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LET dependence of thermoluminescent efficiency and peak height ratio of CaF_2:Tm

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Abstract

First-order thermoluminescence (TL) kinetics computerized glow curve deconvolution (CGCD) and manual analysis of the composite peak structures have been applied to study the behaviour of glow peaks 3 and 5 in CaF_2:Tm (TLD-300, Harshaw-Thermo Fisher Scientific) single crystals after heavy charged particle (HCP) irradiation with respect to ^{60}Co gamma rays for a linear energy transfer (LET) interval from 2.3 to 339.4 keV µm^{-1}. The ratio of peak 5 to peak 3 heights can be used to evaluate effective LET and correct measured doses for TL efficiency. The applicability of this idea to dose equivalent estimation in exotic radiation fields of complex composition is discussed critically.

Keywords: Thermoluminescence; Efficiency; Calcium fluoride

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1. Introduction

The gradually increasing utilization of thermoluminescence (TL) dosimeters in exotic radiation environments, such as in space or around high-energy particle accelerators in medical and industrial installations, implies that greater efforts must be invested in the measurement of TL efficiency for heavy charged particle (HCP) irradiation. Determination of the intrinsic efficiency, i.e. the ratio of the energy emitted as TL light to the energy absorbed during exposure to ionizing radiation, is highly complex since various physical processes are involved (Bos, 2007). Relative TL efficiency, $\eta_{\mathrm{HCP},\gamma}$, with respect to a specified reference radiation—usually ^{60}Co gamma rays—is much easier accessible to the experiment. It is given by

$$\eta_{\mathrm{HCP},\gamma} = \frac{R_{\mathrm{HCP}}/D_{\mathrm{HCP}}}{R_\gamma/D_\gamma}, \tag{1}$$

where R_{HCP} and R_γ are the TL signal intensity created in the dosimeter per unit absorbed dose, D, following HCP and ^{60}Co gamma irradiation, corrected for self-absorption of the TL photons in the detector crystal itself (Horowitz and Stern, 1990). This paper investigates the behaviour of specific glow peaks in thulium-activated calcium fluoride (CaF_2:Tm) with respect to ^{60}Co gamma rays applying computerized glow curve deconvolution (CGCD) and manual analysis of the composite peak structures.

CaF_2 is one of the best studied crystals in solid-state physics. Due to its high effective charge $Z_{\mathrm{eff}} = 16.3$, the phosphor cannot be considered as tissue-equivalent for gamma and X-ray photons and is, probably for this reason, comparatively seldom applied as a TL detector in radiation dosimetry. Single-crystal CaF_2:Tm has been commercialized by Harshaw Chemical Co. (now Thermo Fisher Scientific, Inc.), Cleveland, OH, USA, under the name TLD-300 and benefits from its generally high sensitivity compared, for instance, to widespread LiF:Mg, Ti (TLD-100). At a first glance, the CaF_2:Tm glow curve exhibits two well-resolved peaks (Azorín et al., 1989). An example is given in Fig. 1 for 118.4 mGy of ^{60}Co gamma irradiation at room temperature and readout at a heating rate $\beta = 5\,^{\circ}\mathrm{C\,s}^{-1}$. An in-depth analysis of the experimental curve applying Randall–Wilkins first-order kinetics peak shaping by means of the GlowFit software (Puchalska and Bilski, 2006) reveals the prominent peak 3 at ~ 164 °C and a composite

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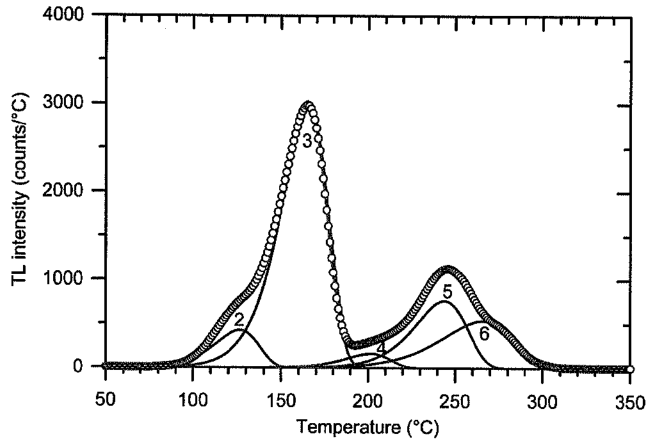

Fig. 1. GlowFit deconvolution using first-order kinetics peak shapes of a glow curve from CaF_2:Tm (TLD-300) following 118.4 mGy of ^{60}Co gamma irradiation. In the fitting procedure, constraints were set for the temperatures, T_m, corresponding to the maxima of peaks 2, 3 and 6. The experimental glow curve (open circles) was recorded at a heating rate $\beta = 5\,°C\,s^{-1}$ using two neutral optical filters (NG3, Schott AG, Mainz, Germany).

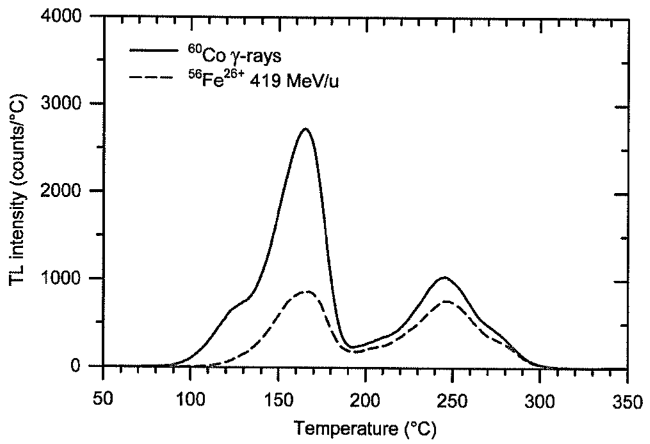

Fig. 2. Sample glow curves from CaF_2:Tm (TLD-300) following 100 mGy of irradiation by ^{60}Co gamma rays (solid line) and $^{56}Fe^{26+}$ ions of 419 MeV u^{-1} (dashed line), respectively. The glow curves were recorded at a heating rate of $\beta = 5\,°C\,s^{-1}$ using two neutral optical filters (NG3, Schott AG, Mainz, Germany).

high-temperature structure (HTS) peaking at ~ 246 °C. The HTS involves primarily peak 5 at ~ 243 °C and peak 6 at ~ 265 °C, with a minor contribution from peak 4. For long-term applications in environmental or space dosimetry, analysis of peak 3 seems inappropriate due to its fading characteristics and the presence of a significant, non-radiation induced TL signal which lies directly under peak 3 (Shachar and Horowitz, 1988). Peaks 5/6, on the other hand, can be used to measure even very low doses down to the µGy level with negligible fading.

The HTS is of considerable interest because of the peculiarities of its ionization density dependence which is essentially different from the behaviour of the main dosimetry peak 3 (Fig. 2). Considering the non-uniform pattern of dose deposition at the microscopic level, the enhanced sensitivity of peak 5 to HCP irradiation can be explained by its linear–supralinear dose response (Olko, 1998, 2007). Several authors have proposed to exploit this increased sensitivity to high linear energy transfer (LET) radiation for neutron–gamma ray discrimination (Loncol et al., 1996) and HCP dosimetry (Hoffmann and Prediger, 1983; Loncol et al., 1996).

2. Materials and methods

2.1. Dosimeter preparation, readout and analysis

Before each exposure, the CaF_2:Tm (TLD-300, Harshaw-Thermo Fisher Scientific) single crystals of size 3.2 × 3.2 × 0.89 mm^3 obtained from the same batch (15RA-566) were annealed according to a well-defined protocol at 400 °C for 1.5 h in air, followed by slow cooling to room temperature for ~ 24 h in the oven. Glow curves were readout by contact heating on a Nikrothal 80 austenitic alloy planchet from room temperature to a maximum temperature of 400 °C at a linear heating rate $\beta = 5\,°C\,s^{-1}$. To minimize spurious chemiluminescence and triboluminescence, the measurement chamber was first evacuated and during readout flooded with ultra-pure (5.0) dry N_2 gas. The in-house developed reader employed the photon counting technique using a Thorn EMI 9635 QB photomultiplier (Thorn EMI Gencom, Inc., Fairfield, NJ, USA) with a bialkali photocathode (Vana et al., 1988). In order to attenuate the light incident on the photomultiplier tube, two neutral optical filters (NG3, Schott AG, Mainz, Germany) were used for doses above ~ 80 mGy. Background subtraction was achieved by an exponential fit with constant offset. This method proved to be superior to manual analysis in which the background would be estimated by a consecutive second readout. The TL glow curves were analysed by means of a CGCD technique developed at the Institute of Nuclear Physics, Krakow, Poland, and based on first-order TL kinetics (Puchalska and Bilski, 2006). A strict deconvolution protocol which set constraints to the maximum temperatures, T_m, of peaks 2, 3 and 6 was followed to ensure consistent description of the glow peaks.

2.2. Irradiation conditions

HCP irradiations were performed at the Heavy Ion Medical Accelerator (HIMAC) of the National Institute of Radiological Sciences (NIRS), Chiba, Japan. Fully ionized nuclei were supplied by three stable, long-lived ion sources of the electron cyclotron resonance (ECR) and the Penning ionization gauge (PIG) type. Before the ionized particles were injected into the synchrotron, they were accelerated in two linear accelerator (LINAC) stages consisting of a radio-frequency quadrupole (RFQ) and an Alvarez LINAC. The heavy ion beams were then transported to a pair of synchrotron rings and further accelerated to a maximum energy of 800 MeV u^{-1}. All irradiations were carried out in the biological irradiation room where a maximum beam diameter of 10 cm can be obtained by using a pair of wobbler magnets and a scatterer. The particle fluence was monitored by a scintillation counter. Reference doses were retrieved from a high-precision Farmer-type ionization chamber of 1 mm water-equivalent thickness installed upstream of the target which was also used to check uniformity of the beam in one dimension, yielding maximum fluctuations of $< 5\%$ over the circular beam area for all ion species (Kanai et al., 1999). The TL crystals were sealed in polystyrene holders of 1 mm thickness and exposed to doses of the order of 100 mGy from the following ion beams (the energy corresponds to the particle energy on exit from the synchrotron): 150 MeV u^{-1} $^{4}He^{2+}$, 400 MeV u^{-1} $^{20}Ne^{10+}$ and 500 MeV u^{-1} $^{56}Fe^{26+}$. The actual energy was calculated for all irradiation conditions from the ion range in water obtained from a Bragg curve measurement. LET variation was achieved by insertion of polymethyl methacrylate (PMMA) binary filters into the beam line. The interval ranged from 2.3 to 339.4 keV μm^{-1} unrestricted LET in water. The residual beam energy and track average LET were calculated for each configuration by means of the SRIM/TRIM 2006 Monte Carlo code (Ziegler et al., 1985). Absorbed doses to water were evaluated from the measured Bragg curve to account for particles scattered off the beam.

^{60}Co gamma rays were used as reference radiation to determine the relative TL efficiency and peak height ratio. Irradiations were performed in air approximately 24 h after annealing of the TLD-300 crystals, using the calibration facility (Philips Theratron) of the Department of Radiotherapy and Radiobiology, Medical University of Vienna, Austria. The detectors were sealed in the same polystyrene holders that had been used for the HCP irradiations. A Farmer-type high-precision ionization chamber calibrated by the Federal Office of Metrology and Surveying (BEV), Vienna, Austria, was employed for determining absorbed dose to water, using correction factors for temperature and air pressure.

3. Results and discussion

3.1. Relative TL efficiency

A great amount of effort was dedicated to the study of TL efficiency, $\eta_{HCP,\gamma}$, with respect to ^{60}Co for HCP radiation fields in attempts to establish techniques of reliable TL dosimetry in exotic environments of mixed particle composition. All results were obtained in the region of linear dose response. Peak 3 efficiency decreases with increasing LET (Fig. 3), while peak 5 efficiency first increases, reaches a maximum at ~ 30 keV μm^{-1} and then decreases with LET (Fig. 4). Hoffmann and Prediger (1983) reported quantitatively similar behaviour for both peaks after exposure to $^{4}He^{2+}$ and $^{20}Ne^{10+}$ available from the LBL Bevalac. Their data were related to absorbed dose to muscle tissue. Loncol et al. (1996) measured peak efficiencies (relative to absorbed dose to water) for an 85 MeV proton beam, thereby complementing the findings of this work in qualitatively good agreement. An in-depth intercomparison of TLD-300 efficiencies determined by different authors can be found in Berger and Hajek (2007). It can be anticipated that the measured response of CaF_2:Tm to different HCPs with the same LET is not a unique function of ionization density (Berger et al., 2006a; Hajek et al., 2006a). A microdosimetric approach (Waligórski et al., 1986) describes the radial dose distribution, $D(r)$, along the particle track as

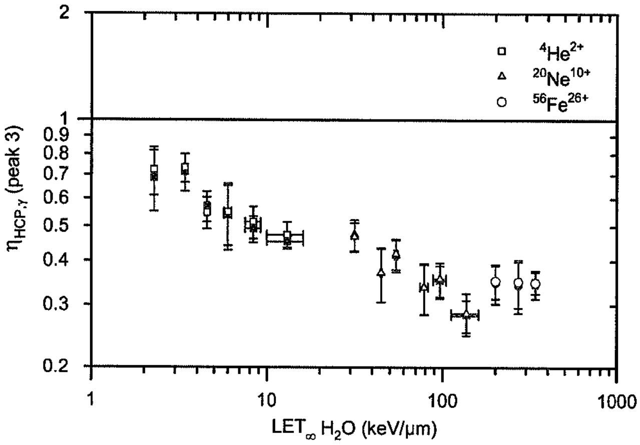

Fig. 3. TL efficiency with respect to ^{60}Co for glow peak 3 in CaF_2:Tm (TLD-300) for several HCPs (solid symbols: CGCD analysis; open symbols: manual analysis).

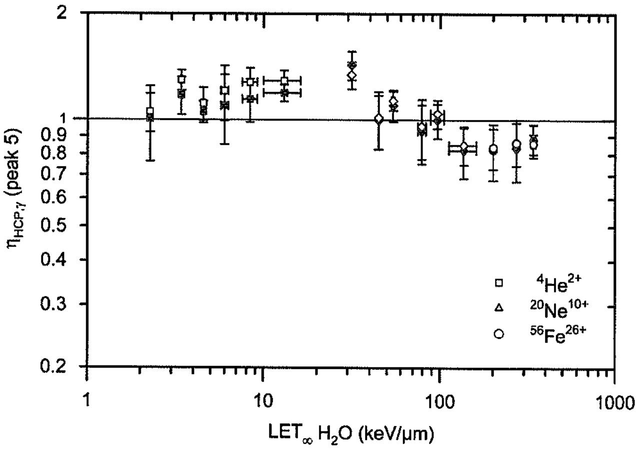

Fig. 4. TL efficiency with respect to ^{60}Co for glow peak 5 in CaF_2:Tm (TLD-300) for several HCPs (solid symbols: CGCD analysis; open symbols: manual analysis).

$$D(r) \approx \frac{Z^{*2}}{(v/c)^2 r^2}, \tag{2}$$

where Z^* is the effective charge, v is the particle's velocity and c is the speed of light in vacuum. For the same value of LET, the velocity of a particle with greater charge must be higher, which leads to lower local ionization density and, consequently, to a higher value of $\eta_{HCP,\gamma}$ (Olko, 2007). Peak efficiencies were evaluated using CGCD (Table 1) and manual analysis of the composite peak structures. For peak 3 as well as for peak 5, both methods agreed within 1 standard deviation (SD). However, due to the composite nature of the HTS the differences tended to be more pronounced for peak 5. In any event, CGCD of the HTS into component glow peaks is time consuming and requires considerable experience—the great number of unknown variable peak parameters results in several possible solutions

Table 1
TL efficiency with respect to ^{60}Co, $\eta_{\mathrm{HCP},\gamma}$, for glow peaks 3 and 5 in CaF_2:Tm (TLD-300) for several HCPs, determined using CGCD analysis

Ion species	LET_∞ H_2O (keV µm^{-1})	$\eta_{\mathrm{HCP},\gamma}$ (peak 3)	$\eta_{\mathrm{HCP},\gamma}$ (peak 5)
$^{4}He^{2+}$	2.3 ± 0.0	0.686 ± 0.134	1.006 ± 0.242
	3.4 ± 0.0	0.714 ± 0.086	1.177 ± 0.144
	4.5 ± 0.1	0.571 ± 0.056	1.056 ± 0.075
	6.0 ± 0.3	0.541 ± 0.112	1.099 ± 0.247
	8.4 ± 0.8	0.493 ± 0.042	1.146 ± 0.160
	13.1 ± 3.1	0.453 ± 0.017	1.193 ± 0.065
$^{20}Ne^{10+}$	31.5 ± 0.1	0.475 ± 0.048	1.429 ± 0.132
	44.8 ± 0.5	0.370 ± 0.064	1.000 ± 0.175
	54.2 ± 1.1	0.415 ± 0.043	1.102 ± 0.112
	78.6 ± 4.1	0.338 ± 0.056	0.928 ± 0.177
	96.5 ± 8.3	0.351 ± 0.037	0.998 ± 0.113
	136.6 ± 24.4	0.281 ± 0.029	0.822 ± 0.137
$^{56}Fe^{26+}$	200.2 ± 0.9	0.347 ± 0.045	0.825 ± 0.148
	270.6 ± 4.3	0.342 ± 0.056	0.836 ± 0.165
	339.4 ± 10.9	0.345 ± 0.032	0.878 ± 0.093

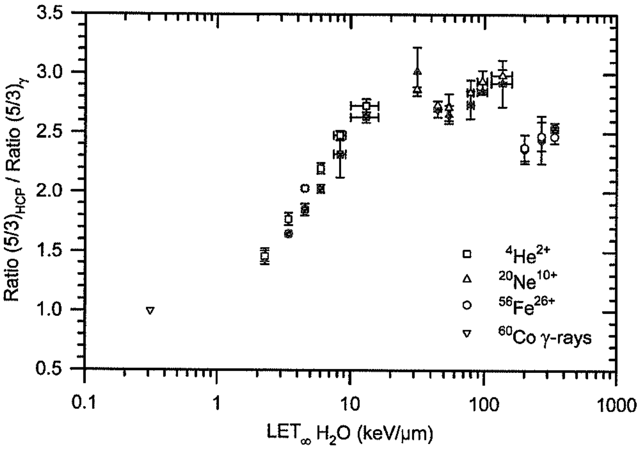

Fig. 5. Relative peak height ratio with respect to ^{60}Co in CaF_2:Tm (TLD-300) for several HCPs (solid symbols: CGCD analysis; open symbols: manual analysis).

of equally good figure of merit (Balian and Eddy, 1977). The results demonstrate that manual analysis can be considered to fulfil the requirements of standard dosimetry.

3.2. Peak height ratio

The relative ratio of peak 5 to peak 3 heights with respect to ^{60}Co in TLD-300 was found to be correlated with LET and further depends on the charge of the particle (Fig. 5). As for ^{7}LiF:Mg,Ti (TLD-700), its slope shows qualitative similarities to the functional dependence of the quality factor, Q, on LET (Hajek et al., 2006b), indicating potential conceptual parallels in the energy deposition on a microscopic level. Due to the non-unique correspondence between the peak height ratio and the LET, a minimum degree of information about the nature of the radiation field is required to use this dependence to evaluate the LET effective in the TL crystal and permit correction of measured doses for TL efficiency (Berger et al., 2006b).

Several authors (Schöner et al., 1999; Yasuda, 2001) have proposed to apply this idea to dose equivalent estimation in the complex space radiation environment using an empiric combination of relative TL efficiencies for specific glow peaks and the HTS as a function of LET. The accuracy of these methods is restricted since ions with charges $Z > 2$ are registered with increasingly lower sensitivity and dose contributions from these ions are consequently underestimated in the integral TL signal.

4. Conclusions

A systematic experimental study of the CaF_2:Tm (TLD-300) response to low doses of HCPs in a wide LET interval from 2.3 to 339.4 keV µm^{-1} revealed remarkable differences between the LET dependence of peaks 3 and 5. TL efficiencies evaluated from CGCD and manual analysis of composite peak structures agreed within 1 SD. The ratio of peak 5 to peak 3 heights depends on ionization density and particle species and can be used to correct measured doses for TL efficiency.

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