# STUDYING AND EXAMINING DOSIMETRIC PARAMETERS OF THERMOLUMINESCENT DOSIMETER USING $CaSO_4$:Dy POWDER PRODUCED AT THE DALAT NUCLEAR RESEARCH INSTITUTE FOR PERSONAL RADIATION DOSIMETRY

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*The research group of the Nuclear Research Institute (NRI) studied and successfully produced thermoluminescent dosemeters (TLDs) using powder material of $CaSO_4$:Dy. For external personal dosimetry in quantity of Hp(10), dosimetric parameters of the TLDs are examined by irradiating with various gamma doses of $^{137}$Cs source and measuring by Rexon-320 reader, including glow curve with temperature, calibration factor (or response value), homogeneity of the batch, reproducibility of measurement, linearity of dose response, limit of detection, fading, light sensitivity, dose-rate dependence, and energy dependence of the response, etc. The study results showed that the dosemeters are ensured for personal dosimetry according to the standards of the IEC-61066:2006. Besides, the TLDs are also irradiated with standard doses of $^{137}$Cs and X rays at the Secondary Standards Dosimetry Laboratory (SSDL) in Hanoi. Comparison results on dose calibrated by the NRI and the SSDL show that the TLDs have been confident. The main advantage of them is that their price has been cheaper in comparison with the same TLDs using $CaSO_4$:Dy solid cards as well as TLDs using other solid cards (as LiF) imported from foreign countries. Therefore, they have been used for routinely external personal dosimetry for radiation workers in the NRI and other radiation installations.*

***Keywords:*** *Thermoluminescent dosemeter, Hp(10), glow curve, calibration factor, fading*

## 1. Introduction

Due to the needs in external personal dosimetry in quantity of Hp(10) for radiation workers contacting with radiation sources at Dalat Nuclear Research Institute (DNRI) and other radiation installations, the research group of DNRI carried out studies and successfully produced thermoluminescent dosimeters (TLDs) using powder material of $CaSO_4$:Dy. The procedure of producing $CaSO_4$:Dy power is implemented as follows [1, 2]:

Basic raw materials: Pure $H_2SO_4$ with 1.84 g/cm$^3$ density, $CaSO_4$ with 99.9% purity, $Dy_2O_3$ (Merck), tri-distilled water (obtained by distillations in usual glassware-and quartz).

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The $CaSO_4$ matrix is activated by a carefully controlled the impurification procedure with Dy atoms, which play the role as thermoluminescence (TL) centers. In this work, $Dy_2O_3$ powder is dissoluted in diluted $H_2SO_4$ (<30%) and tri-distilled water to obtain the dysprosium sulfate solution. The $CaSO_4$ salt is dissolved in the obtained solution, in such an amount as to ensure a controlled Dy impurification of 0.15%. The mixture solution then is evaporated very slowly (20 hours) in air atmosphere at $100^oC$, until a solid crystal is obtained. Temperature is maintained and controlled by using a sand.

After the synthesis procedure, a supplementary thermal treatment is performed in order to remove $H_2SO_4$ traces and interstitial water, which cause poor reproducibility of TL powder from the crystalline matrix in the usual procedures. The treatment is carried out by heating the material at temperature of $700^oC$ for two hours and $400^oC$ for 20 minutes in quartz glassware closed in an electrical oven. The obtained product is the very pure $CaSO_4$:Dy with structure of a hexagonal crystal having pin bright in color. Then, the crystal is crushed in an agate mortar and the powder is separated by sieving in order to select crystals having dimensions of 70 ÷ 200 μm. Finally, the second thermal treatment for TL powder of $CaSO_4$:Dy is carried out at temperature of $400^oC$ for two hours in an oven.

## 2. Experiment and result

### *2.1. Irradiation and measurement of dosimetric parameters*

Experimental procedure established for examining dosimetric parameters consists in the followings. The powder is weighed with amount of 25 mg by a powder dispenser and then is poured into a cylinder resin capsules in black colour. The capsules (as TLDs) are irradiated in a panoramic geometry inside standard irradiation fields of: (i) 662 keV ($^{137}Cs$) gamma rays within dose interval of 0.05–24.24 mSv; Filtered X-rays (Pantak standard X-ray generator) with energies of ISO N40 (33 keV), ISO N60 (48 keV) and ISO N80 (65 keV) for interval dose of 0.3–1.0 mSv. Responsible capacity of TLDs expressed as measured values of equivalent dose (Sv) of Hp(10) is determined by using calibrated Rexon-320 Reader.

Rexon-320 reader with four-step consecultive heating cycle of 30 seconds is as follows: step 1 (increasing from room temperature up to $135^oC$ for 6 seconds), step 2 (keeping at $135^oC$ for 4 seconds), step 3 (increasing from $135^oC$ up to $280^oC$ for 12 seconds as heating rate of $12^oC$/second), and step 4 (keeping at $280^oC$ for 8 seconds). The TL curve of the TLDs ($CaSO_4$:Dy powder) contains the main TL peak at about $220^oC$ and two secondary TL peaks at $70^oC$ and $90^oC$. After short regeneration treatment, the TL peak at $70^oC$ is eliminated in Figure 1 and 2, which show photographs of Rexon-320

Reader and glow curve for a TLD, respectively.


**Figure 1**: *Rexon-320 reader*

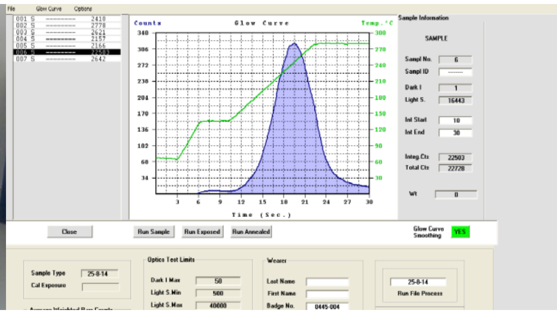


**Figure 2:** *Glow curve for sample of $CaSO_4$:Dy powder*

***2.2. Response value (calibration factor)***

Using 25 annealed TLDs for irradiate with conventional true equivalent dose ($D_{ct}$) of 2.1 mSv generated by the reference source of $^{137}$Cs (gamma activity about 170 mCi in Sep. 2014). Experimental values of dose-rate with different distances from the source to TLDs are measured by Farmer Dosimeter NE 2570/1B) at the Tertiary Standard Dosimetry Laboratory of DNRI. Then, TL signals of all TLDs are measured by Rexon-320 reader. The mean value of the data is $\overline{X}$ = (17370 ± 608) signals with a coverage factor of 2 (corresponding to a 95% confidence level).

Response value (R) is expressed as [1]:

$$R = \frac{\bar{X}}{D_{ct}} \quad (1)$$

So, R is calculated as R = 8281 ± 290 (3.5%) sig./mSv.

***2.3. Homogeneity of the batch (Uniformity)***

Homogeneity of TLD batch (H) is expressed as [1]:

$$H(\%) = \frac{(X_{max} - X_{min})}{X_{min}}.100 \quad (2)$$

From the maximum and minimum X values measured in the equation 2, value of H = 8.2% is obtained.

So, this value is satisfying according to the IEC-61066:2006 standard (H ≤30%) [3].

***2.4. Linearity of the response within the interval dose and uncertainty***

All the selected TLDs grouped on a set of 6 pieces are irradiated at conventional true equivalent dose values ($D_{ct}$) with the interval dose of 0.05–24.24 mSv. The measured dose values ($D_{meas}$) versus $D_{ct}$ values are presented in Table 1 (Due to small activity of $^{137}$Cs source, one could not irradiate TLDs with higher doses).

Uncertainty is defined as [1]:

$$\varepsilon\ (\%) = \frac{D_{meas} - D_{ct}}{D_{ct}}.100 \quad (3)$$

$D_{meas}$ values are calculated as mean values from data obtained with 6 exposured TLDs by using the value of R = 8281 ± 290 (3.5%) sig./mSv above. Table 1 shows the values of $D_{meas}$ using the TLDs irradiated at various $D_{ct}$ values with the interval of 0.05–24.24 mSv.

**Table 1:** *Measured equivalent dose values for TLDs ($D_{meas}$) irradiated at various $D_{ct}$ values within the interval dose of 0.05–24.24 mSv*

| No. | $D_{ct}$ (mSv) | $D_{meas}$ (mSv) | ε(%) | No. | $D_{ct}$ (mSv) | $D_{meas}$ (mSv) | ε(%) |
|---|---|---|---|---|---|---|---|
| 1 | 0.005 | 0.006 | -16.4 | 12 | 0.766 | 0.744 | -2.9 |
| 2 | 0.010 | 0.011 | -14.3 | 13 | 1.128 | 1.244 | +10.3 |
| 3 | 0.020 | 0.019 | -6.0 | 14 | 2.926 | 2.594 | -11.3 |
| 4 | 0.047 | 0.045 | -4.7 | 15 | 3.638 | 3.168 | -12.9 |
| 5 | 0.093 | 0.089 | -4.2 | 16 | 4.978 | 4.592 | -7.8 |
| 6 | 0.154 | 0.137 | -11.2 | 17 | 6.156 | 5.726 | -7.0 |
| 7 | 0.192 | 0.166 | -13.2 | 18 | 8.303 | 8.103 | -2.4 |
| 8 | 0.262 | 0.236 | -9.8 | 19 | 11.932 | 11.501 | -3.6 |
| 9 | 0.375 | 0.357 | -4.8 | 20 | 14.564 | 14.239 | -2.2 |
| 10 | 0.437 | 0.416 | -4.9 | 21 | 18.686 | 17.556 | -6.0 |
| 11 | 0.628 | 0.598 | -4.7 | 22 | 24.244 | 23.254 | -4.1 |

From the data of Table 1, variation of $D_{meas}$ for various $D_{ct}$ is shown in Figure 3.

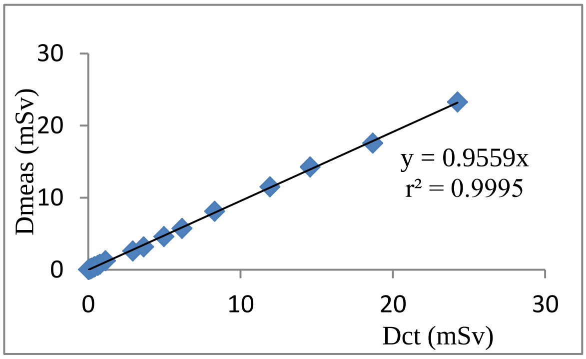


**Figure 3:** *Variation of measured equivalent dose of TLDs for various $D_{ct}$*

By analyzing the data from Table 2, it could be concluded: (i) Within the interval dose of 0.05 - 0.133 mSv, the maximum the uncertainty is 13.6%. This is a good result for low equivalent dose (by comparison with literature data, when 50% uncertainty values are reported); (ii) Within the interval dose of 0.133-4.978 mSv, the maximum the uncertainty is 14.6%; (iii) Within the interval dose of 4.978-24.244 mSv, the maximum the uncertainty is 7%. From calibration curve of Figure 3 calculated as linear regression of the data in Table 1, values of $D_{meas}$ could be examined by $D_{meas} = (0.956 \pm 0.147).D_{ct}$ or $(0.956 \pm 15\%).D_{ct}$.

It may be concluded that the obtained TLDs could quite accurately measure equivalent dose and fully suitable to using as environmental dosimeters. They satisfy requirements on the standards according to [3, 6].

### *2.5. Reproducibility of the response*

The reproducibility is a very important element for measuring low equivalent dose values within interval dose of 0.05–0.3 mSv. The IEC standard [3] defines reproducibility of responses and upper permitted limits by the following relations:

$$S_i = \frac{s_{\overline{Ri}}}{\frac{1}{n}\sum_1^n R_i} \leq 0.075 \quad (4a)$$

$$S_j = \frac{s_{\overline{Rj}}}{\frac{1}{m}\sum_1^m R_j} \leq 0.075 \quad (4b)$$

Where: I = 1 …. n (TLD number); j = 1 …. m (number of irradiation); $s_{\bar{R}}$ is the standard deviation of average values.

These parameters are determined by irradiating n = 20 for each irradiation and m = 5 times at the same values of $D_{ct}$ (0.05 mSv and 0.3 mSv). Experimental values of reproducibility parameters calculated as mean values of the matrices of TLDs and irradiations and are presented in Table 2.

**Table 2:** *Reproducibility data for 20 TLDs irradiated 5 times to $D_{ct}$ = 0.05 mSv and 0.3 mSv*

| $D_{ct}$ (mSv) | $S_i$ | $S_j$ |
|---|---|---|
| 0.05 | 0.056 | 0.064 |
| 0.30 | 0.035 | 0.042 |

From the data in Table 2, it may be concluded that the reproducibility requirement is fully satisfied for the standard according to [3].

***2.6. Limit of detection (Detection threshold)***

The lowest or minimum level of detection (LOD) is defined as three times of the standard deviation from the unexposed signals of annealed TLDs ($s_B$) dividing to response value (R) by the following relation [2]:

$$\text{LOD} = \frac{3.s_B}{R} \quad (6)$$

By selecting 20 annealed TLDs and measuring background TL signals, average value of LOD = 6.1 μSv with uncertainty less than 15% is obtained. So, it means that these TLDs have a quite high sensitivity.

According to recommendations of the IEC 61066:2006 on personal dosimeters (LOD ≤100 μSv) and on environmental ones (LOD ~10 μSv), it may be concluded that these TLDs can be used for both personal and environmental dosimetry [3, 5].

***2.7. Fading***

For this process, 120 annealed TLDs are selected and divided into 6 equal batches. Then, all them are exposured to the similar dose ($D_{ct}$) of 2 mSv and before measuring TL signals of 1$^{st}$ batch after one month, and in turn, 6$^{th}$ batch after six months. The fading data from 1$^{st}$ month to 6$^{th}$ one are 3.3, 4.6, 5.2, 5.9, 6.4 and 6.8%, respectively.

According to recommendations of the IEC 61066:2006 on fading (≤5%/1 month and ≤10%/2 months under standard test conditions such as temperature of 18-22$^o$C, pressure of 86-106 kPa and relative humidity of 50-60%), it may be concluded that fading requirement is fully satisfied.

***2.8. Light sensitivity***

To check the light sensitivity, 80 annealed TLDs are collected and divided into 6 equal batches. By exposuring all them to the similar dose of 2 mSv and storing all them in a room with light of a fluorescent lamp. After that, TL signals of 1st batch are measured after one week, and in turn, 4th batch after four weeks. The data on change of signals from 1st week to 4th one are 2.3, 4.4, 6.7 and 8.5%, respectively.

According to recommendations of the IEC 61066:2006 [3] on effects of light exposure (≤10%/1 week), it could be concluded that requirement on light sensitivity is fully satisfied.

### *2.9. Dose-rate dependency*

In this case, 60 annealed TLDs are selected and divided into 3 equal batches. Al them, then, are exposured to the same dose of 0,5 mSv but 1st batch with dose-rate of 28.8 μSv/h, 2nd one with 125.6 μSv/h and 3rd one with 255.2 μSv/h. By measuring TL signals of the all batches, and consider the 1st batch with relative change as 1 unit. The data on relative change of dose-rate from 1st batch to 3rd one are 1.00, 1.01 and 1.01%, respectively. So, it may be concluded that accumulative dose changes insignificantly with change of the dose-rate range.

### *2.10. Energy dependence of the response*

48 annealed TLDs are selected and divided into 4 equal batches. All them are exposured to the dose of 0.6 mSv with slap phantom at the SSDL of the Institute of Nuclear Science and Technology in Hanoi as follows: 1st, 2nd and 3rd batches for X rays at ISO N40, ISO N60 and ISO N80, respectively, and 4th batch for 662 keV ($^{137}Cs$). By measuring TL signals of the all batches, the data on energy dependence of the response are shown in Figure 4.

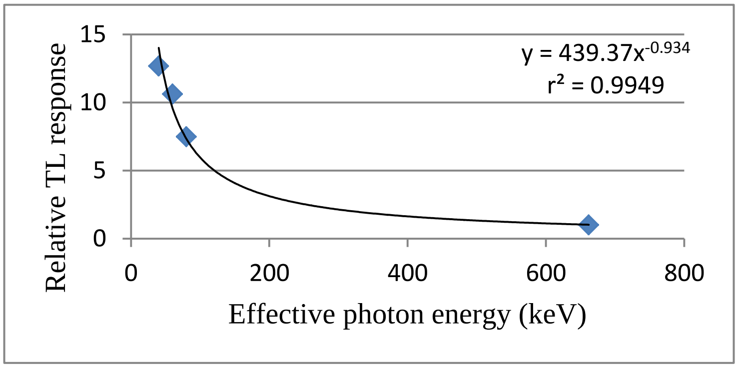


**Figure 4:** *Energy dependence of TLD ($CaSO_4$:Dy)*

It means that response of TLDs depends strongly on low energies in comparison with 662 keV of $^{137}Cs$ (as 1 unit): at 33 keV - about 13 times, at 45 keV - about 11 ones, and at 65 keV - about 7 ones [4, 6].

### *2.11. Dose evaluation with unknown irradiation*

48 annealed TLDs are selected and and divided into 4 equal batches. One half of each batch (6 TLDs) are covered with brass filters of 0.3 mm and 0,5 mm thicknesses. The first 3 batches are irradiated with unknown doses of X

rays at ISO N40, ISO N60 and ISO N80, and 4th batch with unknown dose of 662 keV ($^{137}Cs$) at the SSDL. Then, TL signals of the all TLDs are measured. From the corrected coefficients calculated in the item 10 and ratios between TL signals of the TLDs covered with the filters and the uncovered TLDs, evaluated doses are almostly the same (with uncertainty less than 2.5% for $^{137}Cs$ at 662 keV and about 13% for X ray at 33 keV) in comparison with the dose values provided by the SSDL after that. It proved that personal dosimetry using TLDs with $CaSO_4$:Dy powder has good confidence.

### 3. Conclusion

The TLDs using powder material of $CaSO_4$:Dy studied and successfully produced at DNRI. The results show dosimetric parameters for personal and environmental dosimetry in quantity of Hp(10) are full satisfying comparing with the standards of the IEC 61066:2006 [3]. The main advantage of the TLDs is that their price has been cheaper in comparison with the same TLDs using $CaSO_4$:Dy solid card as well as TLDs using other solid cards (as LiF) imported from foreign countries. Besides, the TLDs produced at DNRI also have some unadvantages as it takes more time for measurement and could cause some errors on weighing and loss of $CaSO_4$:Dy samples. Since 2015, these TLDs have been used for external personal dosimetry for about 140 radiation workers of DNRI and about other 4,300 ones of 450 various radiation installations.

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# NGHIÊN CỨU VÀ KIỂM TRA CÁC THÔNG SỐ ĐỊNH LIỀU CỦA LIỀU KẾ NHIỆT PHÁT QUANG SỬ DỤNG BỘT $CaSO_4$:Dy ĐƯỢC CHẾ TẠO TẠI VIỆN NGHIÊN CỨU HẠT NHÂN ĐỂ ĐỊNH LIỀU BỨC XẠ CÁ NHÂN

***TÓM TẮT***

*Nhóm nghiên cứu ở Viện Nghiên cứu hạt nhân đã tự nghiên cứu và chế tạo thành công liều kế nhiệt phát quang (TLD) sử dụng vật liệu $CaSO_4$:Dy dạng bột. Để định liều cá nhân chiếu ngoài theo đại lượng Hp(10), các thông số định liều của liều kế đã được kiểm tra bằng việc chiếu xạ với những liều gamma khác nhau trên nguồn $^{137}$Cs và đo bằng hệ đọc liều Rexon-320 bao gồm: phổ nhiệt phát quang theo nhiệt độ (hay đường cong phát sáng), hệ số chuẩn (hay giá trị đáp ứng), độ đồng nhất của mẻ TLD, độ lặp lại của phép đo, đáp ứng liều tuyến tính, giới hạn xác định, sự giảm tín hiệu theo thời gian, ảnh hưởng của ánh sáng, sự phụ thuộc vào suất liều, và sự phụ thuộc năng lượng của đáp ứng, v.v… Kết quả nghiên cứu cho thấy liều kế này đảm bảo các đặc trưng về định liều cá nhân theo tiêu chuẩn IEC-61066:2006. Ngoài ra, các liều kế này cũng đã được chiếu chuẩn với nguồn $^{137}$Cs và tia X tại Phòng thí nghiệm chuẩn liều cấp II (SSDL) ở Hà Nội. Kết quả so sánh về liều cho thấy chúng đáng tin cậy. Ưu điểm chính của liều kế loại này là có giá rẻ hơn so với liều kế $CaSO_4$:Dy dạng thẻ cũng như liều kế dạng thẻ dùng vật liệu khác (như LiF). Do đó, chúng đã được dùng để định liều cá nhân chiếu ngoài thường quy cho nhân viên bức xạ ở Viện Nghiên cứu hạt nhân và ở các cơ sở bức xạ khác.*

***Từ khóa:*** *Liều kế nhiệt phát quang, liều sâu (liều toàn thân), đường cong phát sáng, hệ số chuẩn, sự giảm tín hiệu theo thời gian*

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