

EFFECT OF VARIOUS KINETICS PARAMETERS ON THERMOLUMINESCENCE PEAKS AND ELECTRON CONCENTRATION IN TRAPS

By

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Abstract

In this thesis, before we were going to study these effects on thermoluminesce peaks, the phenomenon of both luminescence and thermoluminesce as well as the types of luminescence were explained. we study theatrical and computational methods to investigate the the effects of various kinetics parameters on thermoluminescence peaks and electron concentration in traps using Randall and Wilkins model. The equation of first order kinetics which used for plotting the graph was derived. We implies the methodology and materials used to achieve this work. We find that the effect of heating rate, activation energy, frequency factors and electron concentration on both thermoluminescence peaks and electron concentration in traps were already discussed. As heating rate increase the glow peak shift to higher temperature when heating rate increased and the other parameters also stated under this unit (unit four). The conclusion was explained at the end of this unit.

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Chapter 1 Introduction

1.1 Background of the Study

The phenomenon of thermoluminescence (TL) of minerals was known empirically as early as 1663. It was in this year that Sir Robert Boyle reported to the Royal Society about "Experiments and Considerations upon Colours with Observations on Diamond that Shines in the Dark" (1663) and described how, upon warming a diamond in contact with his body in the dark, he saw a flash. This reflects the definition of thermoluminescence. Not only diamonds but a large number of minerals emit light energy upon warming. Well known examples are quartz, feldspar, calcite and flint. But the phenomenon of thermoluminescence itself was not sufficient to be used as a dating method. It was necessary to establish the mechanisms, which are responsible for resetting the dating clock and link the intensity of the emitted light energy to a time scale. Three centuries later the first law of thermoluminescence was established, which states that the thermoluminescence of minerals is roughly proportional to the irradiation dose to which they had been exposed [1].

Thermoluminescence (TL) is a misnomer in the conventional sense of the names

of luminescence processes like Cathodo-luminescence, Chemi-luminescence, Electroluminescence and Bio-luminescence; heat is not an excitation agent in TL but only a stimulant. The excitation is achieved by any conventional agent like ionizing radiations, UV rays, mechanical vibrations, stress, chemical reactions and so on. Thus Thermoluminescence is the phenomenon of luminescent emission after removal of the excitation under conditions of increasing temperature. Phosphorescence at any temperature after the cessation of the excitation, is nothing but isothermal decay of TL at that temperatures; hence TL should be defined as the thermally stimulated release (in the form of optical radiation) of energy stored in a material by previous excitation. It is also necessary to distinguish clearly between thermoluminescence usually lies in a spectral region where the material is non absorbing; the incandescence emissivity is strongest where absortivity of the material is the maximum.

Thermoluminescence is observed when, in the process of irradiating a material, part of the irradiation energy is used to transfer electrons to traps. This energy, stored in the form of the trapped electrons, is released by raising the temperature of the material, and the released energy is converted to luminescence. This trapping process and the subsequent release of the stored energy find important application in ionizing radiation dosimetry and in the operation of long persistence phosphors. Much information about the trapping process and the release of trapped electrons is obtained from the Thermoluminescence, TL spectrum, in which, after turning off the irradiating source, the thermally stimulated luminescence is monitored under a condition of steadily increasing temperature. The shape and position of the resultant TL glow curves can be analyzed to extract information on the various parameters of the trapping process trap depth, trapping and re trapping rates, etc.

Thermoluminescence (TL) is the thermally stimulated emission of light following the previous absorption of energy from radiation. In TL theory, for first order kinetics, the peak temperature of the glow peaks are expected to change only with the heating rate. The typical glow curve contains one or more glow peaks. Each peak gives information about each trap level and its occupation state, etc in the TL material. The glow peak is analyzed by an empirical method in which a parameter called the order of kinetics is introduced. When the trapped electrons jump up to the conduction band by the thermal energy, they have two kinds of chances to jump down. One is the retrapping process returning to the same kind of traps and another is the recombination with the hole accompanied by the emission of TL light. When the probability of being retrapped is negligible, the glow curve has a narrow peak shape by a rapid recombination process explained by Randall and Wilkins [2].

The heating rate, HR effect on thermoluminescence, TL glow-peaks has been largely discussed by Kitis G. who considers the HR as a dynamic parameter rather than a simple experimental setup variable . His study has been carried out on single, well separated glow peaks, considering the following experimental characteristics; i.e., maximum temperature, T_M full width at half maximum (FWHM), peak intensity and peak integral. The thermoluminescence curve, obtained by plotting the intensity of light emission against temperature for a constant rate of heating, is valuable for finding the distribution of electron traps in a phosphor. Each maximum in the light intensity corresponds to the emptying of a trap whose energy level is a function of the temperature at the maximum. It will be shown in thesis how the trap depth can be calculated using only the positions of the maxima at two different rates of heating [3]. The physical mechanisms governing the trapping and release of electrons are discussed in detail in thesis by [4]. The First-order kinetics equations describing these physical processes at a single type of trap can be solved, under various assumptions, and analytical expressions are obtained for the shape of the resulting glow curve [5].

1.2 Statement of the problem

In this thesis, we want to know the effect of heating rate, activation energy and frequency factor on TL glow-curves and electron concentration in traps. The TL glow curve represents the intensity of emitted light versus temperature; each glow peak is associated with recombination center and related to specific trap. Activation energy and escape frequency factor can be calculated from the glow curve. Various methods can be used to calculate trap parameters based on the kinetics order of glow peaks such as initial rise method and variable heating rates. But in this thesis the effect of various kinetics parameters on TL glow-curves and electron concentration in traps is studied by depending only on first-order kinetics equations in this work.

Therefore, this work focuses on the effect of various kinetics parameters on thermoluminescence glow peaks and electron concentration in traps using Randall Wilkins model with first-order kinetics.

1.3 Basic Research Question

What is the effect of heating rate on TL glow-curves and electron concentration in traps?

What is the effect of frequency factor on thermoluminescence glow curves and electron concentration in traps ? What is the effect of trap depth (activation energy on TL glow-curves and the maximum temperature peak?

1.4 Objectives of the Study

1.4.1 General Objectives

The general objectives of this thesis is to study the effect of various TL kinetics parameters on Thermoluminescence glow curves and electron concentration in traps ...

1.4.2 Specific Objectives

To identify the effect of heating rate on thermoluminescence glow curves and electron concentration in traps.

To describe the proportionality of heating rate with electron concentration in trap.

To analysis the effect of frequency factor on thermoluminescence glow curves and electron concentration in traps.

To examine the effect of activation energy on thermoluminescence glow curves and electron concentration in traps.

To evaluate the relationship between electron concentration in trap and glow-curve peak area by considering initial and final temperatures.

1.5 Significance of the Study

Heating rate has always been an important parameter for the investigation of various kinetic parameters of glow curves in thermoluminescence phenomena. In addition, it plays a critical role in deciding the time required to record the TL glow curves because thousands of dosimeters have to be processed in a short time when personnel monitoring is carried out using TL dosimeters. This is also important because higher heating rate not only increases the glow peak height but also records the glow curves faster, which forms the basis of TL dosimetry in large scale personnel monitoring

1.6 Scope of Study

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The researcher focused on the effects of various TL kinetic parameters on TL peaks and electron concentration in traps with first order-kinetics with using the Randall and Wilkins model .

1.7 Outlines of the Thesis

This thesis is organized into five chapters. In chapter 1, we present some background concepts of thermoluminescence and objectives and significance of the study. In Chapter 2, we review the theoretical concepts of luminescence, in particular the frequency factor in relation to the first order kinetics model calculation. In chapter 3, the material and method used for the accomplishing of this work were explained. In Chapter 4, we explain the effect of various kinetics parameters on the intensity of thermoluminescence and electron concentration in traps by plotting the Figures using the Randall and Wilkins model (first order-kinetics) and finally in Chapter 5, we draw some conclusions.

Chapter 2 Literature Review

What is Luminescence?

The word luminescence is derived from the Latin word for light, 'lumen', and the Latin, 'escentia', meaning the process of and hence is the process of giving off light. Luminescence is any emission of light (electromagnetic waves) from a substance that does not arise from heating. This definition makes luminescence distinct from incandescence which is light emission due to the elevated temperature of a substance, such as a glowing hot ember. There are many types of luminescence which can be classified by the energy source which initiates the luminescence process.

2.1 Major Types Luminescence

They are seven major type of luminescence. These are; Photoluminescence (absorption of photons), Cathodo-luminescence (bombardment by electrons), Chemiluminescence (initiated by chemical reactions), Electroluminescence (applied electric field), Mechano-luminescence (through mechanical action), Radioluminescence (bombardment by ionizing radiation), Thermoluminescence (activated by heating). However, from these types of luminescence Thermoluminescence and Photoluminescence are the most related with our work and we consider them with their types.

2.2 Luminescence Phenomena

In Latin 'lumen' means light, the term 'luminescent' refer to materials such as crystals, minerals, and chemical substances that can emit light, especially in the visible range . Luminescence is the emission of light from certain solids called phosphors. This emission, which does not include black body radiation, is the release of energy stored within the solid through certain types of prior excitation of the electronic system of the solid. This ability to store is important in luminescence dosimetry and is generally associated with the presence of activators.

Luminescence [5-7] is the energy emitted by a material as light, after absorption of the energy from an exciting source which provokes the rise of an electron from its ground energy level to another corresponding to a larger energy (excited level). The light emitted, when the electron comes back to its ground energy level, can be classified according to a characteristic life time, τ , between the absorption of the exciting energy and the emission of light.

The luminescent centers are atoms or group of atoms, called activators, positioned in the lattice of the host material and serve as discrete centers for localized absorption of excitation energy [8]. In other words, a luminescent center is a quantum state in the band gap of an insulator which acts as a center of recombination of charge carriers when it captures a carrier and holds it for a period of time until another carrier of opposite sign is also trapped and both combine. The recombination causes the release of the energy in excess as photons or phonon. A trapping state is that for which the probability of thermal excitation from a localized state into the respective delocalized band is greater than the probability of recombination of the trapped charge with a free charge carrier of opposite sign.

2.2.1 Thermoluminescence

The basic process is the storage energy from radiation in trap. The release of this energy by the application of heat electrons flowing from the traps to where they recombine with the holes (positively charged). Recombination produces light with wavelength of the characteristic luminescent center.

2.2.2 Photoluminescence

Photoluminescence is a result of absorption of photons (units of radiant energy) by substances that are fluorescent or phosphorescent.

Photoluminescence is the emission of light from a material following the absorption of light. The word in itself is interesting in that it the combination of the Latin derived word 'luminescence' and the Greek prefix, 'photo-', for light. Any luminescence that is induced by the absorption of photons is called **photoluminescence**. This could equally be light emission from an organic dye molecule in solution , or band-to-band recombination of electrons and holes following photo excitation of a semiconductor.

Phosphorescence

Phosphorescence takes place for a time longer than $10^{-8}s$ and it is also observable after removal of exciting source. The decay time of phosphorescence is dependent on the temperature. Referring to Fig.2.1, one can observe that this situation arises when an electron is excited (e.g. by ionizing radiation) from a ground state E_o to a meta-stable state E_m (electron trap), from which it does not return to the ground level with emission of a photon (e.g. the transition from E_m to E_o), because it is completely or partially forbidden by the selection rules.

If one supposes that a higher excited level, E_e , exists to which the system can be raised



Figure 2.1: Phosphorescence phenomenon.

by absorption of the energy $E_e - E_m$, and that the radiative transition $E_e - E_m$ is allowed, then one can provide the energy $E_e - E_m$ by thermal means at room temperature. After that a continuing luminescence emission (phosphorescence) can be observed even after the excitation source is removed. This emission will continue with diminishing intensity until there are no longer any charges in the meta stable state. For a short delay time, let us say less than $10^{-4}s$, it is difficult to distinguish between fluorescence and phosphorescence. The only way is then to check if the phenomenon is temperature dependent or not. If the system is raised to a higher temperature, the transition from E_m to E_e will occur at an increased rate; consequently the phosphorescence will be brighter and the decay time will be shorter due to the faster depopulation of the meta stable state. The phosphorescence is then called thermoluminescence. The delay between excitation and light emission is now ranging from minutes to about 10^{10} years. The delay observed in phosphorescence corresponds, then, to the time the trapped charge (i.e. an electron) spends in the electron trap. The mean time spent by the electron in the trap, at a given temperature τ , is expressed by

$$\tau = s^{-1} exp(\frac{E}{kT}) \tag{2.2.1}$$

where

s is called frequency factor (sec^{-1}) , E is the energy difference between E_e and E_m , called trap depth (eV) and k is the Boltzmann's constant which is $(8.62 \times 10^{-4} eV K^{-1})$. Once the electron is in the electron trap, it needs an energy E, provided by thermal stimulation, for rising to E_e from E_m and then to fall back to E_o emitting a photon. The term **phosphor** is used to design all solid or liquid luminescent materials. This term is also used, in particular, for thermoluminescent materials (i.e., TL phosphors). A plot of the light intensity as a function of temperature is called **glow-curve**. A glow-curve may have one or more maxima, called **glow-peaks**, each corresponding to an energy level trap [9, 10].

Fluorescence

Fluorescence is a luminescence effect occurring during excitation. Fluorescence, is the emission of photons (units of radiant energy) that are perceived as colors of light. This occurs as a result of high energy photons, such as Ultraviolet radiation (UV light) striking phosphors, which are chemicals that absorb high energy photons and emit lower energy photons (visible light). Materials that fluoresce only do so during the time that the high energy radiant energy is being absorbed. The light is emitted at a time less than $10^{-8}s$ after the absorption of the radiation. This means that fluorescence is a luminescent process that persists only as long as the excitation is continued. The decay time of fluorescence is independent of the temperature: it is determined by the transition probability of the transition from an excited level E_e to the ground state E_o . The process is shown in the Fig.2.2.



Figure 2.2: Fluorescence process.

2.3 Frequency Factor, (s)

The frequency factor, s, is known as the attempt-to-escape frequency and is interpreted as the number of times per second, (v), that an electron interacts with the crystal lattice of a solid, multiplied by a transition probability K, multiplied by a term which accounts for the change in entropy ΔS associated with the transition from a trap to the delocalized band [11]. S may be written as

$$s = vKe^{\frac{\Delta S}{k}} \tag{2.3.1}$$

where

k is for the Boltzmann constant . **s** is for frequency factor v is for an electron interacts with the crystal lattice of a solid **K** is for a transition probability ΔS is for the change in entropy

2.4 Activation Energy

It is the energy, E, expressed in \mathbf{eV} , assigned to a meta-stable state or level within the forbidden band gap between the conduction band (and the valence band (VB) of a crystal. This energy is also called trap depth. The meta-stable level can be an electron trap, near to the CB, or a hole trap, near the VB, or a luminescence center, more or less in the middle of the band gap. The meta-stable levels are originated from defects of the crystal structure. A crystal can contain several kinds of traps and luminescence centers. If **E** is such that E > several kT, where k is the Boltzmann's constant, then the trapped charge can remain in the trap for a long period. For an electron trap, E is measured, in eV, from the trap level to the bottom of the CB. For a hole trap, it is measured from the trap to the top of the VB.

Bombarding the solid with an ionizing radiation is produces free charges which can be trapped at the meta-stable states. Supposing the solid previously excited is heated, a quantity of energy is supplied in the form of thermal energy and the trapped charges can be released from the traps. The rate of such thermally stimulated process is usually expressed by the Arrhenius equation which leads to the concept of the activation energy, E which can be seen as an energy barrier which must be overcome to reach equilibrium. Considering the maximum condition using the first order kinetics:

$$\frac{\beta E}{kT_M^2} = sexp(-\frac{E}{kT_M}) \tag{2.4.1}$$

Where I_M denotes temperature. it is easily observed that T_M increases as E increases. In fact, for $E \gg kT_M$, T_M increase almost linearly with E. This behavior agrees with the Randall-Wilkins model where, for deeper traps, more energy and, in turn, a higher temperature is required to de-trap the electrons [12].

2.5 The Anomalous Heating Rate Effect of Thermoluminescence,

Heating rate has always been an important parameter for the investigation of various kinetic parameters of glow curves in thermoluminescence phenomenon. In addition, it plays a critical role in deciding the time required to record the TL glow curves because thousands of dosimeters have to be processed in a short time when personnel monitoring is carried out using TL glow curves. This is also important because higher heating rate not only decreases the glow peak hight but also records the glow curves faster, which forms the basis of TL dosimetry in large scale personnel monitoring [13]. One of the additional facts a dosimeter must satisfy is that it should not exhibit thermal quenching, which can be verified by investigating the TL response with increase with heating rates. If at a constant dose, the response of selected glow curve is independent of heating rate, then it does not exhibit thermal quenching. One of the additional considerations is that critical in TL dosimetry is the plotting of light output with time or temperature and the relation between time and temperature is decided by the nature of the heating profile. For a given glow curve at a constant dose (assuming no thermal quenching. The light seen by the photomultiplier remains constant and is related to the under the glow curve.

Now let's drive equation for the maximum case, In order to study the dependence of glow peak height on heating rate, any one of the models of TL, namely Randall-Wilkins, Garlick-Gibson or the general order kinetics, can be used. In this study, the general order kinetics model is used. According to the general order kinetics model, the intensity of TL [14]

$$I = -\frac{dn}{dt} = (\frac{n^b}{N^{b-1}})s.exp(-\frac{E}{kT})$$
(2.5.1)

where

n is the number density trapped electrons (m^{-3}) and

N is the total number of empty traps (m^{-3}) .

This also implies that out of N, n are filled and (N-n) are empty. Further b is the order of kinetics and is dependent on the relative probabilities of retrapping and recombination cross-section and radiation dose, E is activation energy (eV), s is the frequency factor or attempt to escape factor (Hz), $T = T_o + \beta t$ is the linear heating profile and T_o the starting temperature, $\beta = dT/dt$ is the heating rate (K/s) and k is the Boltzmann's constant (eV/K) for b=1, Eq.(2.7.1) reduce to Randall-Wilkins model, which represents first order kinetics:

$$\frac{dn}{dt} = -np = -ns.exp(-\frac{E}{kT})$$
(2.5.2)

whereas for b=2, we have the Garlick-Gibson model representing second order kinetics.

$$I = -\frac{dn}{dt} = \frac{n^2}{N}P = \frac{n^2}{N}s.exp(-\frac{E}{kT})$$
(2.5.3)

For the sake of simplicity, the analytical dependence of peak height on heating rate for first order kinetics seen from Eq. (2.7.2) is

$$I = n_o s.exp(-\frac{E}{kT})exp(-\int_{T_o}^T \frac{s.exp(E/kT)}{\beta}dT)$$
(2.5.4)

From the derivative of Eq. (2.7.4), it follows

$$\frac{\beta E}{kT_m^2} = s.exp(-\frac{E}{kT_m}) \tag{2.5.5}$$

By re-arranging we have,

$$\beta = \left(\frac{sk}{E}\right)T_m^2 exp\left(-\frac{E}{kT_m}\right) \tag{2.5.6}$$

where E(ev) is activation energy, s (s^{-1}) frequency factor, k $(eV.K^{-1})$ Boltszmann's constant and

 $T_m(\mathbf{K})$ is the temperature at the maximum.

When β increases, the right hand-side must increase by the same amount. However, since $T_m^2 exp(-E/kT_m)$ is an increasing function of T_m , the increase in its value is implies that T_m must increase. It has been shown in [14] that although in more complex cases of TL kinetics Eq.(2.7.6) does not hold precisely, it can serve in many an approximation. Anyway, the shift of a TL peak to higher temperatures with increasing heating rates seems to be a general property.

A distinction should be made between two alternative presentations of TL. As an example, let us consider first the simplest example of the first-order kinetics. The governing equations In a fact, a proportionality factor is missing between I(T) and -dn/dt which is set here arbitrarily to unity. As is, the unities of I(t) are cm^{-3} whereas the real intensity is given by photon per second or emitted energy per second. When increasing the heating rate, the maximum intensity increases nearly proportionally [15]. The area under the curve must remain the same for all heating rates and

the simple explanation for the increased intensity is that when heating is faster, the peak gets much narrower on the time scale. An alternative, quite common presentation is reached by normalizing the intensity as defined in Eq.(2.7.7) by dividing it by the heating rate β . This magnitude is usually plotted as a function of temperature rather than time. Thus, the area under the curve remains constant with different heating rates [16], and this normalized intensity, which is $I(T) = -dn/dt(cm^{-3}k^{-1})$ has slightly decreasing maximum value associated with a slight broadening of the peak with increasing heating rate. It should be noted that this property of a decrease of the normalized intensity with heating rate is not limited to the first-order case and is seen in many more complicated situations. [14, 17] reported on thermal quenching of TL in quartz which is the decrease in luminescence efficiency with the rise in temperature. In fact, this effects stated in [18, 19].

By using Eq. (2.7.5) in Eq.(2.7.4), the maximum value of TL intensity or glow peak height I_m at peak temperature, T_m , is

$$I_m = \frac{n_o \beta E}{kT_m^2} exp(-\int_{T_o}^{T_m} \frac{s.exp(E/kT)}{\beta} dT)$$
(2.5.7)

Using $T = T_o + \beta t$, $\beta = dT/dn$ and $T_m = T_o + \beta t_m$, Eq.(2.7.8), the value of the maximum TL intensity or peak height at t_m is

$$I_m = \frac{n_o \beta E}{k(T_o + \beta t_m)} exp - \int_{t_o}^{t_f} s.exp[-E/k(T_o + \beta t_m)]dt$$
(2.5.8)

From Eqs. (2.7.8) and (2.7.9), the value of I_m obtained at T_m is the same as it is at t_m . It follows that **glow peak height** increases with increasing of **heating rate**. Further more from Eqs.(2.7.8) or (2.7.9), it can be seen that radiation doses are inversely proportional to heating rate $(n_o \alpha_{\beta}^{1})$, where α is alpha and even then the resulting glow curves will not exhibit the **same peak height** but **decreasing peak** height pattern with the increase of β . This is because the **peak height** does not increase in **direct proportion** with the increase heating rate β as demonstrated by Eq.(2.7.8) [20,21]

2.6 Models of Thermoluminescence

The typical glow curve contains one or more glow peaks. Each peak gives information about each trap level and its occupation state, etc in the TL material. The glow peak is analyzed by an empirical method in which a parameter called the order of kinetics is introduced. When the trapped electrons jump up to the conduction band by the thermal energy, they have two kinds of chances to jump down. One is the retrapping process returning to the same kind of traps and another is the recombination with the hole accompanied by the emission of TL light. When the probability of being retrapped is negligible, the glow curve has a narrow peak shape by a rapid recombination process explained by Randall and Wilkins [22]. Instead, if the retrapping dominates, the recombination with the holes is suppressed and the curve has a wide peak explained by Garlick and Gibson [23]. These two descriptions are called the first order kinetics and the second order kinetics respectively. Now we will clearly explain these three models i.e., Randall-Wilkins model, and Garlick-Gibson model.

Randall and Wilkins Model

We have found the Randall and Wilkins model for the thermoluminescence in different books [24-26] and journals [27-29]. In 1945, Randall and Wilkins extensively used a mathematical representation for each peak in a glow curve, starting from studies on



Figure 2.3: Randall Wilkins Model.

phosphorescence. Their mathematical treatment was based on the energy band model and yields the well-known first order expression. Figure 2.3. shows the simple model used for the theoretical treatment. Between the delocalized bands, conduction band CB, and valence band VB, two localized levels (meta-stable states) are considered, one acting as a trap Tr, and the other acting as a recombination center R. The distance between the trap Tr and the bottom of the CB is called the activation energy or trap depth E. This energy is the energy required to liberate a charge, i.e. an electron, which is trapped in Tr. According to this model, the TL intensity I, at a constant temperature can be written as

$$I(t) = -\frac{dn}{dt} = nSexp(-\frac{E}{kT})$$
(2.6.1)

where $n(cm^{-3})$ is the electron concentration trapped at time t(s), $k(eVK^{-1})$ is the Boltzmann's constant, $S(sec^{-1})$ is the frequency factor and T(K) is the temperature. If the sample is heated up so that the temperature rises at a linear heating rate

$$\beta = \frac{dT}{dt}(sK)then \tag{2.6.2}$$

$$\frac{dn}{n} = -\left(\frac{S}{\beta}\right)exp\left(-\frac{E}{kT}\right)dT \tag{2.6.3}$$

On integration of Eq. (2.8.2), one obtains

$$n(T) = n_o exp[-(S/\beta) \int_{T_o}^T (-\frac{E}{kT'}) dT']$$
(2.6.4)

where $n_0(cm^{-3})$ is the concentration of traps populated at the starting heating temperature $T_0(K)$. The temperature dependence of the emitted TL is given by

$$I(T) = n_o Sexp(-E/kT) \times exp[-\frac{S}{\beta} \int_{T_o}^T exp(-\frac{E}{kT'})dT']$$
(2.6.5)

Under Randall and Wilkins [30] conditions, the total light output is determined by the trapped charged, which is proportional to the absorbed dose and the heating rate influences only the time at which the trapped charge is released but not the total light output. From the various thermoluminescence dosimetry spectra obtained at different heating rates, the average values of integrated counts are obtained between the two specified temperatures covering the main glow curve (peak).

Garlick-Gibson Model

In 1948 Garlick and Gibson, in their studies on phosphorescence, considered the case when a free charge carrier has probability of either being trapped or recombining within a recombination center. The term second order kinetics is used to describe a situation in which retrapping is present.

They assumed that the escaping electron from the trap has equal probability of either

being retrapped or of recombining with hole in a recombination center.

Let us indicate:

N =concentration of traps,

n = electrons in N,

m = concentration of recombination centers,

n = m for charge neutrality condition.

The probability that an electron escapes from the trap and recombine in a recombination center is

$$\frac{m}{(N-n)+m} = \frac{n}{N} \tag{2.6.6}$$

So, the intensity of phosphorescence, I, is given by the rate of decrease of the occupied trap density, resulting in the recombination of the released electrons with hole in the recombination centers:

$$I(t) = -\frac{dn}{dt} = c(\frac{n}{N}).(\frac{n}{\tau}) = c\frac{n^2}{N}s.exp(-\frac{E}{kT})$$
(2.6.7)

where τ is the mean trap lifetime.

Equation (2.8.2) can be rewritten as

$$\frac{dn}{dt} = cn^2 s_1 \cdot exp(-\frac{E}{kT}) \tag{2.6.8}$$

The quantity $s_1 = s/N$ is called pre-exponential factor and it is a constant having dimensions of cm^3Sec^{-1} . Equation (2.8.3) is different from that one obtained in the case of first order kinetics, where the recombination probability is equal to 1, since no retrapping is possible.

From Eq.(2.8.3), by integration with constant temperature T, we obtain:

$$\int_{n_o}^n \frac{dn}{n^2} = s'.exp(-\frac{E}{kT})\int_0^t dt$$

$$\frac{1}{n_o} - \frac{1}{n} = s't.exp(-\frac{E}{kT})$$
$$n = n_o[1 + s'tn_o.exp(-\frac{E}{kT})]^{-1}$$

and then, the intensity I(t) is:

$$I(t) = -\frac{dn}{dt} = n^2 s' . exp(-\frac{E}{kT}) = \frac{n^2 s' exp(-\frac{E}{kT})}{[1 + s' tn_o . exp(-\frac{E}{kT})]^2}$$

which describes the hyperbolic decay of phosphorescence.

Otherwise, the luminescence intensity of an irradiated phosphor under increasing temperature, i.e. thermoluminescence, taking into account that $dt = dT/\beta$, is obtained as follows:

$$\frac{dn}{n^2} = -\frac{s^{'}}{\beta}exp(-\frac{E}{kT})dT$$

therefore

$$\int_{n_o}^n \frac{dn}{n^2} = \frac{s'}{\beta} \int_{T_o}^T exp(-\frac{E}{kT}) dT$$

and then

$$\frac{1}{n_o} - \frac{1}{n} = \frac{s'}{\beta} \int_{T_o}^T exp(-\frac{E}{kT'}) dT'$$
$$n = n_o [1 + \frac{s'n_o}{\beta} \int_{T_o}^T exp(-\frac{E}{kT'}) dT']^{-1}$$

The intensity I(T) is then

$$I(T) = -\frac{dn}{dt} = n^2 s' exp(-\frac{E}{kT}) = \frac{n_o^2 s' exp(-\frac{E}{kT})}{[1 + \frac{s'n_o}{\beta} \int_{T_o}^T exp(-\frac{E}{kT'}) dT']^2}$$
(2.6.9)

Eq.(2.8.5) can be rewritten as

$$I(T) = \frac{n_o^2 Sexp(-\frac{E}{kT})}{N[1 + (\frac{n_o S}{N\beta})\int_{T_o}^T exp(-\frac{E}{kT'})dT']^2}$$
(2.6.10)

2.7 Calculation of Electron Trap Depths from Thermoluminescence Maxima

Randall and Wilkins [30] derived the equation for the TL or "glow" curve for a single trap depth, assuming first order kinetics and no re-trapping, in the form

$$I = Cn_o s.exp[-\int^T (s/\beta)exp(-E/kT)dT]exp(-E/kT)$$
(2.7.1)

where I is the brightness, C and n_o are arbitrary constants, E is the trap depth, **s** is an atomic frequency factor, and β is the rate of heating. From this they did not solve for the conditions for the maximum (the glow peak) directly, but wrote

$$E = T_m [1 + f(s,\beta)klog_e s \qquad (2.7.2)$$

where

 T_m is the temperature at the maximum; and showed by plotting a numerical example of equation [2.9.1], using Bünger and Flechsig's [40] values for **s** and **E** in KCI(T_o) phosphor, that $f(s, \beta)$ is small compared with unity when β is in the range 0.5 to 2.5 degrees/sec. This linear relationship becomes, with the same value of $s(2.9Xlo^9sec^{-l})$

$$E = 25kT_m \tag{2.7.3}$$

(for β in the range given)

This last result has been widely quoted as a general rule of thumb; but the numerical constant depends on s, and this will be different for each trap, even in the same substance.

2.8 Application of Thermoluminescence

Different types of irradiation sources can be used to excite the samples in TL measurement such as γ -rays, X-rays, and UV radiation. The dependence of energy absorbed during irradiation, which is termed radiation dose and defined as dose rate time of exposure makes TL very effective in radiation dosimetric applications [34, 35] in medicine, industry, biology and agriculture, and also in dating of ancient pottery sherd and certain rocks [36, 37]. Besides dosimetric applications, thermoluminescence spectroscopy is a versatile tool for defect studies in crystals and trapping phenomena at the grain boundaries in nanostructures and soft materials.

Thermoluminescence dosimeters are devices used to measure the dose of ionizing radiation absorbed by matter or tissue. TL output in a TL crystal is directly proportional to the radiation dose received by crystal and hence provides a mean for estimating the amount of unknown radiation dose . Among the other potential dosimeters LiFbased TL are extensively used due to their tissue equivalence, simple glow-curve, high sensitivity relatively less Fading .

TL dosimetry features has many advantages such as small detector size and close tissue equivalence that make it useful for a variety of application in madecine. In medical imaging the sensitivity of thermolumiescence dosimeters, TLD materials such as LiF:Mg, Cu, P and Al_2O_3 :C enables risk assessment even for low dose procedures. In radiotherapy, the fact no cables are required during the measurement allows the use of TLDs inside tissue equivalent phantoms to verify radiation doses delivered in new treatment techniques. These features make TLD also the most versatile in vivo dosimetry tool allowing dose assessments directly on patients during diagnostic or therapeutic procedures. In addition to these clinical uses, TLD is widely employed for quality assurance in medicine [41].

Chapter 3 Materials and Method

3.1 Materials

The study was purely theoretical. The main source of information were the published articles, books, the published thesis and manuscripts and dissertation carried out based on the project title. Softwares such as mathematica for plotting graph and computers for search information concerning with this work and for writing on, are additional instruments, which was used to accomplish the study.

3.2 Methodology

we study the effect of various kinetics parameters on the thermoluminescence peaks and electron concentration in traps by solving the first-order kinetics equation using Randall- Wilkins model, analytically. The Randall and Wilkins equation was derived for the thermoluminescence or glow curve for a single trap depth, in order to study the dependence of glow peak height on heating rate, by assuming first order kinetics and o re-trapping. We have plotted two general graphs of TL glow peak and Various electron concentration in traps which shows their general behavior that support us for individual expression of each effects of various kinetics parameters on these two point. Then, we described the effects of heating rate, initial Concentration of filled traps, activation energy, and frequency factor on thermoluminescence peaks and electron concentration in traps graphically. The effect of these various kinetics parameters were explained from each plotted graph by making vary each value of various kinetics parameter which selected to know its effect on the TL peak and electron concentration in traps and make the others various kinetics parameters value the same. Finally, we study all these effects by analyzing graph.

Chapter 4 Results and Discussion

4.1 Introduction

The effects of various parameters on TL glow peaks and electron concentration in traps were explained with Eq(2.8.3) and Eq(2.8.4) and Figures in Chapter two. But here, the researcher is classified these effects into two, to make easy consideration of this work. The first one is the effects on TL glow peaks by four parameters were explained by plotting the graphs using Bunger and Flechsig's equation Eq(2.7.3), and the effects of various kinetics parameters on electron concentration in traps were examined with materials like as small amorphus silicon quantum [39]. But, for both of these classified subtopic we used the first order-kinetics equation (i.e, Eq. (2.8.3) and eq.(2.8.4)).

4.2 Effect of various kinetics parameters on thermoluminescence peaks and electron concentration in trap

4.2.1 Effect of various kinetics parameters on Thermoluminescence Peaks

Effect of Heating Rate on thermoluminescence peaks

We have seen from first-order kinetics equation Eq. (2.6.7) that, the maximum glowpeak temperature, T_M , is shifted to higher temperatures as the heating rate increases. Equation(2.9.3) shows us that, when β increases, the right-hand side must increase by the same amount. However, since this is an increasing function of T_m , this implies that T_m must be larger.

When increasing the heating rate, the maximum intensity increases nearly proportionally. This indicate that the heating rate has direct relationship with TL peak.

In other way, we have plotted the graphs which are expressing the effects occurred on TL glow peaks by ; heating rate, electron concentration, frequency factor and energy , using the Eq.(2.8.4) and We have seen under Chapter two that, a numerical example of equation [2.9.1], using Bünger and Flechsig's values for **s** and **E** in KCl (T_o) phosphor, that $f(s,\beta)$ is small compared with unity when β is in the range 0.5 to 2.5 degrees/sec was used us for plotting the graph. This linear relationship becomes, with the same value of $s(2.9Xlo^9sec^{-l})$.

The lower temperature peak decreases with increasing heating rate whereas the higher temperature peak increases with the heating rate. The simple explanation for the increased intensity is that when **heating** is faster, the **peak** gets much narrower



Figure 4.1: TL glow curves for vary heating rate values plotted with Eq.(2.8.4) and the values used for plotting are; E = 0.5 eV, $n_o = 10^{11} m^{-3}$, $s = 2.9 \times 10^{10} sec^{-1}$, and $\beta_1 = 0.5k/s$, $\beta_2 = 1k/s$, and $\beta_3 = 1.5k/s$.

on the time scale. An alternative, quite common presentation is reached by normalizing the intensity as defined in Eq. (2.6.7) by dividing it by the heating rate β ; unfortunately, this is also usually termed in the literature intensity. This magnitude is usually plotted as a function of temperature rather than time. When the linear heating rate β changes, the temperature T_m of the maximum TL intensity of the peak also changes: faster heating rates produce a shift in temperature toward higher values of T_m .

The Effect of the Initial Concentration of Filled Traps on the TL Glow Curves

We used the Randall and Wilkins model (first order) TL curves, for plotting by using three different values of the initial concentration of filled traps. All other parameters are the same. As it can be shown in Figure 4.2 The initial concentration of filled traps



Figure 4.2: TL glow curves for initial concentration of filled traps calculated with the help of Eq.(2.8.4) and the values of parameters are; $n_o = 0.5 \times 10^{10} cm^{-3}$, $n_o = 1.3 \times 10^{10} cm^{-3}$ and $n_o = 1.8 \times 10^{10} cm^{-3}$ and E = 1.0 eV, $k = 8.617 \times 10^{-5} eV K^{-1}$ and $s = 10^9 sec^{-1}$, β) = 0.5eV

stated that the (n_o) affects only the maximum height of the TL glow curve, and no affects on the shape of the 1st order TL glow curve and the temperature of maximum TL intensity unchanged.

The Effect of the Activation Energy on The TL Glow Curves

By using mathematica we can plot the TL glow curves for different activation energies. Here is an example of Randall and Wilkins model (first order) TL curves, calculated using three different values of the activation energies. We get the curves as shown in Figure 4.3. It is clearly observed that the TL peaks depend on the trap depth.



Figure 4.3: TL glow curves for the activation energies calculated with the of Eq(2.8.4) and the values of parameters are; $E_1 = 0.75 eV$, $E_2 = 1.0 eV$, and $E_3 = 1.5 eV$, $s = 10^{11} sec^{-1}$, $k = 8.617 \times 10^{-5} eV K^{-1}$ and $k = 8.617 \times 10^{-5} eV K^{-1}$.

The shallowest trap gives the highest TL peak and the deepest trap gives the lowest TL peak in first order kinetics. As the activation energy E is increased, the TL glow curve shifts towards higher temperatures, but the curve maintains its overall shape for these three models.

The Effect of the Frequency Factor on The thermoluminescence Glow Curves

We used the Randall and Wilkins model (first order) TL curves, and calculate using three different values of the frequency factors. We obtained the TL glow curves as shown in Figure 4.4. The Figure 4.4 told us that as the frequency factor s is increased,



Figure 4.4: thermoluminescence glow curves for the frequency factors according to Eq.(2.8.4) the values of parameters used to plot the the graph are: $s_1 = 10^9 sec^{-1}$, $s_2 = 10^{11} sec^{-1}$, and $s_3 = 10^{13} sec^{-1}$, E = 1.0 eV, $k = 8.617 \times 10^{-5} eV K^{-1}$ and $n_0 = 10^{10} cm^{-3}$

the TL glow curve shifts towards lower temperatures, but the curve maintains its overall shape for these three peaks.

4.2.2 Effect of various thermoluminescence kinetics parameters on electron concentration in traps

All the next plotted graphs were from the effect of retrapping on thermoluminescence intensity peak corresponding to each trap of with any small amorphous silicon quantum. For first order kinetics, where there is no effect of retrapping, the thermoluminescence intensity clearly depends on the level of the trap beneath the edge of the conduction band [35].

The effect of Activation Energy on Electron Concentration in Traps

By using eq.(2.8.3) we can plot the graph which show the effect of activation energy on electron concentration in traps, by using various value of activation energy and make other parameters the same. From this Figure 4.5, We obtained that the third



Figure 4.5: Variation of concentration of electrons in traps for the Activation Energy. according to Eq.(2.8.3)the values of parameters used to plot the the graph are: $(E_1 = 0.5eV)$, $E_3 = 1eV$ and $(E_2 = 0.75$ and $s_3 = 10^{11}sec^{-1}$ $k = 8.617 \times 10^{-5}eVK^{-1}$ and $n_0 = 10^9 cm^{-3}$.

trap with trap level $E_3 = 1eV$ provides the highest TL intensity as compared with the first trap ($E_1 = 0.5eV$) and the second trap ($E_2 = 0.75eV$) at higher temperature because of its high trapped electron concentration. The point is that the majority of the electrons released by the first and the second traps will be ret-rapped by the third one.

The effect of Heating Rate on Electron Concentration in Traps

Now let's consider effect of heating rate on electron concentration in traps by using Eq.(2.8.3). The area under the TL peak is not proportional to the initial occupancy



Figure 4.6: Variation of concentration of electrons in traps for heating rate. according to Eq.(2.8.4)the values of parameters used to plot the the graph are: $(\beta_1 = 0.5K/s)$, $(\beta_2 = 1.0K/s)$, $(\beta_3 = 1.5K/s)$ and $n_0 = 10^{11}(cm^{-3})$. $E = 1.25 \times 10^{10} eV$, $s = 2.65 \times 10^{11} sec^{-1}$) and $k = 8.617 \times 10^{-5} eV K^{-1}$.

of this center because during the heating, more holes are added to recombination

center which also contribute to the emitted light. While the heating rate increases, the peak shifts to higher temperature, and more holes move through the valence band to recombination center.

The effect of Frequency Factor on Electron Concentration in Traps

To explain this effects we used the plotted Figure here by three Randall and Wilkins model (first order) TL curves, calculated using three different values of the frequency factors as shown in the caption. We obtained the curves in Figure 4.7. When we



Figure 4.7: Electron concentration in traps with the different values of frequency factor. according to Eq.(2.8.4)the values of parameters used to plot the the graph are: $s_1 = 10^7 sec^{-1}$, $s_2 = 10^{10} sec^{-1}$ and $s_3 = 10^{11} sec^{-1}$ and E = 1.5 eV, $k = 8.617 \times 10^{-5} eV K^{-1}$ and $n_0 = 10^9 cm^{-3}$.

looked this Figure, as the frequency factor s is increased, the TL glow curve shifts towards lower temperatures, but the curve maintains its overall shape for these three peaks.

Chapter 5 Conclusion

We have studied the effect of the Heating rate, activation energy, frequency factor and the initial concentration of filled traps on thermoluminescence peak and electron concentration in trap by using Arhenius equation (first-order kinetics) using Randall-Wilkins model were studied previous in chapter four. In this we have observed the following results from these graph plotted in this chapter again as follow:

(i) When the linear heating rate β changes, the temperature T_m of the maximum TL intensity of the peak also changes: faster heating rates produce a shift in temperature toward higher values of T_m . (ii) when heating rate increased the TL peak also increase. (iii) The glow peak height decreases with increase in heating rate (iv)As the activation energy E is increased, the TL glow curve shifts towards higher temperatures, but the curve maintains its overall shape for the Randall-Wilkins model. (v) As the frequency factor s is increased, the TL glow curve shifts towards lower temperatures, but the curve maintains its overall shape for the Randall-Wilkins model. (vi) The initial concentration of filled traps (n_o) affects only the maximum height of the TL glow curve, and leaves the shape TL glow curve and the temperature of maximum TL intensity (T_{max}) unchanged in the case of the Randall-Wilkins model.(vii)The initial concentration of filled traps (n_o) affects both the maximum height of the TL glow curve and the temperature of maximum TL intensity (T_{max}) .

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