

# **CHAPTER-1**

## **INTRODUCTION**

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## 1.1 Motivation towards the present work and its background

Potential application of Optically Stimulated Luminescence (OSL) in the field of geological and archeological dating is attracting researcher to study OSL of various minerals and natural quartz. Many researchers have also carried out study of OSL on synthetic quartz to use its potential application in radiation measurement and dating. In OSL, the emission of light takes place from luminescent material which is previously exposed to radiations upon stimulation of light. The outcome of OSL is studied in the form of OSL decay curve (graph of OSL Intensity versus stimulation time). The OSL decay curve is usually an exponential decay curve. Researchers found that this OSL decay curve is not usual exponential decay in every condition. To achieve a standard exponential decay curve, current work is being done in the direction of developing an experimental protocol and investigating the thermal transfer effect on the shape of the OSL decay curve.

## 1.2 Luminescence

It is the phenomenon of emission of light from phosphor which is priorly exposed to ionizing radiation when it is stimulated by an external energy source of heat, light, sound, mechanical, chemical etc. It is not an incandescence; light emission takes place till energy stimulation source exists. In luminescence, emission of light continues for a specific period of time only and stops immediately thereafter, presence or absence of the stimulation source does not matter. Further the emission of light from the sample is occurring which is irradiated by ionizing radiation only for a cycle of stimulation. Luminescence is termed as fluorescence for time lag is  $<10^{-8} \text{ s}^{-1}$  and in case of phosphorescence it is more, where time lag is the time duration between excitation and light emission.

Based on nature of stimulation luminescence are of the following types:

Name of the Luminescence	Nature of stimulation /Stimulation Source
Radio Luminescence	Bombardment of Radiations like alpha, beta particles or gamma waves
Photoluminescence	Emission of light by stimulation of light

Bioluminescence	Biochemical reaction in some insects or sea animals
Triboluminescence	Mechanical Treatment to the sample
Chemiluminescence	Chemical reaction (energy liberated in the form of light)
Thermoluminescence (TL)	Energy provided in the form of heat after irradiation
Optically Stimulated Luminescence (OSL)	Stimulated by light after ionizing radiation

**Table 1.1 Types of Luminescence Phenomenon**

The present study is concentrating on study of OSL decay curve from irradiated synthetic quartz and its correlation with TL.

Thermoluminescence (TL) or Thermally stimulated luminescence (TSL) is emission of light energy upon heating a material with uniform heating rate, which had been previously exposed to ionizing radiation. Radiation like gamma, alpha, beta, X-rays and light rays are used to excite different luminescent materials which can exhibiting thermoluminescence. However, essentially the materials have to be semiconductor or insulator as these materials are having metastable energy states presence in between conduction band and valance band.

Generally, the metastable energy states play a crucial role in the production of TL. The metastable energy levels which may be alternatively known as traps are populated due to pre-excitation of the specimens. De-trapping of these traps needs “stimulation” as mentioned earlier to reduce carrier lifetime as they cannot do it by their own. Therefore, on heating, the pre-excited materials depopulate the trapped charges and recombine with opposite charges to emit light. On the other hand, optically stimulated luminescence arises due to the recombination of optically released charges from populated metastable energy levels within a pre-exposed specimen. Intensity of the OSL is purely related to amount of the radiation dose absorbed hence, during illumination with optical source the SOL signal is observed to decreases to a low level as charges in metastable energy levels are depleted.

Emitted light due to recombination of the de-trapped charges is measured within a spectral region different from that of the exciting photons and usually proportional to the amount of radiation dose absorbed by the material.

### **1.3 Defects in Quartz and its luminescence**

In the Earth's crust, Quartz is known to be one of the most abundant minerals. Alpha Quartz is a crystalline variety of silicon dioxide ( $\text{SiO}_2$ ) which is stable at room temperature. Alpha Quartz is found to presence in the form of sand and rock like quartzite, vein quartz and sand in nature. Crystalline Quartz is technologically very important material. For several industries, Single crystal of quartz is an important raw material. Quartz's piezoelectric property can be used to make resonator to control electronic frequency and also in selecting as filters for electronic circuitry. Nowadays it is widely used for mobile and optical communication devices and projection equipment as an optical product, and an electronic component for personal computers and other digital equipment. As per need felt and focusing on its properties, it is synthesized for electronic industry.

Presence of varying concentration of defects/flaws/imperfections in natural and synthetic quartz can create either positive or negative effects. For e.g. due to presence of lower concentration of aluminum ions, which substitute silicon ions produce an unwanted radiation induced frequency shift in quartz electronic oscillators. Same aluminum defects altogether with other defects become cause for electronic trapping and luminescence recombination that will be useful for dosimetry purposes.

Emission of light in most of the luminescence phenomenon occurs due to the presence of defects that are generally involved in the recombination of charge following de-trapping from these defects of higher energy, irrespective of the original energy absorption. The probability of de-trapping from defects must be considered and the phenomenon of OSL generally follows an exponential decay and is temperature dependent: at a fixed temperature the intensity of the emitted light exponentially decreases <sup>2-3</sup>. The wavelengths of emitted light are very often the same in the various types of luminescence of a certain material. Various other

defects are also to be considered specifically those who are responsible for charge trapping looking at OSL and TL.

#### **1.4 Study of Defects in Quartz**

The luminescence properties of quartz defects have been lay open to a large number of studies with aim to understand the complex mechanisms governing the various luminescence emissions and their modifications as a consequence of irradiation and thermal treatments.<sup>4-5</sup>

Through various spectroscopic techniques, it has been identified that many defects related to both intrinsic and impurity related are known to be present or are produced in quartz. Still, it is under consideration to know the role of these defects in producing luminescence emission. A good number of studies have been carried out on the structure properties of defects in quartz, mainly through ESR measurements,<sup>6</sup> only tentative correlations between defects and luminescence emissions have been put forth. Many defects are known to be present in both crystalline (quartz) and the amorphous (silica) forms of SiO<sub>2</sub>, but the luminescence properties of quartz and silica are largely different.<sup>7</sup> Nevertheless there is an agreement on the role of some impurity defects in quartz, particularly so- called Al (aluminum) centers in being related to the emission of dynamics.<sup>8-9</sup>

Defects related to impurity and some intrinsic defects have been notified as responsible for the luminescence emission in quartz while very few are proposed related to trapping centers in the scientific literature so far. It should be borne in mind that it is often the case that traps and recombination centers are strongly related and some OSL and TL have been suggested to be such.<sup>10-11</sup> Two main types of quartz defects have been reported to be related to its luminescence properties: oxygen-free states, the most common internal defects, and Al centers, the ubiquitous impurity center. In addition to these defects, other impurities such as Ti, Fe, and Ge ions cause many defects, and Si jobs are known to occur in response to oxygen-free sites; they give rise to H<sub>4</sub>O<sub>4</sub> centers where the presence of H<sup>+</sup> ions balance the missing charge of Si<sup>4+</sup>.

Naturally occurring single crystals are generally unsuitable due to twins and other imperfections that degrade its electronic properties. For this reason, natural quartz crystals have been gradually replaced by laboratory-grown synthetic quartz

crystals, as synthetic crystals can be produced without twins or other imperfections and can be grown so that minimum material losses can be obtained during the manufacture of electronic components.

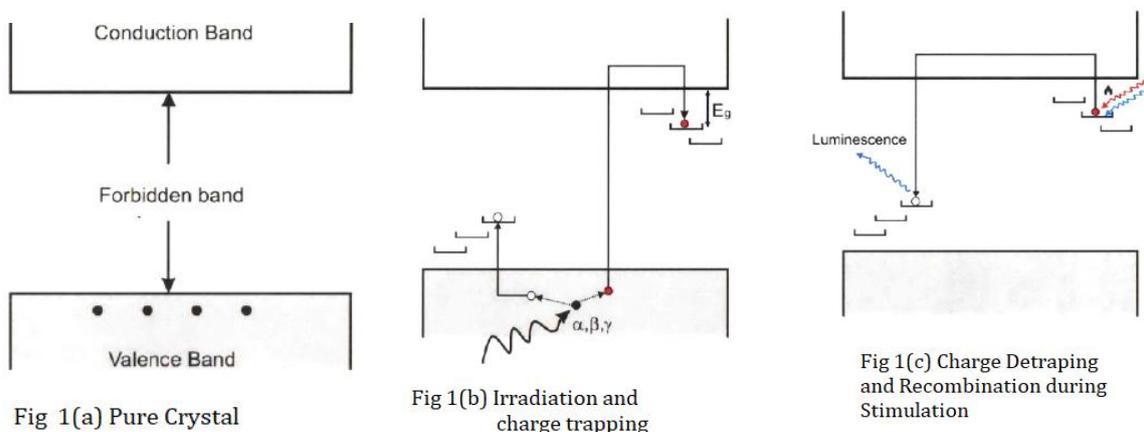
Therefore, it is necessary to identify and characterize point defects in the material. The associated fault dynamics, under different physical conditions, is also important to know, as this information provides an interpretation of the stability of the material in both thermal and radiation backgrounds and thus helps to design the device accordingly.

The literature shows that the thermoluminescence (TL) properties of the material are the most sensitive to the structure. Therefore, TL of natural and synthetic quartz was extensively studied systematically for defect correlation and prior physical treatments.<sup>12-14</sup> Conflicting experimental results have been reported on the same material by various investigators. Mainly they are explained on account of structural changes of minute amount of several metal cations (unavoidably present defect in the material) due to the prior treatments given to the material. The location and concentration of such defects get influenced with different physical conditions such as pre-thermal treatment, irradiation, bleaching and powdering etc.

## 1.5 Thermoluminescence (TL) and Optically Stimulated Luminescence (OSL)

### 1.5.1 Thermoluminescence (TL)

Using solid-state band theory, it is quite easy to understand the luminescence phenomenon (Fig. 1.1 (a)). The valance band is completely filled and the conduction band empty and provides space to free electrons to move at 0 K temperature as shown in the figure 1.1.



**Fig. 1.1 Luminescence phenomenon through band structure**

The forbidden band has no permissible energy levels in pure crystals. Because no crystal is naturally pure and contains defects, some of which provide a local metastable energy level between the conduction band and the valence band, i.e., within the forbidden aperture (Figures 1.1 (b), (c)). During ionization of the crystals, these metastable energy levels act as a trap centre for electrons and holes, called as electron trapping centre and hole trapping centre. During exposure to ionizing radiations of crystals, electrons and holes are created and start moving towards conduction band and valence band respectively. Few of them get trapped in the trapping centers. Depending on the nature of the traps and the charge environment, trapped charges are located in these traps (Garlick and Gibson, 1948; Randall and Wilkins, 1945a; Randall and Wilkins, 1945b).

The expression of lifetime of the electron in a trap is given by

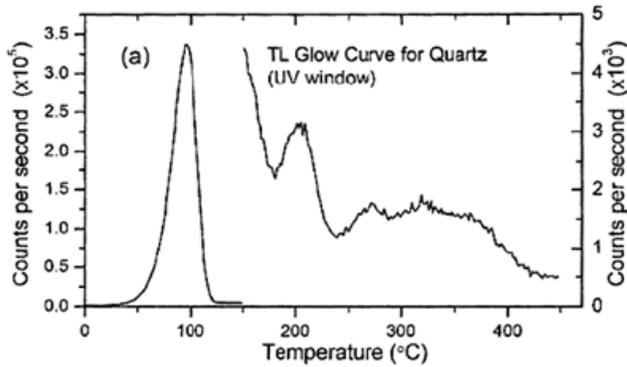
$$\tau = s^{-1} \exp\left(\frac{E_g}{kT}\right) \text{ -----(1)}$$

where  $\tau$  is the life time and  $s$  is the frequency factor which is usually of the order of  $10^{12} \text{ s}^{-1}$ , (Aitken, 1985c)  $E_g$  is called as energy trap depth (Fig. 1.2b),  $k$  is called as Boltzmann constant and  $T$  is the absolute temperature. The lifetime value ranges from seconds to  $10^8$  years and is strongly temperature dependent. When stimulated by heat or light, the charges are evicted from the metastable and some of them radiantly combine with the holes and emit light. Recombination sites are called recombination centers, these centers can be either traps or new traps. Spectral emission characteristics are characteristic of the recombination center. The intensity of the luminescence light is directly proportional to the rate at which electrons are de-trapped during stimulation. (McKeever, 1985; Nambi, 1977)

Expression of the intensity of emitted luminescence light is given by

$$I = C \frac{dn}{dt} \text{ -----(2)}$$

where  $I$  is the intensity of emitted light,  $C$  is the constant of proportionality and  $n$  is the number of electrons present at that instant of time  $t$ . Thermal stimulation (the crystal is heated at a constant heating rate) generates thermo stimulated luminescence and we obtain a TL Glow curve (TL intensity vs. temperature graph, Figure 1.2).



**Fig 1.2 TL Glow curve recorded for Quartz (2 °C/s heating rate)**

In the TL Glow curve, peaks are obtained at different temperatures, these temperatures are related to the energy trap depth of the metastable energy level, and the location of the peak on the temperature axis provides information on the distribution of the trap depth in the crystal (Urbach, 1930). The relationship

between trap depth and TL peak was calculated by Garlick and Gibson (1948); Randall and Wilkins (1945a); Randall and Wilkins, (1945b) based on first-order theory, given that when an electron is detrapped, its recombination produces luminescence. Garlick and Gibson (1948) presented a theory of quadratic kinetics involving the reconnection of disconnected charges.

The equation used for the first order kinetics is

$$I = n_0 \exp\left(-\frac{E_g}{kT}\right) \exp\left[-\left(\frac{s}{\beta}\right) \int_{T_0}^T \left(\frac{E_g}{kT}\right) dT\right] \text{-----(3)}$$

and that for the second order kinetic is

$$I = \frac{n_0^2 s \exp\left(-\frac{E_g}{kT}\right)}{N} \left[1 + \left(\frac{n_0 s}{N \beta} \int_{T_0}^T \left(-\frac{E_g}{kT}\right) dT\right)\right]^2 \text{-----(4)}$$

where I is the intensity,  $n_0$  is the initial concentration,  $E_g$  is the depth, k is the Boltzmann constant, T is the absolute temperature, N is the number of available traps and  $\beta$  is the heating rate. Multiple transitions occur in a two-level system with one trap and one recombination center. The three possible types of transitions are band to band, band to center and center to center transition.

### 1.5.1.1 Literature review on TL of Quartz

Different views and ideas suggested by the several researchers working on Quartz is presented in the following paragraph after reviewing literatures in details. Several TL peaks are found when the irradiated Quartz sample is heated from room temperature to 500°C. Substantial studies have been conducted in the past to investigate the TL properties of different types of quartz.<sup>15-18</sup> Examples of glow

curves from different forms of quartz show large differences in shape. Most of the glow curves reported in the literature for different types of quartz have shown cluster of peaks at temperatures of  $\approx 60^{\circ}\text{C}$ ,  $80^{\circ}\text{C}$ ,  $100\text{--}110^{\circ}\text{C}$ ,  $130^{\circ}\text{C}$ ,  $180^{\circ}\text{C}$ ,  $200\text{--}210^{\circ}\text{C}$ ,  $230^{\circ}\text{C}$ , and  $310^{\circ}\text{C}$ ,  $350^{\circ}\text{C}$ . Although many studies have been performed on the TL properties of quartz, TL peaks were studied particularly well at temperatures of  $100\text{--}110^{\circ}\text{C}$ ,  $200\text{--}210^{\circ}\text{C}$ , and  $350^{\circ}\text{C}$ .<sup>19-20</sup>

#### **1.5.1.2 110°C TL Glow Peak**

110° TL Peak is the one which is found at about  $100^{\circ}\text{C}$  at  $5^{\circ}\text{C} / \text{s}$ . Before TL measurement, natural or synthetic quartz must be irradiated for less than a few hours. This measurement shows the TL Glow peak in this range. The maximum emission of the peak is about 380 nm, which corresponds to the center of the  $\text{H}_3\text{O}_4$  hole in the quartz.

Electron traps corresponding to  $110^{\circ}\text{C}$  peak can be populated by irradiation or by phot transfer. For example, a photo transferred is observed, when green light exposure at room temperature and heating is given to a quartz with a strong peak of  $325^{\circ}\text{C}$  TL peak. Zimmermann explained that the peak response of TL  $110^{\circ}\text{C}$  at a small test dose could be significantly improved at temperatures above  $200^{\circ}\text{C}$ .

#### **1.5.1.3 325°C TL Glow Peak**

The TL peak at  $325^{\circ}\text{C}$  has an emission peak at about 380 nm. The luminescence center used by the electrons, which resulted in a peak at  $325^{\circ}\text{C}$ , had previously been identified from the quartz extracted from the ceramics as a heat treatment. This suggests an increased probability of non-radiative de-excitation in the center during the recombination process.

Even at wavelength as long as 500 nm, the  $325^{\circ}\text{C}$  TL peak is very rapidly bleached. The natural  $325^{\circ}\text{C}$  signal is bleached very quickly when exposed to  $500 \pm 20$  nm light for only 1 minute, when even 1200 minutes of exposure does not cause a decrease in the  $375^{\circ}\text{C}$  peak. The use of higher energy photons bleaches the  $325^{\circ}\text{C}$  peak even faster.

#### **1.5.1.4 375°C TL peak**

It is very slowly bleachable peak. In Australia, this peak has been used widely for dating coastal dune sediment. Scholefield et al., had done spectral studies and observed that 375°C TL peak emission has a maximum at about 480nm in case of Australian sedimentary quartz. McKeever et al, suggested that the emission is attributed to be  $AlO_4$  which act as a hole trap center in the lattice of quartz. Franklin et al., 1992 used green filter (Chance OGr1) in combination with HA3 heat absorption filter to study the behavior of slowly bleachable peak. Green light could be used to remove contribution from 325°C rapidly bleaching peak if any, i.e. using the Chance OGr1 or Schott GG-475 filter in front of broad emission light source. Homyak et al., have reported more complex behavior, which might limit the use of the 375°C TL peak for dating, for quartz from the Kalahari Desert. The TL was observed with either an ultraviolet filter Schott UG-11 centered at 380nm or a green filter OGr1 centered at 480nm to remove 325°C TL peak using green light optical bleaching.

#### **1.5.1.5 Other TL peaks**

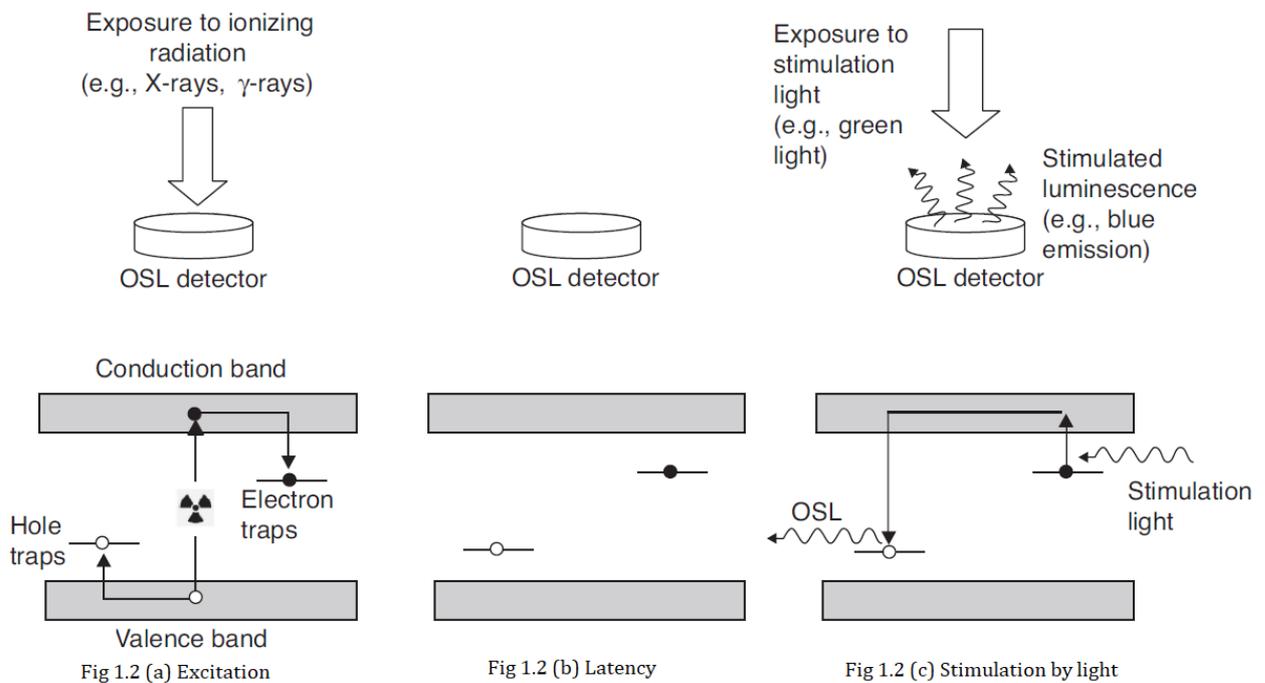
Examination of the glow curve of laboratory irradiated sedimentary quartz shows that there are several other peaks; for example, peaks at 160°C and 280°C the former also being rapidly bleached by exposure to light. Franklin et al, reported that emission wavelengths of 392nm and 410nm are for rapidly bleaching peaks at 150°C-180°C and at 200°C-220°C in the study of emission of wavelength from Australian quartz. From the spectral plots it is concluded that the peaks at 110°C, 180°C, 220°C and 325°C all use the same luminescence center, and its emission peak shifts to higher wavelengths, as the temperature of the sample is increased.

#### **1.5.2 Optically Stimulated Luminescence (OSL)**

Luminescence observed called as optically stimulated luminescence (OSL) upon illumination of luminescent materials that were previous exposed to ionizing radiation illuminated. During excitation by ionizing radiation, electrons and holes separately trapped (metastable states) at defects in the crystal lattice. Upon stimulation by light, release of these electrons and holes from these trapping

centers, resulting in electron/hole recombination and excitation of luminescence centers in the crystal take place. Photons are emitted when these excited luminescence centers decay to the ground state.

Figure 1.2 illustrates the process of OSL described above. In the first stage (Figure 1.2 (a)), luminescent material is exposed to ionizing radiation which results in excitations and ionizations: electrons are promoted to the conduction band, where they can move freely throughout the crystal, leaving behind a hole, which can also move freely in the valence band. These free electrons and holes may get trapped at defects in the crystal lattice, the energy levels for which are represented by the short horizontal lines in the forbidden gap. Second stage is a latency period characterized by concentration of trapped electrons and holes at metastable state (Figure 1.2 (b))

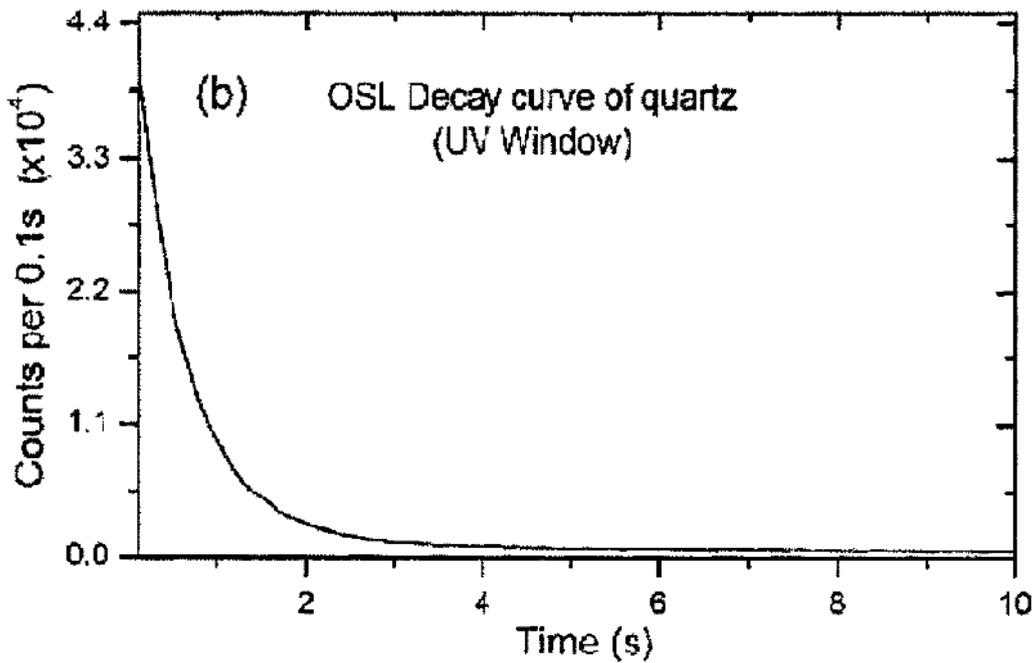


**Fig 1.3 Illustration of OSL Process**

(Eduardo G. Yuhikara and Stephen W. S. McKeever *Optically Stimulated Luminescence: Fundamentals and Applications*, 2011, Ch.2, p.14)

If the trapping centers are sufficiently deep in terms of energy, then the thermally induced escape probability of the trapped charges is negligible at room temperature. This relatively stable concentration of trapped electrons and holes is related to the energy absorbed by materials during the excitation process. As shown in the Figure 1.2 (c), a photon of wavelength  $\lambda$  is used to stimulate the electron to the conduction band. Once electron reach in the conduction band, it is free to move through the

crystal and may reach the trapped hole called recombination process which relaxes an electron to the ground state by the emission of a photon of wavelength  $\lambda_{OSL}$ . With this simple model, it can be proved that OSL intensity decays exponentially during stimulation which is called as OSL Decay curve (A graphical Representation of OSL Intensity versus stimulation time, Fig 1.3).



**Fig1.4 OSL Decay curve recorded for Quartz**

For example, let  $n$  be the concentration of trapped electrons and  $p$  the transition probability per unit time for the trapped electron to escape to the conduction band under optical stimulation. Assuming no possibility of re-trapping,  $n$  will change according to

$$\frac{dn}{dt} = -np \text{ ----- (5)}$$

Solution of this equation shows that the concentration of trapped electrons decays exponentially with the stimulation time:

$$n(t) = n_0 e^{-pt} \text{ ----- (6)}$$

where  $n_0$  is the initial concentration of trapped charges. The intensity of light emitted is proportional to the rate of electrons escaping the trapping centers  $dn/dt$  if all stimulated electrons recombine immediately after stimulation.

$$I_{OSL} \propto \frac{dn}{dt} \text{ -----(7)}$$

$$I_{OSL} = pn_0 e^{-pt} \text{ -----(8)}$$

In the OSL process, the emitted photon to have energy higher than the photon involved in the stimulation (i.e.,  $\lambda_{\text{OSL}} < \lambda_{\text{stim}}$ ) without violation of energy conservation. The energy of the photon emitted is determined by the nature of the transition between the excited and ground states of the luminescence center, and does not depend on the wavelength of stimulation. Part of the energy emitted as OSL originates from the energy absorbed by the material during exposure to ionizing radiation, stored in the form of electrons and holes separately trapped within the crystal.

OSL is a very popular method in the field of dosimetry and dating. Huntley et al.,<sup>21</sup> introduced first time that the technique of OSL can be used for dating. Since then the technique is being used as an important dating technique. Continuous light wave was used for optical excitation which is emitted out from either a high-power arc lamp source, along with filter system or a monochromator to select the excitation wavelength, or a laser operating at or near the desired wavelength. In order to differentiate between the excitation light and the emission light, and to prevent scattering of excitation light from entering the detector, narrow band and/or cut-off filters are used during the recording of OSL counts from materials.

In general, the shape of an OSL decay curve cannot be analyzed using simple description. Shape of OSL decay is found to depend on the type of sample, radiation dose absorbed by the sample and intensity of illuminating light and the temperature. Smith and Rhodes<sup>22</sup> and Bailey et al.,<sup>23</sup> attempted to fit the decay of the green light stimulated OSL from quartz using a summation of three exponentials. One might expect a multi-component OSL decay curve if more than one trap is being emptied at the illumination wavelength being used. Smith and Rhodes also note an increase in the OSL decay rate as the temperature increases. Spooner<sup>24</sup> who presented evidence for a thermally assisted process for OSL production studied the thermal dependence of the OSL from quartz further.

A long tail observed in a OSL decay curve may be due to the influence of shallow trap or a contribution from traps, which are emptying slowly at the excitation wavelength. Since at higher temperatures the shallow traps become less effective, it yield an optically stimulated phosphorescence component to the OSL decay, and they give rise to a temperature dependent OSL component.

### 1.5.2.1 Simplest Model: one trap/one center

Most OSL model assume that in order to reach the recombination site the optically excited charge transport through the delocalized bands. Fig-1.4(a) is simplest a model to understand OSL phenomenon. Trapped electrons (concentration  $n$ ) evicted into the conduction band at rate  $f$ , and get recombine with trapped holes (concentration  $m$ ) to produce OSL of intensity  $I_{OSL}$  upon stimulation with light. With the usual definitions, the rate equation describing the charge flow is

$$\frac{dn_c}{dt} = -\frac{dn}{dt} + \frac{dm}{dt} \text{-----(9)}$$

which can be derived from the charge neutrality condition  $n_c + n = m$  ----- (10)

With the assumptions of quasi-equilibrium ( $\frac{dn_c}{dt} \ll \frac{dn}{dt}, \frac{dm}{dt}$  and  $n_c \ll n, m$ ) and negligible re-trapping we have

$$I_{OSL} = -\frac{dm}{dt} = -\frac{dn}{dt} = nf \text{----- (11)}$$

The solution of which is  $I_{OSL} = n_0 f \exp(-tf) + I_0 \exp(-t/\tau)$  -----(12)

where  $n_0$ , is the initial concentraton of trapped electrons at timet  $t=0$ ,  $I_0$  is the initial luminescence intensity at  $t=0$ , and  $\tau = \frac{1}{f}$  is the decay constant. It is a straight forward relationship in which the initial intensity is directly proportional to the excitation rate and the decay of the OSL with time is a simple exponential. The excitation rate,  $f$  is given by the product of the excitation intensity  $\phi$  and photo ionization cross section  $\sigma$  ( $f = \phi\sigma$ ).

### 1.5.2.2 Competing, deep trap

For two optically active traps (concentration  $n_1$  and  $n_2$  and excitation rates  $f_1$  and  $f_2$  the same assumption yield  $\frac{dm}{dt} = -\frac{dn_1}{dt} - \frac{dn_2}{dt}$  ----- (13)

Along with the charge nutrality conditon of  $n_1 + n_2 = m$ ----- (14)

We have  $n_1 = n_{10} \exp(-tf_1)$  and  $n_2 = n_{20} \exp(-tf_2)$  ----- (15)

Thus  $I_{OSL} = n_{10} f_1 \exp(-tf_1) + n_{20} f_2 \exp(-tf_2)$  ----- (16)

$I_{OSL} = I_{10} \exp\left(\frac{-t}{\tau_1}\right) + I_{20} \exp\left(\frac{-t}{\tau_2}\right)$  ----- (17) and OSL decay curve is the sum of two exponentials.

Alternatively, if the additional trap is optically and thermally inactive, i.e. it acts as a deep, competing trap only, Fig 1.4 (b), the OSL intensity may be written as

$$I_{OSL} = n_{10} f \exp(-tf) - \frac{dn_2}{dt} \text{-----} (18)$$

where  $\frac{dn_2}{dt} = n_c(N_2 - n_2)A_2 \text{-----} (19)$

Adopting the standard definitions. With an additional assumption of  $N_2 \gg n_2$  then  $n_c N_2 A_2 \approx \text{constant } C$  Therefore,  $I_{OSL} = n_{10} f \exp(-tf) - C \text{-----} (20)$

And the OSL is reduced in intensity by the extent of the re trapping into the deep trap. In the limit, as  $t \rightarrow \infty, n_c \rightarrow 0$  Thus,  $C$  is in fact very slowly varying function of time.

### 1.5.2.3 Competing shallow trap

If the competing traps are shallow traps, which are thermally metastable at the temperature of the OSL measurement Fig 1.4 (c), equation (19) becomes

$$\frac{dn}{dt} = n_c(N_2 - n_2)A_2 - n_2 p \text{-----} (21) \text{ , where } p \text{ is the rate of thermal excitation}$$

out of the trap. Now we have

$$I_{OSL} = n_{10} f \exp(-tf) + n_2 p - n_c(N_2 - n_2)A_2 \text{-----} (22)$$

The last two terms in (22) combine to produce a long-lived, temperature dependent tail to the OSL decay. The form of this component will be an initial increase, followed by a decrease at longer times. Depending upon the relative size of this component compared with first term, the overall OSL decay curve may exhibit an initial increase followed by a decrease the relative size of the two components also depends upon the excitation rate/ such that at low values of the temperature dependent term may be significant.

### 1.5.2.4 Competing recombination center

For the case of two recombination centers Fig 1.4 (d), one of which  $m_1$  is radiative and the other  $m_2$  is non radiative, we have  $n = m_1 + m_2$ ,

$$n = n_0 \exp(-tf) \text{ , and } I_{OSL} = n_0 f \exp(-tf) - \frac{dm_2}{dt} \text{-----} (23)$$

As with the case of the deep traps the OSL intensity is reduced by the existence of non-radiative pathway. Since,

$$\frac{dm_1}{dt} = -n_c m_1 A_{m1} \text{-----} (24)$$

and

$$\frac{dm_2}{dt} = -n_c m_2 A_{m2} \text{-----} (25)$$

then, with quasi-equilibrium ( $\frac{dn_c}{dt} \approx 0$ ) we have

$$m_1 \approx m_{10} \exp(-tn_c A_{m1}) \text{-----} (26)$$

$$m_2 \approx m_{20} \exp(-tn_c A_{m2}) \text{-----} (27) \text{ from which we have}$$

$$\frac{m_1}{m_2} \approx \frac{m_{10}}{m_{20}} \exp(-tn_c [A_{m1} - A_{m2}]) \text{-----} (28) \text{ from which we see that the ratio } \frac{m_1}{m_2} \text{ is a}$$

time dependent function .

However, if  $A_{m1} = A_{m2}$ , then  $\frac{m_1}{m_2} = a$  constant, k. In these circumstances the charge neutrality condition is written

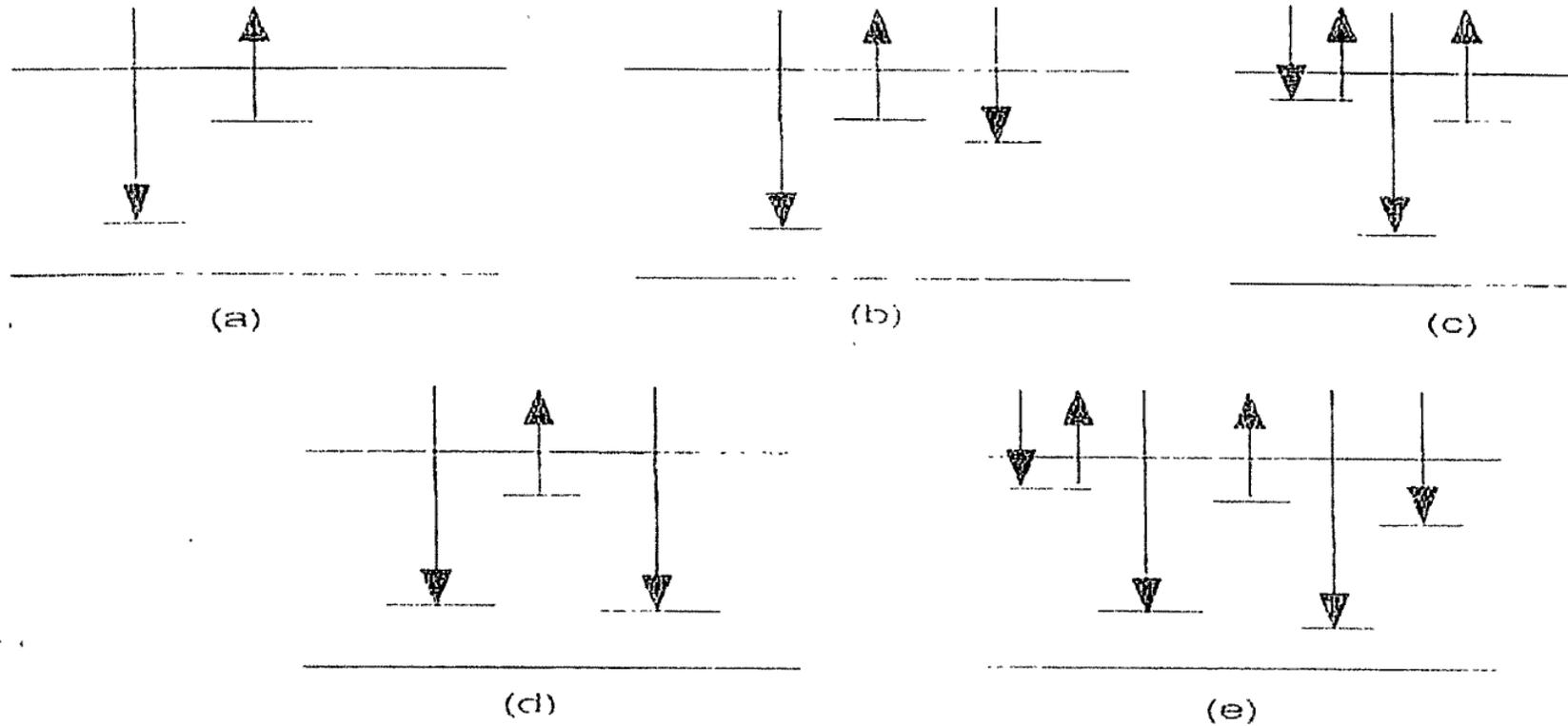
$$m_1 + \frac{m_1}{k} = n \text{-----} (29) \text{ or } m_1 = \frac{n}{1 + \frac{1}{k}} = \frac{n}{K} \text{-----} (30) \text{ where } K = 1 + \frac{1}{k},$$

$$\text{from this } \frac{dm_1}{dt} = \frac{1}{K} \frac{dn}{dt} = -\frac{nf}{K} \text{-----} (31) \text{ and}$$

$$I_{OSL} = \frac{1}{K} n_0 f \exp(-tf) = I_0 \exp(-\frac{t}{\tau}) \text{-----} (32)$$

In the case of two types of recombination centers also, OSL decay curve may still be exponential, but only if the recombination cross section are the same. Comparing (12) and (32) we observed that, as expected the latter case describes a weaker OSL signal by a factor (1/K). An alternative approach is to assume that re trapping into the optically active trap is allowed for the simple case of one trap and one centre we may write

$$I_{OSL} = \frac{dm}{dt} = -\frac{dn}{dt} = nf - n_c(N - n)A \text{-----} (33)$$



**Fig-1.5 OSL Model**

(a) Simplest Model - one electron trap, one hole trap and transport through a de localized band (b) An additional, deep, competing trap (c) A competing, shallow trap (d) Two recombination sites-one radiative and one non-radiative (e) All above elements (traps)

Again the decay is no longer exponential. For the case of  $N \gg n$ ,  $A/A_m = R$  and  $R \gg n/(N-n)$ , a bimolecular second order function results:

$$I_{OSL} = \frac{n^2 f}{NR} = \frac{dn}{dt} = -\frac{dn}{dt} \text{-----(34)}$$

Rewriting  $\frac{dn}{n^2} = -\frac{f}{NRdt}$  -----(35) which yields, after integration,

$$I = I_0 \left(1 - \frac{n_0 f t}{NR}\right)^{-2} \text{-----(36) where } I_0 = \frac{n_0^2 f}{NR}. \text{ For the more general case,}$$

where  $I = \frac{n^b f}{NR}$ , the OSL decay curve is described by  $\left(\frac{I}{I_0}\right)^{\frac{1-b}{b}} = 1 - \frac{n_0 f t}{NR}$  -----(37)

$$I = I_0 \left(1 - \frac{n_0 f t}{NR}\right)^{-b/1-b} \text{----- (38)}$$

### 1.5.2.5 Literature Survey of Optically Stimulated Luminescence of quartz

An exposer with any visible light to priorly irradiated quartz results in to the emission of light known as Optically stimulated Luminescence. This emission of light can be stimulated usually with the light of wavelength 514.5nm(2.41eV) argon line or with the source of broadband green light based on a filtered halogen lamp. As a function of stimulation wavelength, efficiency of OSL production has been measured using other light sources like laser lines from the deep red 646nm (1.92ev) to the blue 458nm(2.71eV) and including 626, 569, 530, and 488nm. A continuous stimulation spectrum from 420-650nm has been obtained for sedimentary quartz. It established that OSL phenomenon is strongly depend on wavelength. Higher energetic photons resulting in higher OSL intensities. Depending upon whether a mono energetic laser line or broad light source is used, it was thought to result in different behavior, such as in the filtered light source of the RISO reader by Duller and Botter-Jenson in 1996 and no difference was observed. It has been concluded by Murray and Wintle that although the lamp gives a stimulation band from 420nm to 550nm, it behaves as a stimulation source with an effective energy of 2.65eV(468nm). It is presumed that the main effect of decreasing stimulation wavelength is to increase the de-trapping probability, and thus the OSL production rate, rather than to access different (deeper) traps. The emission spectrum for Australian sand dune has been observed using a stimulation wavelength of 647nm from a krypton laser. The OSL signal has been extensively studied and used in variety of dating application. No consensus has been reached concerning the long-term stability of the OSL signal. OSL signals have been observed

for quartz grains ranging from a few years to almost a million years old. However, the OSL signal of laboratory-irradiated quartz has been observed to decay substantially on storage over a few weeks at ambient temperatures. Hence the OSL signal appears to be generated from at least one unstable and one stable trap.<sup>25</sup> Huntley et al. first revealed the potential of a dosimetric application of OSL who used the green light from an argon laser (514.5nm) to stimulate luminescence from quartz for dating of sediments.<sup>26</sup> Later studies characterized the OSL properties of quartz with a view to establishing the technique as a dating tool.<sup>27</sup> Later infrared stimulation using IR light emitting diodes was used to measure OSL from feldspars and broad band emitters such as incandescence of arc lamps, in conjunction with carefully selected filters, were used to produce both infrared and visible light stimulated luminescence from feldspar and quartz samples.<sup>28</sup> Most of the commonly used TL materials produced OSL signal to a lesser or greater extent. The applications include retrospective radiation dosimetry and some basic studies of OSL are discussed for a variety of materials, including quartz and feldspars extracted from building materials, and unseparated materials, such as bricks and porcelain.<sup>29</sup>

### **1.6 Scope and Objective of the present work**

The main objective of the work to suggest physical treatment and decide experimental protocol to achieve usual exponential OSL decay curve from synthetic quartz. To achieve this objective, systematic investigations on TL properties of synthetic quartz material is carried out as It may give information about the nature of traps that are responsible for re-tapping of electrons, act as an unstable trap, stable traps, etc. by suggested possibilities. Also, it may become easy to implement for the study of thermal transfer effect on OSL of synthetic quartz samples.

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