TLS II-38: R. Chen, S.W.S. McKeever, and S.A. Durrani

Dose-rate Dependence of Thermoluminescence: The Possibility of Different Behaviours of Different Peaks

Abstract

Our earlier model, accounting for the dose-rate dependence of TL, has been extended to explain the increase with dose-rate of one component of TL in quartz, and the decrease of another. We deal with two recombination centres and one trap. The set of five simultaneous differential equations governing the traffic of charge carriers has been numerically solved for sets of values of the parameters. The final results of hole concentrations in the centres were usually found to depend on the dose-rate in different ways. Under certain choices of the values of the parameters, one concentration increases while the other decreases with the dose-rate.

Introduction

An implicit assumption generally made in TL measurements used for the dating of archaeological and geological materials is that the TL response is independent of the rate at which the radiation is imparted to the material. In their natural state, the samples usually receive irradiation at rates which may vary from $\sim 0.1\,\mathrm{rad\,yr^{-1}}$ to several $\mathrm{rad\,yr^{-1}}$, depending on the exact environment of the sample under study. For the purpose of calibration, an artificial dose of radiation is usually administered to the sample at a rate which may be several million times greater than the natural dose-rate. It is, therefore, important to consider if the TL response of a sample is at all dose-rate dependent.

If a trapping level is quite shallow and the radiation is delivered at a sufficiently low rate (i.e., the excitation takes a relatively long time in relation to the half-life of the trapped carriers, which depends on the ambient temperature), then one might expect some thermal drainage of the filled traps to take place during the irradiation period. In this case, the final level of the trapped charge-carrier population will be dependent on both the ambient temperature and the dose-rate (see Durrani *et al.*, 1976). One can, usually, bypass this difficulty by choosing materials with TL peaks much above ambient temperature for dosimetry and dating purposes.

A more serious problem is anomalous fading (for example see Wintle, 1973, 1977). If this effect is present, a TL peak may fade away even at a temperature which is much below that at which appreciable thermal fading is expected to occur. This phenomenon is usually attributed to a tunnelling effect of the trapped carriers in a manner which is basically temperature independent.

If thermal drainage or anomalous fading does occur, then the net result is that, for a given total dose, one gets more TL for high dose-rates than for lower dose-rates.

An opposite effect, i.e. higher TL level for lower dose-rate, has been reported by Groom *et al.* (1978). In some cases, powder samples of Brazilian crystalline quartz showed a significant decrease of TL with increasing dose-rate, when irradiated with ^{60}Co gamma rays at dose-rates from 0.14 rad s^{-1} to 330 rad s^{-1}.

A model to account for such « genuine » dose-rate effects (i.e. – not related to thermal drainage or anomalous fading) has been suggested by McKeever *et al.* (1980) in a paper presented at the 6th Int. Conf. on Solid State Dosimetry at Toulouse. In this work, the traffic of charge carriers between the valence and conduction bands, one trapping state and one luminescence centre, was considered (see also the companion paper by McKeever and Chen (1982) in these proceedings). The differential equations governing such flow were numerically solved for chosen values of the relevant parameters, and the results of final concentrations of trapped carriers were compared with the TL results. In some cases, the final concentrations at the end of the « irradiation » were found to be dose-rate dependent; and in these cases the concentrations were found to be higher at lower dose rates. McKeever *et al.* (1980) added, however, another element to this model, which seems to be crucial if one wants to simulate the experimental situation properly. This has to do with the concentrations of electrons and holes in the conduction and valence bands, respectively. These concentrations would decay during the time following irradiation, prior to the readout, such that the decay would contribute more electrons to the trapping state. This contribution is expected to be bigger for higher dose-rates, and this may add an element to the dose-rate effect which may oppose the previously described one (viz., higher equilibrium value of TL at high dose-rates). Under very special choices of the set of values of the parameters, the two effects may exactly cancel each other out. However, calculations have shown that sets of parameter values can be chosen such that the final results yield an increase or a decrease of the total concentration of trapped carriers as a function of dose-rate.

Valladas and Ferreira (1980) have done more experiments on TL in quartz. They used three filters : a green, a blue and an ultraviolet one, and studied separately the three spectral components for their dose-rate behaviour. They found that the uv and green components increased with increasing dose-rate, whereas the blue component deccreased. The present work is an extension of the previous theoretical study (McKeever *et al.*, 1980) to this case of different dose-rate dependences of the different spectral components.

The model

Although the previous model did predict an increase of the measured TL with increasing dose-rate for some sets of values of the parameters and a decrease for others, it is not clear, *a-priori*, if the two effects can occur in the same sample under the same excitation conditions. Since the experimental results are of different behaviours of different spectral components, we have to extend the model to include more than one recombination centre. For the sake of simplicity, we restrict the discussion to a model with one trapping state and two kinds of recombination centres. This is shown in Fig. 1. The parameters involved are : N- concentration of electron trapping states (cm^{-3}) ; n- concentration of electrons in traps (cm^{-3}) ; n_c- concentration of free electrons in the conduction band (cm^{-3}); Sn_v-concentration of free holes in the valence band (cm^{-3}) ; N_h and N_k- concentrations of the two recombination centres (cm^{-3}) ; n_h and n_k-concentrations of holes in the recombination centres (cm^{-3}) ; A_r and A_{kk}- transition probabilities of electrons from the conduction band into the centres ($cm^3 s^{-1}$) ; A- transition probability of electrons from the conduction band into the trap ($cm^3 s^{-1}$) ; A_h, A_k-transition probabilities of holes from the valence band into the centres ($cm^3 s^{-1}$).

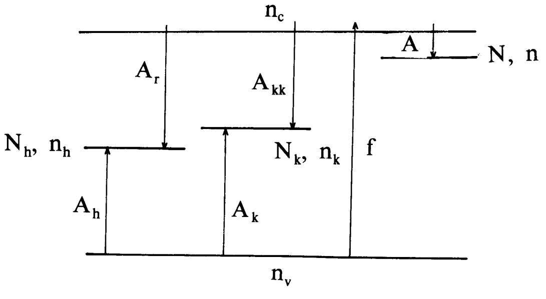

Fig. 1. The energy-level diagram depicting the transitions during irradiation for the one trap - two centres case

We assume that the dose is imparted at such a rate that f electron-hole pairs are produced per cm^3 per second. The dose-rate $r(rad\ s^{-1})$ is thus connected with the « electron-hole generation rate » f by $f = kr$, where k is the ionization efficiency, a constant (for a given material) with units of $cm^{-3}\ rad^{-1}$. Hughes (1975) reported that for quartz, $k = 6\times10^{12}\ cm^{-3}\ rad^{-1}$. Let us denote by $D\,(cm^{-3})$ the « total electron-hole generation »; this magnitude is related to the total dose (in rad) by the same constant k. Assuming f to be constant over the irradiation time, the total generation (cm^{-3}) during t seconds of irradiation is $D = ft$; (if f is not constant, one should take $D = \int_0^t f dt$; but then different possible dose-rate behaviours may occur oven different parts of the irradiation time).

The five differential equations governing the traffic of electrons and holes are the following:

$$dn_c/dt = f - n_cA_rn_h - n_c(N-n)A - n_c\, A_{kk}\, n_k \tag{1}$$

$$dn/dt = n_c(N-n)A \tag{2}$$

$$dn_v/dt = f - n_v(N_h-n_h)A_h - n_v(N_k-n_k)A_k \tag{3}$$

$$dn_h/dt = n_v(N_h-n_h)A_h - n_cA_rn_h \tag{4}$$

$$dn_k/dt = n_v(N_k-n_k)A_k - n_cA_{kk}n_k \tag{5}$$

The simple concept of having the electrons raised into the conduction band accumulating in the trap, and the holes from the valence band accumulating in the centres, would result in the final concentrations of electrons in traps being equal to $D(cm^{-3})$, and the same goes for the total concentration of holes in centres. The additional point here is that recombination is allowed during irradiation (see also McKeever *et al.*, 1980), which seems to be the main reason for the possible dose-rate effect.

For given sets of values of the nine parameters – N, N_k, N_h, A, A_k, A_h, A_r, A_{kk} and f, the equations can be solved numerically. This is done by using the sixth order predictor-corrector Runge-Kutta method. Although the computation procedure is more or less straight-forward, computation is rather long, and in order to get results in a reasonable length of time, the powerful regional CDC 7600 Manchester computer has been utilized.

$D(cm^{-3})$ has been chosen as a parameter for a series of runs. For each calculation in the series, a value of f $(cm^{-3}\ s^{-1})$ is chosen, and the calculation is continued for a time $t = D/f$. In order to simulate relaxation, i.e. the process of the decay of the concentrations of electrons in the conduction band and holes in the valence band following irradiation, the value of f has been set to zero at the end of the time t, and the calculation is continued for a length of time $T = 50$ seconds further. The results are considered as « good » only if n_c and n_v decayed to negligible values during these 50 seconds.

Numerical results

Figure 2 shows the results of $n_h(t+50)$ and $n_k(t+50)$ for the set of parameters: $N = 10^{17}\,cm^{-3}$; $N_k = 10^{16}\,cm^{-3}$; $N_h = 310^{14}\,cm^{-3}$; $D = 10^{16}\,cm^{-3}$; $A = 10^{-16}\,cm^3\,s^{-1}$; $A_{kk} = 10^{-15}\,cm^3\,s^{-1}$; $A_r = 10^{-15}\,cm^3\,s^{-1}$; $A_k = 10^{-15}\,cm^3\,s^{-1}$ and $A_h = 10^{-15}\,cm^3\,s^{-1}$. The value of f changed by a factor of 2 from one calculation to the next, changing overall from $f \cong 10^{14}\,cm^{-3}\,s^{-1}$ to $f \cong 10^{17}\,cm^{-3}\,s^{-1}$. As shown in the figure, n_h changed by a factor of about 6, whereas n_k changed by only ~5 %. The difference in the two dependences is obvious, but both change in the same direction with the dose-rate for the set of parameters chosen.

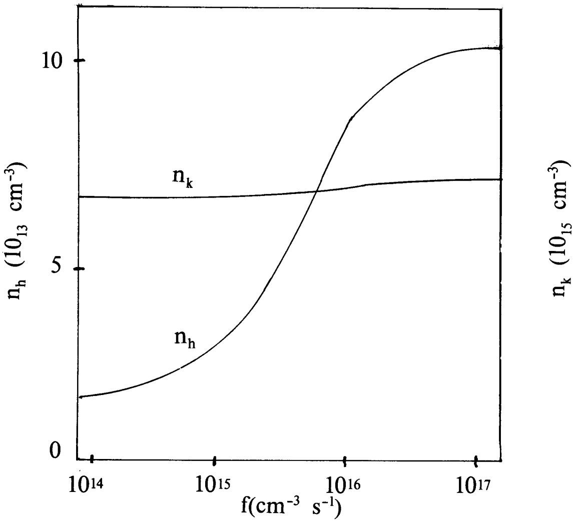

Fig. 2. The final calculated concentrations of holes in the two centres as a function of the dose-rate. Both n_k and n_h change in the same direction, n_h by a factor of ~6 and n_k by only ~5 %. The values of the parameters utilized are given in the text.

Figure 3 depicts the results for a set of values of the parameters that was found to yield « opposite » behaviours of the final filling of the two centres. The values chosen were: $N_h = 10^{15}\,cm^{-3}$; $N = 10^{16}\,cm^{-3}$; $A = 10^{-16}\,cm^3\,s^{-1}$; $A_r = 10^{-13}\,cm^3\,s^{-1}$; $D = 10^{15}\,cm^{-3}$; $N_k = 10^{13}\,cm^{-3}$; $A_{kk} = 10^{-15}\,cm^3\,s^{-1}$; $A_k = 10^{-18}\,cm^3\,s^{-1}$ and $A_h = 1.5\times10^{-15}\,cm^3\,s^{-1}$. The values of f varied from $\sim10^{13}\,cm^{-3}\,s^{-1}$ to $\sim10^{16}\,cm^{-3}\,s^{-1}$.

An effect similar to the experimental variation of TL in quartz with the dose-rate is seen. For a variation of a factor of about 1000 in the dose-rate (or in f), one concentration (n_k) decreases by ~17 % and the other (n_h) increases by a factor of more than 2. This may be compared with a decrease of ~30 % for the blue peak, and an increase of about 70 % in the uv peak and ~10% in the green peak in quartz, when the dose-rate varied by about 1000 as reported by Valladas and Ferreira (1980).

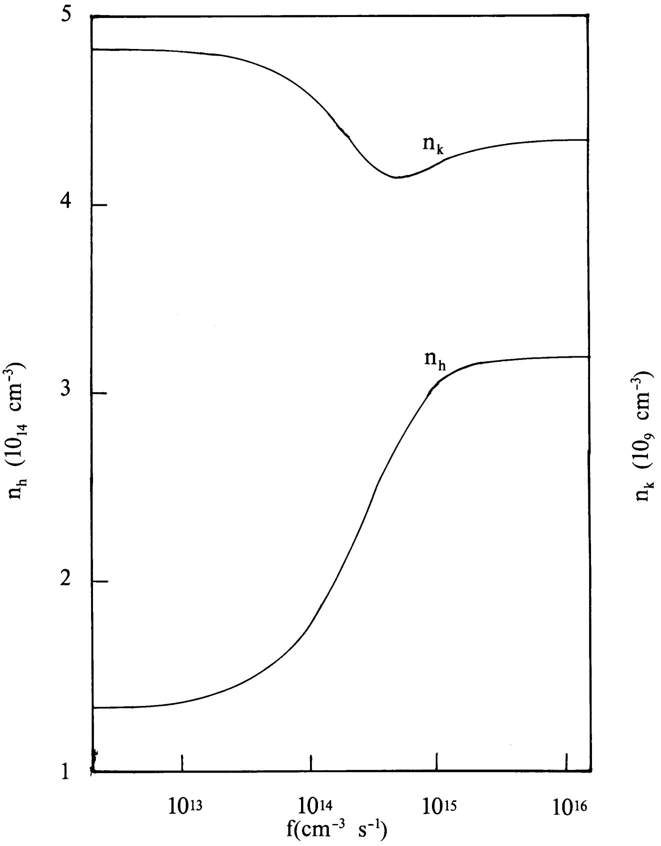

Fig. 3. The final calculated concentrations of holes in the two centres as a function of the dose-rate. Whereas n_h increases with the dose-rate by a factor of >2, n_k first decreases, by ~17 %, and then slightly increases. Note suppressed zero in the ordinate scale. The values of the parameters utilized are given in the text.

Discussion and conclusion

In addition to the previously reported calculation (McKeever *et al.* , 1980), showing that dose-rate effects can be predicted from a simple model of transitions between energy levels, it has been shown above that the model can be extended so as to predict different dose-rate dependences of two TL peaks in the same sample and for the same range of dose-rates. This can qualitatively explain the recent results by Valladas and Ferreira (1980) of different behaviours of peaks having different emission spectra, in quartz. We certainly do not claim that the eight intrinsic parameters chosen (excluding f and D) are necessarily related to quartz, although the ranges chosen for the parameters can be physically considered « reasonable ». The main thing that we have done is merely a theoretical demonstration that such an effect is possible, and therefore should not be considered paradoxical in any sense. This may account for the fact that the agreement between the experiment and the computational results is only qualitative, the experimental effect being less pronounced for one peak and more for the other. It has to be noted, however, that the possibilities of choosing the values of the set of parameters for the computation have, certainly, not been exhausted (and, in fact, cannot really be), and that by no means do we claim that the result given in Fig. 3 is « optimal » in any sense. It is very probable that another set of parameters will approximate the experimental situation much better. In addition, one should remember that, undoubtedly, the present model is still an oversimplification of the real physical situation in quartz, or in any other TL phosphor for that matter. In principle, the treatment given above can be generalized to cover more than the two centres, and, indeed, more than the one trap considered by us : with corresponding increase in the complexity of the solutions. As we have pointed out above, in special cases the net effect may be that of dose-rate independence. If the emission spectrum is not taken into account (e.g. if no filter is inserted between the sample and the photomultiplier), changes in the overall TL ouput will be dominated by those peaks which vary most with dose-rate.

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