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M.Sc in Engineering Physics

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**UNIVERSITY OF GAZIANTEP
GRADUATE SCHOOL OF
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**INVESTIGATION OF THERMOLUMINESCENCE RESPONSE
OF TLD-600 AND TLD-700 DOSIMETERS TO
NEUTRON+GAMMA AND BETA RADIATIONS**

**M.SC THESIS
IN
ENGINEERING PHYSICS**

**BY
BÜŞRA YAZICI
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**Investigation of Thermoluminescence Response of TLD-600 and
TLD-700 Dosimeters to Neutron+Gamma and Beta Radiations**

**Master Thesis
In
Engineering Physics
University of Gaziantep**

Supervisor

Assoc.Prof.Dr. Vural Emir KAFADAR

By

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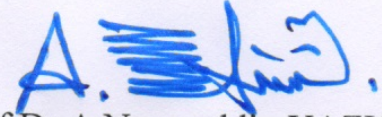
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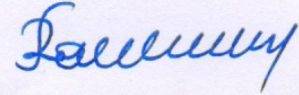
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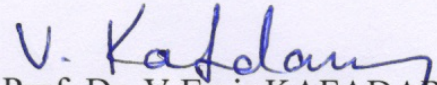
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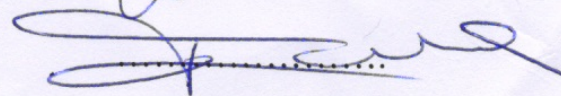
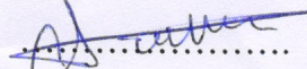
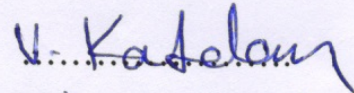
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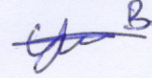
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A handwritten signature in blue ink, appearing to be 'Büşra', with a stylized flourish at the end.

ABSTRACT

INVESTIGATION OF THERMOLUMINESCENCE RESPONSE OF TLD-600 AND TLD-700 DOSIMETERS TO NEUTRON+GAMMA AND BETA RADIATIONS

YAZICI, Büşra

M.Sc. in Physics Engineering

Supervisor: Assoc.Prof.Dr. Vural Emir KAFADAR

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In this study some of the thermoluminescent properties such as glow curve structure, relative thermoluminescence sensitivity, dose response linearity of lithium fluoride thermoluminescent dosimeters TLD-600 ($^6\text{LiF: Ti, Mg}$) and TLD-700 ($^7\text{LiF: Ti, Mg}$) were investigated after irradiation ^{252}Cf neutron+gamma and ^{90}Sr - ^{90}Y beta sources at room temperature and then with obtained results were compared. The kinetic parameters of TL glow peaks namely the order of kinetics (b), activation energy (E_a) and the frequency factor (s) of these dosimeters have also been obtained using the computerized glow curve deconvolution (CGCD) program. In addition, the effect of heating rate on the TL glow curves of dosimeters has been investigated. They were found that the maximum TL glow peak intensities and the total area under the glow curves of both dosimetries are decreasing with increasing heating rate. There is no good agreement with the kinetic parameters calculated by CGCD program for both radiation sources.

Keywords: TLD-600, TLD-700, kinetic parameters, neutron dosimetry

ÖZET

TLD-600 ve TLD-700 DOZİMETRELERİNİN NÖTRON+GAMAVE BETA RADYASYONLARINA KARŞI TERMOLUMİNESANS DAVRANIŞLARININ ARAŞTIRILMASI

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Bu çalışmada ^{252}Cf nötron+gama ve ^{90}Sr - ^{90}Y beta radyasyon kaynakları kullanarak TLD-600 ($^6\text{LiF:Ti,Mg}$) ve TLD-700 ($^7\text{LiF:Ti,Mg}$) lityum florür termoluminesans dozimetrelerinin ışıma eğrisi, termoluminesans duyarlılığı, doz-cevap ilişkisi gibi bazı termoluminesans özellikleri araştırılmış ve elde edilen sonuçlar karşılaştırılmıştır. Bu dozimetrelerin TL ışıma tepelerinin aktivasyon enerjisi (E), frekans faktörü (s), kinetik derece (b) gibi tuzak parametreleri bilgisayar ile ışıldama eğrisi ayrışım (CGCD) metodu kullanılarak bulunmuştur. Ayrıca, ısıtma hızının her iki dozimetrenin TL ışıldama eğrileri üzerine etkisi araştırılmış ve ısıtma hızı arttıkça her iki dozimetrenin de TL ışıldama eğrilerinin altında kalan alan ve maksimum ışıldama tepesi şiddetlerinin azaldığı görülmüştür. Her iki radyasyon kaynağı ile ışılandıktan sonra CGCD programı ile bulunan kinetik parametrelerde uyuma görülmemiştir.

Anahtar Kelimeler: TLD-600, TLD-700, kinetik parametreler, nötron dozimetresi



I dedicate this work to my family....

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LIST OF SYMBOLS

A_h	: Recombination transition coefficient for electrons in the conduction band with holes in centers
A_n	: Transition coefficient for electrons in the conduction band becoming trapped (volume/until time $m^3 sec^{-1}$)
A_m	: Transition coefficient for holes in the valance band becoming trapped in the hole centers
b	: Kinetic order
β	: Heating rate
CGCD	: Computer Glow Curve Deconvolution
E or E_a	: Activation energy (trap depth)
E_c	: Bottom of the conduction band
E_f	: Energy Fermi level
E_g	: Band gap
E_v	: Top of the valance band
ET	: Electron Traps
FOM	: Figure of Merit
HT	: Hole Traps
I	: Thermoluminescence intensity
I_m	: Peak intensity
k	: Boltzman's Constant
N	: Concentration of available electron traps in the crystal (in m^{-3})
n	: Concentration of the filled electron traps in the crystal (in m^{-3})
n_0	: Number of trapped electrons at the initial time $t_0 = 0$
n_c	: Concentration of electrons in the conduction band (per unit volume m^{-3})
N_h	: Concentration of available hole centers
n_h	: Concentration of holes in the recombination centers
n_v	: Concentration of holes in the valance band
m	: Concentration of holes trapped at R (m^{-3})
p	: Probability per unit time of release of an electron from the trap
R	: Recombination centers
s	: frequency factor
T	: Temperature
T_m	: Peak temperature
TL	: Termoluminescence
TLD	: Thermoluminescence Dosimetry

CHAPTER 1

INTRODUCTION

Luminescence refers to emission of radiation in visible or near visible region subsequent to absorption of energy by a substance (excitation). Based on the characteristic time scale (τ) between the excitation and emission of radiation, luminescence can be broadly classified into two types, viz., fluorescence and phosphorescence. The time scales involved in fluorescence are very small of the order of 10^{-8} s, which is almost same as relaxation time of an isolated gaseous ion, whereas for phosphorescence it is in milliseconds. In the case of phosphorescence, the emission occurs even after the excitation has ceased and is also termed as afterglow. The increase in decay time for phosphorescence is because the transition leading to emission of light proceeds via a metastable state. The source of excitation is used as a prefix to luminescence for further classification of luminescence such as photoluminescence if the source of excitation is electromagnetic radiation and chemiluminescence if the source of excitation is chemical reactions and so on. Thermoluminescence (TL) differs from other luminescent phenomena in the sense that the prefix is not a source of excitation, but is a source of stimulation. Explicitly, TL is also referred to as radiothermostimulated luminescence, where the source of excitation is ionizing radiation and thermal energy is used to stimulate the emission [1].

Although the phenomenon of thermoluminescence (TL) was known for many decades, the basic mathematical understanding was first provided by Randall and Wilkins only in 1945 [2] and its applications as a research tool was proposed subsequently in 1953 by Farrington Daniels [3]. There had been considerable efforts in trying to develop enough understanding of the phenomenon both mathematically and physically and these reports are scattered over a spectrum of scientific journals. It is worth mentioning here that TL is perhaps a unique physical phenomenon that

has embraced lot more scientific disciplines than any other process known today; yet there has been no single effort to present all its aspects together in a place.

Thermoluminescence (TL) is a misnomer in the conventional sense of the names of luminescence processes like cathodoluminescence, chemiluminescence, electro-luminescence and bioluminescence. The excitation is achieved by any type of ionizing radiation. It is also necessary to distinguish clearly between thermoluminescence and incandescence emissions from a material being heated. The luminescence usually lies in a spectral region where the material is non absorbing; the incandescence emissivity is strongest where absorptivity of the material is the maximum. There is also a wide gap between the temperatures of occurrences of these two emissions, incandescence occurring at very high temperatures near the melting point of the respective material. The highest temperature at which TL has been reported for any solid is around 650°C. Normally it is understood that no TL can be observed at temperatures far lower than the excitation temperature, there are however exceptions and these are distinguished as cryo-luminescence and cooling-down luminescence. It is heartening to note that in recent times, the phenomenon of TL is being correctly referred as thermally stimulated luminescence (TSL). The phenomenon of TL is usually qualitatively explained with the aid of the band picture of the solid with respect to its electronic energy levels. The forbidden band gap can be imagined to contain some acceptor/donor metastable levels which are basically responsible for the observed TL [4,5].

The ionizing radiations (X-ray, α , β , γ , n) are very harmful on the living organism and people. The amount of biological damage is dependent on the energy of the absorbed radiations and the amount of absorbed dose. The first and the most important method for radiation protection is stayed as minimum as to exposed ionizing radiation. Therefore, the first rule is to prevent peoples from radiation and radioactive contamination. Second rule is that it is very important to measure the amount of ionizing radiation that is absorbed by people [6]. For recording TL, the material exposed to ionizing radiation is heated at constant rate. The intensity of emitted light, referred to as TL intensity is a function of temperature (time), the resulting curve is termed as glow curve. A typical glow curve consists of one or more isolated/overlapping peaks. Glow curve also depends on the light sensitive detector

used to record spectrum and its spectral characteristics. A peak in the glow curve, preferably isolated or well resolved is considered as a dosimetric peak. In TL dosimetry, the TL intensity of the dosimetric peak is directly proportional to the absorbed dose. Thus, by a suitable calibration of dosimeter, unknown doses absorbed by the material can be estimated by measuring TL. For several decades, Thermoluminescent detectors (TLDs) have been widely used in medical, personnel and environmental dosimetry [7]. For dosimetric applications a phosphor should qualify the following important properties. A schematic description of thermoluminescence is presented in figure 1.1. The occurrence of TL following an initial excitation is a 'one shot' effect. Cooling the sample and re-heating it normally does not result in TL emission.

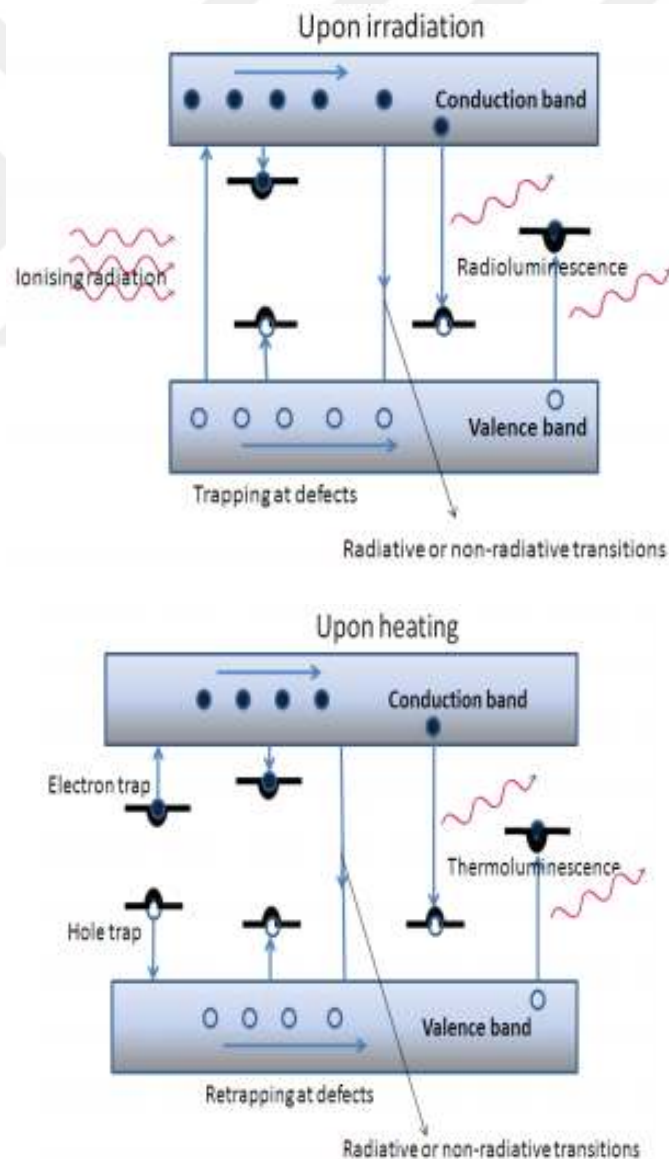


Figure 1.1 Illustration of a simple model for thermoluminescence [8].

Linear Dose response: This is one of the most desirable properties for a phosphor to qualify as a TL dosimeter. That is the phosphor should exhibit a linear relationship between TL output (either in terms of the area under the peak or peak height) and absorbed dose (D). TL output as a function of absorbed dose is known as dose response curve, which linear only over a range of doses. The linear dose response is quantified in terms of dose response function (or supralinearity index) $f(D)$ which is defined as,

$$f(D) = [S(D) / D] / [S(D_1) / D_1] \quad (1.1)$$

Here $f(D)$ is the TL intensity (defined, for the purpose of this work, as the TL peak height) at dose D and $S(D_1)$ is the TL intensity in the linear range of $f(D)$, at dose D_1 . For an ideal dosimeter $f(D)=1$. For a phosphor to be an ideal dosimeter, $f(D = 1)$ is observed only in a very narrow range in most of the TL phosphors. Supralinearity and sublinearity are terms used to describe nonlinear dose response corresponding to $f(D > 1)$ and $f(D < 1)$ respectively. The dose response function, $f(D)$, depends on dose, dose rate, thermal history of the phosphor, type and energy of the radiation field in which the phosphor is irradiated as well as the heating procedure during measurement.

Sensitivity: Sensitivity of any TL phosphor in general is the TL signal strength per unit absorbed dose. This absolute definition of sensitivity is practically inadequate because the TL signal depends on many instrumental parameters used in the TL readout system, heating rate used, and method of measurement (peak height or area under the peak) and so on. Hence, in order to overcome these experimental difficulties, one normally defines what is called as the relative sensitivity by comparing the TL signal of the phosphor of interest with that of the standard phosphor (normally TLD-100 or $\text{CaSO}_4:\text{Dy}$ are used as standard). The strength of TL signals at dose D of the phosphor which is under test and the standard phosphor measured using the same measurement system. This relative sensitivity also depends on dose, dose rate, Linear Energy Transfer (LET), and heating rate.

Fading: Fading is a loss of TL signal with time. It may be caused either due to thermally or optically stimulated release of the electrons or a combination of both.

High thermal fading is observed when the glow curve contains low temperature peaks. If the trap depth E is too small then severe fading of the signal will occur, both during irradiation and between irradiation and readout. Phosphors, which are to be used for dosimetry purposes should possess a glow curve with a dosimetric peak in the range 200°C - 250°C. Usually in this temperature range the trap depth is larger and no appreciable trap emptying can take place.

Thermoluminescence dosimetry is the widely used means of dosimetric measurements especially for personnel monitoring of radiation workers. The materials used for personnel monitoring should meet several requirements like high sensitivity, low fading, and linear response over a wide range of dose and no energy dependence. A single material may not satisfy all the requirements for personnel dosimetry. The field evolves by the development of new materials or improving the characteristics of existing phosphors by using different synthesis technique or by changing the dopants. Thermoluminescence is exhibited by most of the inorganic compounds. However, to use a phosphor for dosimetric applications, it should qualify some properties which limit the choice to only a handful of materials [9].

A radiation dosimeter is an instrument which measures the radiation doses i.e. exposure, kerma, absorbed dose or equivalent dose directly or indirectly [6]. One of the indirect techniques (passive methods) to measure the received dose level by the people, especially employees in the radiation fields, is to use thermoluminescent dosimeters (TLDs) [4,5]. A TL dosimeter along with its reader which is called as TLD reader is referred to as a TL dosimetry system. In TLDs, the energy of ionizing radiations is stored in the crystal defects. This energy is then released back to outside of crystals as a visible photon with heating of them. Then the emitted visible photons are measured by a reader (TL reader). The resulting curve is called as TL glow curve. It includes emitted light intensity from TL dosimeter with respect to temperature. The TL intensity is recorded for applied dose to obtain TL intensity versus dose relationship of each dosimeter. Neutron dosimetric materials have been developed at the standards set by the International Atomic Energy Agency. The main features that the International Atomic Energy Agency (IAEA) identified and recommended were considered [10]:

- The peak temperature (T_m) of the glow curve of the thermoluminescence dosimetry should be between 180 and 250 °C, and possibly a single peak
- The dose-response curve of dosimetry should be linear over a wide dose interval
- Almost no influence from environmental factors
- The wavelength of the emitted light from the dosimeter must be within the visible region boundaries
- Do not change the sensitivity of dosimetry on reuse
- The sensitivity of the dosimetry should not be too dependent on the energy of the applied radiation and the type
- The radiation absorption factor of the dosimetry is the same as the radiation absorption factor of the human body ($Z_{\text{eff}} = 7.42$)
- The dosimeter should be cheap and easily prepared in almost any laboratory environment
- There should not be toxic materials in the dosimetry that could harm the human

In 2007, Triolo *et al*[11] studied the thermoluminescent glow curves of TLD-600, TLD-700 and MCP for mixed area of n and γ . The flux rates of thermal neutrons used to explain the confidently of the thermoluminescent dosimeters in this radiation area and to get knowledge on the dose absorbed rates. The glow curves found have been CGCD (compurized glow curve deconvoluted) program using general order kinetics and the determined variations for the difference linear energy transfer components have been inspected. Yukihaera *et al*[12] explained the probability of using neutron converters consolidated into $\text{Al}_2\text{O}_3\text{:C}$ dosimeters to generate new neutron accurate optically stimulated luminescence dosimeters proper for personal monitoring. A comparison between different neutron converters show that lithium-based compounds result in the best neutron susceptibility, likely due to the comparatively big range of the 2.75 MeV triton manufactured by the ${}^6\text{Li} (n, \alpha){}^3\text{H}$ neutron capture reaction. With these results, neutron susceptible optically stimulated luminescence material for dosimetry systems was manufactured and characterized except for the smaller gamma and enhanced neutron susceptibility. The gamma susceptibility of the new material is $\sim 35\%$ because $\text{Al}_2\text{O}_3\text{:C}$ composition is decreased in the material. The neutron susceptibility performed in this new material

is ~60% of the neutron susceptibility of TLD-600 ($^6\text{LiF:Mg,Ti}$) [13]. Horowitz *et al.* reports in commonly accepted thermoluminescence response of fast neutrons to gamma between 0.05 to 0.3 [14]. A new thermoluminescence dosimeter sample of LiF:Cu,Mg,P , with a γ irradiation thermoluminescence susceptibility ~20 times larger than TLD-100 (LiF:Mg,Ti) is reported. The thermoluminescence response of LiF:Cu,Mg,P to californium-252 (^{252}Cf) (a fast neutron source with an average energy of $E = 2.5 \text{ MeV}$) is measured. Supposition of the thermoluminescence signal was performed using CGCD program [14]. Mittani *et al.* reports the OSL (optically stimulated luminescence) features of neutron dosimeters which is a mixture of C doped Al_2O_3 and neutron converters [15]. The neutron converters researched were maximum density polyethylene, lithium fluoride, LiF 95% enriched with ^6Li , Li_2CO_3 95% enriched with ^6Li , $\text{H}_3^{10}\text{BO}_3$ enriched with 99% of ^{10}B and Gd_2O_3 [16]. The ratio of C-doped Al_2O_3 and neutron converter in the mixture was optimised to minimize the whole optically stimulated luminescence signal and neutron susceptibility. The neutron susceptibility and dose return were reported for the optically stimulated luminescence dosimeters using a bare californium-252 neutron source and similar to the return of Harshaw neutron dosimeters $^6\text{LiF:Mg,Ti}$ and $^7\text{LiF:Mg,Ti}$ [17]. The study indicates the probability of improving an optically stimulated luminescence dosimeter consist of C doped Al_2O_3 sample and neutron converter with a neutron susceptibility and neutron gamma separation similar to the $^6\text{LiF:Mg,Ti}$ and $^7\text{LiF:Mg,Ti}$ combination [17].

Dose can be defined as the amount of accumulated irradiation incident on the sample at low temperature prior to the heating. Indeed, a sample does not have to absorb all incident irradiation during exposure [14]. Dose is simple calculated by the following formula.

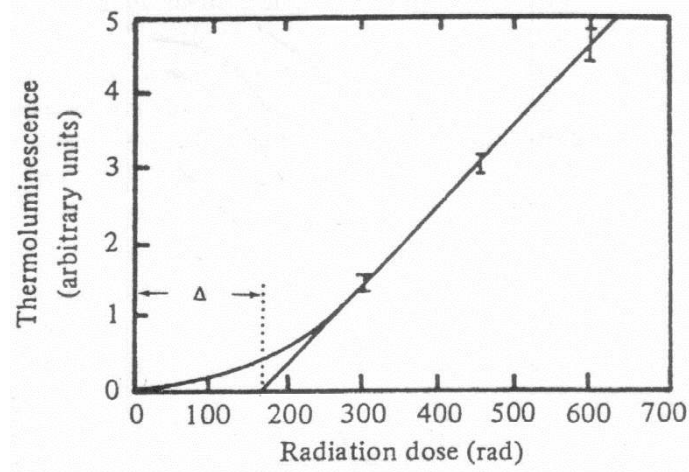
$$D = X.t \quad (1.2)$$

where X is dose rate which is incident ration per unit time and t is the excitation time. Total dose can be found as;

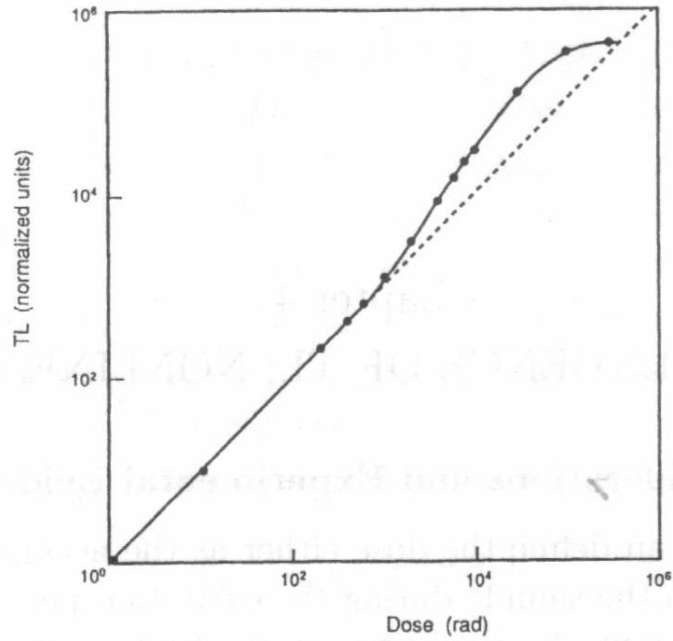
$$D = \int_0^t X(t).dt \quad (1.3)$$

In theoretically, it is expected that the dose and TL intensity graph is linear, but in many materials degrees of non-linearity are seen. In general, there are the two

types of non-linear dose dependence [14]. They are sublinear and superlinear (supralinear) which are shown in below figure 1.2.



(a)



(b)

Figure 1.2 Dose dependence of the TL from (a) pottery quartz, (b) LiF [1].

As seen from figure 1.2(a), the dose dependence of pottery quartz is not linear at low doses up to approximately 250 rad and then becomes linear at higher doses. On the other hand, the characteristic behavior of LiF (see fig.1.2(b)) is that it is initially linear dose dependence, followed by a “supralinearity” region before saturation level is observed at higher dose levels. Non-linear growth of TL as a function of the absorbed doses has been detail discussed in some books [1, 4-5, 7]. Once TL dosimetry techniques and the characteristics of the dosimeters are well

understood, it has become clear that TL dosimeters can be conveniently used in personnel dosimetry services. Previous experiences have been shown that all of the kinds of ionizing and non-ionizing (UV) radiation doses are measured with TLDs according to the type of used crystal. In addition to being used as a personal dosimetry, TL dosimeters are widely used in many areas, such as in the determination of age of archaeological samples, in radiotherapy and in food irradiation because of their linear relationship between dose and thermoluminescence signals, their stability in room temperature after irradiation for months, their tissue equivalence properties [1]. There are many researching groups in the developed countries studying on dosimetry studies using the TL techniques, i.e., Oxford University in the UK, Oklahoma State University in the United States, Tel-Aviv University in Israel, and Radiation Research Department in the Risø National Laboratory in Denmark. In Turkey, radiation dose evaluation using TLDs is carried out only at the ANAEM. In order to be useful, TLDs must exhibit several desirable characteristics [18]. For a suitable radiation dosimeter purposes, TL dosimeters is expected to show relatively simple glow curve structure and the peak temperature (T_m) of the glow curve of the TLD should be between 180 and 250 °C and possibly a single peak. At higher temperature, the infrared emission from the hot sample and sample holder increasingly interferes with the measurement of low doses. TL dosimeters should be high sensitive, which consist of both high efficiencies of light emission and low threshold dose. The sensitivity of dosimetry should not change after re-used. Another desirable property of TLD materials includes a linear response to absorbed dose and an adequate sensitivity. The dose-response curve of dosimetry should be linear over a wide dose interval. However, the dose response of most TL materials is far from linear, which is displaying a variety of non-linear effects depending upon the dose range and irradiation. Almost no influence from environmental factors such as humidity, solvents, light long term stability of stored information at room temperature. A sufficient storage stability of charge carriers in traps to cause no desirable fading even during extended storage at ambient or slightly increased temperatures. Low photon energy dependence of response. The sensitivity of the dosimetry should not be too dependent on the energy of the applied radiation and the type. Depending on the particular use, high or low thermal neutron sensitivity or a certain LET response may also be the desirable characteristic. Also a good TLD should be tissue equivalent having an effective atomic number of nearly $Z_{\text{eff}}=7.42$.

The phosphor should not be very expensive. The dosimeter should be cheap and it may be advantageous if the material can easily be prepared with reproducible properties in a normally equipped chemical laboratory. It should not be toxic that could be harmful for the human and it should be possible to use a small sample only. The wavelength of the emitted light from the dosimeter must be within the visible region boundaries and it should be matched with the high wavelength response of photomultiplier tube used within the TLD reader.

In fact, only a few of materials (CaF_2 and LiF) match to these properties. The materials based on calcium fluoride (CaF_2) with various dopants are well known, highly sensitive dosimeters for personal and environmental monitoring. [19-21]. The synthetic dosimeters have been produced by Harshaw/Bicron Company with the well-known names TLD-200, TLD-300 and TLD-400. The dosimetric characteristics of commercially produced CaF_2 dosimeters have been studied and reported by various authors [19-21]. These dosimeters possess a high sensitivity, wide linearity of dose response, low thermal fading, good chemical stability, and reusability. One of the bad feature of these materials is that their high-energy dependence. However, the high energy dependence of the TL response for exposures to photon radiation can be minimised by the use of appropriate filters. Another interesting feature of these dosimeters is the UV and neutron sensitivity. Especially, neutron dosimetry is an important aspect of radiation protection. Because of the higher biological effectiveness of neutrons than that of beta and gamma radiations [19-22]. LiF based TL Dosimeters are the most used passive detectors in many places such as in industry and medicine to observe the accumulated dose in any radiation field. Since these dosimeters have small dimensions and they are tissue equivalent materials [1].

Neutrons have no charge, are not pushing back if they closing nuclei, and could essentially go into nuclei. It is for this cause that they are of large significant in nuclear physics when you same, they are the blade with which the scientists can nudge about in the nucleus. Because of neutrons are electrically neutral, they reason no ionisation immediately, and thus they could travel large distant whereby substance, more same gamma rays. The most important feature for producing neutrons is that they have to be ejected out of the nucleus. They arise from the nucleus with a large velocity, having energies of up to a few MeV. Majority of these

neutrons are decelerated by collisions with nuclei. Later maybe a hundred of as collisions, the velocity of the neutrons is lower to as a rate that they have concerning the like kinetic energy as the particles of their around. The kinetic energy of these particles is fundamental the temperature of the sample they constitute. For this reason, the neutrons that are slowed down to the similar energy level of the surrounding particles are called thermal neutrons. They are useful for triggering fission reaction. Thermal neutrons continue to drift until they are captured by a particular nucleus [22]. The route in which neutrons interact with substance depends to a major range on their energies. For this cause, neutrons are frequently definite by energy as the under mentioned:

Fast Neutrons - above 8 keV

Slow Neutrons - below 8 keV

Thermal Neutrons - about 0.025 eV

Although the thermal neutrons have as little energy as possible, thermal neutrons should not have a residual effect. They still exists and travel at speeds of 2.2 km/s [22].

Layout of Thesis

This thesis consists of 5 chapters arranged as following:

- Chapter 1 introduces the context and contains historical subject matter, and the other two sections devoted to a description of luminescence and thermoluminescence.
- Chapter 2 separated into two sections: First, thermoluminescence models which described in detail. Second, involved three methods to find out trap parameters.
- Chapter 3 includes the material synthesis and doping procedures, equipment's, radiation source with a brief characterize of irradiation procedure.
- Chapter 4 presents the results of the study of the experiments stated in Chapter 3 with some determination of trap parameters. These results include the outputs from characterization analysis as well as thermoluminescence measurements.
- Chapter 5 discusses the main findings of this study and recommendations.

CHAPTER 2

THERMOLUMINESCENCE THEORY

2.1 Luminescence

Luminescence is a emission of light phenomenon from insulators or wide band-gap semiconductors by excitation energy. Light is a form of electromagnetic wave and it has energy. The luminescence is the emission of optical radiation from matter in the visible, ultraviolet and infrared regions. The emitted wavelength of the light is the characteristic of the material. The luminescence phenomenon should be separated from incandescence phenomena. In the incandescence phenomena, the optical radiation emitted from the material due to temperature (blackbody radiation). The luminescence does not contain the emission of blackbody radiation light. The luminescence mechanism includes two steps. The one step includes the excitation of electrons to higher energy state and the second step includes the subsequent emission of photons from the materials [23].

In solids, there are different luminescence types which are named according to their simulation mechanism causing light emission. Some of them are: bioluminescence stimulated by biochemical energy, sonoluminescence stimulated by sound waves, electroluminescence stimulated electrical energy, triboluminescence stimulated by mechanical energy, cathadoluminescence stimulated by the electron beam, radioluminescence stimulated by ionizing radiation. However, the famous of them are: optically stimulated luminescence (photoluminescence) stimulated by photons and thermally stimulated luminescence (thermoluminescence) stimulated by heat. In the luminescence phenomena, if the delay time between excitation and emission is longer than 10^{-8} sec, it is called as phosphorescence. If it is shorter than 10^{-8} sec, it is called as fluorescence [1, 23].

2.2 Basic Concepts of Thermoluminescence in Solids

The repeated arrangement of atoms in solids forms crystal structures. The crystals are not always perfect structures. Many times, they contain various types of imperfections and defects within them.

These defects interrupt and break the regular patterns of crystal structures. There are many forms of crystal defects. The most important of them is the point defects which plays very important roles in the luminescence phenomena, especially in the thermoluminescence mechanisms. A point defect within the crystal structure is designated as a trap center if it is able to capture a charge carrier and release it back to the band by a stimulation process. A negative ion vacancy acts as an electron trap which may be created by the dislocation of a negative ion. On the other hand, if defect is created by positive ion vacancy, it acts as a hole trap. When a crystal defect captured an opposite sign carrier which is previously trapped by the defect, it results recombination of an electron-hole. This type of defects is classified as a recombination center. If it emits light after this recombination, it is called as luminescent center. It should be noted that non-radiative recombination is also possible [1].

If the energy level of a defect is between Conduction Band (CB) and electron demarcation level, it is called as Electron Trap (ET) (see figure 2.1). But, the level of energy of a defect lies between Fermi energy level and hole demarcation level, it is called as Recombination Center (RC). Point defects can be produced in a crystal by a variety of techniques. The most important type of defect production techniques within the crystals is to utilize the high ionizing radiations. The term ionizing radiation includes all sources that can generate free electrons-holes pair in the crystal. The energy ranges of these radiations are from 10 eV up to high energy ultraviolet photons. However, these high energetic ionizing radiations have the capability to generate point defects in the crystals [1].

The band model describes the localized and delocalized energy levels of crystal structures. It also aids to explain absorption and storage of ionizing radiation energy and then emission of stored energy via photons after the stimulation of crystal with heat or photons. In the band model, absorption of radiation energy creates

electron-hole pairs. The point defects will be able to capture these liberated electrons and holes due to the irradiation process. Thereupon, the presence of point defects such as vacancies and impurities within the crystal store energy and then emit by different stimulation techniques. Figure 2.1 shows the band diagram and typical charge transitions between the bands and traps within the band gap of crystal.

Band gap energy is the energy difference between and conduction band and the valence band (E_g). The excited electrons are freely moving in the CB until they are captured by an electron trap center ET_1 or ET_2 . Similarly, the liberated holes are moved in the VB until they are captured into either a hole trap (HT) or a recombination center R.

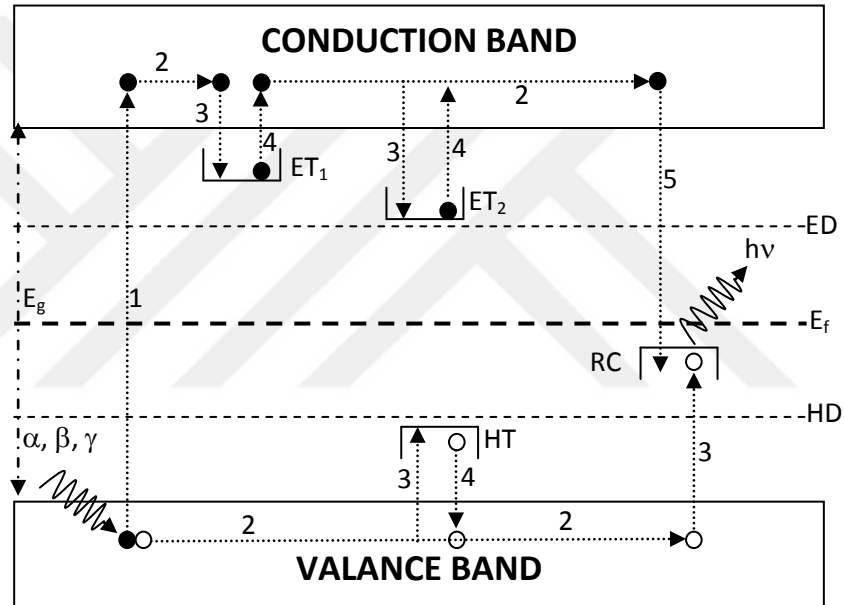


Figure 2.1 Energy band model of solids to show the charge transitions in crystal. (1) generation of electrons-holes; (2) electron and hole migration in their bands; (3) electron and hole trapping; (4) electron and hole release due to thermal stimulation; (5) recombination of electrons with holes in recombination center. (●) shows electrons, (○) shows holes. Level ET_1 and ET_2 are electron traps, HT is hole trap, RC is a recombination centre, E_f is Fermi level, E_g is band gap energy, E_D is electron demarcation level, E_H is the hole demarcation level, α, β, γ are the ionizing radiations and $h\nu$ is the emitted photon.

If the crystals are stimulated by anyone technique, i.e. heat, photon, electrical energies, etc., the captured electron in ET gains energy and can be freed into the conduction band if its energy is enough to pass CB. The trap depth or activation energy E is an energy level between ET and CB. The stimulation of the crystal lattice structure by heating up will excite the electrons out of the traps. The escape

probability of an electron from a trap can be calculated by the Arrhenius equation [23].

$$p = \frac{1}{\tau} = s \exp \left\{ -\frac{E}{kT} \right\} \quad (2.1)$$

where p is the escape probability, τ is lifetime of an electron in trap, E is the depth of trap (eV), k is Boltzmann's constant $= 8.617 \times 10^{-5}$ eV/K, and T is the temperature, s is the attempt to frequency. The frequency factor (s) is assumed to be constant in the simple model, but it can be depending upon the temperature. At the temperature of irradiation T_0 , if $E \gg kT_0$ trapped electrons may remain for a long time in traps. As it is seen in above equation, the escape probability is increased when the temperature of the TL material is increasing above T_0 , or increasing with frequency factor, or decreasing trap depth.

Once electrons are expelled from their respective traps, they diffuse through the crystal lattice until they come across another site that is attractive to electrons and these are referred to as recombination centers. And then it makes a transition to recombination center R. In general, a photon emits after this recombination. If the stimulation energy to the crystal is given by heat process, it is termed thermally stimulated luminescence or simple (TL). Alternatively, if the stimulation energy is given by external light exposure (photon), it is called as optically stimulated luminescence (OSL). The liberated charge carriers in the CB/VB can be measured as electrical current signal when applying voltage across the crystal. These measurements are termed Thermally Stimulated Conductivity (TSC) or optically Stimulated Conductivity (OSC), respectively [23].

The thermoluminescence (TL) phenomena could be observed at insulator and wide bang gap semiconductor materials. Because, the material should have necessary a bang gap to observe TL phenomena. The TL phenomenon in solids is a comparatively complex process. In general, the materials in thermoluminescent dosimetry (TLD) are insulators. The TL materials cannot re-emit the TL light by simply cooling and reheating it without irradiation. To re-exhibit thermoluminescence signal, the material must be re-exposed to ionizing radiation, where upon increasing the temperature will once again produce light emission.

If a wide band gap material is exposed to ionizing radiations at low temperature, the electrons in the valance band are liberated from the VB to the CB (see figure 2.1). That leaves a hole in the valance band. Both types of charge carriers, electrons and holes, become mobile in their respective bands until they recombine with each other or they are trapped in lattice defects in the crystalline solids. These crystal defects have considerable effects in the thermoluminescence process. The trapped electrons could remain therein for a long time when the crystals are kept at low temperatures. If the temperature of the crystal is increased, the trapped electrons will be expelled by thermal vibrations of the lattice (see figure 2.2). These vibrations will be stronger, and the probability of removing increases if the temperature is raised. The liberated electrons pass to conduction band and they may move in this band in crystal solid till they recombine with suitable recombination centers that contain hole with the emission of TL light. The process of light emission from a crystalline solid by increasing the temperature of it with thermal stimulation after irradiation by ionizing radiations is called as TL. To understand TL phenomena, a simplified band diagram is shown in Figure 2.2. If the emitted light intensity is plotted versus time or temperature; this graph is called as the glow-curve (GC) which shows TL peaks occurring at different temperatures [1]. The peak temperature is related with trap depth (E) of electron traps present within the sample, attempt to escape frequency (s), kinetic order (b) and heating rate (β). For a constant rate of heating, the glow curves versus temperature or time are equivalent to each other. The kinetic order is generally obtained $b=1$, or $b=2$ or between them. When the value of $b=1$, it corresponds to a situation where all electrons in the CB recombination with holes. If $b=2$; the electrons in the CB has the same change to recombine with a hole in the recombination center or retrapping by electron traps. When kinetic order is between 1 and 2, it is likely to occur that retrapping and recombination probabilities are different from each other [1].

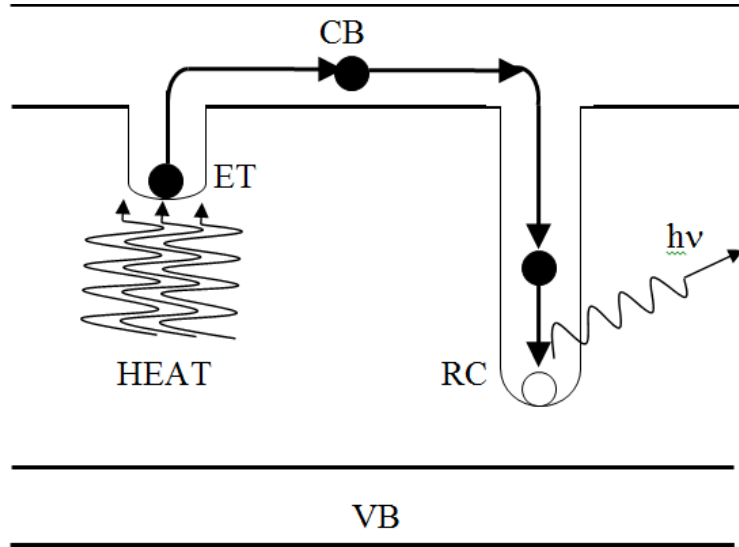


Figure 2.2 Phenomena of thermal excitation of luminescence

The simplest model to describe the physical processes of TL is a single type of recombination center and a trap and model. This model is called as One Trap One Recombination Center (OTOR) model [7]. The analytical form of a TL peak can be described by its the peak position (T_m), the overall peak width and the peak height I_m which depends on the rate of heating and absorbed doses. Its value increases when the absorbed doses and heating rate increased for given experimental conditions.

2.3 Thermoluminescence Kinetic Orders

The first order kinetics is the simplest one which describes the TL process given by Randall and Wilkins [2]. According to them, a TL peak occurs because of the thermal release of electrons from traps and their subsequent recombination with holes trapped in the recombination centers. The assumption that the entities involved are electron traps and hole centers is for the sake of simplicity and the opposite is just as likely to occur with no further complication in the mathematical treatment. This assumes: *(i)* There is one trapping and one recombination center *(ii)* There is no retrapping *(iii)* Electrons cannot accumulate in the conduction band. By assuming negligible retrapping during the heating stage ($mA \gg (N-n)A_r$);

$$I(t) = -\frac{dn}{dt} = sn \exp\left\{-\frac{E}{kT}\right\} \quad (2.2)$$

The solution of equation (2.2) yields the first-order expression of a single glow peak as;

$$I(T) = n_o \frac{s}{\beta} \exp\left\{-\frac{E}{kT}\right\} \exp\left\{-\frac{s}{\beta} \int_{T_o}^T \exp\left\{-\frac{E}{kT'}\right\} dT'\right\} \quad (2.3)$$

One of the most important features of first-order TL kinetics is that T_m is constant as a function of doses (see figure 2.3).

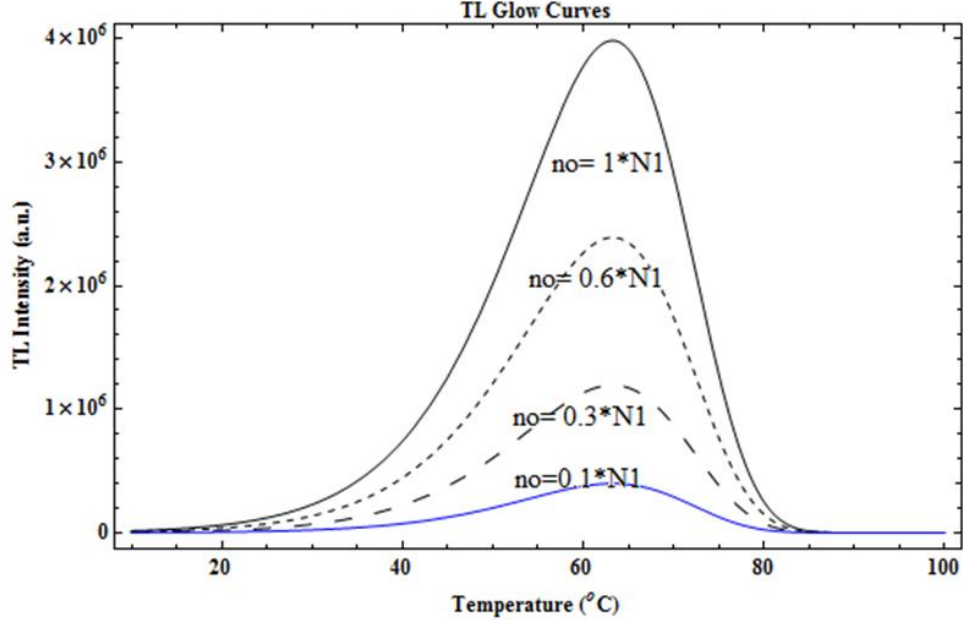


Figure 2.3 The numerical solution of the differential equations for first-order kinetics for the variable initial value n_o ($n_o/N = 1, 0.6, 0.3, 0.1$).

The evaluation of integral in equation (2.3) is not analytically solved. Therefore, some authors have used additional approximations to obtain simpler equations. Because, in dosimetric applications, it is convenient to describe the glow peak in terms of parameters which are easy to derive experimental data. Kitis et al. [24] have shown that equation 2.3 can also be written as,

$$I(T) = I_m \exp\left[1 + \frac{E}{kT} \frac{T - T_m}{T_m} - \frac{T^2}{T_m^2} \exp\left\{\frac{E}{kT} \frac{T - T_m}{T_m}\right\} (1 - \Delta) - \Delta_m\right] \quad (2.4)$$

with $\Delta = 2kT/E$ and $\Delta_m = 2kT_m/E$. On the other hand; Bos et.al[25], were approximated the following first-order TL glow curve expression.

$$I(T) = n_o s \exp\left[-\frac{E}{kT}\right] \exp\left[\left(-\frac{s}{\beta} \frac{kT^2}{E} \exp\left(-\frac{E}{kT}\right)\right) \times \left(0.992 - 1.620 \frac{kT}{E}\right)\right] \quad (2.5)$$

If it was assumed that $mA \ll (N-n)A_r$ and the trap is far from saturation, i.e. $N \gg n$ and $n=m$. Under these assumptions, $I(t)$ becomes [26];

$$I(t) = -\frac{dn}{dt} = s \frac{A}{NA_r} n^2 \exp\left\{-\frac{E}{kT}\right\} \quad (2.6)$$

In this statement; dn/dt is proportional to n^2 and $A=A_r$, the integral of above equation (2.7) becomes;

$$I(T) = \frac{n_0^2}{N} \frac{s}{\beta} \exp\left\{-\frac{E}{kT}\right\} \left[1 + \frac{n_0 s}{N\beta} \int_{T_0}^T \exp\left\{-\frac{E}{kT'}\right\} dT' \right]^{-2} \quad (2.7)$$

The second order kinetics was given by Garlick and Gibson [26] for one trap and one recombination center model. It is based on two assumptions, **(i)** The freed charge carriers have equal probability for retrapping and recombination, **(ii)** The concentration of electrons in traps and holes in recombination center are equal during the entire process. This is the Garlick–Gibson second-order TL kinetics in which the TL glow curve is nearly symmetric. As explained in above sections, the first-order ($b=1$) and second-order ($b=2$) TL kinetic models have been derived after the using of some specific assumptions. However, some experimental studies have been represented that these models do not hold to explain some glow peaks that they will not fitted neither first- nor the second-order kinetics. May and Partridge [28] used an empirical expression which is namely called as general-order ($b \neq 1$ and 2) TL kinetics to analyze these glow peaks,

$$I(t) = -\frac{dn}{dt} = n^b s \exp\left\{-\frac{E}{kT}\right\} \quad (2.8)$$

The solution of above equation (2.9) for $b \neq 1$ produces

$$I(T) = \frac{s''}{\beta} n_0 \exp\left\{-\frac{E}{kT}\right\} \left[1 + (b-1) \frac{s''}{\beta} \int_{T_0}^T \exp\left\{-\frac{E}{kT'}\right\} dT' \right]^{-b/b-1} \quad (2.9)$$

The general-order case is useful since intermediate cases can be dealt with and it smoothly goes to first- and second-orders when $b \rightarrow 1$ and $b \rightarrow 2$ respectively, (Figure 2.5).

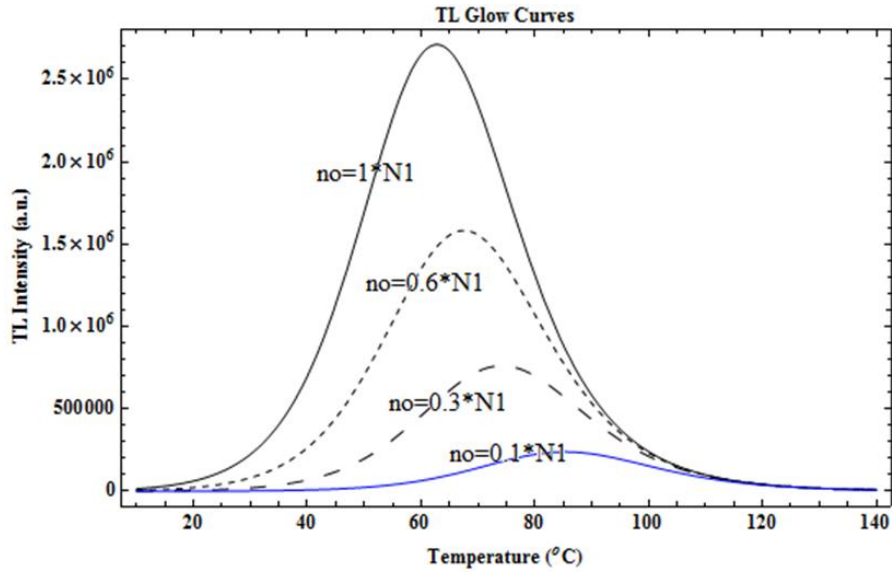


Figure 2.4 The numerical solution of the differential equations for second-order kinetics for the variable initial value n_0 ($n_0/N = 1.0, 0.6, 0.3, 0.1$).

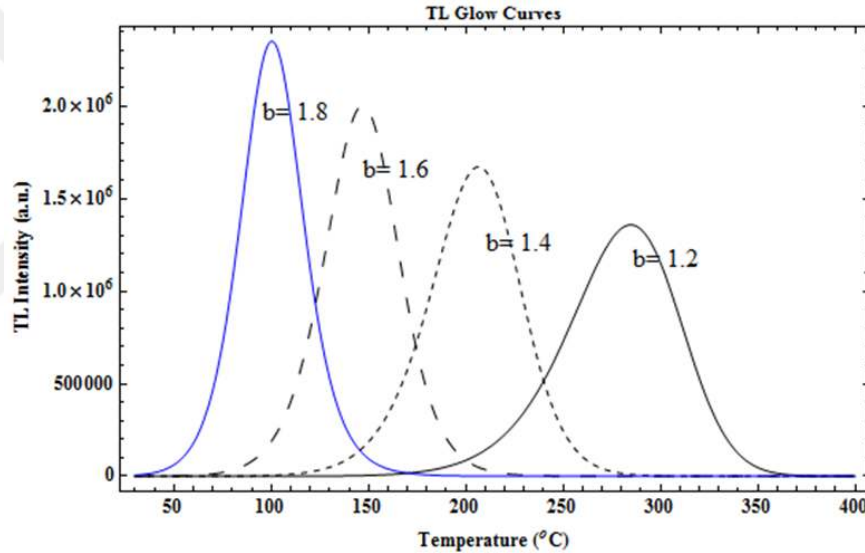


Figure 2.5 The numerical solution of the differential equations for general-order kinetics for the different kinetic-orders ($b = 1.2, 1.4, 1.6$ and 1.8).

2.4 Methods for Determination of Trapping Parameters

The kinetic parameters (the activation energy (E), the frequency factor (s), the kinetic order (b)) of a thermoluminescence material should be calculated for the characterization of dosimetric materials. The glow curve of a sample can be evaluated in a TL experiment for different conditions of measurements. The purpose of these measurements and analysis are to extract the parameters which are used to describe the TL process in the material. The dosimetric characteristic of any TL material depends mainly on these parameters responsible for TL [1].

2.4.1 Peak Shape Method

The shape of a glow peak depends on the kinetic order involved in the TL process. By using the glow peak shape, we can get information about the activation energy, attempt to escape frequency and the kinetic order. Together with the maximum peak temperature (T_m) the half the peak intensity on the rising part, T_1 and falling part T_2 of the glow curve is used [7].

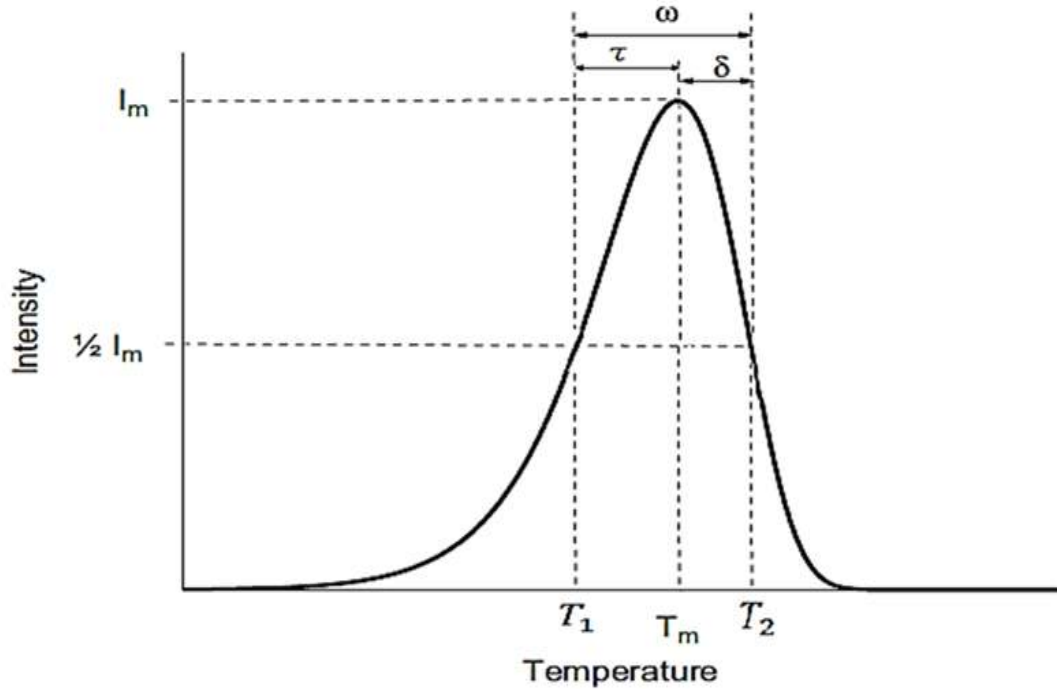


Figure 2.6 Normalized glow peak and shows characteristics of a single glow peak with defining the peak shape parameters. [7]

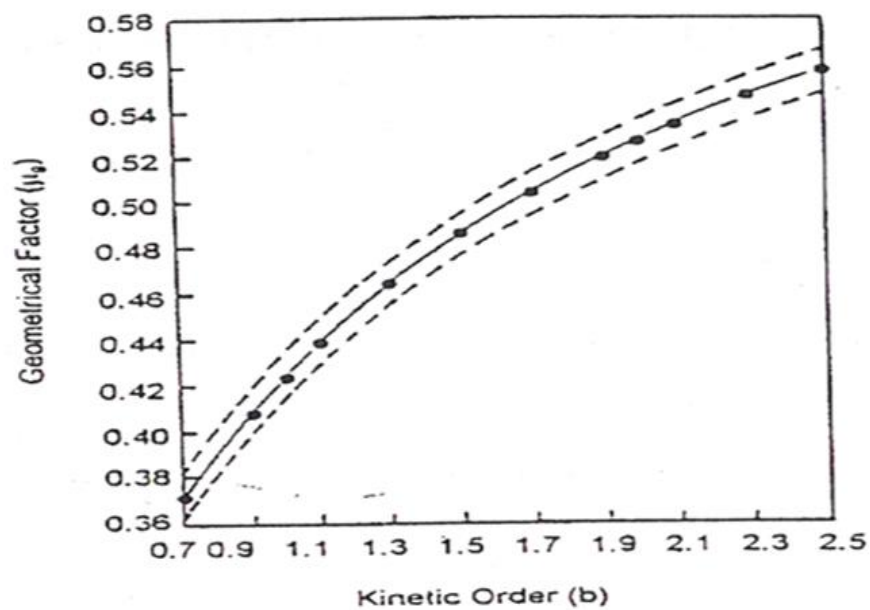


Figure 2.7 Geometrical factors (μ_g), as a function of the given order [7].

Once these values are evaluated from the TL glow curve by using the following equation the kinetic parameters can easily be found. In this equation μ_g is the shape parameter its value is nearly 0.42 for $b=1$ and 0.52 for $b=2$ for linear heating [1, 4, 5, 7];

$$\begin{aligned} E_\tau &= [1.51 + 3(\mu_g - 0.42)] \frac{kT_m^2}{\tau} - [1.58 + 4.2(\mu_g - 0.42)] 2kT_m \\ E_\delta &= [0.976 + 7.3(\mu_g - 0.42)] \frac{kT_m^2}{\delta} \\ E_\omega &= [2.52 + 10.2(\mu_g - 0.42)] \frac{kT_m^2}{\omega} - 2kT_m \end{aligned} \quad (2.10)$$

where; ($\omega=T_2-T_1$); ($\delta=T_2-T_m$); ($\tau =T_m-T_1$) and $\mu_g=\delta/\omega$ called the shape parameter.

The frequency factor can be found as follows;

$$\begin{aligned} s &= \frac{\beta E}{kT_m^2} \exp\left[\frac{E}{kT_m}\right] \\ s &= \frac{\beta E}{kT_m^2} \left[\exp\left(-\frac{E}{kT_m}\right) \left(1 + (b-1) \frac{2kT_m}{E}\right) \right]^{\frac{b}{b-1}} \end{aligned} \quad (2.11)$$

2.4.2. Heating rate method

Another method for determining the kinetic parameters is the heating rate method in which the change of the peak position of TL glow curves during TL readout for different heating rates is used. The maximum temperature of a glow peak changes with the heating rate. Using this property, the activation energy (E) can be obtained using two different heating rates, by

$$E = k \frac{T_{m1}T_{m2}}{T_{m1} - T_{m2}} \ln\left[\left(\frac{\beta_1}{\beta_2}\right)\left(\frac{T_{m2}}{T_{m1}}\right)^2\right] \quad (2.12)$$

When various heating rates for the first-order kinetics are used, the following expression is obtained:

$$\ln\left(\frac{T_m^2}{\beta}\right) = \left(\frac{E}{k}\right)\left(\frac{1}{T_m}\right) + \text{constant} \quad (2.13)$$

Plotting $\ln (T_m^2/\beta)$ versus $(1/T_m)$ will yield a straight line having a slope of E/k . Then if one extrapolated $1/T_m = 0$, $\ln(sk/E)$ can be obtained from here the frequency factor can be found. The various heating rates can also be applicable for general-order kinetics. For the general order case, $\ln [I_m^{b-1} (T_m^2/\beta)^b]$ versus $1/T_m$, will have a slope of E/k .

2.4.3 CGCD Method

One of the most important way that use for obtaining the value of the parameter E_a , s , and b is the Computer Glow Curve Deconvolution (CGCD) method, and instead of heating and adding addition doses by CGCD method can separate many overlapping glow peaks, these computerized curve fitting methods are more accurate with clearly successful value in use. The procedure of this method is firstly from the glow curve the approximation locations of the most evident peak is finding and by using one of the analytical methods is estimated each value of E_a , s , and b . computed curve with the exact result of the experimental curve is compared. Between the two results, the method of root means square is applied.

This method is useful and better than experimental method because without acting to heat treatment can be used in the largely overlapping peak of glow curve [25]. The computerized glow curve deconvolution (CGCD) method has become very popular way to obtain the kinetic parameters from the glow curves of TL materials. Therefore, the kinetic parameters such as number of glow peaks, activation energies (E_a), frequency factor (s) and kinetic order (b) for the dosimetric peaks of TLD-600 and TLD-700 were obtained by CGCD method. In this study, the following general order kinetic analytical equation was used by the approximation for $b \approx 1.1$ [24]

$$I(T) = I_m \times b^{b/b-1} \times \exp\left(\frac{E}{kT} \cdot \frac{T - T_m}{T_m}\right) \times [(b - 1) \times (1 - \Delta \times \frac{T^2}{T_m^2} \times \exp\left(\frac{E}{kT} \times \frac{T - T_m}{T_m}\right) + Z_m]^{-b/b-1} \quad (2.14)$$

where;

$$\Delta = \frac{2kT}{E}, \quad \Delta_m = \frac{2kT_m}{E}, \quad Z_m = 1 + (b - 1) \cdot \Delta_m$$

The number of glow peaks is not a free fitting parameter during the deconvolution of the measured glow curve into individual components in the CGCD programs. If the number of peaks is not known, it can be found by fitting the glow curve several times with a different number of components up to obtaining best-fit result.



CHAPTER 3

EXPERIMENTAL PROCEDURES

The materials, instruments and experimental procedures utilized in this thesis are described in below sections.



Figure 3.1 Thermoluminescence Laboratory.

3.1 Materials

The samples used in this study were commercial TLD-600 (with 95.6% ^6LiF) and TLD-700 (with 99.9% ^7LiF) from the Harshaw Chemical Co., in the shape of chips measuring 3x3x0.9 mm.

3.2 Radiation Source and Irradiation Procedure

The samples were firstly annealed at 400 ± 1 °C for 1 hour prior to irradiation with a specially designed microprocessor controlled electrical oven followed by a fast cool on an aluminum block.

For beta and mixed neutron+gamma irradiations; ^{90}Sr - ^{90}Y ($D \approx 0.04$ Gy/sec) and ^{252}Cf (the average neutron energy of the fission spectrum is ~ 2.3 MeV and $D \approx 0.08$ Gy/sec, $D_n/D_\gamma = 1.2$) sources were used, respectively. The beta source was purchased from Little More Scientific Engineering in UK and calibrated by manufacturer on March 10, 1994. The recommended working life-time of this radiation source is about 30 years. The activity of β -source at this date is about 100 mCi. Nowadays, this source approximately gives 0.015 Gy at per second. Sr-90 emits high energy beta particles from its daughter products (^{90}Sr -0.546 MeV together with ^{90}Y β -2.27 MeV). It is well known that the β -radiation is easily absorbed in any medium, i.e. in air. Therefore, its intensity is quickly decreases with distance much more rapidly than inverse square law calculations. The, the maximum range of Y-90 beta particles in air is approximately 9 meters. The irradiation apparatus is a supplementary part of the 9010 Optical Dating System (Figure 3.2). This system was purchased from Little More Scientific Engineering in England [29] and it is interfaced to a personal computer (PC) using a serial RS-232 port.



Figure 3.2 9010 Optic Dating System and ^{90}Sr - ^{90}Y β -source supplementary on it [18].



Figure 3.3 Neutron + Gamma Source [18].

On the other hand, ^{252}Cf source was used for the production of neutron radiations. The samples were irradiated for 1 min at room temperature with a ^{252}Cf neutron+gamma source delivering about 0.08 Gy/min radiation. The all samples were exposed to neutron, ^{252}Cf radiations at room temperature. The average energy of the neutron source is around 2 MeV. The neutron source was purchased from Eckert & Ziegler Isotope Products GmbH and its calibration was also done by the manufacturer on 01 March 2015 (Figure 3.3).

3.3 TL Analyzer and TL Measurements

The glow curve measurements for quartz crystals were made using a Harshaw TLD System 3500 Manual TL Reader [30]. It economically provides high reliability. The technical architecture of the system includes both the Reader and a DOS-based IBM-compatible computer connected through a standard RS-232 serial communication port to control the 3500 Reader. A picture of the TLD-reader and basic block diagram of this reader is shown in figure 3.4 and 3.5 respectively.



Figure 3.4 Harshaw TLD System 3500 Manual TL Reader.

All the functions are divided between the reader and the specialized TLDSHELL software program that runs on the PC. All data storage, instrument control, and operator inputs are performed on the PC. Signal acquisition and conditioning are performed in the reader. In this way, each glow curve can be analyzed using a best-fit computer program based on a Marquardt algorithm minimization procedure, associated to first-order, second-order and general-order kinetic expressions. The program resolves the individual peaks present in the curve, giving the best values for the different peak

parameters. The instrument includes a sample change drawer for inserting and removing the TLD elements. The reader uses contact heating with a closed loop feedback system that produces adjustable linearly ramped temperatures from 1 °C to 50 °C per second accurate to within ± 1 °C to 400 °C in the standard reader.

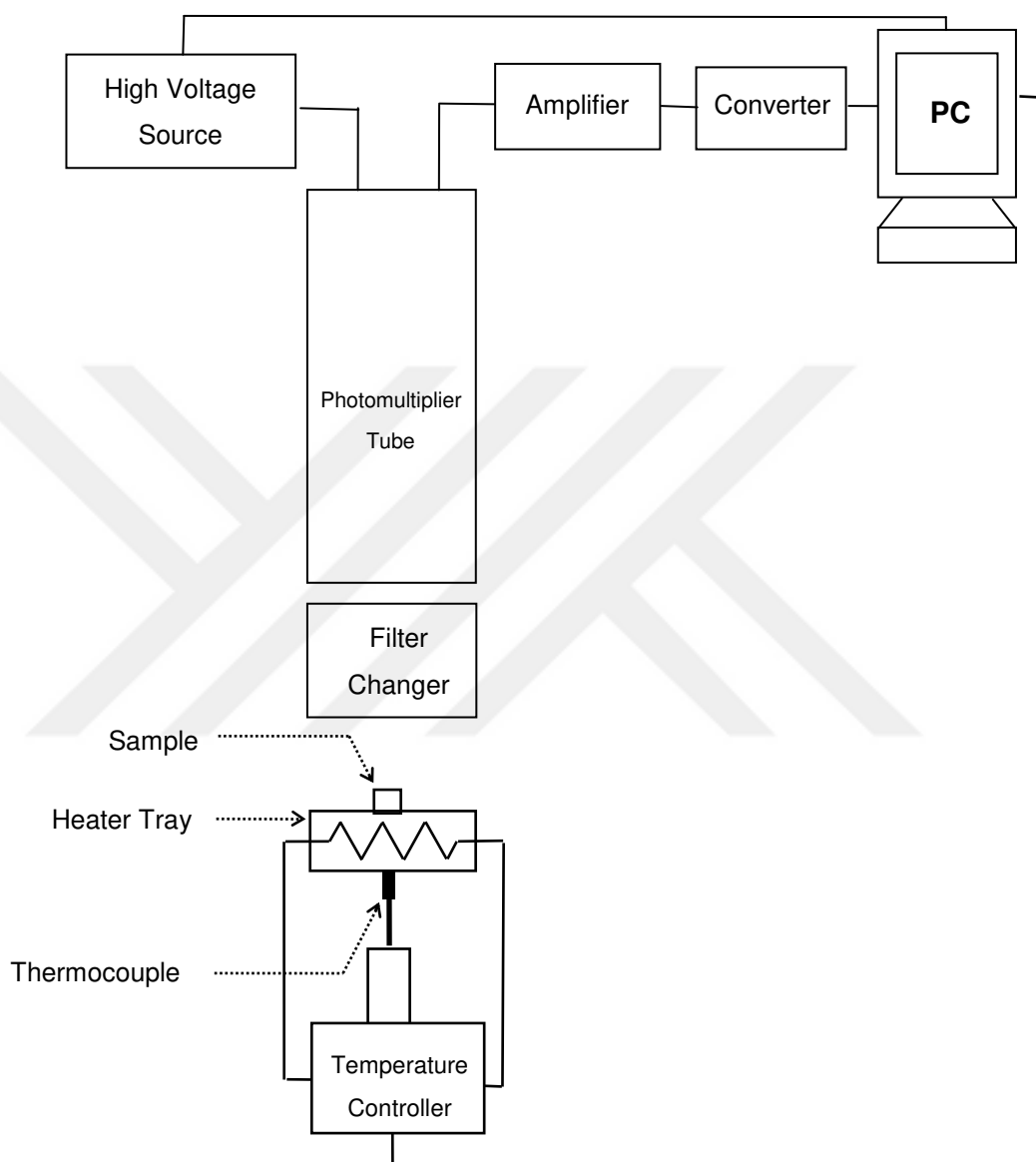


Figure 3.5 Basic block diagram of TL reader.

The Time Temperature Profile (TTP) is user defined in three segments: Preheat, Acquire, and Anneal, each with independent times (Pre-read anneal: adjustable 0 to 1000 sec, Linear ramp: adjustable from 1 °C to 50 °C per second, Post-read anneal: 0 to 1000 sec) and temperature (Pre-read anneal: room temperature to 200 °C, Post-read anneal: up to 400 °C). The typical time temperature profile is shown in figure 3.6. To improve the accuracy of low-exposure readings and to extend

planchet life, the 3500 provides for nitrogen to flow around the planchet. By eliminating oxygen in the planchet area, the nitrogen flow eliminates the unwanted oxygen-induced TL signal. Nitrogen is also routed through the photo-multiplier tube (PMT) chamber to eliminate moisture caused by condensation. Glow curves were measured using a platinum planchet at a linear heating rate of 1 °C/s. The time duration between irradiation and necessary TL operation was always kept constant at about 1 min, except for the storage time experiment. For the variable heating rate method heating rates were varied from 1 to 10 °C/s. At each heating rate, each chip was read out twice and the second readout is considered to be background of the reader plus chip and was subtracted from the first one and all of the analyses have been carried out after subtraction operations

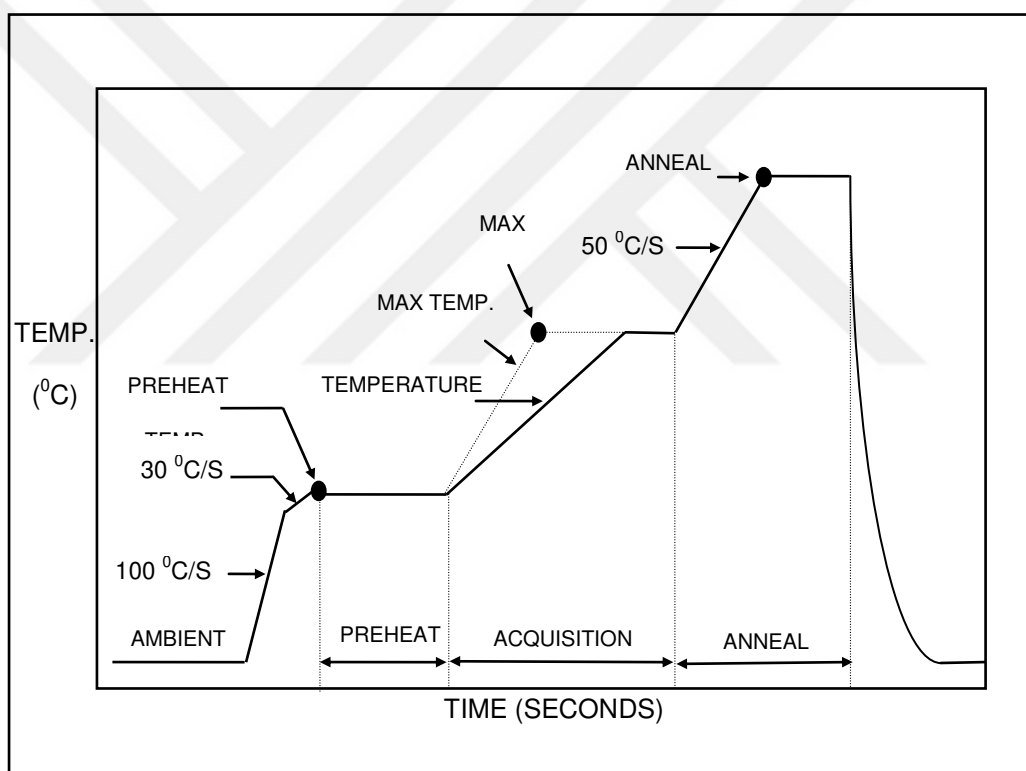


Figure 3.6 Typical time temperature profile (TTP) of a TLF reader.

CHAPTER 4

EXPERIMENTAL RESULTS

The main purpose of this study is to observe the sensitivities, dose response linearity, kinetic parameters and the effect of heating rate on the glow curves of lithium fluoride thermoluminescent dosimeters TLD-600 ($^6\text{LiF:Ti,Mg}$), and TLD-700 ($^7\text{LiF:Ti,Mg}$) after exposed to ^{252}Cf neutron+gamma and ^{90}Sr - ^{90}Y beta-rays.

4.1 Dose Response of TLD-600 and TLD-700

The thermoluminescent dosimeters TLD-600 ($^6\text{LiF:Ti,Mg}$) and TLD-700 ($^7\text{LiF:Ti,Mg}$) were irradiated both ^{252}Cf mixed (neutron (n) +gamma (γ)) and ^{90}Sr - ^{90}Y beta (β)-sources at different doses between ≈ 15 and ≈ 500 Gy for mixed (n+ γ) and β -irradiations to observe the changes in the glow curve structures and to check the glow peak intensities and dose dependence effects of the peak positions as a function of exposed dose levels. Some of the selected glow curves of the TLD-600 ($^6\text{LiF:Ti,Mg}$) after irradiating with different dose levels using mixed (n+ γ) and β -sources are shown in Figure 4.1(a) and (b), respectively.

The experimental observations have clearly shown that there were no significant changes in the glow curve structures and peak temperatures of glow peaks of TLD-600 ($^6\text{LiF:Ti,Mg}$) with increased dose levels after different types of radiation sources and levels. This result shows that all the glow peaks of TLD-600 should have first-order kinetics. The measured peak temperature of highest peak intensity is obtained at around 245 ± 2 °C after both types of irradiations and dose levels. The obtained results of peak temperature for TLD-600 dosimeter are in agreement with the recent studies [31-35]. After being exposed to mixed (n+ γ) and β -sources, the total area under the glow curves of TLD-600 ($^6\text{LiF:Ti,Mg}$) is measured for a variety of exposed dose levels. As seen in Figure 4.2(a) and (b), the total glow curve

area under the glow curve of TLD-600 increases linearly with respect to exposed dose levels in the studied dose levels after both types of irradiations ((n+ γ) and β).

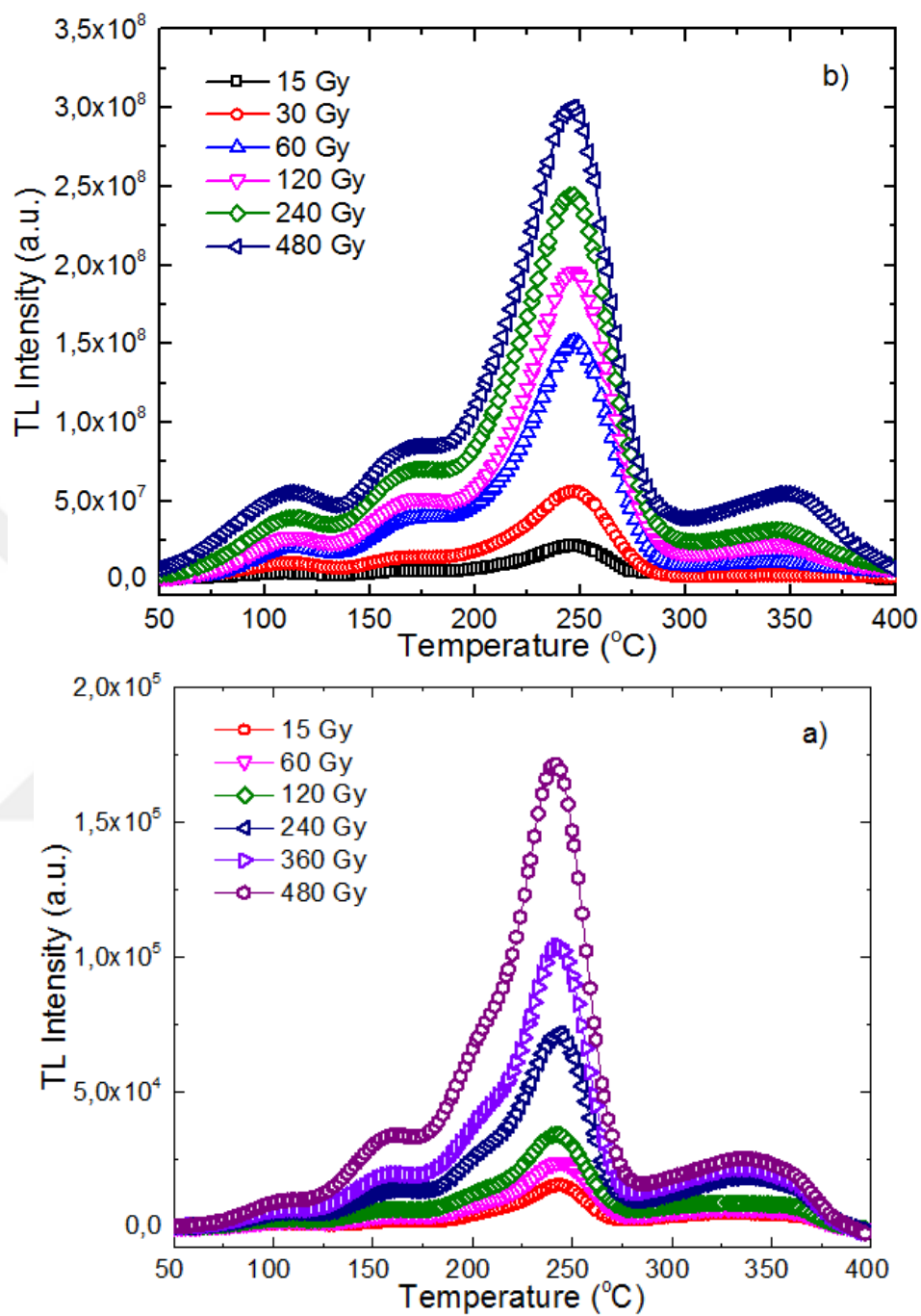


Figure 4.1 The glow curves of TLD-600 ($^6\text{LiF}:\text{Ti},\text{Mg}$) corresponding to various radiation doses using (a) ^{252}Cf mixed (n+ γ) and (b) ^{90}Sr - ^{90}Y β -sources with a heating rate of 1 °C/s.

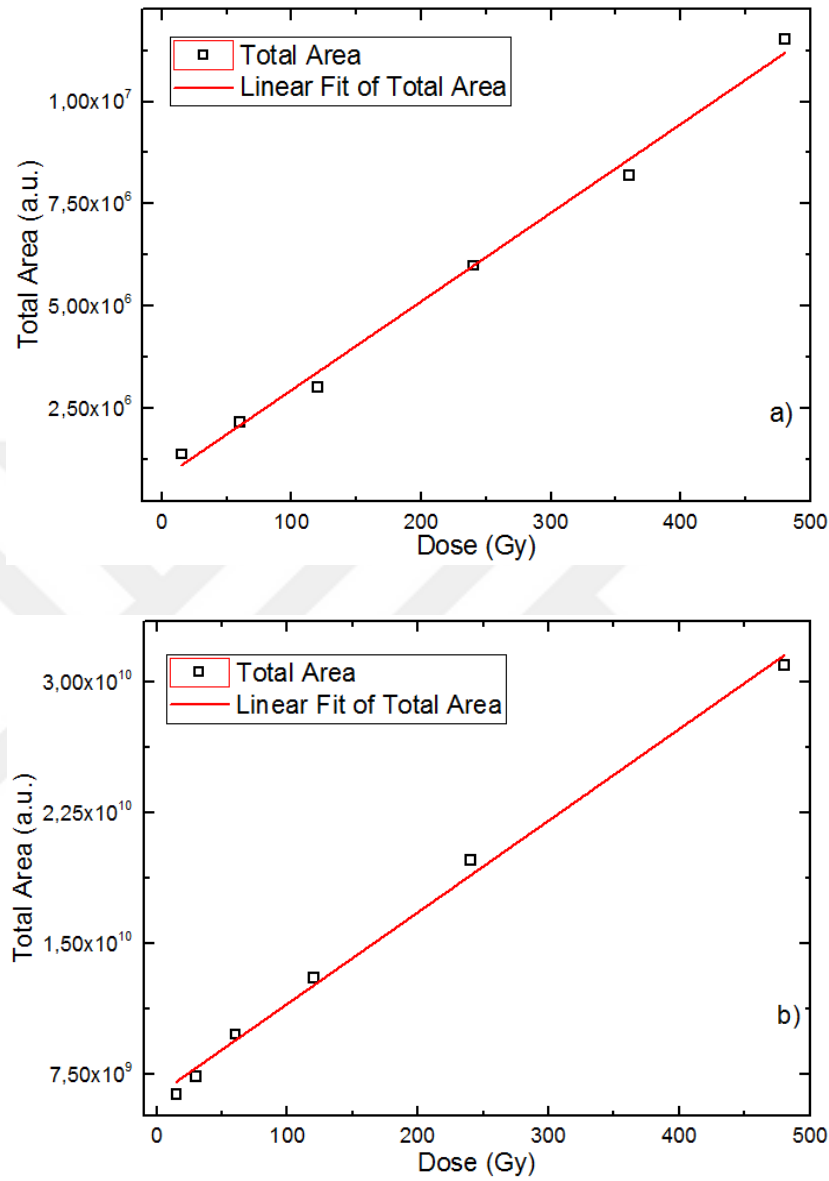


Figure 4.2 The total area under glow curves of TLD-600 ($^6\text{LiF}:\text{Ti},\text{Mg}$) as a function of exposed dose levels using mixed (a) (n+γ) and ^{90}Sr - ^{90}Y β-sources.

Some of the recorded glow curves of the TLD-700 ($^7\text{LiF}:\text{Ti},\text{Mg}$) after irradiating with different dose levels using mixed (n+γ) and β-sources are shown in Figure 4.3(a) and (b). The experimental observations have clearly shown that there were no significant changes in the peak temperatures of highest glow peak of TLD-700 ($^7\text{LiF}:\text{Ti},\text{Mg}$) with increased dose level. This result clearly shows that all the glow peaks of TLD-700 should have first-order kinetics. As seen, the measured maximum peak temperatures of this peak are obtained at around 245 ± 2 °C for both

types of sources. On the other hand, the structures of glow curves are considerably different from each other after both types of sources. On the other hand, the variations of total areas under the glow curves of TLD-700 after being exposed to mixed (n+ γ) and β -sources increase linearly as a function of exposed dose levels (see Fig.4.4(a) and (b)).

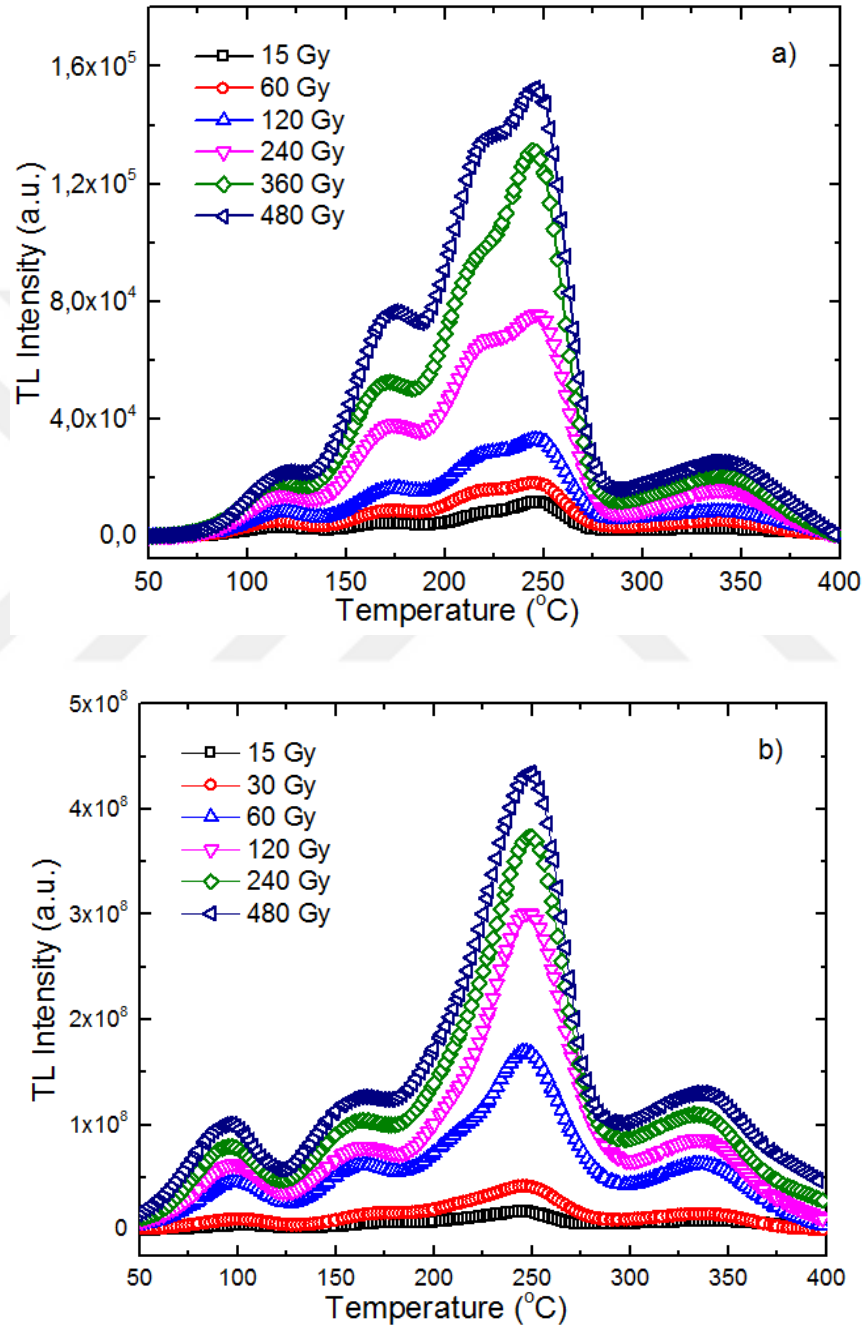


Figure 4.3 a) The glow curves of TLD-700 (^7LiF : Ti, Mg) corresponding to various radiation doses using (a) ^{252}Cf mixed (n+ γ) and (b) ^{90}Sr - ^{90}Y β -sources with a heating rate of 1 $^{\circ}\text{C}/\text{s}$.

When compared the peak intensities of glow curves of TLD-600 ($^6\text{LiF:Ti,Mg}$) and TLD-700 ($^7\text{LiF:Ti,Mg}$) after mixed neutron+gamma and beta sources, it was observed that the glow curve intensities of both dosimeters after irradiation mixed neutron+gamma are always lower than beta-irradiations. The total glow curve areas and at the same time the maximum peak intensities of TLD-600 and TLD-700 after β -irradiations are approximately 10^4 and 10^3 times greater than after mixed n+ γ -irradiations, respectively.

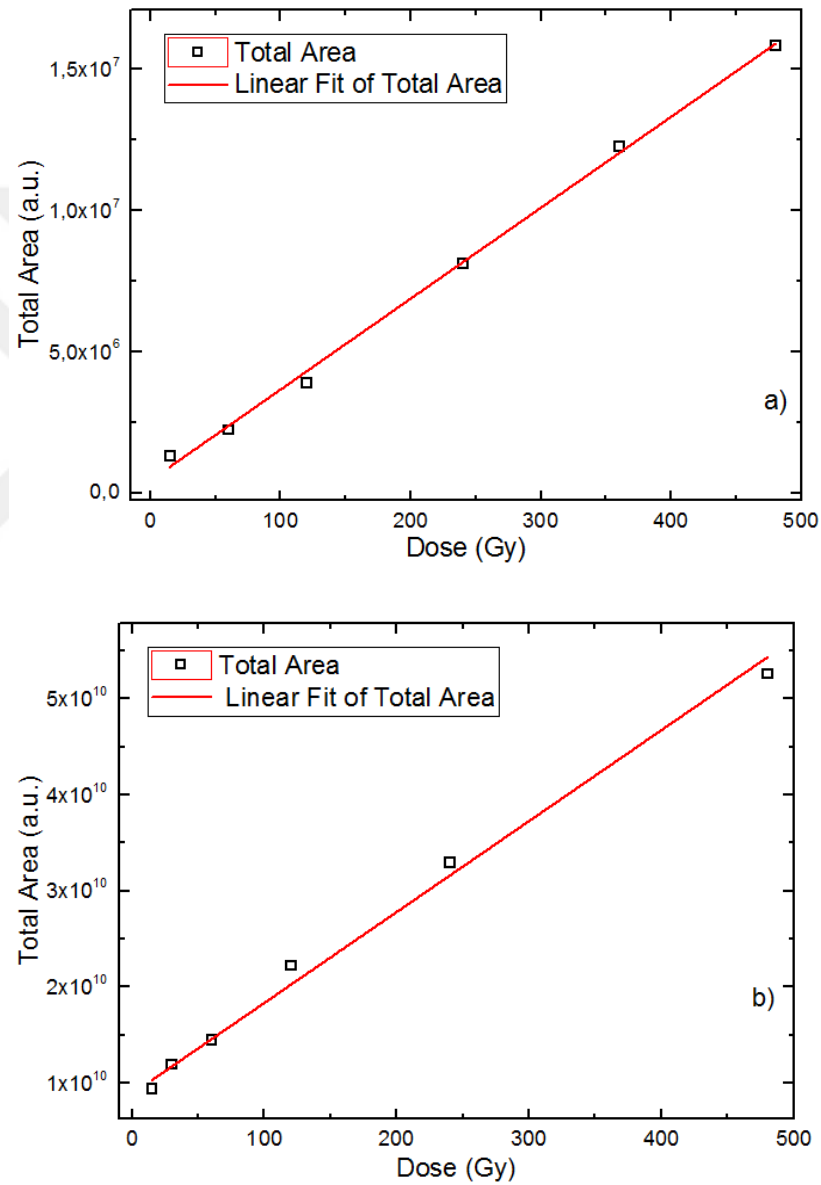


Figure 4.4 The total area under glow curves of TLD-700 ($^7\text{LiF:Ti,Mg}$) as a function of exposed dose levels using (a) mixed (n+ γ) and (b) ^{90}Sr - ^{90}Y β -sources.

4.2. Heating Rate Effects on TLD-600 and TLD-700

It is well known that the heating rate is an important experimental parameter on the glow peak intensities and peak temperatures of many TL dosimetric materials [36-39]. In general, the peak intensity decreases with increasing heating rate due to thermal quenching [36-39]. Therefore, the effects of heating rate on the glow curves of the thermoluminescent dosimeters TLD-600 ($^6\text{LiF}:\text{Ti},\text{Mg}$) and TLD-700 ($^7\text{LiF}:\text{Ti},\text{Mg}$) were also investigated using ^{252}Cf mixed neutron + gamma and ^{90}Sr - ^{90}Y beta sources in this study. Some of the recorded glow curves after various heating rates were shown in Figure 4.5 and 4.6 for TLD-600 ($^6\text{LiF}:\text{Ti},\text{Mg}$) and TLD-700 ($^7\text{LiF}:\text{Ti},\text{Mg}$). It is clearly seen from these figures 4.5 and 4.6 that the peak temperatures of all glow peaks increase while corresponding TL intensities and the total areas under the glow curves of both dosimeters decrease with increasing heating rates (Figures 4.7 and 4.8).

On the other hand, as seen from figures 4.5-4.8, the effects of heating rates on the peak intensities and total area under the glow curves of both dosimeters after β -irradiations are more profound than mixed n+ γ irradiations.

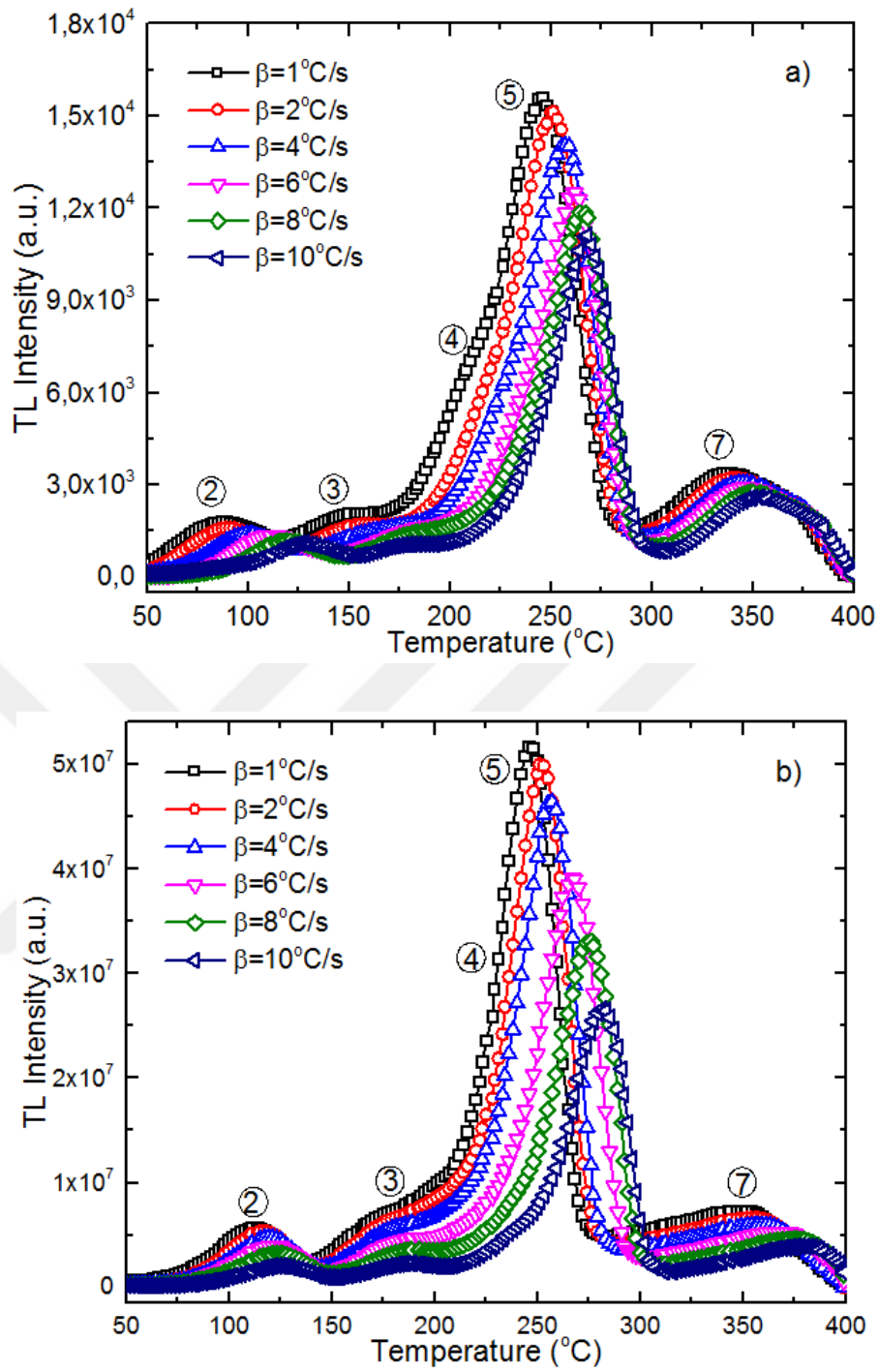


Figure 4.5 The glow curves of TLD-600 ($^6\text{LiF:Ti,Mg}$) measured after various heating rates between 1 and 10 $^{\circ}\text{C/s}$. Measurements are taken after irradiation of 15 Gy using (a) ^{252}Cf mixed n+ γ and (b) β -sources.

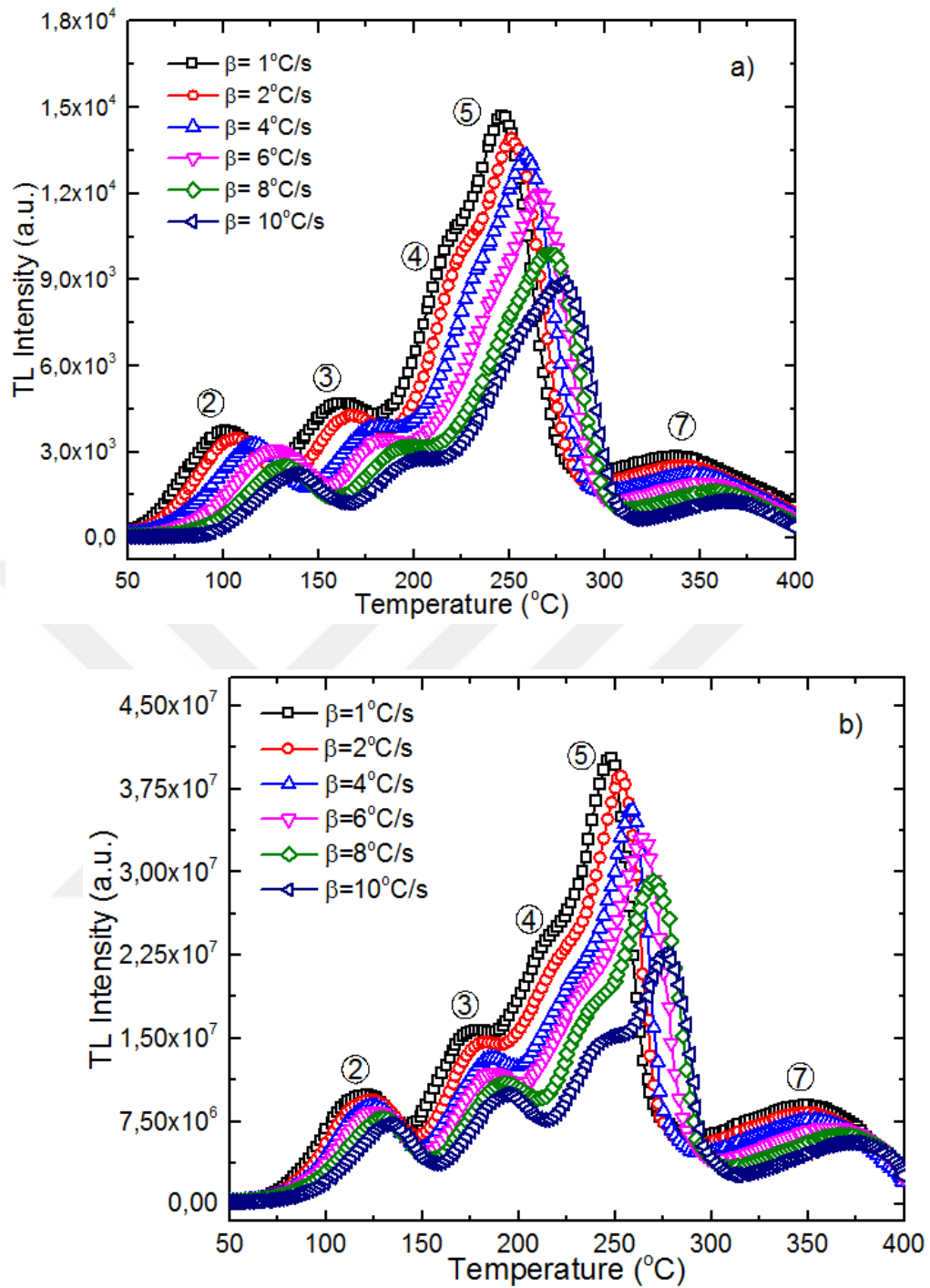


Figure 4.6 The glow curves of TLD-700 ($^7\text{LiF:Ti,Mg}$) measured after various heating rates between 1 and 10 $^{\circ}\text{C/s}$. Measurements are taken after irradiation of 15 Gy using (a) ^{252}Cf mixed n+ γ and (b) β -sources.

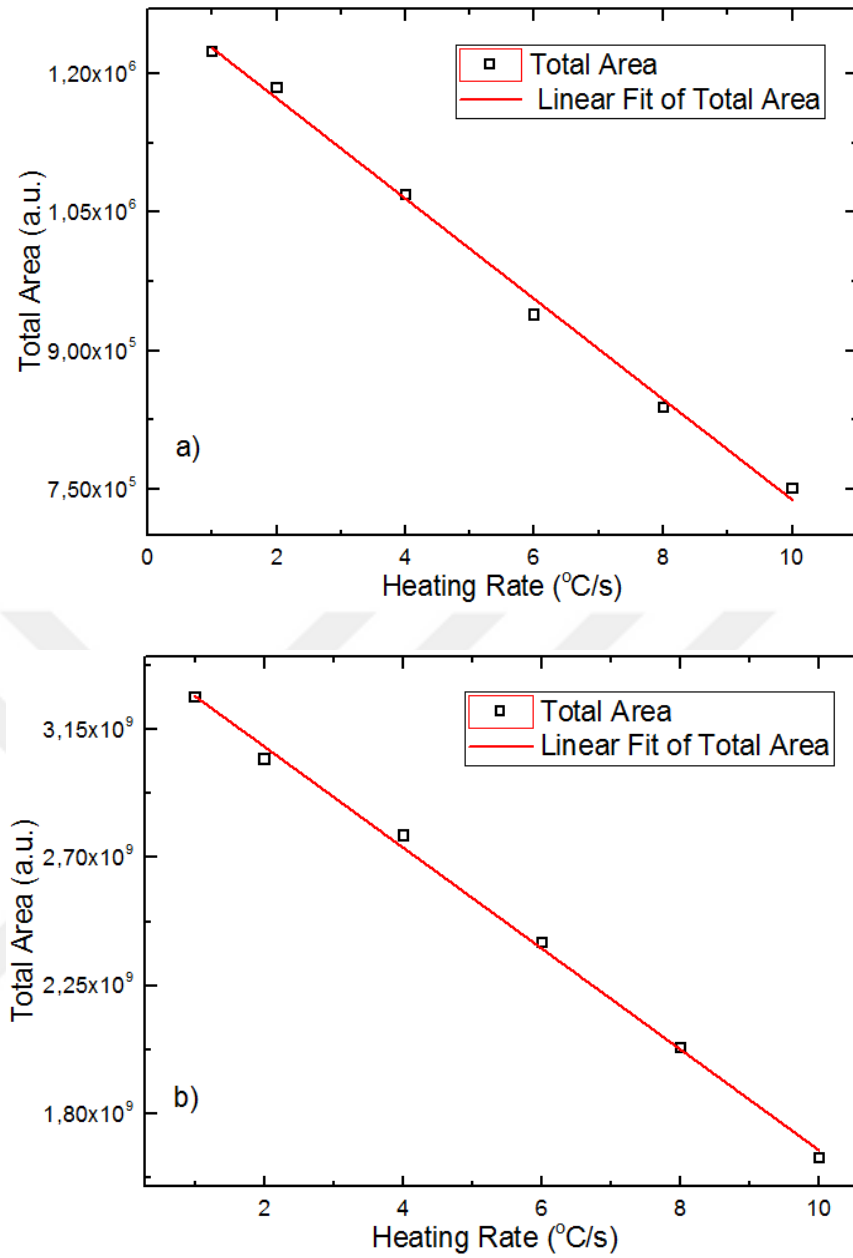


Figure 4.7 The total area under the glow curves of TLD-600 ($^6\text{LiF}:\text{Ti},\text{Mg}$) as a function of heating rates after irradiation of 15 Gy using (a) ^{252}Cf mixed $n+\gamma$ and (b) ^{90}Sr - ^{90}Y β -sources.

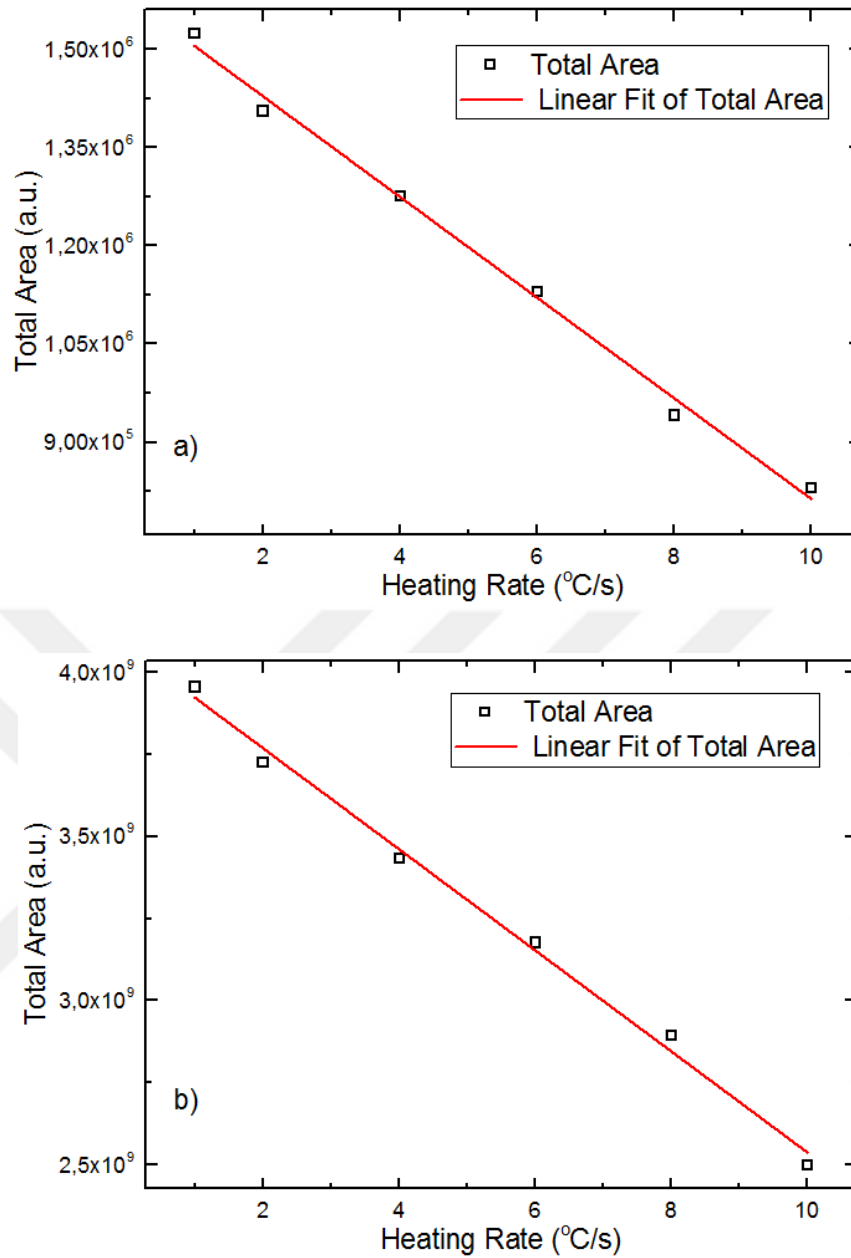


Figure 4.8 The total area under the glow curves of TLD-700 ($^7\text{LiF}:\text{Ti},\text{Mg}$) as a function of heating rates after irradiation of 15 Gy using (a) ^{252}Cf mixed n+ γ and (b) ^{90}Sr - ^{90}Y β -sources.

4.3. Determination of Kinetic Parameters

The computerized glow curve deconvolution (CGCD) method has become very popular way to obtain the kinetic parameters from the glow curves of TL materials [40]. Therefore, the kinetic parameters such as number of glow peaks, activation energies (E_a), frequency factor (s) and kinetic order (b) for the dosimetric peaks of TLD-600 ($^6\text{LiF}:\text{Ti},\text{Mg}$) and TLD-700 ($^7\text{LiF}:\text{Ti},\text{Mg}$) were obtained

by CGCD method. In this study, the following general-order kinetic analytical equation was used by the approximation for $b \approx 1.1$ [42].

$$I(T) = I_m \times b^{\frac{b}{b-1}} \times \exp\left(\frac{E}{kT} \frac{T - T_m}{T_m}\right) \times \left[(b-1) \times (1 - \Delta) \times \frac{T^2}{T_m^2} \times \exp\left\{\frac{E}{kT} \frac{T - T_m}{T_m}\right\} + Z_m \right]^{\frac{b}{b-1}} \quad (4.1)$$

where;

$$\Delta = \frac{2kT}{E}, \quad \Delta_m = \frac{2kT_m}{E}, \quad Z_m = 1 + (b-1)\Delta_m$$

The number of glow peaks is not a free fitting parameter during the deconvolution of the measured glow curve into individual components in the CGCD programs. If the number of peaks is not known, it can be found by fitting the glow curve several times with a different number of components up to obtaining best-fit result. In this study, after many tries with different number of glow peaks, it was observed that the glow curve structures of TLD-600 and TLD-700 are well described by a linear combination of at least seven glow peaks. In this case, a reasonably good fit was always obtained. Some of the analyzed and deconvoluted glow curves of both dosimeters after mixed ($n+\gamma$) and β -irradiations are shown in Figures 4.9-4.10.

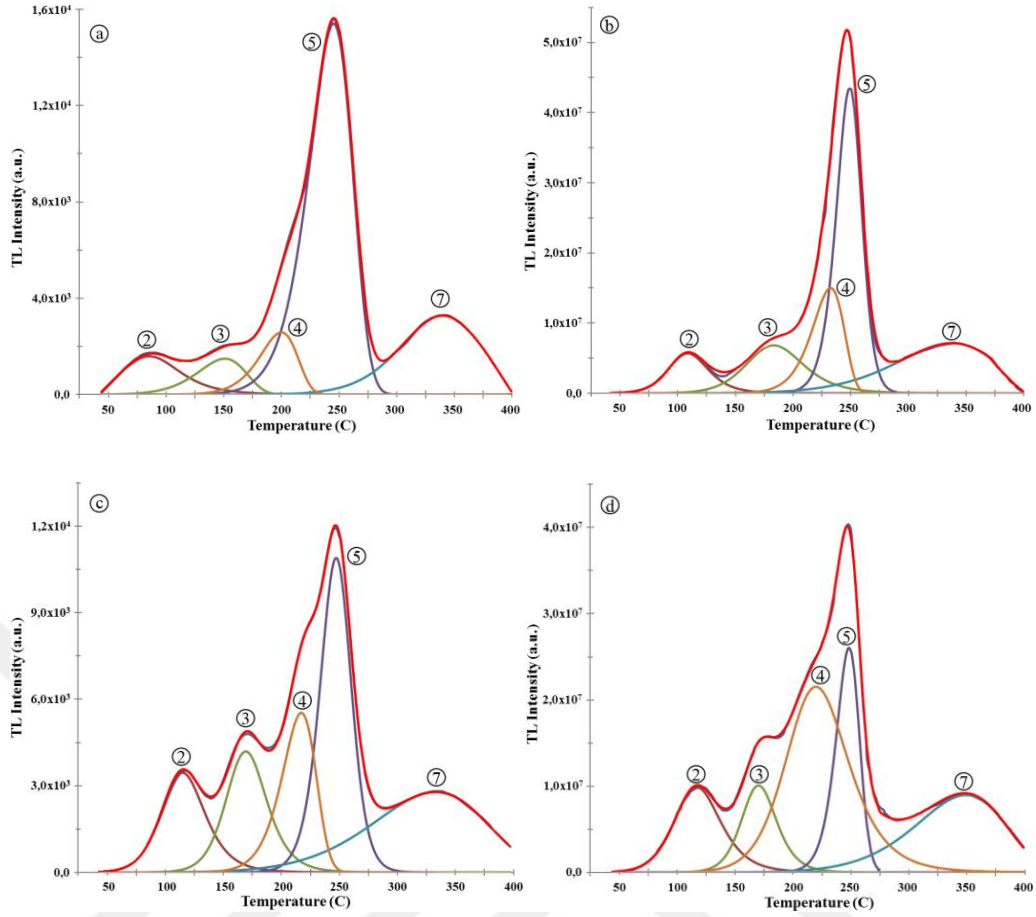


Figure 4.9 Computerised glow curve deconvolution results for TLD-600 (^6LiF : Ti, Mg) and TLD-700 (^7LiF : Ti, Mg) dosimeters irradiated with 15 Gy by a) mixed (n+ γ), b) beta, c) mixed (n+ γ), and d) beta for 1 $^{\circ}\text{C/s}$.

The obtained results of E_a , s and b by using the CGCD methods are shown in Table 4.1 and 4.2. The first column of the table shows the result of neutron + gamma irradiation and the second column beta irradiation of the dosimeters. It is clear that the obtained results are in agreement with each other. These results are also in agreement with the literature [33, 42].

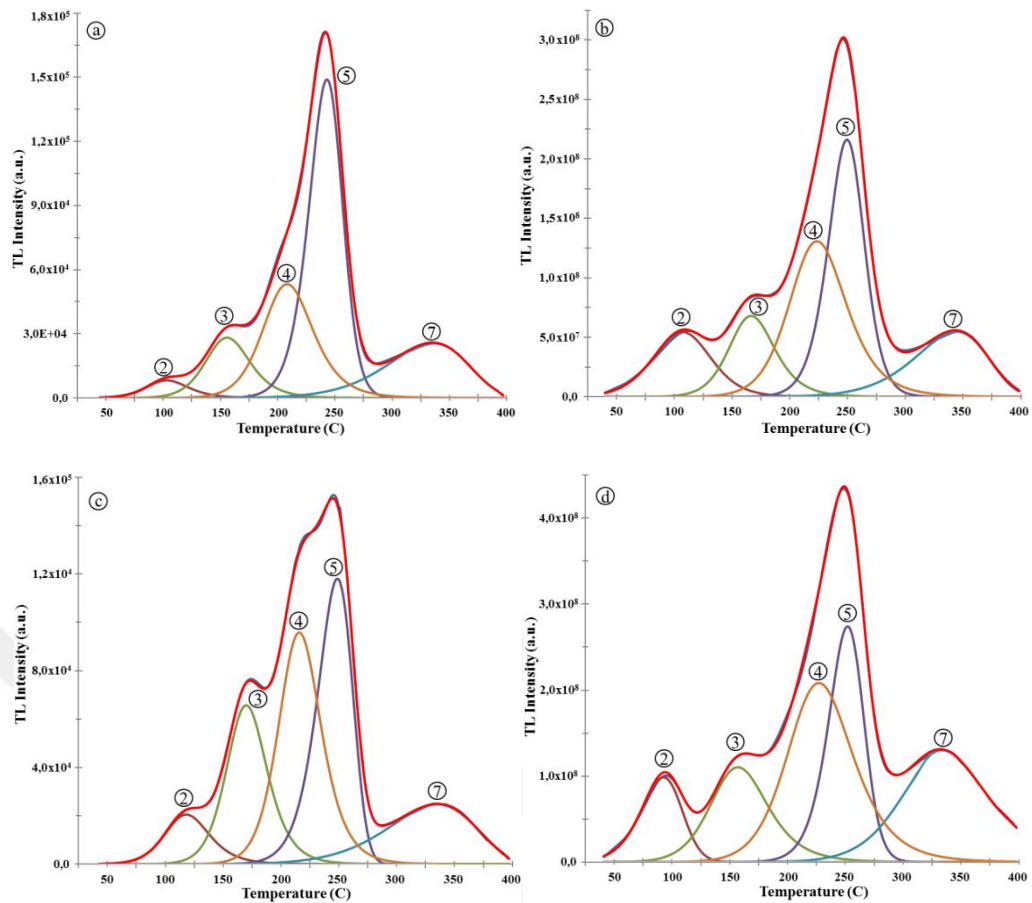


Figure 4.10 Computerised glow curve deconvolution results for TLD-600 (^6LiF : Ti, Mg) and TLD-700 (^7LiF : Ti, Mg) dosimeters irradiated with 480 Gy by a) mixed (n+ γ), b) beta, c) mixed (n+ γ), and d) beta for 1 $^{\circ}\text{C/s}$.

Table 4.1 The obtained kinetic parameters of TL glow peaks of TLD-600 and TLD-700 by CGCD method for 15 Gy for $\beta=1$ $^{\circ}\text{C/s}$.

Peak		Neutron + gamma irradiated glow peaks				β -irradiated glow peaks			
		$T_{\max}$ (K)	E (eV)	s (s^{-1})	b	$T_{\max}$ (K)	E (eV)	s (s^{-1})	b
2 3 4 5 7	TLD-600	357	0.69	1×10^{11}	2.00	383	0.77	2×10^{12}	1.00
		423	0.71	1×10^{11}	1.00	461	1.00	7×10^9	2.00
		472	1.15	1×10^{11}	1.00	506	1.57	4×10^{14}	1.00
		518	1.23	5×10^{10}	1.05	522	2.66	6×10^{24}	1.50
		612	1.08	3×10^{12}	1.59	610	0.71	1×10^{14}	1.00
2 3 4 5 7	TLD-700	$T_{\max}$ (K)	E (eV)	s (s^{-1})	b	$T_{\max}$ (K)	E (eV)	s (s^{-1})	b
		387	0.96	2×10^{11}	2.00	389	0.91	4×10^{10}	2.00
		442	1.36	2×10^{14}	2.00	443	1.59	1×10^{17}	2.00
		490	1.46	7×10^{13}	1.08	492	1.07	4×10^9	2.00
		520	2.26	8×10^{20}	1.63	521	2.64	4×10^{24}	1.26
		603	0.60	1×10^{13}	1.00	621	0.81	1×10^{15}	1.00

Table 4.2 The obtained kinetic parameters of TL glow peaks of TLD-600 and TLD-700 by CGCD method for 480 Gy for $\beta=1$ °C/s.

Peak		Neutron + gamma irradiated glow peaks				β irradiated glow peaks			
		$T_{\max}$ (K)	E (eV)	s (s ⁻¹)	b	$T_{\max}$ (K)	E (eV)	s (s ⁻¹)	b
	TLD-600								
2		375	0.93	2×10^{11}	2.00	379	0.60	4×10^{16}	1.55
3		428	1.15	2×10^{12}	2.00	439	1.18	2×10^{12}	2.00
4		481	1.28	1×10^{12}	2.00	496	1.20	9×10^{10}	2.00
5		515	1.85	1×10^{17}	1.40	522	1.91	2×10^{17}	1.59
7		607	0.87	4×10^{15}	1.00	616	1.01	5×10^{16}	1.00
	TLD-700								
		$T_{\max}$ (K)	E (eV)	s (s ⁻¹)	b	$T_{\max}$ (K)	E (eV)	s (s ⁻¹)	b
2		390	0.95	1×10^{11}	2.00	365	0.70	2×10^{11}	1.23
3		442	1.37	3×10^{14}	2.00	429	0.89	1×10^{14}	2.00
4		488	1.59	2×10^{15}	2.00	499	1.05	2×10^{14}	2.00
5		521	1.64	5×10^{14}	1.13	524	1.82	2×10^{16}	1.36
7		607	0.80	1×10^{15}	1.00	606	1.16	1×10^{12}	1.99

CHAPTER 5

CONCLUSION

TL dosimeters are now being increasingly used to monitor neutron doses. There is a need to monitor neutron doses in mixed n, γ fields. The sensitivity to neutrons of TL phosphors depends on their thickness, isotropic composition, irradiation condition and, to some extent, on the activators. The response of TL materials to fast neutrons is rather low. Various techniques have been tried to increase the neutron response of the main dosimetry peaks such as (i) using a hydrogenous material to provide recoil protons, (ii) using a foil of fissionable material and detecting the fragments and (iii) using a moderator to thermalize the neutrons [43].

A neutron has concerning the like atomic mass same as a proton, however dissimilar a proton, it has no electrical charge. A proton with its plus charge is forcefully repelled from a nucleus; however, a neutron is not due to not having a charge. Neutrons have no charge, are not pushing back if they closing nuclei, and could essentially go into nuclei. Because of neutrons are electrically neutral, they reason no ionisation immediately, and thus they could travel large distant whereby substance, more same gamma rays. The neutrons interact with substance depends to a major range on their energies. For this cause, neutrons are frequently defined as if energy is greater than 8 keV, it is called fast neutrons, if energy is less than 8 keV, it is called slow neutrons, if energy is about 0.025 eV, it is called thermal neutrons [22].

When an energetic neutron is incident on a nucleus and captured by it, the nucleus becomes unstable. This newly formed unstable nucleus immediately ejects a proton or an alpha particle to form a different element, or radiate a gamma photon to lose its excess energy. The reaction which the unstable nucleus undergoes is determined by the excess energy of the incident neutron.

Fast neutrons have energies above 1 keV and the detectors used for them are similar to the ones for slow neutrons but with some modifications due to the higher energy ranges. Fast neutron detectors take advantage of the fact that an important fraction of the neutron's kinetic energy can be transferred to the target nucleus producing an energetic recoil nucleus. This will behave in a similar way to a heavy charged particle, slowly losing energy while passing through the moderator (the moderator is the material which slows down the fast neutrons). Using TL detectors for the dosimetry in mixed neutron-gamma fields requires knowledge of their response to neutrons over a broad energy range. For the calculation of the neutron fluence response, the conversion of the energy deposited by the secondary heavy charged particles to the TL reading effect must be considered. For this purpose, the light conversion factors for all secondaries as a function of their energy must be known. The relative light conversion factor is the ratio of the detector reading coming from a mass element where the dose was absorbed as a result of the deposition of the ion. The energy deposition by secondary ions and gamma radiation generated as a result of neutron interaction in the detector material or in the surrounding material (detector holder, radiator) is of fundamental importance [44].

TLDs readings upon heating come from the light conversion of the energy stored in the material after electronic excitations by the neutron induced charged recoils [45]. Several authors have studied the glow curves of LiF dosimeter with various types and dose of irradiation and found several peaks present [46,47]. It has pointed out that the differences could be arisen by different manners of excitation in thermoluminescent materials for the kinds of radiation when the irradiation dose is not so high. The difference in glow curves of ^6LiF and ^7LiF dosimeters irradiated by neutron may be caused by the $^6\text{LiF} (n,\alpha)^3\text{H}$ reaction in LiF dosimeter [48]. Lithium fluoride thermoluminescent dosimeter TLD-700 ($^7\text{LiF}:\text{Ti},\text{Mg}$) with the enrichment of dose. Unlike TLD-600 with the enrichment of 95.6% ^6LiF and 4.4% ^7LiF , where the thermoluminescence in the TLD chip is primarily generated by densely ionising secondary alpha particles and tritons produced in the dosimeter mass through $^6\text{Li}(^1_0\text{n}, ^4_2\text{He})^3_1\text{H}$ reaction, the thermoluminescence in a TLD-700 chip is created by the $^7\text{Li}(^1_0\text{n}, \gamma)^8_3\text{Li}$ reaction as well as the energy transferred to dosimeter material via elastic and inelastic scattering of the impinging neutrons [46].

In mixed radiation fields, often met in radiotherapy, the difference in the emission due to the different radiation components of the field may be used to relate the contributions of these components to the absorbed dose; but a good knowledge, for each type of radiation, of the characteristics of the peaks in the attribution correctly. In this study, we have investigated the change in the shape of the glow curves and the dose response properties of TLD-600 and TLD-700 dosimeters using mixed neutron + gamma and beta sources. The dose ranges between 15 and 500 Gy for mixed (n+ γ) and β -irradiations were experimented. As can be seen in figures 4.1 and 4.2, there was no significant change in the shape of the glow curves of TLD-600 for exposure to beta and neutron fields, but the difference in the relative intensity of TLD-600 was remarkable for exposure to beta rays. The glow curves of the TLD-700 when exposed to beta rays and neutron field shows some small variations at low temperature side due to gamma contributions. The glow curves obtained using mixed (n+ γ) and β -sources for TLD-600 and TLD-700 exhibits linear dose response curve between 15 and 500 Gy.

The TL glow curves of TLD-600 ($^6\text{LiF}:\text{Ti},\text{Mg}$) and TLD-700 ($^7\text{LiF}:\text{Ti},\text{Mg}$) were also measured after various heating rates between 1 and 10 $^{\circ}\text{Cs}^{-1}$. It was observed that with increasing heating rates, the TL intensities of both samples are reducing and as well as its peak temperatures are shifting into higher temperature side and the total area under the glow curves may be decreases when the heating rate increases. The kinetic parameters were obtained by the CGCD method and the results were tabulated in tables 4.1 and 4.2. As seen from the tables there is no agreement with the kinetic parameters calculated by CGCD program for low and high doses for both radiation sources. The reason for this LiF based dosimeters have very complex glow curves and consist of many glow peaks which are close to each other. Therefore even small changes in the shape of the glow curves significantly affect the calculated kinetic parameters of all glow peaks.

REFERENCES

- [1] McKeever S.W.S. "Thermoluminescence of Solids", Cambridge University Press, Cambridge, (1985).
- [2] J.T. Randall and M.H.F. Wilkins, "Phosphorescence and. Electron Traps: I Study of Trap Distributions," Proc. R. Soc. London Ser. A, **184**, 366, (1945).
- [3] Daniels F., Charles A.B, and Saunders F.D., "Thermoluminescence as a Reaserch Tool", Science, **117**(3040), 343-349, (1953).
- [4] McKeever S.W.S., Moscovitch M. and Townsend, P.D., "Thermoluminescence Dosimetry Materials: Properties and Use", (Nuclear Thechnology Publishing), (1995).
- [5] Mahesh K., Weng P.S. and Furetta C., "Thermoluminescence in Solids and its Applications", (Nuclear Technology Publishing Ashford), (1989).
- [6] Herman C., "Introduction to Health Physics", McGraw-Hill Education; (2008).
- [7] Chen R. and McKeever, S.W.S, "Theory of Thermoluminescence and Related Phenomena", World Scientific, Singapore (1997).
- [8] O.Annalakshmi, "*Dosimetric Characteristics of Borate Based Thermoluminescent Materials*", PhD Thesis, Homi Bhabha National Institute, Mumbai, (2013).
- [9] K.S.V.Nambi, "Thermoluminescence: Its Understanding and Application", Instituto De Energia Atomica, Sao Paulo, Brasil, (1977).
- [10] Spurny, Z. IAEA/SM-160/52 Proc.Symp. "Dosimetry Techniques Applied to Agriculture, Industry, Biology, and Medicine", IAEA, Vienna, 1. (1972).
- [11] Triolo A., Brai M., Gennaro G. and Bartolotta A., "Study of the glow curves of TLD exposed to thermal neutrons" Radiation Protection Dosimetry, **126**(1-4), 333, (2007).
- [12] EG Yukihiro, SWS McKeever, "Optically Stimulated Luminescence: Fundamentals and Applications", John Wiley & Sons, (2011).
- [13] EG Yukihiro, R Gaza, SWS McKeever, CG Soares, "Optically stimulated luminescence and thermoluminescence efficiencies for high-energy heavy charged particle irradiation in $\text{Al}_2\text{O}_3\text{:C}$ ", Radiation Measurements, **38** (1), 59-70, (2004).

- [14] Horowitz Y.S., “Thermoluminescent and Thermoluminescent Dosimetry”, Vols. 1–3, CRC Press, Boca Raton, FL, (1984).
- [15] Mittani, J. C. R.; da Silva, A. A. R.; Vanhavere, F.; Akselrod, M. S.; Yukiara, E. G. “Investigation of neutron converters for production of optically stimulated luminescence (OSL) neutron dosimeters using $\text{Al}_2\text{O}_3:\text{C}$ ”, Nuclear Instruments and Methods in Physics Research B, **260**, 663, (2007).
- [16] Iwai, M., Kobayashi, T. and Furuya H., “Crystal Growth and Optical Characterization of Rare-Earth (Re) Calcium Oxyborate $\text{Re-Ca}_4\text{O}(\text{BO}_3)_3$ (Re=Y or Gd) as New Nonlinear Optical Material”, Journal of Applied Physics, **36**, L276, (1997).
- [17] Vij D.R., “Thermoluminescent Materials”, PTR Prentice-Hall, New Jersey (1993).
- [18] İflazoğlu Sera, “*Development of New Thermoluminescent Dosimeters for Neutron Measurements*”, PhD Thesis, Gaziantep University, (2017).
- [19] Lin, SW; Weng, PS and Hsu PC, “Effects of various storage times and photon energies on the kinetic parameters of glow curves of $\text{CaF}_2:\text{Tm}$ (TLD-300)”, Applied Radiation and Isotopes, **47**(1), 83-91, (1996).
- [20] Hsu, P.C. & Wang, T.K., “On the annealing procedure of $\text{CaF}_2:\text{Dy}$ ”, Radiation Protection Dosimetry, **16**, 253-256, (1986).
- [21] Ben Shachar B., Horowitz Y.S., “Dosimetric characterisation of the high temperature peaks of $\text{LiF}:\text{Mg,Ti}$ and $\text{CaF}_2:\text{Tm}$ using computerised glow curve deconvolution”, Radiation Protection Dosimetry, **22**(2), 87-96, (1988).
- [22] Pownson RA, Pownson ER., “Essential Nuclear Medicine Physics”, Second Edition Published by Blackwell Publishing Ltd., (2006).
- [23] Chen R, Kirsh Y., “Analysis of Thermally Stimulated Processes”, Pergamon Press, Oxford, (1981).
- [24] Kitis G, Gomez Ros J.M., and Tuyn J. W. N., “Thermoluminescence glow-curve deconvolution functions for first, second and general orders of kinetics”, J. Phys. D: Appl. Phys., **31**, 2636, (1998).
- [25] Bos A. J. J., Pijters J. M., Gomez Ros J. M. and Delgado A., GLOCANIN, and Intercomparison of Glow Curve Analysis Computer Programs IRI-CIEMAT Report, 131-93-005 IRI Delft, (1993).
- [26] Garlick G.F.J. and Gibson A.F., “The electron trap mechanism of luminescence in sulphide and silicate phosphors”, Proc.Phys.Soc.**60**, 574, (1948).
- [28] C.E. May and J.A. Partridge, “Thermoluminescent kinetics of alpha irradiated alkali halides”, Journal of Chemical Physics, **40**, 1401-1409, (1964).
- [29] 9010 Optical Dating System User Manual, (1993).

- [30] Model 3500 Manual TLD Reader User's Manual, July 30, Publication No 3500-0-U-0793-005, (1994).
- [31] Yasuda H., Fujitaka K., "Non-linearity of the high temperature peak area ratio of $^6\text{LiF:Mg,Ti}$ (TLD-600)", *Radiation Measurements*, **32**, 355, (2000).
- [32] Y.S. Horowitz and B. Ben Shachar, "Thermoluminescent LiF:Mg, Cu, P for gamma ray dosimetry in mixed fast neutron-gamma radiation fields", *Radiation Protection Dosimetry*, **23**, 401-404, (1988).
- [33] Youssian D. and Horowitz Y. S., "Estimation of Gamma Dose in Neutron Dosimetry Using Peak 4 to Peak 5 Ratios in LiF:Mg, Ti (TLD-100/600)", *Radiation Protection Dosimetry*, **77**, 151–158, (1998).
- [34] Berger T., Hajek M., "On the linearity of the high-temperature emission from $^7\text{LiF:Mg,Ti}$ (TLD-700)", *Radiation Measurements*, **43**, 1467, (2008).
- [35] Yasuda, H., "Peak 4 instability in LiF:Mg,Ti (TLD-600/700): Comparison for gamma-ray and Fe-ion irradiation", *Radioisotopes*, **49**, 260-265 (2000).
- [36] Yazıcı A. N., Hacıbrahimoglu M. Y., Bedir M., "The Effect of Various Experimental Parameters on Glow Peaks and Trapping Parameters of $\text{CaF}_2\text{:Dy}$ (TLD-200) Crystals", *Turkish Journal of Physics*, **24**, 623-649, (2000).
- [37] Kumar M., Chourasiya G., Bhatt B. C., Sunta C. M., "Dependence of peak height of glow curves on heating rate in thermoluminescence" *Journal of Luminescence*, **130**(7), 1216, (2010).
- [38] Kadari A., Kadri D., "New numerical model for thermal quenching mechanism in quartz based on two-stage thermal stimulation of thermoluminescence model", *Arabian Journal of Chemistry*, **8**, 798, (2015).
- [39] Rasheedy M. S. and Zahran E. M., "The effect of the heating rate on the characteristics of some experimental thermoluminescence glow curves" *Phys. Scr.* **73**, 98–102, (2006).
- [40] Horowitz Y.S., Yossian D., "Computerised Glow Curve Deconvolution: Application to Thermoluminescence Dosimetry", *Radiat. Prot. Dosim*, **60**, 1-114, (1995).
- [41] Afouxenidis D., Polymeris G.S., Tsirliganis N.C., Kitis G., "Computerised curve deconvolution of TL/OSL curves using a popular spreadsheet program", *Radiation Protection Dosimetry*, **149** (4), 363-370, (2011).
- [42] Kitis G., Carinou E., and Askounis P., "Glow-curve de-convolution analysis of TL glow-curve from constant temperature hot gas TLD readers", *Radiation Measurements*, **47**, 258-265, (2012).
- [43] Mathur V.K., "Proerties of Principle TL Dosimeters", Research and Technology Department, Naval Surface Weapons Center TR 83-382, (1983).

- [44] Glenn F.K, “Radiation Detection and Measurement”, John Wiley and Sons Inc, (2000).
- [45] Mendez R., Iniguez M.P., Barquero R., Mananes A., Gallego E, Lorente A. and Voytchev M., Response Components of LiF:Mg,Ti Around amoderated Am–Be Neutron Source”, Radiation Protection Dosimetry, **98** (2), 173–178, (2002).
- [46] Yoo Y.S., Kim P.S. and Moon P.S., “A study on the Neutron Dosimetry with LiF Thermoluminescent Dosimeters”, Journal of the Korean Nuclear Society, **7**(3), 191, (1975).
- [47] Proceedings of a Symposium on Neutron Monitoring for Radiation Protection Purposes Organized by the IAEA and Held in Vienna, 11-15 December 1972, http://www.iaea.org/inis/collection/NCLCollectionStore/_Public/05/112/5112898.pdf
- [48] Mukherjee B., “Glow curve analysis of TLD-700 dosimeters exposed to fast neutrons and gamma rays from isotopic sources”, Nuclear Instruments and Methods in Physics Research A, **385**, 179-182, (1997).