CHAPTER-1

INTRODUCTION

1.1 Basic concepts of Luminescence and Thermoluminescence (TL)

When radiation is incident on a material some of its energy may be absorbed & re-emitted as light of a longer wavelength (Stoke's Law). This phenomenon is called as luminescence. There are different types of luminescence processes such as photoluminescence (excitation by optical or ultra violet light), radioluminescence (excitation by nuclear radiation i.e. gamma rays, beta rays, and X- rays) and cathodoluminescence (excitation by electron beam). In addition to excitation by radiation, luminescence can also be generated by chemical energy (chemiluminescence), mechanical energy (triboluminescence), electrical energy (electroluminescence) and even sound wave (sonoluminescence).

Luminescence spectrum is the property of the substance where the absorption of energy and the emission of light photons are dominated by intermediate process between the stages of absorption and emission. The emission of light takes place at characteristic time τ_c after the absorption of the radiation and this parameter allows us to subclassify the process of luminescence. One can distinguish between fluorescence in which $\tau_c < 10^{-8}$ sec and phosphorescence in which $\tau_c > 10^{-8}$sec. The value of $\tau_c < 10^{-8}$sec provides a definition for the essential spontaneous process of fluorescence emission. Thus, the fluorescence emission is during the excitation itself and it ceases after the excitation source is switched off. Phosphorescence on the other hand is characterized by a delay between the radiation absorption and the time T_{max} to reach full intensity. Phosphorescence is seen to continue for sometime after the excitation has been removed.

Phosphorescence itself may be conveniently subdivided into two types namely, short-period ($\tau_c < 10^{-4}$ sec) and long period ($\tau_c > 10^{-4}$ sec) phosphorescence.

The delay observed in phosphorescence corresponds to the time the electron spends in the electron trap. But the mean lifetime (T) is exponentially dependent upon temperature. Therefore, if the trap level is deep enough (energy difference) then T is very large. Thus, luminescence emission can be induced by raising the temperature (thereby reducing T). Since the luminescence in this case, has been stimulated by heating. It is called as thermally stimulated luminescence (TSL). Similarly luminescence can also be observed by another method i.e. stimulation by optical energy. Such a luminescence emission is called Optically Stimulated Luminescence (OSL). In short, delayed phosphorescence can be observed by reducing carrier lifetime by mean of any source of stimulation i.e. thermal or optical etc.[1]

Thermoluminescence (TL) or Thermally Stimulated Luminescence (TSL) is light emission upon heating a material with uniform heating rate, which has been previously excited by the source of radiation. All types of radiation such as gamma, alpha, beta, X-rays and light rays may be used to excite wide range of materials exhibiting thermoluminescence. However, essentially the materials have to be semiconductor or insulator as this material may have metastable energy states in the forbidden gap.

Generally, these metastable energy levels play a vital role in TL production. The metastable energy levels alternatively called as traps are populated due to pre-excitation of the specimens. These traps would not de-trap the charge carriers on its own like any spontaneous phenomenon but need “stimulation” as mentioned earlier to reduce carrier lifetime. Therefore, upon heating, the pre-excited materials depopulate the trapped charges and recombine with opposite charges to emit light. On the other hand, optically stimulated luminescence (OSL) arises from the recombination of charge, which has been optically released from populated metastable energy levels within a pre-exposed specimen. Obviously, the OSL intensity is related to the absorbed radiation dose. During illumination, the OSL signal is observed to decrease to a low level as the metastable charges are depleted.

Luminescence emitted during recombination of the de-trapped charges is usually measured in a spectral region different from that of the exciting photons and is proportional to the radiation dose absorbed in the material. The physical principle of this technique is closely related to those associated with the well-known thermoluminescence (TL) technique.

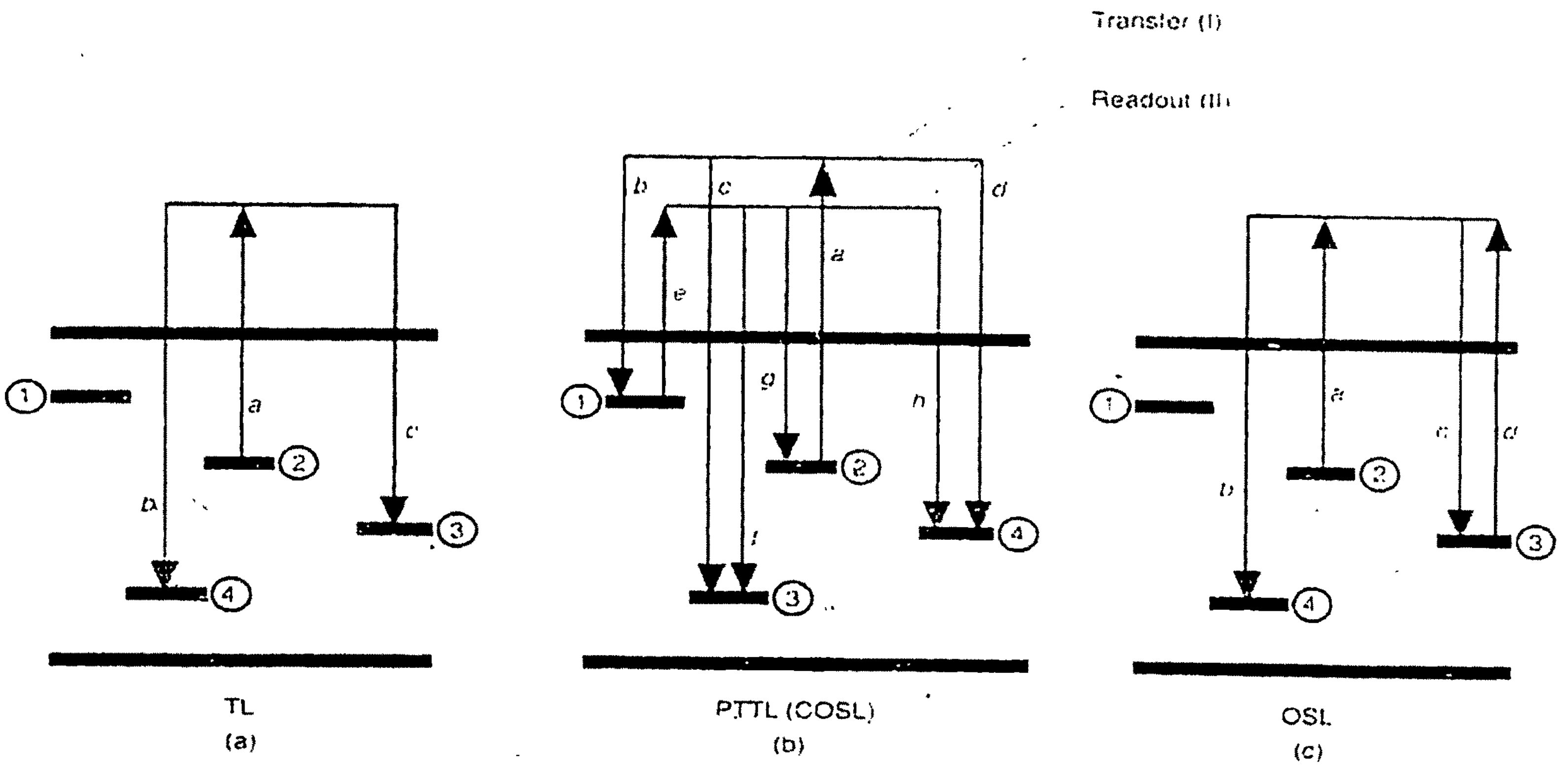
Huntley et al., suggested the essential difference between photoluminescence and OSL that the exciting light causes de-trapping of the electrons so that the light emitted when these reach luminescence centers can be of shorter wavelength than the exciting light.[2]

1.2 Phenomenological Description of OSL and its Advantages over TL

The description of the OSL processes is based on a 'generic' phenomenological model for luminescent dosimetric material (material used in measuring radiation dose) as illustrated in **Fig-1A**.

The model consists of a main dosimetric trap (level-2 in **Fig-1A(a)**) from which electrons may be excited optically (in OSL) or thermally (in TL). Recombination of the released electrons with holes at recombination sites (level-4) produces the OSL or TL emission.

In addition to the main dosimetric trap (or traps) almost all luminescent materials can be expected to contain shallow trapping levels (level-1), which are incapable of storing trapped charges over long periods at ambient temperature. Such levels are normally not directly exploited in most dosimetric



TL (a)

PTTL (COSL) (b)

OSL (c)

Fig-1A: Phenomenological Description of Luminescence Process

(a) TL - 1-Shallow Trap, 2-Dosimetric Trap, 3-Deep Trap, 4-Recombination Center
(b) PTTL- 1-Shallow Trap, 2-Dosimetric Trap, 3-Recombination Center, 4-Deep Trap
(c) OSL - 1-Shallow Trap, 2-Dosimetric Trap, 3-Deep Trap (Advantage over TL), 4-Recombination Center

applications. They do contribute to the overall character of the luminescent behavior of the material. Furthermore, a recent development of cooled optically stimulated luminescence (COSL; a type of PTTL (phototransfer thermoluminescence)) has demonstrated the possibility of using shallow traps directly in dosimetry. Additionally, one can expect the existence of deep, thermally disconnected traps, which are normally too deep to be thermally drained during normal TL applications (level-3).

Although such deep traps may be inaccessible thermally, the optical release of charge from these states is a real possibility in many materials, depending on the chosen stimulation wavelength. This immediately points to one potential advantage of OSL over TL- namely, the possibility of using the deep traps as a source of dosimetric information. **Fig-1A** schematically shows a comparison of the main types of transition occurring during TL, PTTL (COSL) and OSL

In **Fig-1A(a)** transition 'a' represents the thermal excitation of electrons from the dosimetric trap, producing TL via the recombination transition 'b'. However, during this process electrons are also transferred to deep states (transition 'c') and these are lost from the recombination process. Transition 'c' also gives rise to the nonlinear growth of TL with dose.

In **Fig-1A(b)** shows two stages involved in the PTTL (or COSL) process- namely, the transfer stage-I and the read-out stage-II .

During stage-I electrons are removed from the dosimetric trap, 'a', and transferred to the shallow traps, 'b', for a 100% efficient process it is necessary for all electrons to be transferred in this manner. However, a proportion of the electrons are lost via recombination, 'c', and re-trapping, 'd'. During stage-II, the electrons in the shallow traps are thermally released, 'e', and some of this recombine, 'f', to produce the desiré PTTL signal. However, some of them electrons are lost via re-trapping into level-2 and level-4 (transition-'g' and 'h') Thermoluminescence and related photo-transfer thermoluminescence processes can be seen to include several loss processes due to unwanted re-trapping on recombination events.

However, OSL has potential of minimizing the least number of loss processes i.e. **Fig-1A(c)**. Following optical stimulation, 'a', recombination occurs, 'b', to yield the OSL signal. Although the re-trapping into deeper traps may occur, 'c', one also has the potential via a suitable choice of wavelength, to optically release, 'd' the charge from these deep states, either simultaneously with a transition 'a' (like optical stimulation) or subsequent to it. Thus, the loss processes can be dramatically reduced.

In addition, one should recall that many TL materials suffer from the problem of thermal quenching in which the luminescence efficiency is decreased as the temperature while the temperature is increased. Since, OSL is processed at room temperature the problem of thermal quenching can be precluded and one can expect intrinsically higher sensitivity for OSL processes compared with TL processes. On the other hand there is a limitation in thermoluminescence that, once material be heated to excite the light emission; the material cannot be made to emit thermoluminescence again by simply cooling the specimen and re-heating. In order to re-exhibit the luminescence the material has to be re-exposed to radiation, where-upon raising the temperature will once again produce light emission.

The OSL may serve as an alternative to the TSL for radiation dosimetry purposes and as a promising complementary tool for fundamental research.

Instead of heating as in the case of TL, use of light of a fixed wavelength from an effective excited absorption region is made, for OSL to release charge carriers from the traps.

Thus, OSL has also some obvious advantages over TL such as;

- Non-destructive nature of the measurement procedure of the dosimetric signal.
- Possibility of multiple readout.
- Possibility of dealing with deeper trap, which was impossible in case of TL owing to temperature quenching of the corresponding recombination center.
- Higher accuracy and reliability of the results of measurements owing to the absence of chemiluminescence and black body emission.
- Reader can be cheaper due to simple instrumentation system.[3]

1.3 Theoretical concept of OSL

OSL has become a popular method for determining equivalent doses for sedimentary deposits in dating applications and is growing in popularity in dosimetry applications. The method was first introduced for dating by Huntley et al.,[4] and since then the technique has been developed as an important dating procedure. In each of these studies the optical excitation is continuous wave (cw) and is either a high power arc lamp source, along with a monochromator or filter system to select the excitation wavelength, or a laser operating at or near the desired wavelength. The luminescence is monitored continuously while the excitation source is on, and narrow band and/or cut-off filters are used in order to discriminate between the excitation light and the emission light, and to prevent scattered excitation light from entering the detector. Usually, the OSL is monitored from the instant the excitation light is switched on and is observed to take the form of a luminescence decay curve, following an exponential-like function, until all the traps are emptied and the luminescence ceases. The integrated emission signal (i.e. the area under the decay curve) is then recorded and used to determine the absorbed dose.

1.4 OSL Model and its literature survey

General features - In general, the shape of an OSL decay curve dose not lend itself to analysis using simple descriptions. The decay is often non-exponential, typically exhibiting a long tail at long illumination times. Under some circumstances, and for some samples, the OSL may even display an initial slow increase after the illumination is applied, followed by the more usual decrease at longer times. Overall, the decay shape is dependent upon the sample, the absorbed dose, the illumination intensity and the temperature.

Smith and Rhodes[5] and Bailey et al,[6] 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[7] who presented evidence for a thermally assisted process for OSL production studied the thermal dependence of the OSL from quartz further.

A long tail is often observed in the OSL decay curve. This may be caused by either the influence of shallow traps which localize charges released during illumination and slowly release them again at a rate determined by the trap depth and the sample temperature, or a contribution from traps, which are emptying slowly at the excitation wavelength used in the experiment. The shallow traps yield an optically stimulated phosphorescence component to the OSL decay, and they give rise to a temperature dependent OSL component, since at higher temperatures the shallow traps become less effective at trapping charge.

Simplest Model: one trap/one center

Most models for OSL assume transport of the optically excited charge through the delocalized bands in order to reach the recombination site. Thus, the simplest model by which OSL can be produced is shown in **Fig-1B(a).**[8] Here light stimulates trapped electrons, concentration n, into the conduction band at rate f, followed by recombination with trapped holes, concentration m, to produce OSL of intensity I_{OSL}. With the usual definitions, the rate equation describing the charge flow is

$dn_c/dt = -\, dn/dt + dm/dt$ .... (1)

Which can be derived from the charge neutrality condition $n_c + n = m$...(2)

With the assumptions of quasiequilibrium ($dn_c/dt << dn/dt$, dm/dt and $n_c << n, m$) and negligible re-trapping we have

$I_{\mathrm{OSL}} = -dm/dt = -dn/dt = nf$ .... (3)

The solution of which is

$I_{\mathrm{OSL}} = n_o f \exp\{-tf\} + I_o \exp\{-t/\tau\}$...........(4)

Here, n_o is the initial concentration of trapped electrons at time $t = 0$, I_0 is the initial luminescence intensity at $t = 0$, and $\tau = 1/f$ is the decay constant. One can observe a straightforward relationship in which the initial intensity is directly proportional to the excitation rate and the decay of the OSL with time is a

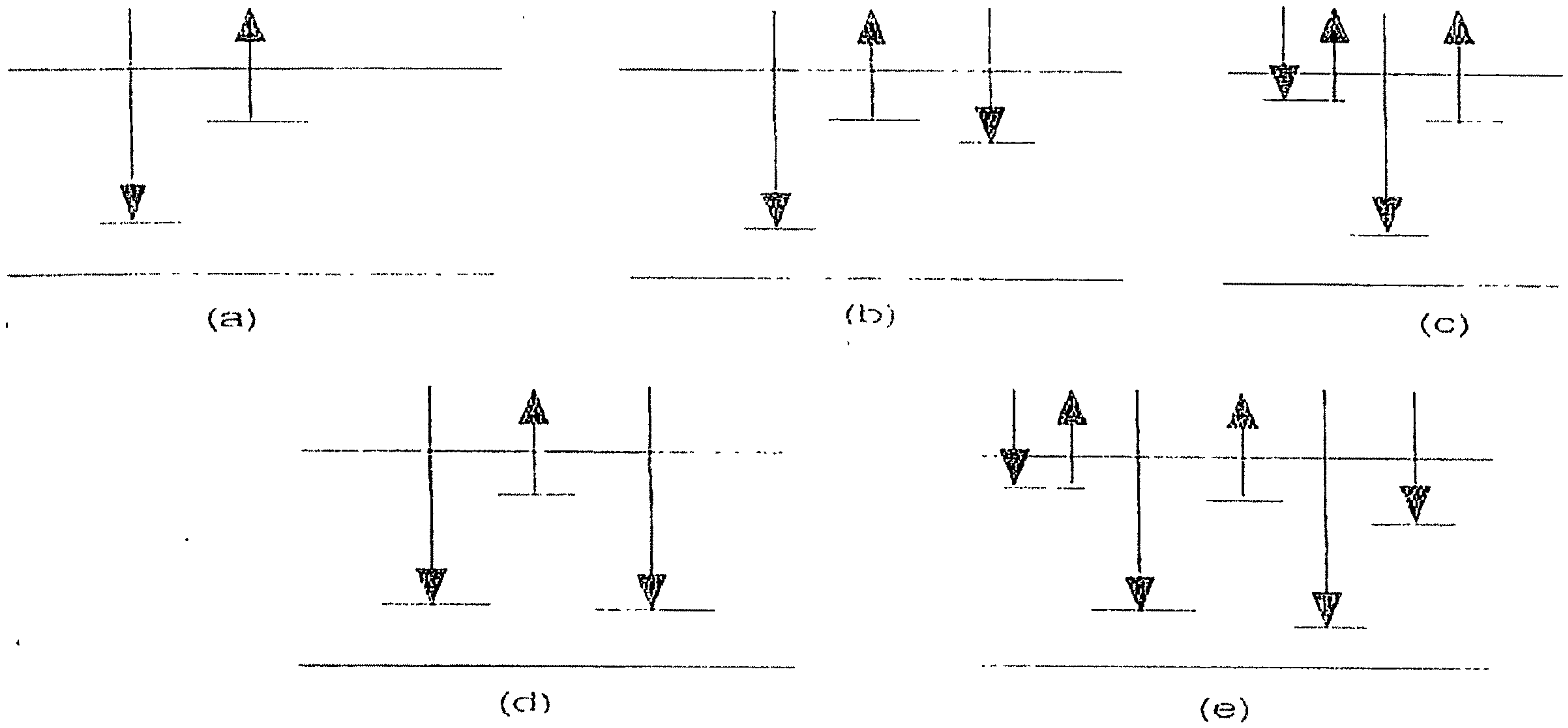


Fig-1B: OSL Model
(a) Simplest Model - one electron trap, one hole trap and transport through a delocalised band (b) An additional, deep, competing trap (c) A competing, shallow trap (d) Two recombination sites-one radiative and one non-radiative (e) All above elements (traps)

simple exponential. The excitation rate, f is given by the product of the excitation intensity, φ and the photoionization cross-section ($f = \varphi\sigma$).

Competing, deep trap

For two optically active traps (concentrations n_1 and n_2 and excitation rates f_1 and f_2) the same assumption yield

$dm/dt = -dn_1/dt - dn_2/dt$....(5)

Along with the charge neutrality condition of $n_1 + n_2 = m$

We have $n_1 = n_{10} \exp\{-t f_1\}$...(6) and $n_2 = n_{20} \exp\{-tf_2\}$ ...(7)

thus $I_{OSL} = n_{10}f_1\exp\{-tf_1\} + n_{20}f_2\exp\{-tf_2\}$

$= I_{10} \exp\{- t/\tau_1\} + I_{20}\exp\{- t/\tau_2\}$...(8)

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-1B(b)**, the OSL intensity may be written as

$I_{OSL} = n_{10}f\exp\{-tf\}-dn_2/dt$...(9)

Where $dn_2/dt = n_c(N_2-n_2)A_2$...(10)

Adopting the standard definitions. With an additional assumption of $N_2 >> n_2$ then $n_c N_2 A_2 \approx$ constant C. Therefore,

$I_{OSL} = n_{10}f\exp\{-tf\} - C$.... (11)

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.

Competing shallow trap

If the competing traps are shallow traps, which are thermally metastable at the temperature of the OSL measurement **Fig-1B(c)**, equation (10) becomes

$dn_2/dt = n_c(N_2-n_2)A_2-n_2p$....(12)

where p is the rate of thermal excitation out of the trap.

Now we have $I_{\mathrm{OSL}} = n_{10}f\exp\{-tf\} + n_2p\text{-}nc(N2\text{-}n_2)A_2$...(13)

The last two terms in (13) 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 f such that at low values of f, the temperature dependent term may be significant.

Competing recombination center

For the case of two recombination centers **Fig-1B(d)**, one of which m_1 is radiative and the other m_2 is non radiative, we have $n = m_1 + m_2$, $n = n_0\exp\{-tf\}$, and $I_{\mathrm{OSL}} = n_0f\exp\{-tf\}\text{-d}m_2/\mathrm{d}t$ ....(14)

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

$\mathrm{d}m_1/\mathrm{d}t = -\ n_cm_1A_{\mathrm{m1}}$...(15) and $\mathrm{d}m_2/\mathrm{d}t = -\ n_cm_2A_{\mathrm{m2}}$....(16)

then, with quassiequilibrium ($\mathrm{d}n_c/\mathrm{d}t \approx 0$) we have

$m_1 \approx m_{10}\exp\{-\ tn_cA_{m1}\}$...(.17)

$m_2 \approx m_{20}\exp\{-\ tn_cA_{m2}\}$....(18) from which we have

$m_1/m_2 \approx m_{10}/m_{20}\exp\{-\ tn_c[A_{m1}\text{-}A_{m2}]\}$...(19) from which we see that the ratio m_1/m_2 is a time dependent function.

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

$m_1 + m_1/k = n$....(20) or $m_1 = n/1+1/k = n/K$....(21) where $K = 1+1/k$, from this $\mathrm{d}m_1/\mathrm{d}t = 1/K\mathrm{d}n/\mathrm{d}t = -\ nf/K$....(22)

and $I_{OSL} = 1/Kn_0f\exp\{-tf\} = I_o \exp\{-t/\tau\}$....(23)

In this way we see that the OSL decay curve may still be exponential in the case of two types of recombination centers, but only if the recombination cross section are the same. Comparing (4) with (23) 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 center we may write

$I_{OSL} = \mathrm{d}m/\mathrm{d}t = -\mathrm{d}n/\mathrm{d}t = nf - n_c(N - n)A$....(24)

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

$I_{OSL} = n^2 f/NR = \mathrm{d}m/\mathrm{d}t = -\mathrm{d}n/\mathrm{d}t$....(25)

Rewriting $\mathrm{d}n/n^2 = -f/NR\mathrm{d}t$....(26)

Which yields, after integration,

$I = I_o(1 - n_0ft/NR)^{-2}$......(27)

Where $I_o = n^2_0 f/NR$. For the more general case, where $I = n^b f/NR$, the OSL decay curve is described by

$(I/I_o)^{1-b)/b} = 1 - n_0ft/NR$...(28)

$I = I_o(1 - n_0ft/NR)^{-b/(1-b)}$....(29)

1.5 Quartz and its importance

A crystalline variety of silicon dioxide, stable at room temperature, is known as alpha-quartz. In nature, alpha-quartz is found in the form of sand and rock like sandstone, quartzite and vein quartz. Single crystal of quartz is an important raw material for the several industries. Electronic grade quartz single crystals exhibit piezoelectric property, which allows them to be used for accurate electronic frequency control as resonator, and for selection as filters, in electronic circuitry. Recently, it is also used for mobile and optical communication devices and projection equipments as an optical product, and an electronic component for personal computers and other digital equipments. Thus, quartz crystal is a technologically important material.

However, single crystals as found in nature, are generally not suitable because of the presence of twins and other imperfections which degrades its electronic properties. It is because of this reason that natural quartz single crystals have gradually been replaced by laboratory grown synthetic quartz single crystals, since the synthetic crystal can be produced without any twins or other imperfections, and can be grown in a manner such that the material loss can be kept to be minimum during fabrication of electronic components.

A large portion of research into defects structure of quartz and its amorphous silica has received its impetus from the need to develop optical and electronic devices, which are able to perform reliably in metal oxide semiconductor (MOS Devices) as a thermal oxide film. Point defects play an important role in many of the optical and precision frequency control applications of quartz crystal.[9] Therefore, it is necessary to identify and characterize point defects present in the material. The related defects dynamics, under different physical conditions are also important to know since this information provide the interpretation about stability of the material in thermal as well as radiation background and hence help to design the device accordingly.

Literature shows the thermoluminescence (TL) properties of the material is the most structure sensitive. Therefore, TL of the natural as well as synthetic quartz was widely studied systematically from the correlation of defects and prior physical treatments point of view.[10-12]

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.

1.6 Thermoluminescence of quartz

A brief literature survey is presented in the following paragraph in order to analyze the different views and ideas suggested by the several researchers working on these technologically important materials and subsequently pinpoint the objective of the present work.

Quartz exhibits a number of TL peaks when irradiated grains are heated from room temperature to 500°C. Two peaks above 300°C were observed for sand sized grains (quartz inclusion) extracted from coarse pottery by sieving and etching in concentrated HF.[13] The preferred peak for dating was observed at 375°C, with a heating rate ~5°C /sec. A peak occurring on the lower shoulder of this peak, around 325°C, was found to show an unreliable dose response and was termed 'malign'. Another peak which attracted attention for dating was only found in laboratory irradiated sample; it occurred at about 110°C and become the basic of the "pre-dose dating method" when it was demonstrated that the peak height could be used to monitor dose-dependent sensitivity changes which were observed after heating to 500°C.

The suits of peaks observed, and their relative intensities can be altered by the choice of detection filter, this arises from the three main emission bands in quartz.[14] Natural quartz has two emission bands, in the blue (460-480nm) and in the orange (610-630nm) for selected temperature range above 300°C.[15] Lower temperature peaks resulting from laboratory irradiation also show an emission band in the near UV to violet (360-420nm), and it is this waveband, which is observed for OSL. The natural TSL for another Australian quartz is obtained using a corning 7-59 filter and (inset) a combination of this filter and an Schott UG-11. The 375°C peak can scarcely be seen on the high temperature edge of the 325°C peak when the Schott UG-11 filter is added.

The two main quartz peaks above 300°C show different bleaching behaviour under sunlight, leading them to be described as the "slowly bleaching peak" (SBP) and the "rapidly bleaching peak"(RBP).[16] Using the Corning 7-59 and Schott UG-11 filter combination for yet another Australian quartz, Spooner et al.,[17] showed that the ease with which the 325°C peak could be bleached by light of 500±20nm.

375°C TL peak (SBP)

The TL peak at 375°C has been used extensively for dating coastal dune sediment in Australia. It bleaches more slowly than the 325°C peak, but wind blown grain and even sediment from major river system are well bleached down to a non-reducible level by the high intensity sunlight incident on Australia. Spectral studies by Scholefield et al., have shown that the 375°C peak emission for their Australian sedimentary quartz has a maximum at about 480nm. The emission has been suggested by Mckeever et al., to be AlO_4 acting as a hole trap in the quartz lattice. The behavior of this slowly bleaching peak has been studied by observing it with a green filter (Chance OGr1) in combination with the Chance HA3 heat absorption filter. Any contribution from the 325°C rapidly bleaching peak could be removed by bleaching with green light, i.e. using the Chance OGrl or an Schott GG-475 filter in front of broad emission light source.

Hornyak 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. Following a green light bleach to remove the 325°C TL peak, the TL was observed with either an ultraviolet filter Schott UG-11 centered at 380nm, of a green filter OGr1 centered at 480nm.

325°C TL peak (RBP)

The TL peak at 325°C has its emission peak at about 380nm. The signal from this peak can be enhanced relative to, but not totally separated from, the 375°C peak by using a detection filter pack made up of an Schott UG-11 and a Corning 7-59 filter, which passes from 329nm to 380nm. The luminescence center used by electrons giving rise to the 325°C peak had previously been identified in quartz extracted from pottery as exhibiting thermal quenching. This refers to the increased probability of non-radiative de-excitation of the center during the recombination process.

The 325°C TL peak bleaches vary rapidly, even at wavelength as long as 500nm. The natural 325°C signal is bleached very rapidly when exposed to 500±20nm light for only 1min, when even exposure for 1200min doesn't cause any reduction in the 375°C peak. Use of higher energy photons will bleach the 325°C peak even more rapidly.

110°C TL peak (pre-dose peak)

The TL peak, which is found at just about 100°C for a heating rate of 5°C/sec is known as the 110°C peak. A peak in this region can be seen in all quartz, whether natural or artificial provided that it has been irradiated less than a few hours before measurement.

The peak has a maximum emission at about 380nm corresponding to the H_3O_4 hole center in quartz. The 110°C peak electron traps can be populated by irradiation or by photo-transfer. For instance, when quartz with a strong 325°C TL peak is exposed briefly to green light at room temperature and then heated, photo-transfer is observed. Zimmermann explained the response of the 110°C TL peak to a small test dose could be enhanced significantly to temperatures above 200°C.

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. In their study of emission wavelengths from another Australian quartz, Franklin et al., reported rapidly bleaching peaks at 150°C-180°C and at 200°C-220°C having emission wavelengths of 392nm and 410nm, respectively. 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.

1.7 Optically Stimulated Luminescence of quartz

Optically stimulated Luminescence can be observed when irradiated quartz is exposed to light of any visible wavelength. Stimulation is usually achieved with either the 514.5nm(2.41eV) argon line or a broadband green light source based on a filtered halogen lamp. However, the efficiency of OSL production as a function of stimulation wavelength has been measured using other 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. These measurements showed that OSL production is strongly dependent upon wavelength, with more energetic photons resulting in higher OSL intensities. This might be thought to result in different behavior, depending upon whether a mono-energetic laser line is used or a broad light source, such as in the filtered light source of the RISO reader by Duller and Botter-Jenson in 1996 and no difference was observed.

Murray and Wintle concluded 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 a laboratory-irradiated quartz has been observed to decay substantially on storage over a few weeks at ambient temperatures. Hence the OSL signal appears able to be generated from at least one unstable and one stable trap.[18]

1.8 Literature survey of OSL

Optically stimulated luminescence technique have now been applied extensively to dating for many years, these techniques have seen only limited use in personal and environmental radiation dosimetry despite several potential advantages. However, the instrumental simplicity of optical stimulation makes this technique attractive for radiation dosimetry, where using materials collected in the environment (for example, in retrospective dose reconstruction after radiation accident) of synthetic materials manufactured specifically for luminescence dosimetry applications. 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.[19] Later studies characterized the OSL properties of quartz with a view to establishing the technique as a dating tool.[20]

More recently, 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.[21] 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 items.[22]

The optically stimulated luminescence technique developed at RISO National Laboratory Denmark and at Oklahoma State University (O.S.U.), and recent collaborative investigation on the properties of variety of natural and synthetic materials are reported. The single green line 514.5nm from an argon laser was initially used as OSL stimulation light source for quartz sample. Luminescence can be excited in feldspars with light energies in the red/near-infrared, owing to the occurrence of one or more excitation resonance in this material.

At RISO demand for OSL measurement of quartz has led to the development of a compact OSL system based on a simple low power halogen lamp (75W) rather than using a complex and expensive argon laser installation. In contrast to the single line output from a laser, the halogen lamp provides a broadband light from which a suitable spectrum can be selected by using optical transmission filters. An infrared diode system in the halogen lamp unit delivers infrared stimulation and has resulted in an OSL/TL system of high flexibility for studying both quartz and feldspar.[23]

A recent development at RISO in OSL instrumentation is scanning of sediment cores, with the aim of being able to determine rapidly the radiation dose acquired by sedimentary materials since deposition and to obtain information about mineralogy. This technique also lends itself to the continuous scanning of, for example, brick, or of tile cross sections taken from building in nuclear accident zones, allowing radiation depth dose profile to be measured directly.[24] At O.S.U. recently reported the use of pulsed stimulation method in which the stimulation source is pulse.[25-26] Phototransferred TL (PTTL) is a well known phenomenon caused by the optically stimulated transfer of electron from deep trap to low temperature TL trap. It has been observed and studied in a wide variety of materials and has been suggested as a tool in radiation dosimetry and in dating of ceramic sample.[27-28] In addition to potential dating and dosimetry applications, however, PTTL is intrinsically interesting in its own right. Investigations on PTTL in quartz are appropriate not only because of its potential advantages for age determination but also due to its relevance to sedimentary dating.

1.9 Objective, the present work and results

Systematic efforts are made to establish a definite correlation between defects, color centers and TL/OSL (under different physical conditions) of synthetic quartz in the present work. This study helps to understand the influence of different protocols followed for sample preparation in different applications. It can suggest the best protocol to be followed for the specific application areas of this material. This study is also aimed to understand the stability of various color centers responsible for the luminescence emission.

Optically stimulated luminescence decay characteristics of powdered synthetic quartz crystal have been studied using 470nm blue light for 100sec stimulation for various protocols at and above room temperature with respect to different physical conditions such as pre-thermal treatment, radiation and duration of pre-thermal treatment systematically. The following significant experimental results are observed.

1.10 Effect of different protocols on synthetic quartz

[A] OSL measured at room temperature reveal:

The nature of curves as well as signals is highly sensitive to pre-thermal treatment, irradiation dose and duration of pre-thermal treatment.

The shape of decay curves are non-exponential for lower irradiation dose and lower pre-thermal treatment i.e. 400°C. For high temperature of annealing such as 600°C and above, the shape of decay curves become exponential and the width of curve decreases systematically.

Either the dose, temperature of pre-thermal treatment or duration of pre-thermal treatment increases the initial rate of OSL decay.

A linear growth in OSL intensity was observed with either increase in beta dose or temperature of pre-thermal treatment. However, specimens pre-heat-treated at 1000°C show significant growth in OSL sensitivity.

Thermoluminescence (TL), (after measuring OSL at room temperature) reveal following facts:

The changes in TL sensitivity and area under the TL glow curves with respect to physical conditions are in accordance with that of OSL.

Systematic shifting and growth in TL sensitivity after OSL at room temperature compared to TL emission before OSL under identical conditions were observed. Similar pattern of observations are observed with different physical conditions also.

[B] The OSL measured at 160°C for 1 min reveal:

Except for the faster decay rate, almost identical pattern of observations are displayed for the OSL measured at high temperature (160°C), compared to the OSL measured at room temperature for different protocols

Absence of low temperature TL peak and shifting as well as growth in TL intensity were observed for the specimens after OSL at 160°C.

[C] The OSL measured at 160°C of pre-heat treated beta irradiated and quickly annealed at 290°C for different durations of 0 to 30sec reveals:

The OSL decay becomes still faster for the pre-heat treated beta exposed and further quickly annealed at 290°C.

Along with shifting and growth in TL sensitivity for the above said protocol, a stable 230°C peak was observed for the specimens after OSL at high temperature.

[D] Cyclic OSL at 160°C for 1 min reveals:

Under identical protocol, OSL intensity decreases systematically when OSL decay at 160°C was repeated on the same sample. While the TL intensity is increased with shifting of glow curve with respect to rise in temperature of annealing after the cyclic OSL measurement.

The observed results are explained on the basis of changes in color centers and dynamics of defects with respect to the different protocols followed by the sample. The results are confirmed with Electron Spin Resonance (ESR) techniques also. On the basis of the observed experimental facts, efforts are made to suggest the right protocol for the use of synthetic quartz as a promising dosimetric material.

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