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THERMOLUMINESCENCE SYSTEMS WITH TWO OR MORE GLOW PEAKS
DESCRIBED BY ANOMALOUS KINETIC PARAMETERS*

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Abstract

The thermoluminescence (TL) glow peaks obtained from materials exhibiting two or more glow peaks, e.g. NaCl, quartz and other minerals, often appear to be well described by the well-known first and second order TL kinetics. However, in many cases physically unrealistic E (activation energy) and s (pre-exponential factor) parameters are obtained. In addition, the kinetic parameters often depend on the pre-measurement radiation dose that controls the initial trapped charge concentrations. The usual first and second order TL kinetic expressions are based on a number of assumptions, including the usually unstated assumption that charges released from one type of trap, giving rise to one glow peak, are not retrapped on other types of traps, associated with other glow peaks. Equations have been developed describing TL systems in which charges released from one type of trap may be retrapped in other types of traps. Called interactive kinetic equations, they are quite simple but have been studied by numerical methods. In particular, glow curves computed from the interactive kinetic equations have been regarded as "data" and analyzed by fitting them to the usual first and second order kinetic expressions. All of the anomalous features described above are reproduced. For example, usually the computed glow peaks are well fitted by the first and second order expressions over their upper 60 to 80 percent but not in the "wings." This explains why the usual analysis methods, especially those utilizing peak temperature, full width, etc. appear to describe such peaks. Often unrealistic kinetic parameters are often obtained. Furthermore, the computed glow curves often reproduce the observed dependence on dose.

1. Introduction

Thermoluminescence, which is the light emitted by a substance when it is heated and is the result of previous exposure to ionizing radiation, has been understood qualitatively and, to some extent, quantitatively since the late 1940's. A typical thermoluminescence study consists of several steps. First, the sample is exposed to a known amount of ionizing radiation at a temperature appreciably lower than that producing glow peaks. Second, as the sample temperature is increased, almost always linearly, the emitted light is recorded. The resulting curve of intensity vs. temperature, or time, is called a glow curve and contains one or more glow peaks. Third, as discussed in some detail below, the resulting glow curve is regarded as the superposition of a number of glow peaks and each peak subject to analysis to determine the kinetic parameters determining the peak shape, peak temperature, etc. If the recorded data is actually a curve of phototube current vs. time, or temperature, to make a valid kinetic analysis it is necessary to assume that the current is strictly proportional to the number of thermoluminescence processes occurring per unit time. Usually this condition is reasonably well fulfilled. Below, thermoluminescence will be abbreviated TL.

In almost all studies the analysis of the recorded glow curves are based on the first order TL kinetics originally derived by Randall and Wilkins [1] or the second order TL kinetics developed by Garlick and Gibson.[2] These kinetics are given by expressions that describe the shape, peak temperature, full width at half maximum (FWHM), dependence on heating rate and other characteristics of a single glow peak. They

do not provide information on the absolute intensity of the glow peaks. As will be explained in detail in later sections, under proper conditions the kinetic analysis can provide: 1) the order of the kinetics, 2) the activation energy for thermal untrapping E , and 3) the preexponential or "attempt-to-escape" frequency s .

During the past decade or two a large number of TL publications have appeared which contain kinetic results that are questionable for one or more reasons. These include:

a) Cases where physically unrealistic kinetic parameters are obtained. More specifically, often the s parameters lie outside of the range $10^8 \leq s \leq 10^{13}$ which can be considered physically realistic. Also, although the range of reasonable E values is not welldefined, unusually small E values are often observed. The small E values are often accompanied by abnormally large s values.

b) The peak temperatures and kinetic parameters of some glow peaks vary little with the pre-measurement dose while, in contrast, the parameters for other peaks depend strongly on dose. For example, in natural quartz [3] glow curves obtained after the samples had been exposed to a variety of doses contained some glow peaks that could be well fitted by first order kinetics and others that could be fitted by second order kinetics. However, the measured kinetic parameters depend--in an erratic manner--on the pre-measurement dose.

c) Glow peaks are observed that cannot be fitted by either first or second order kinetics. Recent TL measurements on melt grown NaCl crystals and natural rock salt [4] produced results similar to those obtained from quartz and, in addition, a large fraction of the measured

glow curves contained glow peaks that could not be fitted by either kinetics. The reliability of these anomalous results is strengthened by the fact that they were made with equipment which produced reasonable kinetic parameters when other TL systems were studied.

The studies described below were undertaken in an effort to explain these anomalous TL results. Although carried out in a somewhat different sequence, the development in this paper can be considered the development of TL kinetics for systems not described by the usual first and second order TL kinetics equations.

2. The basic single glow peak TL kinetic equation

To provide a basis for the more general TL kinetic equations described below, the well known first and second order TL kinetic equations will be derived in a way that emphasizes the approximations involved. The treatment follows closely that given by Randall and Wilkins [1] and Garlick and Gibson [2]. Also, although a number of different generalizations of these equations can be considered, the one given below appears to be the simplest possible. It is a obvious extension of the development in references 1 and 2.

Consider a TL system consisting of a single electron trap and a single hole trap. Also, assume that TL emission occurs when thermally untrapped electrons recombine with trapped holes at the hole traps, i.e. the hole traps are also recombination centers. Let N and N_T be the electron and hole trap concentrations. In the case given here N_T will not appear in the equations. After the sample has been irradiated the trapped electron and hole concentrations will be n_0 and n_{T0}

and, $n_0 = n_{T0}$. The thermal untrapping of electrons is given by the usual expression, $dn_1/dt = -n_1 s_1 \exp(-E_1/kT)$ where E_1 and s_1 were described above, n_1 is the trapped electron concentrations at time t , and n_r is the corresponding hole concentration. For simplicity it is assumed that hole untrapping is negligible in the region where $dn_1/dt \neq 0$, i.e., only electron untrapping is contributing to the glow peak. If at time t the trapped electron concentration is n_1 , the probability that an electron in the conduction band will be trapped on an empty electron trap is proportional to $\sigma_{t1}(N_1 - n_1)$ where $(N_1 - n_1)$ is the concentration of empty electron traps and σ_{t1} is the cross section for electron retrapping. Likewise the probability that a conduction band electron will undergo a light emitting recombination is proportional to $\sigma_r n_r$, where σ_r is the cross-section for electron-hole recombination. Also, assume that the system is irradiated at a temperature where all thermal untrapping processes are negligible and, at a later time, the sample is heated linearly during the TL measurement, i.e. the sample temperature is given by $T = T_0 + \beta t$ where β is the heating rate. Then the equation describing the trapped electron concentrations during heating is

$$\frac{dn_1}{dt} = -n_1 s_1 \exp(-E_1/kT) \left(1 - \frac{\sigma_{t1}(N_1 - n_1)}{\sigma_{t1}(N_1 - n_1) + \sigma_r n_r} \right) \quad (1)$$

the term preceeding the brackets is the number of electrons thermally released to the conduction band per unit time. The bracketed term is the fraction of conduction band electrons undergoing recombination.

The TL light intensity, I , at time, t , is

$$\frac{dn_1}{dt} = \frac{dn_r}{dt} = I(t) \quad (2)$$

In other words, in this simple derivation it is assumed that all recombination events give rise to TL emission.

2.1 The first order TL expression

If in eq. (1) it is assumed that $\sigma_r \gg \sigma_{t1}$ or $\sigma_r n_r \gg \sigma_{t1}(N_1 - n_1)$, which is physically equivalent to assuming that retrapping is negligible, then eq. (1) becomes

$$\frac{dn_1}{dt} = -n_1 s_1 \exp(-E/kT). \quad (3)$$

Since n_1 appears with exponent 1, this eqn. and its solution, obtained from the linear heating condition $T = T_0 + \beta t$, namely

$$I(T) = n_{01} s_1 \exp(-E_1/kT) \exp\left(-\int_0^T \frac{s_1}{\beta} \exp(-E_1/kT) dT\right). \quad (4)$$

are usually referred to as the first order TL kinetic expressions.

2.2 The second order TL expression

In contrast, if in equation (1) it is assumed that retrapping occurs and, in addition, it is assumed that $n_1 = n_r$ (or equivalently that $n_{01} = n_{r0}$) and that $\sigma_{t1} = \sigma_r$, i.e. the cross sections for retrapping and recombination are equal, then eq. (1) becomes

$$\frac{dn_1}{dt} = -dn_1/dt = (n_1^2/N_1) s_1 \exp(-E_1/kT) \quad (5)$$

In as much as n_1 appears to the power 2, this eqn. and its solution, using $T = T_0 + \beta t$, namely

$$I(T) = \frac{n_{01} s_1^* e^{-E_1/kT}}{\left(1 + \frac{s_1^*}{\beta} \int_0^T \exp(-E/kT) dt\right)^2} \quad s_1^* = \frac{n_{01}}{N_1} s_1. \quad (6)$$

are known as the second order TL kinetic equations.

The principal properties of eqns. (4) and (6) are described in reference 2 and in a number of books. [5-8] Unfortunately, it appears that they have been used extensively without regard to the assumptions invoked in their derivation. As will be explained, these equations require an additional assumption that is, as far as can be determined, seldom mentioned explicitly. Namely, the derivation assumes that only one type of electron trap is present, i.e. interactions involving more than one type of trap are ignored. It should be emphasized that these equations describe the shape, peak temperature, dependence on heating rate, etc. They do not provide information on the intensity of individual glow peaks. The most important properties of the first order expression, eqn. (4) are:

- a) The shape and peak temperature depend on heating rate.
- b) For a fixed heating rate, the peak temperature and shape are independent of the initial trapped electron concentration n_{01} . In turn, the value of n_{01} depends on the pre-measurement dose. The TL glow curve obtained for any n_{01} value can be superimposed on the curve obtained for a different n_{01} by multiplying by an appropriate factor.

The principle properties of the second order glow curve expression, eqn. (6) are:

- a) For all other parameters kept constant, the shape and peak temperature depend on heating rate.
- b) For a fixed heating rate, the peak temperature and shape are strongly dependent on the initial trapped charge concentration n_{01} . Peaks obtained for different initial trapped charge concentrations cannot be superimposed by multiplying by a factor.

c) The glow peaks obtained for different n_0 values tend to superimpose at the high temperature extremity of the glow peak.

Additional details on the derivations given in this section and graphical demonstration of the properties described above are contained in recent articles.[9-11]

2.2 Analysis of Glow Curves Using the First and Second Order Kinetic Expressions

Since their introduction, the first and second order kinetic expressions have been the basis for a large number of methods for extracting the kinetic parameters, s_1 and E_1 , from measured glow curves. These can be divided into two categories. First, numerous formulas have been derived for computing s_1 and/or E_1 values from measured parameters such as: 1) variations of peak temperature with heating rate and 2) the peak temperature and full width at half-maximum (FWHM) or the half width at half-maximum (HWHM). These methods can, and very often are, applied without determining if the glow peak shapes are described by equations (4) or (6). Heuristically, they can be applied if any of the necessary parameters such as the peak temperature or FWHM, can be extracted from the measured glow curves. Recent reviews of these methods are contained in reference 8 and 12.

Almost all recorded glow curves contain two or more glow peaks that usually overlap. Thus it is obviously advantageous to have methods for resolving such curves into individual peaks and determining the kinetic order and kinetic parameters of each peak. Consequently, in the late 60's methods were developed for fitting combinations of the first and

second order kinetic expressions to measured glow curves using computerized best-fit procedures, e.g. see Mattern et. al.[13-14], Fairchild et al.[15] and Fuller and Levy [3]. These fitting procedures often make it possible to determine if a given glow peak is best fitted by first or second order kinetics, or not fitted by either. A good example is reference 14. In recent years these fitting procedures have provided information to conclude that some glow peaks are not well described by either of the usual expressions or, in some instances the glow curves are well fitted but that physically unrealistic kinetic parameters are obtained [3,4,15].

2.4. Properties of single glow peaks not described by first and second order kinetics

To determine the properties of single glow peaks that are not described by the usual first and second order kinetics, equation (1) was solved numerically for a variety of parameters that do not fulfill the assumptions described above. These solutions are described in some detail, along with graphical illustrations, in references 9, 10 and 11. The computed glow peak shapes differ markedly from those computed from eqns. (4) and (6). For example, the glow peak shapes depend strongly on the σ_{t1}/σ_r ratios, i.e. on the trapping and recombination cross sections. Also, they depend on the relative size of n_1 and n_r . More specifically if the $n_{01}=n_{r0}$ condition is not fulfilled the glow peak shapes differ from those for which $n_{01}=n_{r0}$. Obviously, as σ_{t1}/σ_r approaches one and/or n_1 approaches n_r the computed curve more closely resemble the usual curves.

3. Interactive TL kinetics: properties of TL systems containing more than one type of trap

As mentioned above, the usual first and second order kinetic expressions and, most importantly, the various kinetic formulas derived from them make use of an often unstated or implicit assumption. Namely, it is assumed that the TL system contains only one type of trap or, if more than one type of trap is present, it is assumed that the processes involving one type of trap do not interact with other types of traps. To investigate systems in which interactions between different types of traps are allowed a number of generalizations of eqn. (1) could have been studied. The system chosen for study appears to be the simplest possible generalization of the one trap system originally considered by Randall and Wilkins and by Garlick and Gibson.

Consequently, consider a system consisting of j different types of electron traps and a single type of hole trap. Let N_i be the concentration of the i th type of electron trap, N_T the concentration of the single type of hole trap with $N_T \geq N_1 + N_2 + N_3 + \dots$, n_i the trapped electron concentrations and n_T the trapped hole concentration at time t , n_{0i} and n_{r0} the corresponding concentrations at $t=0$, σ_{ti} the cross section for electron retrapping on the $(N_i - n_i)$ empty electron traps of the i th type, and σ_T the cross section for TL light emitting electron-hole recombination. Lastly, assume that the system is irradiated at a temperature where all thermal untrapping processes are negligible. Thus, before heating the initial trap charge concentrations are n_{0i} , n_{r0} and $n_{01} + n_{02} + n_{03} + \dots = n_{r0}$. Then, by strict analogy with the equations given above, the equations for the trapped electron and hole concentrations during heating are:

$$\frac{dn_i}{dt} = -n_i s_i e^{-E_i/kT} + \left(\sum_{i=1}^j n_i s_i e^{-E_i/kT} \right) \frac{\sigma_i (N_i - n_i)}{n_r + \sum_{i=1}^j \sigma_i (N_i - n_i)} \quad (7)$$

$$I(t) = -\frac{dn_r}{dt} = -\sum_{i=1}^j \frac{dn_i}{dt} \quad (8)$$

$$\sigma_i = \sigma_{ti}/\sigma_r \quad (9)$$

In eqn. (7) the first term describes the thermal untrapping from the ith type of trap, the first bracket gives the total charge concentration in the conduction band at time t and the second bracket is the fraction of conduction band electrons retrapped on the ith electron trap. The conduction band electrons that are not retrapped will recombine with holes to produce TL light I(t), as described by eqn. (8).

Note that $\sigma_i = \sigma_{ti}/\sigma_r$ throughout. Thus only the retrapping-recombination cross section ratios appear, not the actual cross sections. It is particularly important to emphasize the fact that these equations describe a TL system in which charges released from one type of electron trap may be retrapped on all of the empty electron traps present. The lifetime of the trapped electrons in the low temperature traps may be very short. However, the dominant effect is the retrapping of charges released at low temperatures by traps operative at high temperatures. The term interactive kinetics will be used to describe TL systems in which more than one type of trap is present and charges may be trapped on all empty traps. In contrast, the term non-interacting kinetics will be used to describe the usual kinetics in which it is implicitly assumed that interactions between different types of traps do

not occur. When $n_r \sigma_r \gg \sum_{i=1}^j \sigma_{ti} (N_i - n_i)$ these equations reduce to a set of j independent first order kinetic equations. In a sense, this can be regarded as justification for analyzing certain glow curves on the basis that they contain j non-interacting first order glow peaks.

4. Properties of TL systems described by interactive kinetics

The properties of interactive TL systems have been studied by obtaining computerized numerical solutions of eqns. (7), (8), and (9). For example, Fig. 1 demonstrates that the glow curves (solid lines) depend strongly on the σ_{ti}/σ_r cross section ratios. To compute these glow curves the following numerical parameters were used in eqns. (7), (8) and (9): $N_1 = N_2 = N_3 = 10^{16}$, $n_{01} = n_{02} = n_{03} = 5 \times 10^{15}$, $\sigma_{ti}/\sigma_r = \sigma_i$, $n_{r0} = 1.5 \times 10^{16}$, $s_1 = s_2 = s_3 = 10^{10}$, $E_1 = 1.0$ eV, $E_2 = 1.25$ eV, and $E_3 = 1.50$ eV, and the σ_{ti}/σ_r ratios are as given. Also, to compare the interactive kinetic glow curves with the usual kinetics, typical first and second glow peaks are shown. The latter were computed from eqns. (4) and (6) using numerical values appropriate for the values given above, namely $N_i = 10^{16}$, $n_{0i} = 5 \times 10^{15}$, $s_i = 10^{10}$ and $E_1 = 1.0$ eV, $E_2 = 1.25$ eV and $E_3 = 1.50$ eV. These first and second curves also appear in other figures. In all cases the shape and position of these peaks are significant, not the height or intensity.

This figure also demonstrates that the interactive kinetics glow curves approach the usual first order glow curves as the σ_{ti}/σ_r ratios decrease, i.e. as the retrapping probability decreases or, alternatively, as the recombination probability increases. Recall that the usual first order kinetics is obtained when retrapping is negligible compared to recombination.

However, one might expect that putting $\sigma_{ti} = \sigma_r$, which is one of the principle conditions used to obtain the usual second order kinetic expression, would produce glow curves resembling the non-interaction second order glow peaks. Figure 1 clearly demonstrates that this does not occur, at least in this example. In fact, this figure suggests that, even though interactive retrapping occurs, the interactive kinetic glow peaks more closely resemble first order than second order peaks.

In the derivation of the usual second order kinetics it is explicitly assumed that the concentration of trapped electrons and hole remain equal. Fig. 2 demonstrates that when interactive kinetics apply the shape and position of the glow peaks depend on the distribution of the initial trapped charge concentrations, i.e. on the n_{0i} values. In this example the total electron concentration remains equal to the total trapped hole concentration. However, the initial distribution clearly influences the glow peak properties.

To demonstrate how the properties of interactive kinetics glow curves depend on the magnitude of the initial trapped charge concentration a number of different situations have been studied by obtaining numerical solutions of (7), (8) and (9). This case is particularly important since it demonstrates how the glow curves and glow peak properties depend on the dose imparted to the sample prior to measurement. (The manner in which the premeasurement dose and dose rate controls the initial trapped charge distribution is an important aspect of TL which will not be discussed here.) Primarily to include the simplest possible instructive case, the system to be studied in detail will consist of three electron traps N_1, N_2, N_3 , and one hole trap N_r with $N_1 + N_2 + N_3 < N_r$, the $\sigma_r/\sigma_{ti} = 0.1$, the E_i and s_i values the same as

above, and the initial trap concentrations equal, i.e., $n_{01} = n_{02} = n_{03}$. The resulting glow curves, computed from (7), (8) and (9), are shown in Fig. 3 for n_{0i} values ranging from 5×10^{11} to 5×10^{15} .

The computed glow curves shown in Fig. 3 contain appreciable information of which only the most important features, from the point of view of understanding TL measurements, are sketched below. 1) The glow peak shapes depend on the initial trapped charge concentrations. 2) As the n_{0i} values decrease the relative heights of the peaks change drastically. At low n_{0i} values the height of the lowest temperature peak is so low compared to the other peaks it would not be detected with typical TL apparatus with dynamic range less than 10^3 . 3) At high n_{0i} values the computed glow curves most closely resemble non-interactive first order peaks. This is not surprising; as the n_{0i} values increase re-trapping becomes likely. 4) Most importantly, the peak temperatures are functions of the n_{0i} values. In this example the peak temperatures of the two lower temperatures undergo relatively little change as the n_{0i} values change. In marked contrast, the peak temperature of the highest temperature peak changes markedly with changing n_{0i} values. 5) Similar glow peak shape changes occur. 6) At high temperatures all of the glow curves tend to superimpose. As noted previously, [2,9-10] this is a property of single glow peaks described by the usual second order kinetics.

5. Analysis of interactive kinetics TL glow curves

To determine the properties of interactive kinetic glow curves and to establish their applicability to the numerous analyzed glow curves in the literature, the computed glow curves shown in Fig. 3 were regarded

as "data" and analyzed, where possible, using the computerized best-fit methods described in the Introduction. It is essential to outline the steps included in this analysis:

a) The peak temperature and FWHM are determined. These can be determined for all of the curves in Fig. 3. Thus E_i and s_i values, whether they are physically realistic or not, can be computed for all peaks using the approximate formulas [8,12]. In other words, even if the measured glow peaks are not shaped in a manner consistent with the usual kinetic expressions, E_i and s_i values can be obtained.

b) The E_i and s_i values computed from the approximation formulas are used as "starting-values" in the computerized procedure that determines the values of E_i and s_i that provide a best fit of eqns. (4) and (6) to the measured glow peaks.

c) The computerized best fit procedures continues until one of three things occur. 1) The program converges to a best fit. 2) The program indicates that the convergence procedure cannot be initiated. This usually occurs when the measured glow peak shapes differ appreciably from the shapes that are consistent with eqns. (4) or (6). Or, 3) The convergence process is continuing but has not converged when an arbitrary number of parameter adjustment cycles, usually 50 or 100, have been completed. Usually, in this case, additional cycles do not alter significantly the kinetic parameter values obtained previously.

The results obtained by fitting the usual first and second order TL expressions to the computed glow curves in Fig. 3 are illustrated, by Fig. 4 and summarized in Tables I and II. The computed best fit to the $n_{01} = 1 \times 10^{14}$ curve in Fig. 3 is shown in Fig. 4. This figure illus-

trates three important points. 1) Both the usual first and second order expressions describe the interactive kinetic glow curves very well over the upper 60 percent or more of each peak. However, neither expression simultaneously describes both wings of the peaks. 2) As a consequence of 1) it is apparent that TL kinetic analyses based on peak temperature, FWHM, etc., will appear to provide meaningful kinetic parameters in many cases. 3) The kinetic parameters obtained from the best fit procedures differ, by varying amounts, from those used to compute the glow curves. These conclusions apply, to varying degrees, to most of the computed "data" curves in Fig. 3.

The data summarized in Tables I and II provide additional significant conclusions. 1) The higher intensity peaks appear to resemble the usual curves more than the lower intensity curves. This is demonstrated by the fact that the high intensity curves are readily fitted to the kinetic expressions whereas the low intensity curves are not. 2) The E_i and s_i values depend on the initial trapped charge concentrations. The highest temperature peak depends very strongly on the n_{0i} values. 3) Physically unrealistic E_i and s_i values are obtained, particularly for the highest temperature peak. 4) As the n_{0i} values increase the curves more closely resemble the first order kinetic curves.

SUMMARY

In an attempt to explain various anomalous TL results, the basic one trap TL equations were studied for cases in which the usual approximations did not apply. Some of the anomalous results appear to correspond to these cases. Consequently, TL systems were studied that are not

subject to the seldom mentioned assumption that applies to the usual one-trap kinetics. Namely, in systems with more than one type of trap it is implicitly assumed that charges released from one type of trap are not retrapped on other types of traps. For this purpose, the basic one trap TL equation has been generalized to include trapping between different types of traps. Called interactive TL kinetics, the properties of such systems have been studied by regarding computed glow curves as "data" and analyzing each glow peak by fitting it to the usual first and second order kinetic equations. The major results are: 1) The relative peak heights, shape, peak temperature and other properties depend strongly on the pre-measurement trapped charge levels, i.e. on the dose. 2) Most glow peaks, particularly at high trapped charge levels, are well fitted by the usual expressions over their upper 60 percent or more; but not in the wings. 3) The kinetic parameters, E_1 and s_1 , depend strongly on the pre-measurement dose and often differ, appreciably, from the values used to compute the "data" curves.

Thus, interactive kinetic glow curves reproduce many of the anomalous features of recent TL measurements. In particular, they give rise to glow curves that depend on the pre-measurement dose and glow peak shapes not described by the usual kinetics.

6. Acknowledgement

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Table 1

Kinetic parameters obtained from a computerized procedure for determining the E_i and s_i values that provide a best fit of the usual first order kinetic expression, eqn. (4) in the text, to the glow curves computed from interactive TL kinetics shown in Fig. 3.

		E_1	E_2	E_3	s_1	s_2	s_3
Assumed Values		1.00	1.25	1.50	10^{10}	10^{10}	10^{10}
Initial Trapped Charge, n_{oi}	Fraction of Traps Initially Filled, $n_{o1}/10^{16}$						
5×10^{15}	5×10^{-1}	0.984	1.225	1.311	6.1×10^9	5.4×10^9	2.4×10^8
1×10^{15}	10^{-1}	0.978	1.236	1.158	4.6×10^9	6.0×10^9	8.9×10^6
5×10^{14}	5×10^{-2}	0.981	1.259	1.106	4.7×10^9	9.3×10^9	2.6×10^6
10^{14}	10^{-2}	0.989	1.361	1.032	5.2×10^9	8.2×10^{10}	3.2×10^5
5×10^{13}	5×10^{-3}	0.990	1.423	1.018	5.2×10^9	3.0×10^{11}	1.7×10^5
10^{13}	10^{-3}	<u>0.985</u>	0.789	1.085	6.5×10^9	3.2×10^2	2.0×10^5
5×10^{12}	5×10^{-4}	(0.972)	<u>1.100</u>	1.025	(2.9×10^9)	<u>1.8×10^8</u>	4.1×10^4
10^{12}	10^{-4}	(1.002)	(1.726)	(0.940)	(6.9×10^9)	(1.4×10^9)	(2.5×10^3)
5×10^{-11}	5×10^{-5}	(0.968)	(1.841)	(0.928)	(1.2×10^9)	(3.6×10^9)	(1.4×10^3)
10^{-11}	10^{-5}	(0.994)	(1.590)	(0.925)	(3.2×10^9)	(1.3×10^7)	(4.2×10^2)
5×10^{-10}	5×10^{-6}	(0.990)	(1.479)	(0.940)	(2.2×10^9)	(1.3×10^6)	(2.9×10^2)
10^{-10}	10^{-6}	<u>1.046</u>	<u>1.170</u>	<u>1.030</u>	<u>2.1×10^{10}</u>	<u>8.0×10^8</u>	<u>7.2×10^2</u>

X.XXX = Best fit parameters

(X.XXX) = Best fit parameters, convergence not complete

X.XXX = Fitting procedure does not converge; parameters obtained from peak temperature and FWHM.

Table 2

Kinetic parameters obtained from a computerized procedure for determining the E_i and s_i^* values that provide a best fit of the usual second order kinetic expression, eqn. (6) in the text, to the glow curves computed from interactive TL kinetics shown in Fig. 3.

		E_1	E_2	E_3	$s_1^*(a)$	s_2^*	s_3^*
Assumed Values		1.00	1.25	1.50	10^{10}	10^{10}	10^{10}
Initial Trapped Charge, n_{oi}	Fraction of Traps Initially Filled, $n_{01}/10^{16}$						
5×10^{15}	5×10^{-1}	1.432	1.778	2.125	2.7×10^{15}	2.1×10^{15}	1.8×10^{15}
10^{15}	10^{-1}	1.413	1.754	1.850	1.3×10^{15}	9.4×10^{14}	5.7×10^{12}
5×10^{14}	5×10^{-2}	1.410	1.758	1.745	1.1×10^{15}	9.0×10^{14}	5.7×10^{11}
10^{14}	10^{-2}	1.399	1.808	1.576	6.6×10^{14}	2.1×10^{15}	8.7×10^9
5×10^{13}	5×10^{-3}	1.393	1.834	1.540	5.3×10^{14}	3.5×10^{15}	2.5×10^9
10^{13}	10^{-3}	1.383	1.876	1.505	3.9×10^{14}	8.2×10^{15}	3.1×10^8
5×10^{12}	5×10^{-4}	1.381	1.885	1.501	3.6×10^{14}	9.9×10^{15}	1.5×10^8
10^{12}	10^{-4}	1.378	1.896	1.497	3.4×10^{14}	1.3×10^{16}	2.8×10^7
5×10^{11}	5×10^{-5}	(1.251)	(1.814)	(1.497)	(5.8×10^{11})	(1.9×10^{15})	(1.4×10^7)
10^{11}	10^{-5}	<u>1.489</u>	<u>1.673</u>	<u>1.552</u>	<u>5.1×10^{15}</u>	<u>6.1×10^{13}</u>	<u>5.6×10^6</u>
5×10^{10}	5×10^{-6}	<u>1.489</u>	<u>1.667</u>	<u>1.524</u>	<u>5.1×10^{15}</u>	<u>6.7×10^{13}</u>	<u>2.1×10^6</u>
10^{10}	10^{-6}	<u>1.498</u>	<u>1.678</u>	<u>1.502</u>	<u>5.7×10^{15}</u>	<u>6.5×10^{13}</u>	<u>3.2×10^5</u>

^a Since $s_i^* = (n_{oi}/N_i)s_i$ the s_i^* from non-interacting kinetics are always $< 10^{10}$

X.XXX = Best fit parameters

(X.XXX) = Best fit parameters, convergence not complete

X.XXX = Fitting procedure does not converge; parameters obtained from peak temperature and FWHM.

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Figure Captions

- Fig. 1. The usual first and second order TL kinetics glow peaks, computed from eqns. (4) and (6) in the text, are shown by points. The glow curves computed from interactive TL kinetics, eqns. (7), (8) and (9) in the text, are shown by solid lines. This figure demonstrates that the glow curve properties of TL systems described by interactive kinetics depend markedly on re-trapping-recombination cross section ratios.
- Fig. 2. Glow curves computed from interactive TL kinetics showing that the individual glow peak properties reflect the initial trapped charge concentrations.
- Fig. 3. Interactive TL kinetic glow curves computed for the specified initial trapped charge concentrations. The peak temperatures, shapes, relative intensities and other properties are strong functions of the initial trapped charge concentrations. The kinetic parameters obtained by fitting these curves with the usual first and second order TL kinetic expressions are given in Tables 1 and 2.
- Fig. 4. The $n_{01} = n_{02} = n_{03} = 10^{14}$ curve in Fig. 3, shown as individual "data points", fitted to the usual first and second order TL kinetic expressions. Both the first order (solid lines) and second order (dashed lines) kinetics fit the computed glow peak quite well over their upper 60 percent or more. However, neither expression adequately describes the glow peak "wings."

FIG. 1

FIG. 2

FIG. 3

FIG. 4