

ADVANTAGES AND DISADVANTAGES IN THE UTILISATION OF THERMOLUMINESCENCE (TL) AND OPTICALLY STIMULATED LUMINESCENCE (OSL) FOR RADIATION DOSIMETRY

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Abstract

Thermoluminescence is an established method for radiation dosimetry as well as retrospective dosimetry. In spite of its great success, different difficulties are associated with its application. These include anomalous fading, changes in the properties of the sample due to its heating in the readout phase, including a sensitization due to the combined effect of irradiation and heating and non-linear dose dependence observed in some potential dosimetric materials. Another point is that once the sample is heated during the read-out, the TL signal is erased; this is of help for re-use of the sample, but prevents a repeated reading of the measured quantity. The alternative of using optically stimulated luminescence has become very popular in recent years. Here, the ill effects associated with the heating of the sample are avoided. Also, the possibility of reading out the sample several times exists. This can be done by using short pulses of stimulating light, which deplete only a small part of the trapped charge carriers. The present work points out some difficulties that may take place with OSL. These include a possible superlinearity in the dose dependence as well as dose-rate dependence. Also is discussed the difference between pulsed and integral OSL signals.

Introduction

Thermoluminescence is the physical phenomenon in which a solid sample absorbs energy while irradiated at a given temperature, and releases this energy in the form of light while heating the sample. The emitted light is recorded as intensity vs. temperature in the shape of one or more TL peaks. Under favorable conditions, the emitted TL light intensity is proportional to the absorbed dose, and thus, using an appropriate calibration, one can evaluate the applied dose in the given radiation field. The TL signal may be the intensity at the maximum or the area under the TL glow peak, which are usually nearly proportional to each other. In “regular” dosimetric applications, one can choose an appropriate material with reproducible results in repeated measurements, linear dose dependence for the kind of radiation in question as well as dose-rate independence and long time stability. The situation is significantly more complicated in retrospective dosimetry such as in accident dosimetry where the exposure to radiation is to be determined by using existing materials such as porcelain (see e.g. [1]), which cannot be chosen according to their thermoluminescent properties, and cannot be pre-calibrated. In this sense, the problem is similar to the one occurring in archaeological and geological dating [2].

The main features required for making a material a good TLD (TL dosimeter) candidate are:

1. *Reproducibility.* This sounds like an obvious requirement. However, in some materials e.g. quartz, a sensitization effect has been found [3]. This is an increase of sensitivity as a result of irradiation followed by an annealing at, say, $\sim 500^\circ\text{C}$.
2. *Stability.* A desirable feature is that once the sample in hand is irradiated, the potential TL signal should be stable prior to the beginning of the heating. A fading of the expected signal always occurs to some extent. This includes “normal” fading which is the expected thermal fading commensurate with the trapping parameters, for the given temperature at which the

sample is held (e.g. RT). Anomalous fading has also been detected in some materials, e.g. feldspars. Here, the expected signal is decaying with time much faster than warranted by the peak parameters. It has been explained [4] as being due to the tunneling of carriers from the trap to the center during the waiting period. An alternative account has been given [5] which explained anomalous fading as being a normal fading in disguise, being associated with apparent high values of the activation energy and frequency factor which result from competition with radiationless centers.

3. *Linearity*. For the sake of simple reconstruction of the absorbed dose, linearity of the TL signal with the dose is very desirable. However, superlinearity has been found in many materials, even in the very well recognized dosimetric material LiF [6]. This has been explained by different models based on the competition of the radiative transitions with transitions into non-radiative centers either during the excitation or during the heating (or both). Of course, at high doses, sublinear dose dependence always occurs, when the TL intensity goes toward saturation.
4. *Dose-rate independence*. Although we usually consider the total dose applied to the sample in hand, it has been found that in certain cases, the measured TL is a function of the dose rate. Thus, applying a given dose at a small rate for a long period of time may result in a different amount of emitted TL than when the same dose is applied at a high rate during a short period of time [7]. A theoretical-numerical account explaining the effect has recently been given [8], based on the fundamental understanding of the traps and centers in the sample in hand, and the transitions between them.

More recently, the use of optically stimulated luminescence (OSL) for dosimetry as well as archaeological and geological dating has become very popular. This began with the pioneering work by Huntley et al. [9] on the optical dating of sediments. The advantages of OSL over TL are rather obvious. There is no need to heat the sample, thus avoiding the blackbody radiation occurring at relatively high temperatures. Also, possible thermal quenching of luminescence is avoided, and the sensitization effect taking place in TL is significantly less likely to occur in OSL.

In nearly all the reports on OSL, it is assumed and sometimes shown (see e.g. McKeever and Akselrod [10]) that the initial dose dependence is linear, followed by an approach to saturation. It is also *assumed* that there are no dose-rate effects and therefore, one can calibrate the sample at high dose-rates and deduce the dose (archaeological, geological or “regular” dosimetric) imparted at a much lower rate. There are, however, some reports in the literature on superlinear dose dependence of OSL. In the study of OSL of quartz and mixed feldspars from sediments, Godfrey-Smith [11] found linear dependence on the dose of the unheated samples. However, following a preheat at 225°C, the samples showed a clear superlinearity of the OSL signal at low excitation doses of γ irradiation. Roberts et al. [12] have also found superlinear dose dependence of quartz OSL in several samples. For samples preheated at 160°C, they reported a quadratic equation, $S=aD^2+bD+c$, which describes the dose dependence of the OSL signal S where D is the dose and a , b , c are positive quantities. Bøtter-Jensen et al. [13] further studied the OSL of quartz and found that it was slightly superlinear for the annealed specimens. They explained this using a multiple level model. Banerjee [14] also observed superlinearity in annealed quartz and explained it by adopting the previous models of competition employed for the explanation of superlinear dose dependence of TL. Chen and Leung [15] studied theoretically the dose dependence and dose-rate dependence of OSL. They showed that there is a significant difference between the behavior of the area under the decaying OSL signal and the OSL response to a short stimulating pulse. In the simplest model of one trapping state and one kind of recombination center, the former is linear with the dose whereas the latter is superlinear. Also, it has been shown numerically that for short pulse OSL, and assuming this simplest possible (one trap-one center) model, a dose-rate effect can be expected. These points will be elaborated upon below.

The relevant TL models

In the models of both TL and OSL, we will make the simplifying assumption that the whole sample is uniformly excited by the irradiation. This is basically the case for x-rays, γ -rays and to a good approximation β -rays, and may be a rather poor approximation for heavy particles such as α particles. The possibility, however, of having non-linear effects in the very basic processes described below may have some relevance even in the case of heavy particle bombardment where the excitation is mainly performed by secondary and tertiary electrons. The basic model describing superlinear dose dependence has to do with competing levels. These may be either non-radiative competing centers or disconnected competing traps. The models involve either competition during the excitation stage or during the heating stage or both. These models will be briefly described here.

The first model has to do with competition during excitation [16,17]. A schematic energy model of the levels in the forbidden gap of the thermoluminescent model is shown in Figure 1.

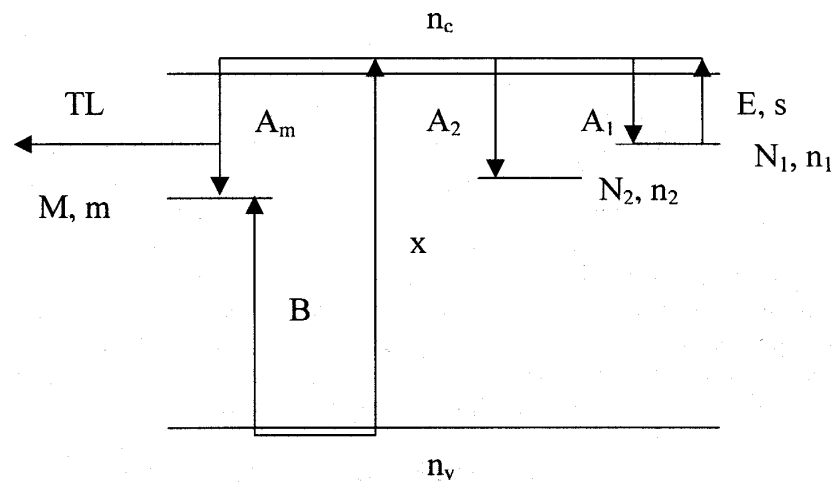


Fig. 1: An energy level scheme for the model of two competing traps and one recombination center.

In this model, it is assumed that the measured TL is proportional to the filling of the active trap $N_1(\text{m}^{-3})$ with instantaneous occupancy $n_1(\text{m}^{-3})$. Here, $N_2(\text{m}^{-3})$ (with instantaneous occupancy of $n_2(\text{m}^{-3})$) plays the role of “competitor during excitation”. Intuitively speaking, let us assume that the trapping probability coefficient $A_2(\text{m}^3\text{s}^{-1})$ into the competitor is larger than that into the active trap $A_1(\text{m}^3\text{s}^{-1})$. At low doses, both n_1 and n_2 grow linearly. The capacity of the competitor N_2 , however, is relatively small and therefore, at higher doses n_2 approaches saturation. More electrons are made available to n_1 and therefore, n_1 is being filled faster. When n_2 is entirely saturated, n_1 grows linearly but at a faster rate than at the low dose range. The intermediate dose range is of interest; when n_2 approaches saturation, n_1 is in the range of transition from small slope linearity to high slope linearity. In this range, the TL intensity is thus necessarily superlinear with the dose. Chen and Bowman [17] used some conventional simplifying assumptions and found an approximate expression which demonstrated the initial linear dose dependence, followed by superlinearity which, in turn, was followed by a second linear range and an approach to saturation.

An alternative model of competition during heating can be intuitively explained as follows. Suppose a certain TL peak is related to a trap with initial concentration n_{1o} and a center with an initial concentration m_o . Normally, one would expect the TL signal (measured either by the maximum intensity I_{max} or the area under the glow peak S) to be proportional to the smaller of the two, $\min(n_{1o}, m_o)$ (see e.g., Chen and McKeever [18]). In other words, S

should be proportional to n_{1o} or m_o , depending on whether the trap or center is depleted first. it may happen, however, in the presence of a competitor with initial concentration of n_{2o} that, say, $n_{1o} > m_o$ but n_{1o} is depleted first since the released electrons may go either to the recombination center or into the competitor. This leads to the dependence of the measured TL on n_{1o} which, in turn, may depend linearly on the dose. In addition, the ratio of the total number of electrons going into the center will be proportional to the relative concentration of holes m_o to that of the empty competitors $N_2 - n_{2o}$. If the latter is far from saturation, then the measured TL is proportional to m_o , which, in turn, may be proportional to the dose. These two linear dependencies on the dose of n_{1o} and m_o in the same dose range combine to quadratic dose dependence. An approximate expression given by Kristianpoller et al. [19] has been

$$S \cong (A_m / A_2 N_2) m_o n_{1o} \quad (1)$$

which explained the quadratic dose dependence previously reported by Rodine and Land [20] for one of the TL peaks occurring in ThO₂. As further shown by Chen et al. [21], a better approximation is

$$S \cong \{ A_m / [A_2 (N_2 - n_{2o})] \} m_o n_{1o}. \quad (2)$$

Since n_{2o} is an increasing function of the dose, $N_2 - n_{2o}$ is decreasing, and since this factor appears in the denominator of the expression, it adds an increasing element to S with the dose. Thus, S is expected to be growing faster than quadratically with the dose. It is to be noted that this extra effect is strong in the range where n_{2o} approaches saturation. As described above, however, the range where n_{2o} approaches saturation is that where competition during excitation plays an important role. This shows that in this region, n_{1o} by itself grows superlinearly with the dose and therefore, the measured TL may be more than quadratic for two different reasons. Here, the approach to saturation of the competitor may have effects both during the excitation and the heating. An important point to understand is that under these circumstances, the two effects of competition during excitation and heating are rather difficult to distinguish.

In order to follow the process without making any simplifying assumptions, one should solve the appropriate sets of simultaneous rate equations. This should be done in three stages of excitation, relaxation and heating. Since the relevant equations are nonlinear, the only reasonable thing that one can do is to solve the equations numerically. This has been done [22] for given sets of trapping parameters, and will be re-iterated briefly here. The set of equations governing the process during excitation, in agreement with Fig. 1 is

$$- dm / dt = A_m n_c m - B n_v (M - m), \quad (3)$$

$$dn_v / dt = x - B n_v (M - m), \quad (4)$$

$$dn_1 / dt = A_1 n_c (N_1 - n_1), \quad (5)$$

$$dn_2 / dt = A_2 n_c (N_2 - n_2), \quad (6)$$

$$dm / dt + dn_v / dt = dn_c / dt + dn_1 / dt + dn_2 / dt. \quad (7)$$

The relaxation period is the period during which electrons and holes relax from the conduction and valence bands respectively until the concentrations of free carriers is negligibly small. This simulates the process taking place between the excitation and heating in the measurement, and is performed by solving the same set of equations (3-7) setting $x=0$, and taking the final values of the concentration functions during the excitation period, as initial values for the relaxation period. In the last stage, the TL behavior during heating is governed by the following set of equations; an additional quantity introduced here is I , the TL intensity given in arbitrary units:

$$dn_1 / dt = -s \exp(-E / kT)n_1 + A_1(N_1 - n_1)n_c, \quad (8)$$

$$dn_2 / dt = A_2(N_2 - n_2)n_c, \quad (9)$$

$$I = -dm / dt = A_m m n_c, \quad (10)$$

$$dm / dt = dn_1 / dt + dn_2 / dt + dn_c / dt. \quad (11)$$

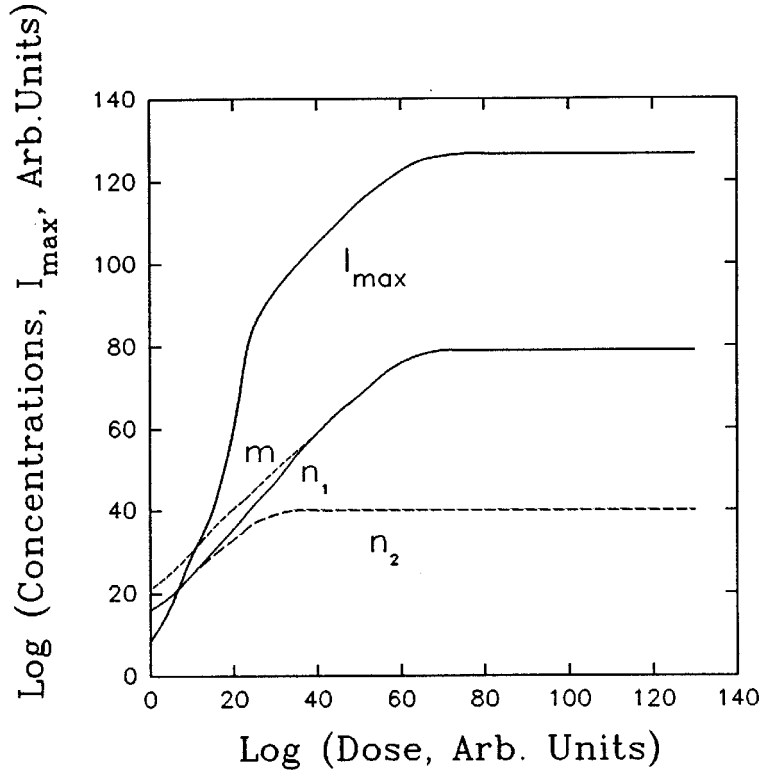


Fig. 2: A sample result based on the model depicted in Figure 1 (Eqs. 3-11). The parameters used were $E = 1.0$ eV, $s = 10^{13} \text{ s}^{-1}$, $B = A_1 = A_m = 10^{-21} \text{ m}^3 \text{ s}^{-1}$, $A_2 = 10^{-19} \text{ m}^3 \text{ s}^{-1}$, $N_1 = 10^{23} \text{ m}^{-3}$, $N_2 = 10^{21} \text{ m}^{-3}$, $M = 1.01 \times 10^{23} \text{ m}^{-3}$, $x = 10^{21} \text{ m}^{-3} \text{ s}^{-1}$. The x-axis gives, on a logarithmic scale, the irradiation time t_D in seconds, from which the relevant dose $D = x t_D$ is readily derived. The curves show the dose dependence of n_{1o} , n_{2o} , m_o and I_{max} .

When this set is solved numerically, the final values of the concentration functions during the relaxation period are taken as initial values here. A heating function is to be assumed; usually linear function, $T = T_o + \beta t$, where β is the heating rate, is taken and the TL intensity as function of temperature, $I(T)$ can be evaluated. As mentioned above either the maximum value, I_m , or the area under the curve, S , can be taken as the TL signal; as pointed out before, these values are nearly proportional to each other in most cases. A sample result is shown in Fig. 2.

The OSL model

At first sight it looks that TL and OSL are supposed to behave at the same way as far as the signal intensity vs. the dose is concerned. Indeed, the excitation stage is exactly the same, no matter whether the read-out is being performed thermally or optically. Banerjee [14] has made this analogy and suggested that the superlinearity observed is due to exactly the same reasons as in TL. He has stated that superlinearity of OSL can take place only when a competitor participates in the process and stated that similarly to TL, strong superlinearity

takes place when the competition is between two trapping states whereas only weak superlinearity can be expected when the competition is between recombination centers. Chen and Leung [15] have suggested that there is a difference between the measurement of pulsed OSL and integral OSL. We are going to elaborate on this point here, and will show that there is a significant difference between the two cases. Let us demonstrate first that when the integral under the OSL decaying curve is concerned, a similar behavior to that of TL can be expected. The same set of equations (3-7) should be numerically solved in the two stages of excitation and relaxation. As for the read-out phase, Eq. (8) should be replaced by

$$dn_1 / dt = -fn_1 + A_1(N_1 - n_1)n_c \quad (12)$$

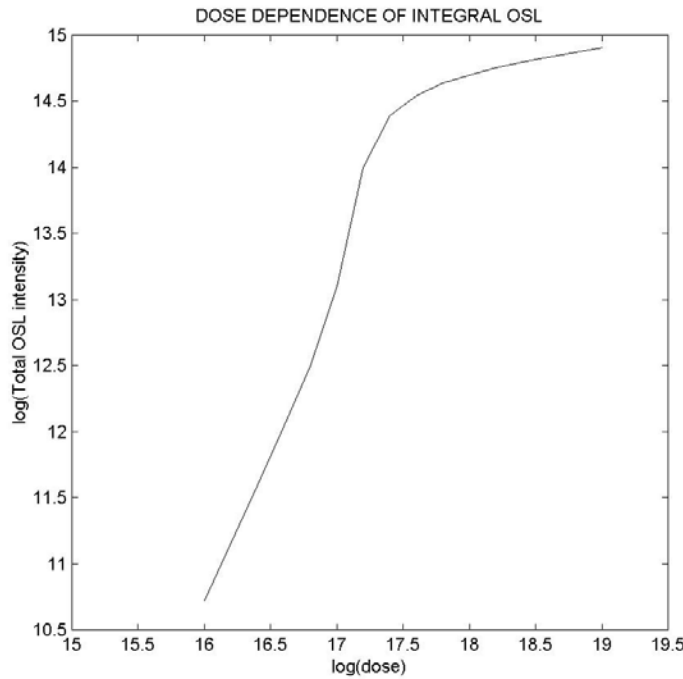


Fig. 3: Simulated dose dependence of integral OSL for the case of two trapping states and one kind of recombination center as calculated from Eqs (3-7) and (9-12). The parameters chosen were $A_m = 10^{-17} \text{ m}^3 \text{ s}^{-1}$, $B = 10^{-16} \text{ m}^3 \text{ s}^{-1}$, $N_1 = 10^{17} \text{ m}^{-3}$, $N_2 = 10^{16} \text{ m}^{-3}$, $A_1 = 10^{-16} \text{ m}^3 \text{ s}^{-1}$, $A_2 = 10^{-15} \text{ m}^3 \text{ s}^{-1}$, $M = 10^{18} \text{ m}^{-3}$, $f = 1 \text{ s}^{-1}$ and x varied from 10^{16} to $10^{19} \text{ m}^{-3} \text{ s}^{-1}$.

where the intensity of the stimulating light is represented by $f(\text{s}^{-1})$. It should be noted that fn_1 in Eq. (12), having units of $\text{m}^{-3} \text{ s}^{-1}$ is analogous to x in the excitation stage, and is the rate of release of trapped electrons per second and per m^3 . The main difference between the two is that x is usually constant whereas fn_1 varies with time when n_1 decreases. An example of the calculated integral dose dependence is shown in Fig. 3. The similarity with the TL dose dependence under the same circumstances is obvious; it is seen to be quadratic at low doses, more than quadratic at intermediate doses and tends to saturation at high doses.

It is quite obvious that in the absence of competing levels, both TL and integral OSL should depend linearly on the dose. However, it has been suggested by Chen and Leung [15] that as long as pulsed OSL is utilized, superlinear dose dependence may sometimes take place even when only one trapping state and one kind of recombination center are involved. In particular, in cases where retrapping plays an important role in the process and both the trapping state and recombination center are empty at the beginning of the excitation, quadratic dose dependence of the OSL signal can be expected. An example is shown in Fig. 4.

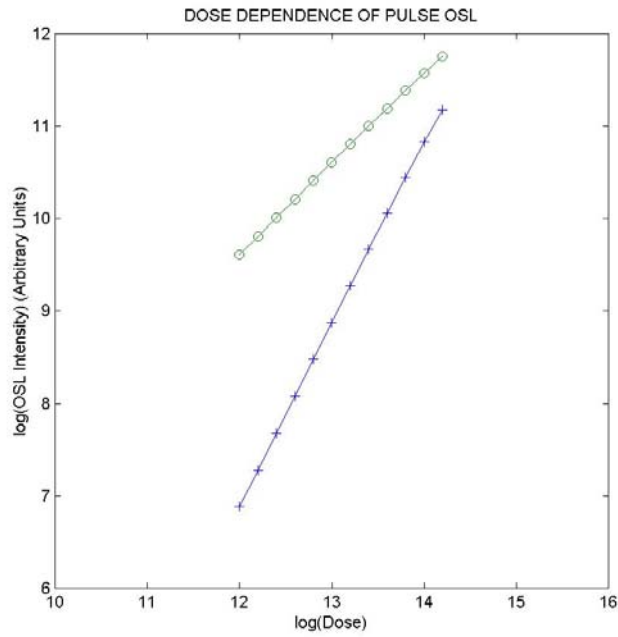


Fig. 4: Dose dependence of pulse OSL when only one trapping state and one kind of recombination center are involved. (+) represent the results when excitation starts with empty traps and (o) when $n_o=0.9N$. The values of the chosen parameters are the same as in Fig. 3. Eqs. (3-5) and (7) have been solved for excitation and relaxation and (10-12) for the read-out.

Summary

The possibility of non-linearity of the dose dependence of TL and OSL has been considered. As previously noted, TL intensity may be superlinear with the dose, and it has been pointed out that the integral OSL may behave in a similar way in the presence of a competitor. The additional point discussed here is that even when only one trapping state and one kind of recombination center are involved in the process, quadratic dose dependence may be expected. Using the quite well known quasi-equilibrium assumption, namely that $n_c \ll n$ and $|dn_c/dt| \ll |dn/dt|$, one can easily get, in analogy with TL, the approximate expression

$$I = \frac{fA_m m n}{A_m m + A_n (N - n)}. \quad (13)$$

It can be seen from this that in the case of dominating retrapping, $A_n(N-n) \gg A_m m$ and when the trap is far from saturation, $N \gg n$, this yields a quadratic behavior as long as m and n grow linearly with the dose. Moreover, when the trap approaches saturation, a small range of more than quadratic behavior is expected when $N-n$ in the denominator decreases, thus adding a super-quadratic element to the dose dependence prior to its saturation when n and m saturate.

It should be noted that whereas the integral OSL is analogous to the area under the TL curve, the pulse OSL is analogous to the intensity at a certain temperature within the initial-rise range of TL. It has been shown [23] that for second order kinetics peaks, the dependence of the intensity in the initial-rise range on the dose is quadratic. The discussion given here concerning Eq. (13) has to do with the similar situation when the filling of traps and centers is only sampled using the stimulating pulse.

Finally, it is only mentioned briefly that by using the same simple model, a certain amount of dose-rate effect can also be predicted.

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