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Research Article

A STUDY IN COMPUTATIONAL AND EXPERIMENTAL PHOTON ENERGY DEPENDENCE FOR $CaSO_4$:Dy AND Al_2O_3:C MATERIALS

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ABSTRACT

In this work, the energy dependence of thermoluminescence dosimeters (TLDs) and optically stimulated luminescence dosimeters (OSLDs) for photon by the computational and experimental results were studied. Mass energy-absorption coefficients [in μ_{en}/ρ (cm^2/g)] for calcium, sulfur, oxygen, aluminum, carbon, and dysprosium using a Mathematica software were calculated. For materials composed of various elements, it is assumed that the contribution of each element to the total interaction of the photon is additive "mixture rule." *The results obtained from the experiments and the computation were normalized to ^{137}Cs energy response. Within method uncertainty, the calculated energy dependency shows an agreement with experimental results. Both $CaSO_4$:Dy powder (Made in Dalat Nuclear Research Institute) and Al_2O_3:C dosimeters (InLight Basic, Landauer Inc., USA) showed very good uniformity, sensitivity, batch reproducibility, linearity, and low fading for a wide range of doses. Choosing the correct energy for TLDs' calibration is an important factor that can affect the accuracy of the absorbed dose. The results showed that TLDs and OSLDs have a non-uniform response at different energies and both types of dosimeters are quite sensitive in the low photon energy region.*

Keywords: Dosimeter; dosimetry; energy dependence; optically stimulated luminescence dosimeter (OSLD); thermoluminescence dosimeter (TLD)

1. Introduction

1.1. Principles of TL and OSL

Calcium sulfate doped with various lanthanides and aluminum oxide doped with carbon are well-known and extensively studied thermoluminescence (TL)/optically stimulated luminescence (OSL) materials in radiation dosimetry (Harvey, 2010; Knezevic, 2013; Guckan & Volkan, 2017), and many other researchers studied and discussed the

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mechanism of TL/OSL. The basic principles of TL/OSL are described in Fig. 1 in terms of the energy band model of electron-hole production following irradiation. Ionizing radiation creates electron-hole pairs. These electrons and holes become trapped at defects T and H. The trap T_s represents an unstable trap, from where the probability of escaping is large. T_t is a trap for the storage of electrons where the probability of escaping (without external stimulation) is negligible. By stimulating the sample either thermally (TL) or optically (OSL), electrons gain sufficient energy to escape from the trap and recombine with holes in recombination centres (R). The recombination is followed by the emission of light. E_f is the Fermi level.

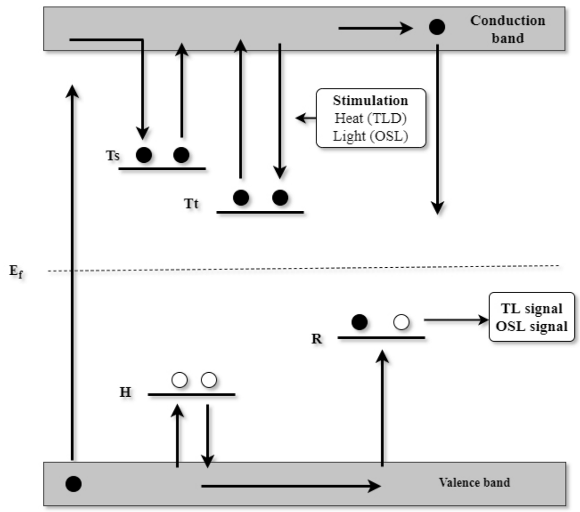

Figure 1. *Basic principles of TL/OSL processes*

1.2. Characteristics of TL for $CaSO_4$:Dy and of OSL for Al_2O_3:C

Calcium sulfate doped with dysprosium ($CaSO_4$:Dy powder) produced at Dalat Nuclear Research Institute (DNRI) has been used for external personal dosimetry in quality *Hp(10)* and environmental monitoring. Landauer Inc. has developed a dosimetry system (called InLight microStar) based on its OSL dosimeter technology, using aluminum oxide doped with carbon (Al_2O_3:C) as an OSL detector material for external personal radiation dosimetry in quality deep dose *Hp(10),* eye-lens dose *Hp(3),* shallow dose *Hp(0.07)* and beta or neutron doses. Each InLight dosimeter contains a slide with four OSL detectors *(E1, E2, E3, E4).* Filters, placed in front of each detector, provide different radiation attenuation conditions. The signal from each OSL detector is used in conjunction with the dose algorithm to evaluate different dosimetric quantities. Due to the wide (9.5 eV) energy band gap, Al_2O_3:C is used popularly as a high sensitive OSL material in personnel dosimetry (Akselrod & Botter-Jensen, 2007). Basic characteristics of the above detectors are presented in Tab. 1 and Tab. 2 (Stanford Landauer Dosimetry, 2008).

Table 1. *Characteristics of detectors used for field measurements*

Detector	Range of photon energy	Form	Dimension	Z_{eff}	Reader
$CaSO_4$:Dy	10 keV-20 MeV	Powder	Grain size: 70÷200 µm Mass of each capsule: 25	15.62	Toledo-654D Vinten
Al_2O_3:C	5 keV-20 MeV	Chips	Grain size < 105 µm Diameter: 5 mm	11.28	Landauer's Inlight microStar

Table 2. *Uniformity, sensitivity, linearity, reproducibility and fading for TL and OSL materials*

Detector	Uniformity	Sensitivity	Linearity	Reproducibility	Fading
$CaSO_4$:Dy	8.2%	10 µSv - 10 Sv	3.5%	5.6%	3.3%/month. 6.8%/6
Al_2O_3:C	Less 0.4% for reads on the strong beam. Less 0.1% for reads on the weak beam.	50 µSv-10 Sv	Less 1% in range of 10 µSv-10 Sv. 5% in range of 1-5 Sv. 10% in range of 5-13	10% in range of 70-140 keV. 5% in range of 5-20 MeV.	3-5%/year.

2. Materials and methods

2.1. Dosimeters

$CaSO_4$:Dy powder (0.15% mass of Dy concentration) is used for calculating energy dependence. $CaSO_4$:Dy powder is mechanically divided into black plastic capsules with an amount of 25 mg (Hung et al., 2019). OSL dosimeter consists of a PVC plastic holder, which snaps shut to hold a plastic dosimeter packet. The dosimeter packet holds the metal/polystyrene plastic filters and a plastic slide containing the detector elements. Each Inlight dosimeter contains a slide with four of such OSL elements, as shown in Fig. 2. When the slide is inside the case, each detector is positioned behind different filters providing different radiation attenuation conditions. The detector element is a layer of Al_2O_3:C sandwiched between two layers of polyester for a total thickness of 0.3 mm. Al_2O_3:C crystals have luminescence emission wavelength centered at 420 nm (blue), optical separation realized be stimulating at 532 nm (green) wavelength with filtration to pass only blue emission, luminescence lifetime 35 msec.

Tab. 3 contains the naming convention used together with the approximate filtration for the four positions of this dosimeter.

Table 3. Design of InLight OSL dosimeter by Landauer Inc.

Name and primary filtration	**Position**	**Density thickness** *(mgxcm^{-2})*		
Absorber (including holder)		Front	Back	Use
Open window (OW)	E1	29	29	Beta response
Plastic filter (PL)*	E2	275	275	Beta characterization, photon response
Aluminum filter (Al)*	E3	375	375	Photon characterization
Copper filter (Cu)*	E4	545	545	Photon characterization

* Add approximately 120 *mgxcm*$^{-2}$ for the outer holder.

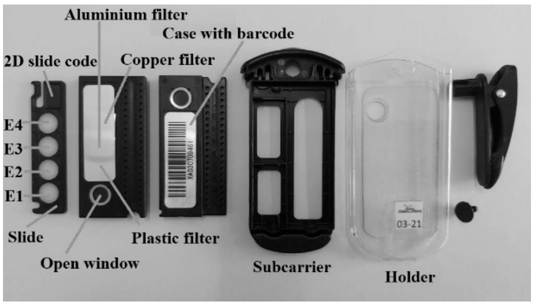

***Figure 2.** Main components of Inlight OSL dosimeters by Landauer Inc.*

2.2. Mathematica software

The mass energy-absorption coefficient and photon energy dependence calculations are carried out by using Mathematica software (ver. 12.1) as an environment for programming.

The function MixtureMAC [mixture, energy] of the package “XRayAttenuation.m” has been used for calculating μ_{en}/ρ coefficients (Schweppe, 2002). A mixture is defined by the form {{material1, fraction1}, {material2, fraction2}, ...}, where each fraction is the fraction by weight of that material in the mixture. The package XRayAttenuation.m needs to be added to the directory .../AddOns/Applications, and then the package must be loaded.

2.3. Irradiation, annealing, and measurement of $CaSO_4$:Dy and Al_2O_3:C dosimeters

Dosimetry with $CaSO_4$:Dy dosimeters requires complex thermal annealing steps (Tab. 4). The annealing procedure was aimed at removing all the previous irradiations and signals to increase the TLD sensitivity. After annealing, these dosimeters were irradiated with ^{137}Cs gamma radiation source at the Tertiary Standard Dosimetry Laboratory (TSDL), DNRI, and X-ray irradiator with various energies at the Secondary Standard Dosimetry Laboratory (SSDL), Hanoi. After irradiation, the TL intensity of these dosimeters was measured (using Toledo-654D system, Vinten, England) after at least 24 hours, to stabilize the fading rate of the TL center in all the dosimeters. According to the Portal, the glow curve presents peaks at 80, 120, 220, and 250 ^{o}C (Souza et al., 1993). To select the best readout parameters, the time-temperature profile (TTP) was chosen as in Tab. 4 and plotted in Fig. 3.

Table 4. *Evaluated parameters for TLDs used in measurements*

Temperature		Time	
Preirradiation annealing	700ºC + 400ºC	120 min + 20 min	
Preheat	160ºC	Preheat	10 sec
Maximum	280ºC	Acquire	27 sec
Anneal	no	Anneal	0 sec
		Rate	8 ºC/sec

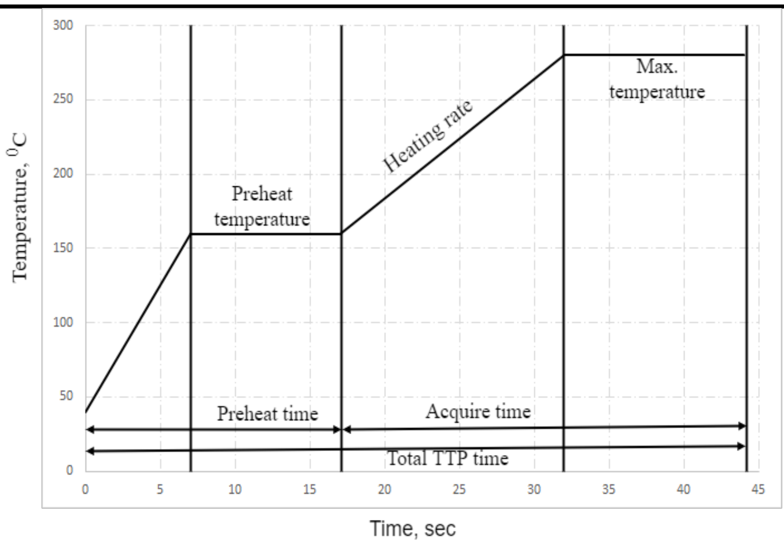

Figure 3. *Time-Temperature Profile*

For the energy dependence experiments of TLDs, the dosimeters were irradiated to 3 mSv with X-ray irradiator at the SSDL, Hanoi and the following ISO reference radiations were used as ISO N40, N60, and N80 fields (ISO 4037-1, 1996). Irradiation with ^{137}Cs in gamma dose at 3 mSv was performed at the TSDL, DNRI.

For OSLDs, irradiations using a ^{137}Cs and X-ray irradiator (at ISO N60, N80, and N120 fields (ISO 4037-1, 1996)) were performed in SSDL, Office of Atomic Energy for Peace, Thailand for following dose values: 0.4 mSv at a low dose for all energies. At a high dose, OSLs were exposed to: 2 mSv at N60 and N120, 4 mSv at N80, 3 mSv at ^{137}Cs. The readout was performed with InLight Basic reader (Landauer Inc., USA)

Both TLDs and OSLs are irradiated with PMMA phantom (30x30x15 cm^3).

2.4. Method

This work proposes determination of the energy dependence of $CaSO_4$:Dy and Al_2O_3:C at low photon energy and the mass energy-absorption coefficient for calcium, sulfur, oxygen, aluminum, and phosphor materials from their compounds. The results are compared with the experimental values.

The mass energy-absorption coefficient (μ_{en}/ρ) for the chemical composition one may assume that the contribution of each element to the total interaction of the photon is additive "mixture rule". In accordance with this rule, the total mass energy-absorption coefficient of a compound is the sum of the weight proportion of each atom present in it (Morabad & Kerur, 2010).

$$\frac{\mu_{en}}{\rho} = \sum_i w_i \left(\frac{\mu_{en}}{\rho}\right)_i \qquad (1)$$

where w_i is the fraction by weight of the *i-th* atomic constituent, ρ is the density of the material, and the $(\mu_{en}/\rho)_i$ is the mass energy-absorption coefficient of the *i-th* atomic constituent. The atomic number and atomic mass of elements were taken from the atomic weight of elements 2011, IUPAC report by (Michael & Holden, 2013).

The energy dependence of the phosphor materials from 0.001 to 20 MeV is calculated using the formula given by F. H. Attix as follows (Attix, 1986):

$$R_{TLD/OSL} = \frac{0.869 \times (\mu_{en}/\rho)_{TLD/OSL}}{(\mu_{en}/\rho)_{air}} \; rad/R \qquad (2)$$

where $R_{TLD/OSL}$ is the energy absorbed in the material of the TLDs or OSLDs per unit exposure, $(\mu_{en}/\rho)_{TLD/OSL}$ is the mass energy-absorption coefficient for the TLDs or OSLDs obtained by adding the weighted average of the μ_{en}/ρ values of the various component elements of TL or OSL materials and $(\mu_{en}/\rho)_{air}$ is mass energy-absorption coefficient values for dry air (Up to now, there are no measured data available, but numerous calculated values exist in different tabulations).

3. Results and discussion

3.1. Mass energy-absorption coefficient

Fig. 4 shows the mass energy-absorption coefficient of various TL/OSL elements for the energy range from 0.001 to 20 MeV in each step of 0.001 MeV. μ_{en}/ρ coefficient decreases when energy increases from 0.001 to 0.03 MeV and then falls to a constant value.

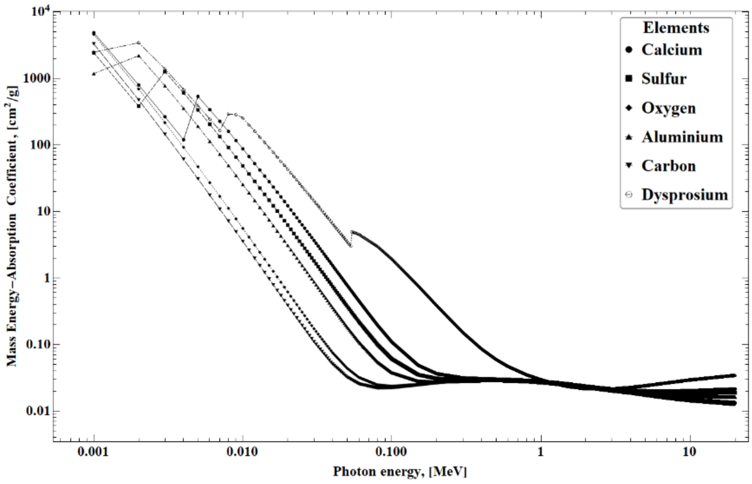

Figure 4. *Plots of mass energy-absorption coefficient of various elements for photon energy from 0.001 to 20 MeV*

The energy dependence of phosphor materials for the photon is plotted in Fig. 5. It is seen that the mass energy-absorption coefficients depend on the photon energy and chemical content.

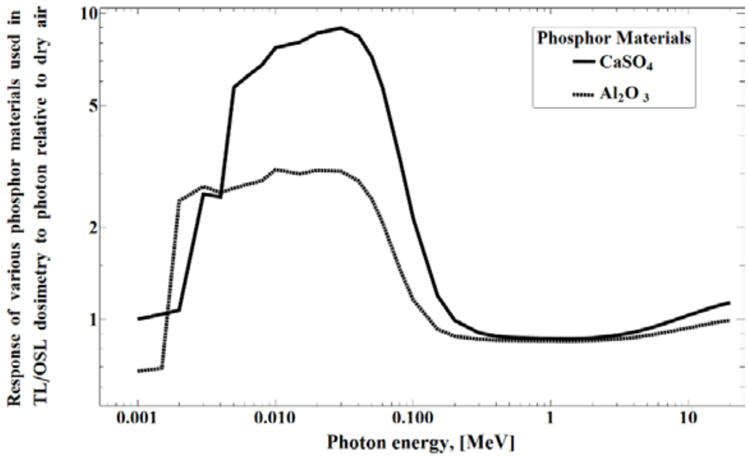

Figure 5. *Calculated energy dependence for photon energy from 0.001 to 20 MeV, and from these data, the energy dependence values of TL response is taken and plotted in Fig. 6 and Fig. 7*

Fig. 5 shows that the response of the phosphor materials is maximum between 10 keV and 30 keV and then gradually becomes constant from 300 keV to 10 MeV. As photon energy increases from 30 keV to 300 keV the relative TL/OSL response for photon energy in the materials decreases. The maximum responses of the phosphor materials are shown in Tab. 5.

Table 5. *The results calculated the response of the phosphor material*

TL materials	Maximum response, rad/R	Energy, MeV
Al_2O_3	3.089	0.010
$CaSO_4$	8.973	0.030

The energy dependence curves show that both types of dosimeters are quite sensitive in the low photon energy region. The slight rise of energy dependence after 2 MeV is due to the slight increase in the stopping-power values above that energy.

3.2. Experimental photon energy dependence of $CaSO_4$:Dy and Al_2O_3:C

Fig. 6 shows the energy dependence of $CaSO_4$:Dy to X-rays (N40, N60, N80) and ^{137}Cs radiation fields through the Mathematica software and experiments. The energy dependence results were normalized to ^{137}Cs response as a unit. The experimental TL response is taken three measurements of each energy to calculate standard deviations.

The experimental energy dependence with a higher value than the theoretical one for all irradiation energies. TL response increases 24.41% at N40, 0.00% at N60 and 26.28% at N80. As expected, the TL response shows a decreasing trend with the increase of energy in both experiment and computation results. The calculated energy dependencies show good agreement with the experimental results within 20% of uncertainty. These differences are due to the non-homogeneous distribution of Dy concentration in the powder and the irradiation geometry. Another factor is the real grain size in phosphor, thus the grain size is not considered in the calculation.

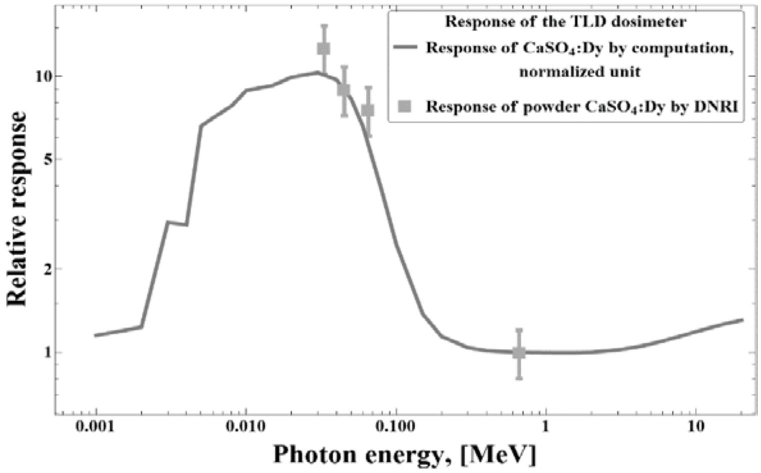

Figure 6. *Comparison between experimental and computational energy dependence of $CaSO_4$:Dy to photon energies at N40, N60, N80 and* ^{137}*Cs*

Fig. 7 shows that the experimental energy dependence of low dose is higher than theoretical values (with a relative error of 7.92% at N60, 1.79% at N80, and 9.31% at N120). At a high dose, OSL takes a lower response than theoretical and varies with relative error -14.05% for N60, -21.27% for N80 and -18.50% for N120. Three dosimeters were irradiated for each energy. The energy dependence results were normalized to ^{137}Cs response as a unit. Measurements were repeated 5 times for each dosimeter and the average percentage error was found to be nearly 16% of the mean delivered dose values.

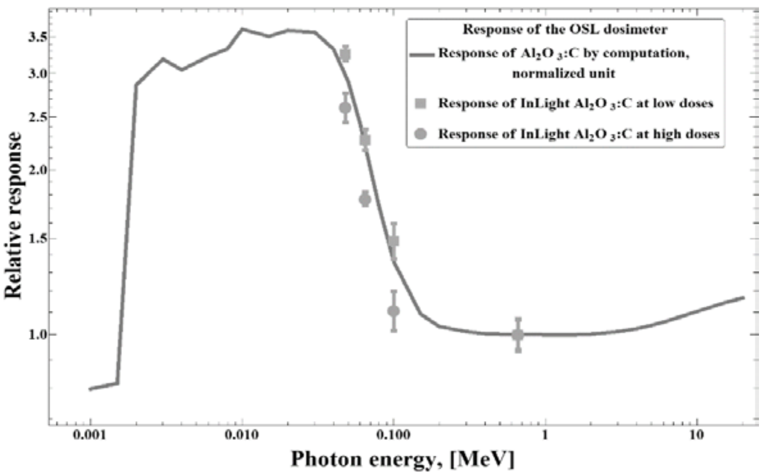

Figure 7. *Experiment energy dependence of Al_2O_3:C to photon energies at N60, N80, N120, and* ^{137}*Cs, compared with computational values*

The response of the four positions *(E1, E2, E3, E4)* to photon radiation from 16 to 1250 keV is shown in Fig. 8. The data for these plots were taken from (Stanford Landauer Dosimetry, 2008).

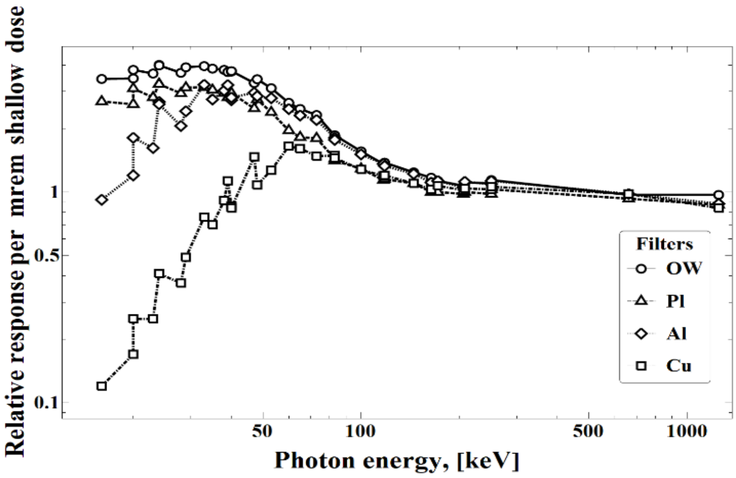

Figure 8. *Relative element response for photon fields of InLight Landauer*

The relative responses of the elements are used to characterize the fields. In this case, the ratio of E3 to E4, which is filtered with aluminum and copper, is used to characterize the photon fields. For three low energy fields as NS25 (16 keV), NS30 (20 keV), and H30 (20 keV), the ratio of E3 to E4 is equal to 7, and then increasing the energies will decrease the ratio. This ratio is a strong indicator for photon energies up to several hundred keV. For higher photon energies, this ratio becomes constant and is equal to 1.

In Fig. 9, the experimental energy response of Al_2O_3:C is plotted with four energies (ISO N60, N80, N120, and ^{137}Cs). The experimental results were satisfactory and were in agreement with the data from Stanford Landauer Dosimetry (2008) with a standard deviation of ratio E3/E4 is less than 9%.

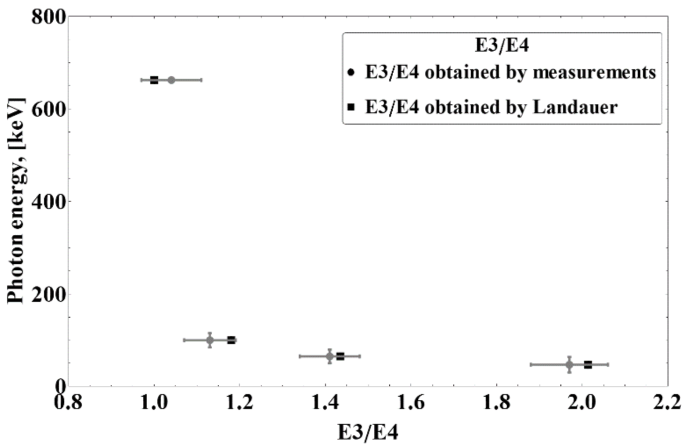

Figure 9. *Photon energy as a function of ratio E3/ E4 obtained by Landauer Inc. and experiment*

4. Conclusion

The energy dependence of TLDs developed at DNRI and OSLDs of Landauer Inc. was evaluated by using the Mathematica software and experiments. Experiments can be compared with the computational data, the agreement is very good with uncertainties of 20% for TLDs and 25% for OSLDs. However, the energy dependence of TLDs is different from the theoretical one because of the effect of the grain size in TL powder and the non-homogeneous distribution of Dy concentration in the powder. The energy dependence curves show that TLDs and OSLDs are quite sensitive in the low-energy region.

The evaluation of E3/E4 ratio helps to determine the energy characteristics of the photon fields, thereby helping to determine the radiation dose more accurately. The measurement results show high accuracy compared to the declared data of Landauer Inc. This work can develop algorithms for external photon dosimetry in medical imaging, nuclear medicine dosimetry, and dosimetry in high-energy radiation fields.

A further study is needed for the effects of grain size and composition ratio of Teflon, which is useful in the development of new $CaSO_4$:Dy Teflon disk dosimeters for external personal monitoring.

❖ ***Conflict of Interest:*** *Authors have no conflict of interest to declare.*

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NGHIÊN CỨU SỰ PHỤ THUỘC NĂNG LƯỢNG PHOTON BẰNG THỰC NGHIỆM VÀ TÍNH TOÁN ĐỐI VỚI VẬT LIỆU $CaSO_4$:Dy VÀ Al_2O_3:C

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TÓM TẮT

Trong công trình này, sự phụ thuộc năng lượng của liều kế nhiệt phát quang (TLD) và liều kế quang phát quang (OSLD) đối với photon *bằng kết quả đo thực nghiệm và tính toán đã được nghiên cứu. Hệ số hấp thụ năng lượng khối [đơn vị μ_{en}/ρ (cm^2/g)] đối với canxi, lưu huỳnh, oxy,* nhôm, cacbon *và* dysprosi *đã được tính toán bằng phần mềm Mathematica. Đối với vật liệu gồm các nguyên tố khác nhau, có thể giả sử rằng đóng góp của mỗi nguyên tố vào tương tác tổng cộng của photon là* "quy tắc trộn thêm". *Kết quả thu được từ phép đo thực nghiệm và tính toán được chuẩn hóa với đáp ứng năng lượng của ^{137}Cs. Trong phạm vi độ không bảo đảm đo của phương pháp, sự phụ thuộc năng lượng được tính toán đã chỉ ra sự phù hợp tốt so với kết quả đo thực nghiệm. Cả hai loại liều kế bột $CaSO_4$:Dy (chế tạo ở Viện Nghiên cứu hạt nhân) và liều kế Al_2O_3:C (loại InLight Basic, hãng Landauer, USA) đã chỉ ra sự đồng nhất, độ nhạy, độ lặp lại theo mẻ chế tạo, độ tuyến tính là tốt và sự giảm tính hiệu theo thời gian là nhỏ ở dải liều rộng. Việc chọn năng lượng đúng để hiệu chuẩn TLD là yếu tố quan trọng mà có thể ảnh hưởng đến độ chính xác của liều hấp thụ. Kết quả đã chỉ ra rằng các TLD và OSLD có đáp ứng không đồng nhất ở những năng lượng khác nhau và cả hai loại liều kế là khá nhạy ở vùng năng lượng photon thấp.*

Từ khóa: liều kế; định liều; phụ thuộc năng lượng; liều kế nhiệt phát quang (TLD); liều kế quang phát quang (OSLD)