THERMOLUMINESCENCE AND DEFECT CENTERS IN EUROPIUM DOPED YTTRIUM OXIDE NANOPHOSPHOR

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A Thesis Submitted to

The Department of Physics

PRESENTED IN FULFILMENT OF THE
REQUIREMENTS FOR THE DEGREE OF MASTER OF SCIENCE
JIMMA UNIVERSITY
JIMMA, ETHIOPIA
NOVEMBER, 2021

JIMMA UNIVERSITY
DEPARTMENT OF
PHYSICS

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Dated: November, 2021

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JIMMA UNIVERSITY

Date: **November, 2021**

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Title: **Thermoluminescence and defect centers in Europium doped Yttrium oxide nanophosphor**

Department: **Physics**

Degree: **M.Sc.** Convocation: **November** Year: **2021**

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Abstract

In this work ,we have employed for the analysis of TL glow curves of $Eu^{3+} : Y_2O_3$ material. The continuous promotion in numerical methods has greatly helped to solve the complex TL kinetic equations (rate equations) which can not be solved analytically. The complexity of the rate equations increase with the number of traps considered. In this thesis,the properties of the thermoluminescence glow curves of $Eu^{3+} : Y_2O_3$ has been employed theoretically in view of first order kinetics. As activation energy increases the glow curve shifts to higher temperatures with a decrease in the height. For higher E values or deeper traps more energy or higher temperature is needed to release the charge carriers. Therefore, Similar changes can be noticed as the parameter s is varied but now in the opposite way: As frequency factor s increases the peak shifts to lower temperatures with an increase of the height and a decrease in width. A trapping center with a high frequency factor needs less energy or it needs lower temperature to free a charge carrier.

Keywords: TL (Thermoluminescence), Eu^{3+} (Europium ion), Y_2O_3 (Yittrium oxide)

Acknowledgements

First of all, I would like to thanks my God for his presence in my work.
In particular ,I would like to express my deep gratitude to my advisor and instructor **Dr.Nebiyu Gemechu** for his constant assistance,guidance and encouragement without any reservation starting from the planning up to the development of this thesis work.
Next, my sincere thank goes to my co-advisor **Dr.Menberu Mengesha** for his insightful comment and continuous encouragement to the accuracy of this thesis.
My sincere thank also goes to **Dr.Tolu Biressa** for his nice approach, and giving important softwares..
Finally I would likes to thank **my family** for their moral and economical support for the successes of my work.

Chapter 1

Background of the Study

1.1 Introduction

In order to understand how thermoluminescence (TL) can be used to study luminescent materials, a theoretical background of the phenomenon is important [1]. The luminescence process usually takes place at low temperatures (i.e. it is cold body radiation) in contrast with incandescence which occurs at very high temperatures only is caused by heat. From the type of luminescence, Thermoluminescence is one of them for this case it has also definition. The term "thermoluminescence" (TL) consists of two words: thermo, meaning heat and luminescence, meaning emission of light [2]. These words may appear to mean that the emission of luminescence also called thermally stimulated luminescence is widely used to study defects in insulators and semiconductor material, applicable in field of radiation dosimetry. Thermoluminescence (TL) is a luminescence phenomenon of an insulator or semiconductor which can be observed when the solid is thermally stimulated. TL should not be confused with the light spontaneously emitted from a substance when it is heated to incandescence. At higher temperatures (say in excess of 200^0C) a solid emits (infra) red radiation of which the intensity increases with increasing temperature. This is thermal or black body radiation. TL, however, is the thermally stimulated emission of light following the previous absorption of energy from radiation . From this description the three essential ingredients necessary for the production of TL can be deduced. Firstly, the material must be an insulator or a semiconductor metals do not exhibit luminescent properties. Secondly, the material must have at some time absorbed energy during exposure to ionizing radiation. Thirdly, the luminescence emission is triggered by heating the material [3] . A thermoluminescent material is

thus a material that during exposure to ionizing radiation absorbs some energy, which is stored. The stored energy is released in the form of visible light when the material is heated. Note that TL does not refer to thermal excitation, but to stimulation of luminescence in a sample which was excited in a different way. This means that a TL material cannot emit light again by simply cooling the sample and reheating it another time .It should first be re-exposed to ionizing radiation before it produces light again. The storage capacity of a TL material makes it in principle suitable for dosimetric applications.It should first be re-exposed to ionizing radiation before it produces light again. The storage capacity of a TL material makes it in principle suitable for dosimetric applications.

1.2 Statement of the Problem

Many researchers have been done on the nanoparticles of materials and dosimetery of thermoluminescence materials. The luminescence investigations were carried out using thermoluminescence (TL) methods. But luminescence properties of Eu^{3+}:Y_2O_3 with thermoluminescence(TL) methods plays significant role in the area of the application of this material Eu^{3+}:Y_2O_3. In addition to this the knowledge of the luminescence properties of Eu^{3+}:Y_2O_3 is very important to distinguish it from other materials. To the best of our knowledge this problem has not be studied in other researcher. Therefore, the major concern of this research is to compare the experimental value of luminescence properties of Eu^{3+} : Y_2O_3 with theoretical values. In this context, this statement of the problem expected to answer the following basic questions.

• What is the effect of activation energy on the thermoluminescence glow curves of Eu^{3+} doped Y_2O_3nanophosphor in view of first order kinetics?

•How to describe effect of variation of electron concentration on the thermoluminescence glow curves of Eu^{3+} doped Y_2O_3 nanophosphor in view of first order kinetics ?

• What is the effect of frequency factor on the Thermoluminescence (TL) glow curves of Eu^{3+} doped Y_2O_3 nanophosphor in view of first order kinetics?

• What is the effect of heating rate on the Thermoluminescence glow curves of Eu^{3+} doped Y_2O_3 nanophosphor in view of first order kinetics?

1.3 Objectives of the Study

1.3.1 General Objectives

The general objective of this study is to investigate theoretically the thermoluminescence properties of the material Eu^{3+} doped Y_2O_3 nanophosphor in view of first order of kinetics.

1.3.2 Specific Objectives

• To describe effect of activation energy on the thermoluminescence glow curves of Eu^{3+} doped Y_2O_3 nanophosphor in view of first order kinetics.
• To obtain the effect of variation of electron concentration on the thermoluminescence glow curves of Eu^{3+} doped Y_2O_3 nanophosphor in view of first order kinetics.
• To study effect of frequency factor on Thermoluminescence (TL) glow curves of Eu^{3+} doped Y_2O_3 nanophosphor in view of first order kinetics.
• To examine the effect of heating rate on the Thermoluminescence glow curves of Eu^{3+} doped Y_2O_3 nanophosphor in view of first order kinetics.

1.4 Significance of the Study

This work helps us:
• To understand the peculiar properties of Thermoluminescence glow curve of Eu^{3+} :Y_2O_3 in view of first order kinetics theoretically.
• To investigate the error of theoretical result when compared with experimental results of the Thermoluminescence properties of Eu^{3+} :Y_2O_3 in view of first order kinetics.
•To be used as a reference for the scientific community to know about thermoluminescence properties of Europium doped Yttrium oxide nanophosphore. Also helps as base for other researchers.

1.5 Scope of the Research

The scope of this study is theoretical investigation of thermoluminescence and defect centers in Europium doped yttrium oxide nanophosphore.

1.6 Limitation of the Study

Due to lack of suitable apparatus that helps practical measurements, the study is depend on the theoretical calculation and the scope of the study is limited to investigate, effect of heating rate, effect of frequency factor, activation energy and concentration of electron in trap on the material Europium doped yttrium oxide nanophosphore by using rate equation in view of first order kinetics.

1.7 Thesis Outline

This study contains five chapters and organized as follows: In chapter 1, we have discussed some of the background of Thermoluminescence, including the statement of the problem, objectives, significance and scope of the study. In chapter 2, we review the theoretical backgrounds of Luminescence, Thermoluminescence process, definition of phosphor, Thermoluminescence application, simple model of thermoluminescence, methods of determining kinetic parameters and the properties of Eu^{3+} :Y_2O_3. In chapter 3, We discuss some of the methodology we used for our work In chapter 4, We discuss the properties and the effect of varies heating rate on the Thermoluminescence glow curve of Eu^{3+} : Y_2O_3 with parameter activation energy (E), frequency factor (s), the dose and the concentration of the trapped electron in the trap and compare the theoretical and experimental result on Eu^{3+} : Y_2O_3 material. Finally in chapter 5, we draw some conclusions.

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Chapter 2

Review of Related Literatures

2.1 Luminescence Phenomena

Luminescence is defined as the emission of light above that expected for black body, from some solids commonly called phosphors. This emission is the release of energy stored within the solid through some type of prior excitation of the solid electronic system, i.e. by visible, infrared (IR) or ultra violet (UV) light and ionizing radiation. The light emitted has a longer wave length of the one of the incident radiation (Stoke's law). Furthermore, the wavelength of the emitted light is a characteristic of the luminescent material. The ability to store the radiation energy is important in luminescence dosimetry and is generally associated with the presence of activators (i.e., impurity atoms and structural defects) which act as trapping levels for the free electrons generated by excitation. Luminescence is the emission of light by a material as a consequence of it absorbing energy. A material that emits light is called luminescent material. The emission of light takes place in a characteristic time t after the absorption of the radiation and this parameter allows us to sub classify the process of luminescence. Thus we can distinguish between fluorescence in which $t < 10^{-8}$ s and phosphorescence in which $t > 10^{-8}s$ [4]. To describe light emitting or luminescent material the Greek word 'phosphor' is usually used and it means 'light bearer'. A phosphor emits energy from an excited electron as light and the excitation of the electron is triggered by absorption of energy from one of the external excitation sources. An excited electron occupies a quantum state whose energy is above the minimum energy ground state. In semiconductors and insulators, the electronic ground state is commonly referred to electrons in the valence band, which is completely filled with these electrons. The excited quantum state often lies in the conduction band, which

is empty and separated from the valence band by gap. Therefore unlike metallic materials, small continuous change in electron energy within the band is not possible. Instead a minimum energy equal to the band gap energy is necessary to excite an electron in semiconductors and insulators, and the energy released by de-excitation is often nearly equal to the band gap. The band gap of semiconductor material is such that at room temperature, very few electrons are promoted from the valance band to the conduction band leaving holes in the valence band. In general, luminescence emission is explained by the transfer of energy from radiation to the electrons of the solid, thus exciting the electrons from a ground state to an excited state. The emission of a luminescent photon takes place when an excited electron returns to a lower energy state [5].

2.2 Definition of Phosphor

• Phosphors (or luminophors) may be defined as solid materials showing luminescence. Phosphors essentially consist of very pure inorganic materials doped with suitable ions called activators [6, 7].

•The activator is usually present in concentration levels varying from one to five parts per million of the host lattice. Often, additional ions act as charge compensators or donors in the lattice. These are termed co-activators. Luminescence [8] is produced when activators are inserted into the host lattice. They create local centres that can be excited to produce Luminescence. Normally phosphors are made from crystalline materials that act as host crystals. They contain x amount of controlled impurities which are called activators, that generate the luminescence. Different methods and type of excitation gives rise to various kinds of luminescence [9]. Emission obtained

a) From the absorption of photons (light) is called photoluminescence.

b) By applying electric currents or electric fields (a.c or d.c) is called electroluminescence.

c) By bombardment with an electron beam is called cathodoluminescence.

d) From the use of pressure is called triboluminescence.

e) By the use of heat is called thremoluminescence.

f) From a chemical reaction is called chemiluminescence.

Phosphors have seen wide spread use in for example televisions tubes, cathode ray tubes, fluorescent lighting (strips and compact light bulbs).The cathode ray tubes

have now been almost totally replaced by flat screen technologies [9,10]. Colour television technology was invented and started by John.L.Baird in 1928 and was further developed in 1940 by Peter Goldmark. Initially the colour television was transmitted in red, blue and green [11].

2.3 Thermoluminescence Process

Thermoluminescence(TL)is defined as the emission of light from a semiconductor or an insulator when it is heated, due to the previous absorption of energy from irradiation. The graph of the amount of light emitted during the Thermoluminescence process as a function of the sample temperature is known as a "Thermoluminescence glow curve." On subsequent heating the energy may be released and some of it may be in the form of light, which we call thermoluminescence (TL). The underlying mechanism involves the role of:-

• crystal defects which allows the storing of energy derived from exposure to radiation through the trapping of carriers at these defect centers

• Subsequent release of stored energy as visible light when these trapped carriers, after having been freed by thermal stimulation, recombine at the luminescent centers provided by impurity atoms in the solids [12-13].

The fundamental principles which govern the production of Thermoluminescence are essentially the same as those which govern all luminescence processes and hence Thermoluminescence is one member of a large family of luminescence. However, the application of heat stimulates the release of the stored energy in the sample due to pre-exposure to the ionizing radiations which in turn produces luminescence. The basic effect leading to the production of Thermoluminescence is the trapping of charge carriers, i.e. electrons and holes, produced during exposure to an external source at defect sites in the material. Defect sites can be divided into two categories:- i, Those inherently present in the material ii, Those produced by external means, such as deliberately doping the sample with impurities. A well-known example of the first category is a negative ion vacancy. Such a vacancy can trap an electron and is then called an F center. An example of the second category is a lattice vacancy caused by a higher valence impurity ion at the position of a lattice ion. A trivalent cation impurity in a divalent lattice, for example, would induce the formation of a cation vacancy in the lattice to maintain charge neutrality. Cationic vacancies are potential sites for trapping holes. There are many other types of defects that can act as electron

or hole trapping centers. Revealing the nature of those defects is part of the research of luminescent materials. Heating of the material causes the release of the trapped charge carriers and the recombination of electrons and holes at a luminescent center. The release of some of the stored energy excites the center and relaxation may lead to the emission of light. The nature of the luminescent center can be revealed by measuring the emission spectrum which is in many cases characteristic for a specific element. We note that not every recombination event leads to luminescence. If, for example, there is not enough energy available to excite the center, the recombination will be non-radiative. The process leading to recombination includes, in many cases, the transition of charge carriers through the conduction or valence band, but localized transitions may also take place [14-15].

2.4 Application of Thermoluminescence

The modernization and development in the instrumentation and better understanding of Thermoluminescence have helped the professionals to solve their problems in many fields. The present research on Thermoluminescence has explored its very high application potential in various fields such as archaeology, analysis of defects in solids, radiation dosimetry, geology, forensic sciences, quality control in industry for controlling the quality of many glass, ceramics, and semiconductor products; biology and biochemistry for studying the properties (involving different chemical reactions) or contents of proteins and leaves. Moreover, Thermoluminescence has also interesting applications in space science,thermostimulated luminescence (TSL) photography, radiation physics, petroleum exploration.

2.4.1 Archeology

The thermoluminescence output from the specimen is mostly attributed to Thermoluminescence sensitive mineral inclusions (mostly quartz) in the host clay matrix of the pottery fired in the kiln sometime in the long past. That event is considered to be the starting of the 'Thermoluminescence clock' for archaeological dating [16].

2.4.2 Defects in Solids

TL is very sensitive to traces of impurities or defects within the host material of a given sample and experiments on TL yields useful information on the properties

of the various types of defect present within an insulator or semiconductor. This includes the position of the defect within the energy gap and sometimes the type of defect itself. There are early reports on the effect of impurities on the TL properties of various materials. In general the impurities give rise to the localized energy levels within the forbidden energy gap and that these are crucial to the TL process. For the purpose of detecting the presence properties of these defect levels, the analysis of TL glow curves is important. In addition to defect levels produced by external means such as irradiation or doping, there are also those due to defects, such as lattice vacancies and interstitial inherently present in the material. The presence of this type of imperfection is also crucial to the TL process in many materials[17].

2.4.3 Radiation Dosimetry

Thermoluminescence has an interesting application in the field of radiation dosimetry [18]. It is clear that the absorption of radiation increases the level of Thermoluminescence observed from a given sample by filling the localized energy levels with trapped electrons while the absorption of heat from the environment tends to reduce the numbers of trapped electrons by de-trapping them.

2.4.4 Geology

In geology the sun light bleaching is considered to be the basis for dating the geological event. The exposure of sand grains to sun light during their weathering and transport through wind and water results in bleaching of their geological Thermoluminescence. This bleaching is effective enough to reduce Thermoluminescence level to a negligible value [19].

2.5 Properties of Eu^{3+}doped Y_2O_3 Nanophosphor

In recent years, some attention has been paid to the study of radiation induced defects in laser and luminescent materials, since they affect the optical and stimulated emission properties. Like many oxide based materials rare earth doped Y_2O_3 has a resilience to ionized radiation [20].The wide variety of materials of dopants that can be incorporated allows the material to be tuned to emit in the red Eu^{3+} region of electromagnetic spectrum[21,22].Additionally ,the wide energy gap of the Y_2O_3 material system reduces the effect of optical absorption by the host. $Y_2O_3 : Eu^{3+}$

has attracted much attention because of its high brightness as a red color phosphor under UV or cathode ray excitation ,acceptable atmospheric stability and reduced degradation under applied voltages[23-25]. $Y_2O_3 : Eu^{3+}$ were found to be suitable for field emission display (FED), Vacuum fluorescent display panel (PDP) devices. It has also been reported to show high photo and cathodoluminescence efficiency for $Y_2O_3 : Eu^{3+}$.Europium is efficiently used as a luminescent center in phosphors for various purposes. Phosphors doped with europium ions are of greater importance for observing red colors on the monitors of various display devices [26]. Red phosphor of Y_2O_3 doped with trivalent ions such as Eu has in particular attracted considerable interest in terms of its high chemical durability and thermal stability [27]. This phosphor has been widely used in CRT displays. Nanoparticles have gained an immense interest, in anticipation that this unexplored range of material dimensions will yield size-dependent properties. The physical and chemical properties vary drastically with size, which clearly represents a fertile field for materials research [28-30]. Producing nano scale materials opens new opportunities in the creation of product with enhanced properties for applications such as electronics, optics, medicine and magnetism. Luminescent phosphors are among the current nanostructures of materials that can be incorporated into various applications,viz., the development of flat-panel displays depends critically on the design of bright and stable phosphors [31]. Nanocrystalline phosphors are suitable for high definition television (HDTV) where conventional bulk phosphor cannot be used [32]. The morphology and the particle size affect the emission intensity of phosphor [33-35]. In general, the luminous efficiency of phosphor reduces with decreasing particle size as long as the quantum size effect does not occur [36]. Optically transparent yttrium oxide Y_2O_3 appears to be a perspective laser material, because its thermal conductivity is two and ten times higher than thermal conductivity of YAG and glass respectively [37]. Nanophosphor Y_2O_3 crystallites have high luminescence efficiency in the orange-red, high purity, good chemical resistance and thermal stability. Therefore the $Y_2O_3 : Eu3+$ powder is largely used in optical display technology, medical image and illumination [38]. Eu^{3+}activated Y_2O_3 nanophosphors have been studied for a long time because of their efficient luminescence under UV excitation. In recent years, the nanoparticles of Y_2O_3 :Eu has been rewarded much attention for its incredible potential applications in optical display and lighting materials and dosimetry due to their excellent chemical and thermal stability, good corrosion resistivity, broad optical absorption and low phonon

energy which reduces the multiphonon relaxation process which leads to high luminescence efficiency [39,40]. Rare earth ions have long been used for producing phosphor materials. Doping appropriate host matrix (usually oxide) with these ions products sharp and intense emission under UV excitation [41]. Researchers have succeeded to produce many kinds of rare earth-doped oxide phosphors emitting various colors. To date,they succeeded to produce blue phosphor of (Zn,Mg)O:Zn, green phosphor of ZnGa2O4:Mn, red phosphor of Y2O3:Eu and CrTiO3:Pr [42, 43], red phosphor of SrTiO3:Pr,M(M = Al or Ga) [44], and yellow-green phosphor of (Y,Gd)Al5O12:Ce [45, 46].

2.6 Simple Thermoluminescence Model

The process by which materials emit light when heated can be understood by considering the simplest possible model consisting of two localized levels, an isolated electron trap (T) and a recombination center (RC), as shown in Figure 2.1. This is commonly referred to as the one-trap-one-recombination center model (OTOR). Let us denote by N the total concentration of the traps in the crystal m^{-3}, by $n(t)$ the concentration of filled traps in the crystal in,m^{-3} at time t,and by $n_h(t)$ the concentration of trapped holes in the recombination center in, m^{-3}. The initial concentration of filled traps at time t=0 is denoted by n_o. In a typical thermoluminescence experiment the sample is heated with a linear heating rate $\beta = \frac{dn}{dt}$ from room temperature up to a high temperature usually around 500^oC. As the temperature of the sample is increased, the trapped electrons in T are thermally released into the conduction band, as shown by the arrow for transition 1 in Figure 2.1. These conduction band electrons can either recombine with holes in the recombination center RC (transition 2), or they can be retrapped into the electron trap T (transition 3), as shown in Figure 2.1. The intensity of the emitted light is equal to the rate of recombination of holes and electrons in the recombination center, and is given by

$$I(t) = \frac{dn_h}{dt} \tag{2.6.1}$$

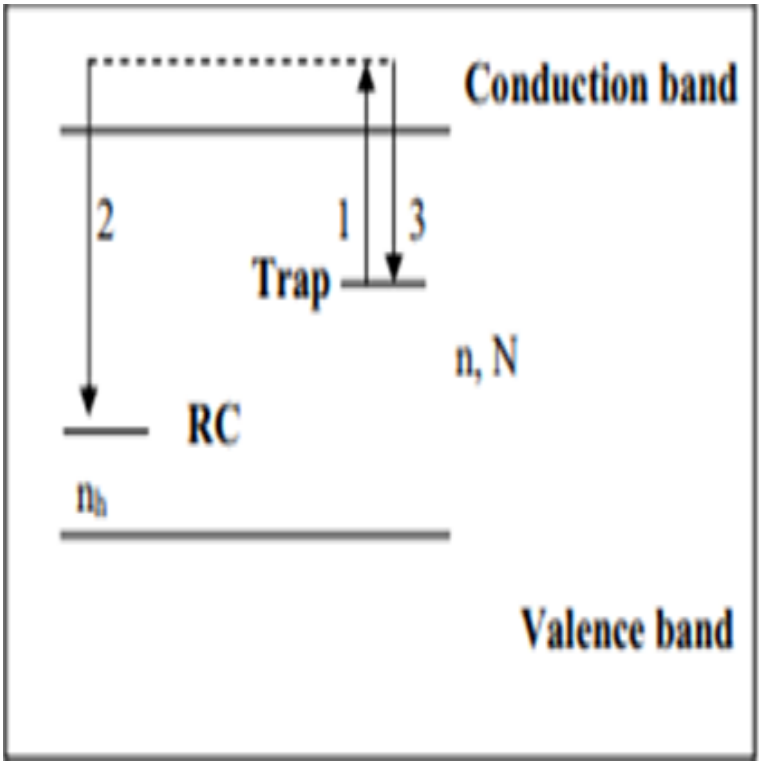

Figure 2.1: The simple two-level model for the thermoluminescence process[47]

2.7 Expressions for First-, Second-, and General-Order Thermoluminescence Kinetics

The equations governing the thermoluminescence processes have been given by Randall-Wilkins [48] ,Garlick-Gibson [49] and May-Partridge [50] for first, second, and general orders.

2.7.1 Thermoluminescence Equations For First-Order Kinetics

In 1945, Randall and Wilkins [48]extensively used a mathematical description for a TL peak in a glow curve. Their mathematical treatment was based on the energy band model and yields to the well-known first order expression. Between the delocalized bands, conduction band, CB, and valence band, VB, two localized levels

(metastable states)are considered, one acting as a trap, TR, and the other acting as a recombination center, RC. The distance between the trap TR and the bottom of the CB is called activation energy or trap depth: E. This energy is the energy required to liberate a trapped charge.

The probability p, per unit of time, that a trapped electron will escape from the trap, or the probability rate of escape per second, is given by the Arrhenius equation, having considered that the electrons in the trap have a Maxwellian distribution of thermal energies .

$$p = s.exp\frac{-E}{KT} \tag{2.7.1}$$

Where:E is the trap depth (eV), k is the Boltzmann's constant= 8.61710^5 ev/k, T is the absolute temperature (K), S is the frequency factor sec^{-1}, depending on the frequency of the number of hits of an electron in the trap, seen as a potential well. If n is the number of trapped electrons in the trap, and if the temperature is kept constant, then n decreases with time t according to the following expression

$$\frac{dn}{dt} = -pn \tag{2.7.2}$$

Integrating Eq. (2.7.2) one obtains

$$n(T) = n_o exp[exp[(\frac{-s}{\beta})] \int_{T_O}^{T} exp(\frac{-E}{KT}dT)] \tag{2.7.3}$$

Where n_0 is the number of trapped electrons at the initial time $t_o = 0$. Assuming now the following assumptions:

•Irradiation of the thermoluminescent material at a low enough temperature so that no electrons are released from the trap,

•The life time of the electrons in the conduction band is short,

• All the released charges from trap recombine in luminescent center,

• The luminescence efficiency of the recombination centers is temperature independent,

• The concentrations of traps and recombination centers are temperature independent,

• No electrons released from the trap is retrapped according to the previous assumptions, the TL intensity, I, at a constant temperature, is directly proportional to the detrapping rate;$\frac{dn}{dt}$:

$$I = -C[\frac{dn}{dt}] = cpn.... \tag{2.7.4}$$

Where c is a constant which can be set to unity. Eq. (2.7.4) represents an exponential decay of phosphorescence. Introducing now a linear constant heating rate, $\beta = \frac{dT}{dt}$ one obtains

$$I(T) = n_o s exp\frac{-E}{KT} exp[\frac{-s}{\beta}\int_{T_O}^{T} exp\frac{-E}{KT}dT'] \tag{2.7.5}$$

This expression can be evaluated by mean of numerical integration.

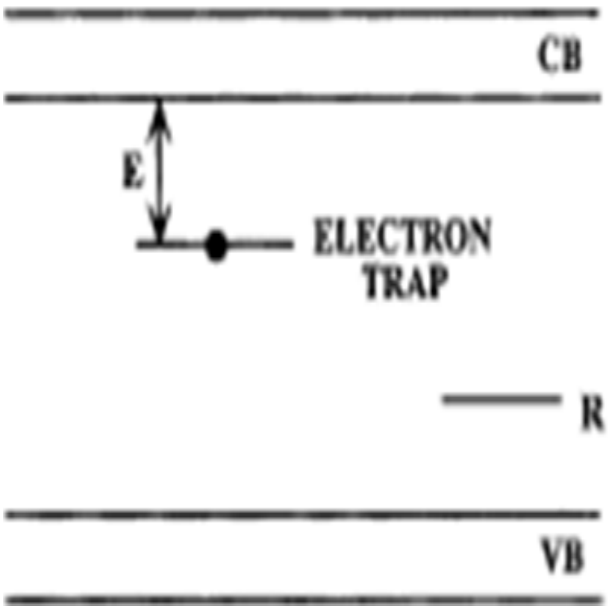

Figure 2.2: Randal and wilkins model[48]

2.7.2 Thermoluminescence Equations For Second-Order Kinetics

Using the same OTOR model, Garlick and Gibson (GG) modified the model of TL intensity proposed by RW [48]. According to GG model, an electron which is detrapped into the conduction band from the trap centers after absorption of thermal energy has two options i.e., it may either recombine with a hole trapped at recombination center to produce luminescence or may be retrapped by any of the vacant traps. However, in RW model, retrapping is assumed to be negligible and the detrapped electrons are

assumed to recombine directly with the trapped holes giving luminescence.. The term second order kinetic is utilized to explain a behavior in which retrapping is present

$$I(T) = \frac{-dn}{dt} = -s^{'} n^2 exp\frac{E}{KT} \tag{2.7.6}$$

Where $s' = \frac{S}{N}$ is called pre-exponential factor and constant having dimensions of $cm^3 s^{-1}$. A second order kinetics based on such as Eq (2.7.6) are said to follow second order kinetics. The shape of the TL glow curve is given by Garlik and Gibson[49]is

$$I(T) = -s' n^2 exp\frac{-E}{KT} = \frac{n_o^2 s^{'} \exp(\frac{-E}{KT})}{N[1 + \frac{n_o s}{N\beta}\int_{T_O}^{T} \exp(\frac{-E}{KT'})dT']^2} \tag{2.7.7}$$

again equation(2.7.7) can be written as;

$$I(T) = s^{'} n^2 exp\frac{-E}{KT} = n_o^2 \frac{S}{N} exp\frac{-E}{KT}[1 + \frac{n_o s}{N\beta}\int_{T_o}^{T} exp\frac{-E}{KT'}dT']^{-2} \tag{2.7.8}$$

2.7.3 Thermoluminescence Equations For General Order Kinetics

May and Partridge (MP) suggested a more general expression for TL emission which would satisfy not only the first order and second order kinetics expressions when b = 1 and b = 2 respectively, but would also include all other possible values of b including its non-integral values between 1 and 2 or even outside this range. Accordingly, they proposed the following expression for TL intensity

$$I(t) = \frac{-dn}{dt} = n^b s^{'} exp\frac{-E}{KT} \tag{2.7.9}$$

s' frequency factor b kinetics, Therefore, this expression for TL intensity is called general order kinetics. Rearranging

$$\frac{dn}{n^b} = -s^{'} exp\frac{-E}{KT}dT \tag{2.7.10}$$

which gives by integration

$$n = n_o[1 + s^{'} n_o^{b-1}(b-1)texp\frac{-E}{KT}]^{\frac{1}{1-b}} \tag{2.7.11}$$

can be written;

$$n = n_o[1 + s^{''}(b-1)te\frac{-E}{KT}] \tag{2.7.12}$$

where $s" = s^{'} n_o^{b-1}$ The frequency factor s" is constant for a given dose and would vary when the dose is varied. The intensity is given by

$$I(t) = \frac{-dn}{dt} = s^{'} n^b exp\frac{-E}{KT} = s^{''} n_o exp\frac{-E}{KT}[1 + S^{''}(b-1)texp\frac{-E}{KT}] \tag{2.7.13}$$

Assuming a Linear heating rate $dT = \gamma dt$

$$n = n_o[1 + s^{''}(\frac{b-1}{\beta}) \int exp\frac{-E}{KT} dT^{'}]^{\frac{1}{1-b}} \tag{2.7.14}$$

The intensity is given by;

$$I(T) = S^{''} n_o exp\frac{E}{KT}[1 + S^{''}(\frac{b-1}{\beta}) \int exp\frac{-E}{KT} dT^{'}]^{\frac{1}{1-b}} \tag{2.7.15}$$

To get the parameter in general order kinetics the equation above can experienced in the form of

$$\frac{KT_M^2 bs^{''}}{\beta E} exp\frac{-E}{KT_M} = 1 + S^{''}\frac{b-1}{\beta} \int exp\frac{-E}{KT} dT^{'} \tag{2.7.16}$$

2.8 Methods of Determining Kinetic Parameters

Many differentmethods have been proposed to synthesize Y_2O_3-based phosphors such as the sol-gel method [51], chemical vapour deposition [52], combustion synthesis [53], precipitation [54], spray pyrolysis [55], simple heating of precursor in a polymer solution [56], hydrothermal [57-59], and bicontinuous cubic phase [60,61]. Among various methods combustion method has been studied extensively due to its simplicity and easiness to control the particle size of the products [62-64].The success of this process is due to intimate blending among the constituents using suitable fuel or complexing agent (e.g. citric acid and urea etc) in an aqueous medium followed by exothermic redox reaction between fuel and an oxidizer (i.e. nitrates) [65]. The thermoluminescence technique plays a vital role in getting the information about the trapping parameters, such as activation energy, frequency factor, and order of kinetics, of the traps present in the materials. These parameters quantitatively describe the trapping and emitting centers, the knowledge of which is essential for the understanding of luminescence properties of materials. In the present studies, the kinetic parameters are calculated using peak shape methods [66]. The TL glow curve is related to the trap levels lying at different depths in the band gap between the conduction and the valence bands of a solid. These trap levels are characterized by different trapping parameters such as trap depth, order of kinetics, and frequency factor [67].

Chapter 3

Research Methodology

This study has been carried out by using the following procedures. These are: materials used and, the methodology.

3.1 Materials

An intensive survey of literature from published articles, books, Latex software ,journals, and computers were additional instruments to accomplish this project.

3.2 Methodology

3.2.1 Analytical

Analytical solution of the rate equations are achieved by assuming negligible retrapping in case of first order kinetics, whereas, the consideration of re-trapping complicates the rate equations and hence numerical approaches are employed. Therefore, for numerical calculations, Mathematica Software will be used to determine the effect of heating rate, Activation energy(E) and Frequency factor(s) on Thermoluminescence glow curves of $Eu^{3+} : Y_2O_3$ in view of first order kinetics.

3.2.2 Computational(Graphical)

The properties of Thermoluminescence of the material $Eu^{3+} : Y_2O_3$ with linear heating rate will be interpreted graphically with Mathematica.

3.2.3 Ethical Consideration

The University guidelines and regulations are strictly followed and respected. Ethical authorization like proper citations and appropriative verification will be obtained from research review and Ethical committee of College of Natural Science, Jimma University. Any concerned will be informed about the purpose of the study.

Chapter 4

Result and Discussion

4.1 Thermoluminescence Glow Curve Properties of Eu^{3^+}:Y_2O_3 nanophosphor

In this chapter, we seek to determine the effect of activation energy,frequency factor, heating rate and concentration of electrons in the traps , the TL intensity as a function of temperature (the glow curve) of Europium doped yttrium oxide nanophosphor in the view of first order kinetics by employing the method which is based on mathematica software and the input parameters used for first order kinetics are: $E_1 = 0.49eV, E_2 = 0.53eV, E_3 = 0.57eV, S_1 = 1 * 10^7 s^{-1}, S_2 = 7 * 10^7 s^{-1}, S_3 = 2 * 10^8 s^{-1}, \beta = 6.7^0 cs^{-1}, n_o = 10^{10} cm^{-3}$

4.1.1 Effect of the activation energy (E) on the Thermoluminescence glow curve of Europium doped yttrium oxide nanophosphor

The Thermoluminescence intensity as a function of temperature with different value of E. Calculations are carried out using equation (2.7.5) which has been generated by using Mathematica.

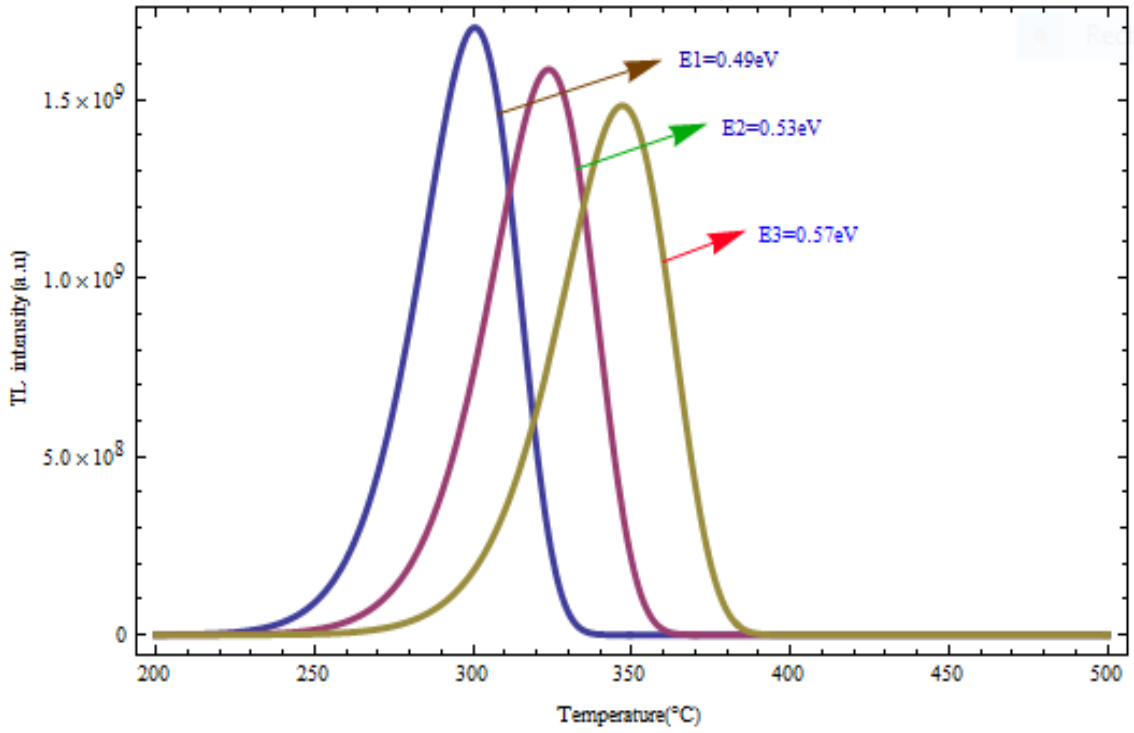

Figure 4.1: The effect of activation energy on the Thermoluminescence Properties of Eu^{3+}:Y_2O_3 at three different (E) values
$E_1 = 0.49eV, E_2 = 0.53eV, E_3 = 0.57eV, S = 7 * 10^7 s^{-1}, \beta = 6.7^o cs^{-1}, n_o = 10^{10} cm^{-3}$

4.1.2 Effect of variation concentration of electron in traps on the Thermoluminescence glow curve of Europium doped yttrium oxide nanophosphor

It is interesting to note that there was no observed shift of the intensity peak positions to high or low temperature region due to the variation of the initial filled concentration of electron n_o in traps by using equation (2.7.3).This means that the peak characteristics are independent of n_o in the sample, which indicates the population of electron in traps. It is observed that the concentration of trapped electrons population decreases as the temperature increases just above 270^oC, reaching its minimum values approximately between 345^oC and 350^oC and at temperature of $150^oC - 270^oC$ the trapped electron is not released from the center of trap because the temperature is

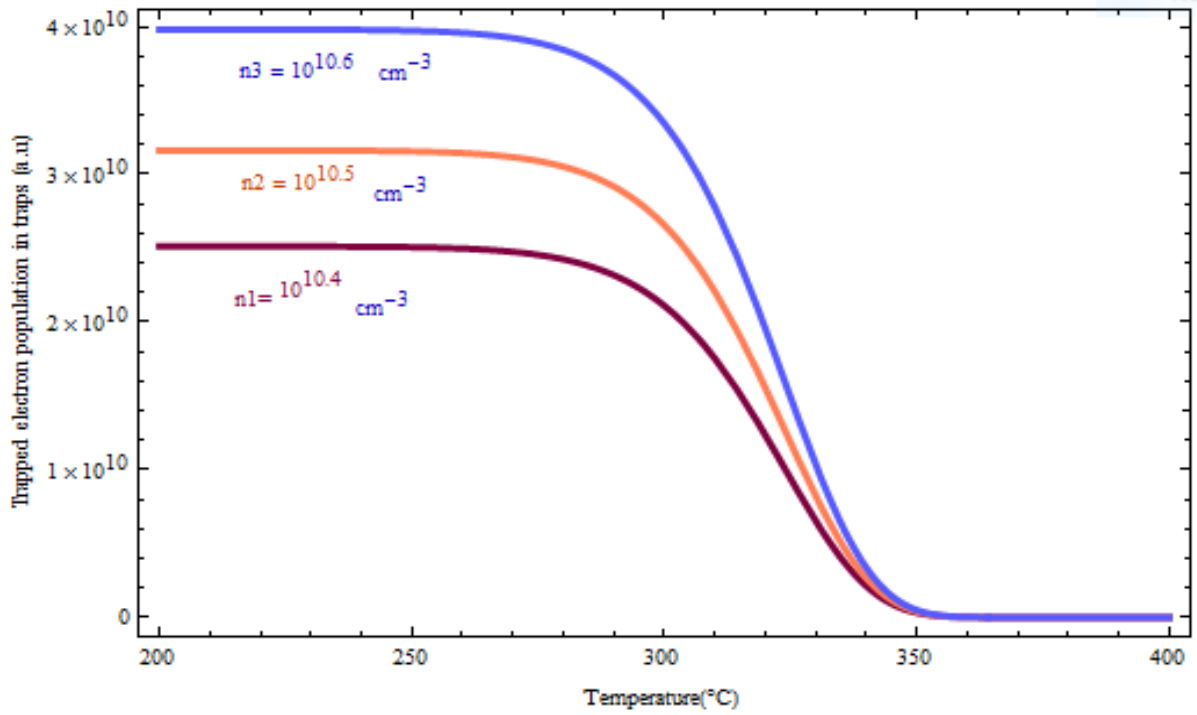

Figure 4.2: The effect of variation of electron concentration (n_o) in trap on the Thermoluminescence (TL) Properties of Eu^{3+}:Y_2O_3 at different initial concentration (n_o) of the electron in traps

$E = 0.53eV, S = 7 * 10^7 s^{-1}, \beta = 6.7^o cs^{-1}, n_1 = 10^{10.4} cm^{-3}, n_2 = 10^{10.5} cm^{-3}, n_3 = 10^{10.6} cm^{-3}$

not enough to eject it, but above the temperature of 270^oC the trapped electron start to excite from trap center.

4.1.3 Effect of frequency factor (s) on Thermoluminescence glow curves of Europium doped yttrium oxide nanophosphor

Properties of TL glow curve for Eu^{3+}:Y_2O_3 nanophosphor, showing the variation with the frequency factor (s) from equation 2.7.5 given in literature. Parameter values we used are; $E = 0.53eV, S_1 = 1 * 10^7 s^{-1}, S_2 = 7 * 10^7 s^{-1}, S_3 = 2 * 10^8 s^{-1} \beta = 6.7^o cs^{-1}, n_o = 10^{10} cm^{-3}$. For a given values of 'E' and 'β', the glow peak shifts to

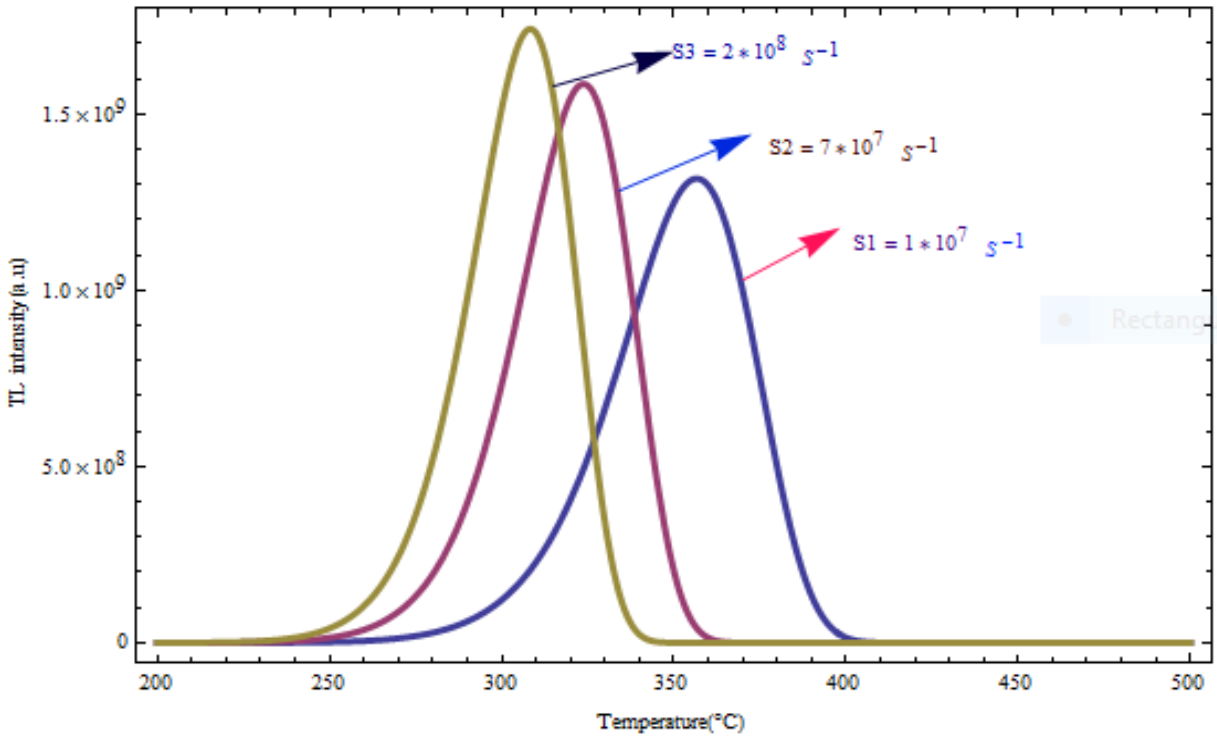

Figure 4.3: The effect of frequency factor on the Thermoluminescence Properties of Eu^{3+}:Y_2O_3 at three different values

lower temperature as 's' is increased. Thus, it is clear that 'E' and 's' have opposite effects on temperature. This is simple to understand since higher 's' means faster escape of the trapped change from the excited state of the trap. Higher frequency factor (s) leads also to increase in peak height. Therefore, Similar changes can be noticed as the parameter 's' is varied but now in the opposite way as frequency factor

(s) increases the peak shifts to lower temperatures with an increase of the height and a decrease in width. A trapping center with a high frequency factor needs less energy (lower temperature) to free a charge carrier.

4.1.4 Effect of heating rate on Thermoluminescence glow curves of Europium doped yttrium oxide nanophosphor

Heating rate is an important parameter for the determination of the various kinetic parameters of Thermoluminescence glow curves. The glow peak height decreases or increases with increasing heating rate. In this study, The Thermoluminescence glow curves intensity of Eu^{3^+}:Y_2O_3 varies at three different value of heating rates. As we expected from the result, the intensity maximum shifted to higher temperatures as the heating rate was increased. Moreover, increase of the glow peak height was observed as increasing heating rate. Change in the linear heating rate (β) results in the change of the increase of the Thermoluminescence glow curve and faster heating rates produce a shift in temperature relatively towards higher values of temperature. Generally, Fig.4.4 shows corresponding Thermoluminescence glow curves of with different heating rate and from the figure we conclude that:-

✓Glow peak height increases with the increase of the heating rate.

✓Peak position shifts toward higher temperature.

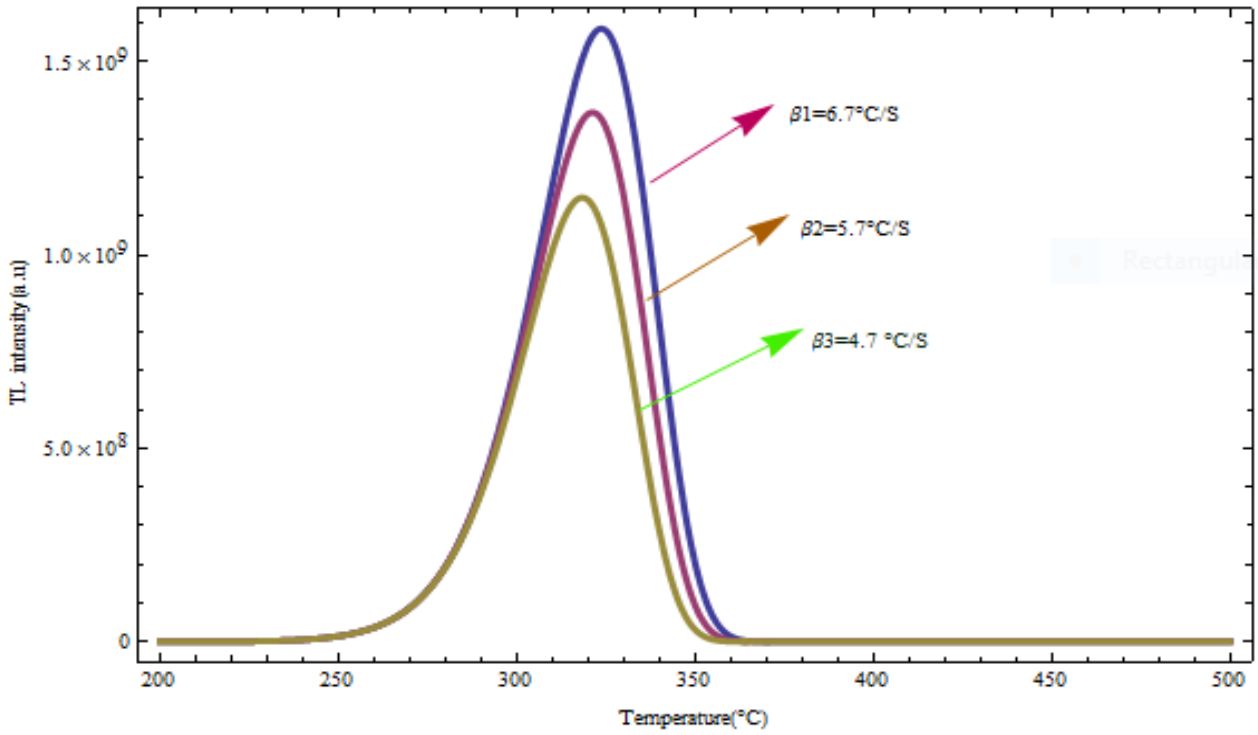

Figure 4.4: The effect of heating rate on the Thermoluminescence properties of Eu^{3+}:Y_2O_3 at three different values
$E = 0.53eV, S = 7 * 10^7 s^{-1}, \beta_1 = 6.7^o cs^{-1}, \beta_2 = 5.7^o cs^{-1}, \beta_3 = 4.7^o cs^{-1}, n_o = 10^{10} cm^{-3}$

4.2 Comparison of theoretical and experimental result on the Thermoluminescence glow curve of Eu^{3+}:Y_2O_3 nanophosphor

When we compare the theoretical and experimental result on the Thermoluminescence glow curve of Eu^{3+}:Y_2O_3:-

✓For given values of 'E' and 'β', the glow peak shifts to lower temperature as 's' is increased. Thus, it is clear that 'E' and 's' have opposite effects on temperature.

✓when the value of the dose was increased then the peak is decreased. This decrease of the Thermoluminescence glow peak can be attributed to the stronger competition with non-radiative centers at higher doses.

✓As we conclude from our work as heating rate increased the TL glow peak of Eu^{3+}:Y_2O_3 was increased and the glow peak is shift to the maximum temperature as we observe from fig.4.4

Chapter 5

Conclusion

In this thesis, the Thermoluminescence kinetic parameters such as activation energy (E), concentration of trapped electron population (n_o), heating rate (β) and frequency factor (s) was used to express the Thermoluminescence glow curve of $Eu^{3^+} : Y_2O_3$. The thermoluminescence intensity increases with an increase in frequency factor (s) and then decreases with increase in activation energy . As frequency factor (s) increases the peak shifts to lower temperatures with an increase of the height and a decrease in width. A trapping center with a high frequency factor needs less energy or it needs lower temperature to free a charge carrier. When the initial concentration of electron in traps (n_o) changed the peak characteristics are independent of radiation dose given to the sample. Also the vertical axis represents the trapped electron population in traps versus temperature . The concentration of trapped electrons population decreases as the temperature increases just above 270^oC, reaching its minimum values approximately between 345^o_C and 350^oC and at temperature of $150^oC - 270^oC$ the traped electron is not released from the center of trap because the temperature is not enough to eject it, but above the temperature of 270^oC the traped electron start to excite from trap center.

Therefore, Similar changes can be noticed as the parameter (s) is varied but now in the opposite way: As frequency factor (s) increases the peak shifts to lower temperatures with an increase of the height and a decrease in width. A trapping center with a high frequency factor needs less energy or it needs lower temperature to free a charge carrier.

Generally, The appearance of a peak in the Thermoluminescence intensity (glow peak) indicates the presence of a charge carrier trap in the material. The temperature at which this peak is located is a measure for the trap depth, since it is the temperature

at which enough thermal energy is available for the trapped charge carriers to be released and recombine at luminescent centers. The shape, height, and location of the peak all contain information on the number and depth of the traps.

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JIMMA UNIVERSITY
COLLEGE OF NATURAL SCIENCES
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Graduate Program: **Summer, M.Sc.**

1. Course Work Performance

Course Code	Course Title	Cr. hr	Number Grade	Rank **	Remark
Phys699	**M.Sc. Thesis**	**6**			

** Ecellent, Very Good, Good, Satisfactory, Fail.

Thesis Title

Thermoluminescence and defect centers in Europium doped yttrium oxide nanophosphor

2. Board of Examiners decision Mark × in one of the boxes. Pass [×] Failed [ ]

   If failed, give reasons and indicate plans for re-examination.

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3. Approved by: Name and Signature of members of the examining Board, and Department Head

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External Examiner	_____	_____	_____
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DECLARATION

I here by declare that this M.Sc thesis is my original work and has not been presented for a degree in any other University and that all source of materials used for the dissertation have been duly acknowledged.

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