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VARIATION IN RESPONSE AND PHYSICAL PROPERTIES OF SOME COMMERCIAL THERMOLUMINESCENT DOSIMETERS LiF (TLD-100), CaF₂: Dy (TLD-200) and CaSO₄: Dy (TLD-900) AFTER GAMMA IRRADIATION

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ABSTRACT

For routine dosimetry, several TLD materials have been used, each having its own definite properties. In this paper, the physical TL properties of the three used commercial thermoluminescent (TL) materials, [LiF (TLD-100), CaF₂:Dy (TLD-200) and CaSO₄:Dy (TLD-900)], have been studied in order to identify the efficiency of the optimum material for gamma radiation measurements. The main investigated dosimetric properties are glow curve structure, sensitivity, dose response, fading, memory effects and sensitization.

The experimental results show that the glow curve structure of the , TLD-100 includes 4 glow peaks at 135, 182, 210 & 230 °C , TLD-200 includes 2 peaks at 179 & 240 °C and TLD-900 includes 3 peaks at 123, 220 & 338 °C. The sensitivity of TLD-200 was found to be 9-20 times greater than the standard one (TLD-100), but for TLD-900, it was found 8-13 times greater than the standard one. It was found that the TL response of the three phosphors fit to a straight line in the logarithmic scale from, 50 μGy – 5 Gy , 50 μGy – 10 Gy and 5 μGy – 10 Gy, respectively. The fading values were found to be ≈ 7.5 & 20.1 % , 14 & 32% and 10.4 & 12 % for the three phosphors, respectively, after the storage for 3 months at 25 & 50 °C, respectively. From the results which previously mentioned, calcium Sulphate doped by dysprosium has been selected for local preparation.

Key words: Thermoluminescent Dosimeters, TL-Sensitivity, memory effects, LiF (TLD-100), CaF₂:Dy (TLD-200), CaSO₄:Dy (TLD-900).

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INTRODUCTION

A lot of materials have been suggested for use in thermoluminescence materials, some of which appear to show excellent properties. The main dosimetric characteristics such as glow curve structure, sensitivity, dose response, fading [1], memory effect and sensitization for the most commonly used TL materials [LiF (TLD-100), CaF₂:Dy (TLD-200) and CaSO₄:Dy (TLD-900)] were analyzed and compared [2].

Dosimetric grade LiF exists in a variety of forms, including LiF:Mg,Ti and LiF:Mg,Cu,P[3]. Various groups have reported that the dose response of the TLD-100 is characterized by the linear-supralinearity-sublinear response. Sensitivities are adequate for personnel dosimetric usage, with atypical lowest level of detection of 100 μ Gy. It is usual to refer to TLD-100 as the standard TLD materials and to assign it a sensitivity of 1.0 against which the sensitivities of the materials can be compared [4, 5].

Calcium fluoride comes in several forms, a wide of different impurities acting as activators such as Mn, Dy or Tm, or natural fluorite. Because of the high Z_{eff} for this materials, all CaF₂ TLDs exhibit a significant over response, which limiting their usefulness in personnel dosimetry [4]. The sensitivity of CaF₂:Dy is \approx 30 times greater than TLD-100 [6].

CaSO₄:Dy TL phosphor have found increasing use in various applications in the field of radiation dosimetry due to its high sensitivity[7], stability, reusability and ease of preparation[8, 9]as compared to many other sensitive TL phosphor. The sensitivity is dependent up on the concentration of Dy and optimum values lead to minimum measurable doses of 2 μ Gy [10, 11].

The objective of this work was to investigate the TL properties for the most commonly used TL materials [LiF (TLD-100), CaF₂:Dy (TLD-200) and CaSO₄:Dy (TLD-900)] to identify the efficiency of the optimum material for measurements in gamma radiation fields.

EXPERIMENTAL AND METHODS

The three commercial phosphors [LiF (TLD-100), CaF₂:Dy (TLD-200) and CaSO₄:Dy (TLD-900)]used in this study was obtained in conventional ribbon form (3.2 x 3.2 x 0.9 mm³), from the Harshaw chemical company. These phosphors was ground and sieved to obtain a powder with grain sizes from 75 – 180 μ m. The three phosphors were selected after 400, 500 and 600 °C annealing for 1 hr for each, respectively, which reveals good reusability and lowest residual TL with respect to gamma radiation. In every irradiation run, the mass of the sample dispensed was about 5 mg

for each TL read out and at least 10 measurements were made for each experiment. All measurements were taken 24 hr after the end of the irradiation.

The sources used for irradiation during this study were Cs-137, Co-60 gamma rays irradiators. The intensity of the Co-60 γ - source (Gamma chamber 5000) was 12 000 Ci, giving a dose rate of 6.02 KGy /hr. The intensity of Cs-137 γ - source was 1.5 Ci, giving a dose rate in air changes with the distance of the samples from the surface of the source as shown in fig.1. The irradiation conditions covered the regions from 5 μ Gy to 5×10^4 Gy.

A NABER muffle oven, model D-2804 with a temperature range from room temperature to 1100 °C, and was used for annealing and for applying thermal treatment procedures. A TLD reader model 4000 (Harshaw) was used for TL readout.

RESULTS AND DISCUSSION

Glow curve structure:

Fig. 2.a represents the glow curve structure of TLD-100 after pre-irradiation anneal at 400 °C for 1 hour, followed by cooling to room temperature, and irradiation with 1 Gy dose from ^{137}Cs source. The glow curve is characterized by 4 glow peaks at ~ 135, 182, 210 and 230 °C. The main glow peak is at 230 °C (for heating rate of 10 °C/sec). Fig.2.b represents the glow curve structure of $\text{CaF}_2:\text{Dy}$ (TLD-200), after pre-irradiation anneal at 500 °C for 1 hour followed by cooling to room temperature, and irradiated with 1 Gy dose from ^{137}Cs source. The glow curve of $\text{CaF}_2:\text{Dy}$ (TLD-200) consists of 2 glow peaks at ~ 179 and 240 °C. Fig.2.c represents the glow curve structure of $\text{CaSO}_4:\text{Dy}$ after pre- irradiation anneal at 600 °C / 1 hr. and irradiated with 1 Gy dose from ^{137}Cs source. Three glow peaks at 123, 220 and 338 °C were obtained. The main peak at 220 °C is observed independently of the dopant ion, but with minor differences in the low and high temperatures features.

Sensitivity:

The sensitivity of $\text{CaF}_2:\text{Dy}$ and $\text{CaSO}_4:\text{Dy}$ phosphors is compared to that of the standard LiF (TLD-100) phosphor instead of measuring its absolute sensitivity. The three phosphors were subjected to their own optimum annealing treatment before irradiation. The samples were irradiated with a dose from 5 μ Gy to 5×10^4 Gy as shown in table 1. and measured after 24 hours in order to stabilize the fading rate. LiF (TLD-100) used as the standard reference with relative sensitivity of 1.0

From table 1, the sensitivity of the $\text{CaF}_2:\text{Dy}$ (TLD-200) phosphor during this scheme irradiation was 9-20 times greater than the standard LiF (TLD-100) phosphor and the sensitivity of the $\text{CaSO}_4:\text{Dy}$ (TLD-900) phosphor was 8-13 times greater than (TLD-100) during this irradiation scheme.

Dose Response:

This experiment was carried out to study the TL-response of LiF (TLD-100), CaF₂:Dy (TLD-200) and CaSO₄:Dy (TLD-900) phosphors to different doses of ionizing radiation with a dose ranges from 5×10^{-6} Gy to 5×10^4 Gy .

Fig.3.a shows the TL response of LiF (TLD-100) as a function of irradiation doses in the above ranges. It is found that there is a linear TL-response in the range from minimum detectable exposure dose 5×10^{-5} Gy to about 5 Gy. Above 5 Gy, the response is supralinear up to 2×10^3 Gy and the saturation starts from 5×10^2 Gy.

Fig.3.b shows that the TL response of CaF₂:Dy (TLD-200) is linear in the range from minimum detectable exposure dose 5×10^{-5} Gy up to about 10 Gy . Above 10 Gy, the response is supralinear up to 2×10^2 Gy and the saturation starts from 100 Gy.

Fig.3.c shows the TL response of CaSO₄:Dy (TLD-900) phosphor is linear in the range from minimum detectable exposure dose 5×10^{-6} Gy up to about 10 Gy. Above 10 Gy, the response is supralinear up to 2×10^3 Gy and the saturation starts from 10^2 Gy up to 5×10^4 Gy and after that the TL- response is decreasing.

Fading:

The storage effect (fading) on the TL response of the LiF (TLD-100), CaF₂:Dy (TLD-200) and CaSO₄:Dy (TLD-900) phosphors has been studied. The samples of each type were annealed at their own optimum condition, then irradiated with 0.1 Gy gamma dose, and then stored for a time extending to 90 days under both room temperature (~ 25 °C) and 50 °C in a dark box and oven respectively.

Fig.4.a shows the relation between the relative TL response (%) of LiF (TLD-100) and storage time in days. It is clear from this figure that the fading is time and temperature dependent, in which it reaches 7.5 ± 1.15 % at room temperature (25 °C) and 20.1 ± 3.6 % at 50 °C. Figure (4.b) shows the fading of CaF₂:Dy (TLD-200), the initial fading rate is relatively high, the fading is approximately 8 ± 2.2 % and 16 ± 3.8 % at room temperature (25 °C) and at 50 °C respectively in the first 10 days, and reaches 14 ± 2.7 % and 32 ± 8.3 % respectively near the end of the period (90 days).

Fig.4.c shows the fading of CaSO₄:Dy (TLD-900), the fading is 10.4 ± 1.8 % and 12 ± 2.2 % for a storage period of 90 days at room temperature (25 °C) and at 50 °C respectively. The rate of fading of CaSO₄:Dy is less than the rate of fading of CaF₂:Dy at room temperature and at 50 °C, and less than the fading of LiF at 50 °C. The rate of fading of TLD-100 is dependent on a variety of parameters such as: storage temperature, annealing, radiation type, and glow curve integration region.

Memory Effect:

Fig.5. shows the effect of previously absorbed dose, from 0.1 Gy to 10^3 Gy, on the TL efficiency of LiF (TLD-100), CaF₂:Dy (TLD-200) and CaSO₄:Dy (TLD-900) phosphors. From this figure, the TL response of LiF (TLD-100) increases sharply as the previous absorbed dose increases and reaches the maximum response at 1000 Gy pre-irradiation dose .

The TL response of CaF₂:Dy (TLD-200) increase up to 90 Gy and attains a maximum response at 100 Gy . Further increase in the previous absorbed dose results in a decrease in TL response. At 1000 Gy pre-irradiation dose the TL response is nearly equal to the response at 0.1 Gy pre-irradiation doses. After high pre-irradiation dose, radiation damage effects in the dosimeters may change the response either by a reduction or a creation of electron traps, which results in a lower response or in a sensitization of the TL material.

The TL response of CaSO₄:Dy increases slowly with the increase in the pre-irradiation dose up to 100 Gy, after that the increase in TL response is high with the increase in the previously absorbed dose until CaSO₄:Dy reaches the maximum response at 1000 Gy (8.4 times greater than with 0.1 Gy pre absorbed dose)

Sensitization Radiation:

The effect of previous exposure to γ -rays on TL response of LiF (TLD-100), CaF₂:Dy (TLD-200) and CaSO₄:Dy (TLD-900) phosphors have been studied here .Fig.6.a shows the relation between the pre-irradiation doses (1 Gy, 100 Gy and 1000 Gy) on TL response of LiF (TLD-100) at different doses as follows: 10^{-3} Gy, 10^{-2} Gy, 0.1 Gy, and 1 Gy, respectively. From this figure, it is clear that the TL response increases as the pre irradiation doses increases for the four different doses, but it increases highly at the dose 10^{-2} Gy (The response at 1000 pre-irradiation dose is 2.6 times greater than that without sensitization) and the lowest TL response is at a dose of 1 Gy.

Fig.6.b shows that up to 10^2 Gy pre-irradiation dose, the rate of increase in the TL response of CaF₂:Dy (TLD-200) is very small as compared with TLD-100. After that as the pre-irradiation dose increase above 10^2 Gy the TL response is decreased. At the doses 10^{-3} , 10^{-2} and 0.1 Gy there is nearly no effect of the pre-irradiation dose on the TL response.

Fig.6.c shows that the TL-response increases slowly as the pre- irradiation dose increases up to 10^2 Gy, after that the TL response increases highly until it reaches the maximum value at 10^3 Gy pre-irradiation dose . At 10^3 Gy, as the dose increases the value of sensitization is decreased. At 1 Gy pre-irradiation dose, there is no effect on TL-response at a dose of 1 Gy and the TL-response is decreased to 0.86 % at 0.1 Gy γ - dose.

CONCLUSIONS

This paper concerned with the variation of the three used commercial thermoluminescent materials, [LiF (TLD-100), CaF₂: Dy (TLD-200) and CaSO₄: Dy (TLD-900)], in response to gamma radiation and their dosimetric characteristics which lead to choose a phosphor for locally prepared. The results of the dosimetric characteristics of the three phosphors showed that CaSO₄: Dy (TLD-900) is a highly sensitive and stable TL Dosimeter, which meets the most important requirements for radiation measurements. TLD-900 has a TL response which fits a straight line on the logarithmic scale from 5 μGy to 10 Gy and a fading rate of about 10.4 & 12 % after storage for 3 months at 25 °C & 50 °C, respectively. According to these results, TLD-900 has been selected for locally prepared.

REFERENCES

- [1] Bacci, C.; Furetta, C.; Rispoli, B.; Raubaud, G.; and Tuyn, J.W.N.; "The Effect of storage Temperature on the thermoluminescence Response of some phosphors ", Radiat. Prot. Dosim., 43, 25, (1988).
- [2] Prokic, M.; and Botter – Jensen, L., "Comparison of Main Thermoluminescence properties of some TL Dosimeter ", Radiat. Prot. Dosim., 195, 47, (1993).
- [3] Mohamed, A.H.H; "Preparation and dosimetric properties of LiF (Mg,Cu,P) TLD, And its application in radiation protection", Ph.D. in Science (Physics); Ain Shamss University; Faculty of Girls for Art, Science & Education, (1997).
- [4] McKeever, S.W.S.; Chen, R., "Theory of thermoluminescence and related phenomena", published by World Scientific Publishing Co. Pte.Ltd. (1997).
- [5] Wu, D.K.; Sun,F.Y. & Dai, H.C.; " A high sensitive LiF thermoluminescence dosimeter ", Health Physics, 64 , P.P. 1063-1067, (1984).
- [6] Weng, P.S.; Li, S.H.; Hsu, P.C.; Hsu, K.C.; " Response of thermoluminescence dosimeter CaF₂:Dy to ionizing and non ionizing radiations ", Int. J.Appl. Radit. Isot. , 43, P.P. 717-720, (1992).
- [7] Mathur, V.K.; Lewandowski, A.C.; Gauradala, N.A.; Price, J.L.; "High dose measurements using thermoluminescence of CaSO₄: Dy", Radi. Meas., 735-738, 30, (1999).
- [8] Azorin, J.; and Gutierrez, A.; " Preparation and performance of a CaSO₄:Dy, Tm Thermoluminescence phosphor for long-term Gamma Measurements ", Health Physics, 56 (4), P.P. 551-559, (1989).
- [9] Yamashita, Y.; Nada, N.; Anichi, H. and Kitamura, S.; "Calcium Sulphate activated by thulium or dysprosium for thermoluminescent dosimetry ", Health Phys., 295, 21, (1971).
- [10] Guelev, M. G.; Mischev, I. T.; Burgkhardt, B.; and Piesh, E.; " A Two- element CaSO₄:Dy Dodimeter for Environmental Monitoring ", Radiat. Prot. Dosim., 35, 51, (1994).
- [11] Lakshmanan, A.R.; Shinde, S.S.; Bhatt, R.C.; and Supe, S.J.; "Grain size and Dy Concentration Effects in Thermoluminescent CaSO₄:Dy ",Radiat. Prot Dosim., 173, 22, (1988).

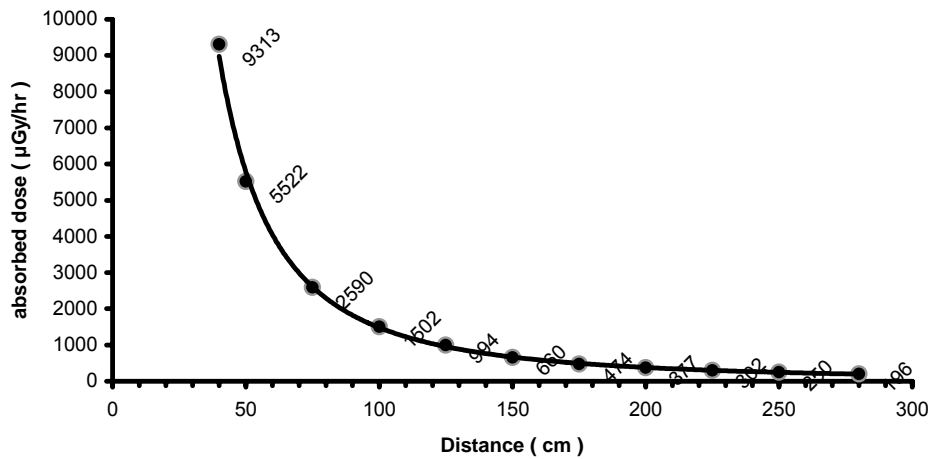


Fig.1. Absorbed dose rate changes with the distance of the samples from the surface of the source

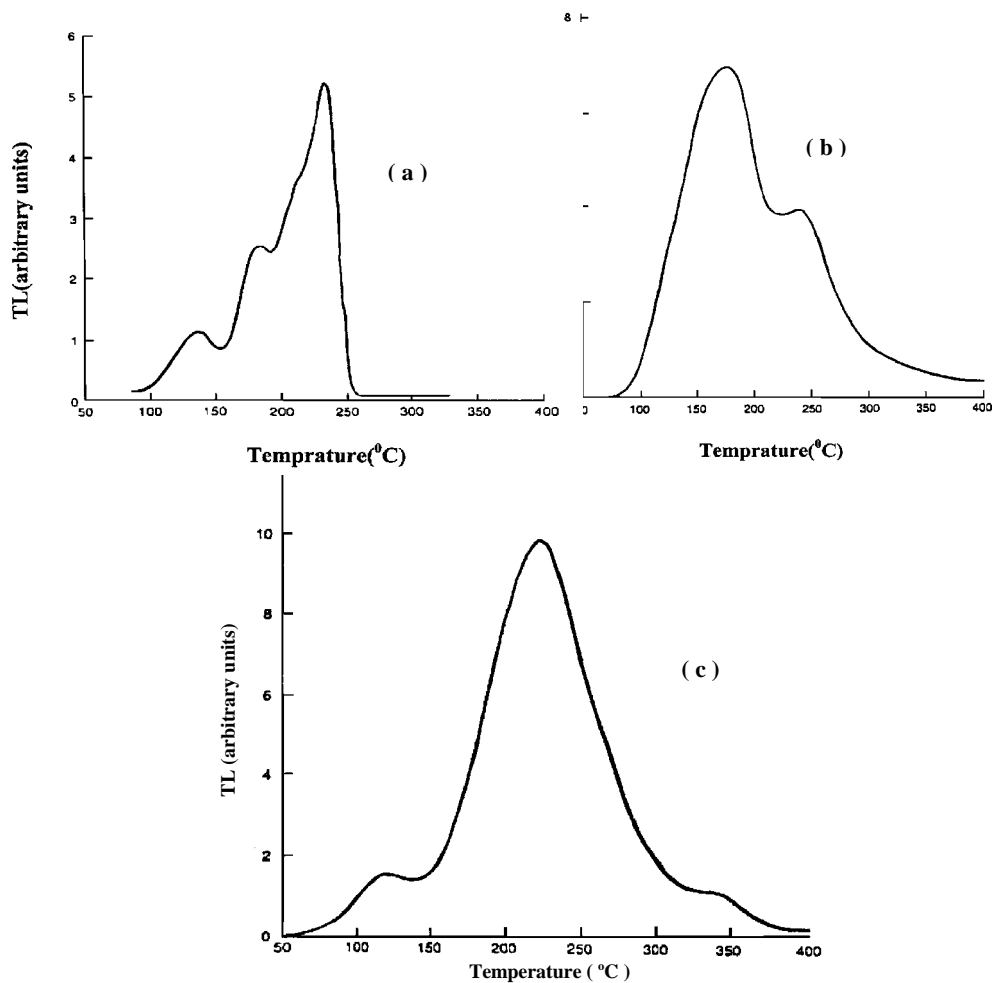


Fig. 2. Glow curve structure of the three phosphors: (a) LiF, (b) CaF₂: Dy, (c) CaSO₄: Dy after irradiating with 1 Gy from ¹³⁷Cs at room temperature and the heating rate was 10 °C/sec.

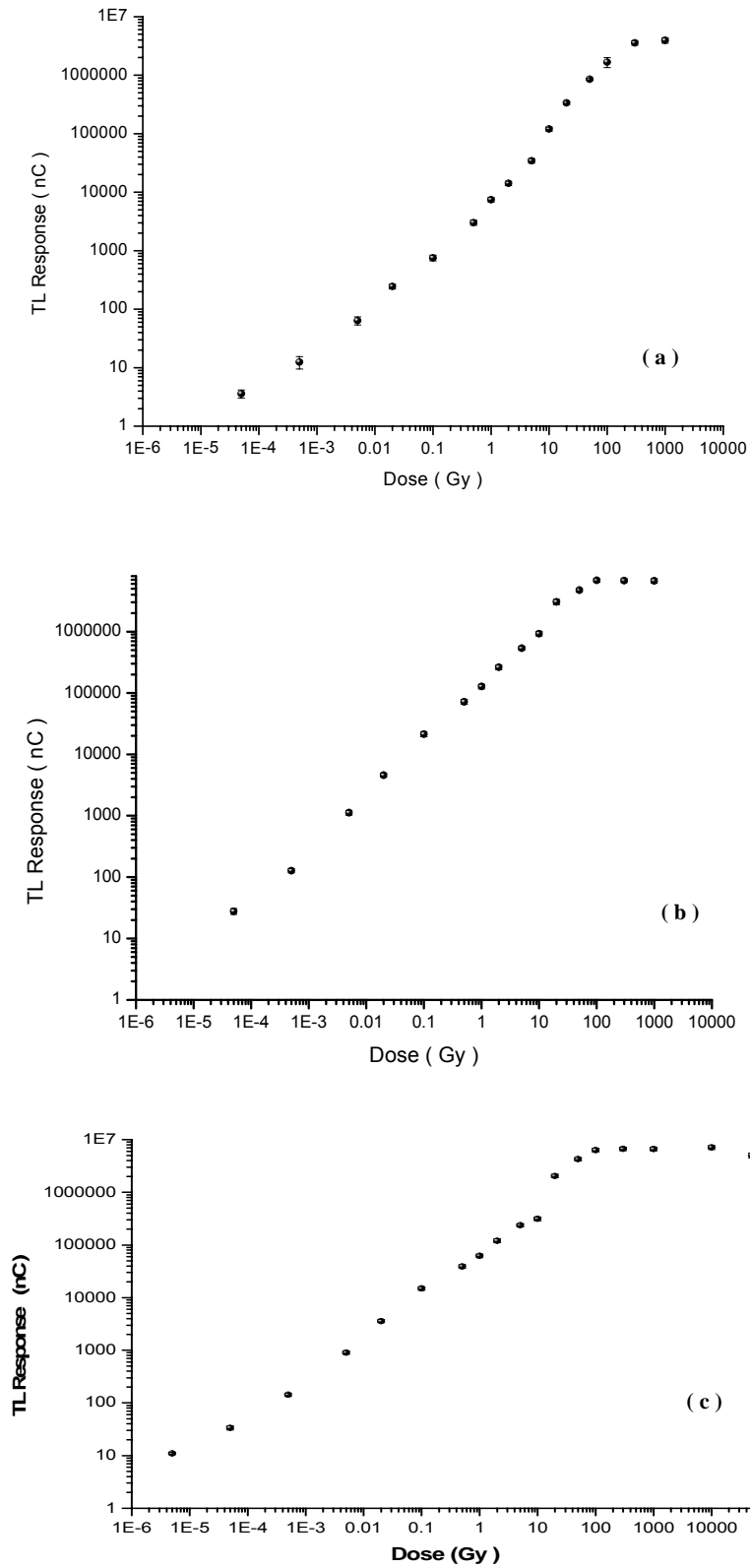
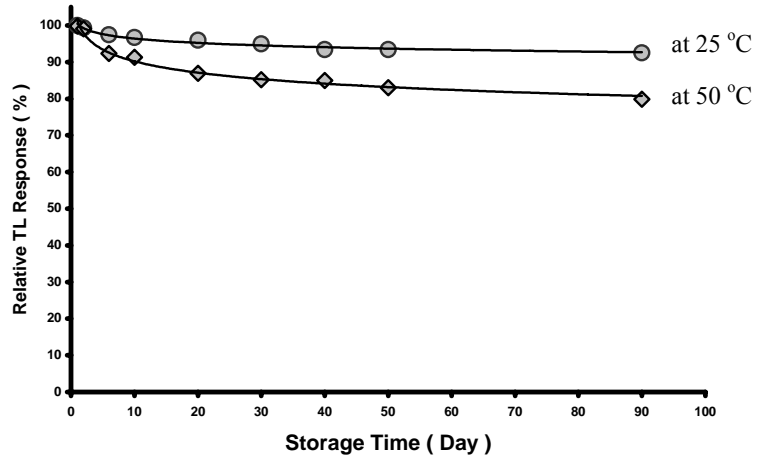
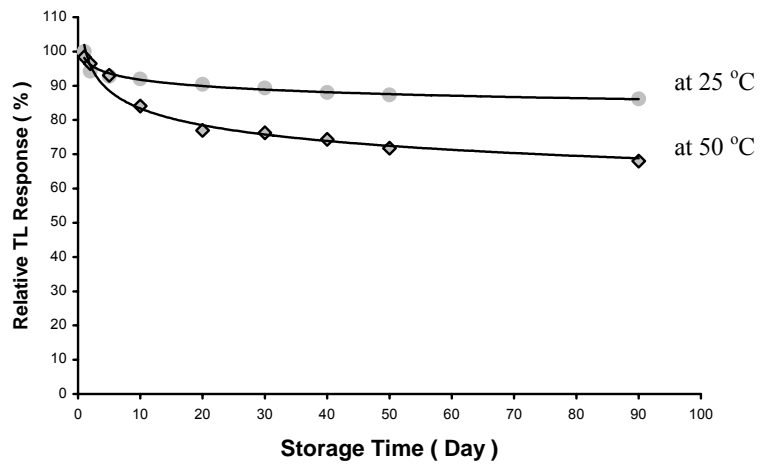


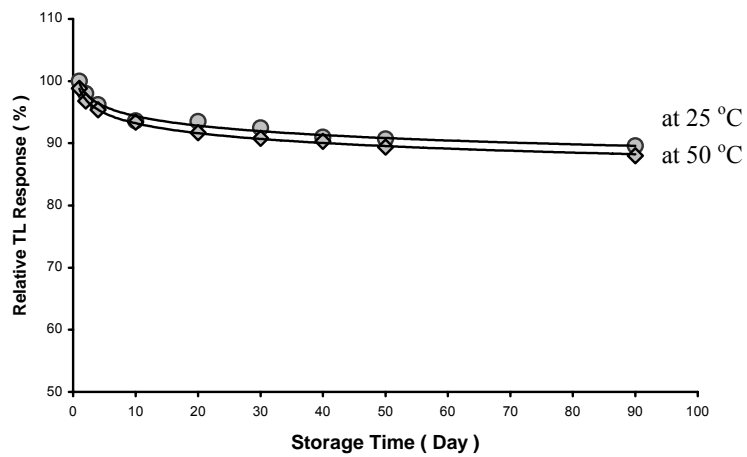
Fig.3. TL-response of the three commercial phosphors :
 (a) LiF (TLD-100), (b) CaF₂:Dy (TLD-200) and (c) CaSO₄: Dy (TLD-900)
 as a function of the irradiation dose in gray



(a)



(b)



(c)

Fig.4. The percentage of the TL sensitivity of the three phosphors: (a) LiF (TLD-100), (b) CaF₂: Dy (TLD- 200), (c) CaSO₄: Dy (TLD-900) as a function of storage time and temperature at 25 °C & 50 °C.

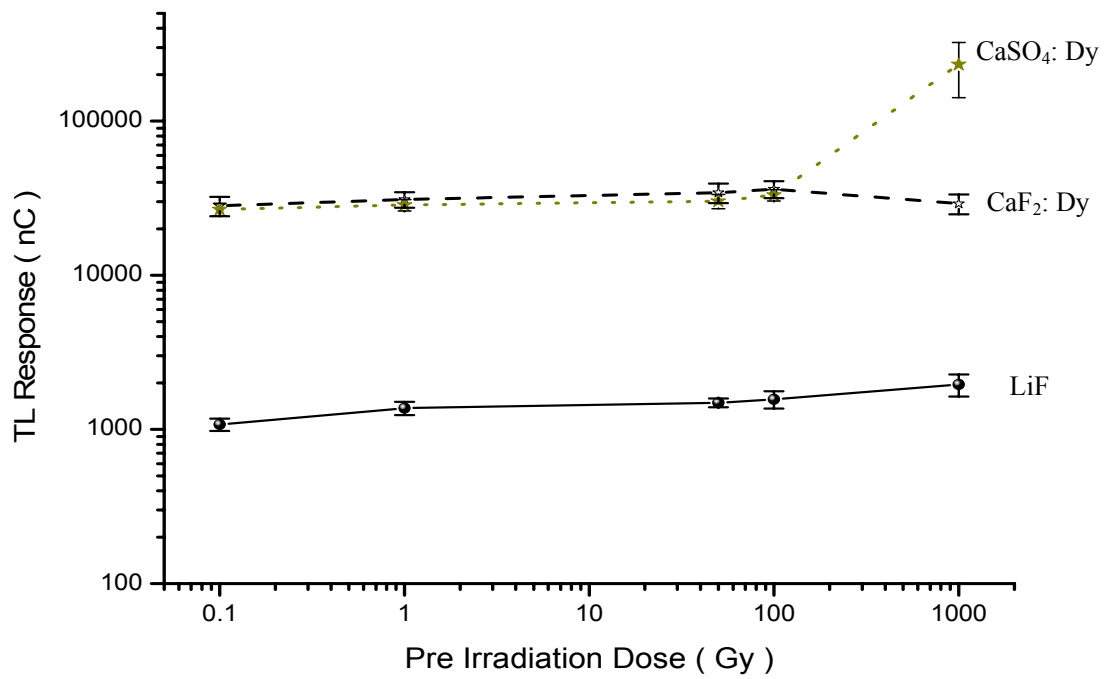


Fig. 5. The effect of previously absorbed dose on the TL efficiency of the three phosphors, LiF (TLD-100), CaF₂: Dy (TLD-200) and CaSO₄: Dy (TLD-900) at a test gamma dose of 0.1 Gy.

Table 1. Sensitivity of the LiF (TLD - 100), CaF₂:Dy (TLD - 200) and CaSO₄:Dy (TLD - 900) as a function of the irradiation dose.

Dose (Gy)	LiF (TLD - 100)	CaF ₂ :Dy (TLD - 200)	CaSO ₄ :Dy (TLD - 900)
5 x 10 ⁻⁶	-	-	11.1
5 x 10 ⁻⁵	3.6	27.7	33.7
5 x 10 ⁻⁴	12.5	127.3	143
5 x 10 ⁻³	64	1 123	902
2 x 10 ⁻²	245	4 587	3 582
0.1	755	21 376	14 880
0.5	3 021	71 978	39 105
1	7 454	127 688	62 320
2	14 214	262 123	120 141
5	34 487	537 234	238 052
10	108 231	926 972	315 223
2 x 10 ¹	335 688	3 042 835	2 048 287
5 x 10 ¹	853 188	4 749 497	4 258 544
10 ²	1 671 078	6 846 420	6 348 715
5 x 10 ²	3 571 692	6 797 522	6 715 523
10 ³	3 946 920	6 721 045	6 656 060
10 ⁴	-	-	7 117 097
5 x 10 ⁴	-	-	5 015 884

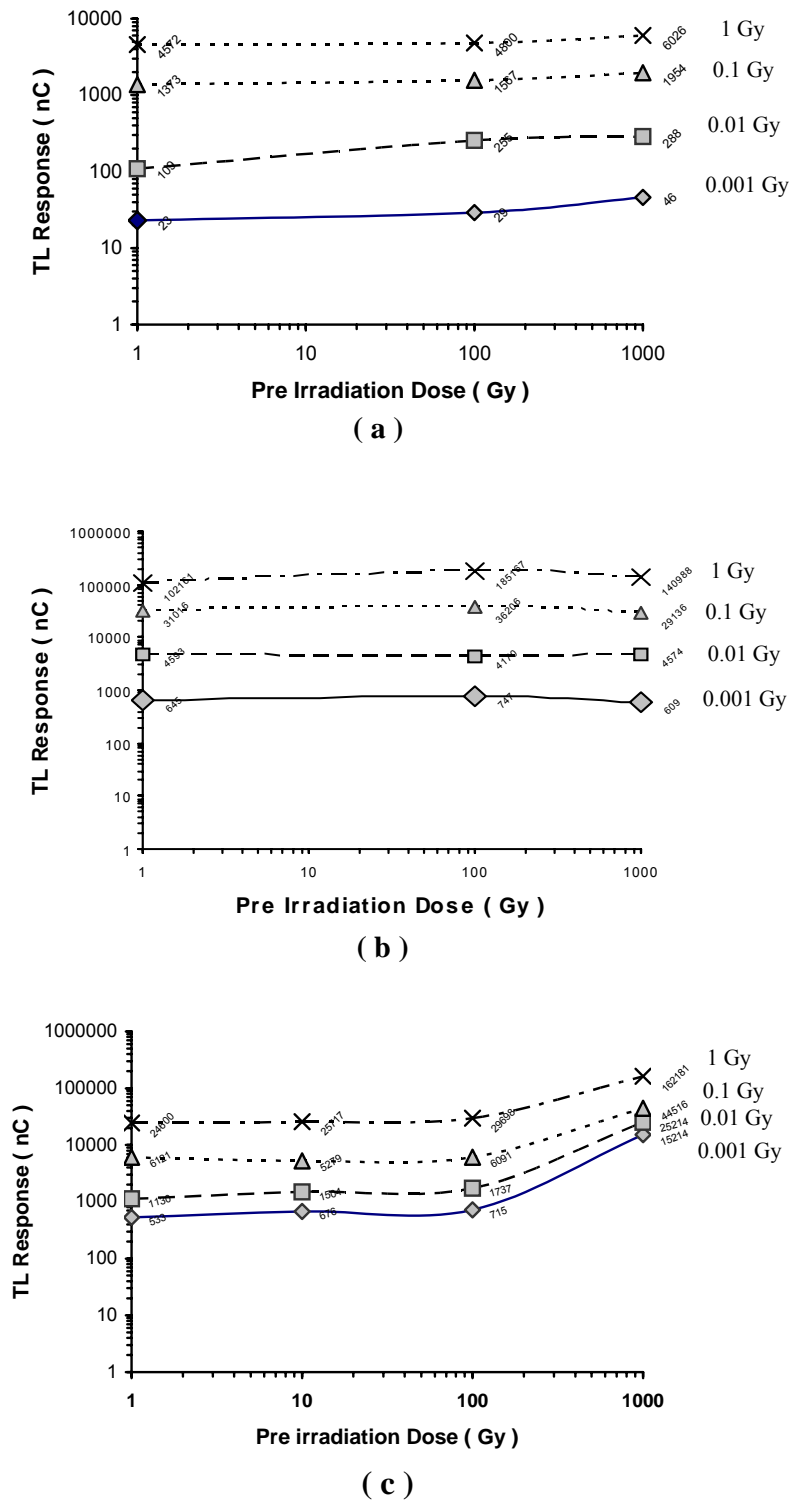


Fig. 6. TL-response of the three phosphors: (a) LiF (TLD-100), (b) CaF₂: Dy (TLD-200), (c) CaSO₄: Dy (TLD-900) (normalized to that response without sensitization as unity) as a function of sensitizing radiation.