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ScienceDirect
Radiation Measurements 43 (2008) 180–184
Radiation Measurements
www.elsevier.com/locate/radmeas

Dimensionless study of the total response of thermally stimulated luminescence

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Abstract

The response of thermally stimulated luminescence (TSL) is investigated theoretically. An original dimensionless treatment is proposed, first to highlight the key dimensionless factors rationalizing the competition between the mechanisms at play during the TSL readout, and second to clean up the approach from any particular values of the relevant physical parameters. The approach holds for an unlimited number of interactive traps. The total response from a current trap is derived and cast into simple formal expressions as a function of the recombination cross-section, the set of trapping cross-sections and of initial trap occupancies (i.e. those existing before the readout of the current peak). This allows one to calculate each peak response by taking proper account of carrier transfers accompanying the successive peak readouts.

Keywords: Thermally stimulated luminescence; Response calculation

1. Introduction

Thermally stimulated luminescence (TSL) is used as a routine solid-state dosimetry technique. Nevertheless, its modelling still suffers from a lack of dimensionless analysis-highlighting the key factors which govern its response. Basic mechanisms at play—trapping, detrapping and recombination—are usually accounted for by a set of differential kinetic equations, but their resolution has not provided the sought factors: simplest formal treatments were based on severe approximations (well-known kinetic order models with single active traps and quasi-equilibrium approximation). More realistic models, involving several interactive traps, have been treated numerically for restricted sets of parameter values (see, e.g. Chen et al., 1981; Bull et al., 1986; Berkane-Krachai et al., 2002; Mady et al., 2006), or formally with significant complexity (Lewandowski and McKeever, 1991).

In dosimetry, what is expected from a TSL theory is basically a relation between the absorbed dose and the measured response. The total response integrated over a relevant temperature range is only governed by intrinsic dosimeter properties, as trapping and recombination cross-sections or trap concentrations, and by trap occupancy at the onset of this range. This occupancy depends on the radiation dose and dose-rate given to the dosimeter prior to the readout (Chen et al., 1981). Thus, the dose response modelling first requires the determination of the initial trap occupancy yielded by irradiation. It then demands to calculate the total response as a function of initial trap occupancy. The present work focuses on the latter stage.

Since we deal with integrated responses, there is no point in considering a time- or temperature-resolved approach here. This frees us from the mathematical complexity that forces to resort to numerical resolution and makes the theory independent of the nature or kinetics of the stimulation allowing the readout. Then, general expressions of the TSL response are derived within a simple formalism. The key coefficients governing the response are naturally pointed out by the dimensionless treatment.

2. Theoretical route of attack

A typical system similar to that of usual TSL models is illustrated in Fig. 1. TSL is assumed to originate from the stimulated release of trapped electrons which subsequently

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doi:10.1016/j.radmeas.2007.11.022

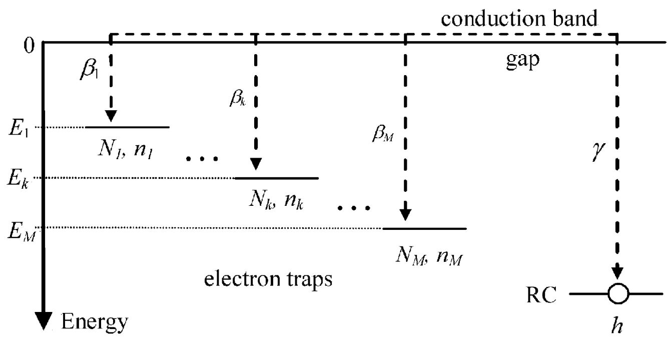

Fig. 1. Typical energy scheme showing traps, recombination centres (RC), trapping and recombination transitions. Trap depths E_k increase with k.

recombine with holes on recombination centres (RC). The treatment being independent of the carrier polarity, it would hold for holes as well. A set of M discrete gap levels is considered. Each level is denoted by its order index k which increases with the trap depth ($1 \leqslant k \leqslant M$). β_k is the trapping coefficient of traps k (electron thermal velocity multiplied by the capture cross-section of traps k), while γ is the recombination coefficient (electron thermal velocity multiplied by the recombination cross-section). N_k and n_k are the densities of traps k and of electrons in traps k, respectively, and h is the concentration of holes on RC.

The total TSL response of the pth peak, due to the emptying of trap p, results from a conversion between two equilibriums characterized by the absence of carriers in the conduction and valence bands. At initial state, electrons are distributed on traps p to M, levels 1 to $(p-1)$ having been emptied at lower temperature. At final state, electrons that have escaped recombination are retrapped on traps $(p+1)$ to M.

In what follows, initial concentrations are indicated by a superscript (p). Final populations are denoted by a superscript $(p+1)$ since they also define initial occupancies relative to the emptying the $(p+1)$th trap. The sample neutrality is then written as

$$h^{(p)} = \sum_{k=p}^{M} n_k^{(p)} \quad \text{or} \quad h^{(p+1)} = \sum_{k=p+1}^{M} n_k^{(p+1)}. \tag{1}$$

During the readout of the current peak p, n_k (for $p+1 \leqslant k \leqslant M$) and h obey usual equations, where n_c is the transient electron concentration in the conduction band:

$$\frac{\mathrm{d}n_k}{\mathrm{d}t} = \beta_k n_c (N_k - n_k), \tag{2}$$

$$\frac{\mathrm{d}h}{\mathrm{d}t} = -\gamma n_c h. \tag{3}$$

For levels $p+1 \leqslant k \leqslant M$, thermal release is neglected within the temperature range of the pth peak. Then, partial integration of Eqs. (2) and (3) between initial and final states gives

$$n_k^{(p+1)} - n_k^{(p)} = (N_k - n_k^{(p)}) \left[1 - \left(\frac{h^{(p+1)}}{h^{(p)}} \right)^{\beta_k/\gamma} \right],$$
$$k \geqslant p+1. \tag{4}$$

3. Dimensionless formulation and total response

After Eq. (4), competition between recombination with holes on RC and retrapping on levels $k > p$ is naturally rationalized by the $\alpha_k = \beta_k/\gamma$ ratios. We furthermore define trap occupancies φ_k as the fraction of occupied traps n_k/N_k, and introduce the proportion ξ_k of traps k with respect to the total density of traps: $\xi_k = \frac{N_k}{\sum_{j=1}^{M} N_j}$.

The total TSL response of the pth peak is equal to the total number of recombination taking place throughout the peak readout. If every recombination is radiative, this response is equal to $h^{(p)} - h^{(p+1)}$, the variation of the hole concentration on RC between initial and final states. We then define the dimensionless total response R_p of the pth peak by

$$R_p = \frac{h^{(p)} - h^{(p+1)}}{N_p}. \tag{5}$$

The normalization is such that $0 \leqslant R_p \leqslant 1$. The case $R_p = 1$ is achieved when all traps $k \geqslant p$ are initially saturated.

From (1), (4) and (5), one obtains the formal equation of R_p for initial occupancy rates $\varphi_k^{(p)}$, trap weights ξ_k and cross-section ratios α_k $(k = p, \ldots, M)$:

$$R_p = \varphi_p^{(p)} - \frac{1}{\xi_p} \sum_{k=p+1}^{M} \xi_k (1 - \varphi_k^{(p)}) \times \left[1 - \left(1 - \frac{\xi_p R_p}{\sum_{k=p}^{M} \xi_k \varphi_k^{(p)}} \right)^{\alpha_k} \right]. \tag{6}$$

This leads to $R_p = \varphi_p^{(p)}$, that is $h^{(p)} - h^{(p+1)} = n_p^{(p)}$, in the absence of traps deeper than the pth one ($\xi_k = 0$ for $k > p$), or if deep traps are initially saturated ($\varphi_k^{(p)} = 1$ for $k > p$). Then, carriers released from the pth trap cannot be retrapped (except on the pth trap itself from which they are rapidly re-emitted). Finally, they all recombine and take part in the total response.

4. Case of a single trapping cross-section

When traps have a same trapping cross-section, i.e. $\alpha_k = \alpha$ for $k = 1, \ldots, M$, Eq. (4) shows that $(\varphi_k^{(p+1)} - \varphi_k^{(p)})/(1 - \varphi_k^{(p)})$ does no depend on k. $\varphi_k^{(p+1)}$ is therefore independent of k if $\varphi_k^{(p)}$ is the same for any k. This is actually true regardless of initial and final moments, even between $\varphi_k^{(1)}$ and trap occupancy before irradiation: if traps are all initially empty, they will fill up parallel to one another under irradiation. Thus, the $\varphi_k^{(1)}$ and the subsequent $\varphi_k^{(p)}$ for $p > 1$ will be all independent of k and