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Thermoluminescence dose response of photon irradiated NaCl: Unified interaction model analysis of the dependence of the supralinearity on photon energy

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HIGHLIGHTS

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- Dose Response.
- Unified Interaction Model.

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ABSTRACT

The thermoluminescence (TL) dose response of a common Israeli table salt (NaCl) was measured following ^{137}Cs gamma ray and 125 kVp X-ray irradiation. The dose response is linear, then turns supralinear and finally enters into saturation. The phenomenon of TL linear/supralinear response and the dependence of the supralinearity on ionization density/photon energy is of major interest theoretically and also impacts the practical use of the material as a reliable dosimeter. Prior to this investigation only the supralinearity of the glow peaks of TLD-100 had been investigated. The supralinearity of the major glow peaks (peaks 3 and 5) of the investigated NaCl is strongly dependent on photon energy which lends credence to the probability that the ionization density dependence of TL supralinearity is a common if not universal phenomenon. The Unified Interaction Model used herein is based on the joint action of geminate and conduction band mediated recombination and was used to simulate the dose response of peaks 3 and 5. The simulated dose response allows a reliable estimate of the dose threshold where the supralinearity begins to appear. This information is useful in the evaluation of the Israeli salts as pragmatic retrospective dosimeters in the event of a radiological event.

1. Introduction

Many irradiated thermoluminescence (TL) materials exhibit a linear/supralinear/saturation dose response (Horowitz, 1984). The supralinearity of the glow peaks in the dose response of LiF:Mg, Ti (TLD-100), perhaps the most investigated of all the TL materials, has been shown to be dependent on photon energy/ionization density (ID) (Horowitz, 2001; Livingstone et al., 2010) The normalized dose response, *f(D)* is given by:

$$f(D) = [F(D)/D]/[F_l(D_l)/D_l] \quad (1)$$

where *F(D)* is the TL signal at dose, *D*. $F_l(D_l)$ is the TL signal at low dose, D_l, preferably in the region of linear dose response.

The characteristics of the dose response can be described by D_{th}, the dose threshold at which *f(D)* becomes greater than unity

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(supralinearity begins); $f(D)_{max}$ – the maximum value of $f(D)$ and D_m, the level of dose at which $f(D)_{max}$ occurs. D_m is approximately the same for the three photon energies, the value of $f(D)_{max}$ for the ^{60}Co gamma rays is ~3.5 and for the X-rays ~1.5–2.5. The different values of $f(D)_{max}$ are correlated to differences in D_{th} – an important parameter describing the onset of deviation from linear behaviour. In TLD-100 One finds for glow peak 5 a linear region of constant TL efficiency [$f(D) = 1$] from the lowest dose levels measurable of ~3 μGy up to a dose level of ~1 Gy for ^{60}Co and ^{90}Sr/^{90}Y irradiation followed by a supralinear region in which $f(D)$ reaches a maximum value of ~3–4 at dose levels of approximately 200 Gy. The dependence of the supralinearity on ID is not peculiar to composite peak 5 but rather occurs for all the investigated glow peaks of TLD-100 (Datz et al., 2011).

The dependence of the supralinearity on photon energy/ID has not been investigated in many TL materials. Prior to the work on LiF:Mg, Ti (TLD-100) only one phosphor, CaSO4:Dy (Srivastava and Supe, 1980), was reported to demonstrate such a dependence. A cardinal question is, therefore, whether the ID dependence of the supralinearity is a quirk of these materials or rather a common, perhaps even a universal, phenomenon. LiF:Mg, Ti is known to be a rather quirky material (Horowitz, 1997).

2. Theoretical modeling

Conduction band/valence band kinetic modeling based on delocalized recombination has been very successful in describing some specific features of the TL characteristics, especially the shape of the glow peaks (Chen and Pagonis, 2011). It has not been as successful in describing the linear/supralinear behaviour of the dose response in LiF:Mg,Ti. In fact, delocalized recombination models predict supralinear growth over the entire dose range. These characteristics of linear/supralinear behaviour and especially the dependence of the supralinearity on photon/electron energy arise from the non-uniform spatial ID characteristics following irradiation and have been successfully modeled in the framework of the Unified Interaction Model (UNIM) (Horowitz, 2001). The UNIM is based on nanodosimetric considerations and spatial correlation/coupling between the trapping centers - TC (donors) and the luminescent centers - LC (acceptors) and has been successful in describing many of the features of the supralinearity and sensitization dose response behaviour of LiF:Mg, Ti following photon/electron irradiation. Following irradiation, the spatially correlated TC/LC complex is populated by an electron(e)-only, a hole(h)-only, or an electron-hole (e-h) (Horowitz, 2001).

The relative population of the e-h to the e-only complex is determined by the ID. Only recently has geminate/localized recombination been incorporated in kinetic simulations which successfully simulate linear/supralinear behaviour and its dependence on ID (Eliyahu et al., 2014a, 2014b). UNIM fits to the experimentally measured dose response of peak 5 for various photon energies, achieved by variation of the parameter, *ks*, representing the relative strength of the localized recombination (eq. (2)).

2.1. The unified interaction model: mathematical formulation

In the UNIM, the TL signal intensity is given by

$$F(D) = ksn_e + (1-ks)n_e \sum_{i=1}^{i=4} \int_{r_0}^{R_{max}} g(r_h, R_i) e^{-\frac{R_i}{\lambda}} \times P_i(n_{LC}, R_i) dR_i \quad (2)$$

The first term represents the contribution from localized (geminate) recombination. The second term estimates the increased TL intensity at high dose levels due to conduction band mediated recombination in the presence of competitive processes. n_e is the number density (cm^{-3}) of occupied TCs and its dose dependence is given by $N_e[1-\exp(-\beta_{TC} D)]$ as demonstrated by optical absorption studies (Biderman et al., 2014) where β_{TC} is the dose filling constant of the TC and N_e is the total number density of available TCs. k is the relative probability of geminate/localized recombination in the vicinity of the TC/LC complex not subject to competitive processes and s is the fraction of TC/LC complexes which have captured an e-h pair following irradiation. r_o is the distance between the paired TC and LC and is some measure of the average distance over which geminate recombination can occur. g (r_h,R_i) is an approximate three dimensional solid angle factor between two neighbouring TC/LC pairs so that

$$g(r_h, R_i) \approx \left[\pi r_h^2\right] \Big/ 4\pi R_i^2 \quad (3)$$

$S_{LC} = \pi r_h^2$ is the cross-section for capture of an electron by the h-occupied LC. R_i is the distance between neighbouring TC/LC pairs; λ, the mean free path of the electrons between the TC/LC pairs, is an increasing function of dose (due to the filling (de-activation) of the competitors with increasing dose) and its dose dependence for gamma rays is given by

$$\lambda_g = \lambda_0 \exp(\beta_{cc} D) \text{ where } \lambda_0 = (N_{cc} S_{cc})^{-1} \quad (4)$$

Where β_{cc} is the dose filling constant of the competitive center (CC), N_{cc} is the total number density of available CCs and S_{cc} is the cross section for charge carrier capture by the CC. Returning to eq. (2), $P_i(n_{LC},R_i)dR_i$ is the i'th nearest-neighbour distance probability distribution function (PDF) which have been calculated elsewhere. Fourth and higher-order nearest neighbour interactions do not contribute significantly to the supralinearity of peak 5.

There are thus 9 parameters in the UNIM (β_{TC}, β_{LC}, β_{CC}, ks, r_o, S_{LC}, λ_o N_{LC} and β_{RD} where β_{RD} is used to model the effects of radiation damage at very high levels of dose beyond 1000 Gy. The TC and LC dose filling constants have a dramatic effect on both $f(D)_{max}$ and D_m with a much smaller effect, however, demonstrated by the CC filling constant. The value of the other parameters has a negligible effect on D_m. This selective effect on D_m is significant since the experimental results on NaCl show much lower values of D_m than in TLD-100.

3. Experimental measurements of TL characteristics of an Israeli table salt

The investigation of Israeli common table salts was carried out as part of a comprehensive project to determine the potential of these salts as retrospective dosimeters in the case of a radiological emergency (Datz et al., 2016; Druzhyna et al., 2016). Several brands of salts were investigated and the dose response of one of the major commercial brands irradiated at two photon energies is reported herein. The batch (designated A1) was irradiated with ^{137}Cs gamma rays at the Soreq Research Reactor in Israel and with 125 kVp (HVL of 6.23 mm in Al) X-rays at the Illawarra Cancer Care Center in Australia.

3.1. Glow curve analysis

A typical deconvoluted glow curve following ^{137}Cs gamma irradiation to a dose-level of 1 Gy is shown in Fig. 1. Glow curve analysis was carried out using the Ben Gurion University code with peak shapes based on first order kinetics (Horowitz and Yossian, 1995). The major glow peaks are peaks 3 and 5 at maximum temperatures of 180 °C and 235 °C respectively. The glow curve