose was found suitable for stability conditions and repeated measurements indicate that the annealing temperature results in good of the recorded GL-peaks. The delivered test dose to samples was 1 rad. This test dose, however, was found to show all GL-peaks in the GL-curve of both sample types, and no need to

reach high irradiation doses to observe all peaks. TL measurements were made using a Harshaw 3500 TLD reader using a linear heating rate of 2°C/s.

3. RESULTS AND DISCUSSION

3.1 Effect of Annealing Temperatures on Single Doped and Co-doped CaSO₄ of Dy(0.2), Tm(0.2) and Dy/Tm(0.2) wt%

Annealing refers to a heat treatment given to a material so as to modify some of the material properties. It is a process in which the material is heated above its recrystallization temperature, for certain duration, maintaining a suitable temperature, and then finally cooled. Annealing is used to relieve internal stresses in the material thereby refining the structure by making it more homogeneous. Considering the above facts the phosphor was annealed at different temperatures and compared. the effects of annealing temperatures on the structure of the TL glow curves not only depend on redox reactions taking place during annealing/readouts but also depend on certain other phenomena, such as, aggregation of impurities, the diffusion of atmospheric oxygen during annealing and so on [22-27]. The glow curve of the base material CaSO₄ which gives background signals, initiated the work through the selection of arbitrary concentration (0.2 wt%), of dopant based on the approach and method followed generally by Yamashita [20-21]. Atypical glow curve which represents the intensity of emitted light versus temperature of CaSO₄: Dy, Tm and Dy/Tm. Fig. 1 shows the TL glow curves of the phosphor annealed at different temperatures ranging from 200°C to 700°C. TL glow curves were recorded using a recorded using a Toledo (Model 654) and Harshaw TLD reader (Model 3500). The figure also includes the unannealed sample for comparison. It was found that the maximum TL sensitivity of the phosphor was for an annealing temperature of 400°C, so the rest of the experiments were performed with phosphors annealed at this temperature of saved samples, while was 500°C for fresh synthesis. Fig. 1 (a1-a3 and b1-b3), it could be observed from these figures that all the samples annealed from 200-700°C, there are two dosimetric peaks, while for fresh synthesis, which were observed three peaks, position of these peaks detected as shown in Table 1 with different impurity and annealing temperatures.

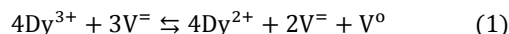
3.2 Effect of Annealing Temperatures on Single Doped of Standard Samples CaSO₄ of Dy, Tm

Two standard samples, CaSO₄:Dy and CaSO₄:Tm prepared at our laboratory procured from Harshaw Bicorn, USA (currently M/s Thermo Scientific) were used for the TL comparison study. CaSO₄:Dy and CaSO₄:Tm were subjected to the standard annealing treatment of 400°C for 1 h. Before TL readout, all the samples were irradiated from ⁹⁰Sr-⁹⁰Y-source beta ray with a dose of 1rad in air at RT. Fig. 2 (a and b) shows the TL glow curves of the phosphor annealed at different temperatures ranging from 400°C to 700°C. TL glow curves were recorded using a Harshaw TLD reader (Model 3500). Fig. 2(a) shows the effects of thermal treatment temperature on the shapes of the glow curves and the TL intensity of CaSO₄:Dy at different temperatures applied, showing growth in the TL intensity as the temperature increases.. As expected CaSO₄:Dy exhibits two peaks at around 115° C and 225° C with main peak at 225° C. The same effect has also been observed for CaSO₄:Tm. The temperature of thermal treatment affects the TL intensities; it decreases as the temperature increases up to maximum at 700°C as shown Fig. 2(b).

3.3 Comparison Synthesis CaSO₄:Dy and CaSO₄:Tm with Standard Samples

Fig. 3(a and b) shows a comparison graph of synthesis of CaSO₄:Dy, CaSO₄: Tm and standard CaSO₄:Dy, Tm, respectively, for a dose of 1rad. As expected CaSO₄:Dy exhibits two peaks at around 129°C and 208°C with main peak at 208°C, for synthesized material, while 110°C and 223°C of CaSO₄:Dy standard. CaSO₄:Dy(0.2) showed a higher TL intensity than standard sample. A similar shape is seen in CaSO₄:Tm phosphor but with enhanced intensity with change in the peaks positions to shift lower temperature comparisons with CaSO₄:Tm standard. It can be seen that the present sample is approximately 6.5, 8.4 and 13 times the intensity in comparison to CaSO₄:Tm, Dy and Dy standard respectively. The first peak was observed to be of lower intensity than the second one. Generally there is agreement with literature about the main dosimetric peaks observed, but disagreement in relation to the detailed structures. The origin of the first peak is known to be related to a defect center of the type SO₄⁻-V_{Ca} (V) with each center trapping a single hole

[28-29]. The second peak, on the other hand, is due to holes being released from Ca²⁺ vacancy-related two-holes trapping centers [28-29]. The kinetic equation governing the TL process in CaSO₄: Dy is based on the equilibrium equation, namely [30].



Where V[·] is cation vacancies, V^{··} centers are vacancies which have captured a hole and V[°] center is a vacancy that has captured two holes. The above reaction indicates the formation of V-type centers when cation vacancies capture holes released from the impurity ions by radiation. Such centers are, however, annealed during TL readout. The released holes recombine with the impurity ions, given off light [30-31].

3.4 Comparison of Synthesis of Single and Co-doped CaSO₄

After optimizing the concentration of 0.2 wt%, further samples were prepared by doping Dy in the form of Dy₂O₃ to prepare CaSO₄:Dy (0.2 wt %) and CaSO₄:Dy, Tm (0.2 wt%, 0.2 wt%). All the three prepared samples CaSO₄:Dy, CaSO₄:Tm and CaSO₄:Dy(0.2)/Tm(0.2) were subjected to a beta dose of 1 rad from a ⁹⁰Sr-⁹⁰Y-source. Fig. 4a shows the comparative TL glow curves for the three samples. It was observed that the Dy doping in CaSO₄ (i.e., CaSO₄:Dy) exhibits two peaks at around 129°C and 208°C with main peak at 208°C while the Tm doping (CaSO₄:Tm) showed a higher TL intensity than the Dy doping with two peaks at around 125 and 204°C, as reported elsewhere [4-5,12]. Interestingly, the co-doped sample i. CaSO₄:Dy (0.2), Tm (0.2) resembles the TL glow curve of the Dy doped sample with no significant change in the second peak position with shift to higher temperature of first peak and with an enhanced intensity compared with CaSO₄:Dy and CaSO₄:Tm both, this results reading of samples stored. While fresh samples, as shown in Fig. 4b, it was observed that the Dy doping in CaSO₄ (i.e., CaSO₄:Dy) exhibits three peaks at around 148, 240 and 263°C with main peak at 240°C while the Tm doping (CaSO₄:Tm) showed a lower TL intensity than the Dy doping with three peaks at around 147, 242 and 272°C, as reported elsewhere [8-32]. Interestingly, the co-doped sample i. CaSO₄:Dy(0.2), Tm (0.2) resembles the TL glow curve of the Dy doped sample with no significant change in the first peak position with shift to higher temperature of

second and third peaks and with an enhanced intensity compared with CaSO₄:Dy and CaSO₄:Tm both.

The 240°C TL peak in Rare Earth (RE) doped CaSO₄ has been correlated with the holes trapped by sulfate radicals near Ca²⁺ ion vacancies created due to charge imbalance between RE³⁺ ions and Ca²⁺ ions. Since the incorporation of Tm³⁺ ions does not create cation vacancies, the high temperature TL peak at 240°C is absent in CaSO₄:Tm. But the enhanced TL intensity advocates that although CaSO₄ doped with Tm does not have a 240°C peak, Tm is playing a significant role in the TL of 240°C peak in CaSO₄:Dy/Tm phosphor.

3.5 Effect of Activator Concentration

Figs. 5(a, b) and 6(a, b) show the glow curves of the CaSO₄:Dy/Tm, the ratio of Dy and Tm was again varied as CaSO₄:Dy(x)/Tm (1-y) and Tm(x)/Dy(1-y) where x is a constant value (x = 0.1 wt%) and y varies in steps from 0 to 0.99, total additive quantity of dopants being 0.01 wt%. Fig. 5a shows the TL glow curves of CaSO₄:Dy/Tm phosphor samples with Dy(0.1) and variation in Tm doping for a dose of 1 rad. From the same batch, three samples were taken every time to ensure accuracy in the readings. It was observed that the TL intensity increases with the increase in the Tm concentration from 0.01 to 0.1 and decrease of charge density above the dopant concentration 0.1%. In particular, at this concentration however, the glow curves show an unexpectedly enormous drop in the overall TL-intensity. The highest intensity ratio between the main peak detected near 210°C to the first peak at 130°C amongst the group of two peaks detected is about 2 times. The glow curves indicate that with increasing the concentration of co-dopant Tm, not only extra defects are created, but also changes within the distribution of existing defect structure is noted. In addition, one also notices that the major group of two peaks detected are preserved, indicating that within the host, the co-dopant traps are thermally disconnected but there is a probability of charge carriers transferring between existing traps. An almost similar behavior is noted below and above the dopant concentration of 0.2%, as shown in Fig. 5b illustrate the results of varying the concentration of the co-dopant Dy, when incorporated to the Tm doped based material. Results, which were noted in Fig. 5 (a and b) for storage samples.

While, Fig. 6 (a, b) illustrates the results of fresh samples for co-dopant same synthesized samples of CaSO₄: Dy/Tm with different concentrations as shown in Fig. 6 (a, b). Comparison with results of storage samples. TL-glow curves exhibit three peaks. It was observed that the TL intensity increases with the increase in the Dy and Tm concentration, except 0.5 wt% concentration TL-intensity was decreased. The maximum TL intensity was seen in CaSO₄:Dy(0.1)/Tm(0.05) and CaSO₄:Tm(0.1)/Dy(0.05) combination.

4. ANALYSIS OF TL-GLOW CURVES BY GCCD CURVE FITTING AND TRAPPING PARAMETERS

The escaping electron from the trap has equal probability of either being re-trapped or of recombining with hole in a recombination center. For this, computerized glow curve fitting methods have been used by physicists studying TL mechanisms and have helped better understanding and important advances in TLD. Considering this fact, the glow curve convolution deconvolution (GCCD) curve fitting in CaSO₄: Dy, Tm and Dy/Tm samples were done using glow curve deconvolution (GCD) functions [Eqs. (1) and (2)], suggested by Kitis et al., [33] for first, second and general order glow curves, respectively. They are applied to the experimentally obtained glow curves to separate each peak.

The kinetic parameters namely activation energy (E), Tm, order of kinetics (b), n₀, frequency factor (s) and life time (τ) for any experimental TL glow curve can be determined using various methods reported in literature (Chen and McKeever, 1997). In the present study, we chose CaSO₄:Dy, Tm synthesized and commercial and CaSO₄:Dy/Tm annealed at 400°C and exposed to 1 rad dose of beta radiations to determine the trapping parameters. It is worth to mention that all the glow curves were deconvoluted also using the general-order kinetics (GOK) equations. For linear heating rate, the GOK equation is given by [34] is generally given by:

$$I(T) = s^n n_0 e^{-E/kT} \left[1 + \frac{s(b-1)}{\beta} \int_{T_0}^T e^{-E/kT} dT \right]^{b-1} \quad (1)$$

Where: n₀ = initial concentration (cm⁻³) of trapped charge carriers at time t=0 and initial temperature T₀ = 0K.

s = a constant characteristic of the electron trap, called the pre-exponential-frequency factor or attempt-to-escape frequency" (s^{-1}). This parameter is proportional to the frequency of the collisions of the electron with the lattice phonons. Typically the maximum values of s correspond to the values of the lattice vibration frequency, i.e. 10^{12} – 10^{14} s^{-1} , s' is the effective pre-exponential factor for general order kinetics ($cm^{3(b-1)}s^{-1}$), $s'' = s'n_0^{(b-1)}$ = an empirical parameter acting as an "effective" frequency factor for general-order kinetics (in s^{-1}), β = heating rate (in $K.s^{-1}$); assumed linear: $T = T_o + \beta t$, E = activation energy or trap depth (in eV);

b = kinetic order; k = Boltzmann constant ($=8.617 \times 10^{-5}$ eV.K $^{-1}$); and T = final temperature (in K).

The frequency factor s and n_0 are therefore given by:

$$s = \frac{\beta E}{kT_M^2} \left[1 + \frac{(b-1)(2kT_M)}{E} \right]^{-1} e^{\frac{E}{kT_M}} \quad (2)$$

And the life time, which is the time the electron spends in the electron trap is given by

$$\tau = s^{-1} e^{E/kT} \quad (3)$$

Figs. 7 and 8 show the de-convoluted curves and the theoretical curve fitted with the experimental curve after convolution for $CaSO_4:RE^{3+}$ powder samples exposed to a beta ray. All glow curves are deconvoluted through four main peaks labeled P_1 through P_4 , except $CaSO_4:Dy/Tm$, of five peaks. Such peaks are correlated to crystal defects within the material which are responsible for TL emission.

Trapping parameters of all the above peaks are also calculated and are summarized in Table 2. From the data, which was chose of $CaSO_4:Dy$ fresh and stored samples. It is clear that all the peaks follow first, second and mixing order of kinetics and hence there is no retrapping taking place. The energy levels (activation energy) of various traps (corresponding to various peaks) are very much different. Therefore, it is clear that there are some deep and shallow traps. The competition among them might be giving various releasing and retrapping probabilities, which might have resulted in different frequency factors. The traps could be either electron traps or hole traps or of both kind.

Table 1. Effect annealing temperatures on position of main traps of $CaSO_4:Dy$, Tm and Dy/Tm and standard samples of Dy, Tm

Fresh samples										
T_a °C	T_m (°C) of $CaSO_4:Dy$			T_m (°C) of $CaSO_4:Tm$			T_m (°C) of $CaSO_4:Dy/Tm$			
	P_1	P_2	P_3	P_1	P_2	P_3	P_1	P_2	P_3	
400	148	240	263	147	242	272	145	236	260	
500	148	239	274	147	242	268	143	231	264	
600	148	237	274	138	236	263	142	236	266	
700	148	239	274	154	233	261	144	236	266	
Stored samples										
T_a °C	T_m (°C) of $CaSO_4:Dy$		T_m (°C) of $CaSO_4:Tm$		T_m (°C) of $CaSO_4:Dy/Tm$		Standard samples			
							$CaSO_4:Dy$		$CaSO_4:Tm$	
	P_1	P_2	P_1	P_2	P_1	P_2	P_1	P_2	P_1	P_2
0	223	-	233	-	226	-	-	-	-	-
0-1rad	127	217	127	226	129	216	-	-	-	-
200	125	233	125	229	127	222	-	-	-	-
300	117	230	125	227	125	223	-	-	-	-
400	127	210	124	203	134	208	110	223	143	255
500	132	209	137	207	136	209	115	227	133	252
600	129	212	129	209	133	206	111	224	137	240
700	123	204	119	207	125	207	112	225	130	236

Table 2. TL parameters for eight characteristic of CaSO₄:Dy of fresh and stored samples

Samples	Ps	E(eV)	T _m (°C)	b	n ₀ (cm ⁻³)	I _m (a.u)	ω(°C)	s(s ⁻¹)	t*
CaSO ₄ :Dy (Fresh)	P ₁	0.831	140	2.5	1.974×10 ⁴	520	64.73	1.43×10 ⁹	19.77 h
	P ₂	0.865	221	1.00	4.324×10 ⁴	1427	55.82	5.499×10 ⁷	2.67 m
	P ₃	0.952	259	1.00	6.035×10 ⁴	1853	59.89	8.154×10 ⁷	52.74m
	P ₄	1.161	304	1.19	2.603×10 ⁴	764	62.23	1.102×10 ⁹	1.06×10 ³ y
CaSO ₄ :Dy (Stored)	P ₁	0.673	126	1.40	4.055×10 ⁴	133655	55.00	2.988×10 ⁷	2 h
	P ₂	0.816	195	1.11	4.487×10 ⁴	147666	55.77	5.024×10 ⁷	13 days
	P ₃	1.035	225	1.33	5101×10 ⁴	168180	55.08	2.812×10 ⁹	3 y
	P ₄	1.132	284	2.5	1.8×10 ⁴	35597	86.31	1.314×10 ⁹	290 y

[Reference to Fig. 7(a and c) and Eqs. (2) and (3)]

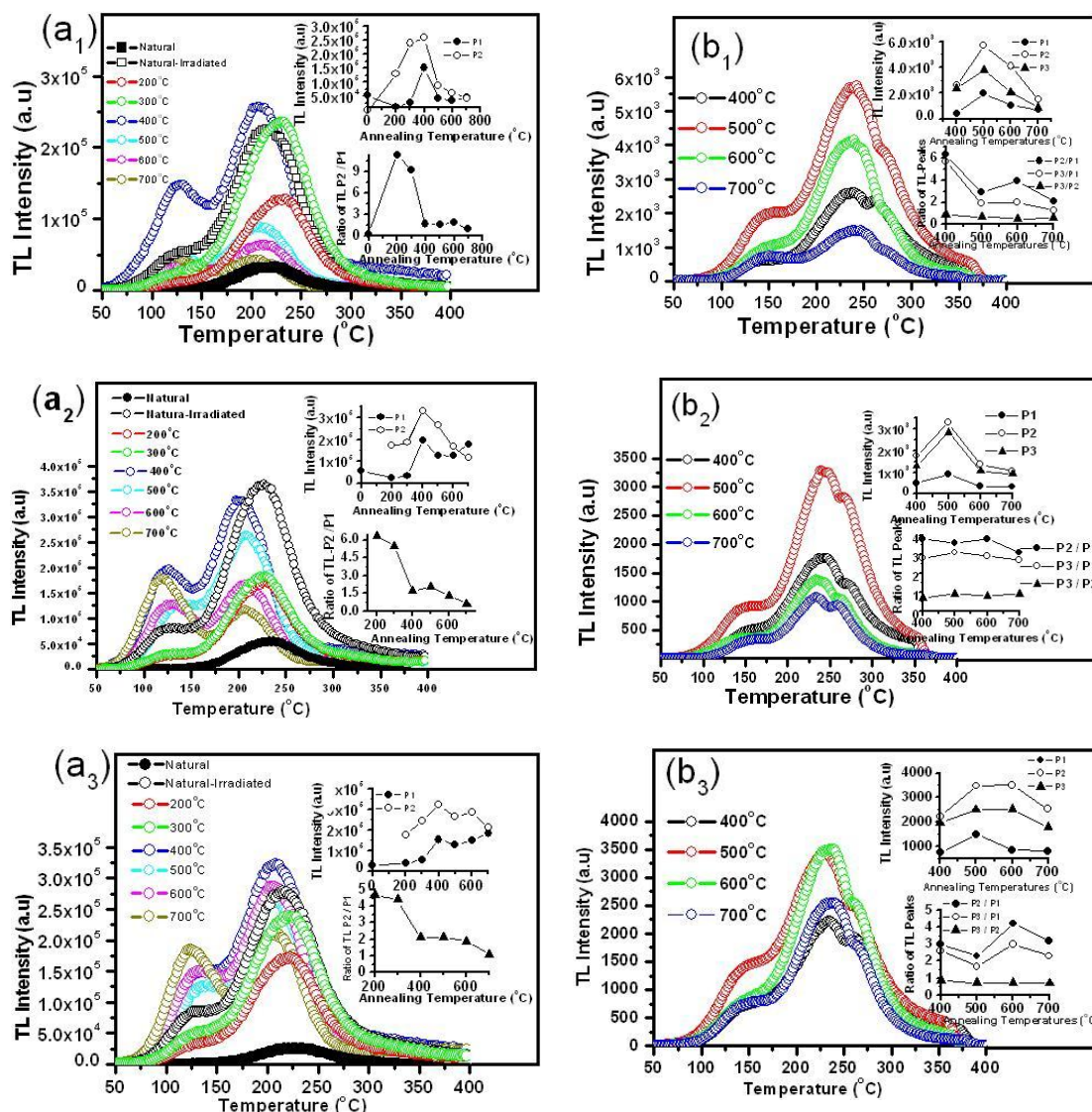


Fig. 1. Influence of annealing temperatures on TL glow curve of CaSO₄: Dy, T_m and Dy/T_m exposed to 1 rad of beta radiation. Stored samples (a1-a3) and Fresh samples (b1-b3)

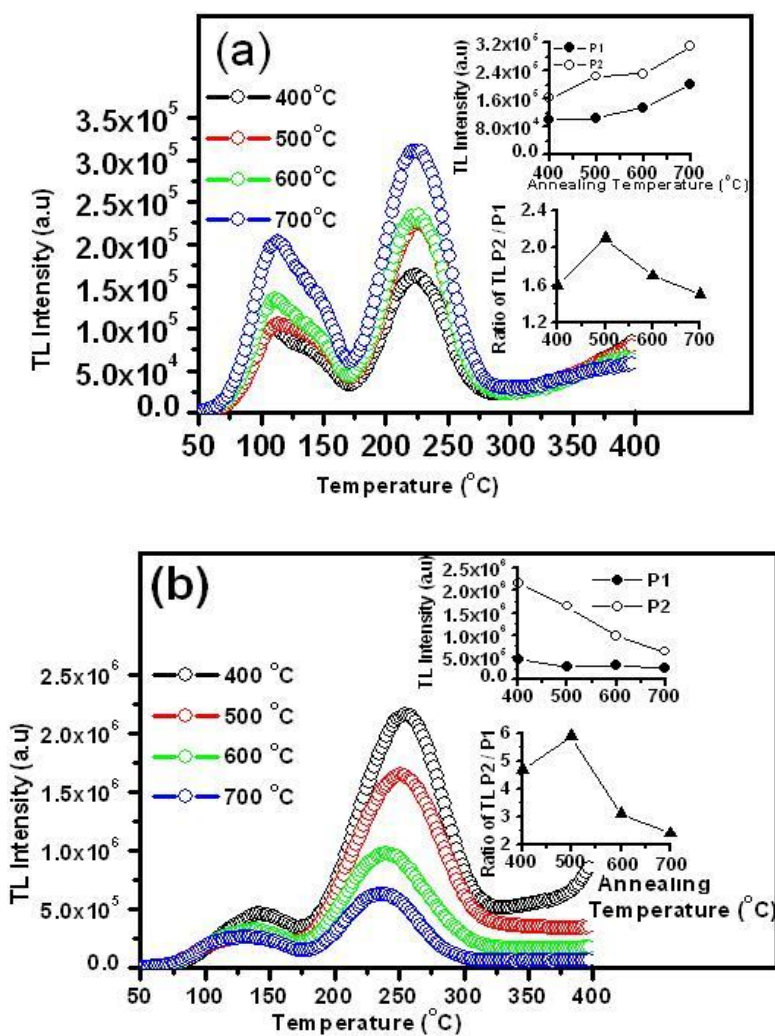
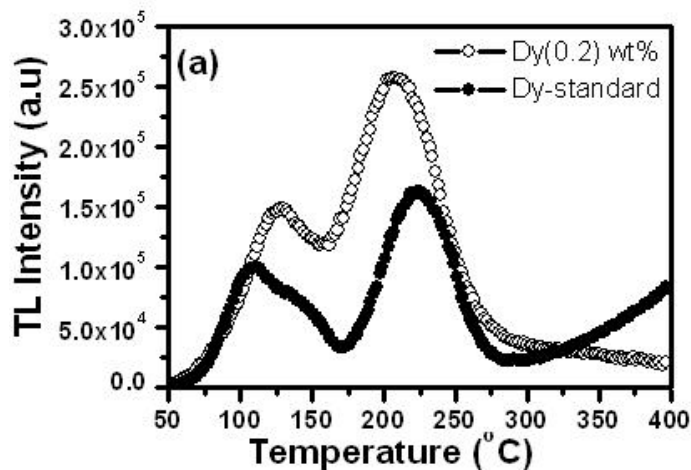


Fig. 2. Effect annealing temperature on TL glow curves of standard CaSO₄:Dy (curves (a)) and CaSO₄:Tm (curves (b)) a beta dose of 1 rad.



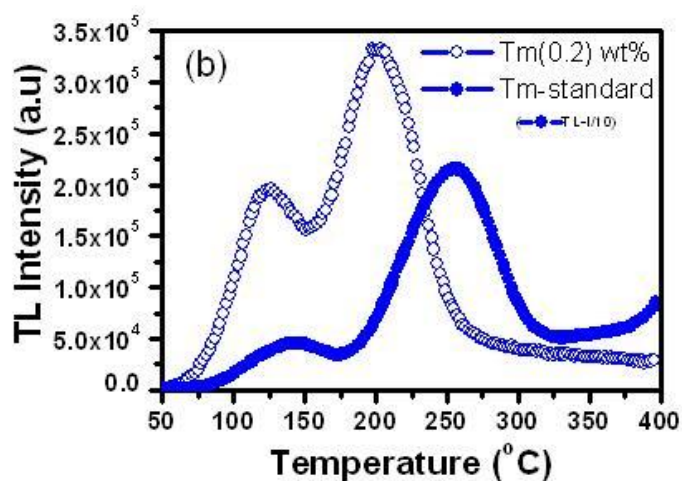


Fig. 3. TL glow curves of comparison between standard samples and synthesis samples: (a) CaSO₄:Dy, CaSO₄:Dy(0.2); (b) CaSO₄:Tm, CaSO₄:Tm(0.2) wt% for a Beta dose of 1 rad annealed at 400°C/1 h

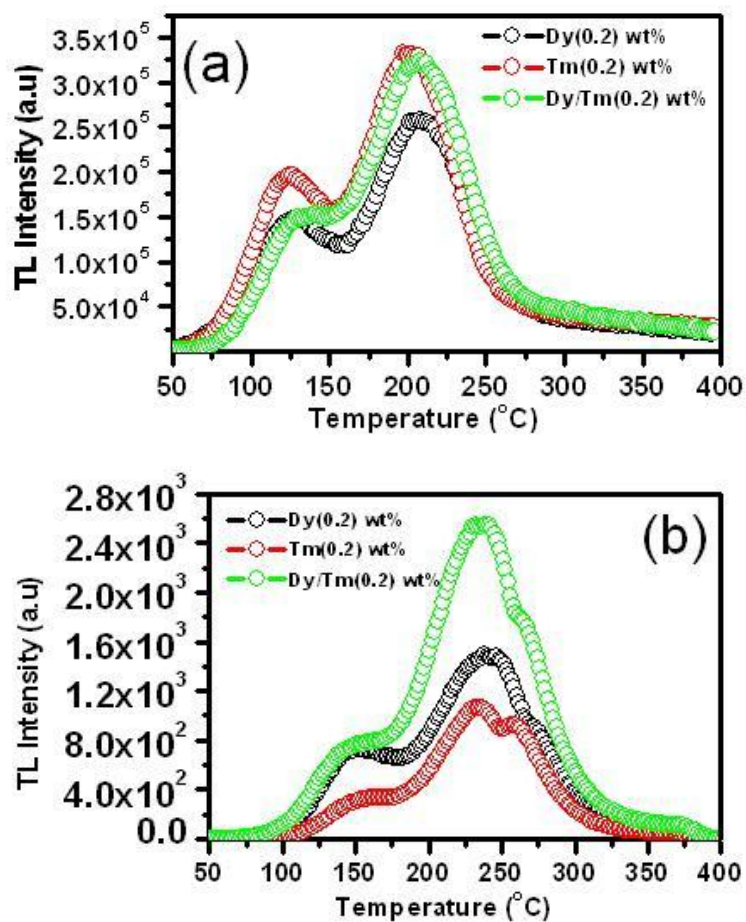


Fig. 4. TL glow curves for CaSO₄: Dy, CaSO₄: Tm and CaSO₄:Dy (0.2)/Tm (0.2) annealed at 400°C/1 h and irradiated with 1rad. (a) Stored samples. (b) Fresh work

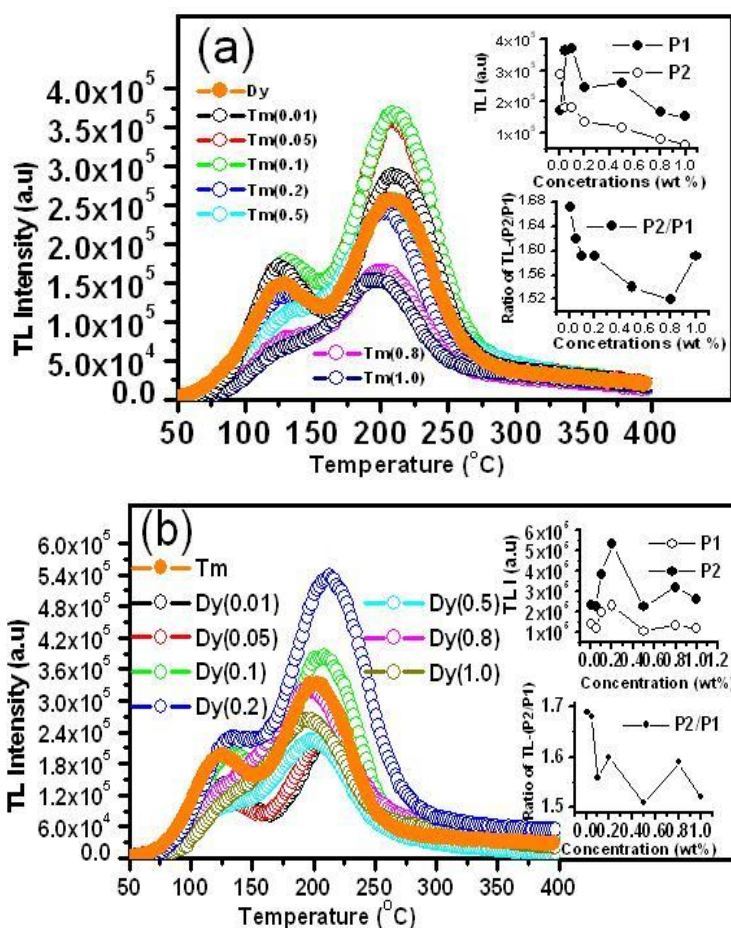
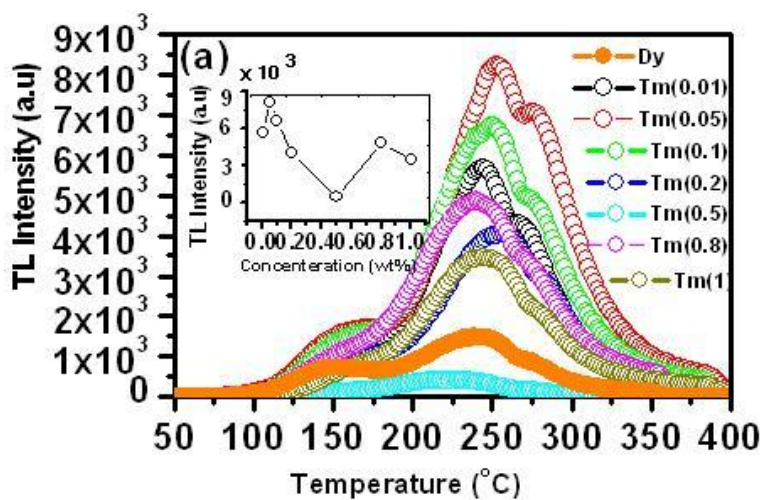


Fig. 5. TL glow curves for CaSO₄: Dy(0.1)/Tm(x) (curves a) and Tm(0.1)/Dy(x) (curves b) for x varying as shown for a Beta dose of 1 rad. [stored samples]



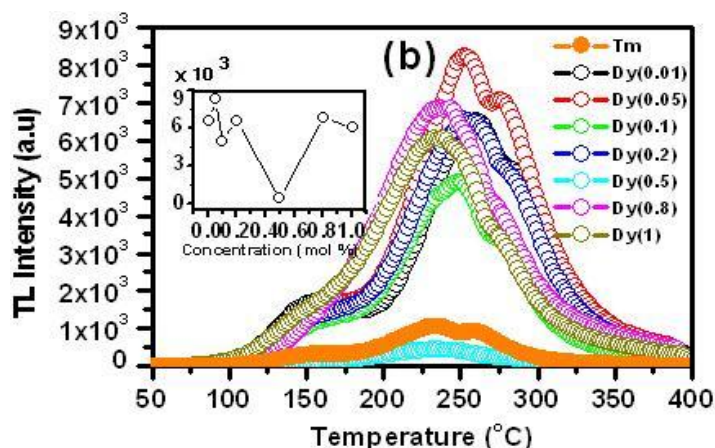


Fig. 6. TL glow curves for $\text{CaSO}_4: \text{Dy}(0.1)/\text{Tm}(x)$ (curves a) and $\text{Tm}(0.1)/\text{Dy}(x)$ (curves b) for x varying as shown for a Beta dose of 1 rad. [Fresh samples]

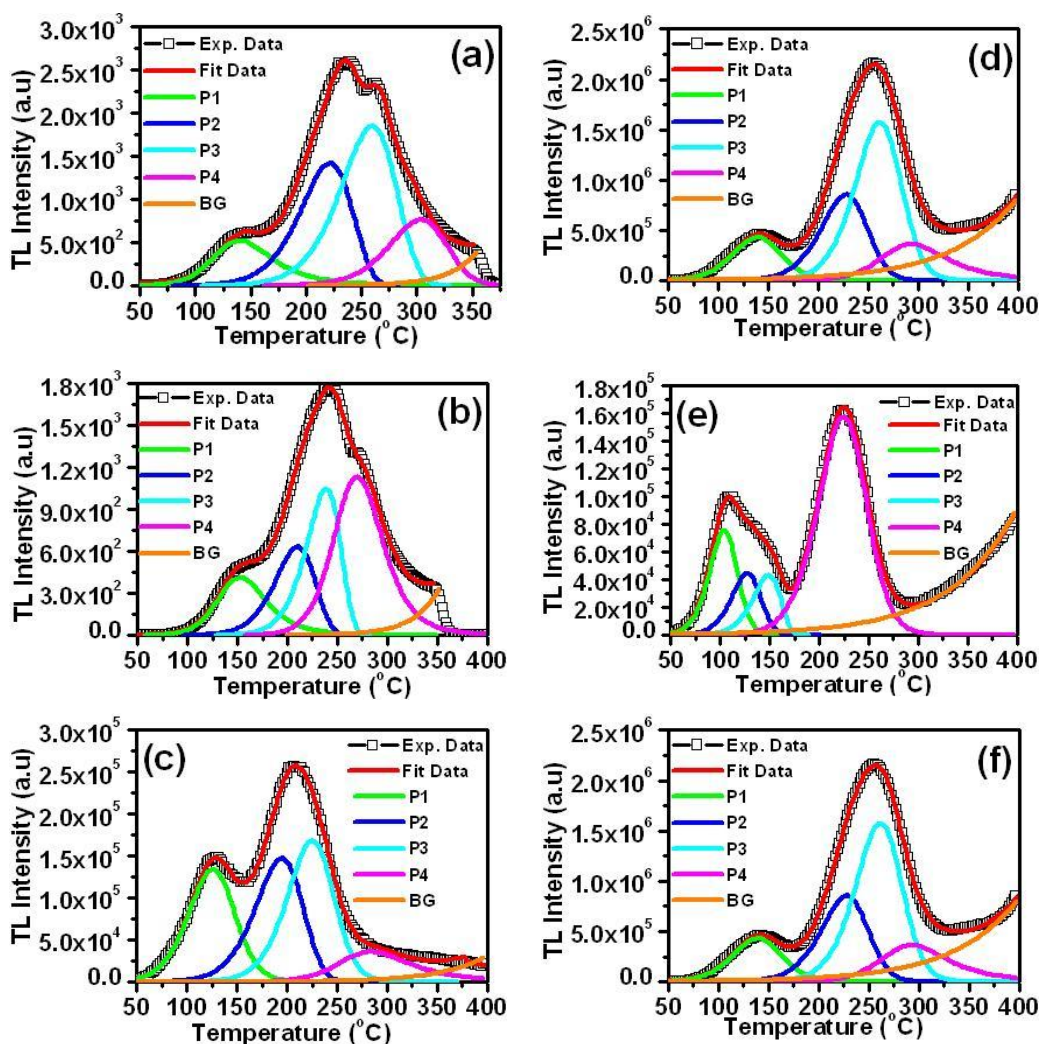


Fig. 7. De-convolution of the experimental TL glow curves of $\text{CaSO}_4: \text{Dy}$, Tm synthesis of a fresh, saved and standard sample [curves (a-f)] respectively

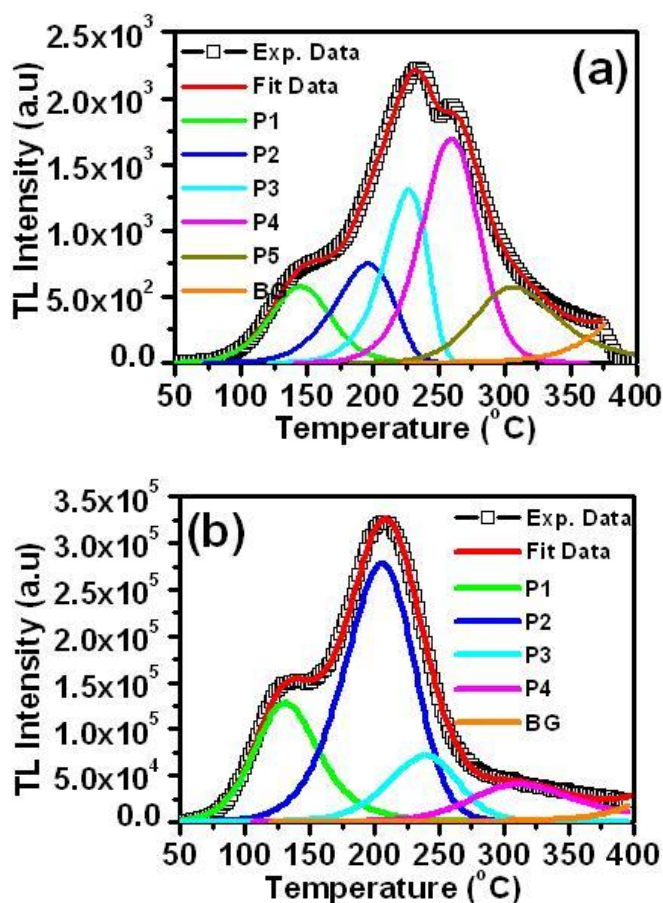


Fig. 8. De-convolution of the experimental TL glow curves of $\text{CaSO}_4:\text{Dy/Tm}$ synthesis of a fresh and saved sample [curves (a-b)] respectively

4. CONCLUSION

Glow curve structure of $\text{CaSO}_4:\text{Dy, Tm, Dy/Tm}$ undergoes reversible changes owing to the treatments in the temperature range from 200 to 700°C. The changes could be attributed to the interconversion of defect complexes responsible for the low temperature and main dosimetric glow peaks. Thermal treatment of 400°C for 1 h was found to be optimum for maximizing the dosimetric peak and minimizing the low temperature peak, which is of significance for its use in radiation dosimetry. Beyond 700°C, the change in the glow curve structure could be correlated to the partial phase transition in $\text{CaSO}_4:\text{Dy, Tm, Dy/Tm}$ phosphor of stored and $\text{CaSO}_4:\text{Tm}$ standard. While, thermal treatment of 500°C and 700°C for 1 h of a fresh and $\text{CaSO}_4:\text{Dy}$ standard sample were found to be optimum for maximizing the dosimetric peak and minimizing the low temperature peak, which

is of significance for its use in radiation dosimetry.

Trap distribution changes due to diffusion may be the main reason for the cause of the change in the binding energies of trapped charge carriers. Besides, trapped charged conversion and competing non-radiation defect interactions, storage may influence the fitted activation energy. Temperature dependence of frequency factor, variations of temperature lag and fluctuation and shift of the distribution frequency factor can be used to interpret the drift phenomenon of frequency factor. The variation of kinetic order during the heating stage can also be used to interpret the drift phenomenon of kinetic order.

COMPETING INTERESTS

Authors have declared that no competing interests exist.

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