PRODUCTION OF THERMOLUMINESCENCE DOSIMETER MATERIALS [$CaSO_4$]

A MASTER'S THESIS
in
Engineering of Physics
University of Gaziantep

By
Ayda KÖKSAL
February, 1991

Approval of the Graduate School of Natural and Applied Science

Prof.Dr. Mazhar ÜNSAL

I certify that this thesis statisfies all the requirements as a thesis for the degree of Master of Science in Engineering of Physics Department.

Assoc. Prof. Dr.Ata SELÇUK
Chairman of The Department

We certify that we have read this thesis and in our opinion it is fully adequate, in scope and quality, as a thesis for the degree of Master of Science in Engineering of Physics Department.

Assoc. Prof. Dr. Ata SELÇUK
Supervisor

Examining Committee in Charge

Assoc. Prof. Dr. Ata Selçuk

Assoc. Prof. Dr. İ. Engin Türe

Assist. Prof. Dr. Muktim Erdoğmuş

ABSTRACT

PRODUCTION OF THERMOLUMINESCENCE DOSIMETER MATERIALS
[$CaSO_4$]

KÖKSAL Ayda
M.S. İn Engineering of Physics
Supervisor: Assoc.Prof.Dr.Ata SELÇUK
February, 1991 104 pages

The accurate measurement of exposure and dose is important in the monitoring of persons who are accupationally exposed to ionizing radiations. Such measurements have traditionally been carried out using devices such as air ionisation chambers and film badges, and also thermoluminescence (TL) dosimeters are increasingly being used.

The purpose of this thesis is to provide an information about the use of TL teflon dosimeters in ionising radiation dose measurements after preparations of the dosimeters.

In this study, luminescence phenomena, theoretical aspection to TL, prepration of phosphors, measurements, calculations and also conclusions have been discussed.

In this work, the crystal growth of $CaSO_4$ is performed dopping some impurities in different percentage. These impurities are Mn, Mg, Zn, Se and Mn,Mg (double activated). Different properties of the phosphor depending on these dopped materials are investigated and some characteristics like glow curve, dose-response curve and sensitivity stability are also investigated. Some kinetics constans of teflon dosimeters are calculated. These constants are trap energy and frequency factor.

Keywords: $CaSO_4$, thermoluminescence (TL), dosimeter,

ÖZET

TERMOLÜMİNESANS DOZİMETRİK MATERYALLERİN ÜRETİLMESİ
[$CaSO_4$]

KÖKSAL Ayda
Yüksek Lisans Tezi, Fizik Mühendisliği Bölümü
Tez Yöneticisi:Doç.Dr.Ata SELÇUK
Şubat 1991, 104 sahife

Radyasyona muhatap olan kişiler için, doz miktarlarının hassas olarak ölçülmesi önemlidir. Şimdiye kadar radyasyon ölçümleri için uygulanan yöntemlerden bazıları, iyonlaşma odaları, film rozetleri ve dozimetrelerdir.

Bu çalışmanın esas amacı termoluminesans teflon dozimetrelerin hazırlanması ve bunların doz ölçümleridir.

Işıldama olayları, termoluminesansın teorisi, elde edilen fosforların hazırlanışı, ölçümleri,hesaplamaları ve sonuclar bu tezin kapsamındadır.

$CaSO_4$ kristaline çeşitli safsızlıklar katılması ile uygun bir dozimetre maddesi haline gelmesine çalışılmıştır. Kullanılan bu safsızlıklar için Mn,Mg,Zn,Se kullanılmış ve bunlara ilaveten çift safsızlık etkisini görmek içinde Mn ve Mg beraberce kullanılarak denenmiştir.

Dozimetrik özellikler olan hassasiyet, kararlılık ve doza karşı elde edilen ışık şiddetindeki orantılılık incelenmiştir. Dozimetrelerin bazı kinetik sabitleri hesaplanmıştır. Bu sabitler trap enerjileri ve frekans faktörleridir.

Anahtar kelimeler: $CaSO_4$, termoluminesans, dozimetre.

ACNOWLEGEMENT

I would like to thank very deeply the following people:

Assoc. Prof.Dr. Ata SELÇUK for his supervision, help, advice, guidance and his helpfull comments while preparing the present thesis.

Assist Prof.Dr. Muktim ERDOĞMUŞ for his advice during my work, helpfull discussions and providing documents.

Mrs. Servet CANBOLAT for her typing.

I would like to express my gratitude to the reseach assistants and other staff of Department of Engineering of Physics for the help and friendship they have shown herself.

This study would have never been completed without moral support, continuons help and encorgement of my dearest family, therefore my very special thank are due to them.

TABLE OF CONTENTS

LIST OF TABLES

LIST OF FIGURES

PAGE

CHAPTER 1

INTRODUCTION TO LUMINESCENCE PHENOMENA

Most substances that absorb ultraviolet or visible light energy dissipate excess energy as heat, through collisions with neighbouring atoms or molecules. However, a number of important substances lose only part of this excess energy as heat and emit the remaining energy as electromagnetic radiation of a longer wavelength than that absorbed. This process of emitting radiation is collectively termed luminescence, further classified as either fluorescence or phosphorescence. The distinction between the two is based on the mechanism or path by which the substance returns to the ground state, although a practical distinction is based on how soon after absorbtion the emission occurs.

Luminescent materials (luminophors) can absorb energy, store a fraction of it, and convert it into optical radiation which is then emitted. A number of examples of this nomenclature are shown in Table 1.1.

Table 1.1. Luminescence effects

Luminescence effect	Means of excitation
Photoluminescence	Optical photons (ultraviolet,visible, infrared)
Triboluminescence	Rubbing or grinding
Chemiluminescence	Chemical energy
Bioluminescence	Biochemical energy
Cathodoluminescence	Cathode rays
Electroluminescence	Electric field
Radioluminescence	Ionizing radiation
Sonoluminescence	Sound waves
Fluorecence (prompt emission)	Various
Phosphorescence (delayed emission)	
Thermoluminescence (thermally accelerated emission)	

1.1. FLUORESCENCE AND PHOSPHORESCENCE

Fluorescence and phosphorescence are particular forms of luminescence related not to the means of excitation (which for all two may be varied), but to the time scale over which the emission of luminescence takes place.

Luminescence excitation involves the transfer of energy to the electrons and their displacement to a higher energy state. The excess vibrational energy will be dissipate by collisions while the electron remains in the excited electronic state by emission of radiation. Should the electrons return promptly to their original energy state (ground state G) with emission of optical radiation, then this process is called fluorescence Figure 1.1.a.

However, if, due to the presence of an electron trap (metastable state M), the return of electrons to the ground state is delayed, the process is termed phosphorescence, as shown in Figure 1.1.b. The transition of electrons directly from a metastable state to the ground state is forbidden. The metastable state represents a shallow electron trap and electrons returning from it to the excited state require energy. This energy can be supplied in the form of optical radiation (photostimulation) or as the heat (thermal stimulation).

A typical lifetime for an excited electronic state is about 10^{-8} sec.and materials exhibiting fluorescence generally reemit excess radiation within 10^{-8} to 10^{-4} sec. of absorption. The lifetime of phosphorescence is much longer than fluorescence, generally ranging from 10^{4} to 20 sec. or longer.

Before the luminescence can occur, a substance must absorbed radiation of proper wavelength. When a quantum of light of the proper wavelength impinges on a molecule, it will be absorbed in about 10^{-5}sec (the time required for a molecule to go from one electronic state to onother). An excited molecule can exist as such for only about 10^{-7} and 10^{-8} sec, before it will eliminate some or all of the excess energy. One of the tree possibilities will happen to excited molecule:

1. It will reemit a photon of the frequency as it absorbed (resonance emission).
2. It will emit an infrared photon, thus losing vibrational energy and ending up at the lowest vibrational level of the excited electronic state or.
3. It will undergo radiationless loss of vibrational energy through collisions, ending up in the lowest vibrational level of the excited electronic state.

The first two alternatives are extremely rare, and in practice these occur only with gases at very low pressure.

Once a molecule arrives at the lowest vibrational level of an excited state, it again has three possible modes by which it may lose the remaining excess energy

1. It may undergo radiationless loss of electronic energy through collisions or other interactions.
2. It may emit an ultraviolet or visible (fluorescent) light photon; or
3. It may undergo a transition to a metastable triplet state and some time there after return to the ground state, usually by emission of an ultraviolet or visible (phosphorescence) light photon.

1.2 ENERGY ROTATIONS FOR SOME KIND OF LUMINESCENCES

According to another expression, fluorescence and phosphorescence change of energy for atoms and molecules. Some energy transformations are shown in Figure 1.1. In this Figure, G is ground energy state of atom or molecule, D and M are the semistable energy states. There are resonance radiation , irradiation, and reflection radiation within the part of a. However, because of the absorbtion of the radiation at the energy state E_f, the changing of energy occurs on the atom or molecule. Therefore, the molecule which absorbed energy, excited to F energy state from G. Excited molecule immediately return to the G state in about 10^{-3} second, before it can eliminate the energy which is equal to its absorbed energy [2].

Delayed fluorescence (phosphorescence) effect is shown in Figure 1.1 b. At this position the radiation energy which have same wavelength with absorbed energy, is emitted. The absorbed energy is stored during a period of time at semistable state M. The atoms or molecules return back to the M level after secondary collisions. The energy difference between them is refered as the kinetic energy of the collided molecules. The molecules at M state immediately excited to the energy level F from semistable state. For this transition position, the required energy taken from the kinetic energy of the collided molecules. The raised atoms to the F level emit irradiation and return to the G ground state. According to Stokes law, the wavelength of the emitted radiation at fluorescence effect is greater than the wavelength of the excited radiation. This means that, after the absorbtion of radiation, the molecules excite to F state, they return to the low energitic state C during irradiation. The frequency of this radiation, is $h\nu = E_f - E_c$.

The fluorescence effect does not agree with Stokes'law as shown in Figure 1.1 d and e. The excited molecules on the H state is translated to the F state with known wavelength of radiation. After then this molecule which emits a low wavelength radiation, falls to the G ground state. Sometimes, as in Figure 1.1.e, the excited molecules at F raise to the upper energy state O with first type collisions or absorbtion of additional radiation. And then it makes irradiation which wavelength is smaller than the first absorbed radiaton wavelength.

The thermolumincscence and phosphorescence effects are as shown in Figure 1.1.f.. The molecules or atoms absorb the energy which come from the raising temperature and then may excite to the F state. They may fall back to the another law energy state with irradiation.

The molecule collides to other one, (so that they are on the level D,C, or M), and then one of them rasises to the non-semistable state F In a period of time it may return to the level G with irradiation, (delayed fluorescence).

At a phosphor, the electrons are normally in the filled band and they have constant energies. Becouse of the rays (X or gamma rays), they raise to the upper energy level that is conduction band during irradiation. The moving electrons at the conduction band are captured by electron traps or level F in the crystal. They emit their energies in the form of the light or the other ways.

The electrons which captured in the electron traps, are excited to the conduction band by heating of the crystal. Until they are captured by the trap centers, they make motion. However they emit radiation and settle certain position in the emission centers.

1.3. THERMOLUMINESCENCE

When a substance is exposed to ionizing radiation or ultraviolet light, or when it undergoes certain chemical reactions or mechanical stress, a varying percentage of liberated electrons (or holes) may become traped at certain imperfections in the latice which are called electron (or hole) traps. If the temperature is low and/or the traps are deep enough, they may remain trapped before they are released by a

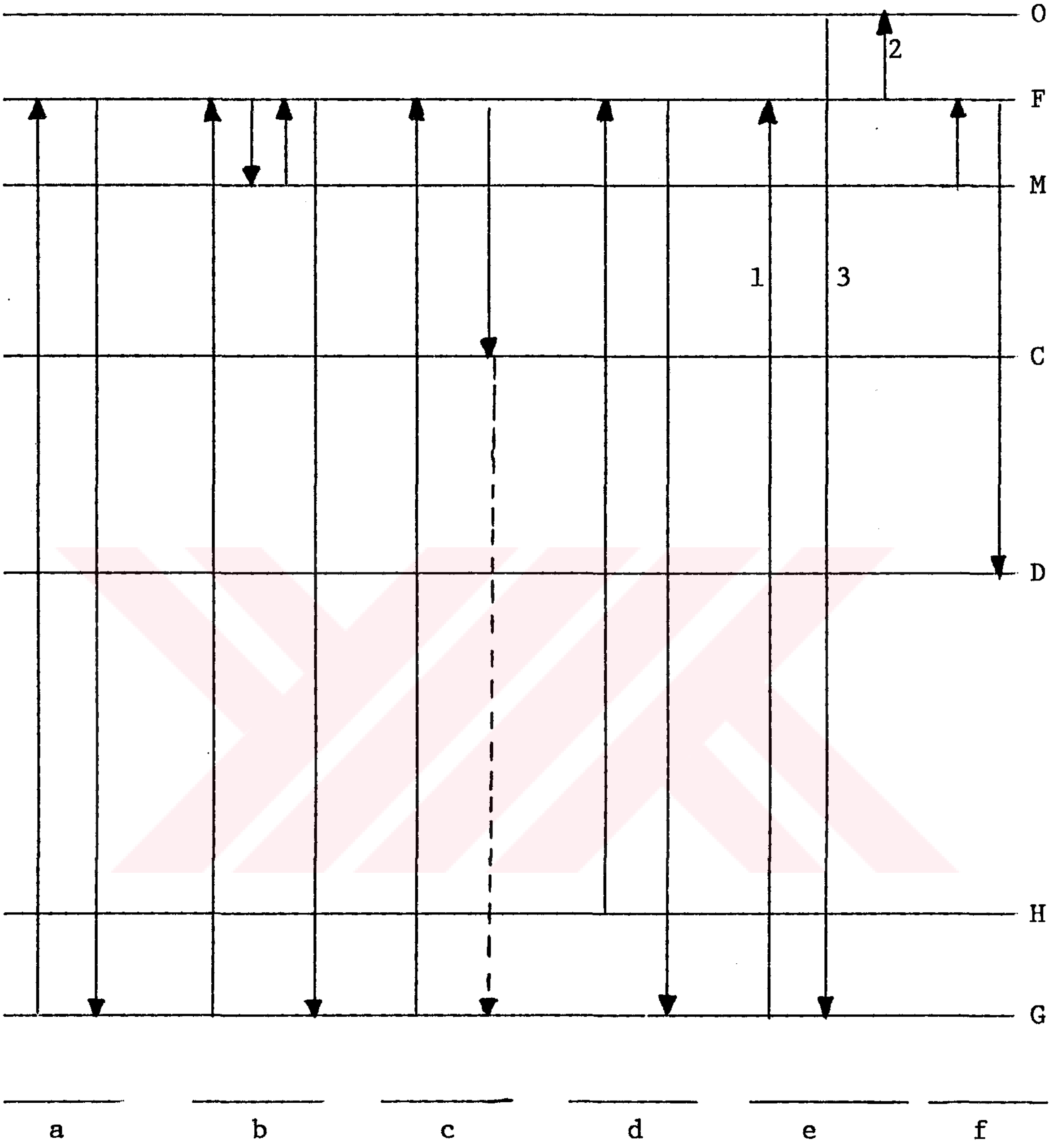

Figure 1.1 Energy rotations for some kind of luminescences.

sufficient "stimulation" which increases their probability of escaping from the traps. Stimulation usually consists of the transfer of optical (light) or thermal energy to the substance . The probability 'p' of escape of an electron from a metastable state to an excited state is governed by the Boltzmann equation.

$$p = S \exp (- \Delta E/kT)$$

where S is a frequency constant, ΔE is the energy difference between states E (excited state) and M,(commonly called the trap depth), k is Boltzmann's constant, and T is the temperature in Kelvin [1].

The return of the electrons (or trapped holes) to the stable state, which is called annealing (or fading if it proceeds slowly), is associated with a released as visible or ultraviolet light, which can be observed as a transient "glow" of the pre-irradiated crystal or glass at a charecteristic temperature during heating. By raising temperature, the probability of escape of an electron is increased, and this effectively accelerates the phosphorescence process as progressively deeper metastable states empty with increasing temperature. This process is called thermoluminescence and is quite different from incandesence, in which the application of heat causes vigorous vibrations, collisions and excitation of all the atoms of a material. The thermoluminescence emission spectrum of a material depends on the species of luminescent atoms present. The term thermoluminescence produced as a result of the absorbtion of ionising radiation, in accordance with common practice. If after removal of excitation, the delay of excited atom is of the order of 10 ns or so, the process is undoubtedly fluorescence, whereas should the emission persist for a period longer than 100 m sec (which the eye will resolve) it is undoubtedly phosphorescence. For intermediate decay periods it may be necessary to examine the effect of raising the temperature of the material to decide whether the emission is fluorescence or phosphorescence. Fluorescence is essentially independent of temperature, whereas, as we have seen, phosphorescence is accelerated by a rise in temperature according to Boltzmann's equation.

If the several traps of different depths are involved in the process, the recording of the light emission as a function of heating time (or temperature) results in a "glow curve" consisting of several "glow peaks". For quantitative measurements, the obtained signals are amplified and recorded. When the intensity of radiation is drawn versus time or temperature, a curve is obtained which name is GLOW CURVE. The graphic obtained by drawing light intensities versus radiation doses is the CALIBRATION CURVE or DOSE-RESPONSE curve.

The peak location is fairly constant for a given TL crystal or glass (with or without "activators") and the light sum under the peak can be made proportional to the radiation dose [3].

1.3.1. THEORY OF THERMOLUMINESCENCE

The theory of thermoluminescence and some closely associated phenomena that are also based on trapping processes, including phosporescence, thermally stimulated current and thermally stimulated exoelectron emission are not yet completely understood.

When some materials are heated in a dark place, they emit radiation during raising of temperature. If the temperature is held constant during radiation, it was seen that the intensity of radiation will be decreased with respect to time. The velocity of the decreasing intensity is depend on the temperature which held constant.The intensity of radiation obtained from heating depends on material, temperature and the velocity of raising temperature.

Usually, discrete energy levels and certain transitions are assumed, and the parameters are adjusted to fit experimental glow curves and fading rates.

The production of thermoluminescence in a material by exposure to ionizing radiaton may be divided into two stages:

a) Ionisation and electron trapping,

b) Electron and hole recombination with photon emission.

As seen in Figure 1.2a. interaction of ionizing radiation with the phosphor results in the transfer of sufficient energy to electrons in valence band (VB), for transffering them (δ) into conduction band (CB). This process, which requires about 10 to 15 eV is usually due to the effect of secondary electrons which are produced in environmental of primary photoelectron or charged particle tracks. The delibrate introduction of impurities into crystals increases the number of traps and can increase the thermoluminescent efficiency of the material. Most thermoluminescence dosimeter materials contain these "activators". The centers to which they give rise can have their own characteristic energy levels and transitions between them give rise to emission of photons of corresponding characteristic energies. A varying percentage of thus "liberated" electrons recombine within a very short time with activators A(γ) ,releasing some of its energy in the form of light (fluorescence), while the remaining electrons are captured in electron traps H(β). The trapped electrons will remain in their traps provided that they do not acquire sufficient energy to escape. This will be determined by two main factors; the depth of the traps, and the temperature of the material. If the temperature of the material is raised trapped electrons may acquire sufficient thermal energy and be released. Released electrons may recombine with holes at luminescence centers and the excess energy is radiated as visible or ultraviolet photons. While electron capture and delayed recombination with a hole at luminescence center is mechanism of TL. [5] Figure 1.5.

If the traps are not very deep, detrapping and recombination may already occur at a substantial rate around room temperature, resulting a short halflife of the stored energy (phosphoresence). Only if the traps are deep enough to result in sufficient storage stability at room temperature (halflives of trapped electrons at least several weeks or months, normally corresponding to glow peaks at temperatures exceeding

100 to 200^{o}C) , the effect is called thermoluminescence and becomes of dosimetric interest.

In the case of true "thermoluminescence", the electrons are liberated (α) by the transfer of thermal energy (heating) as in Figure 1. b, representing a "bimolecular" model. The electrons either are retrapped (β) or recombine with activator (γ), the emitting part of their energy (< 1%) as visible or ultraviolet light. Before retrapping or recombination takes place, the electron population in CB can be observed as a thermally stimulated electrical conductivity. Such conductivity curves closely parallel to the TL glow curves under certain conditions.

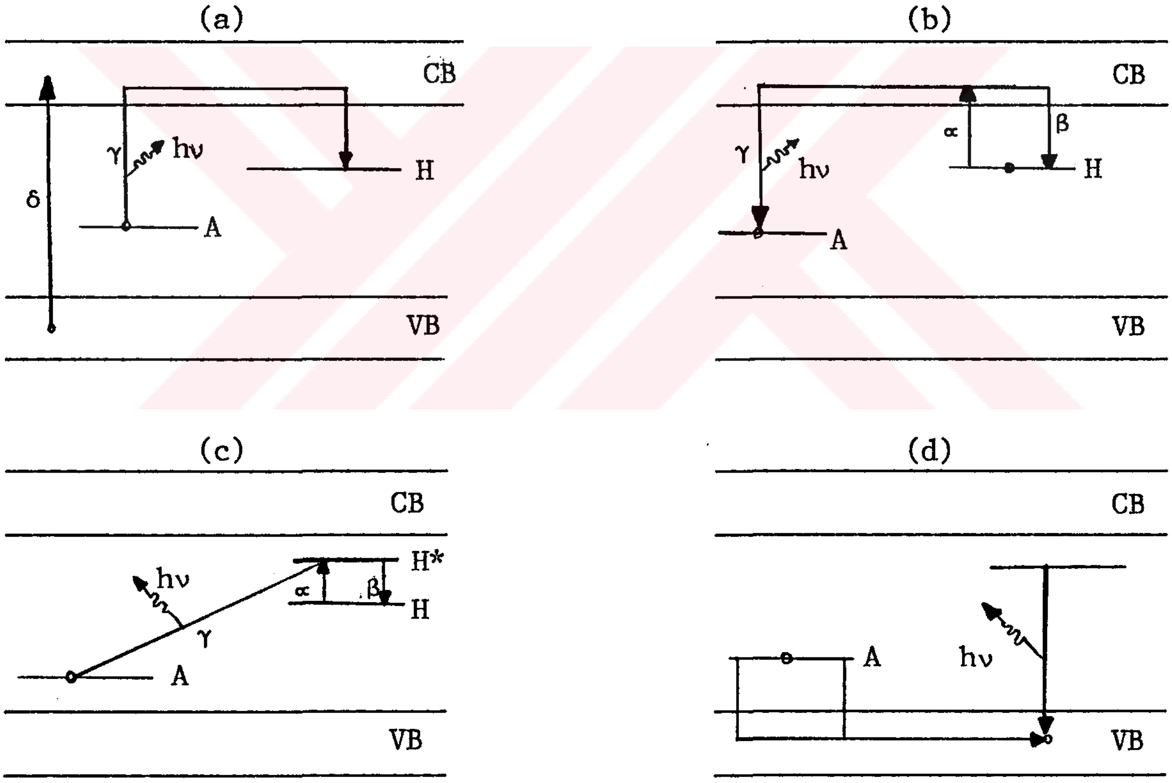

Figure 1.2 a) Simplified model of the processes in a TL phosphor during exposure to ionizing radiation

b) And during heating according to the bimolecular model

c) The monomolecular model

d) If the hole trap (defect electron trap) is stable, it can be released and recombine with an electron

1.3.2 THE ROLE OF LATTICE DEFECTS IN THE THERMOLUMINESCENCE

All real crystals contain lattice defects of various kinds and these play an all-important role in the thermoluminescence process. There are three types of lattice defects:

a) Thermal or intrinsic defects;

b) Extrinsic defects or substitutional impurity ions,

c) Radiation-induced defects.

The presence of defects in a material is important for the TL process. For the moment we will consider only the role of intrinsic defects in the electron trapping process.

A negative ion vacancy, illustrated in Figure 1.3b.is essentially a region of excess positive charge and as such may be regarded as a potential trap for a free electron. Electron capture creates an F centre. Similarly,a region of excess negative charge will be a potential trap for free positive charges (holes). The analogue "anti-centre" of an F centre whould be a hole trapped at a positive ion vacancy (a V_F centre), but there is doubt as to whether this particular centre generally exists [4].

The energy band structure for an ideal crystal may be represented by an energy diagram as shown in Figure 1.4a. The valence band is representative of all electrons held in bound states, and the conduction band is representative of all electrons in unbound states which are free to migrate through the crystal lattice. In the case of ideal electrically insulating crystal under discussion the CB (conduction band) will be empty and all electrons will reside in the VB (valence band). The CB and VB are widely seperated in energy by so-called "forbidden gap". Without the influence of external forces it is highly improbable for an electron to be able to traverse the forbidden gap from VB to CB. However, in the case of a real disordered crystal containing defects of a simple and/or complex nature, other allowed energy levels exist in the forbidden gap region,as seen in Figure 1.4b

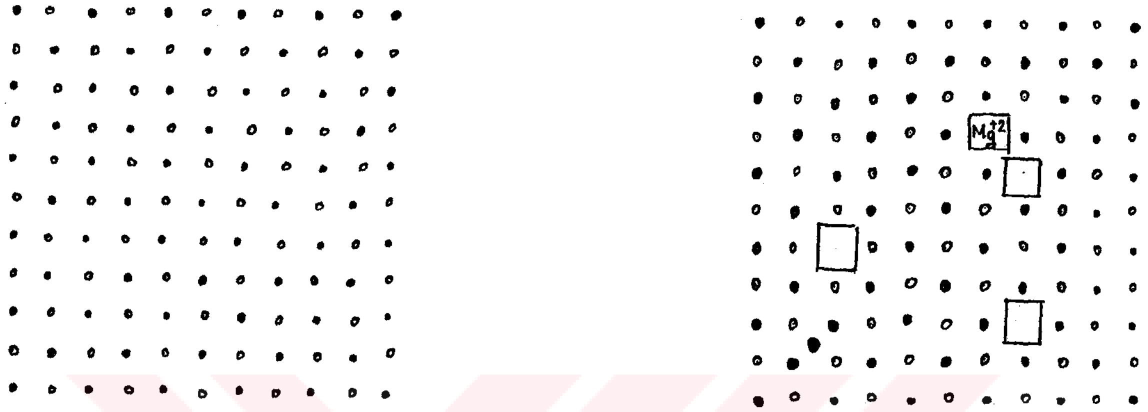

Figure 1.3 Ionic structure of
a) an "ideal" perfect alkali halide crystal,
b) a "real" imperfect crystal containing defects of various types. Also shown is a divalent magnesium ion-alkali ion vacancy dipole. ● halogen ion, o alkali metal ion.

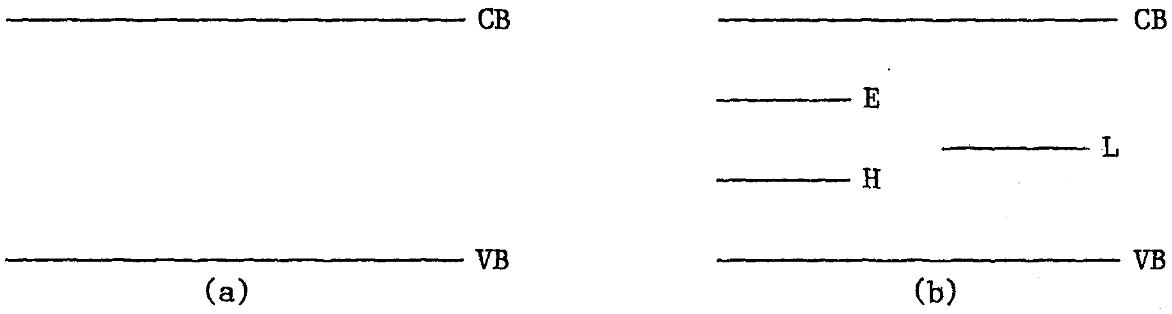

Figure 1.4 a) Energy band structure for an ideal crystal,
b) Energy band structure for a real disordered crystal containing defect.

In the description of the general model which follows we shall suppose that the energy level labelled E represents an electron trap and that level H represents a hole trap. L is a luminescence centre where electrons and holes may recombine with photon emission.

1.3.3 A GENERAL MODEL FOR TL.

The production of thermoluminescence in a material by exposure to ionizing radiation may be divided into two stages:

a) Ionization and electron trapping,

b) Electron and hole recombination with photon emission Figure 1. 5 , illustrates the energy band configuration for each stage.

Ionizing radiation is absorbed in the material and free electrons are produced. With respect to the energy band diagram this is equivalent to transferring electrons from the valence band to the CB (step 1). These electrons are now free to move through the crystal (step 2), but if trapping levels such as E are present, the electrons may become trapped (step 3). The production of free electrons is associated with the production of free positive holes which may also migrate, in energy terms, via the valence band (step 2'). The holes may become trapped (step 3').

The trapped electron centers produced, such as F centers, are lattice defect centers, and such their properties are primarily determined by the lattice and the defect. Many hole centres are thermally unstable and may decay rapidly at normal room temperature (step 4). If the temperature of the material is increased, the electron which is trapped may acquire sufficient thermal energy and be released (step 4'). The electrons which are released can recombine with holes at luminescence centers such as L, and the excess energy is radiated visible or ultraviolet photons (step 5). There is TL mechanism in where electron capture and delayed recombination with a hole at luminescence centre. The electron hole recombination processes are possible. For example, immediate are delayed recombination with subsequent thermal degration of energy without photon emission, and fluorescence caused by the immediate recombination of holes and electrons at luminescence centers [5].

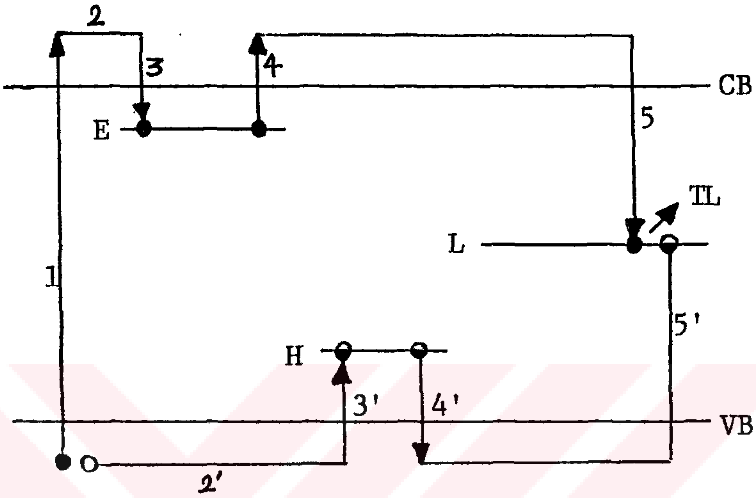

Figure 1.5. A simple energy band model for TL, ● electron, o hole.

In real phosphors many different trapping levels will be present, each one due to a particular lattice defect or complex of defects. Each trapping level will give rise to an associated glow peak maximum which may or may not be resolved during readout. The area and peak height of each glow peak depends on the number of associated electron traps present. This in turn depends on the type and amount of impurity atoms present, on the thermall history and treatment of the material.

The TL phosphor material that is in the form of a loose powder a plastic compound, glass, sintered disk, or single crystal (or encapsulated permanently in a glass capillary, glass bulb, etc.) is heated by electrical resistance heating or exposure to a hot gas. Usually the temperature of the phosphor carrier is monitored during the heating. After passing through infrared and/or additional optical filters, the emitted light is detected by a photomultiplier and the light output is recorded either on a current versus time or a current versus temperature chart, whith a peak-height-recording microamper, or with a charge-integrating circuit.

Varying dose, linear energy transfer (LET),phosphor pretreatment, heating rate, and reader desing (PM tube, filters, etc) can all modify peak height and location.

CHAPTER 2

RADIATIONS AND UNITS

Radiations can be classified into two main groups as electromagnetic radiations and particles.

2.1. ELECTROMAGNETIC RADIATIONS

Radio waves, infrared, visible light, ultraviolet, X rays and gamma rays are types of electromagnetic radiations. Electromagnetic radiation travels through space in a wave motion. The classification of the electromagnetic radiations is shown in Figure 2.1.

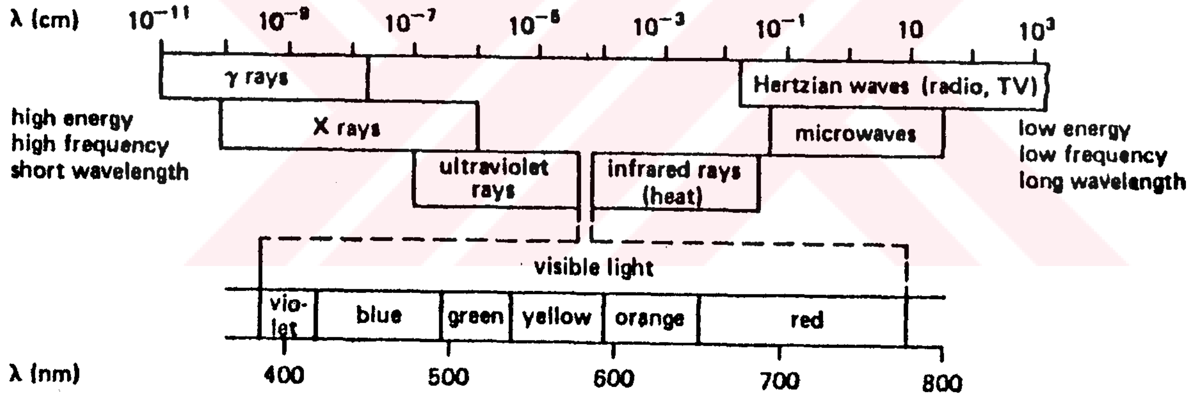

Figure 2.1 Electromagnetic radiation. Note that the approximate ranges of electromagnetic radiations are plotted on a logarithmic scale in the upper part of the diagram. The spectrum of visible light is not plotted in this way.

2.1.1 RADIO WAVES

It can be produced by fast electrical vibrations. Radio waves have very long wavelengths. They are determined by electrical devices.

They have low energy and low frequency. They make reflection from conductors and ionized atmosphere layers.

2.1.2 INFRARED

Infrared lights are produced by molecular vibrations or by the exitation of outer electron of the atom. They are determined by thermometer and some kind of photograph films. They have longer wavelength than red light.

2.1.3 VISIBLE LIGHT

Visible light are produced by molecular vibration and the excitation of the outer electron of electron layer. They can be observed by eyes, films,and photoelectric cell. They have seven colours between red and violet.

2.1.4 ULTRAVIOLET LIGHT

Ultraviolet lights are produced by the excitation of the outer electrons of the atom. It can be seen by photograph films, photoelectric cell. Their effects are on the bacteria and skin.

2.1.5 X-RAYS

The emission of characteristic X-rays induced by charged-particle beams has been used for elemental analysis of thin samples (≤ 1 mg/cm^2) and small areas (a few square milimeters). The technique inuolves the observation of characteristic X-rays emitted when atomic inmer-shell vacancies created by particle bombardment are filled from outer shells. Therefore, X-rays produced by the bombardment between cothod rays and target. This ray is an electromagnetic radiation. The cathod rays are electron beam, sothat, the fast electrons slowly down by target and X-rays are produced.

2.1.6 GAMMA RAYS

A nucleus in an excited state may give up its excitation energy and return to the ground state in a variety of ways. The most obvious, and the most common, transition is by the emission of the high energitic electromagnetic radiation. Such radiation is called γ radiation, the gamma rays have a frequency determined by their energy $E = h\nu$. Frequently the transition does not proceed directly from an upper state to the ground but may go in several steps involving intermediate excited states. Gamma rays with energies between a few thousand electron volts and about 7 MeV have been observed in radioactive processes. All the processes just deseribed we call γ transitions, although only in the first is a γ ray emitted by the nucleus. All are characterized by a change in energy without change in Z and A [6].

Photons passing through matter do not lose energy continuously along their paths as do charged particles. On the contrary, in two of the three fundamental processes through which photon interact with matter the entire energy of the photon is transferred to the medium in a single interaction; in the third there is a small probability for a few energy- degrating encounters. The ionization observed for γ rays is almost entirely secondary in nature, from the three processes by which γ rays (and X rays) lose their energy. These affects are

a) Photoelectric effect,

b) Compton effect,

c) Pair production

2.2 PARTICLES

Electrons, protons, neutrons, photons, and neutrinos are abundant in our environment. Electrons,protons, and neutrons are the constituents of the atoms in our own bodies and in everything that surrounds us. Photons and neutrinos are emitted copiously by the sun; furthermore, high-energy photons and neutrinos are emitted by natural radioactive substances. All these particles with the exception of free neutrons, are stable: If left to themselves, they live forever. Extraordinary particles that are created by the conversion of energy into mass in high energy collisions between the everyday particles. Most of new particles are extremely unstable. A few are stable when left to themselves, but they are made of antimatter, which annihilates when it comes into contact with the ordinary matter of our immediate environment.

2.2.1 ELECTRON

This is the unit of the electrical charge which associated with atom. The charge on a single electron is

$$e = - 1.602\text{x}10^{-19} \text{ coul.}$$

and the mass of the electron is given by

$$m_e = 9.1096\text{x}10^{-28}\text{gr.}$$

2.2.2 PROTON

If one or more electrons are removed from a neutral atom or molecule, the residue has a positive charge equal to the sum of the negative charges of the electrons removed. When different gases are used in discharge tube, different types of positive ions are produced. When the hydrogen gas is used a positive particle results that has the smallest mass of any positive ion can be observed. The proton has a charge equal in magnitude to that of the electron but opposite in sign,

$$e = +\ 1.602\text{x}10^{-19}\text{coul.}$$

This is called unit electrical charge. The mass of proton which is 1836 times electron mass.

$$m_p = 1.672\text{x}10^{-24}\ \text{g.}$$

2.2.3 NEUTRON

This is an uncharged particle. It has mass which is very slightly larger than that of proton.

$$m_n = 1.6749\text{x}10^{-24}\ \text{g.}$$

2.2.4 PHOTON

The photon as a particle is the quanta of the electromagnetic field. Monochromatic radiation of freguency w comes in packets carrying an energy $E = hw$.

2.2.5 ALPHA RAY

It consists of particles that carry a 2^+ charge and have a mass about four times that of proton. These particles are ejected from the radioactive substance at speeds around 16,000 km/s. They have two protons and 2 neutrons.

2.2.6 BETA RAY

They are stream of electrons, that travels at appreximately 130,000 km/s. H have the charge of componen 1^-.

2.2.7 COSMIC RAY

They were subsequently produced artificially by the bombardment of targets with beams of high-energy protons are electrons from an accelerator.

2.3 RADIATION UNITS

Radiation units classified into two main groups. These are absorbtion units and emission units.

2.3.1 ABSORBTION UNITS

The absorbtion units are rontgen, rad and rem. These are explained below.

2.3.1.a RONTGEN

A unit of radiation exposure is the Rontgen, R. The rontgen is defined as "that quantity of X or γ radiation such that the associated corpuscular emission per 0.001293 gr of air produces, in air, ions carrying 1 esu of electricity of either sign". This means that 1 R produces 1.61×10^{12} ion pairs per gram of air, which corresponds to the absorption of, 84 ergs of energy per gram of air. In water the energy absorption cornesponding to 1 R is about 93 ergs/gram or 0.93 rad for all X or γ ray energies above about 50 keV.

2.3.1.b RAD

The rad (radiation absorbtion dose) is a quantitative measure of radiation energy absorption (usuall called the dose). A dose of 1 rad deposits 100 ergs/gr of material.

In IS system, the unit of dose is the gray (Gy) which is defined as 1 J/kg, so that 1 Gy = 100 rad.

2.3.1.c REM

The unit of radiation dosage that is used in radiation is the rontgen equivalent man (rem). The dosage in rems is equal to that in rads, multiplied by the relative damage cavsed by various kinds of radiation. The latter quantity depends on several factors, the most important of which is the density of ionization that in biological studies is often measured by the linear energy transfer (LET) the energy that is deposited per unit path.

2.3.2 EMISSION UNITS

The emission units are curie and becquerel. They are discussed below.

2.3.2.a CURIE

The curie (Ci) is a unit of radioactivity originally based on the disintegration rate of 1 gr of radium, but now defined as the quantity of any radioactive nuclide in which the number disintegrations per second (dis/sec) is 3.7×10^{10}.

2.3.2.b BECQUEREL

The IS unit of radioactivity is the becquerel, defined as 1 dis/sec.

CHAPTER 3

THERMOLUMINESCENCE KINETICS

3.1 INTRODUCTION

The kinetics of a reaction are related with the information about its reaction velocity, order, and reaction mechanism. There are some factors, sothat they increase or decrease the reaction velocities named as cathalist. All of the steps within the reactions represent the mechanism of the reactions.

The glow curve theory of Randall and Wilkins (1945) is used and assumes that no retrapping of released electrons occurs (first order kinetics) [7] . The case of equal retrapping and recombination probabilities is called second-order kinetics. Indeed the trapping and recombination processes may not be limited to first- or second-order kinetics but may follow a general "order" [8].

In this chapter, there are some thermoluminescence models and some methods of determination of the kinetics constants, e.g. E (trap energy) and S (frequency factors), etc.

3.2 MODELS OF THERMOLUMINESCENCE

3.2.1 SEITZ MODEL

According to "SEITZ" model, the electrons which are excited from stable state to the semistable state can be captured by traps or can back to the ground state [9].

If the electrons release from traps, they make some translations. These transitions are optical transitions that electrons return to the ground level without radiation or translate to the any level by "$h\nu$"

photon radiation.

3.2.2 RANDALL - WILKINS MODEL

At this model, the probability of the thermal release of an electron from a trapping centre is represented by the equation 3.1.

$$p = S\ \exp(-E/kT) \qquad 3.1$$

where, S is the frequency factor,(1/sec)
T is absolute temperature (^{o}K)
k is the Boltzmann's constant (8.6×10^{-5} eV/^{o}K)
E is the trap dept (eV)

If n is the number of the traps having electrons, the luminescence intensity I is given by

$$I = -C\,(dn/dt) \qquad 3.2$$

where C is the proportionality constant which is related with luminescence efficiency.

The reaction velocity is

$$-\frac{dn}{dt} = np \qquad 3.3$$

where n is the number of the occupied traps at any time t.

If the material is heated at a uniform heating rate,(b),

$$\frac{dT}{dt} = b \qquad \text{(degree/second)} \qquad 3.4$$

substituting b and p in equation 3.2

$$\frac{dn}{dT} = (n\frac{S}{b}) \times (e^{-E/kT}) \qquad 3.5$$

integration of the equation

$$n = n_o \exp - \int_{T_o}^{T} (S/b) \exp(-E/kT)\, dT \qquad 3.6$$

is obtained. From equations 3.1, 3.2, 3.4,

$$I = C\, n\, S \exp(-E/kT) \qquad 3.7$$

is found. If n replaces by its value in equation 3.6

$$I = C n_o\, S e^{-E/kT} \int_{T_o}^{T} (S/b)\, e^{-E/kT}\, dT \qquad 3.8$$

and, dI/dT = 0 gives the maximum point of the glow curve,

$$E/kT^2 = -\frac{S}{b}\; e^{-E/kT} \qquad 3.9$$

where, T is maximum temperature, E trap dept for this peak.

When two different temperatures for heating rates used and when one of the equation divided by the other side by side, as a result equation of.

$$\frac{T_2^2}{T_1^2} = \frac{b_2}{T_1^2}\, e^{-E/k\,(1/T_1 - 1/T_2)} \qquad 3.10$$

$$\ln \frac{b_1}{b_2}\; \frac{T_2^2}{T_1^2} = \frac{E}{k}\left(\frac{1}{T_2} - \frac{1}{T_1}\right) \qquad 3.11$$

and

$$E = k \frac{T_1 - T_2}{T_1 \times T_2} \ln\frac{b_1}{b_2} + 2 \ln \frac{T_2^2}{T_1^2} \qquad 3.12$$

is obtained. This formula can be used for calculation of the trap depts (trap energy).

3.3 TRAP ENERGY DETERMINATION METHODS

3.3.1 PEAK TEMPERATURE METHOD

In this method, the temperature which corresponds to the maximum peak of glow curve is the maximum temperature. For the calculation of the activation energy URBAH [10] made a good approximation.

$$E = \frac{T_m\ (^{o}K)}{500} \qquad 3.13$$

3.3.2 SINGLE PEAK INITEGRATION THERMOLUMINESCENCE METHOD

The single peak integration thermoluminescence method can be used for calculation of the trap energies.

Since the complete peak is used for the calculations the method is named as "Single Peak Integration Method" [2] .

According to RANDALL-WILKINS [7] model of thermoluminescence, the light intensity detected by heating the sample is given as

$$I = - \frac{dn}{dt} C \qquad 3.14$$

where n is the number of the electrons located at the traps at moment t; C is the proportionality constant.

As known the luminescence intensity I is proportional to the escape speed of the electrons from the traps, and the escape speed is also proportional to both the number of electrons trapped and the possibility of escape of the electrons from the traps. So

$$-\frac{dn}{dt} = np \qquad 3.15$$

is valid. For the monoenergetic traps escaping possibility is given as

$$p = S \exp(-E/kT) \qquad 3.16$$

where p is the escaping possibility, S is the frequency factor, E is the trap energy k is the Boltzmann constant, T is the absolute temperature. From the equation 3.14

$$I\, dt = -C\, dn$$

integrating the equation

$$\int_0^t I\, dt = \int_{n_o}^{n} -C\, dn$$

is obtained. The left side of the equation is the area under the glow curve for the time interval of (0-t). So

$$A_1 = C\,(n_o - n)$$

is obtained. Where A_1 is the area under the glow curve peak in the interval of (0-t). As a result, the area under the glow curve peak in any time interval have been found proportional to the number of the traps have been emptied. When all occupied traps being empty, the full area under the complete glow curve peak can be expressed as

$$A = C n_o \qquad 3.17$$

In case the constant heating rate is used for the experiment the equation of the heating rate

$$\frac{dT}{dt} = b \qquad 3.18$$

is valid. Using equation 3.15 and 3.18

$$-\frac{dn}{dT} = n\frac{p}{b}$$

is obtained. Integrating the equation

$$\int_{o}^{n} \frac{dn}{n} = \int_{T_o}^{T} -\frac{p}{b}\,dT$$

$$\Big|_{n_o}^{n} \mathrm{Ln}\, n \Big| = \frac{p}{b}\,(T_o - T) \text{ and } \mathrm{Ln}\,\frac{n}{n_o} = \frac{p}{b}\,(T_o - T) \qquad 3.19$$

is found. Since $A_2 = Cn$ and $A = Cn_o$ were already defined

$$\ln \frac{A_2}{A} = \frac{p}{b}\,(T_o - T) \qquad 3.20$$

is obtained. Defining an a value as

$$a = \frac{A_1}{A} \qquad 3.21$$

and substituting $A_2 = A - A_1$ (as seen from Figure 3.1) into the equation the result is obtained as

$$\ln (1-a) = \frac{p}{b}\,(T_o - T) \qquad 3.22$$

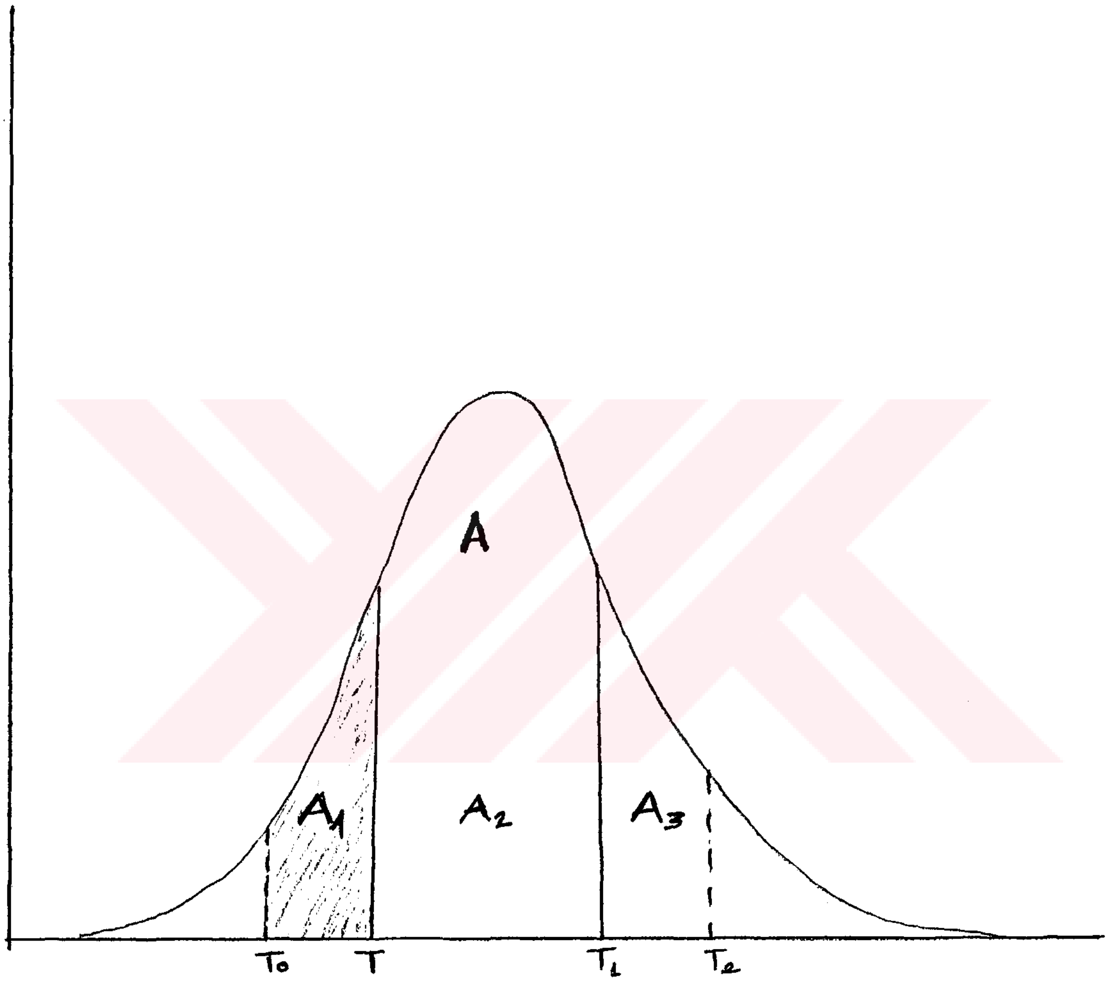

Figure 3.1 A graph which can be used for calculation of area in single peak integration TL method.

Assuming that the reaction is isothermal in a differential temperature or time interval it will be possible to find out a 'p' value for every one temperature and corresponding 'a' value. So 'p' values of different temperatures can be evailable.

Since the probability is given as

$p = S \exp(-E/kT)$ the natural logarithm yields the following equation

$$\mathrm{Ln}\, p = \mathrm{Ln}\, S - \frac{E}{kT} \qquad 3.23$$

The graph of lnp versus 1/T gives a straight line having a slope of (-E/k) where the trap energy E can be calculated. Equation 3.23 also gives the frequency factor by the y intercept which is equal to ln S.

CHAPTER 4

EXPERIMENTS

4.1 INTRODUCTION

In this chapter, the preparations of the crystals and dosimeters are explained.The information of thermoluminescence dosimeters produced in ionizing radiation dose measurements are investigated.

The kinetics constants of the thermoluminescence dosimeters are calculated for different types of materials. For the calculation of E (trap enegy) and S (frequency factor), the single peak integration method is mainly preferred. $CaSO_4$ is used for preparation of the phosphors with some activators. The impurities are Mn, Mg, Se, Zn and both Mn,Mg to investigate the performance of the product thermoluminescence materials, fixed teflon dosimeters prepared.

The accuracy, sensitivity, linear response of the dosimeters are investigated to decide the convenient material.

4.2 PREPARATIONS OF THE THERMOLUMINESCENCE DOSIMETER PHOSPHORS

For the preparation of $CaSO_4$ teflon dosimeter with impurities, the general form is given in the first sample. For the other dosimeters, there are only special prepration properties are listed.

4.2.1 $CaSO_4$:Mn-1

$CaSO_4.2H_2O$ and %1 mole $MnSO_4.H_2O$ are mixed with 2 ml H_2SO_4 which are diluted to %25. The mixture is left at room temperature for 24 hours. The liquid in the mixture is driven off by heating for 2 hours at 100 °C , and then 3 hours at 300 °C for H_2SO_4. The residue is heated for two hours at 900 °C. And it is grounded and sieved by 230 mesh. After that the sample is pressed with %75 teflon powder to form of 10 mm diameter and 1 mm thick disc. The total weight of this dosimeter is 200 mg.After then it is heated for 20 minutes at 310 °C.

4.2.2 CaSO$_4$:Mn - 2

% 2 mole MnSO$_4$.H$_2$O
20 hours at room temperature
3 hours at 100 °C
2 hours at 300 °C
2 hours at 900 °C
Mesh no: 230
%76 teflon and %24 phosphor
Total weight of the sample is 230 mg.

4.2.3 CaSO$_4$:Mg

%1 mole MgSO$_4$.H$_2$O
24 hours at room temperature
3 hours at 100 °C
2 hours at 300 °C
2 hours at 900 °C
Mesh no: 230
%70 teflon and %30 phosphor
Total weight of the sample is 210 mg.

4.2.4 CaSO$_4$Zn

%5 mole ZnSO$_4$.H$_2$O
20 hours at room temperature
3 hours at 100 °C
3 hours at 300 °C
3 hours at 900 °C
Mesh no: 230

%70 teflon and %30 phosphor
Total weight of the sample is 250 mg.

4.2.5 $CaSO_4$:Se

%1 mole SeO_2
21 hours at room temperature
3 hours at 100 °C
3 hours at 300 °C
2 hours at 900 °C
Mesh no: 200
%80 teflon and %20 phosphor
Total weight of the sample is 230 mg.

4.2.6 $CaSO_4$:Mn,Mg

%3 mole of $MnSO_4.H_2O$
%3 mole of $MgSO_4.H_2O$
23 hours at room temperature
3 hours at 100 °C
3 hours at 300 °C
2 hours at 900 °C
Mesh no: 230
%76 teflon and %24 phosphor
Total weight of the sample is 250 mg.

4.3 PROPERTIES OF DOSIMETERS

4.3.1 $CaSO_4$:Mn - 1

This dosimetric sample is exposed to Sr-90 β radiation. The glow curves of this crystal are shown in Figure 4.1. In this figure, the curves 1, 2, 3, and 4 represent the glow curves of 7.5 Rad, 15 Rad, 22.5 Rad and 33 Rad, respectively. The dose response curve of this teflon dosimeter is shown in Figure 4.2. The maximum peaks are 137,5 °C for 7,5 Rad and 15 Rad, 134.5 °C for 22.5 Rad, and 140 °C for 33 Rad.

4.3.2 $CaSO_4$:Mn - 2

The light signals from the dosimeter exposed to Sr-90 β radiation are shown in Figure 4.3. This sample is exposed to the radiation for three times. First curve is for 30 Rad, second curve is for 75 Rad, and the third one is for 150 Rad. The maximum peak temperatures are 133.5 °C, 135 °C, and 137 °C for 1., 2., 3., curves respectively. The dose response curve of $CaSO_4$.Mn - 2 is shown in Figure 4.4.

4.3.3 $CaSO_4$:Mg

This sample is exposed to the Sr-90 β radiation. The glow curves of the phosphor are shown in Figure 4.5. The first curve which is represented by 1 is for 75 Rad, the second curve 2 is for 150 Rad, third curve 3 is for 225 Rad, and, the fourth one is for 300 Rad. The maximum peak temperature is 160 °C for four curve. From the glow curves of $CaSO_4$.Mg, the dose response curve is drawn as seen in Figure 4.6. The correlation coefficient for dose response curve is 0.9957.

4.3.4 $CaSO_4$:Zn

The glow curves of this sample are shown in Figure 4.7. They are for 15 Rad, 22.5 Rad, 30 Rad. The maximum peak temperature is 120 °C. The dose response curve which is a straight line is shown in Figure 4.8. The correlation coefficient of the linear is 0.9998.

4.3.5 $CaSO_4$:Se

The glow curves of the dosimeters are shown in Figure 4.9. The curves 1, 2, 3 are drawn for 15 Rad, 30 Rad and 52,5 Rad respectively. The dose response curve of this sample is shown in Figure 4.10. The maximum peak occur at 132 °C. The correlation coefficient is 0.99945.

4.3.6 $CaSO_4$:Mn,Mg

The glow curves are plotted in Figure 4.11. There are three types of glow curves. The first glow curve is plotted for 45 Rad, the second one for 75 Rad, and the third one for 120 Rad. The dose response curve of these curves are drawn in Figure 4.12, which is drawn dose versus relative light intensity. The maximum peak is at 140 °C. The correlation coefficient of dose response curve is 0.9984.

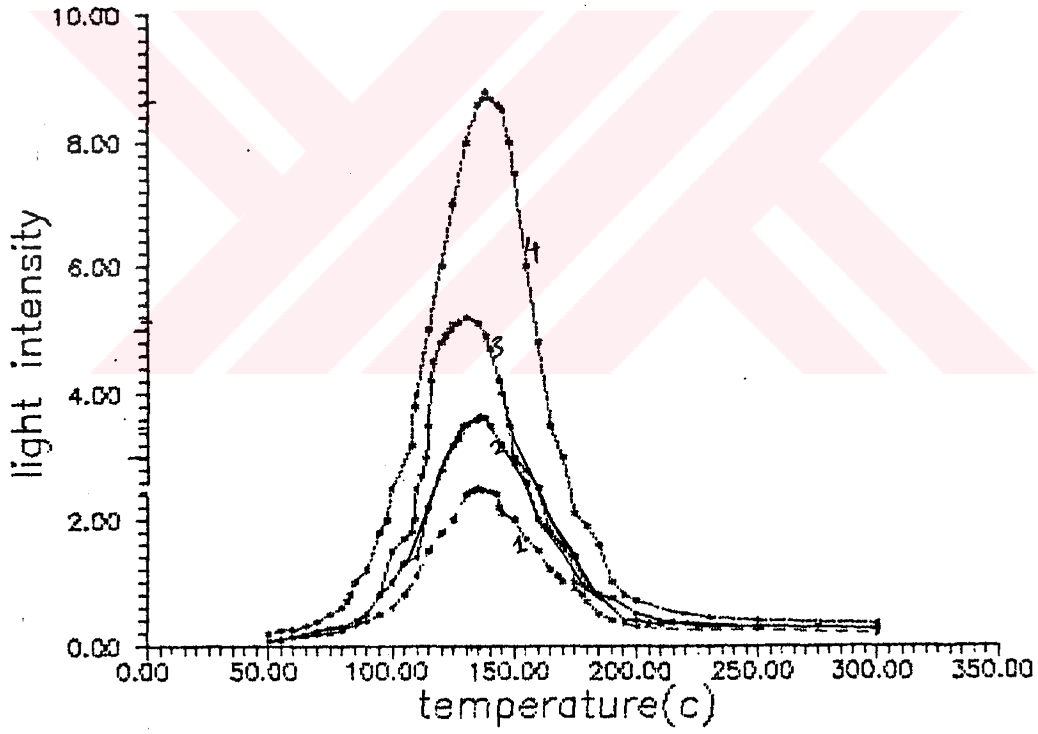

Figure 4.1. Glow curves $CaSO_4$:Mn - 1 teflon dosimeter exposed to Sr-90 β .

1) 7,5 Rad
2) 15 Rad
3) 22.5 Rad
4) 33 Rad.

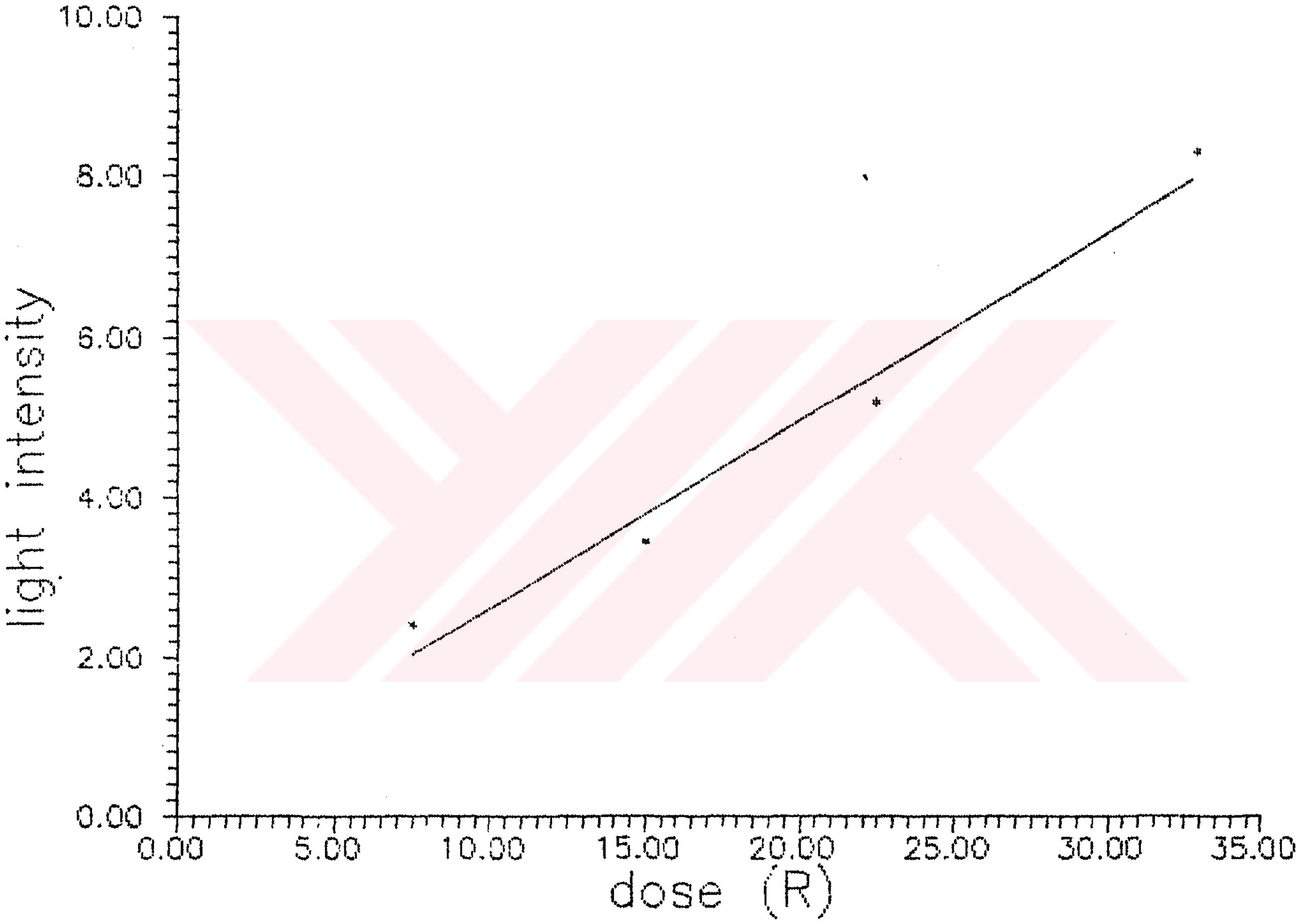

Figure 4.2 Dose response curve of $CaSO_4$:Mn - 1

$y = 0.23x + 0.28$

$r = 0.9878$

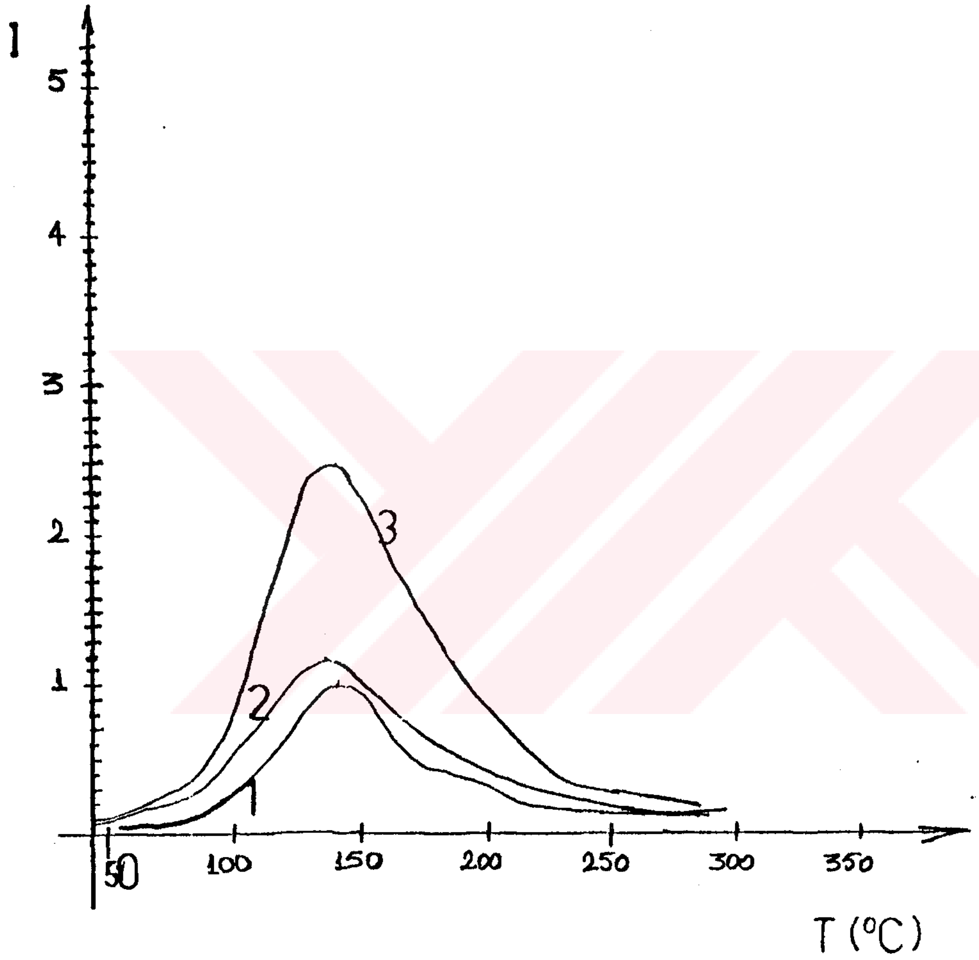

Figure 4.3 The glow curves of the $CaSO_4$:Mn - 2 teflon dosimeter.They are exposed to Sr-90 β radiation.

1) 30 Rad

2) 75 Rad

3) 150 Rad.

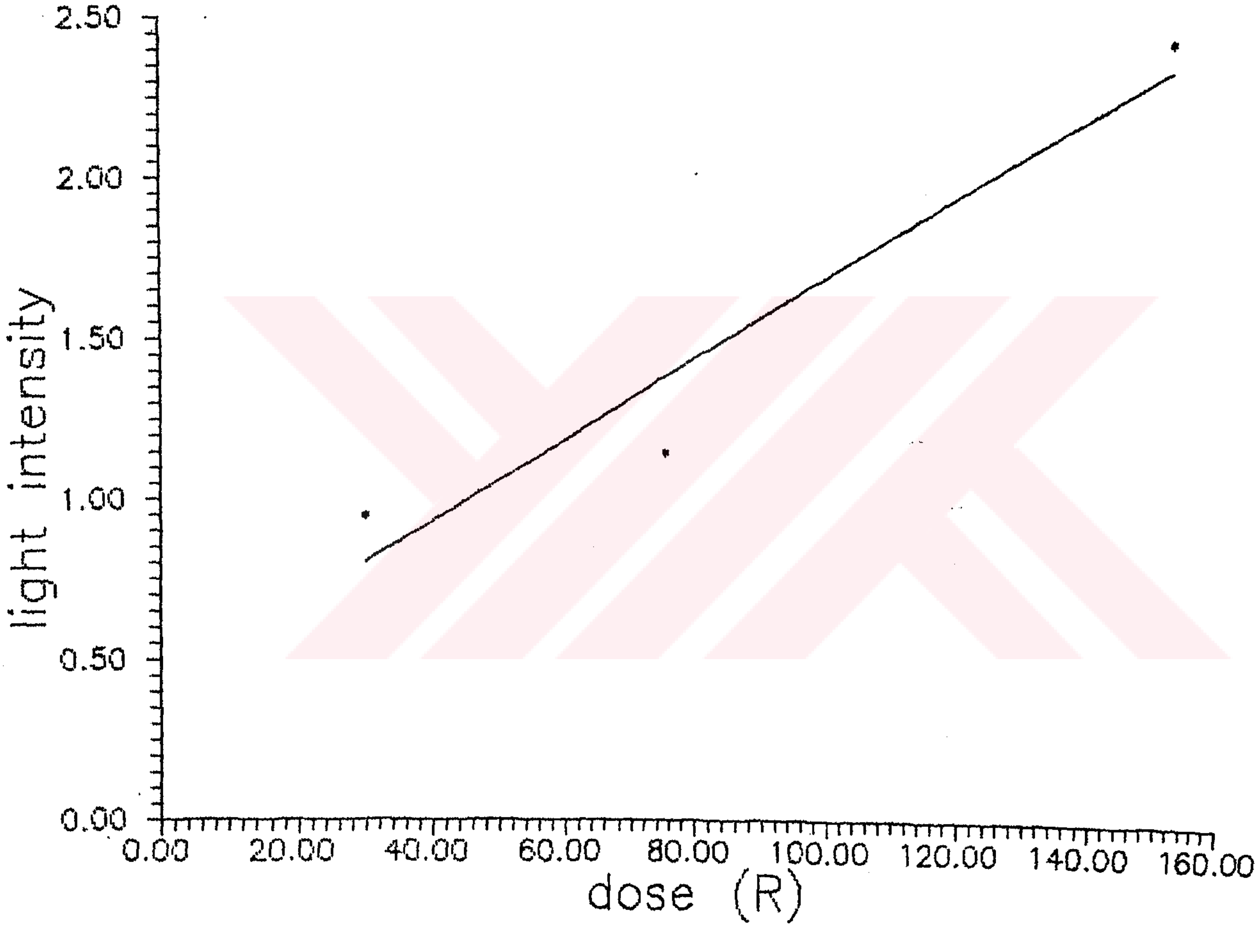

Figure 4.4. The dose response curve of $CaSO_4$:Mn - 2.

$y = 0.0129x + 0.4122$

$r = 0.9671$

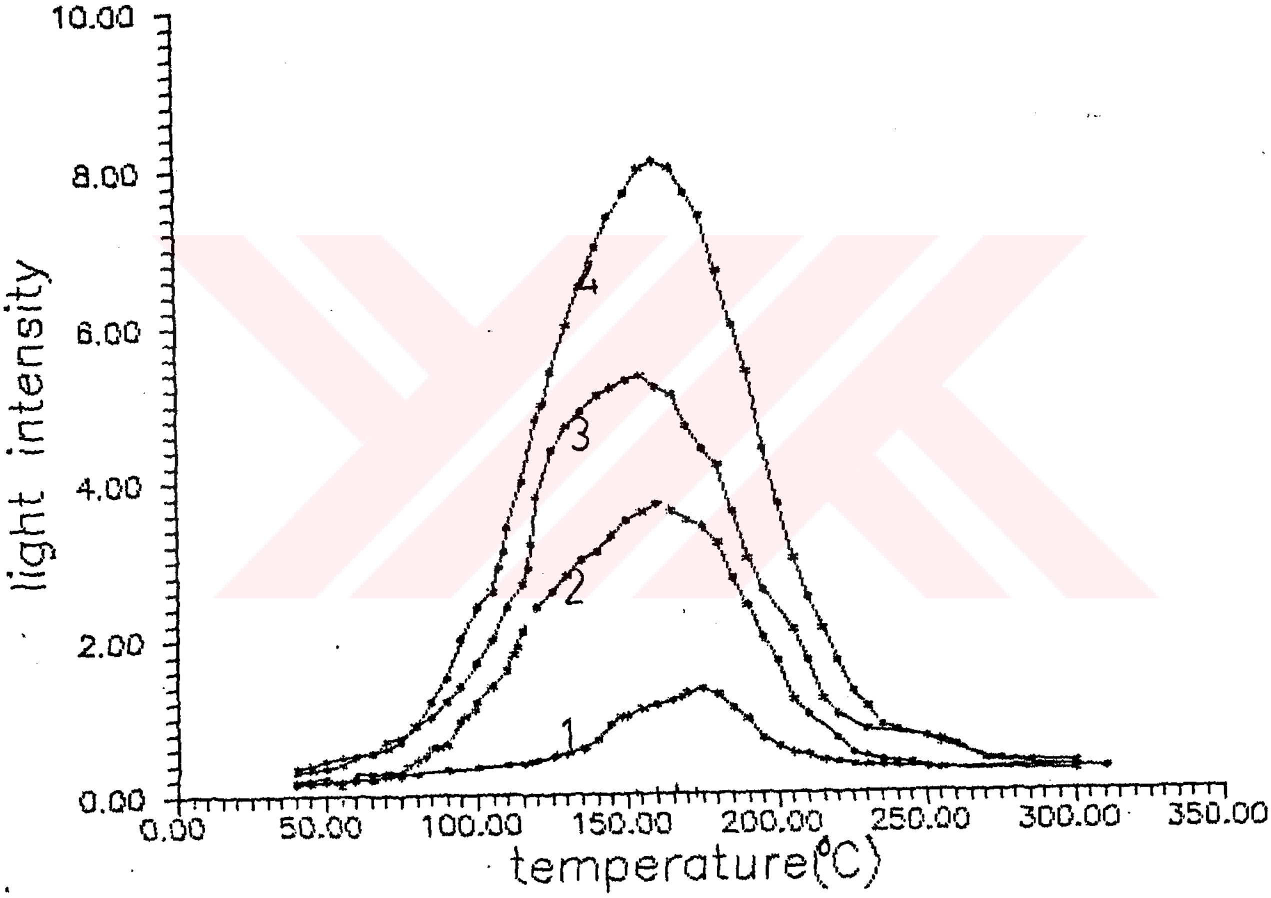

Figure 4.5 The glow curves of $CaSO_4$:Mg teflon dosimeter for,

1) 75 Rad
2) 150 Rad
3) 225 Rad
4) 300 Rad.

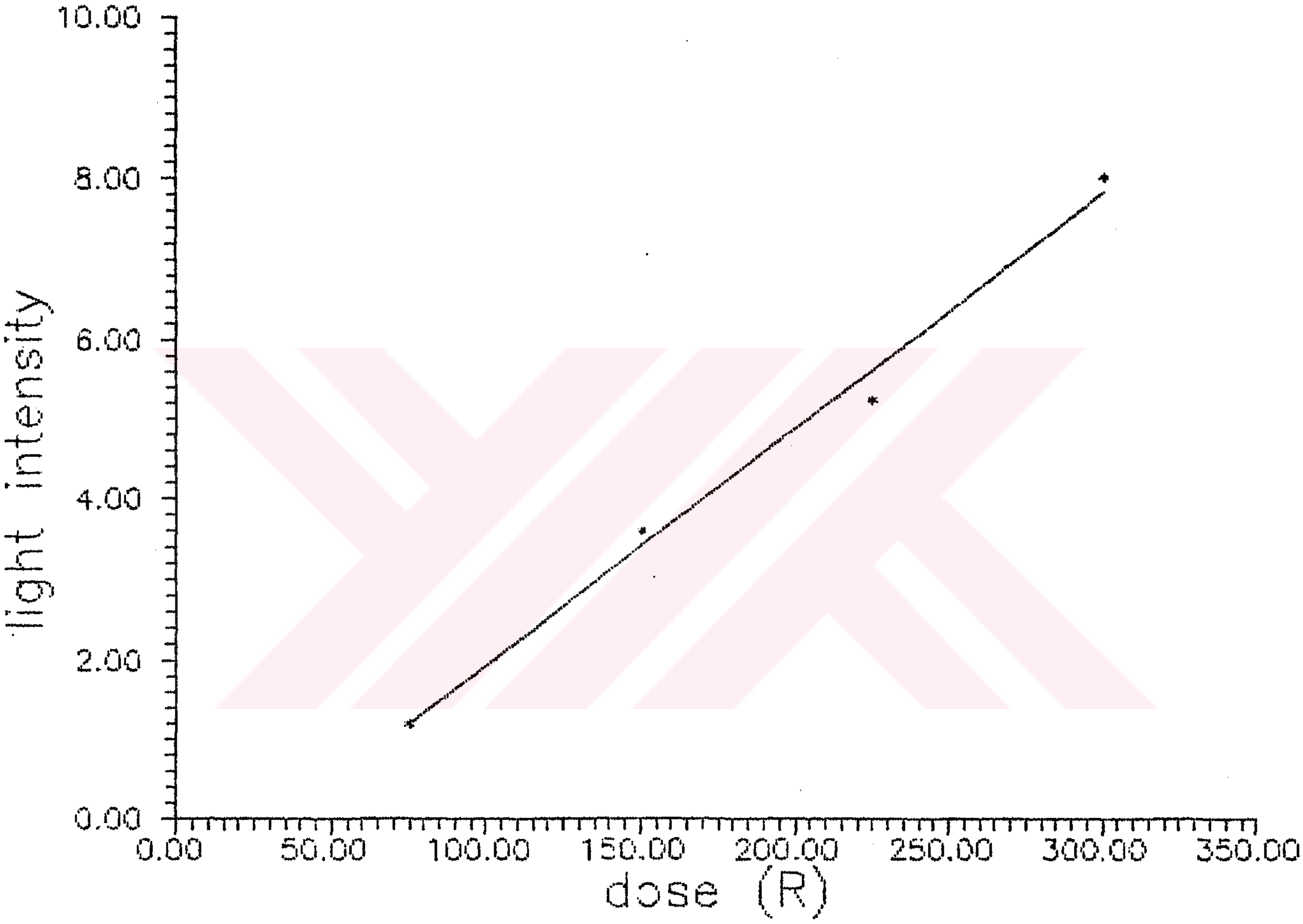

Figure 4.6 Dose response curve of $CaSO_4$:Mg

$y = 0.029x - 1.01$

$r = 0.9957$

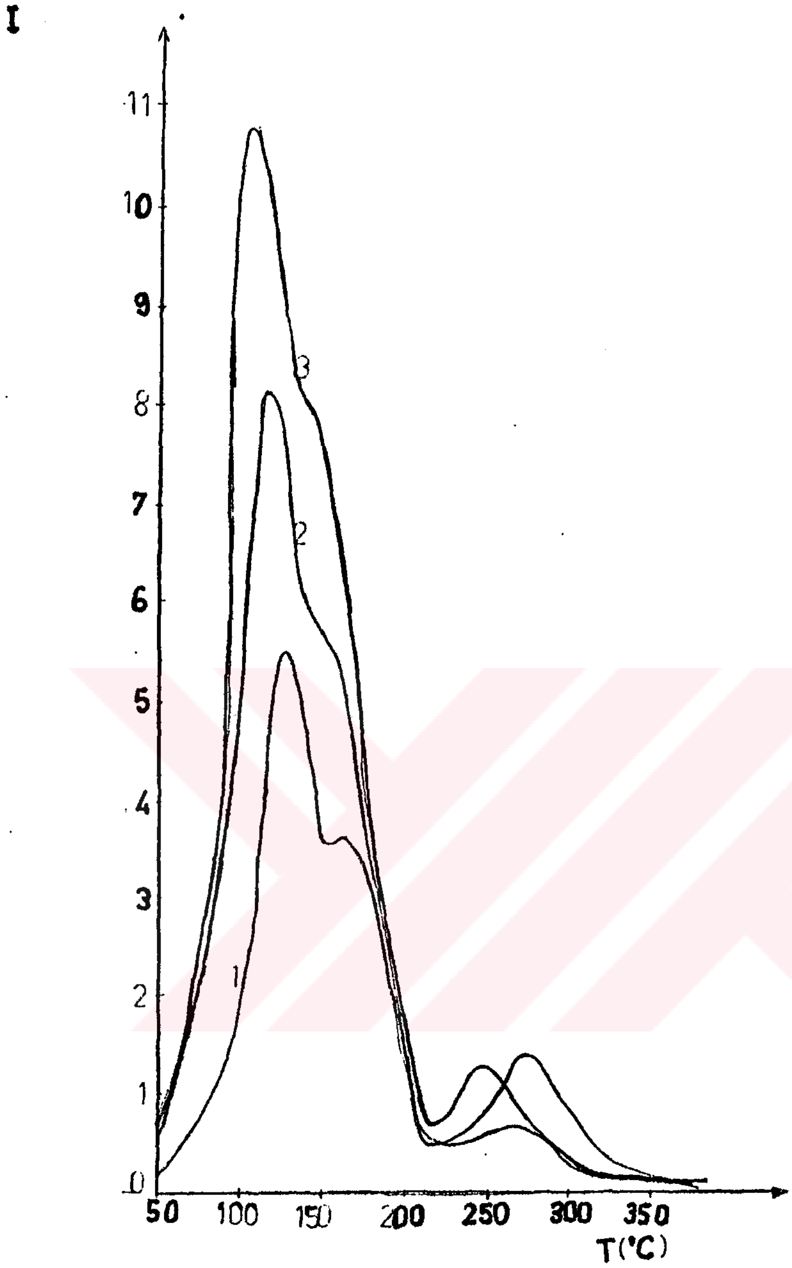

Figure 4.7 The glow curves of $CaSO_4$:Zn teflon dosimeter

1) 15 Rad

2) 22.5 Rad

3) 30 Rad

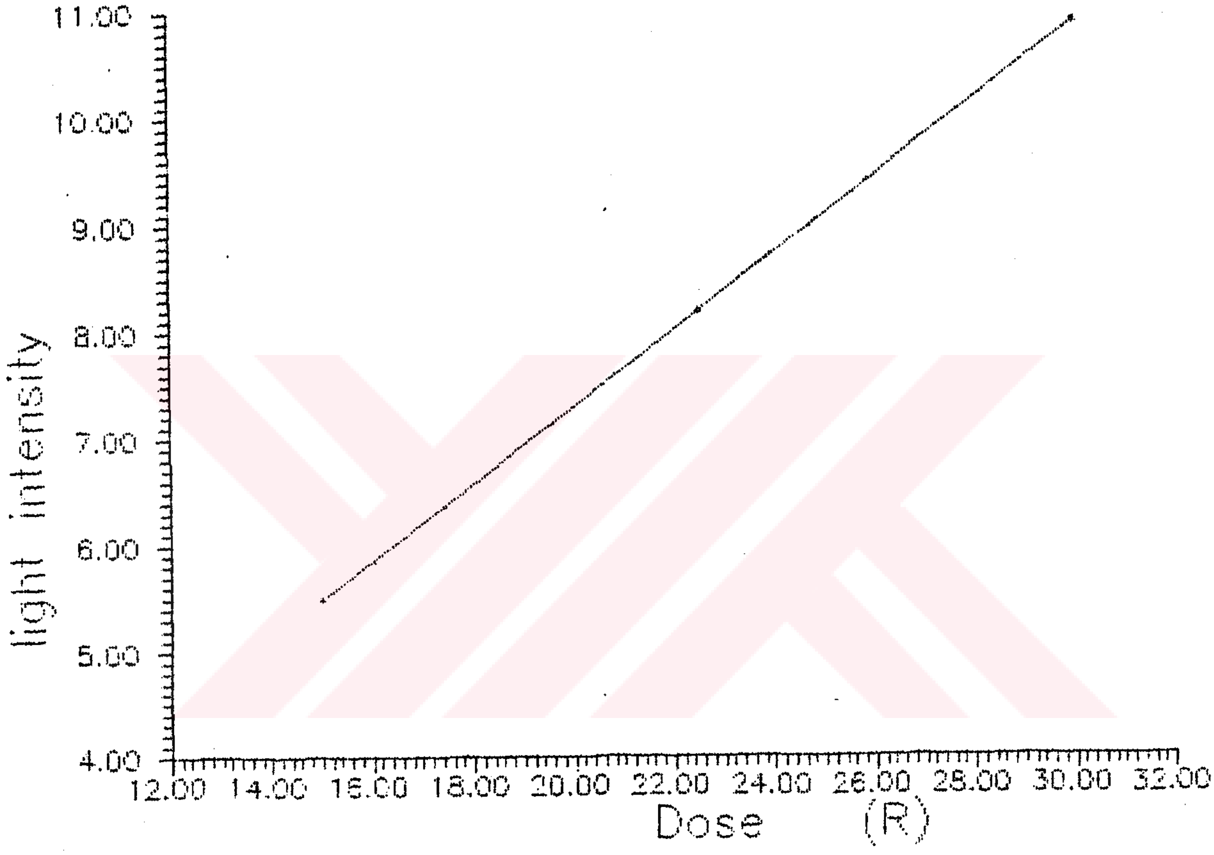

Figure 4.8 The dose response curve of $CaSO_4$:Zn

$y = 0.33x + 0.23$

$r = 0.999$

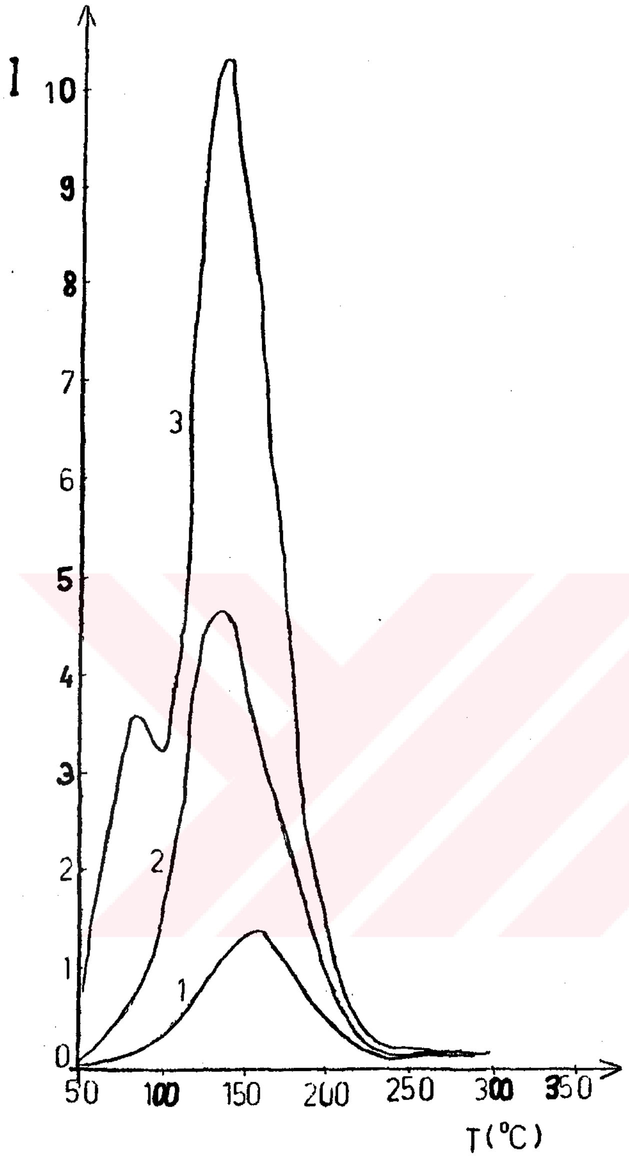

Figure 4.9 The glow curves of $CaSO_4$:Se

1) 15 Rad

2) 30 Rad

3) 52.5 Rad

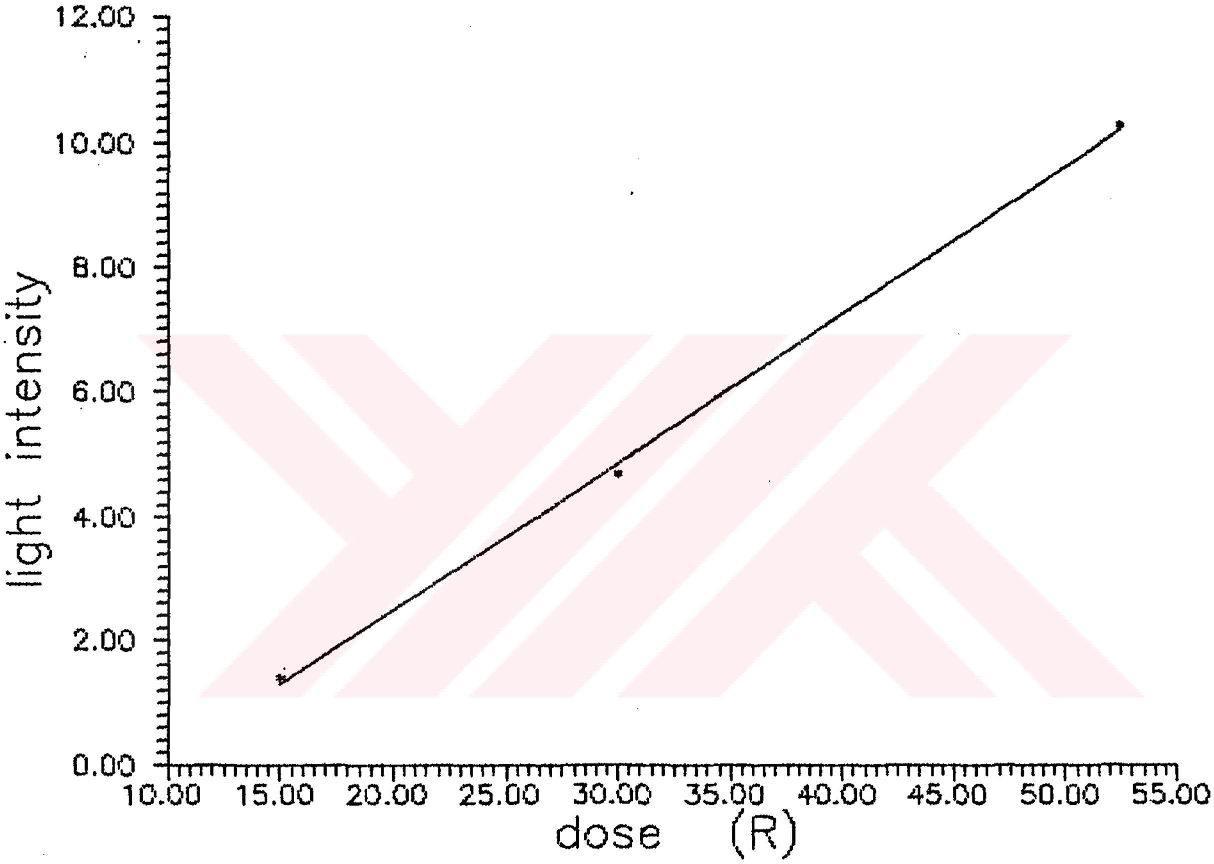

Figure 4.10 The dose response curve of $CaSO_4$:Se

$y = 0.2382x - 2.2763$

$r = 0.99945$

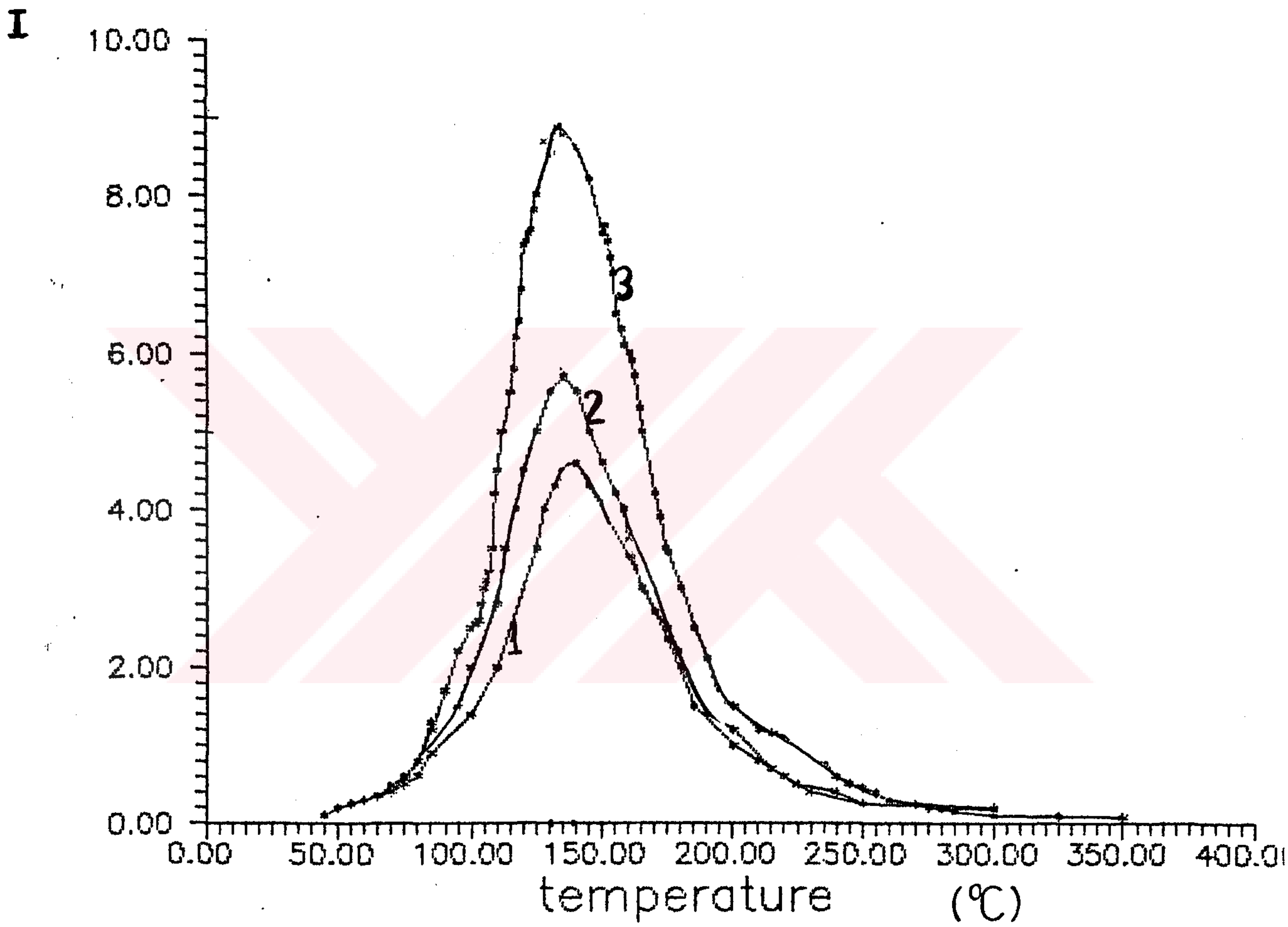

Figure 4.11 The glow curves of $CaSO_4$:Mn,Mg teflon dosimeter

1) 45 Rad

2) 75 Rad

3) 120 Rad.

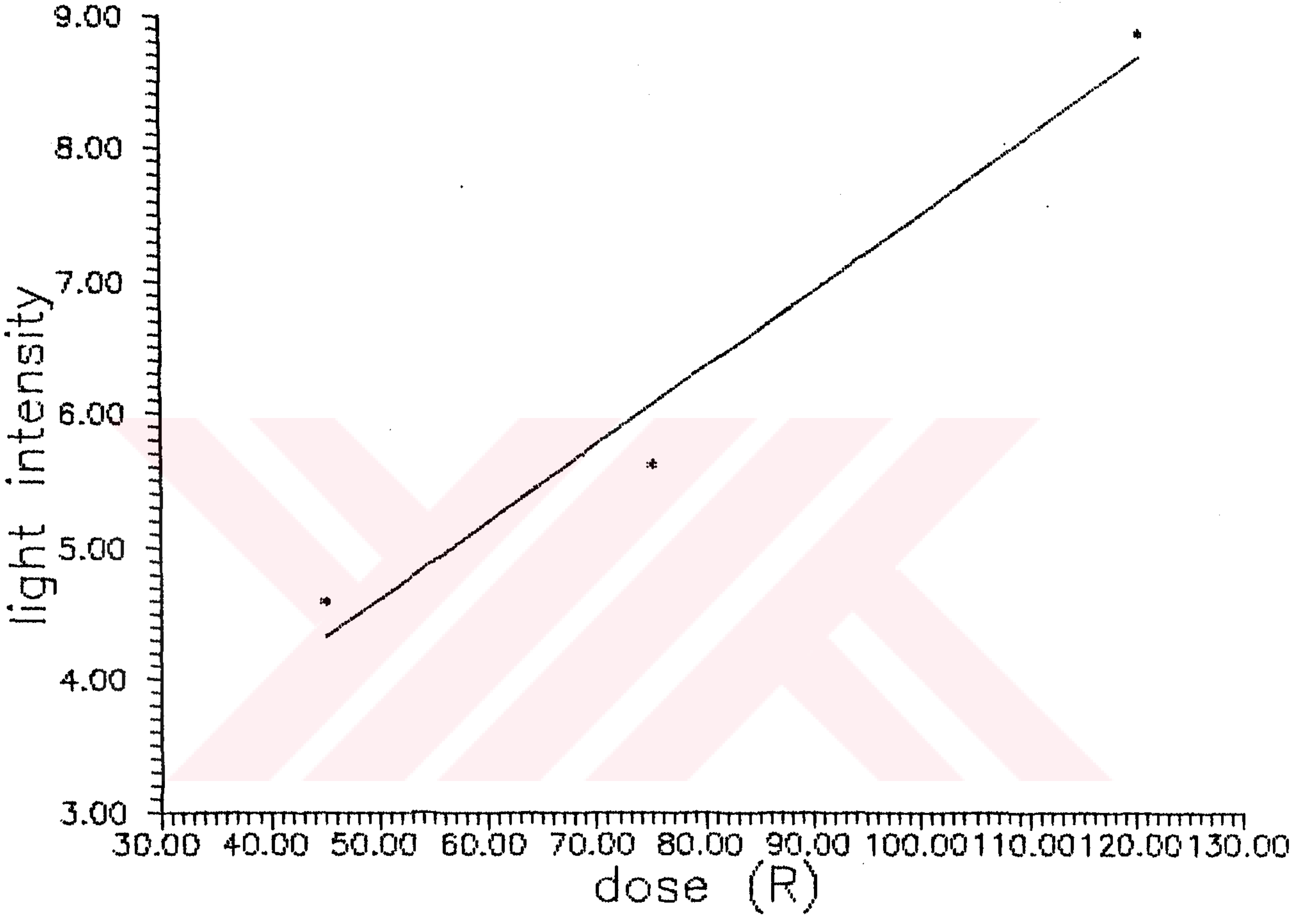

Figure 4.12 The dose response curve of $CaSO_4$:Mn,Mg
$y = 0.058x + 1.72$
$r = 0.9984$

4.4 CALCULATIONS OF KINETICS CONSTANTS

For the calculation of trap energy and frequency factor the single-peak-integration method are used.

From the glow curves, the value of 'a' , which is explained at chapter 3, must be calculated. For the calculation of 'a' , the area under the glow curve is divided into some part same as Figure 3.1. Therefore the value of the temperature T corresponds to 'a' can be selected. The calculation 'a' value are made by TI-58 calculater. 'a' can be expressed by A_1/A, where A_1 is the area which corresponds to the temperature T_1. And A is the total area between T_o and T. For each dosimeter, the 'a' values correspond to T, and calculated 'p' values are tabled.

According to this method, the graph 1/T versus lnp gives a straight line. From the slope of the tangent, the trap energy can be calculated, because of the slope of this line is equal to E/k, where E is trap energy and k is the Boltzmann constant. The frequency factor is calculated from the y-intercept of this line so that the value of y-intercept is equal to lnS.

The information about calculated trap energy and frequency factor for each dosimeter are given below.

4.4.1 $CaSO_4$:Mn - 1

The graph of 'a' versus T is shown in Figure 4.13. Table 4.2 shows the calculated 'p' values with respect to the 'a'. Figure 4.14 is related with lnp versus 1/T graph for this sample. The fourth curve is used.

correlation coefficient, r, (Figure 4.14): 0.9987
y - intercept : 11.12
slope : - 6097.3

E (trap energy) : 0.52 eV

S (Freqency factor) : 6.76×10^4 1/sec

4.4.2 $CaSO_4$:Mn - 2

The a values are calculated from the glow curve so that, the third curve is used. The 'a' value and corresponding T volues are shown in Table 4.3. From the graph of a versus T, the a values are determined according to desirable values of T, Figure 4.15. Table 4.4 contains the experimental results of this sample. According to plot of lnp versus 1/T, Figure 4.16, E and S are calculated.

correlation coefficient r: 0.993

y - intercept : 3.90

Slope : -3376

E : 0.291 eV

S : 51 1/sec.

4.4.3 $CaSO_4$:Mg

For the calculation of 'a' and corresponding 'p' values, the fourth curve is used. The Table 4.5 shows 'a' values with respect to T . The calculated 'p' values and corresponding T values are listed in Table 4.6. The graph of 'a' versus T is shown in Figure 4.17. The graph of lnp versus 1/T is as seen in Figure 4.18.

correlation coefficient r: 0.9879

y - intercept : - 3.10

slope : -3229

E : 0.278 eV

S : 22.4 1/sec.

TABLE 4.1 'a' and T values of $CaSO_4$:Mn-1 teflon dosimeter for 4^{th} curve

T_o = 343.15 °K
T_m = 413.15 °K

T (°K)	a	Area	I h (cm)
358.15	0.01493	6.15	0.66
368.15	0.04465	18.38	1.15
378.15	0.10561	43.48	2.43
388.15	0.20826	85.75	4.24
398.15	0.35770	147.28	6.27
408.15	0.53171	218.93	7.24
418.15	0.70240	289.21	7.12
428.15	0.83213	342.63	5.40
438.15	0.90544	372.81	2.91
448.15	0.95353	392.61	1.97
458.15	0.98299	404.75	1.22
473.15	0.99732	410.65	0.63
483.15	1.00000	411.75	0.12

TABLE 4.2 T and p values of $CaSO_4$:Mn-1 teflon dosimeter for 4^{th} curve

T_o = 343.15 °K
T_m = 413.15 °K

a	T	1/T	p	lnp
0.1	377.15	0.00265	0.006197	- 5.083
0.2	387.15	1.00258	0.001014	- 4.591
0.3	394.15	0.00253	0.01398	- 4.269
0.4	400.15	0.00249	0.01792	- 4.021
0.5	407.15	0.00245	0.02166	- 3.832
0.6	412.15	0.00242	0.02636	- 3.635
0.7	418.15	0.00239	0.03202	- 3.441

4.4.4 $CaSO_4$:Zn

The third curve is used for calculation. Table 4.7 contains the 'a' values with respect to T and Table 4.8 contains the 'p' values with respect to 'a'. The graphs of 'a' versus T and lnp versus 1/T are plotted in Figure 4.19 and 4.20 respectively.

correlation coefficient r: 0.9909
y - intercept : 11.84
Slope : -6668
E : 0.575 eV
S : 1.39×10^5 1/sec.

4.4.5 $CaSO_4$:Se

For this dosimeter, the 'a' values corresponding to T, 'p' values corresponding to T, a-t graph, and lnp-1/T graph are illustrated in Table 4.9, Table 4.10, Figure 4.21, and Figure 4.22, respectively. The second glow curve peak is used.

Correlation coefficient: 0.978
y - intercept : 4.9
Slope : -3767
E : 0.325 eV
S : 138 1/sec.

4.4.6 $CaSO_4$:Mn,Mg

The third curve is used.The Table 4.10 contains 'a' and T values Table 4.11 is for lnp and T values. Figure 4.23 is plotted a versus and Figure 4.24 shows lnp versus 1/T graph.

Correlation coefficient: 0.979
y - intercept : 3.105
slope : -3027.6
E : 0.261 eV
S : 22.3 1/sec.

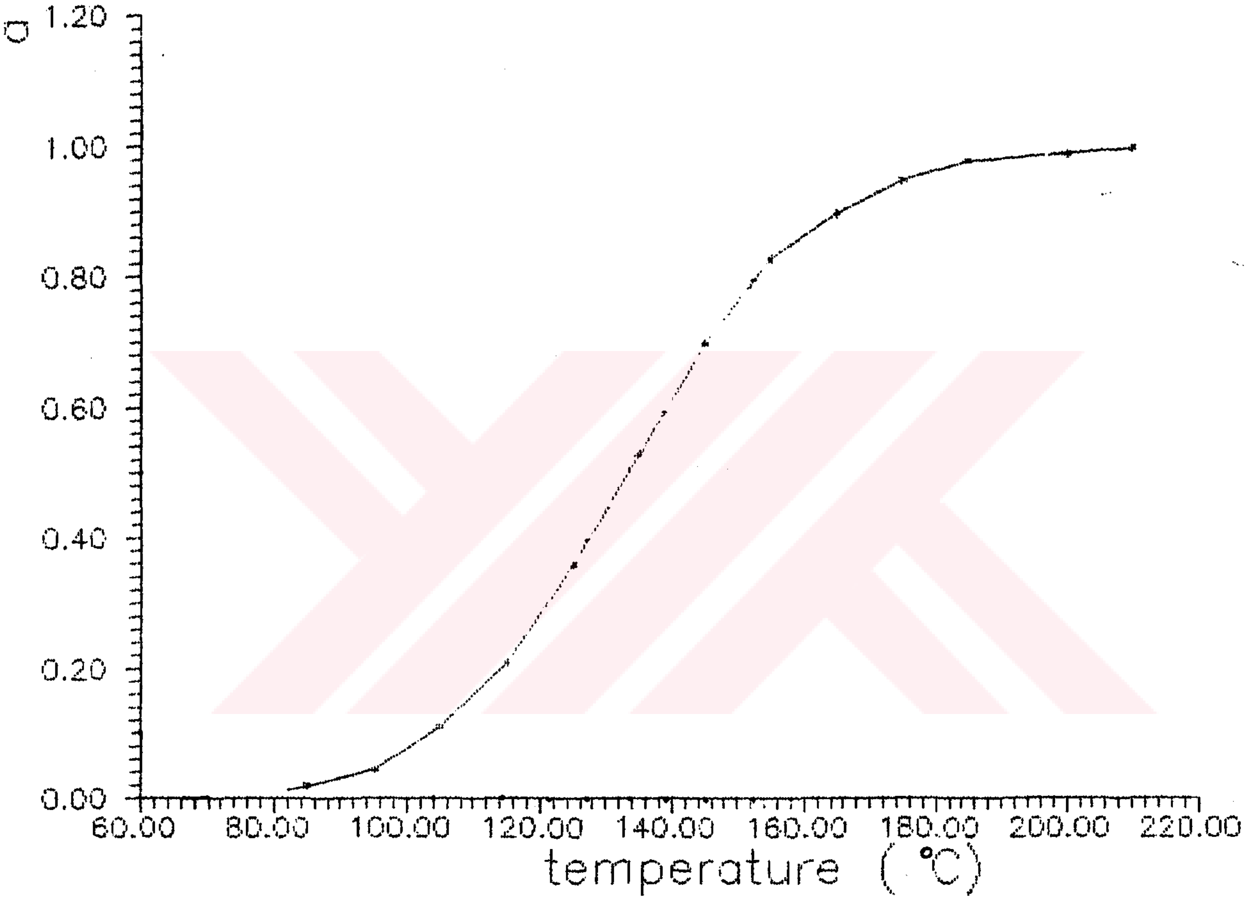

Figure 4.13 (a-t) graph of $CaSO_4$:Mn - 1

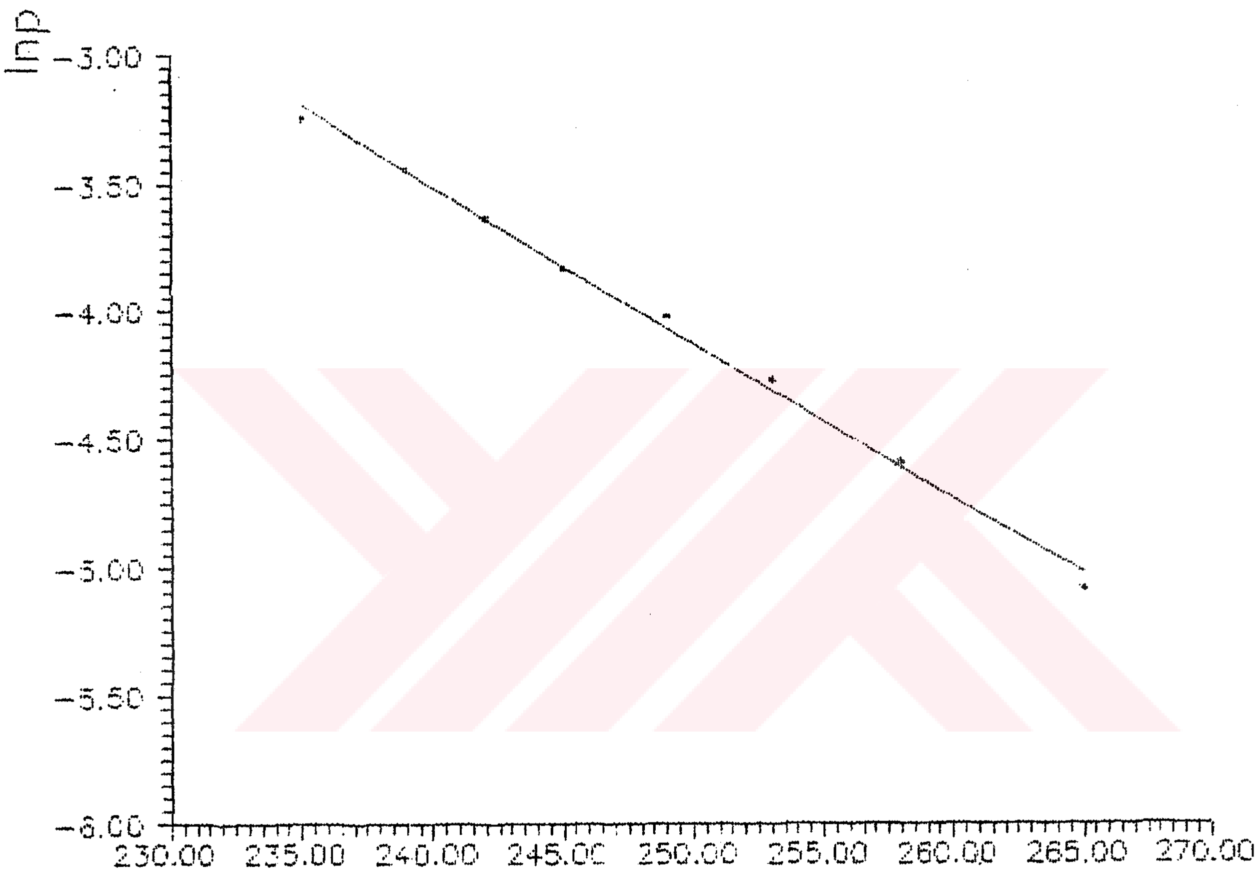

Figure 4.14 (lnp - 1/T) graph of $CaSO_4$:Mn - 1

b : 2 °C/sec.

slope : -6097.3

TABLE 4.3 'a' and T values of $CaSO_4$:Mn - 2 teflon dosimeter for 3rd curve

T_o = 335.15 °K
T_m = 410.15 °K

T (°K)	a	Area	I.h(cm)
360.65	0.02892	11.916	0.76
373.15	0.08260	34.041	1.78
385.65	0.17344	71.479	3.02
398.15	0.29592	121.958	4.03
410.65	0.44343	182.75	4.93
423.15	0.58249	240.02	4.61
435.65	0.70149	289.104	4.02
448.15	0.79026	325.687	2.92
458.65	0.86073	354.729	2.34
473.15	0.91416	376.75	1.74
485.65	0.94651	390.083	0.97
498.00	0.97937	403.625	0.87
546.65	1.00000	412.125	0.75

TABLE 4.4 T and p values of $CaSO_4$:Mn - 2 teflon dosimeter for 3rd curve

T_o = 335.15 °K
T_m = 410.15 °K

a	T (°K)	1/T	p	lnp
0.1	376.27	0.00265	0.00512	-5.273
0.2	390.65	0.00256	0.00804	-4.823
0.3	399.65	0.00250	0.01100	-4.504
0.4	408.40	0.00245	0.01390	-4.272
0.5	417.15	0.00239	0.01690	-4.080
0.6	427.15	0.00234	0.01990	-3.916
0.7	437.15	0.00228	0.02340	-3.752

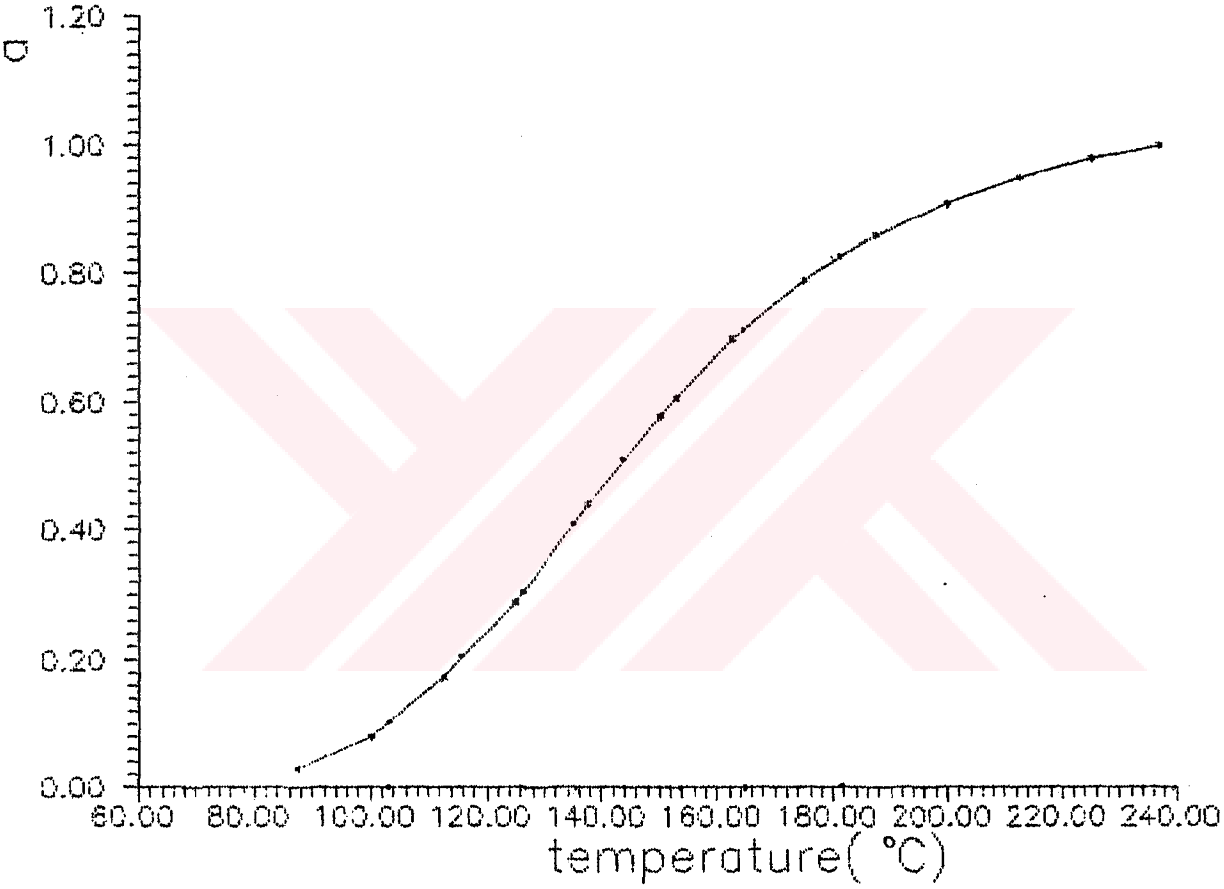

Figure 4.15 (a-T) graph of $CaSO_4$:Mn - 2

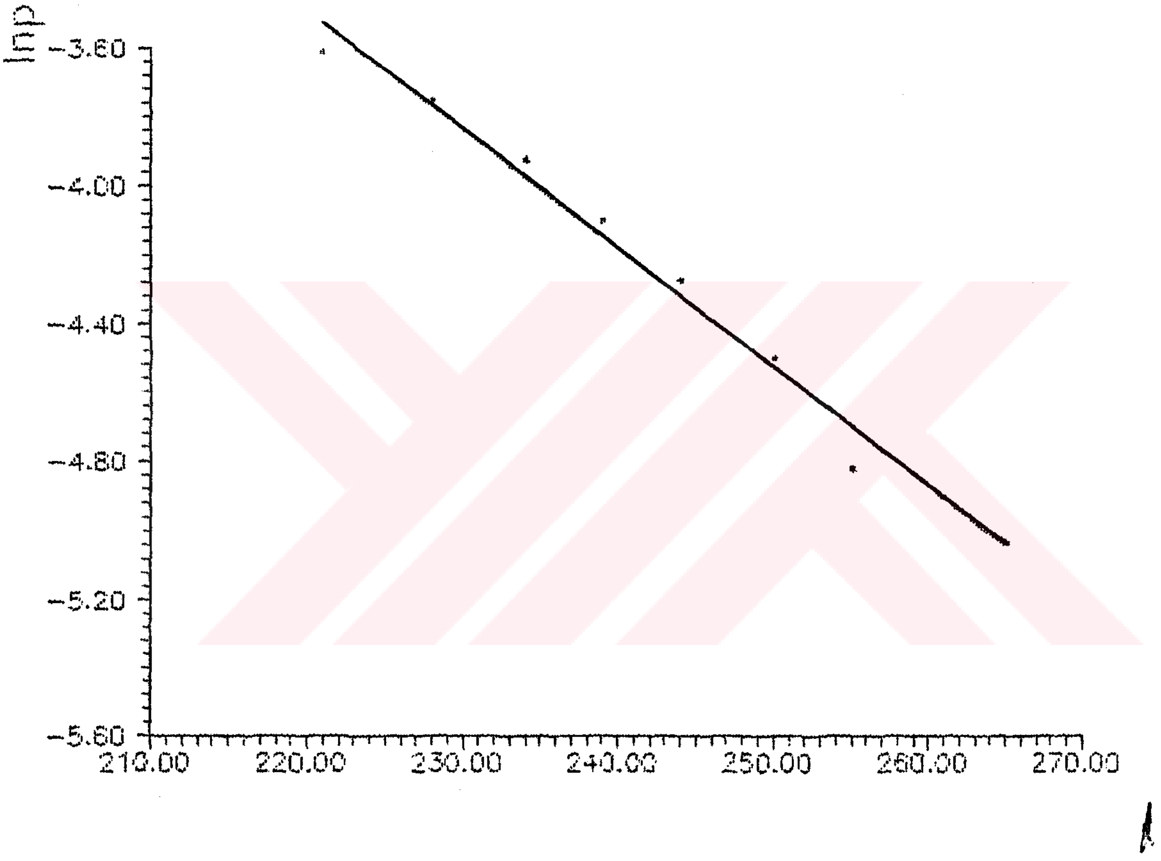

Figure 4.16 (lnp- 1/T) graph of caSO$_4$:Mn - 2

b : 2 ^{o}C/sec.

slope : - 3838

TABLE 4.5 'a' and T values of $CaSO_4$:Mg teflon dosimeter for 4th curve

T_o = 333.15 °K
T_m = 433.15 °K

T (°K)	a	Area	I.h (cm)
363.15	0.04154	46.36	2.35
383.15	0.11615	129.63	3.98
403.15	0.25056	279.66	7.20
423.15	0.45768	510.83	11.78
443.15	0.76488	753.20	12.35
463.15	0.85464	953.90	10.17
483.15	0.95848	1069.80	5.68
503.15	1.00000	1116.13	2.52

TABLE 4.6 T and p values of $CaSO_4$:Mg teflon dosimeter for 4th curve

T_o = 333.15 °K
T_m = 433.15 °K

a	T	1/T	p	lnp
0.1	385.25	0.00262	0.00438	-5.430
0.2	397.55	0.00251	0.00692	-4.970
0.3	409.45	0.00244	0.00934	-4.763
0.4	428.65	0.00233	0.01069	-4.538
0.5	437.35	0.00228	0.01330	-4.319
0.6	447.03	0.00223	0.01605	-4.132
0.7	455.35	0.00229	0.01970	-3.927
0.8	467.40	0.00213	0.02397	-3.730

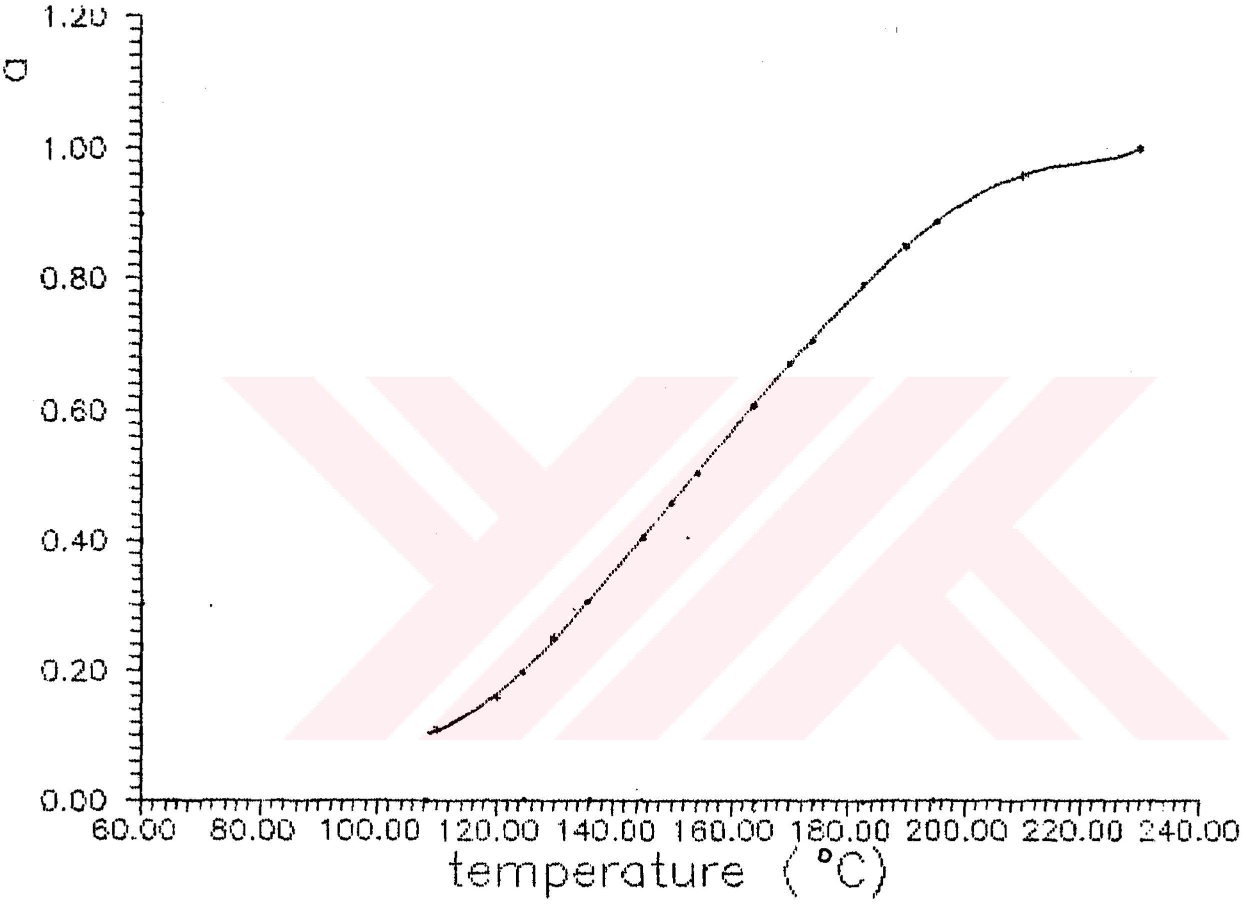

Figure 4.17 (a-T) gragh of $CaSO_4$:Mg

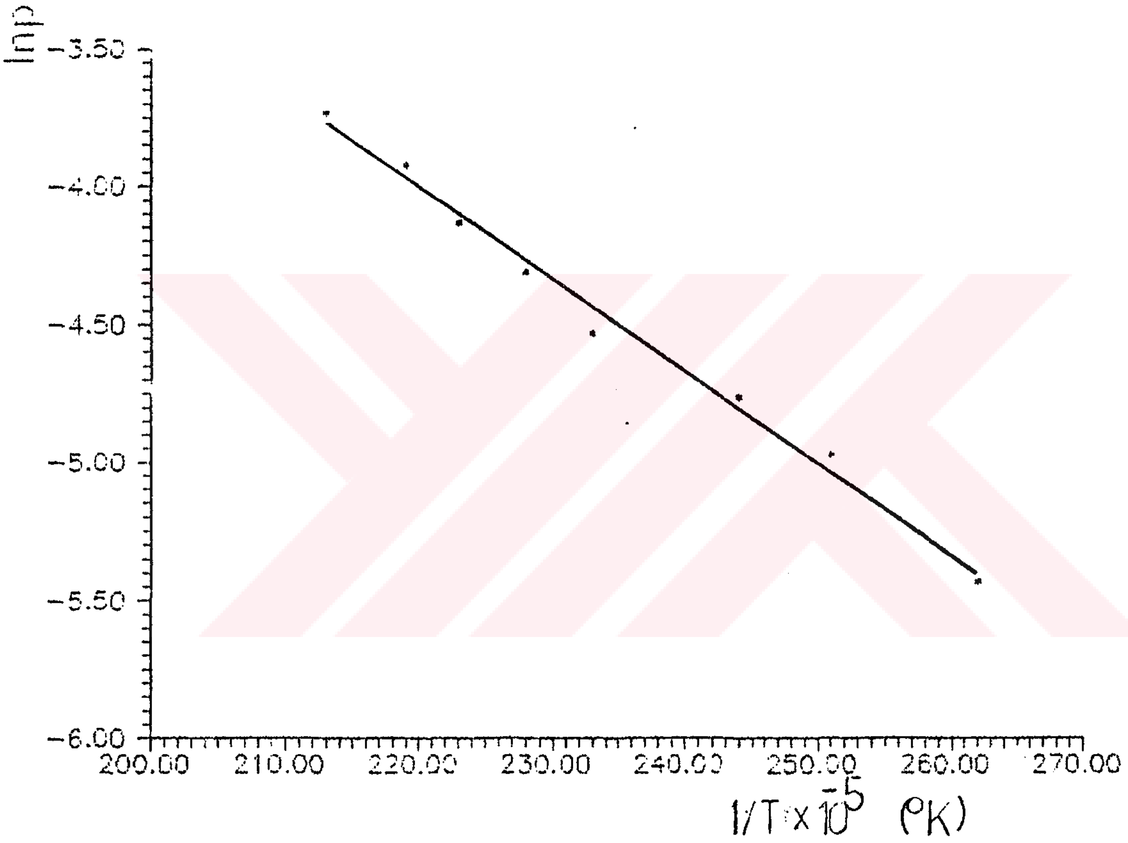

Figure 4.18 (lnp - 1/T) graph of $CaSO_4$: Mg

b : 2 °C

slope : - 3229

TABLE 4.7 'a' and T values of CaSO$_4$:Zn teflon dosimeter for 3rdcurve

T_o = 323.15 ^{o}K
T_m = 393.15 ^{o}K

T (K)	a	A	I.h
329.40	0.00513	5.583	0.46
341.75	0.00121	13.200	0.62
354.40	0.02268	24.64	0.84
366.90	0.04490	48.87	1.84
379.40	0.09247	100.45	4.03
391.90	0.18800	204.25	8.25
404.40	0.34769	377.70	13.32
416.90	0.56056	608.95	18.50
429.40	0.73412	797.50	15.00
441.90	0.85110	924.62	10.18
454.40	0.92386	1003.60	6.38
466.90	0.96630	1049.72	3.54
479.40	0.98523	1070.29	1.34
491.90	0.99570	1081.41	0.90
504.40	1.00000	1086.33	0.44

TABLE 4.8 T and p values of CaSO$_4$:Zn teflon dosimeter for 3rd curve

T_o = 323.15 ^{o}K
T_m = 393.15 ^{o}K

a	T	1/T	p	lnp
0.1	0.00260	0.00348	-5.65	383.65
0.2	0.00254	0.00630	-5.05	393.25
0.3	0.00249	0.00910	-4.69	401.15
0.4	0.00245	0.01210	-4.41	407.25
0.5	0.00241	0.01510	-4.18	414.65
0.6	0.00238	0.01860	-3.97	421.15
0.7	0.00234	0.02310	-3-76	427.15

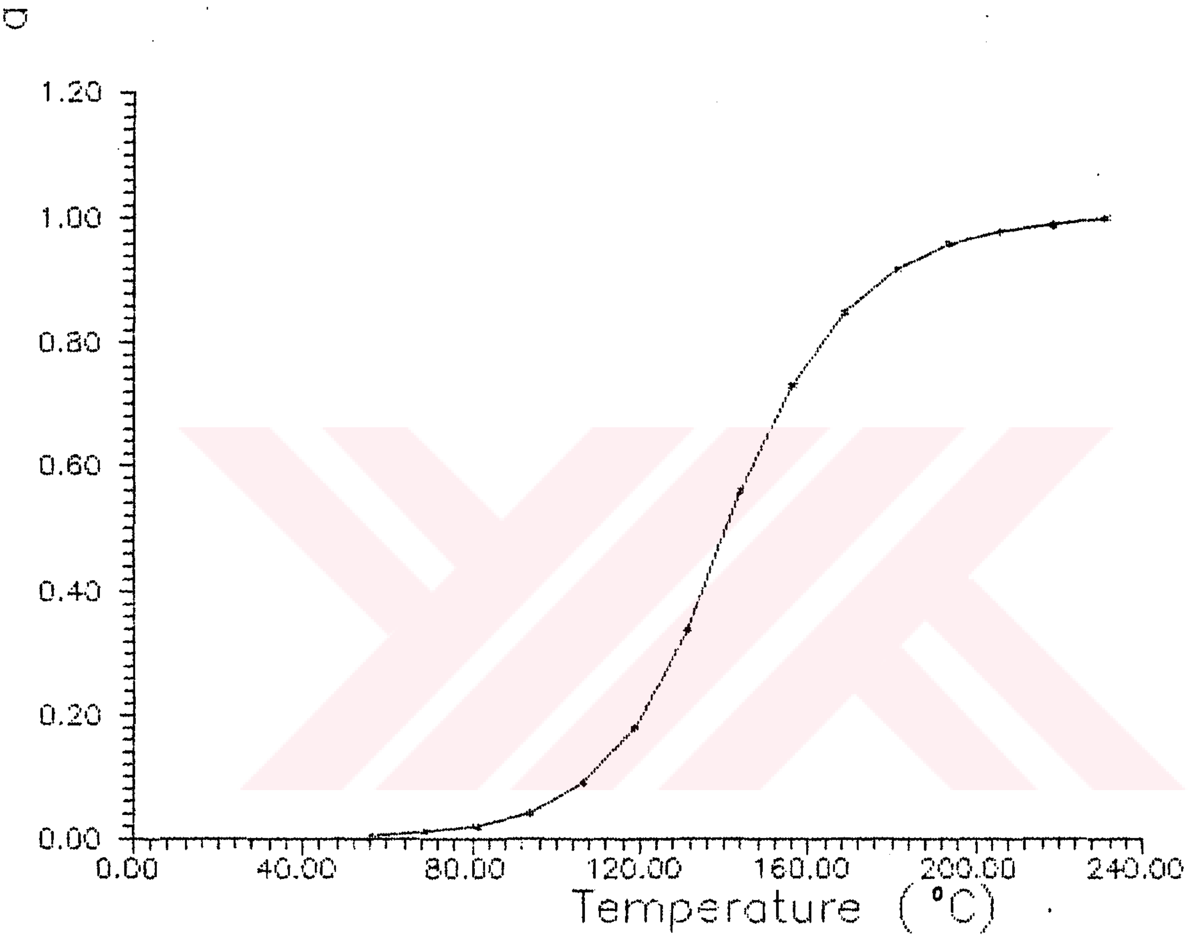

Figure 4.19 (a-T) graph of $CaSO_4$:Zn

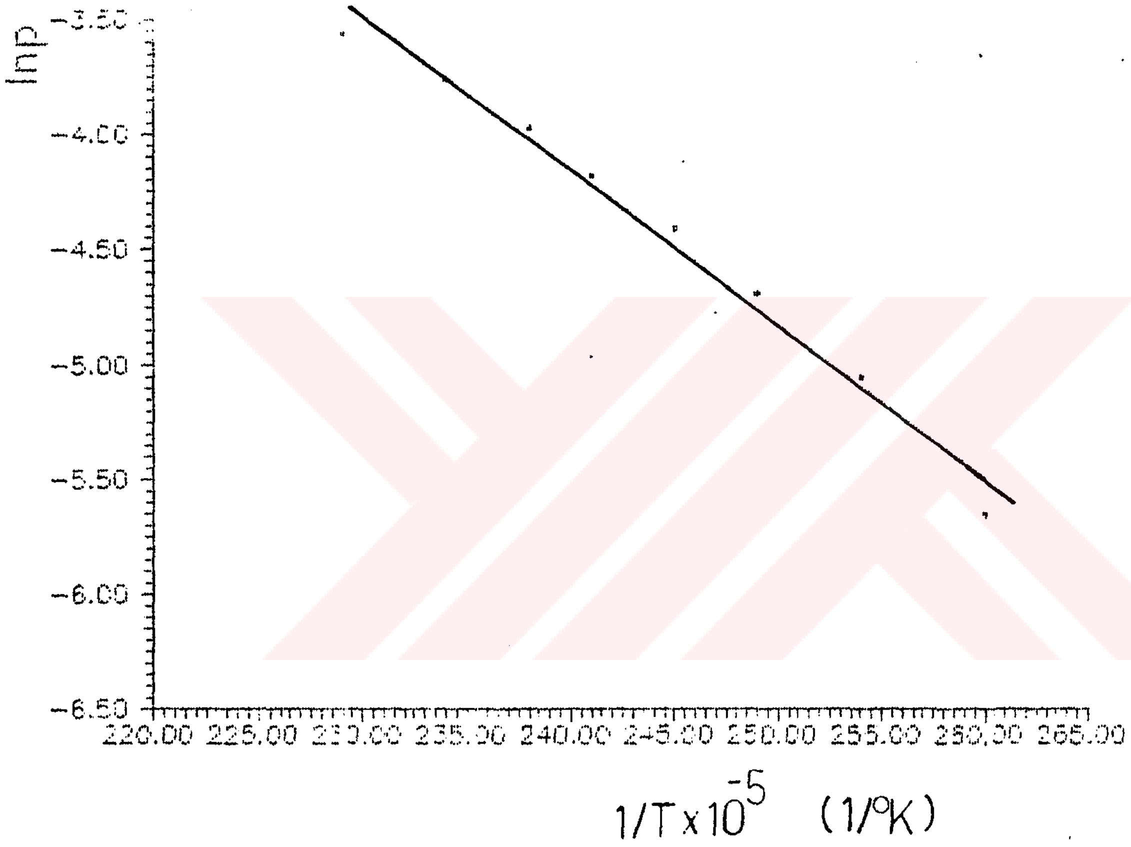

Figure 4.20 (lnp-1/T) graph of $CaSO_4$:Zn

b : 2°C

Slope : -6668

TABLE 4.9 'a' and T values of $CaSO_4$:Se teflon dosimeter for 2^{nd} curve

T_o = 323.15 °K
T_m = 405.15 °K

T (°K)	a	Area	I h (cm)
329.40	0.00593	4.66	0.36
341.90	0.01643	12.92	0.60
354.40	0.03801	29.87	1.32
366.90	0.08450	66.42	2.90
379.40	0.16880	132.68	5.24
391.90	0.29530	232.08	8.02
404.40	0.44109	346.66	9.23
416.90	0.57273	450.12	8.24
429.15	0.68440	537.91	7.00
441.90	0.77980	612.91	6.00
454.40	0.85340	670.70	4.61
466.90	0.90960	714.91	3.52
479.40	0.95334	749.25	2.80
491.90	0.98080	770.85	1.74
516.90	1.00000	785.90	0.42

TABLE 4.10 T and 'p' values of $CaSO_4$:Se teflon dosimeter for 2^{nd} curve

T_o = 323.15 °K
T_m = 405.15 °K

a	T	1/T	p	lnp
0.1	369.65	0.00270	0.00450	-5.396
0.2	384.35	0.00260	0.00729	-4.920
0.3	393.25	0.00254	0.01010	-4.580
0.4	398.35	0.00251	0.01350	-4.298
0.5	410.40	0.00243	0.01580	-4.142
0.6	422.65	0.00236	0.01840	-3.990
0.7	431.90	0.00231	0.02200	-3.810
0.8	445.30	0.00224	0.02630	-3.635

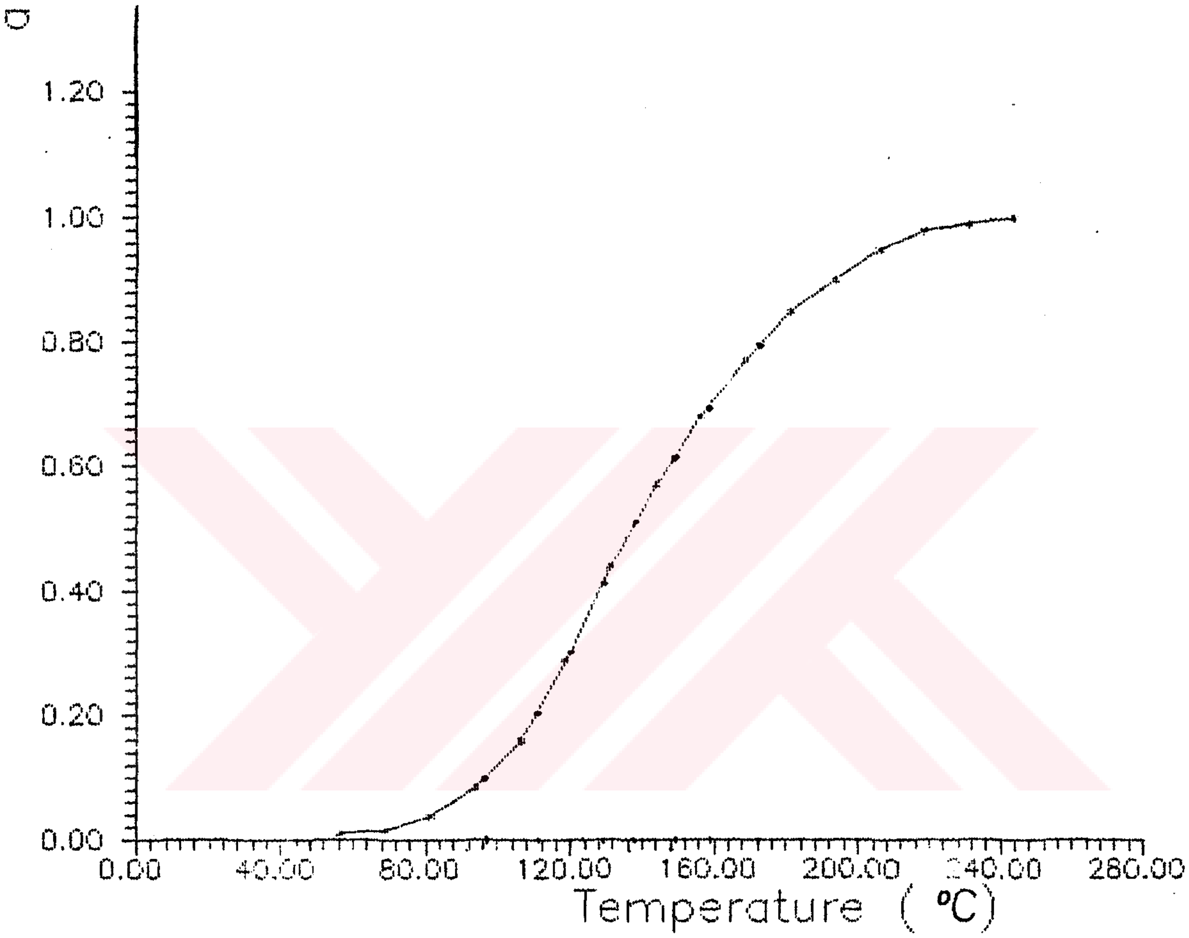

Figure 4.21 (a-T) graph of $CaSO_4$:Se

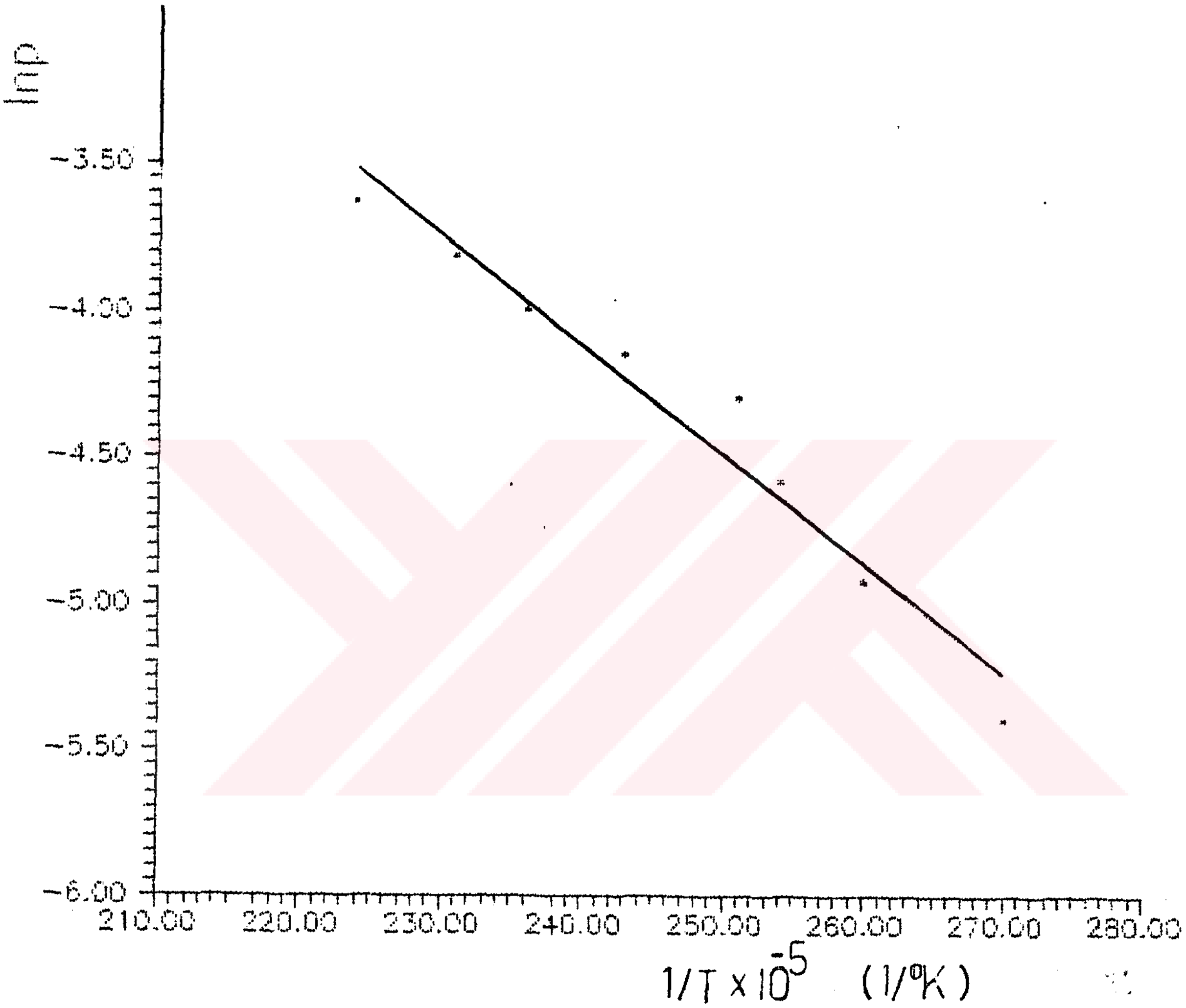

Figure 4.22 (lnp - 1/T) graph of $CaSO_4$:Se

b : 2^{o}C

slope : - 3767

TABLE 4.11 'a' values of $CaSO_4$:Mn,Mg teflon dosimeter for 3^{rd} curve

T_o = 323.15 ^oK
T_m = 408.15 ^oK

T (^oK)	a	Area	I h (cm)
358.15	0.03807	25.40	1.22
378.15	0.13801	92,06	3.15
398.15	0.36918	246.26	7.91
418.15	0.62892	419.53	8.74
438.15	0.81601	544.33	6.18
458.15	0.90886	606.26	2.82
478.15	0.95393	636.33	1.44
498.15	0.98576	657.56	1.08
518.15	1.00000	667.06	0.52

TABLE 4.12 T and p values of $CaSO_4$:Mn,Mg teflon dosimeter 3^{rd} curve

T_o = 323.15 ^oK
T_m = 408.15 ^oK

a	T	1/T	p	lnp
0.2	383.15	0.00261	0.00744	-4.901
0.3	392.15	0.00255	0.01033	-4.571
0.4	401.15	0.00249	0.01309	-4.355
0.5	412.15	0.00242	0.01506	-4.195
0.6	427.15	0.00234	0.01760	-4.039
0.7	436.15	0.00229	0.02130	-3.848

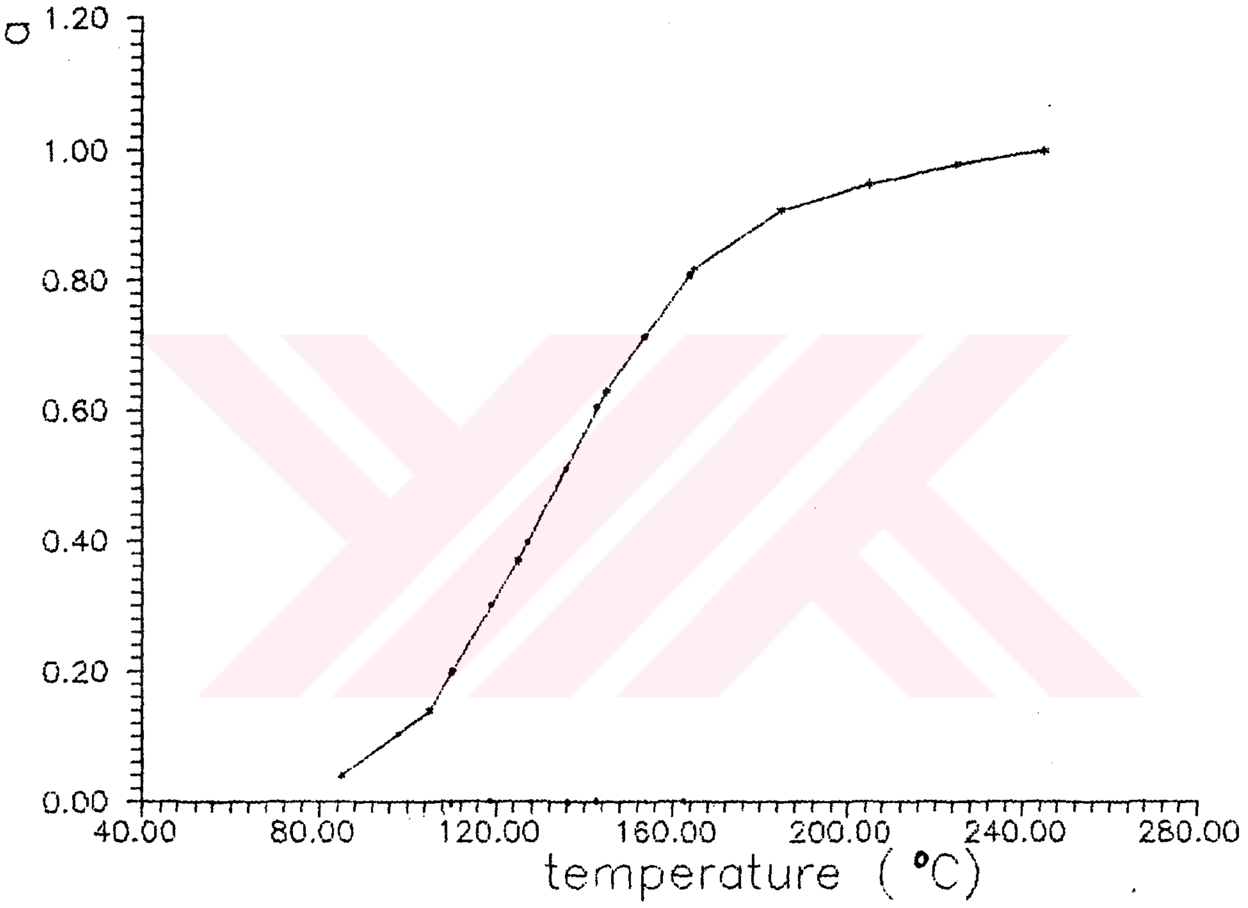

Figure 4.23 (a-T) graph of $CaSO_4$:Mn,Mg

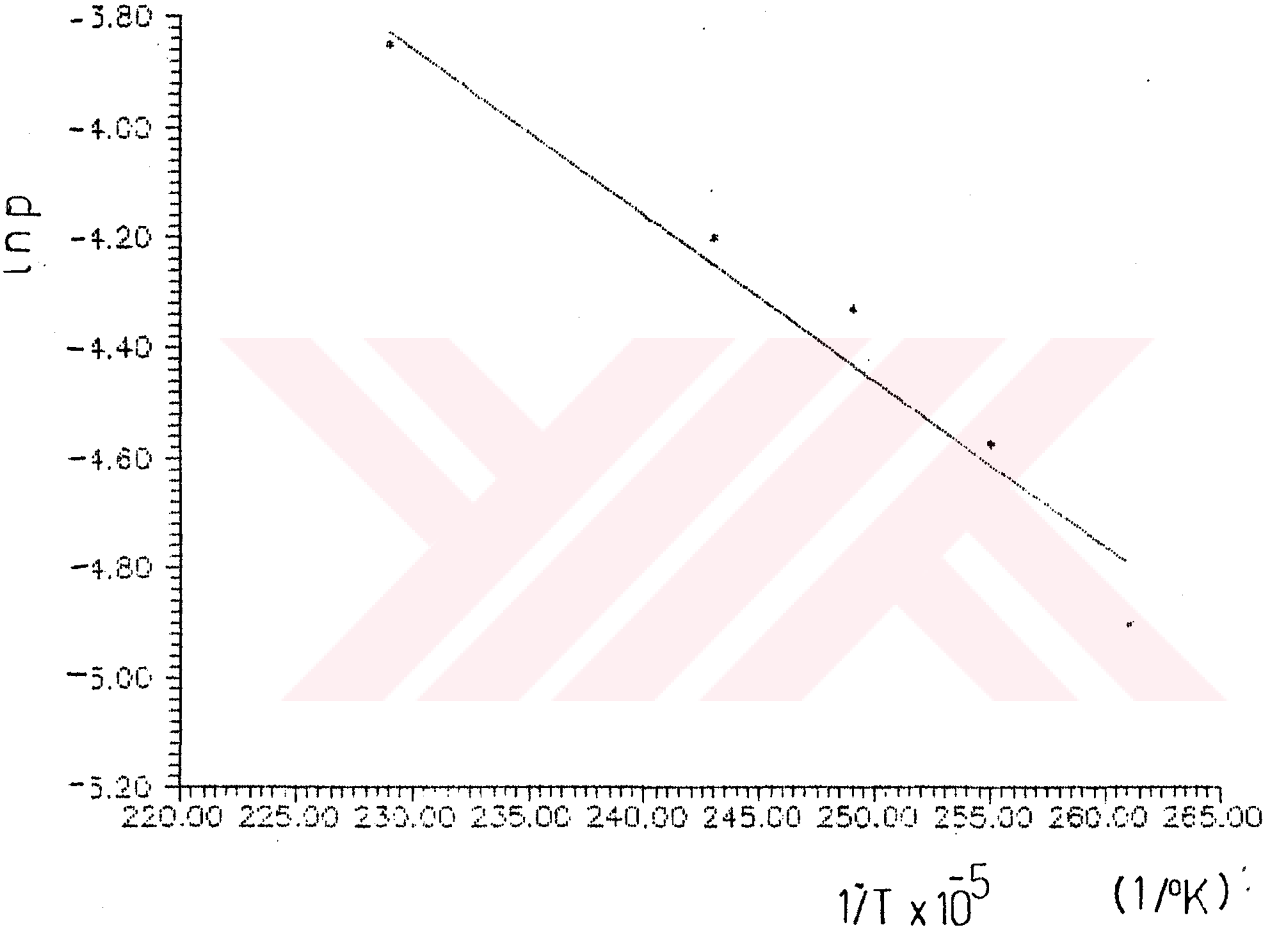

Figure 4.24 (lnp - 1/T) graph of $CaSO_4$:Mn,Mg

4.5 GLOW AND DOSE RESPONSE CURVES OF OTHER SAMPLES

At this section, some glow curves and their dose response curves are plotted. And some detail information about samples are listed below.

4.5.1 $CaSO_4$:Mg - 1

% 1 mole $MgSO_4 . H_2O$
22 hours at room temperature
2 hours at 120 °C
3 hours at 300 °C
2 hours at 900 °C
mesh no : 200
total weight of the sample : 230 mg.
% 76 teflon and % 24 sample are used.
heating rate, b: 2 °C/sec.

This sample is exposed to Sr-90 β radiation, first curve is for 30 Rad, second curve is for 75 Rad, third curve is for 150 Rad, fourth curve is for 225 Rad, and fifth curve is for 255 Rad. The maximum peak height of the glow curves are 150 °C, 175 °C, 160°C, 150°C and 153 °C for 1.,2., 3.,4.,5., curves, respectively. The glow curves are shown in Figure 4.25. Dose response curve is shown in Figure 4. 26. The correlation coefficient of dose response curve which is a straight line, is 0.9938.

4.5.2 $CaSO_4$:Mg -2

% 1 mole $MgSO_4 . H_2O$
24 hours at room temperature
3 hours at 100°C
3 hours at 300°C
2 hours at 900°C

mesh no: 230
total meight:210 mg.
% 60 teflon and % 40 sample are used.
heating rate, b:2 °C /sec.

This sample is exposed to Sr-90 β radiation, first curve is for 30 Rad, second curve is for 60 Rad, third curve is for 105 Rad, fourth curve is for 180 Rad. The maximum peak height of the glow curves are 165.5 °C, 175 °C, 165.5 °C and 175 °C for 1., 2., 3., 4., curves, respectively. The glow curves are shown in Figure 4.27. The dose response curve is shown in Figure 4.28, which is drawn dose (R) versus light intensity. The correlation coefficient of dose response curve which is a straight line, is 0.99468.

4.5.3 $CaSO_4$:Mg - 3

% 1 mole $MgSO_4.H_2O$
23 hours at room temperature
3 hours at 100 °C
3 hours at 300 °C
1.5 hours at 900 °C
mesh no: 230
total weight of the sample:275 mg.
% 76 teflon and % 24 sample are used.
heating rate, b: 2 °C/sec.

The glow curves are shown in Figure 4.29. The exposure doses and their maximum peak temperatures are 7.5 Rad and 200 °C for first curve, 15 Rad and 200 °C for second curve 30 Rad. and 205 °C for third curve. The dose response curve is as shown in Figure 4.30. Its correlation coefficient is 0.9689.

4.5.4 $CaSO_4$:Mg - 4

% 2 mole $MgSO_4 . H_2O$
21 hours at room temperature
2 hours at 100 °C
3 hours at 300 °C
2 hours at 900 °C
Mesh no: 230
total weight of the sample:230 mg.
% 76 teflon and % 24 sample are used.
heating rate, b: 2 °C/sec.

The glow curves are shown in Figure 4.31. The exposure doses and their maximum peak temperatures are 30 Rad and 132 °C for first curve, 45 Rad and 135 °C for second curve 75 Rad and 132 °C for third curve, 150 Rad and 130 °C for fourth curve.The dose response curve is as shown in Figure 4.32. Its correlation coefficient is 0.9997.

4.5.5 $CaSO_4$:Mn - 3

% 1 mole $MnSO_4 . H_2O$
24 hours at room temperature
3 hours at 100 °C
3 hours at 300 °C
2 hours at 900 °C
mesh no: 230
total weight of the sample:230 mg.
% 70 teflon and % 30 sample are used.
heating rate, b: 2 °C/sec.

The glow curves are shown in Figure 4.33. The exposure doses and their maximum peak temperatures are 60 Rad and 137 °C for first curve, 90 Rad and 140 °C for second curve,127.5 Rad and 136 °C for third curve, 165 Rad and 137 °C for fourth curve.The dose response curve is as shown in Figure 4.34. Its correlation coefficient is 0.9987.

4.5.6 $CaSO_4$:Mn - 4

% 1 mole $MnSO_4.H_2O$
24 hours at room temperature
3 hours at 100 °C
3 hours at 300 °C
2 hours at 900 °C
Mesh no: 230
total weight of the saple: 230 mg.
% 70 teflon and % 30 sample are used.
heating rate, B: 2 °C/sec.

The glow curves are shown in Figure 4.35. The exposure doses and their maximum peak temperature are 22.5 Rad and 140 °C for first curve, 45 Rad and 140 °C for second curve, 75 Rad and 130 °C for third curve, 150 Rad and 137.5 °C for fourth curve. The dose response curve is as shown in Figure 4.36. Its correlation coefficient is 0.9981.

4.5.7 $CaSO_4$:Mn,Mg - 1

% 1 mole $MnSO_4.H_2O$
% 1 mole $MgSO_4.H_2O$
22 hours at room temperature
2 hours at 100 °C
3 hours at 300 °C
2 hours at 900 °C
Mesh no:230
total weight of the sample: 300 mg.
% 76 teflon and % 24 sample are used.
heating rate, B: 2 °C/sec.

The glow curves are shown in Figure 4.37. The exposure doses and their maximum peak temperatures are 15 Rad and 150 °C for first curve, 45 Rad and 130 °C for second curve, 48 Rad and 125 °C for third curve, The dose response curve is as shown in Figure 4.38. Its correlation coefficient is 0.996.

4.5.8 $CaSO_4$:Mn,Mg - 2

% 3 mole $MnSO_4.H_2O$
% 3 molu $MgSO_4.H_2O$
23 hours at room temperature
3 hours at 100 °C
3 hours at 300 °C
2.5 hours at 900 °C
Mesh no: 230
total weight of the sample: 250 mg.
% 76 teflon and % 24 sample are used.
heating rate, b: 2 °C/sec.

The glow curves are shown in Figure 4.39. The exposure doses and their maximum peak temperatures are 30 Rad and 155 °C for first curve, 60 Rad and 150 °C for second curve 165 Rad and 150 °C for third curve, The dose response curve is as shown in Figure 4.40. Its correlation coefficient is 0.997.

4.5.9 $CaSO_4$:Mn,Mg - 3

% 3 mole $MnSO_4.H_2O$
% 3 mole $MgSO_4.H_2O$
23 hours at room temperature
3 hours at 100 °C
3 hours at 300 °C
2.5 hours at 900 °C
mesh no: 230
total weight of the sample:275 mg.

% 75 teflon and % 25 sample are used.
heating rate b: 2 °C/sec.

The glow curves are shown in Figure 4.41. The exposure doses and their maximum peak temperatures are 45 Rad and 128 °C for first curve, 60 Rad and 128 °C for second curve, 75 Rad and 127 °C for third curve, The dose response curve is as shown in Figure 4.42. Its correlation coefficient is 0.9532.

4.5.10 $CaSO_4$:Mn,Mg - 4

% 3 mole $MnSO_4.H_2O$
% 3 mole $MgSO_4.H_2O$
23 hours at room temperature
3 hours at 100 °C
3 hours at 300 °C
2.5 hours at 900 °C
mesh no: 140
total weight of the sample:230 mg.
% 76 teflon and %24 sample are used.
heating rate, b: 2 °C/sec.

The glow curves are shown in Figure 4.43. The exposure doses and their maximum peak temperatures are 60 Rad and 132.5 °C for first curve 90 Rad and 137.5 °C for second curve, 150 Rad and 128 °C for third curve The does response curve is as shown in Figure 4.44. Its correlation coefficient is 0.971.

4.5.11 $CaSO_4$:Mn,Mg - 5

% 1 mole $MnSO_4.H_2O$
% 1 mole $MgSO_4.H_2O$
22 hours at room temperature
2 hours at 100 °C

3 hours at 300 °C
2 hours at 900 °C
mesh no:200
total weight of the sample: 275 mg.
% 76 teflon and % 24 sample are used.
heating rate, b: 2 °C/sec.

The glow curves are shown in Figure 4.45. The exposure doses and their maximum peak temperatures are 4.5 Rad and 150 °C for first curve, 7.5 Rad and 140 °C for second curve, 21 Rad and 137 °C for third curve, 30 Rad and 138 °C for fourth curve., The dose response curve is as shown in Figure 4. 46. Its correlation coefficient is 0.989.

4.5.12 $CaSO_4$:Se

% 1 mole SeO_2
20 hours at room temperature
3 hours at 100 °C
3 hours at 300 °C
2 hours at 900 °C
mesh no: 140
total weight of the sample: 230 mg.
% 76 teflon and % 27 sample are used.
heating rate, b: 2 °C/sec.

The glow curves are shown in Figure 4.47. The exposure doses and their maximum peak temperature are 7.5 Rad and 125 °C for first curve, 15 Rad and 127 °C for second curve, 30 Rad and 125 °C for third curve, The does response curve is as shown in Figure 4.48. Its correlation coefficient is 0.999.

4.5.13 $CaSO_4$:Zn

% 1 mole $ZnSO_4.H_2O$
23 hours at room temperature
2 horus at 100 °C
3 hours at 300 °C
2 hours at 900 °C
mesh no: 200
total weight of the sample:250 mg.
% 76 teflon and % 24 sample are used.
heating rate, b: 2 °C sec.

The glow curves are shown in Figure 4.49. The exposure doses and their maximum peak temperatures are 7.5 Rad and 146 °C for first curve, 22.5 Rad and 145 °C for second curve, 30 Rad and 145 °C for third curve, The does response curve is as shown in Figure 4.49. Its correlation coefficient is 0.998.

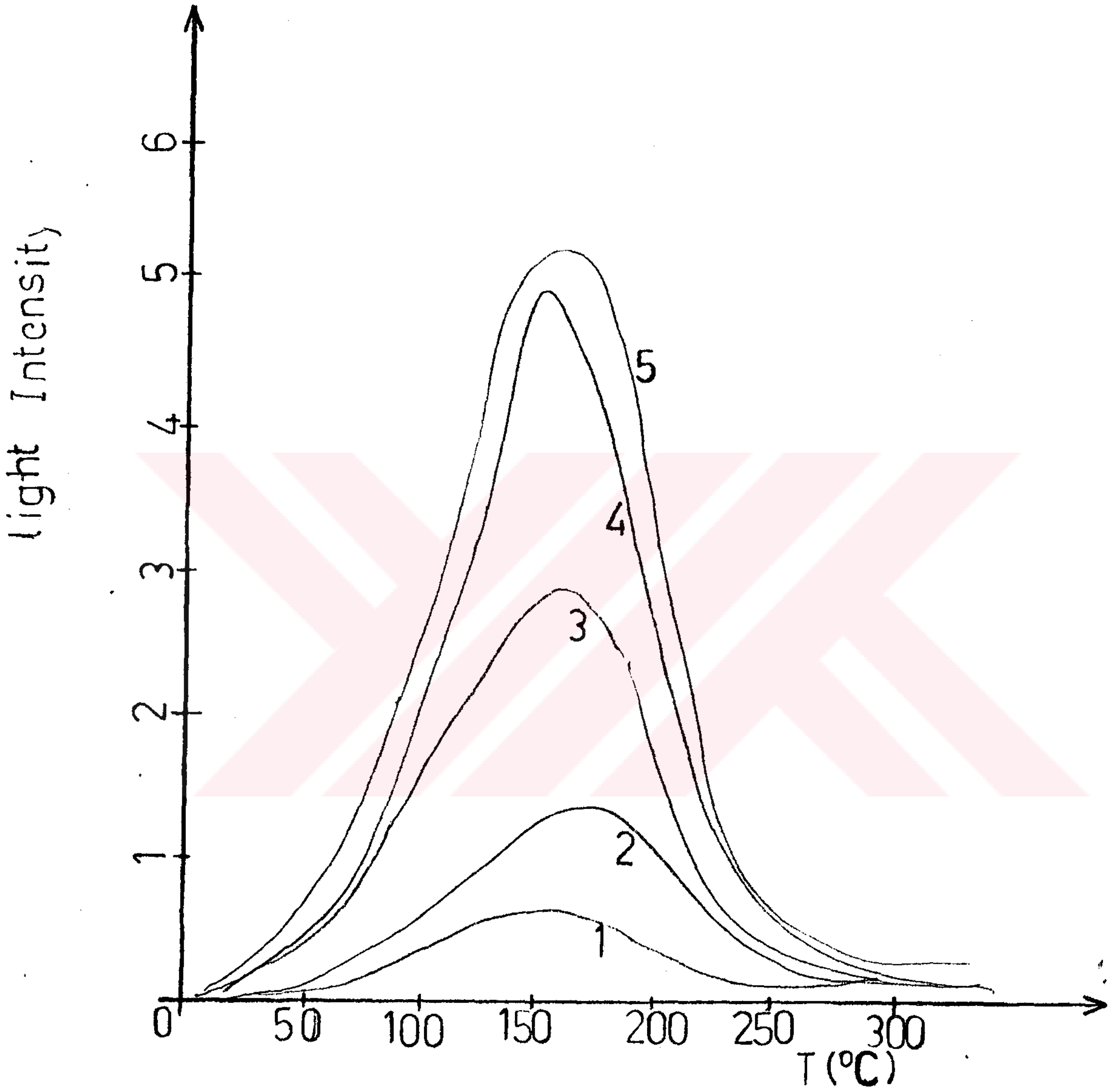

Figure 4.25 Glow curves of $CaSO_4$:Mg - 1, exposed to Sr-90 β radiation;

1) 30 Rad
2) 75 Rad
3) 150 Rad
4) 225 Rad
5) 255 Rad

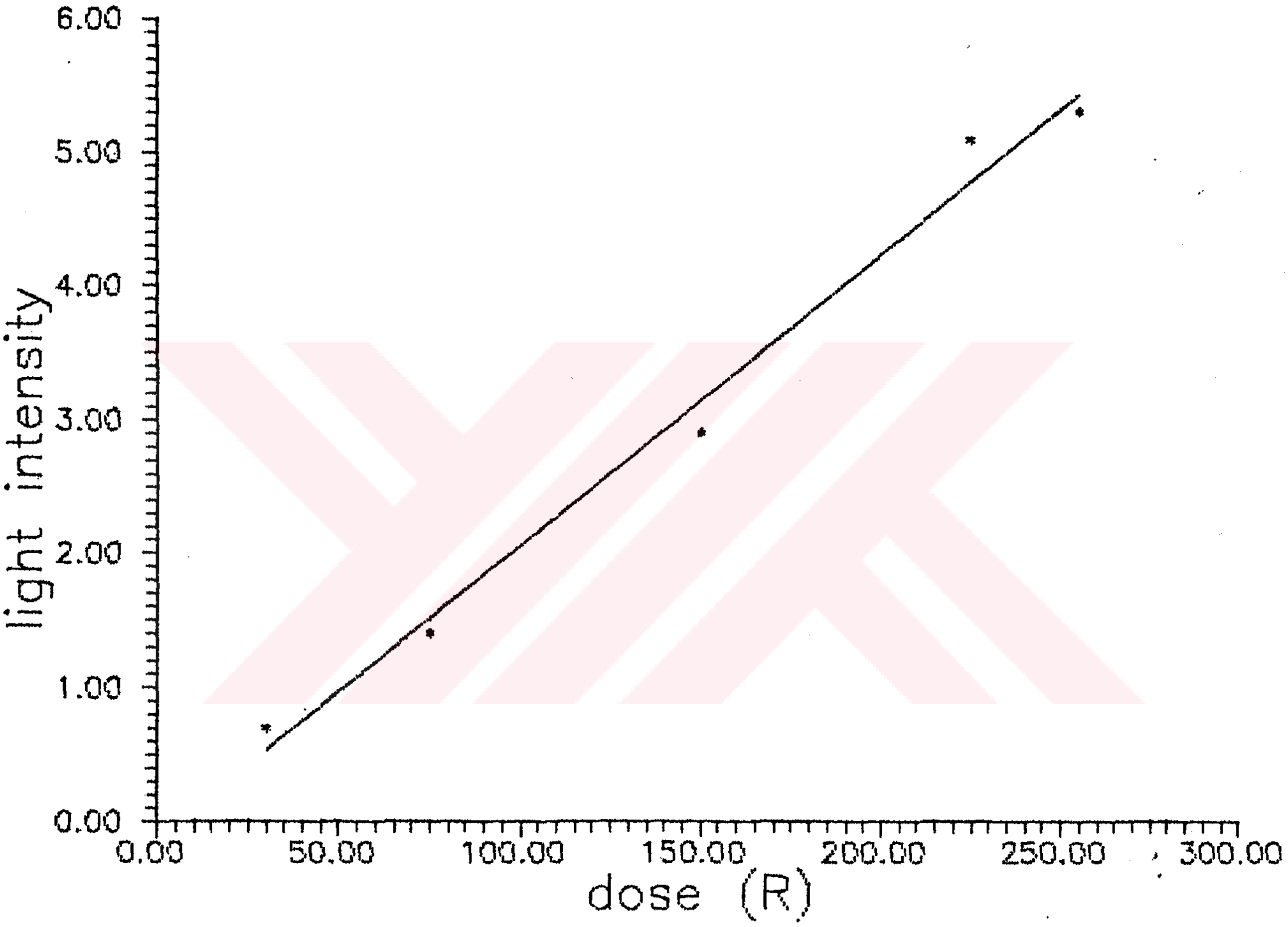

Figure 4.26 The dose response curve of $CaSO_4$:Mg -1 teflon dosimeter.
r = 0.9938

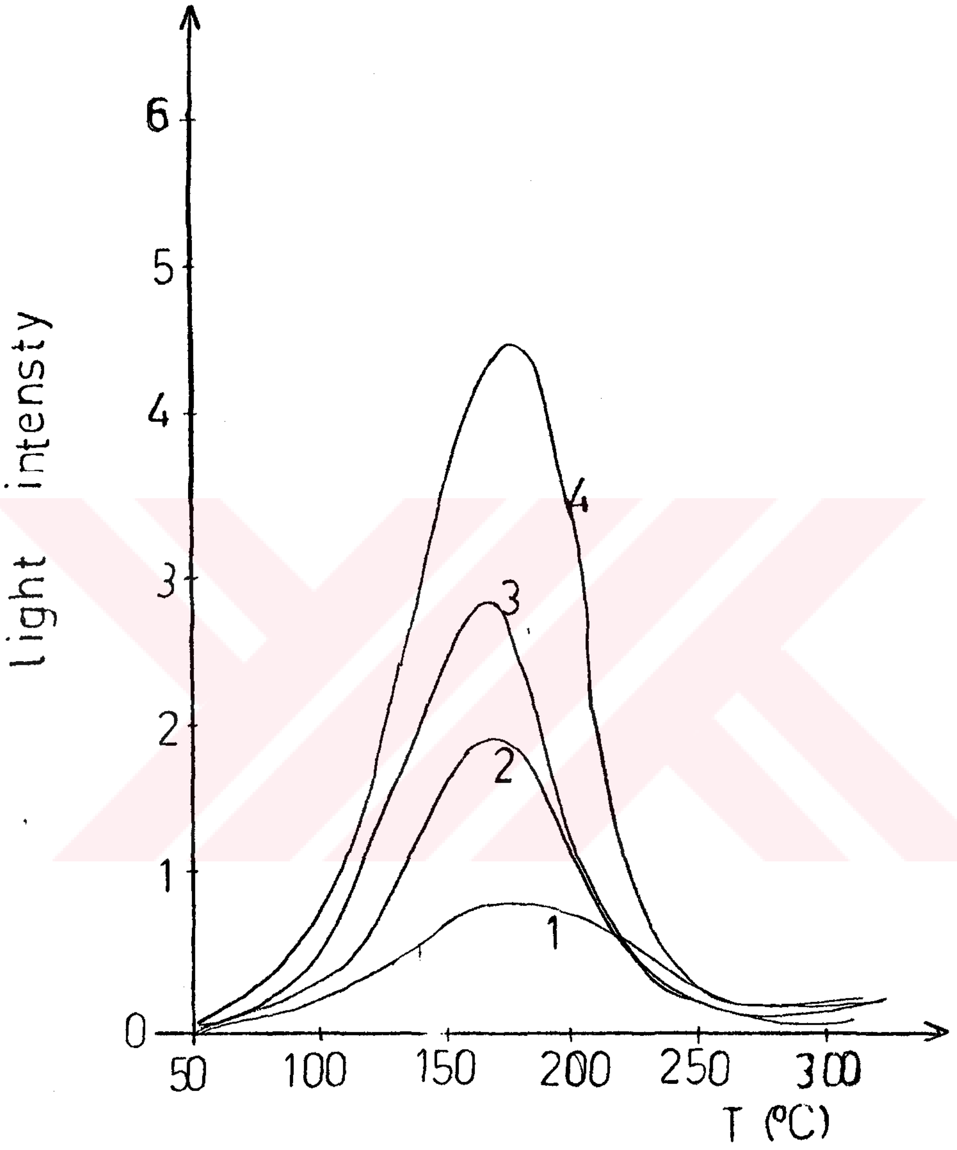

Figure 4.27 Glow curves of $CaSO_4$: Mg - 2, exposed to Sr-90 β radiation;

1) 30 Rad
2) 60 Rad
3) 105 Rad
4) 180 Rad

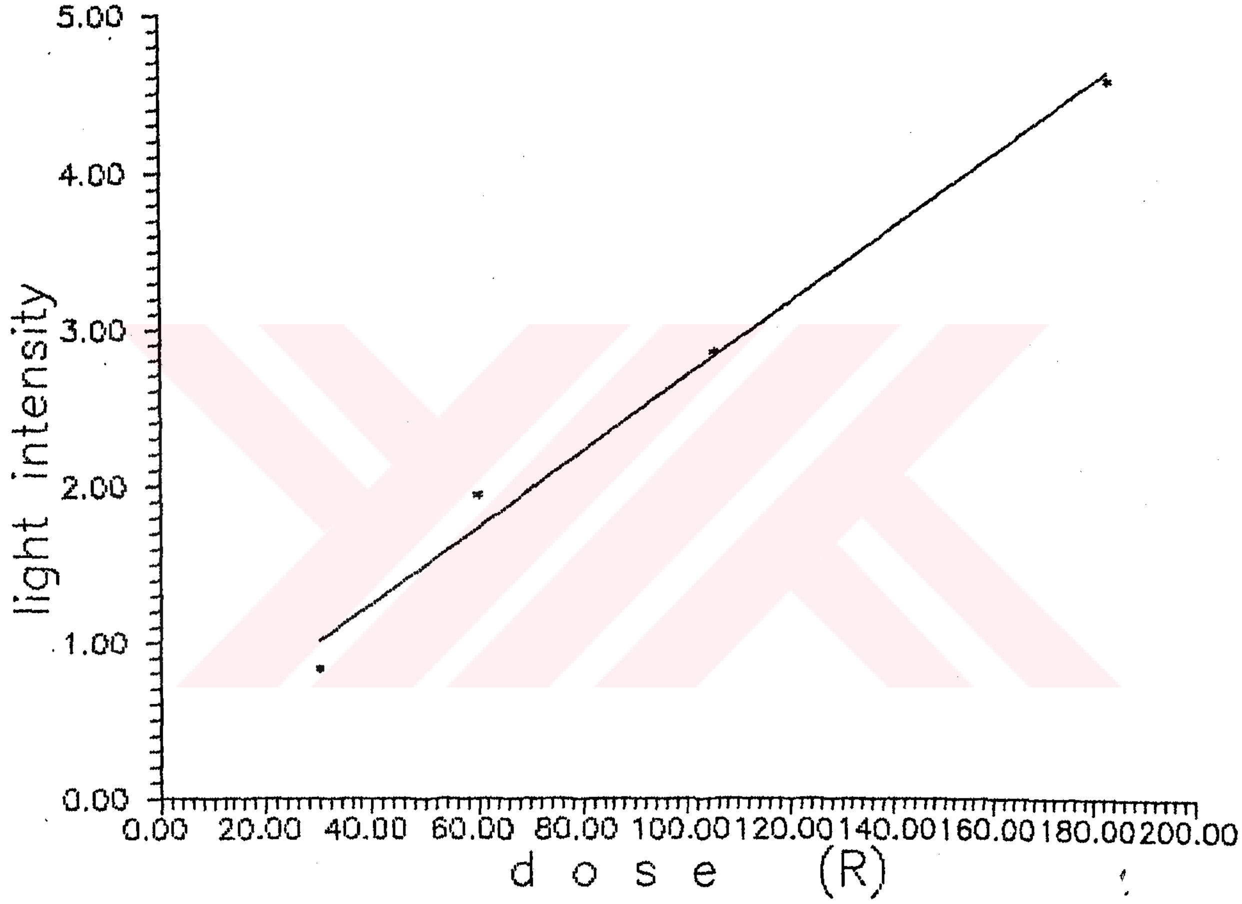

Figure 4.28 Dose response curve of $CaSO_4$:Mg - 2 teflon dosimeter.
r = 0.99468

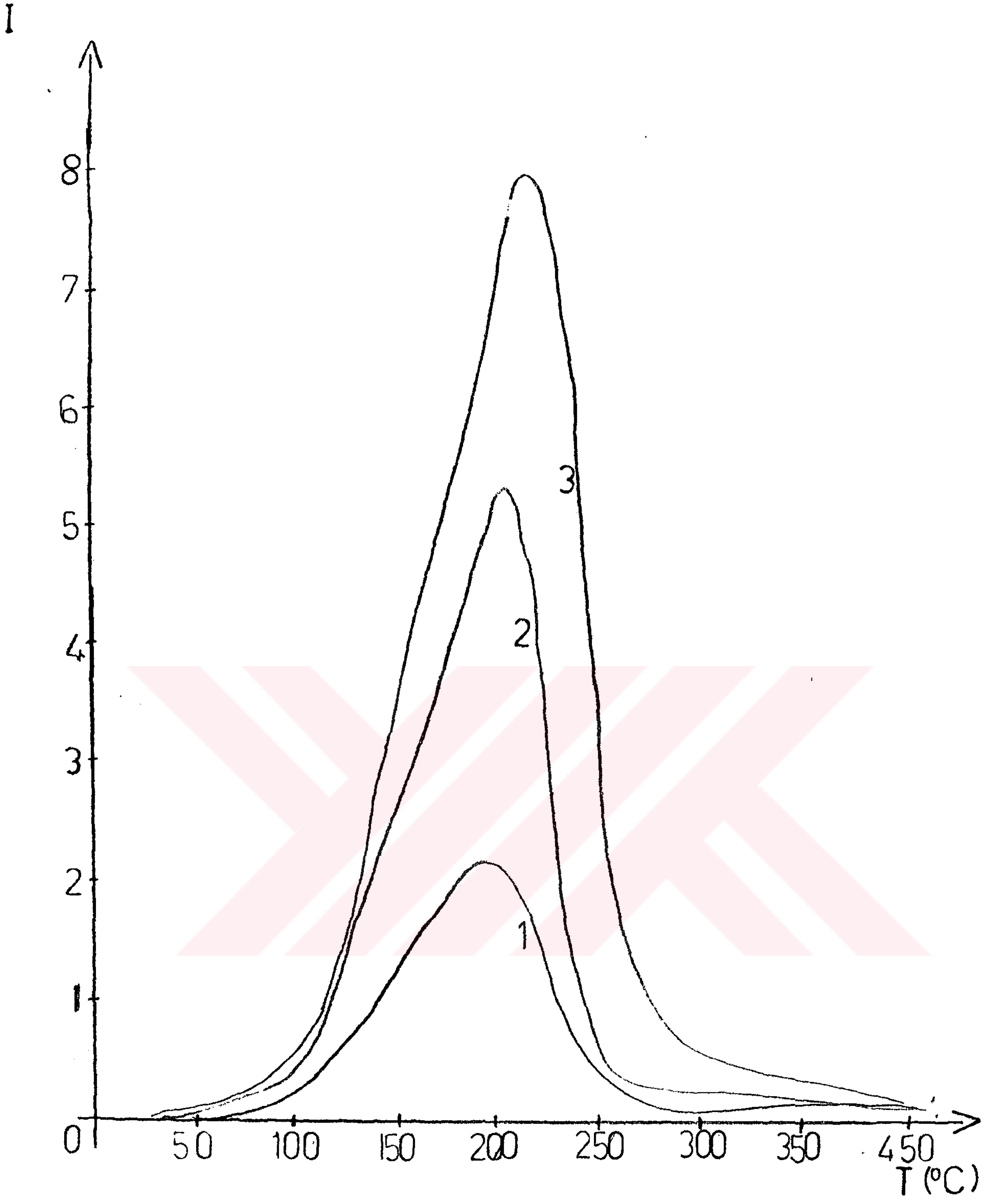

Figure 4.29 Glow curves of $CaSO_4$:Mg - 3, exposed to Sr-90 β radiation;

1) 7.5 Rad

2) 15 Rad

3) 30 Rad

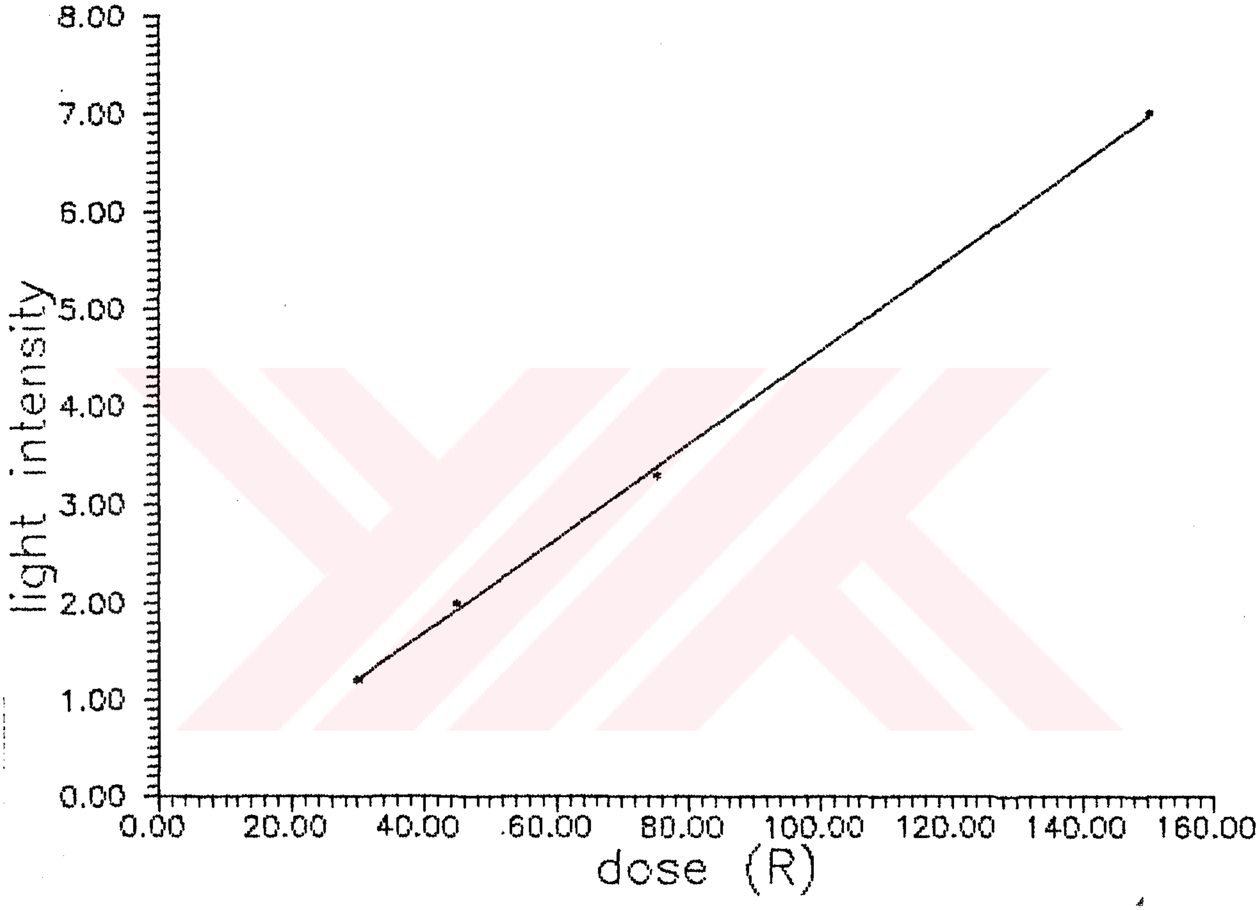

Figure 4.30 Dose response curve of $CaSO_4$:Mg - 3 teflon dosimeter.
r = 0.9689

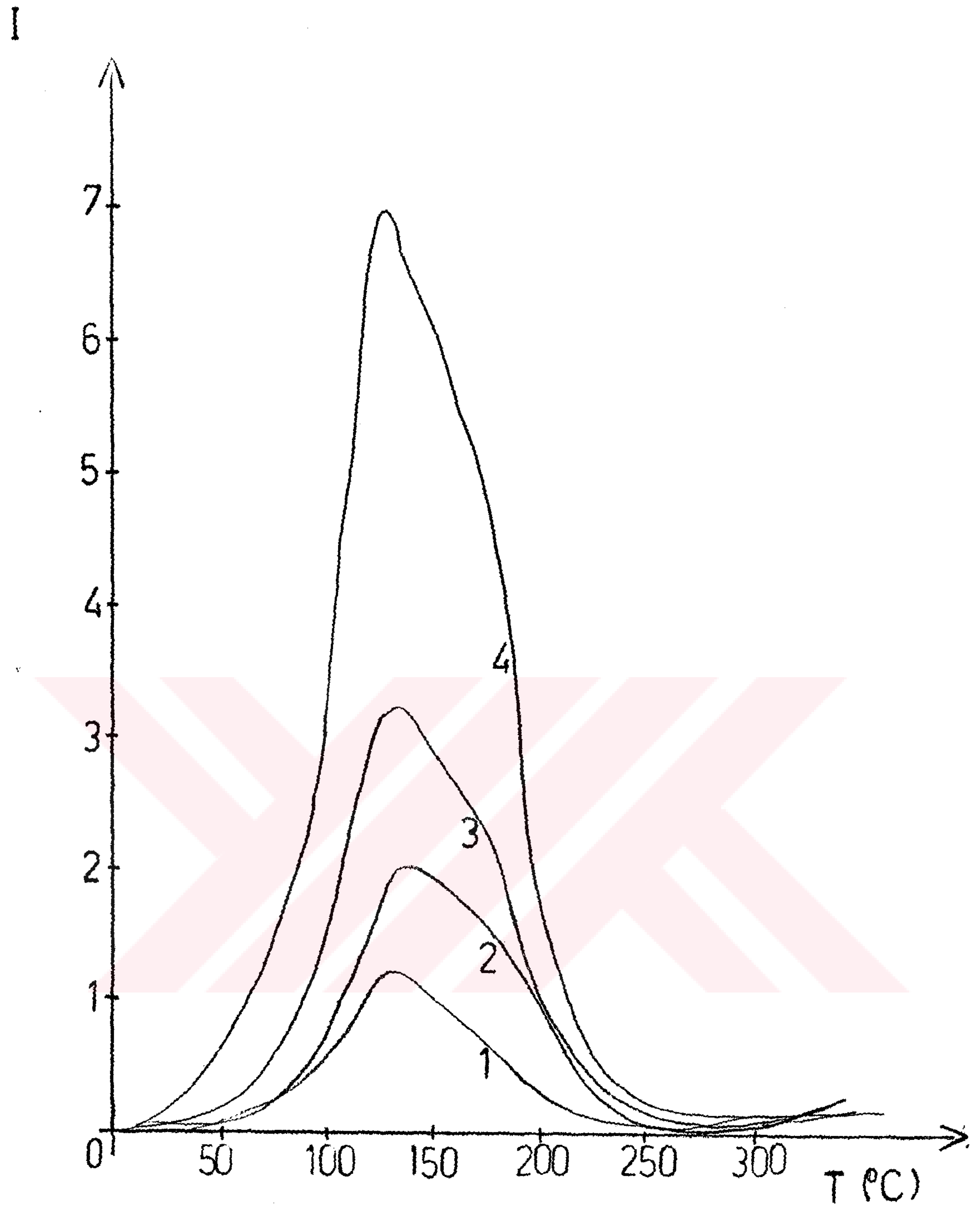

Figure 4.31 Glow curves of $CaSO_4$:Mg - 4, exposed to Sr-90 β radiation;

1) 30 Rad
2) 45 Rad
3) 75 Rad
4) 150 Rad

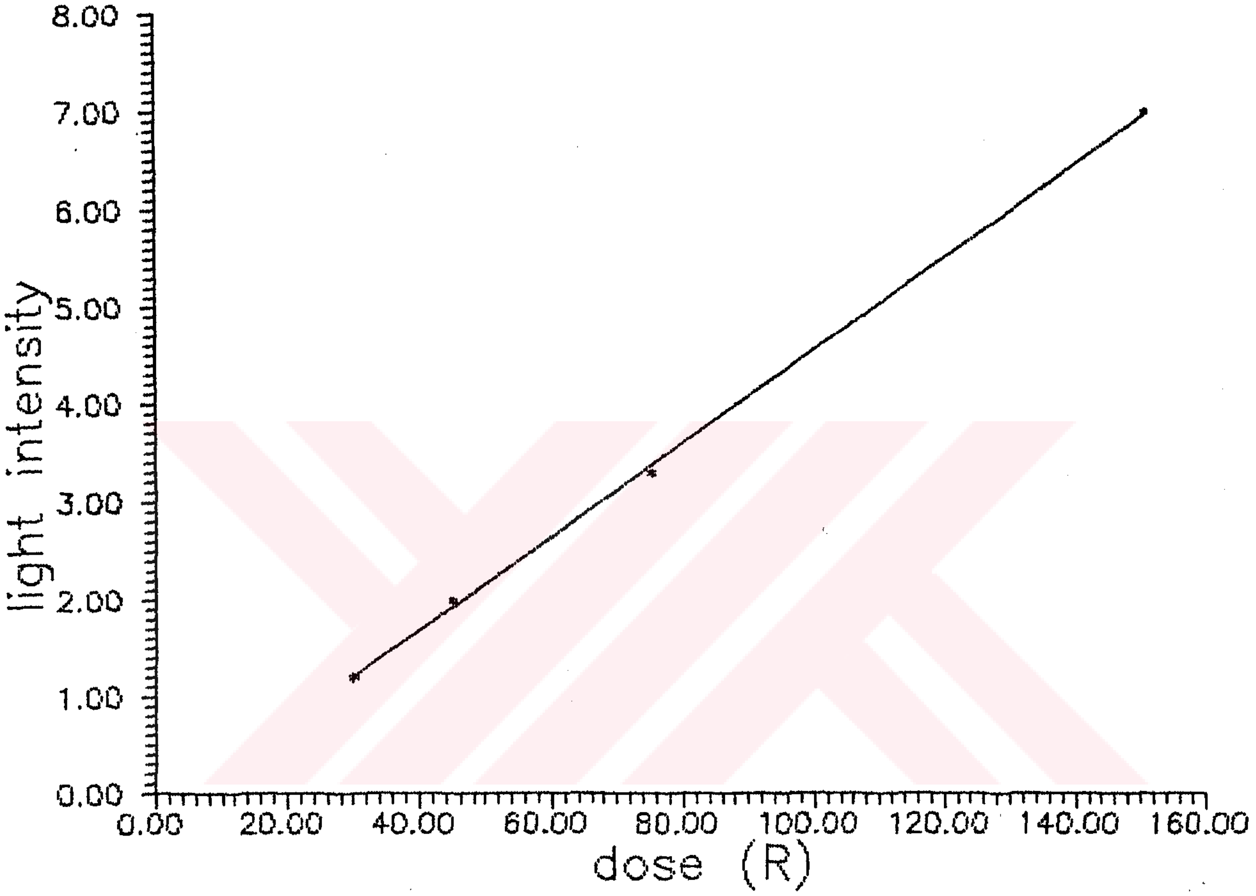

Figure 4.32 Dose response curve of $CaSO_4$:Mg - 4 teflon dosimeter.
r = 0.9997

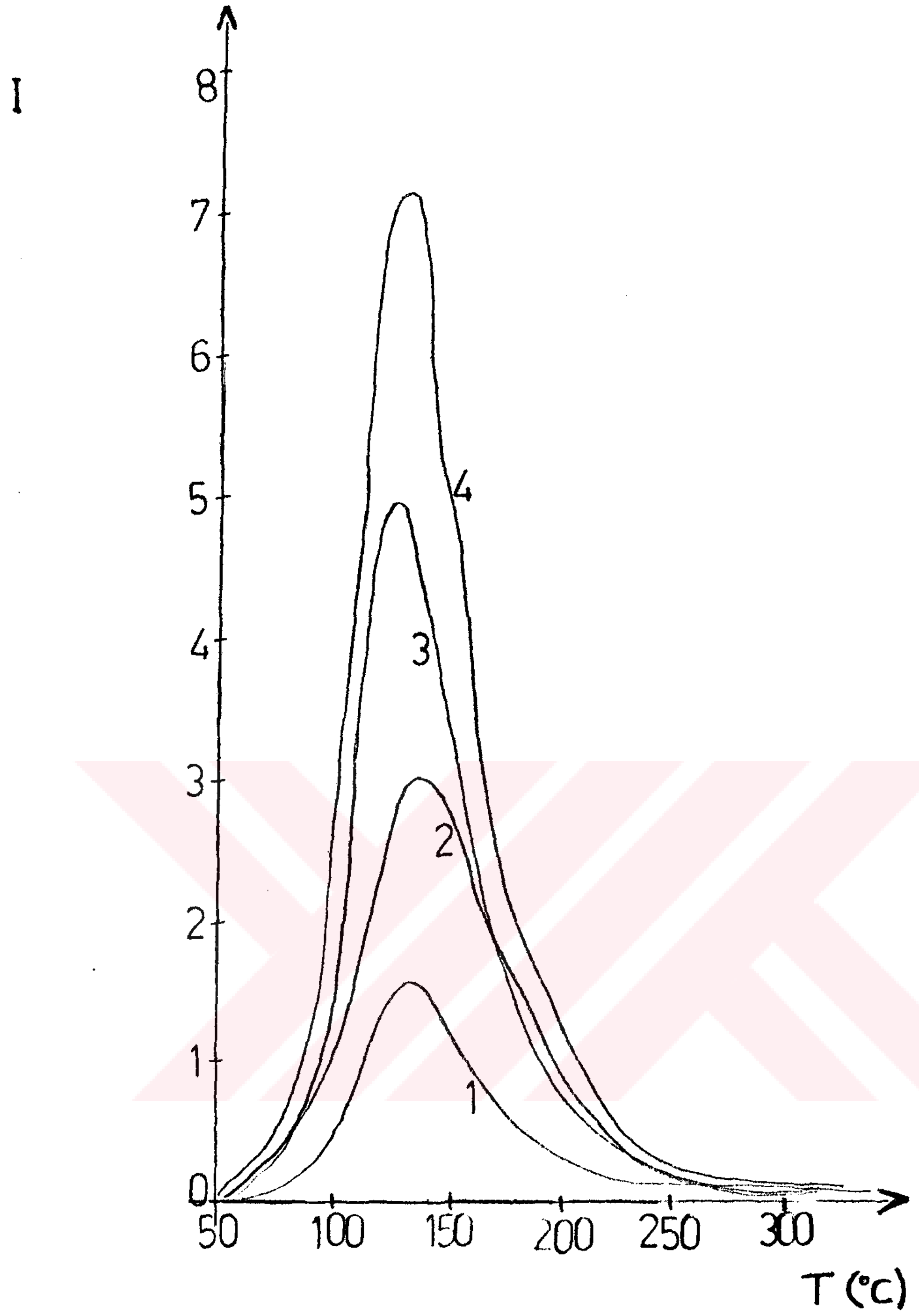

Figure 4.33 Glow curves of $CaSO_4$:Mn - 3, exposed to Sr-90 β radiation;

1) 60 Rad
2) 90 Rad
3) 127.5 Rad
4) 165 Rad

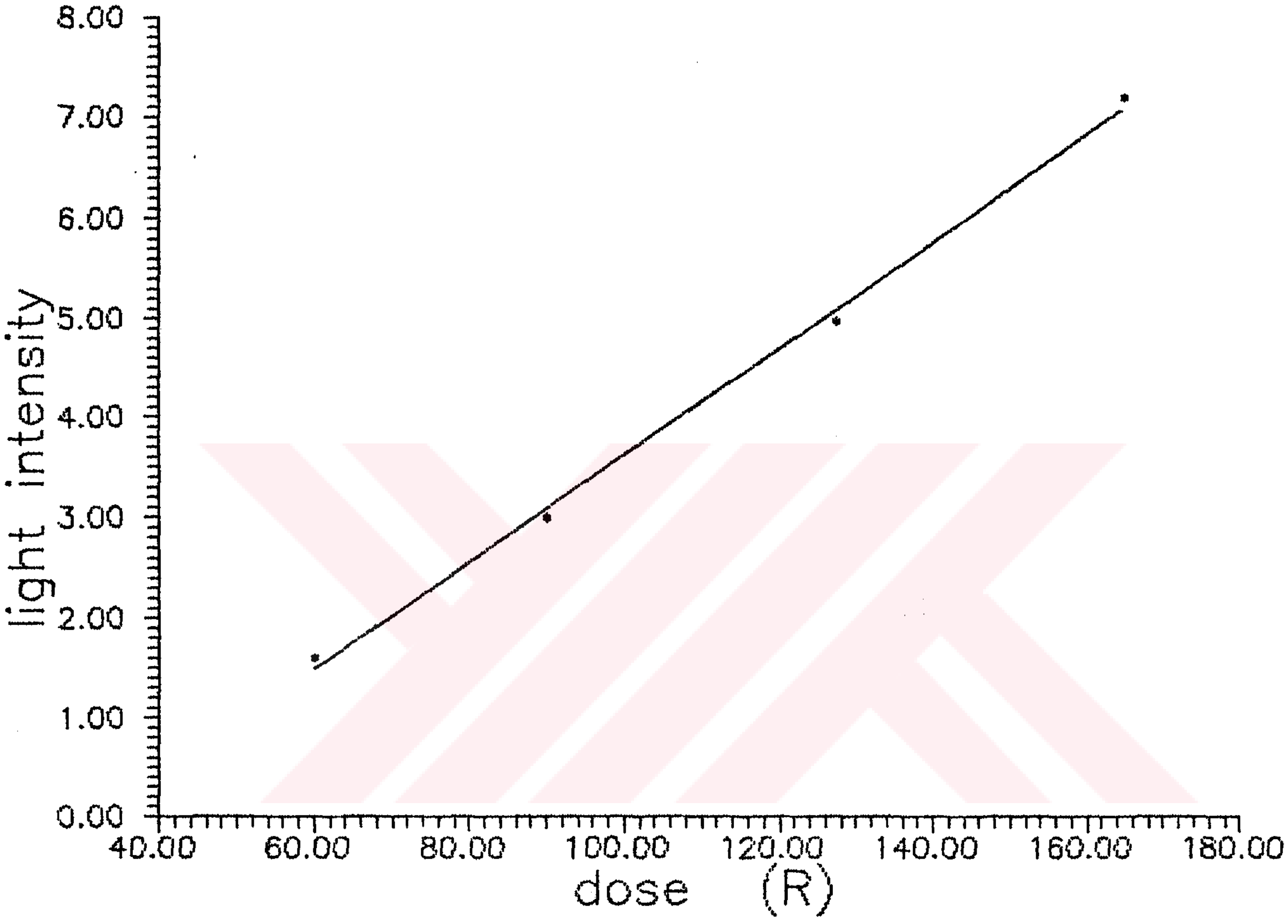

Figure 4.34 Dose response curve of $CaSO_4$:Mn - 3 teflon dosimeter.
r = 0.9987

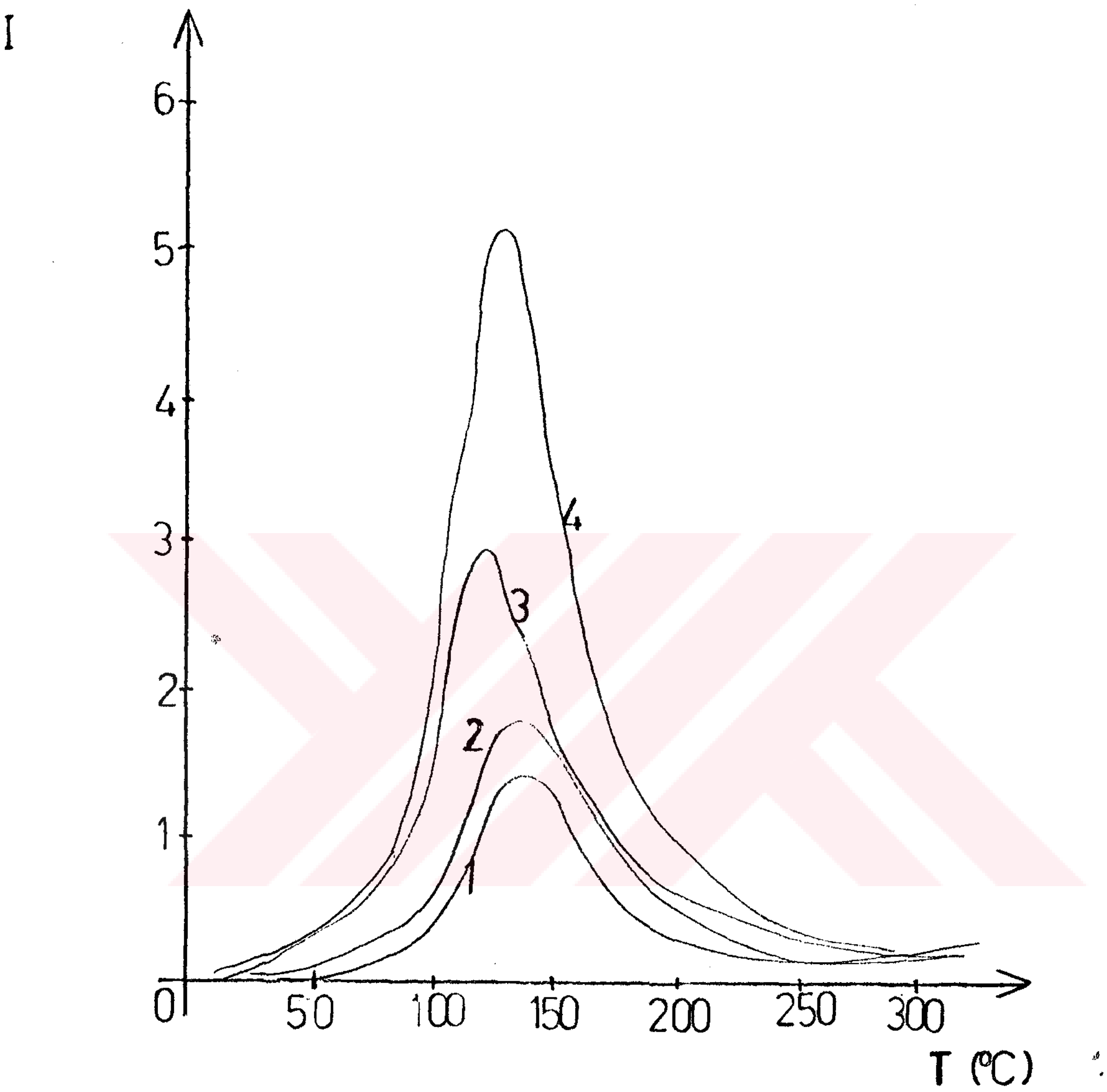

Figure 4.35 Glow curves of $CaSO_4$:Mn - 4, exposed to Sr-90 β radiation;

1) 22.5 Rad

2) 45 Rad

3) 75 Rad

4) 150 Rad

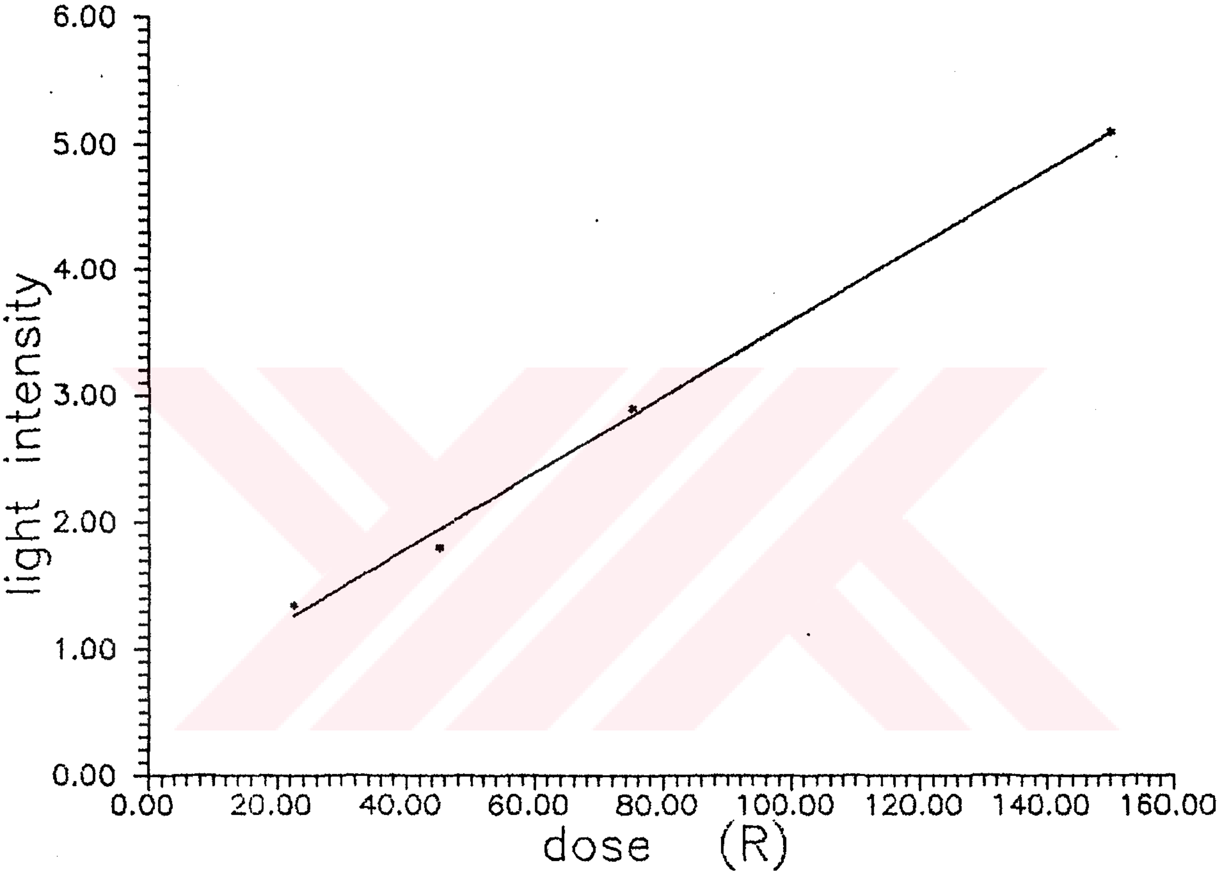

Figure 4.36 Dose response curve of $CaSO_4$:Mn - 4 teflon dosimeter.
r = 0.9981

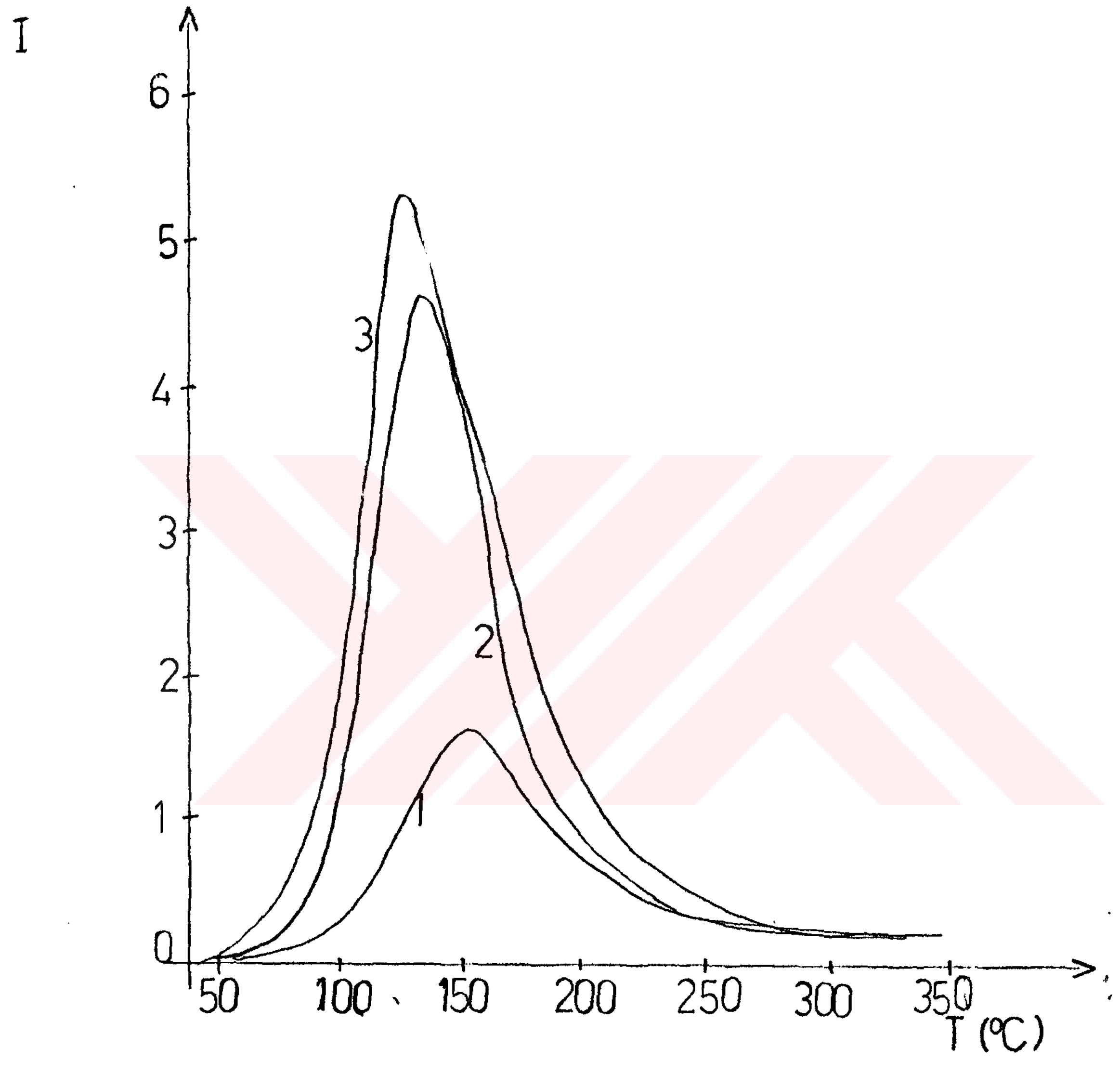

Figure 4.37 Glow curves of $CaSO_4$:Mn,Mg - 1 exposed to Sr-90 β radiation;

1) 15 Rad
2) 45 Rad
3) 48 Rad

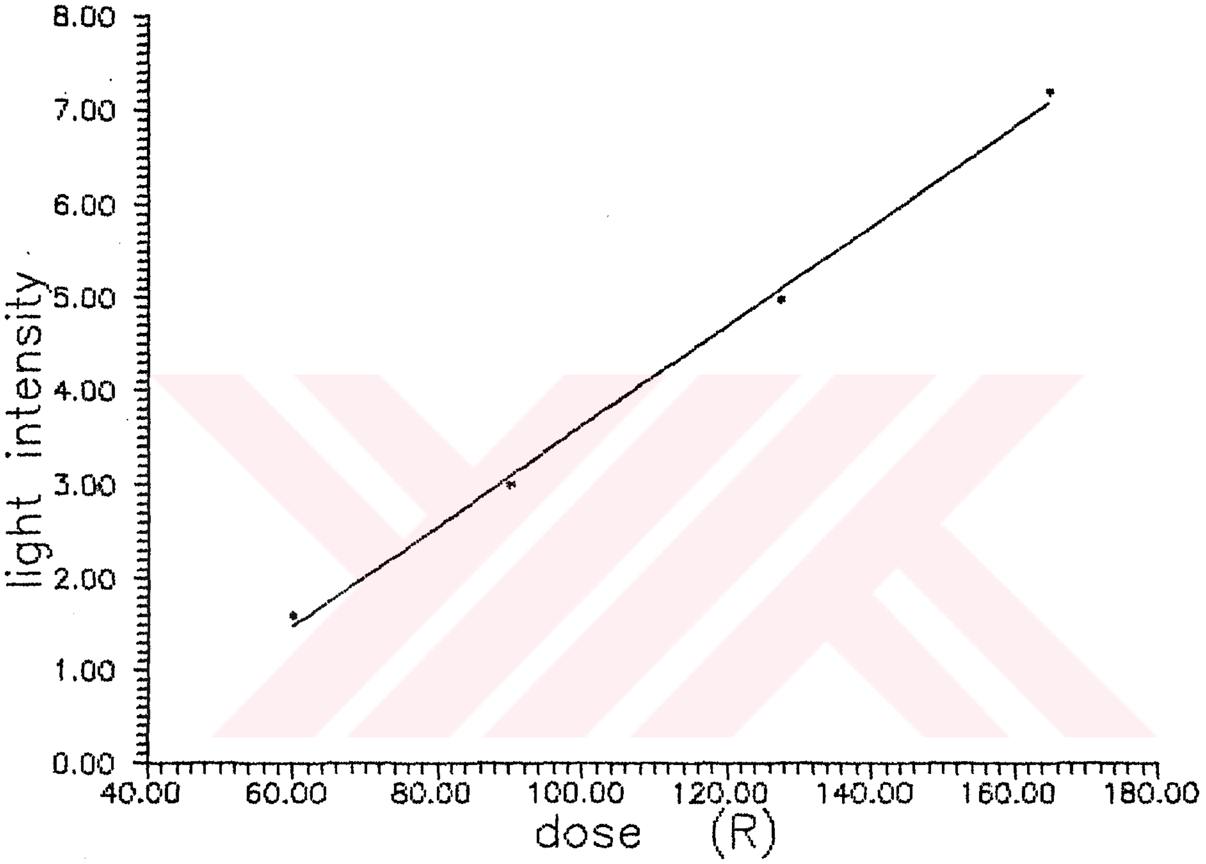

Figure 4.38 Dose response curve of $CaSO_4$:Mn,Mg - 1 teflon dosimeter.
$r = 0.996$

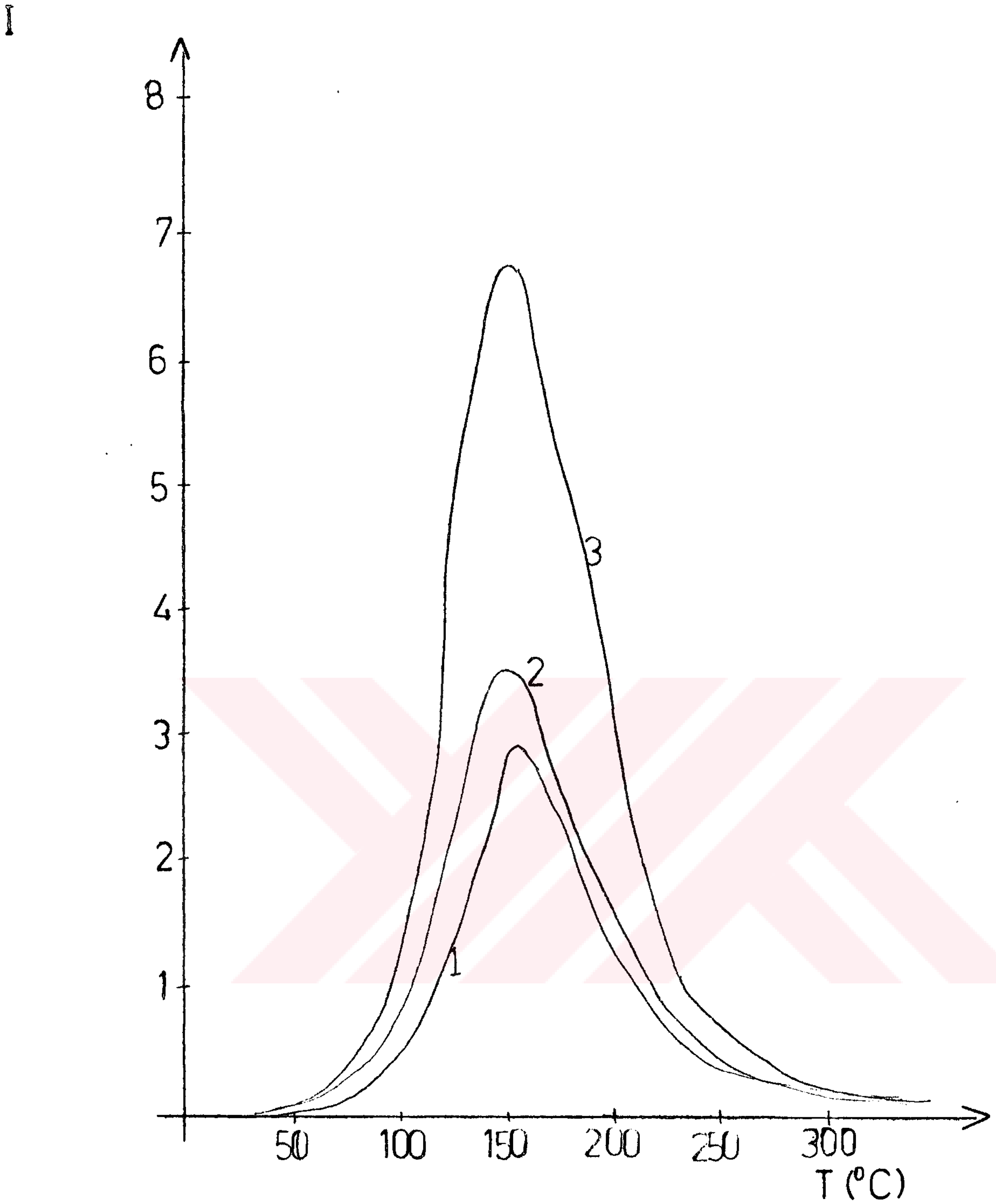

Figure 4.39 Glow curves of $CaSO_4$:Mn,Mg - 2 exposed to Sr-90 β radiation;

1) 30 Rad
2) 60 Rad
3) 165 Rad

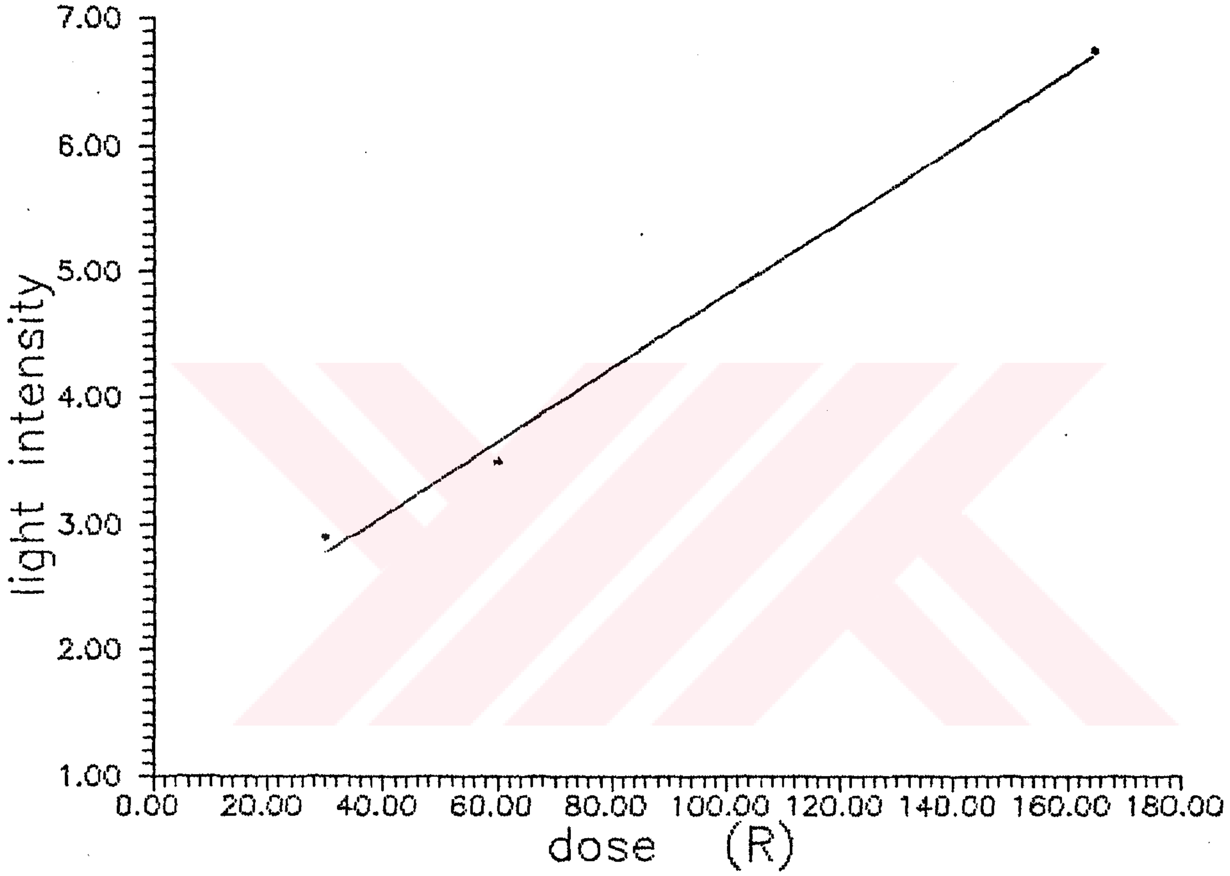

Figure 4.40 Dose response curve of $CaSO_4$:Mn,Mg -2 teflon dosimeter.
r = 0.997

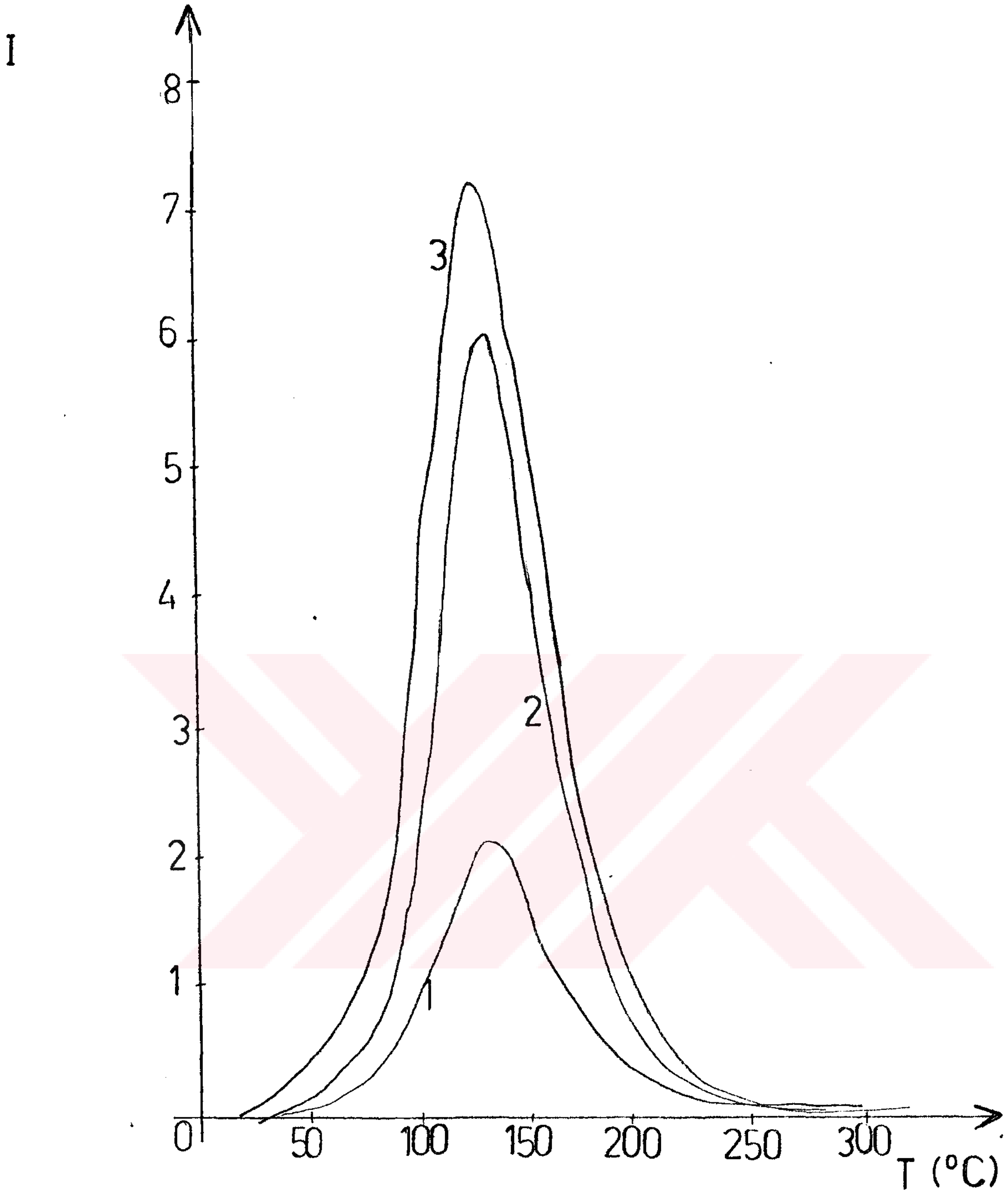

Figure 4.41 Glow curves of $CaSO_4$:Mn,Mg - 3, exposed to Sr-90 β radiation;

1) 45 Rad
2) 60 Rad
3) 75 Rad

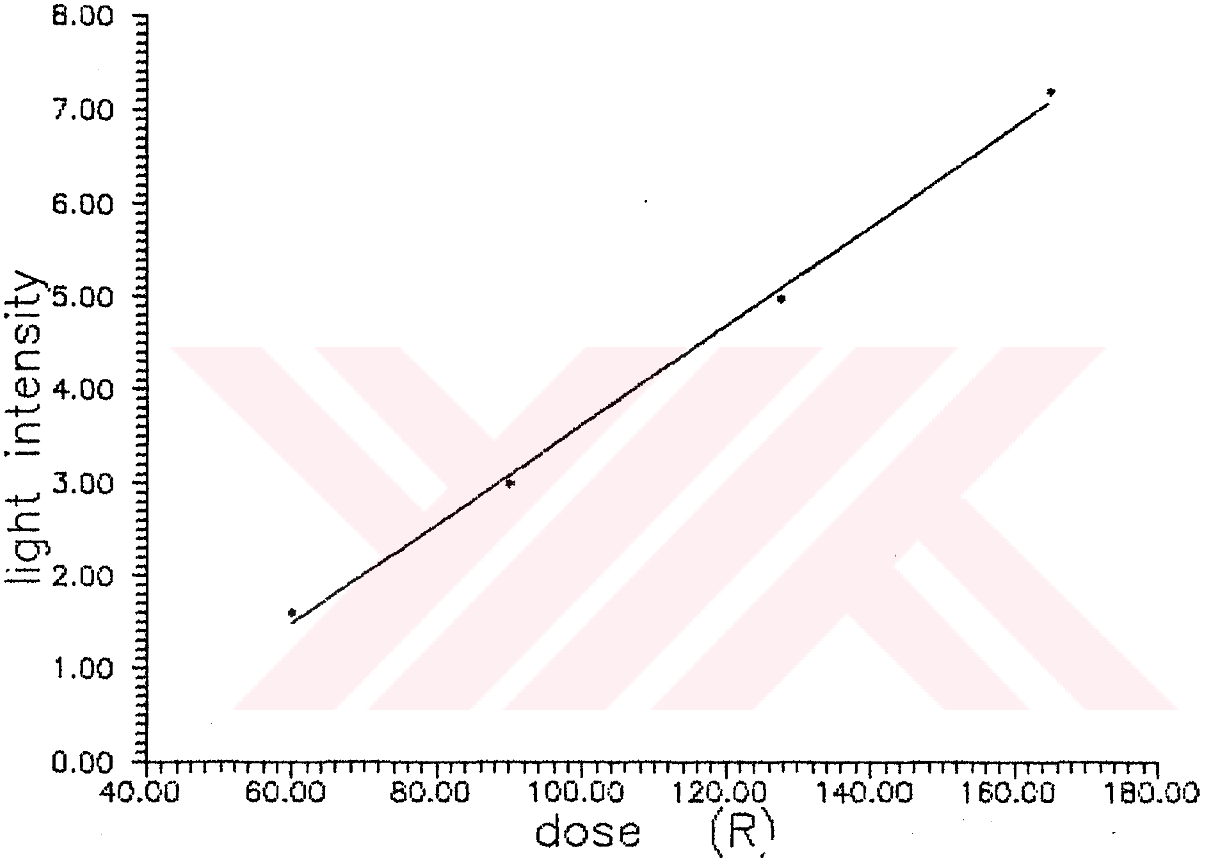

Figure 4.42 Dose response curve of $CaSO_4$:Mn,Mg -3 teflon dosimeter.
r = 0.9532

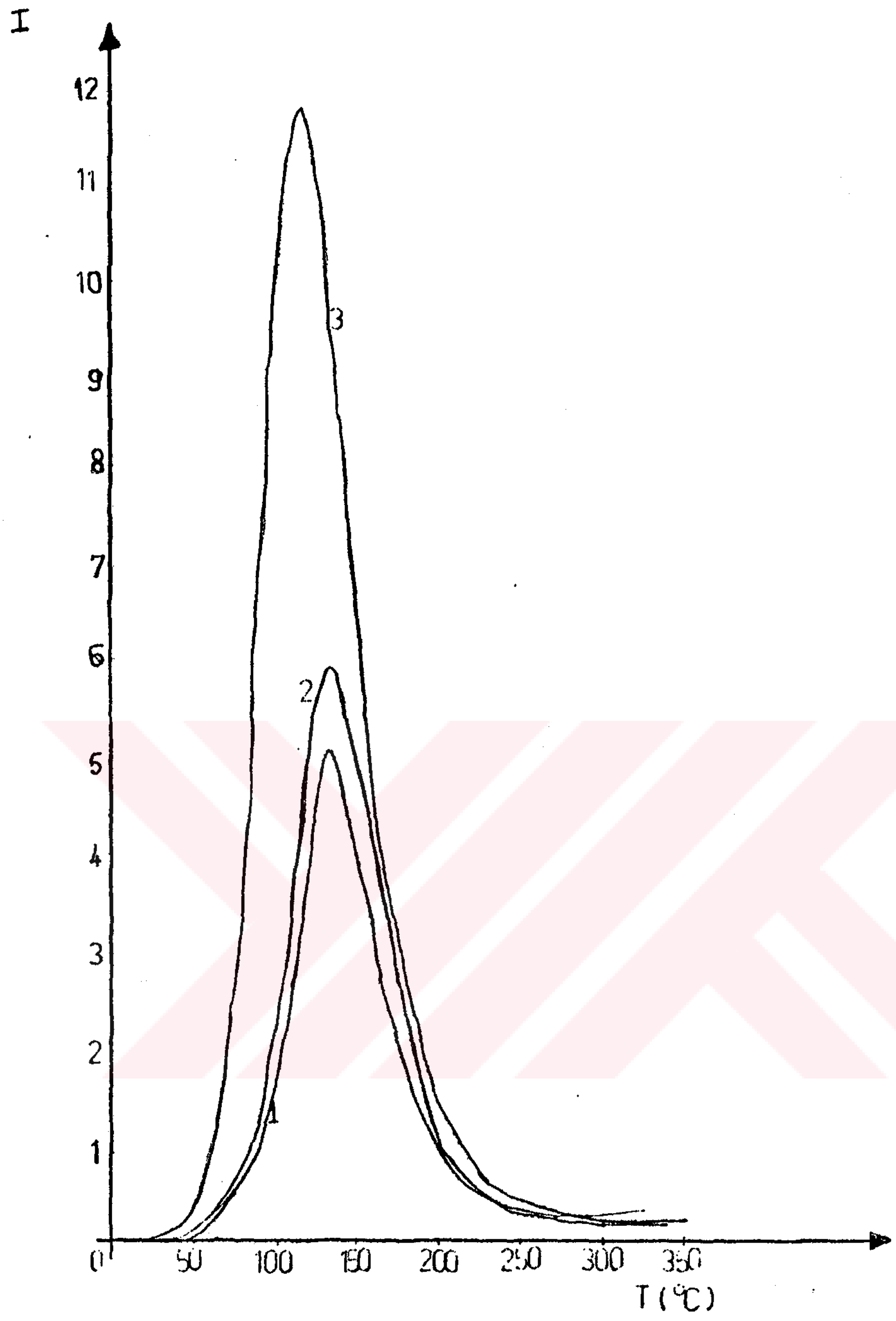

Figure 4.43 Glow curves of $CaSO_4$:Mn,Mg - 4,exposed to Sr-90 β radiation;

1) 60 Rad
2) 90 Rad
3) 150 Rad

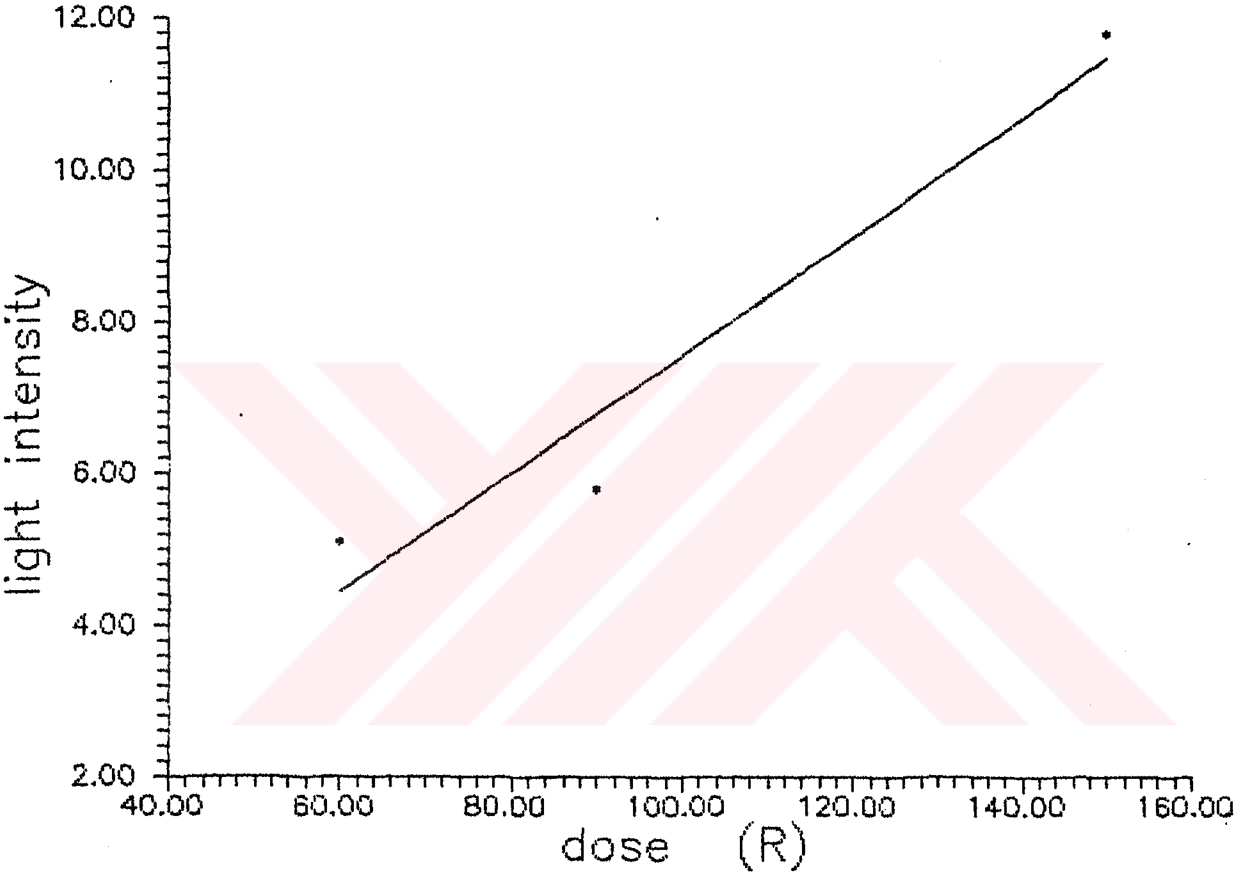

Figure 4.44 Dose response curve of $CaSO_4$:Mn,Mg - 4 teflon dosimeter.
r = 0.971

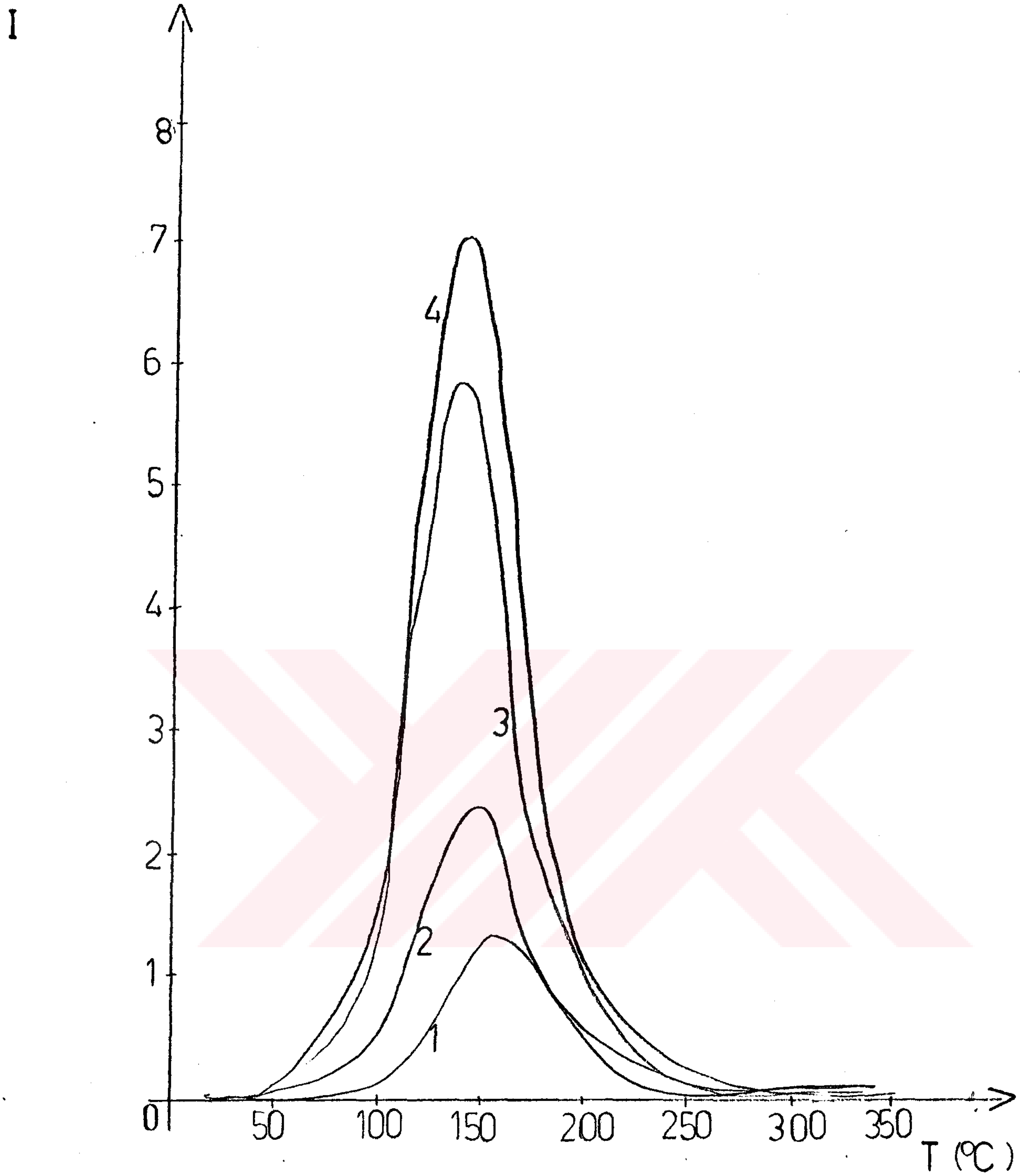

Figure 4.45 Glow curves of $CaSO_4$:Mn,Mg - 5, exposed to Sr-90 β radiation;

1) 4.5 Rad
2) 7.5 Rad
3) 21 Rad
4) 30 Rad

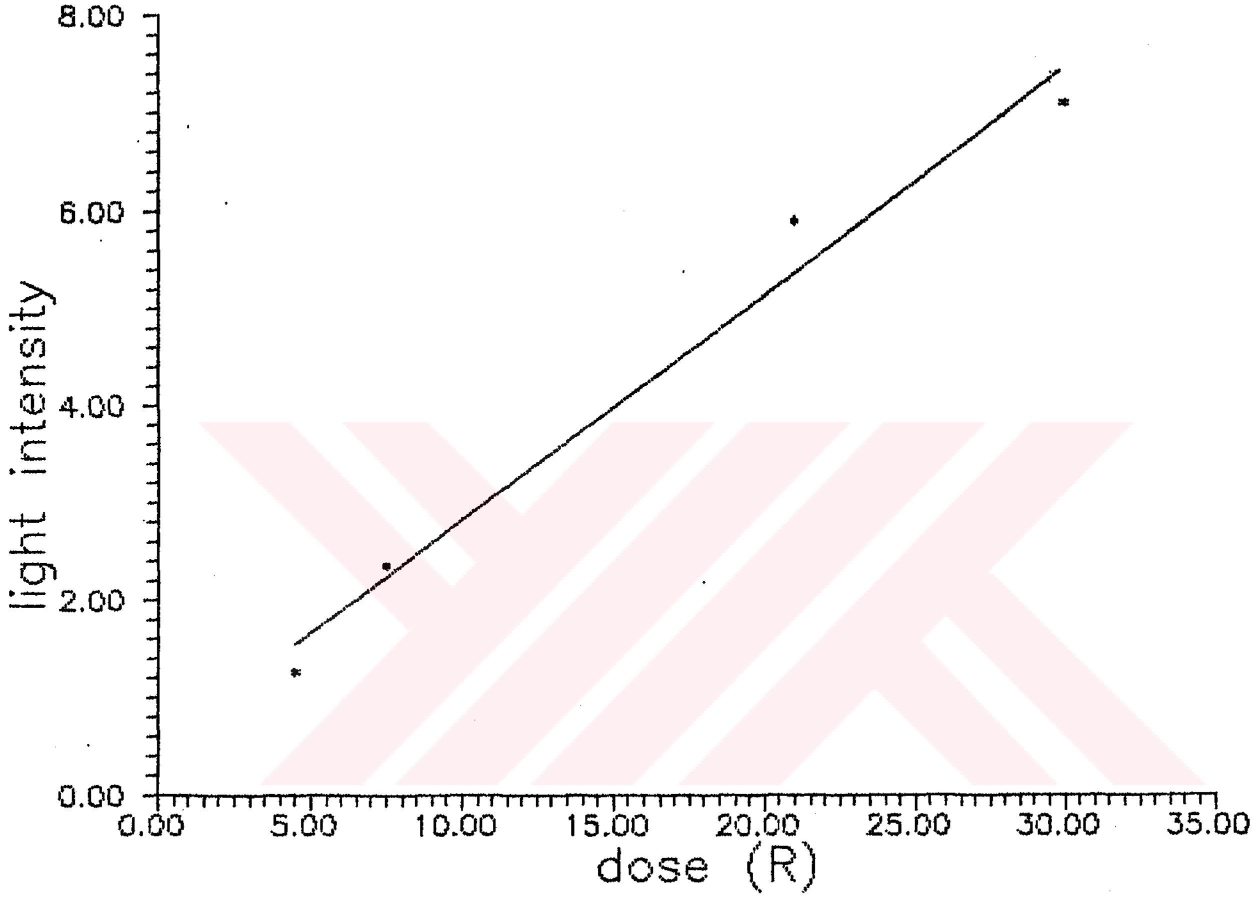

Figure 4.46 Dose response curve of $CaSO_4$:Mn,Mg - 5 teflon dosimeter.
r = 0.989

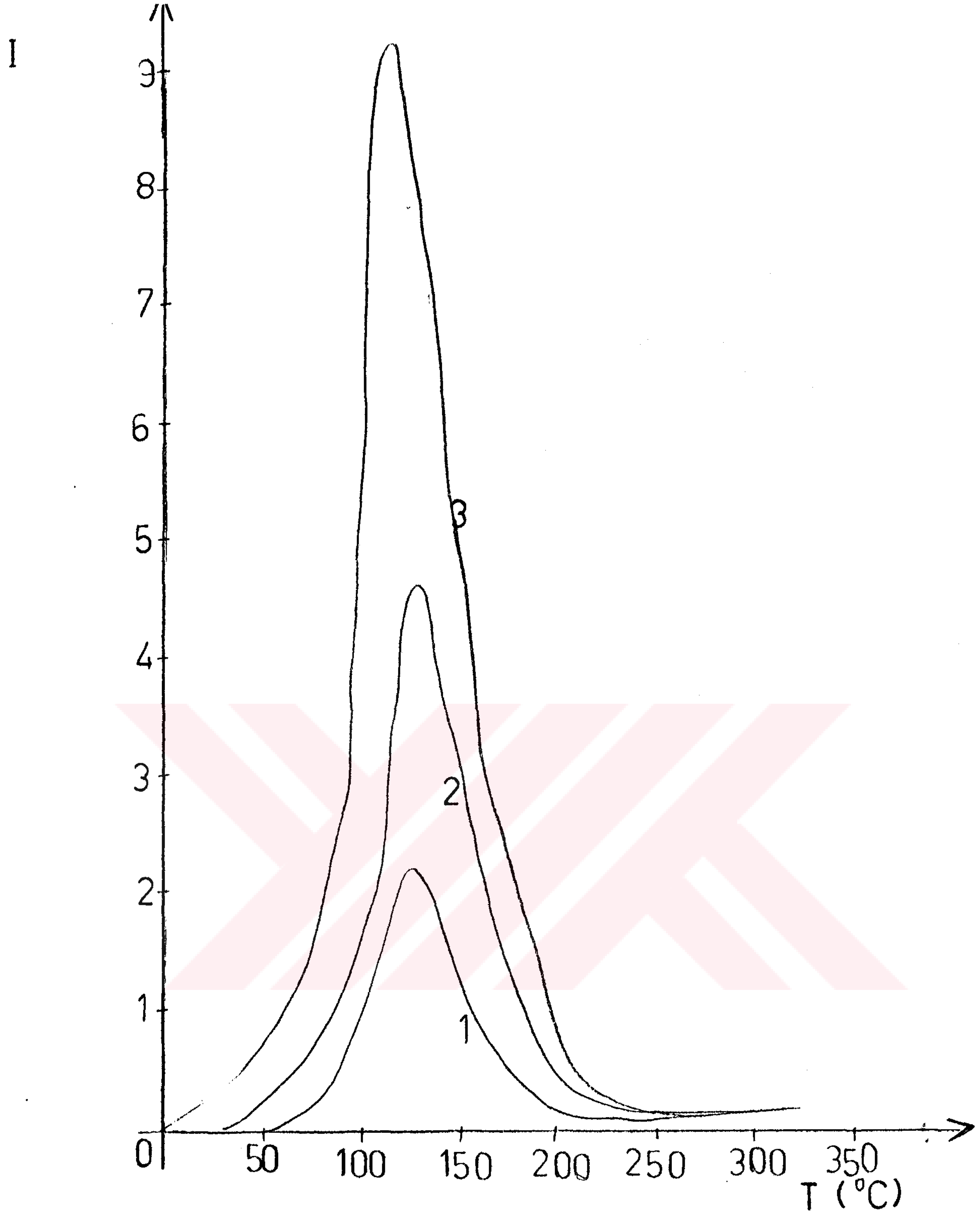

Figure 4.47 Glow curves of $CaSO_4$:Se, exposed to Sr-90 β radiation;

1) 7.5 Rad
2) 15 Rad
3) 30 Rad

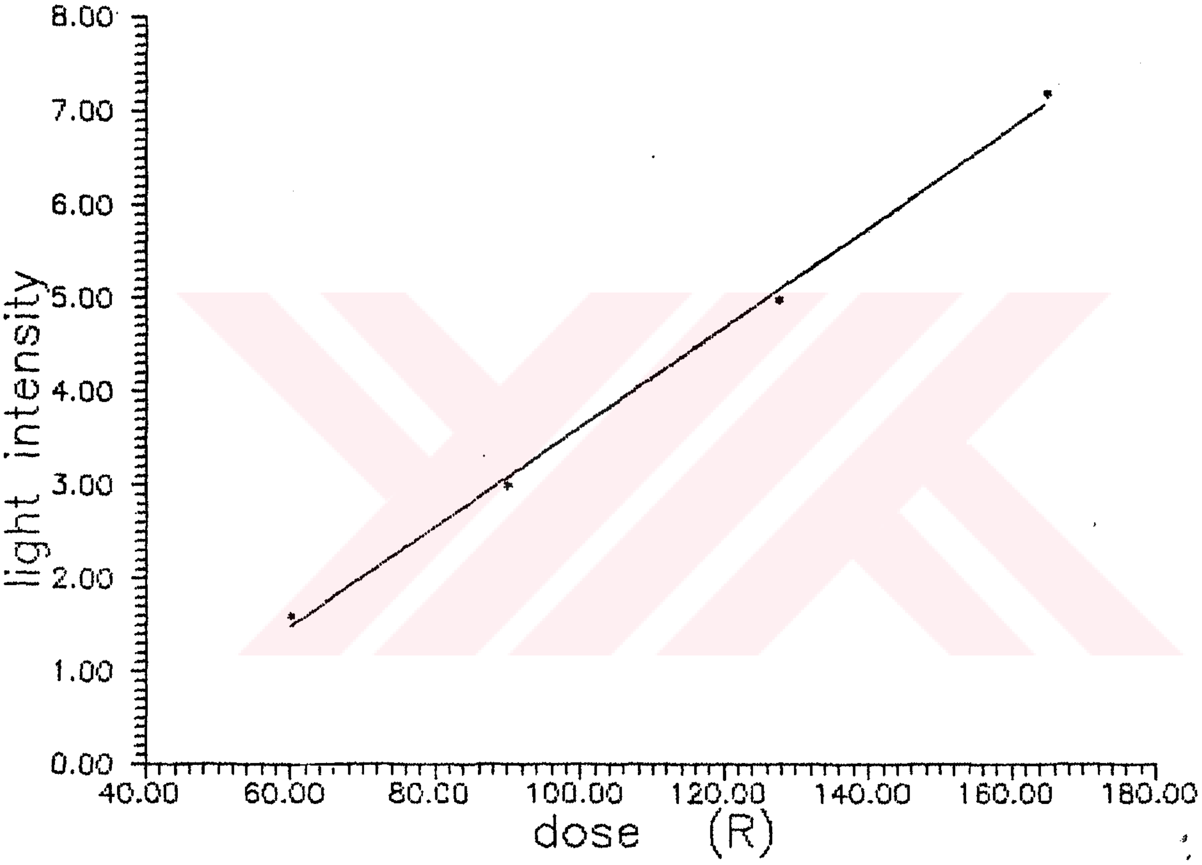

Figure 4.48 Dose response curve of $CaSO_4$:Se teflon dosimeter.
r = 0.999

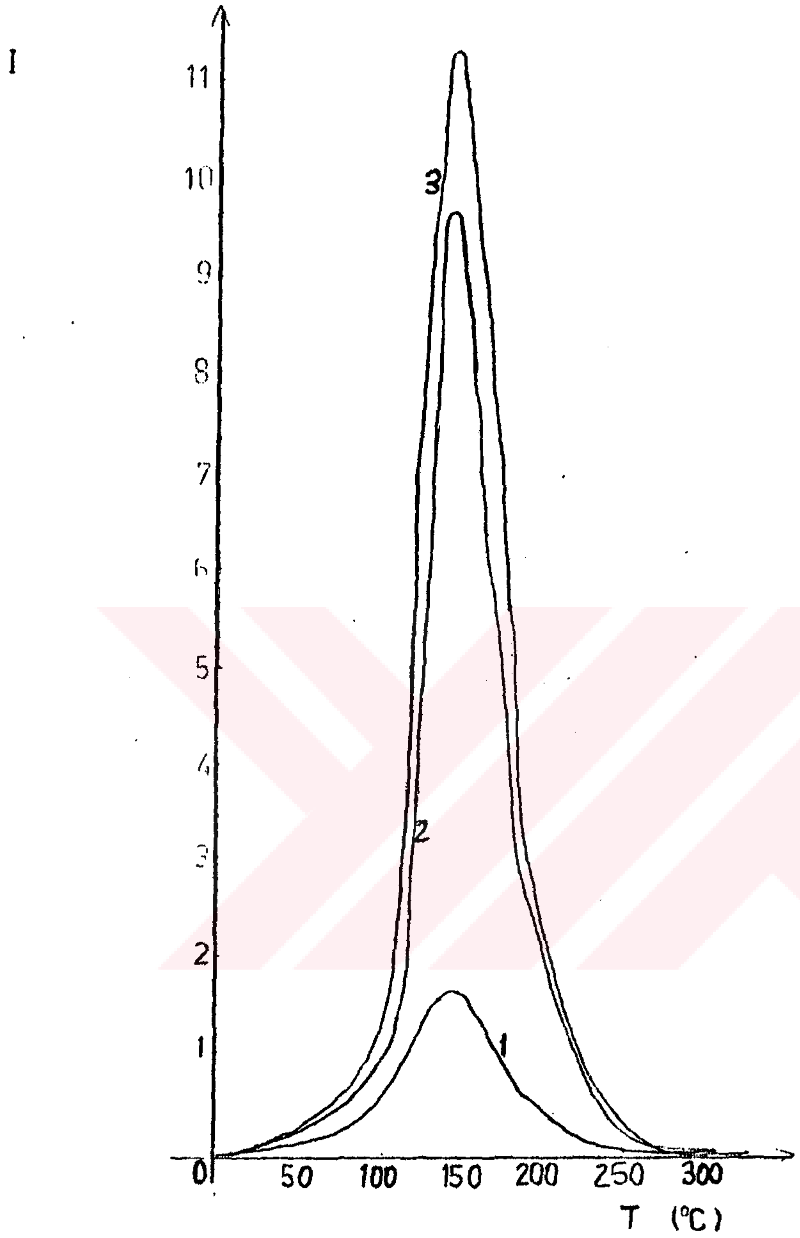

Figure 4.49 Glow curves of $CaSO_4$:Zn, exposed to Sr-90 β radiation;

1) 7.5 Rad
2) 22.5 Rad
3) 30 Rad

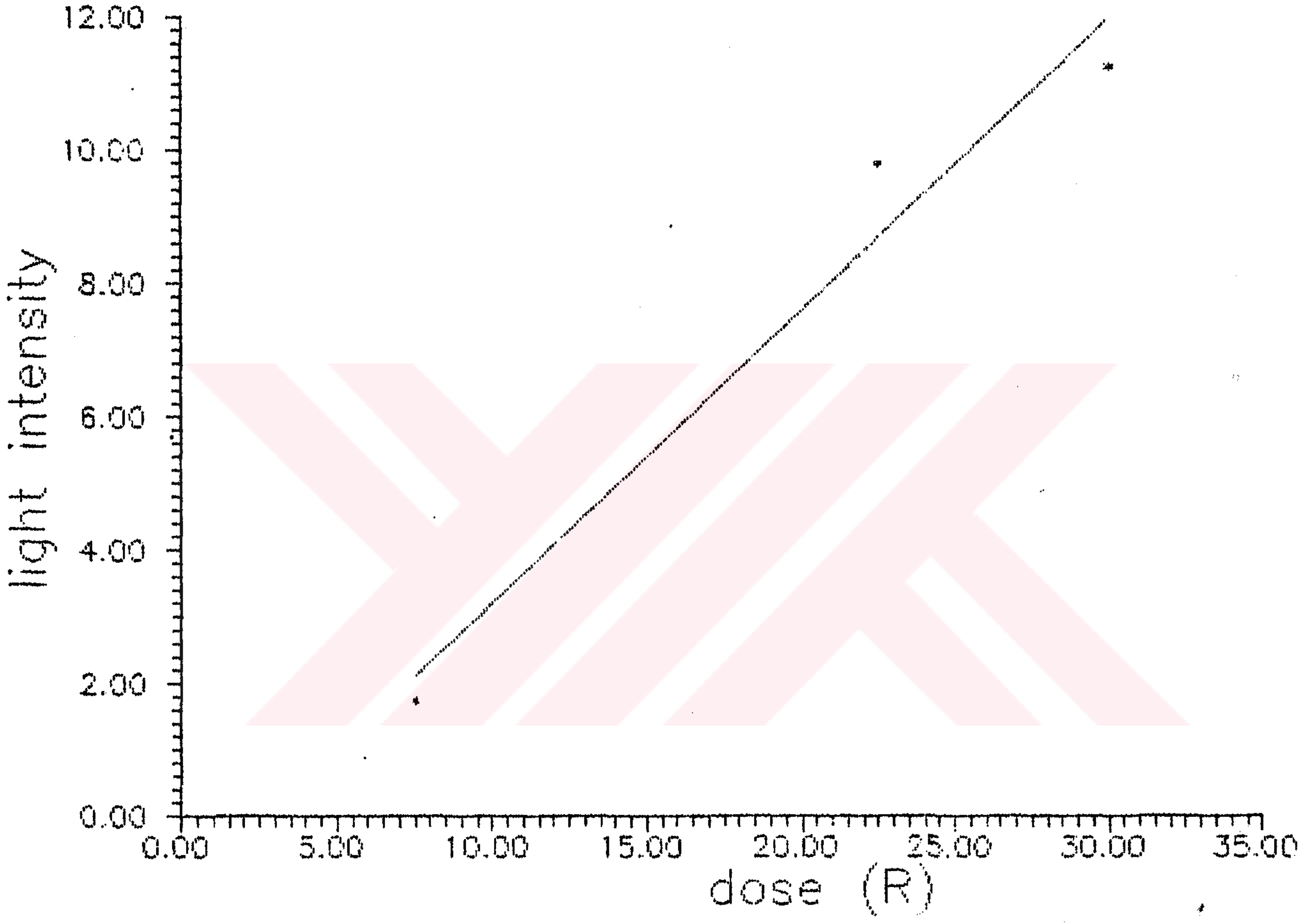

Figure 4.50 Dose response curve of CaSO$_4$:Zn teflon dosimeter.
r = 0.998

CHAPTER 5

RESULTS AND DISCUSSION

In general thermoluminescence phosphors are produced by using crystal growth methods. Some naturally found materials have thermoluminescence properties. It is possible to control the impurity content in artifically grown materials.

The materials obtained in this work have good proportionality, reproducibility, stability but have different values of sensitivities in thermoluminescence dosimetric characteristics.

As seen from Table 13, the sensitivity is affected by the type of impurity and its percentage in $CaSO_4$ phosphor. The best sensitivity which is defined as the proportion of the area under the glow curve to the mass of the sample, is found for the sample having % 1 Mg + % 1 Mn. From the list it is seen that the most effective impurity material is Mg, and Se is the next. When Mn is added to the sample, the sensitivity of the phosphor is increased.

As a result, all the samples can be used as dosimetric material and $CaSO_4$:(Mn+Mg) is found to be the most appropriate one.

TABLE 5.1 Sensitivity of the dosimeters

Sample	Sensitivity
$CaSO_4$:Mg - 1	2.355
$CaSO_4$:Mg - 2	0.64
$CaSO_4$:Mg - 3	4.545
$CaSO_4$:Mg - 4	4.166
$CaSO_4$:Mn - 4	2.445
$CaSO_4$:Mn,Mg - 2	1.33
$CaSO_4$:Mn,Mg - 4	6.159
$CaSO_4$:Mn,Mg - 5	3.030

REFERENCES

1. A.F. McKınlay
"Thermoluminescence Dosimetry" National Radiological Proction Board, Harwell 1981

2. Ata Selçuk "$Li_2B_4O_7$,LiF ve CaF_2 Kristallerinin Thermoluminesans Dosimetrik Özellikleri, Kinetik Sabitleri Tayin Methodları" Ph.D. BDMM Akademisi, Balıkesir 1982

3. Klavs Becker, "Solid State Dosimetry" 1973

4. H.Pick, "Optical Properties of Solid" 1972

5. F.H. Atrix, "Luminescence Dosimetry" Proc. 4th. Int Conf on "Luminescence Dosimetry 1974

6. Gerhart Friedlander, at all, "Nuclear and Radiochemistry" 1981

7. J.F. Randall and M.H.F.Wilkins 1945

8. R. Chen and S.A.A.Wiher 1970

9. F.Seitz Trans. Faraday Soc. 35 (1939) 79

10. F.Urbah 139 (1930) 363

11. B.BJarngard (1963)