

Application of cavity theory to $\text{CaSO}_4:\text{Dy}$ thermoluminescent dosimetry

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Abstract. We discuss the influence of particle size on thermoluminescent (TL) response of $\text{CaSO}_4:\text{Dy}$. The cavity theory was applied in order to calculate the variations in the TL response of this (phosphor powder) bound in KBr. The sizes of the particles considered ranged from 50 to 300 μm in diameter, and the irradiations were effectuated with ^{60}Co Gamma rays. Calculations were made using the well known Burlin's cavity theory and the new one proposed by Kearsley. We make a comparison between experimental results obtained for different particle size intervals, irradiating with ^{60}Co gamma rays, and the calculations using to the two theories. Experimental results show that Kearsley theory fits better the observed data.

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1. Introduction

Calcium sulfate doped with dysprosium is a thermoluminescent material which is being increasingly used in radiation dosimetry, particularly for low dose measurements, because of the great number of favorable properties it has. In addition to its small size, low cost and easy reading, it has a good reproducibility and a high degree of both, sensitivity and stability.

Generally calcium sulfate is used in powdered form, embedded in plastics or resinous substances, or bound in alkali halides.

Since particle size is an important factor affecting the response of TL material, the particle size of calcium sulfate, as a TL dosimeter, has to be kept in mind in order to determine its response. Those variations found in the response of TL materials of the same compounds can be attributed to differences in the absorbed dose in powder grains, as particle size is varied.

This study analyses the dependence of the $\text{CaSO}_4:\text{Dy}$ TL response as a function of particle size of the phosphor powder used to make pellets of $\text{CaSO}_4:\text{Dy}$ bound in KBr. The TL response was determined by means of the ratio of the absorbed dose in a grain of $\text{CaSO}_4:\text{Dy}$ to the absorbed dose in KBr (f_z^c) calculated using the Burlin [1] cavity theory and the recent one proposed by Kearsley [2, 3]. Experimental determinations of these variations were carried out by irradiation locally made pellets of $\text{CaSO}_4:\text{Dy}$ bound in pure KBr.

2. Theory

In order to explain TL response-particle size variation by the cavity theory we consider a grain of TL powder situated in a medium of different atomic number and density as a "cavity", and assume that the thickness of the medium surrounding the grain is sufficient to establish electronic equilibrium.

The energy deposited in the grain depends on the energy balance among the sum of the energies of the electrons leaving the grain, the sum of those energies of the electrons generated within the grain, and the sum of those energies of the electrons entering the grain from the surrounding medium.

The total energy of the electrons generated within a grain depends on the mass energy absorption coefficient of the grain material, while the total energy of the electrons entering a grain is dependently related to the mass energy absorption coefficient of the surrounding medium. The amount of energy removed from these electrons and absorbed by both, the grain and the surrounding medium, depends on the individual mass stopping powers of the two materials.

The absorbed dose in a grain of TL material (D_c) surrounded by a medium of mass energy absorption coefficient ($\mu en/\rho$) is proportional to:

$$D_c \propto (\mu en/\rho)_c f_z^c,$$

where f_z^c is the ratio between the absorbed dose in the cavity to the absorbed dose in the surrounding medium.

When the size of the TL material is very large the absorbed dose is proportional to:

$$D_{c(\rightarrow\infty)} \propto (\mu en/\rho)_c.$$

This expression represents the dose a grain of any size would absorb if it were surrounded by a medium of identical composition (perfectly matched combination). The TL response of different grain sizes relative to that of a grain of a very large size, becomes:

$$TL = \frac{(\mu en/\rho)_z}{(\mu en/\rho)_c} f_z^c. \quad (1)$$

In accordance with Burlin's cavity theory, the basic equation for evaluating stopping powers ratio is [1]:

$$f_z^c = \frac{(Z/A)_c}{(Z/A)_z} \left\{ 1 + \frac{d}{T_0} \left[\int_{\Delta}^{T_0} R_z(T_0, T) \left(\frac{B_c(T)}{B_z(T)} - 1 \right) dT + \Delta R_z(T_0, \Delta) \right. \right. \\ \left. \left. \times \left(\frac{B_c(\Delta)}{B_z(\Delta)} - 1 \right) \right] + (1-d) \left(\frac{(\mu en/\rho)_c (Z/A)_z}{(\mu en/\rho)_z Z/A_c} - 1 \right) \right\}, \quad (2)$$

where T_0 is the initial energy of the electrons, Z , the atomic number, A the atomic weight, and $R(T_0, T)$ the ratio of the total electron flux to the flux of primary electrons at an energy T when the initial electrons energy is T_0 , $B(T)$ is the stopping number of the electrons of energy T , Δ is the average energy of an electron which will just, on the average, cross the cavity, d is a weighting factor, and $\mu en/\rho$ is the mass energy absorption coefficient; the subindexes c and z are referent to the cavity and to the medium respectively.

The weighting factor, d , determines the contribution to the dose within the grain due to the electrons generated in the surrounding medium. This weighting factor represents the average attenuation of the electron spectrum which passes through the cavity, and is expressed as:

$$d = \frac{\int_0^g e^{-\beta x} dx}{\int_0^g dx} \tag{3}$$

$$1 - d = \frac{\int_0^g (1 - e^{-\beta x}) dx}{\int_0^g dx}, \tag{4}$$

where g is the average path length of electrons crossing the cavity, expressed in $g \cdot \text{cm}^{-2}$, and β is the effective mass absorption coefficient for the electrons.

Equation (2) can be expressed in a more simple form as:

$$f_z^c = \frac{1}{g} \int_0^g e^{-\beta x} dx \frac{\langle S/\rho \rangle_c}{\langle S/\rho \rangle_z} + \frac{1}{g} \int_0^g (1 - e^{-\beta x}) dx \frac{(\mu en/\rho)_c}{(\mu en/\rho)_z}. \tag{5}$$

The ratio of the average dose in the cavity to the absorbed dose in the medium according with Kearsley's theory [2, 3] is given by:

$$f = \frac{\frac{1}{g} \int_0^g \zeta(x) dx \cdot \langle s/\rho \rangle_c}{\psi(\mu en/\rho)_z},$$

where $\zeta(x)$ is the electron fluence at a point x in a cavity surrounded by wall material expressed in cm^{-2} , and given by the following expression:

$$\begin{aligned} \zeta(x) = & \frac{\psi(\mu en/\rho)_c}{\langle s/\rho \rangle_c} \left[1 + (1 - e^{-\gamma g}) \frac{F_c}{U_c} (b_z \lambda_2(z, c) - b_c \lambda_2(c, c)) \right] \\ & - \frac{F_c}{U_c} \lambda_1(c, c) + (1 - e^{-\gamma g}) \frac{B_c}{U_c} (b_z \lambda_1(z, c) - b_c \lambda_1(c, c)) \\ & - \frac{B_c}{U_c} \lambda_2(c, c) \frac{\psi(\mu en/\rho)_z}{\langle s/\rho \rangle_z} \left| \frac{F_z}{U_z} \lambda_1(z, c) + \frac{B_z}{U_z} \lambda_2(z, c) \right|. \end{aligned}$$

In the above expression, $\psi(\mu en/\rho)/\langle s/\rho \rangle$ represents the secondary electron fluence throughout the cavity or the surrounding medium, where: ψ is the photon energy fluence expressed in $\text{MeV} \cdot \text{cm}^{-2}$; γ is the mass absorption coefficient of the secondary electron, in $\text{cm}^2 \cdot \text{g}^{-1}$; F is the component of the secondary electron fluence produced upstream of a reference plane at the interface, in cm^{-2} ; B is the component of the secondary electron fluence produced downstream of the reference plane at the interface, cm^{-2} ; U is the equilibrium fluence in an homogeneous material throughout cavity or the surrounding medium, in cm^{-2} ; b is the backscatter factor for a semi-infinite thickness of cavity or wall material (dimensionless); and the coefficients λ_1 and λ_2 are given by

$$\lambda_1(z, c) = \frac{b_z e^{-2\beta g} e^{\beta x} + (1 - b_c b_z) e^{-\beta x}}{(1 - b_c b_z)^2 - b_z^2 e^{-2\beta g}},$$

$$\lambda_2(z, c) = \frac{b_z e^{-\beta g} e^{-\beta x} + (1 - b_c b_z) e^{-\beta g} e^{\beta x}}{(1 - b_z b_c)^2 - b_z^2 e^{-2\beta g}},$$

g , β , $\langle s/\rho \rangle$ and $(\mu en/\rho)$ have their usual meaning.

In the above equations the subindexes c and z refer to cavity material and to the surrounding medium respectively.

The mean difference between the two theories is that Kearsley's theory takes into account the effect of secondary electron scattering at cavity boundaries and can be used to calculate the average cavity dose. The dose distribution inside the cavity, as well as the relative contributions of the wall and the cavity to the cavity response. Meanwhile, the Burlin's model defines the factor d as the average reduction in the wall spectrum, which evidently means the average reduction in the fluence of secondary electrons emitted into the cavity from the wall, assuming a constant differential energy spectrum throughout the cavity. The average build up of the cavity secondary spectrum turns out to be $(1 - d)$, as long as the argument of the build up exponential is identical to the argument of the attenuation exponential. The attenuation and production of the secondaries are independent processes which seems like an unnecessary restriction.

The most important perturbation to energy deposition in the cavity, ignored by Burlin's model, is the influence of scattering at the boundaries. This effect almost completely determines the amount and direction of energy entering and leaving the cavity.

3. Calculations

The TL response of the $\text{CaSO}_4:\text{Dy}$ bound in KBr pellets made with different grain powder sizes as a function of the particle size was calculated by computer using equation (1), calculating f_z^c by means of equation 5 and 6.

The particle sizes considered were in the range from 50 to 300 μm in diameter, since TL dosimeters having grain sizes in this range are widely used.

The g and β values were calculated as follows:

$g = \rho x$, where ρ is the phosphor density in $\text{g} \cdot \text{cm}^{-3}$ and was obtained experimentally for each particle size considered, and x is the grain diameter in cm, $\beta = \gamma = 14/(E_{\text{max}})^{1.09}$, as suggested by Paliwal and Almond [7] where E_{max} is the maximum energy of the electron.

The other parameters were taken from the literature as indicated below:

$(\mu\text{en}/\rho)$ values from Hubell [4], 1982,

$\langle s/\rho \rangle$ values from Seltzer [5], 1982,

F/U , B/U and b values from Dutreix and Bernard [6], 1966.

Once calculations were completed, the TL response (which was determined using f_z^c values calculated by means of the two theories) was plotted as a function of particle size.

4. Experiment

In order to determinate TL response of CaSO₄:Dy bound in KBr experimentally, pellets of CaSO₄:Dy + KBr were made by pressing a mixture (3:1) of CaSO₄:Dy powder of different grain sizes with pure KBr. This procedure was reported before [7].

Irradiations were done with a ⁶⁰Co encapsulated source (16.5 GBq) in electronic equilibrium conditions.

The particle size intervals used were 50–75; 75–100, 100–120, 120–150, 150–175, 175–190, 190–210, 210–250, 250–280, 280–300 μm and were selected by sieving.

Adsorbed doses in CaSO₄:Dy and in KBr were determined by measuring the exposure with an ionization chamber.

TL readings were made in a Harshaw 2000 A/B TL analyser at a heating rate of $6.7 \text{ K} \cdot \text{s}^{-1}$, integrating from room temperature ($\sim 293 \text{ K}$) to 523 K over 60 seconds.

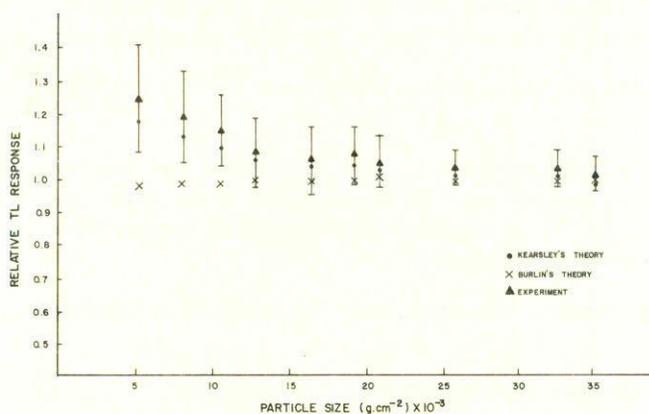


FIGURE 1. Relative TL response of $\text{CaSO}_4:\text{Dy} + \text{KBr}$ pellets as a function of particle size of the phosphor powder.

Particle size (g/cm^{-2}) $\times 10^{-3}$	Experimental TL response (relative value)	Theoretical TL response		Percentual deviations from experimental values	
		Kearsley	Burlin	Kearsley	Burlin
5.26	1.24 ± 0.15	1.18	0.97	4.3	21.5
8.15	1.20 ± 0.13	1.12	0.97	6.7	18.7
10.34	1.15 ± 0.11	1.09	0.97	4.9	14.9
12.84	1.08 ± 0.11	1.06	0.97	1.3	9.2
15.95	1.07 ± 0.10	1.05	0.97	1.4	8.3
18.64	1.09 ± 0.08	1.04	0.98	3.9	9.8
20.40	1.04 ± 0.08	1.02	0.98	2.2	6.0
25.05	1.02 ± 0.05	0.99	0.98	2.5	3.2
33.62	1.01 ± 0.05	0.99	0.98	2.5	2.9
36.79	1.00 ± 0.05	0.97	0.99	3.0	1.3

TABLE I. Comparison between experimental and theoretical TL response of $\text{CaSO}_4:\text{Dy}$ bound in KBr as a function of particle size of the phosphor powder.

5. Results and conclusions

Theoretical and experimental values of TL response as a function of particle size obtained for $\text{CaSO}_4:\text{Dy}$ bound in KBr are plotted in figure 1.

A comparison between experimental results and the calculations by means of the two theories were made in Table I.

The deviations obtained show better agreement of the experiment with Kearsley's theory than with Burlin's for particle sizes ranging from 5 to 12 mg/cm^{-2} . Figure 1 suggests that for ^{60}Co the spectrum at the front side of the cavity, using

Kearsley's theory, appears to be harder than at the back side. Meanwhile, the apparent exponential attenuation of secondary electrons in the Burlin's theory is probably a geometric effect and has little to do with the shape of a secondary electron equilibrium spectrum.

These results support those obtained in previous work [8] for CaSO₄:Dy phosphor powder in air.

Reference

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Resumen. Presentamos la influencia que el tamaño de las partículas tiene sobre la respuesta termoluminiscente del CaSO₄:Dy. Se utilizó la teoría de cavidad para calcular las variaciones en la respuesta TL del polvo de material TL en KBr. Los tamaños de las partículas consideradas van de 50 a 300 μm de diámetro, y las irradiaciones fueron efectuadas con rayos gama de ⁶⁰Co. Los cálculos fueron hechos utilizando la teoría de cavidad de Burlin y la nueva teoría propuesta de Kearsley. Hacemos la comparación entre los resultados experimentales obtenidos y los cálculos hechos con las dos teorías. Los resultados experimentales muestran que la teoría de Kearsley se ajusta mejor a los datos observados.