

EFFECTS OF VERY HIGH DOSE RATES ON THE
RESPONSE OF LiF THERMOLUMINESCENT DOSIMETERS

by 817

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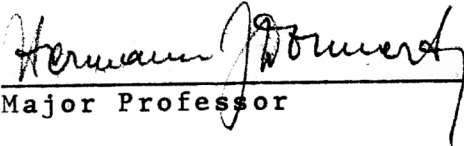
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1. INTRODUCTION

Radiation dose is the term used for the actual radiation energy absorbed per unit mass of an absorbing medium. It is one of the physical quantities, like temperature, pressure, etc., that is measured indirectly. It is not the radiation dose itself but it is the effect of radiation dose on certain substances which is measured to assess the amount of total dose. Accordingly the definitions of the units of radiation dose are also based upon the effects of radiation dose, like for example the unit of exposure dose, 1 roentgen, is defined as "the quantity of gamma radiation which will produce, by ionization, one electrostatic unit of electricity of either sign in 1 cm³ of dry air, measured at standard conditions of temperature and pressure", and the unit of absorbed dose in a material, 1 rad, is defined as the "absorbed energy of 100 erg per gram of material."

The art of measurement of radiation dose is known as radiation dosimetry. The need for appropriate dosimetry was recognized soon after the discovery of ionizing radiation. The radiation hazard involved in the use of ionizing radiation and its controlled use in biology, industry, medicine, research and military applications required accurate measurement of the radiation energy absorbed. Various dosimetry systems utilizing various effects of ionizing radiation on materials were developed. Early dosimetry devices consisted of ionization chambers coupled with an electrometer and photographic emulsions⁽¹⁾. Then "chemical dosimetry" was developed during the years 1948 -

1958. "Solid-state dosimetry" has been developed quite recently exploiting the following effects of radiation in solids.

- (a) radiation-induced coloration or decoloration;
- (b) radiation photo luminescence;
- (c) radiation thermoluminescence;
- (d) luminescence degradation;
- (e) miscellaneous (for instance: changes of conductivity, exo-electron emission, blackening of photographic emulsions, etc.)

Thermoluminescent dosimeters, utilizing the effect (c) mentioned above, have been the subject of great interest because of their exceptional properties such as:

1. wide range (10^{-4} R upto 10^8 R)
2. small size (needle shape dosimeters, 0.9 mm diameter, 6.0 mm long, are being manufactured⁽²⁾)
3. sensitivity to all kinds of ionizing radiation,
4. accuracy
5. re-usability
6. convenient and quick readout.

They are being used with advantages in the fields of personnel dosimetry, health physics, and research dosimetry. They are, however, coupled with some drawbacks which are yet to be overcome. Two main drawbacks are:

1. thermoluminescent phosphors are extremely sensitive to minute impurity concentrations and thus it is difficult to reproduce the phosphors and obtain the same properties,

2. dosimeter reading devices are quite complicated and expensive.

A long list of thermoluminescent phosphors suggested for use in dosimetry is given in ref. (1). Each phosphor is associated with its own advantages and disadvantages. Dosimetry using lithium fluoride (LiF) was first developed by Bräunlich and Scharmann⁽³⁾ and Cameron et al.⁽⁴⁾. The specific merits of LiF compared to other phosphors are its low dependence on photon energy, extremely good storage properties and its tissue equivalence which makes it extremely useful in personnel dosimetry. Two types of LiF phosphors, TLD-100 and TLD-700, have been developed specifically for use in dosimetry by Harshaw Chemical Company, Cleveland, Ohio. TLD-100 contains ^6Li and, hence, is quite sensitive also to thermal neutrons; TLD-700 is made from pure ^7Li and is rather insensitive to thermal neutrons.

All the dosimetry techniques are empirical in nature in the sense that they require calibration in the laboratory with some standard radiation source before they can be actually put to use. Several factors such as temperature, type of radiation, dose rate, etc., may change when the dosimeters are put to practical use, and it is of importance to know the effects of all these factors individually for obtaining meaningful results. It is the purpose of this research mainly to study the effect of high dose rates on the response of LiF dosimeters. This effect has been studied by several investigators in the low range of dose rates. But as the scope of application of LiF dosimetry is being increased looking at its potentialities it is now

important to know the effect of dose rate even in the high dose rate range. Importance of such studies also lies in the fact that the mechanism of thermoluminescence of LiF is still not understood and studies in various directions may give important clues towards understanding the mechanism.

2. THEORY

2.1. DEFINITION OF TERMS

2.1.1. Radiation Thermoluminescence:

Certain types of insulating crystals store energy when they are irradiated by nuclear radiation. They release the stored energy in the form of light at a later time when they are heated to higher temperatures. Thermoluminescence (TL) is the name given to such phenomenon.

2.1.2 Phosphor:

A material which exhibits TL is known as a phosphor. The basis of a phosphor is a pure insulating crystal. It is made luminescent by the addition of small proportions of impurity atoms.

2.1.3. Traps:

Structural imperfections found frequently in crystals, and crystal lattice defects form localized centers of positive or negative charge. These centers are capable of trapping electrons or holes, depending upon the charge they have accumulated. When these are set free they come down to ground level transferring their energy either in the form of light photons (luminescence) or in some other form. In case of luminescence the light emitted shows the color characteristic of the trap. These traps are therefore also known as color centers. The particular center consisting of an electron trapped at an anionic vacancy is called an

F-center, after the German name for color center: "Farbzentrum."⁽⁵⁾
 Traps are also formed by some impurity atoms.

2.1.4. Trap depth:

Traps discussed above are located in the forbidden energy gap of crystals. The energy E (Fig. 1) between the lower edge of the conduction band and the trap location is termed as the

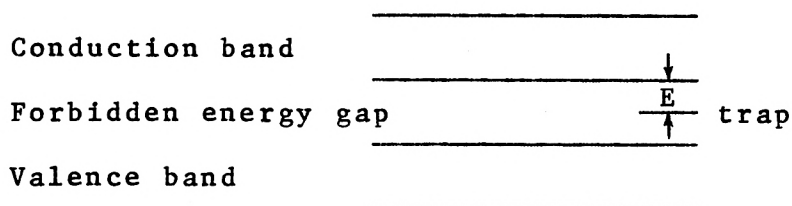


Fig. 1. Energy band model of a crystal.

trap depth because of the fact that a trapped electron requires energy at least equal to E to be freed from there.

2.1.5. Glow Curve:

The intensity of light given out from an irradiated phosphor when it is heated varies with the temperature. A plot of the light intensity given out from a phosphor against time, or against temperature if the rate of heating is constant, is known as the glow curve.

2.2. THE PROCESS OF TL

The phenomenon of TL is not completely understood even today. The varying shapes of glow curves obtained with different types of phosphors led people to suggest various models for TL,

the validity of any of which is not fully ascertained. However, there are some ideas which are generally accepted.

2.2.1. TL Process In General Terms:

Numerous traps or localized energy levels are essential in a phosphor if it is to be used for the dosimetry of ionizing radiation. Carefully fabricated high-purity crystals contain only a limited number of these traps. Dosimetry then, in principle, could be based upon the production of traps by irradiation, the concentration of which could be determined by measuring their luminescence. In practice, however, this is not feasible, because neither the efficiency nor the reproducibility of trap formation in pure salts is adequate for most dosimetric applications, and the act of measuring the luminescence bleaches them out at normal temperatures. Therefore in order to have sufficient number of traps available in a crystal it is doped with some impurity atoms. For a description of various types of traps and their characteristics the reader is referred to the review article by Schulman⁽⁵⁾.

The effect of radiation on phosphors is then to excite electrons from the filled valence band to the empty conduction band, from where they can and do fall into the traps located in the forbidden energy gap (refer Fig. 1). How long the electrons remain trapped depends upon the trap depth. In semiconductors, for example, the traps are so shallow that trapped electrons are excited back to the conduction band by thermal motion at the room temperature. In phosphors the traps are much

deeper and the probability of an electron going back to conduction band at the room temperature is negligibly small. How well a particular phosphor is suitable for dosimetry purposes is actually determined in part by the depth of traps it contains.

In a phosphor the number of trapped electrons is proportional to the radiation dose received by the phosphor. To determine this radiation dose the phosphor is heated. Receiving the thermal energy the electrons are excited back to the conduction band from where they come down to ground levels in the valence band transferring their energy in the form of light photons. The total light given out is proportional to the number of electrons released from the traps and gives a measure of the radiation dose when all or a well defined fraction of all the trapped electrons are released.

This represents an over-simplified picture of TL. The complicated shapes of glow curves obtained with different phosphors suggest that the mechanisms of excitation of electrons, their trapping and their release are more involved. They might even be different for different phosphors. A brief description of the theories developed so far to explain the glow curve and some other characteristics of TL is given in the following sections.

2.2.2. Model Suggested By Randall and Wilkins⁽⁶⁾

Randall and Wilkins developed for the first time a mathematical equation for the glow curve. One of the main assumptions they make for developing the model is that the electrons released from the traps have negligible probability of getting trapped again. The rate of release of electrons and hence the

intensity of light given out by the crystal is then proportional only to the concentration of trapped electrons. The kinetic order of the readout process is thus assumed to be unity.

According to this model several peaks appear in the glow curve each corresponding to a particular trap depth.

2.2.3. Model Developed by Bonfiglioli et al.⁽⁷⁾:

Hill and Schwed⁽⁸⁾ found experimentally that the activation energy (defined earlier as trap depth) is unique even though several peaks appear in the glow curve of NaCl. Taking this for granted Bonfiglioli et al.⁽⁷⁾ develop a model for TL which explains the appearance of several glow peaks corresponding to a unique activation energy. They assume that the electrons released from the traps go to populate the conduction band. They are then trapped in some kind of "Luminophor Centers," and each glow peak corresponds to a particular type of Luminophor centers. The intensity of light given out by the crystal then depends not only upon the concentration of trapped electrons but also upon the concentration of Luminophor centers, thus making the readout process second order kinetically.

2.2.4. Model Developed By Cameron et al.⁽⁹⁾:

Apart from exciting electrons to the conduction band, which later fall into traps, radiation quanta are supposed also to create new traps in the phosphor, thus increasing the total number of empty traps in it. These 'created' traps are, however, supposed to bleach out at high temperatures of annealing. For developing some mathematical equations the following assumptions are made:

(1) a fixed number of initial unfilled traps, (2) creation of traps by irradiation with a proportionality constant, (3) filling of traps by irradiation with a proportionality constant, and (4) a maximum number of total traps. For processes that occur during and after heating the Randall and Wilkins model is supposed to hold true.

This is the only model proposed so far which suggests some details of the irradiation process. But with the new ideas introduced here many more questions arise: whether or not the proportionality constants for the creation of traps and filling up of traps depend upon the total amount of dose given to the phosphor or upon the rate at which the dose is delivered? And if they do depend upon these factors in what way do they change with increasing dose and dose rate? The answers to such and many other questions are yet to be found.

2.2.5. Predictions of Theory for the Dose Rate Effect:

The main interest in the current investigation is to see whether or not the response of phosphors (hereafter referred to as dosimeters), i.e., the quantity of light given out by the dosimeters depends upon the rate at which they are supplied with the dose. In other words it is desired to see when two identical dosimeters are supplied with the same total amount of dose but at different dose rates, whether their response remains the same or not? For making predictions to this effect the actual processes that take place in the dosimeter during irradiation should be known. Unfortunately, details of such processes

are missing. Most of the models proposed for TL are intended only to explain the glow curve, and as such the details of irradiation mechanisms are ignored.

Generally the effect of radiation on dosimeters is assumed to be only exciting electrons from the valence band to the conduction band. Total number of electrons trapped and hence the response R of the dosimeter is then proportional to the concentration of electrons in the conduction band n and the concentration of empty traps T :

$$R \sim n \cdot T$$

If T is very large, as generally is the case, it practically remains constant with time especially at low doses. The process of irradiation at low doses then remains pseudo-first-order kinetically. At higher doses it is possible that T changes with time leading to second order kinetics for the process of irradiation. Response in such cases should decrease because a portion of the excited electrons will recombine as the probability of recombination no longer remains negligible compared with the probability of trapping.

If Cameron's model is assumed to be true then several questions already posed in the preceding section arise and nothing about the dose rate dependence of response can be predicted.

2.2.6. Dose Rate Dependence Vs. Reaction Kinetics:

A theory developed by Donnert^{(10)*} links up the dose rate effect with the kinetic order of the irradiation process. It is shown that:

- (1) The response of any dosimetry system for ionizing radiations will exhibit an observable dependence on dose-rate if and only if its reaction mechanism involves any processes which are not kinetically of first order.
- (2) An existing dose-rate effect for a dosimetry system is necessarily a dose-dependent phenomenon; the response of these dosimetry systems must, therefore, be considered as a function of dose and dose-rate, two independent parameters.

2.2.7. Mathematical Model for the Dose Rate Effect:

Donnert's theory leads to a mathematical expression for the response which may be tested experimentally. Since the functional relationship between the response R of a dosimeter to a dose D delivered at a dose-rate θ is not known, we choose to express the response R by an n th degree polynomial in D and θ :

$$R = R_{00} + R_{10}D + R_{01}\theta + R_{11}D\theta + R_{20}D^2 + R_{02}\theta^2 + \dots \\ + R_{on}\theta^n \quad (2.2.7-1)$$

where R_{ij} ($i = 0, 1, \dots, n; j = 0, 1, \dots, n$) are the expansion coefficients independent of D and θ . In an actual experiment

*The reference is not available readily because it is classified. The theory is outlined in Appendix A.

it is difficult to obtain significant data for determining large number of expansion coefficients, therefore the expression should be restricted to a small number of terms:

$$R = R_{00} + R_{10}D + R_{01}\theta + R_{11}D\theta \quad (2.2.7-2)$$

The reason why only the interaction term ($R_{11}D\theta$) has been included out of the three second order terms is evident from the Appendix A. Actually according to Donnert's theory if the response is dose-rate dependent, which is ascertained from a non-zero R_{01} coefficient, then the R_{11} coefficient should also be non-zero.

3. THERMOLUMINESCENCE DOSIMETRY

3.1. THERMOLUMINESCENT RESPONSE OF LiF:

Five peaks appear in the glow curve obtained from LiF. A typical glow curve obtained by Zimmerman et al.⁽¹¹⁾ is reproduced in Fig. 2. Figure also shows the temperatures at which the various peaks appear in the glow curve. It has been found that peaks corresponding to low temperatures decay out fast even at room temperature. The approximate half lives of the peaks 1 through 5 are found to be 5 min., 10 hr., 1/2 yr., 7 yr., and 80 yr., respectively.

True shape of the glow curve is obtained only when the rate of temperature rise of the dosimeter is constant. The positions of the glow peaks on the temperature scale, and their heights also depend upon the heating rate used. The heating rate used in EG and G model TL-3B reader (see section 4.2 for detailed description) was found to be very close to linear. The glow curve obtained with the TL-21 dosimeters is reproduced in Fig. 3. The peak 1 indicated in Fig. 2 does not appear in Fig. 3. It probably decayed out before the dosimeter was read. Otherwise the close resemblance of the glow curve of TL-21 dosimeter with the typical glow curve of Fig. 2 is remarkable.

Peak 5 which is the largest and most stable peak is generally used for dosimetry purposes.

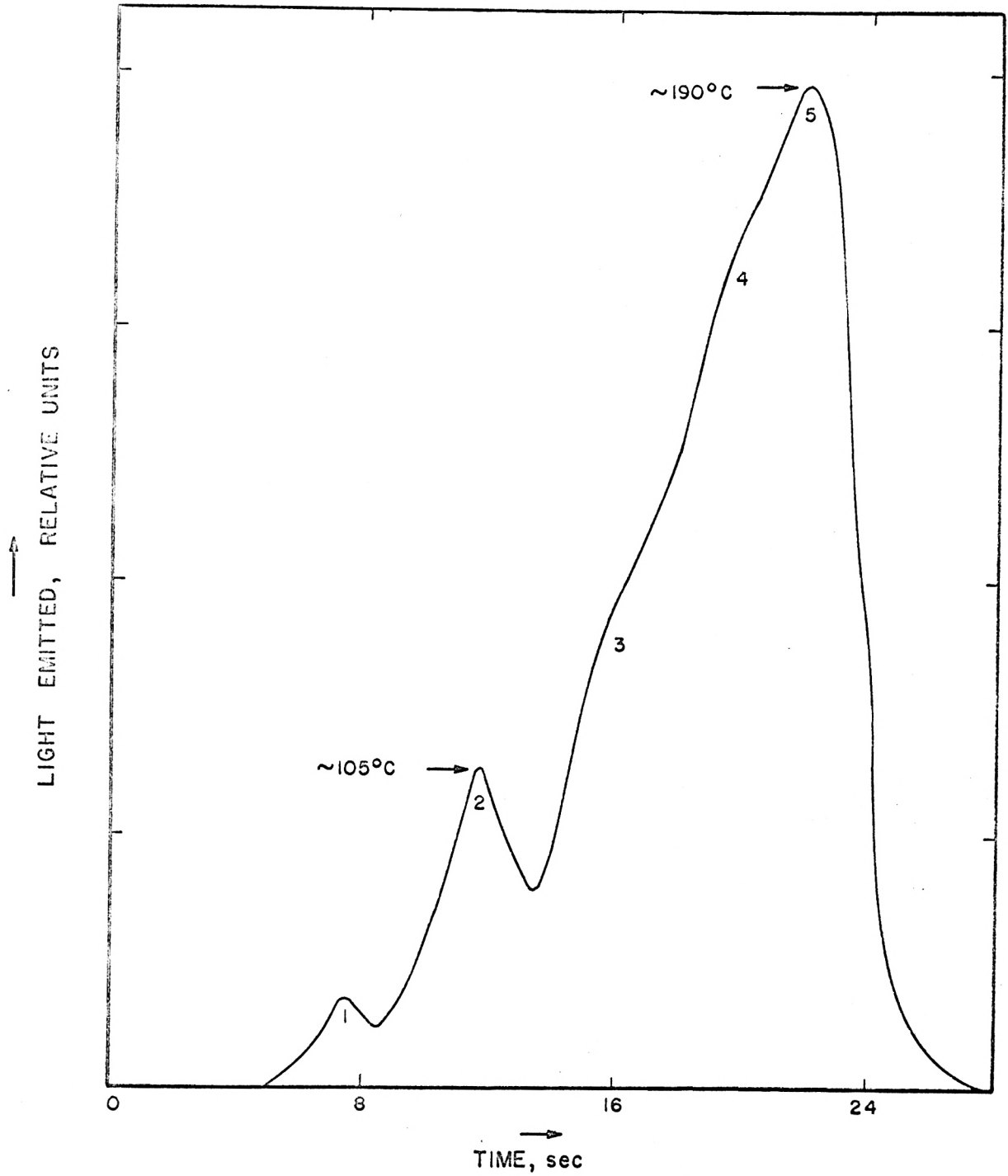


Fig. 2. Typical glow curve of TLD-100 after annealing 1 hr at 400°C and irradiation to 100 R (reproduced from ref. 11).

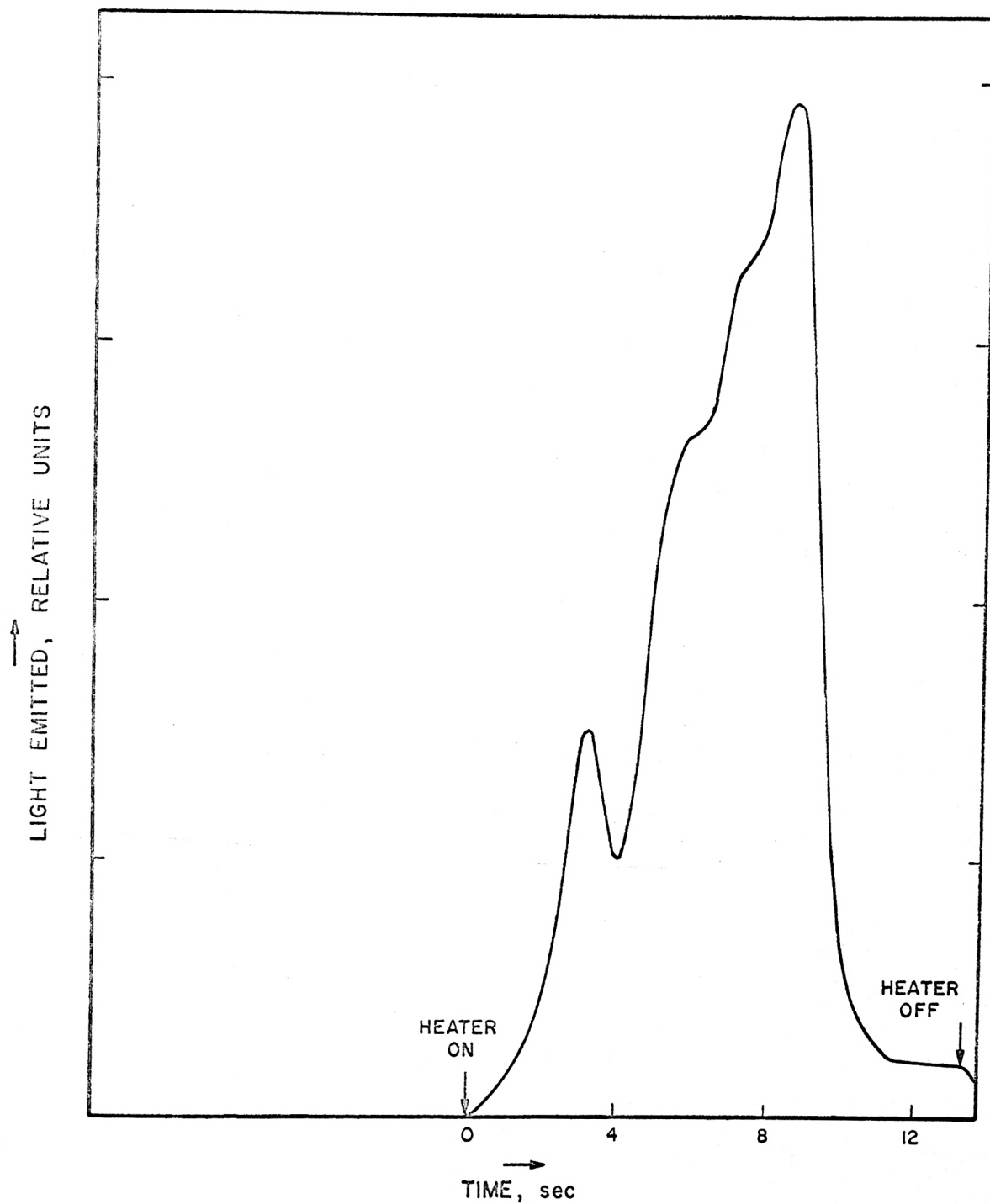


Fig. 3. Typical glow curve obtained on EG and G model TL-3B reader from EG and G model TL-2 dosimeters irradiated to about 600 R.

3.2. FACTORS AFFECTING THE DOSIMETER RESPONSE:

Although much research work has been and is being done on TL dosimetry, standard methods of dosimetry have not yet been established. Some of the factors that affect the response critically along with the reasons for selecting particular methods in this research are discussed in the following sections.

3.2.1. Type of Radiation:

Morehead and Daniels⁽¹²⁾ determined the energy required to produce an F-center in LiF by different types of radiation at different amounts of doses already given to the phosphor and report the results as in Table 1. From their results it is easily

Table 1. Effects of Different Types of Radiation on LiF (taken from ref. 12)

Radiation	Energy required to produce an F-center (eV)		
	Initial	after 10^6 R	after 10^8 R
2 MeV alpha particles	700	700	700
2 MeV electrons	140	140	700
1 MeV gamma photons	62	160	700
Thermal neutrons	65	100	700

seen that the types of radiation has pronounced effects on the response of dosimeters, but it is difficult to find out these effects quantitatively.

Pinkerton and Laughlin⁽¹³⁾ compared the response of LiF dosimeters to gamma-rays of ^{60}Co and to electron of energies 10

MeV and 15 MeV. They report the following response ratios when the same amount of dose was supplied in all cases:

$$\frac{\text{Response to 15 MeV electrons}}{\text{Response to } ^{60}\text{Co gamma rays}} = 0.893 \pm 0.018$$

$$\frac{\text{Response to 10 MeV electrons}}{\text{Response to } ^{60}\text{Co gamma rays}} = 0.876 \pm 0.021$$

Some more data of this type is available in ref. (14). The response to different types of radiation differed by a constant factor, but the shapes of glow curves obtained for different heating times remained the same in all cases. This shows that the final effects of the interaction of high energy electrons with LiF are same as those in case of ^{60}Co gamma-rays, but only a little less in magnitude. It was on this basis that in this research the calibration curve obtained with ^{60}Co gamma-rays was assumed to hold good for the responses obtained with 14 MeV electrons.

3.2.2. Quantity of Phosphor Used:

Obviously, larger amounts of phosphor will store larger amounts of energy and hence will give higher response. But this again depends upon the physical arrangement of phosphor when it is heated. If it is arranged in a thick layer the layers at the top may screen the light emitted by the layers at the bottom, thus affecting the total light measured.

Correct amount of phosphor in all dosimeters is therefore essential in order to obtain the same results. Based upon this factor EG and G selects its dosimeters to yield response within $\pm 10\%$ in a particular batch of dosimeters.

3.2.3. Quantity Taken to Represent Response:

Strictly speaking the total light emitted by the dosimeter during the complete heating cycle, which is the same as the area under the glow curve, gives the response which is proportional to the radiation dose. But in practice it turns out that this is not the quantity measurable very easily. At high temperatures the heating element also emits infra-red light and it is difficult to separate this from the light emitted by the dosimeter. At low doses the main radiation induced peak is quickly followed by a non-radiation-induced tribothermoluminescent peak* and it becomes difficult to decide at which point the heating should be discontinued to avoid this peak⁽¹⁵⁾.

It has been found that when the heating cycle is reproducible the peak heights are also related to the radiation dose applied. Therefore when this is taken as a measure of dose then the problems of tribothermoluminescent peaks etc. are avoided.

3.2.4. Method of Annealing:

Necessity for annealing dosimeters before re-use arises from the fact that all the traps are not fully emptied by the small heating cycle used for reading the dosimeters out. But then the way thermal annealing is carried out has marked effects on the response of dosimeters⁽¹¹⁾. It is, therefore, quite important that a suitable annealing procedure be decided upon and adhered to to get consistent results.

*induced in loose powder by mechanical handling after the powder is reprocessed.

3.2.4(a). Annealing Methods Suggested in Literature:

For the particular type of dosimeter used in these studies (see section 4.1 for description) ref. (2), suggests a secondary annealing procedure consisting of heating the dosimeters at 80°C for 24 hrs, preceded by some standard primary annealing method which could be heating the dosimeters at 350°C or 400°C for 1 hour. Zimmerman et. al.⁽¹¹⁾ studied in detail the effects of pre-irradiation annealing on the response of LiF dosimeters and reported that annealing at 400°C for 1 hour removes all the trapped electrons and also the effects of any other previous thermal annealing procedure, and that increase in the annealing time beyond 1 hour has little effect on the dosimeter response. They also recommend an annealing procedure consisting of heating the dosimeters for 1 hr. at 400°C followed by a heating for 24 hrs. at 80°C. The 80°C annealing is supposed to reduce the low temperature peaks 1 and 2 (Fig. 2) relative to higher temperature peaks.

3.2.4(b). Experimental findings:

In the present investigation the problem of removing the low temperature peaks was solved in another way (see section 3.2.6.) and hence it was necessary to see if the 80°C annealing in the procedure recommended by Zimmerman et. al.⁽¹¹⁾ was still helpful in any way. Two batches of 8 dosimeters each; one annealed at 400°C for 1 hr., and the other annealed at 400°C for 1 hr. followed by annealing at 80°C for 24 hrs., were irradiated by ⁶⁰Co. gamma-rays to about 500 R. Later they were

read out and the standard deviations in the response values of each batch were compared. No significant difference in the standard deviations was observed. Hence the 80°C annealing was considered superfluous and was avoided.

3.2.5. Heating Rate:

The shape of the glow curve depends critically upon the heating cycle used in reading out the dosimeter. True shape of the glow curve is obtained only when the rate of temperature rise is constant. Changing the heating rate has several effects on the response. If the heating rate is increased the peaks in the glow curve appear at lower temperatures and the peak heights increase even by orders of magnitude.

Linearity of the rate of temperature rise is not important in dosimetry. Any rate which is close to being linear can be used. Results obtained will be consistent with the particular heating cycle used. But the reproducibility of the heating cycle is very important for the results to be consistent. Care should be taken to ensure this especially when the position of dosimeter in the heating pan or heating element is not fixed. The EG and G system used in this work uses a heating cycle which gives almost linear rate of temperature rise, and also the heater coil arrangement assures the reproducibility of the heating cycle.

3.2.6. Fading:

To function as a satisfactory dosimeter, a phosphor should have a TL response that is independent of the storage time at room temperature between exposure and read out.

3.2.6(a). Theoretical Background:

Phosphors containing shallow traps fade out much faster than those containing deep traps, and are not suitable for dosimetry purposes. Phosphors containing both shallow and deep traps present some problems connected with fading which must be avoided carefully. Effects of fading that may be expected in such phosphors are:

1. a reduction in the total light output due to the de-excitation of electrons trapped in shallow traps,
2. an increase in the height of the high temperature peaks due to the electrons, released from shallow traps, falling into deeper traps (assuming that the high temperature peaks are caused by deeper traps).

If the peak height of the high temperature peak is taken to give the response the first effect does not create any problem; however, positive measures must be taken to prevent the second effect from affecting the results.

3.2.6(b). Previous Investigations:

Endres⁽¹⁶⁾ studied the fading effects on the TL response of LiF over a period of 15 days. Using the total light output as the response he observed a 5% decrease in the response over the period of study.

Observing the same integrated light output of type N LiF powder Karzmark et. al.⁽¹⁵⁾ report a 10% to 20% reduction in response in 3 weeks followed by an increase towards the initial value over a period of 6 to 8 weeks.

To remove the fading effects and low temperature peaks Cameron et al.⁽⁹⁾ partially annealed the dosimeters after irradiation at 100°C for 10 min., while Dean and Larkins⁽¹⁷⁾ anneal the dosimeters approximately 1-1/2 hr. after the irradiation at 110°C for 7 min.

3.2.6(c). Experimental Findings:

Because of the inconsistencies in the results of fading studies reported in the literature and the differences in the procedures used to remove the fading effects by the previous investigators, it was decided to determine experimentally how the fading effects, if they were present, could be avoided best.

To determine whether any fading effects were present at all, 42 dosimeters were irradiated at one time to about 600 R using the radiation facility described in section 5.2. They were stored at room temperature. Batches of 6 dosimeters were read out allowing different fading times over a period of 48 hours. The results are presented in Fig. 4. The fluctuations in the response values of different batches and the large error bars indicated that possibly fading did effect the response.

The procedure used by Dean and Larkins to remove the fading effects appeared more logical. From the typical glow curve in Fig. 2, it was seen that the distinct low temperature peak appeared at 105°C. Annealing at 110°C, therefore, was assumed to be effective in removing this peak. To determine whether in removing this peak this procedure does help the fading effects, 42 dosimeters were irradiated at one time to about 600 R. They

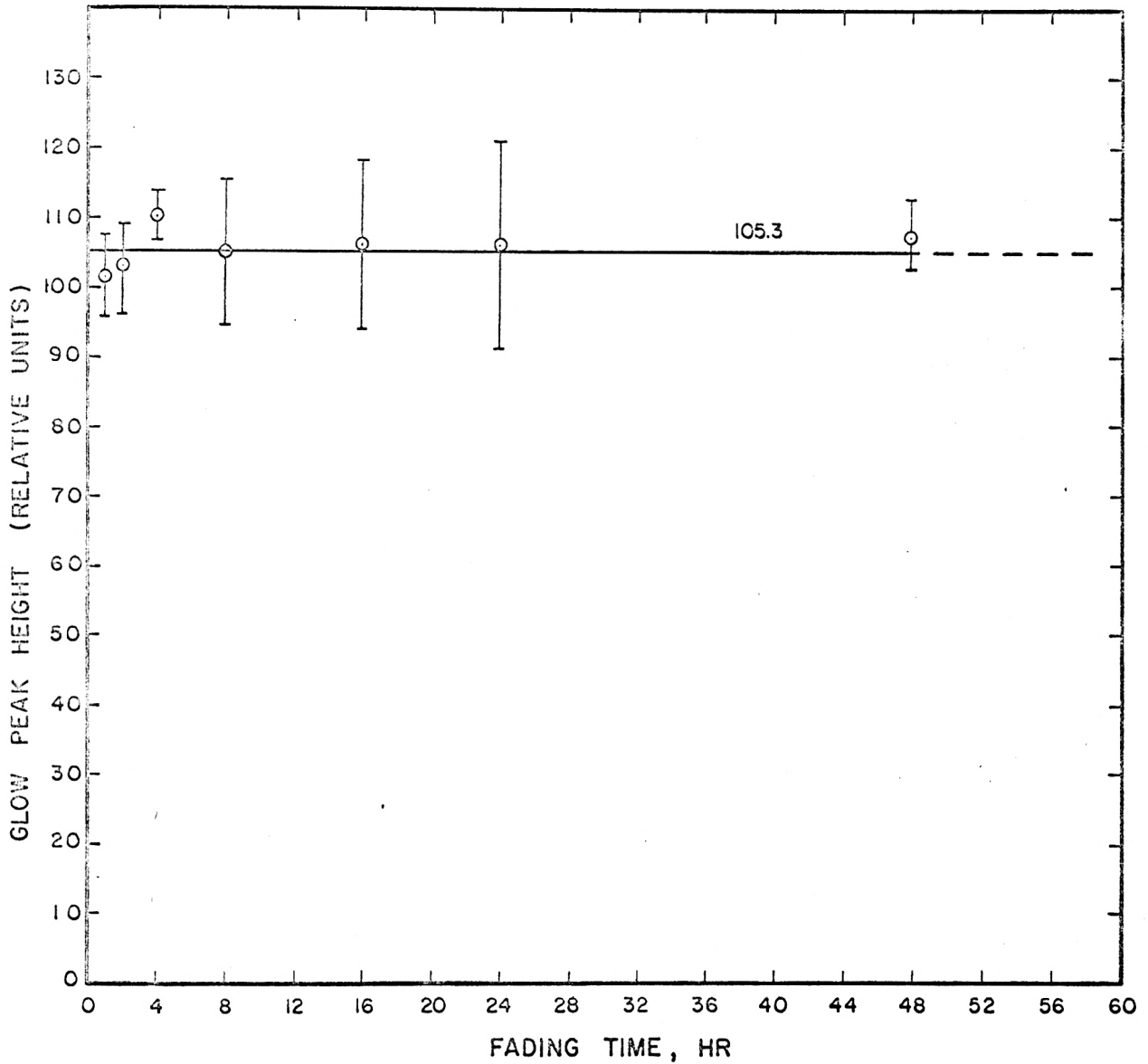


Fig. 4. Changes in the dosimeter (EG and G model TL-21) response with increasing fading time.

were all annealed at 110°C for 7 min and stored at room temperature. Batches of 6 dosimeters were read out over a period of 48 hours allowing different fading times, after the partial annealing. The results are presented in Fig. 5. The comparatively smaller fluctuations in response and the smaller standard deviations in this case indicate that the partial annealing did help in removing the fading effects. It was also found experimentally that small errors in temperature or duration of annealing did not affect the response and hence exact reproduction of the partial annealing cycle was found to be not critical.

Partial annealing also removed the low temperature peaks. A glow curve obtained after partial annealing is reproduced in Fig. 6.

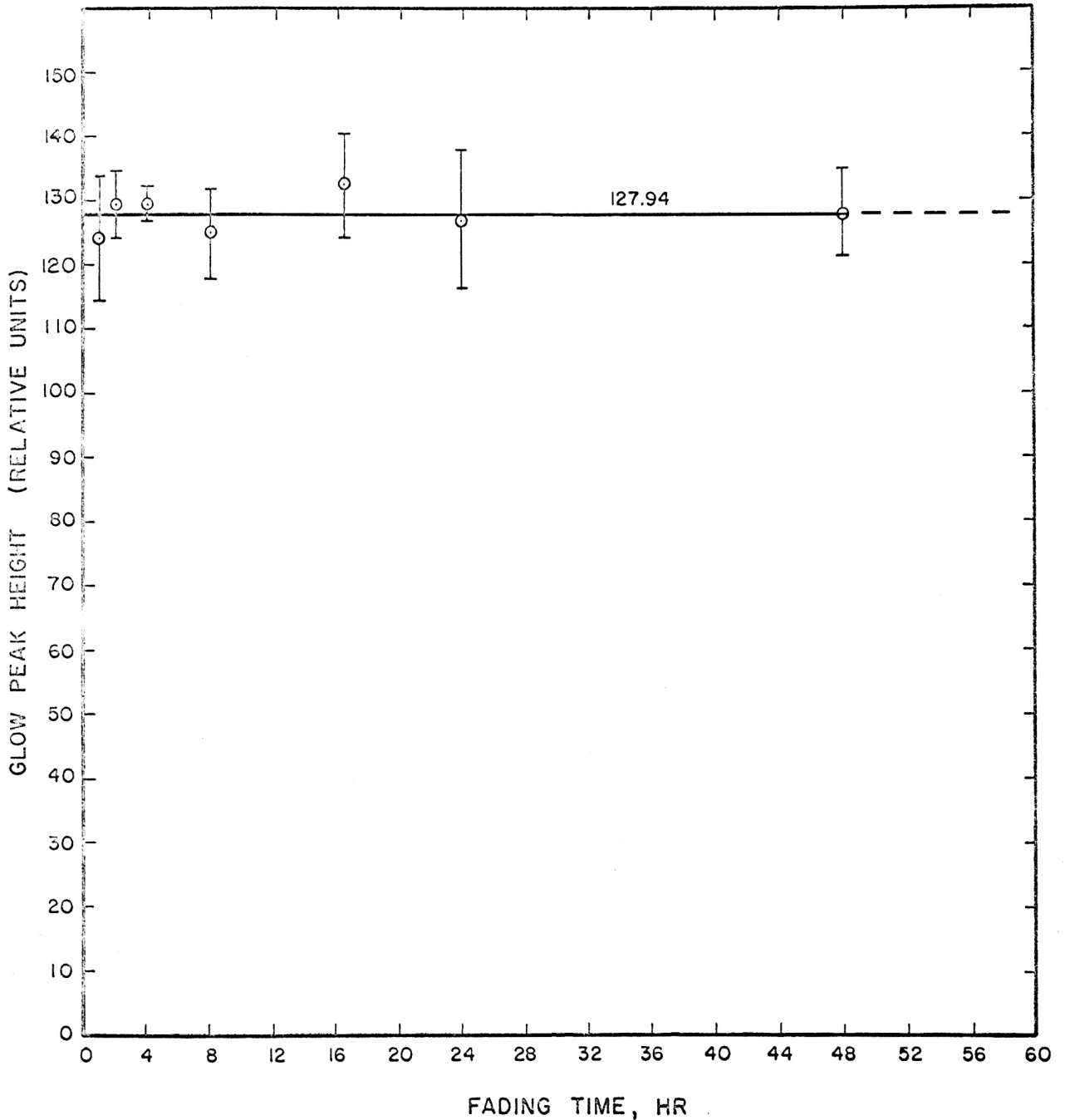


Fig. 5. Changes in TL dosimeter (EG and G model TL-21) response. Dosimeters were partially annealed after irradiation at 110°C for 7 min.

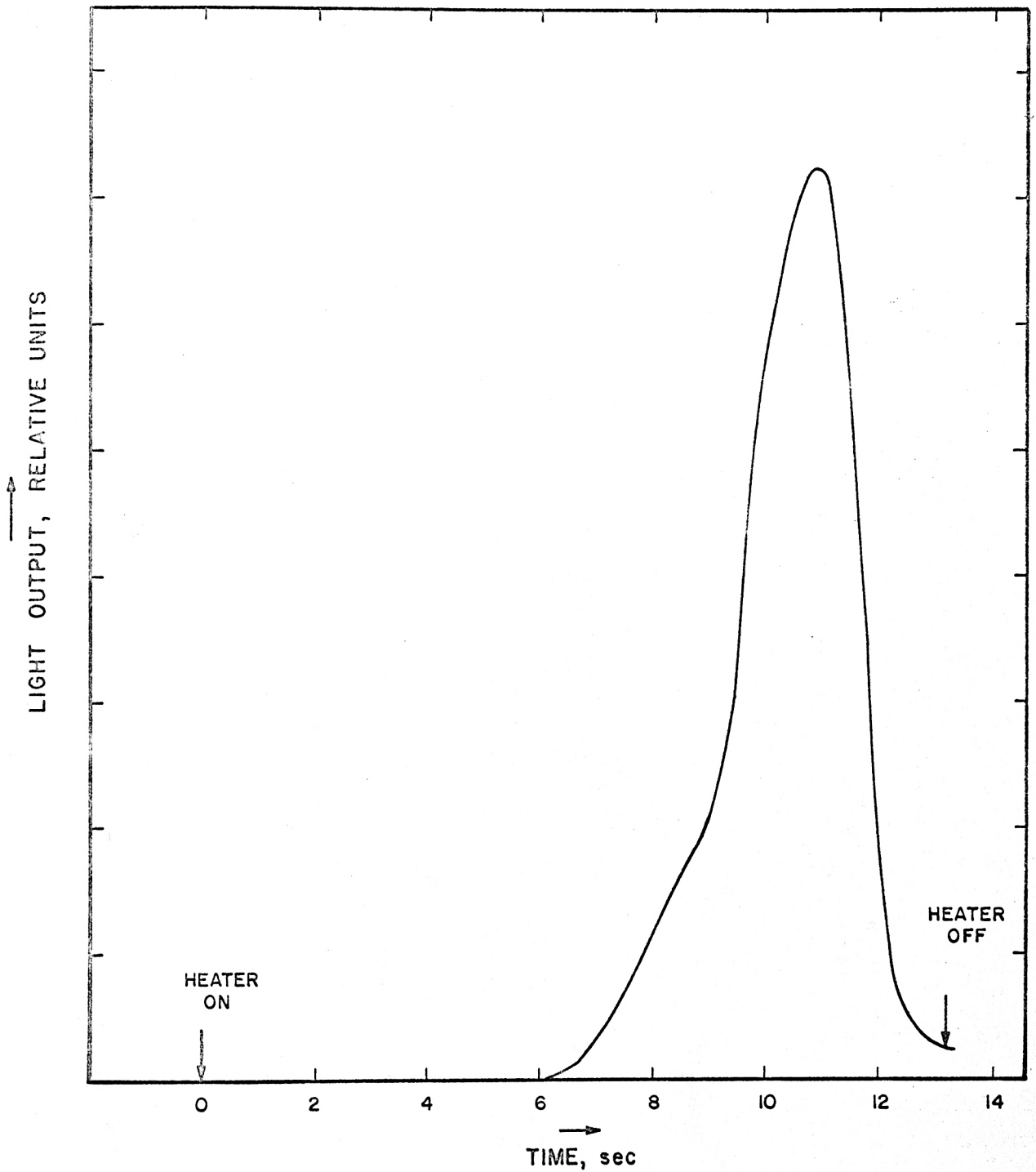


Fig. 6. Glow curve obtained on EG and G model TL-3B reader from EG and G model TL-21 dosimeter. Dosimeters were partially annealed at 110°C for 7 min. after irradiation.

4. DESCRIPTION OF APPARATUS

4.1. DOSIMETERS.

EG and G model TL-21 "LiF Miniature Dosimeters" were used in these studies (See Fig. 11). The dosimeters consist of about 10 mg of LiF phosphor vacuum sealed in a glass capillary. The size of the dosimeters is 1.4 mm in diameter and 12 mm in length. The type of phosphor used was natural LiF, containing 7.42% ^6Li and 92.58% ^7Li , obtained from Harshaw Chemical Company as TLD-100 brand.

Useful range of the dosimeters was from 10 mR to 10^5R . Dosimeters were supposed to give a response linear with respect to dose upto about 1000 R. Response in the higher dose range was reproducible, but not linear. Pre-calibration was required to use the dosimeters in the higher dose range. Response was dependent upon the energy of the radiation, but was independent of ordinary temperature changes.

Reproducibility of the dosimeters was $\pm 3\%$ above a dose of 1 R and $\pm 20\%$ at a dose of 10 mR. Due to small fluctuations in the weight of the phosphor, grain size, geometry of ampule, and phosphor sensitivity, the response of the dosimeters from a batch selected by the manufacturers were supposed to be within $\pm 10\%$ of the mean.

4.2. READER UNIT:

EG and G model TL-3B reader (Fig. 7) was used to read out the dosimeters.

Basically the operation of the reader unit was to heat up the dosimeter, supply the light emitted to a photo multiplier tube, use the output signal of PM tube to drive a chart recorder and thus obtain a glow curve.

A block diagram of circuitry of the reader unit is shown in Fig. 9. The regulated current supply fed the read head adapter (described below) in which the dosimeter was positioned. Light emitted by the dosimeter was reflected towards the PM tube. The output of the PM tube was given to a pen-servomechanism in the recorder unit via an automatic ranging circuit. Initially PM tube was set to its maximum sensitivity, which gave the lowest range on the recorder. When the light output increased beyond this range and the pen reached full scale on the recorder, the automatic ranging circuit lowered the reader sensitivity by a factor of 10. In this way readings from the lowest full scale range to of 50 mR to the highest full scale range of 500 R were possible. The control logic sequenced the operations of the recorder, detector heating cycle, automatic ranging circuits and the status indicator to provide a chart record of the detector light emission. Status indicator gave the full range of the recorder.

A standard C-14 source, whose light output was equivalent to 340 mR with TL-32A reference dosimeter, was provided for the

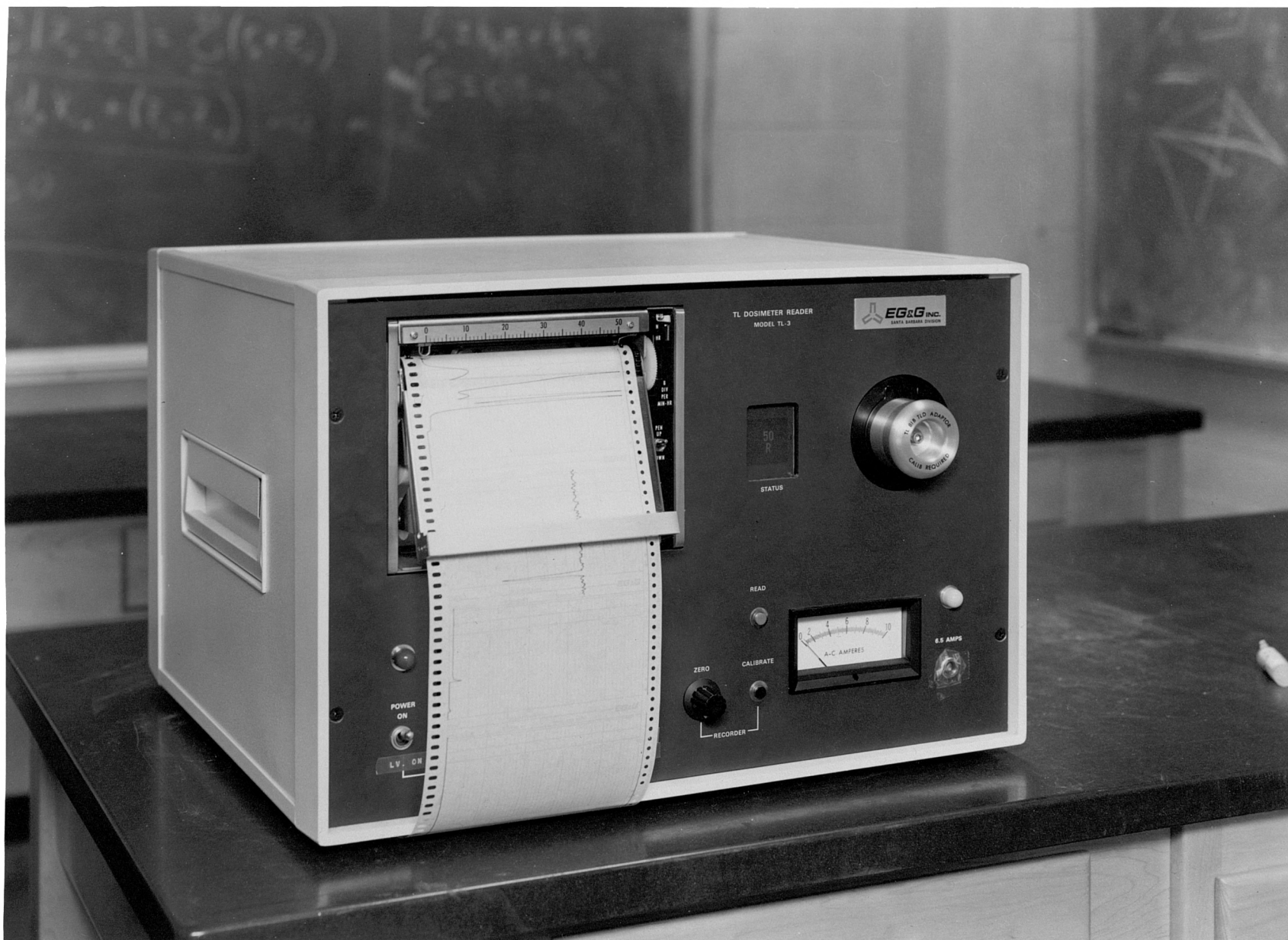


Fig. 7. A view of the EG and G model TL-3B thermoluminescent dosimeter reader.

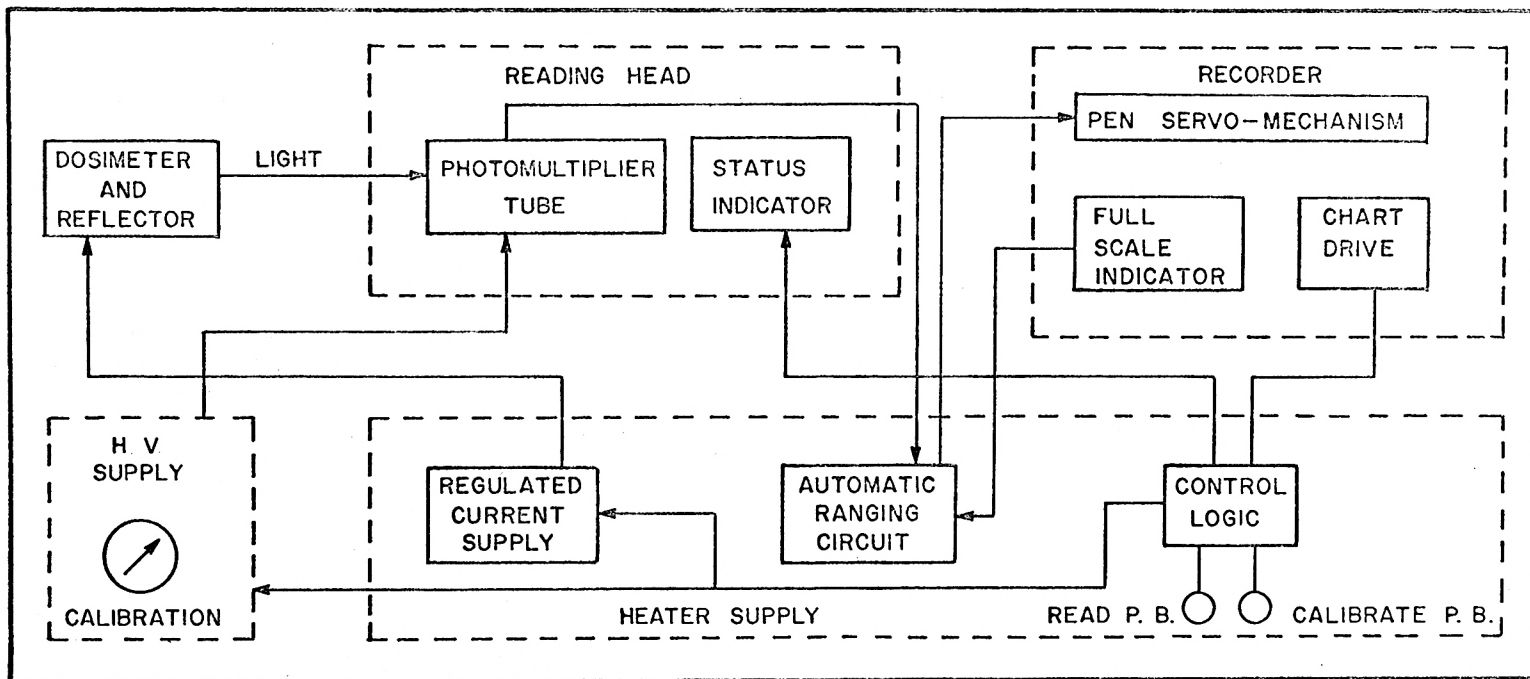


Fig. 8. A block diagram of the EG and G model TL-3B reader.

calibration of the reader unit. The calibration was accomplished by changing the high voltage across the PM tube. The normal useful range of the reader was 5 mR to 5 kR with the standard dosimeter, but it could be extended down to 0.5 mR or upto 50 kR by respectively increasing or decreasing the gain of the PM tube.

A typical chart record obtained on this reader with TL-21 dosimeters was reproduced in Fig. 9. Heater turned on when the pen reached the short line at the base near the middle of the chart and turned off at the end of the chart. Each time the range switch operated, the event marker pen produced a vertical "pip" in the continuous line at the top of the chart. Thus the final range of the recorder could be determined by counting the number of "pips", besides observing it on the status indicator.

EG and G Model TL-81B read head adapter (Fig. 10) was used to hold and heat up the model TL-21 dosimeters in the reader unit. It consisted of a heating coil, and a shunt resistance. It received a constant current of 6.5 A from the regulated current supply. The shunt resistance could be adjusted such that a correct amount of current, which would produce the glow peak on the chart in the middle of the three lines to the left of the chart (see Fig. 9), flew through the heating coil.

4.3. GAMMA-CELL:

Gamma-cell-220 of the Atomic Energy, Canada, was used to calibrate the dosimeters.

The gamma-cell was loaded with a 3,963 Ci ^{60}Co source on March 15, 1965. The source was in the form of a hollow circular cylinder located inside thick water shield. A 6 inch diameter by

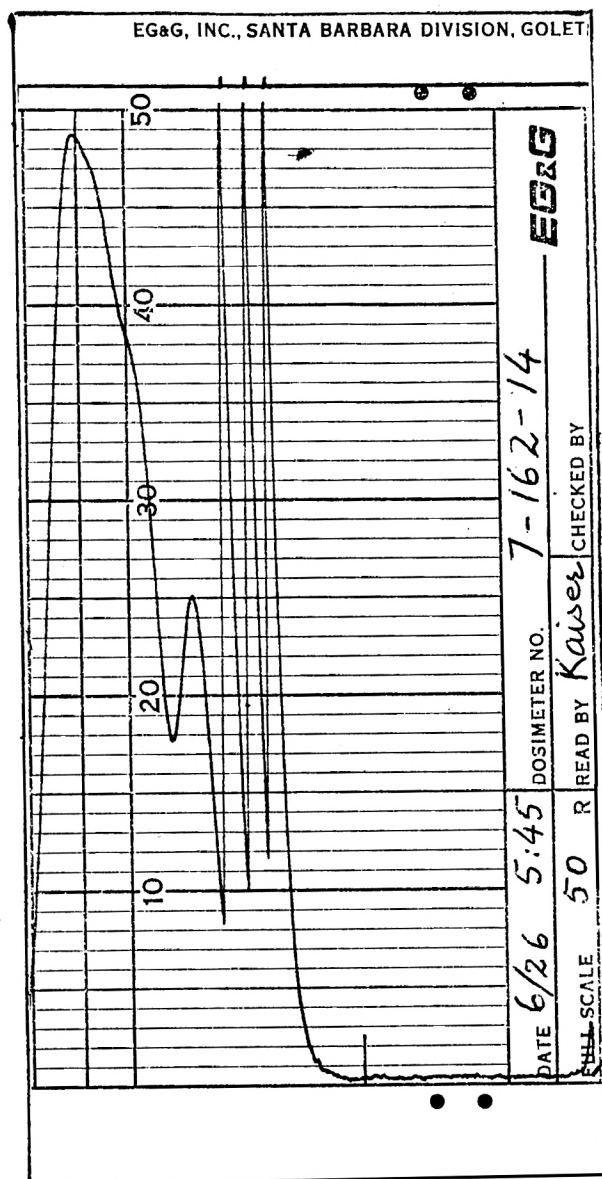


Fig. 9. A typical chart record obtained for EG and G model TL-3B TL dosimeter reader.

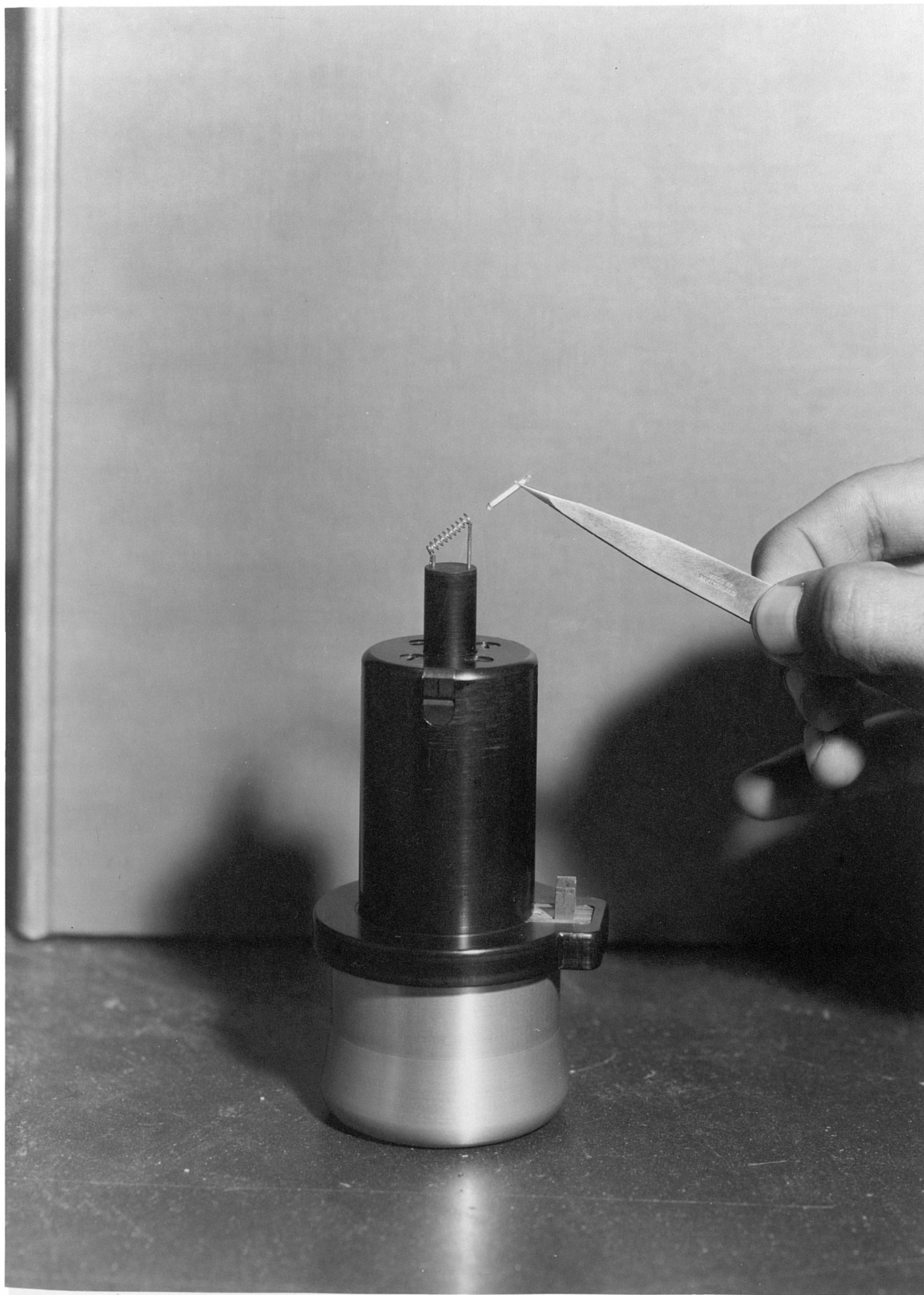


Fig. 10. EG and G model TL-81B read head adapter. Also showing model TL-21 dosimeter held in a pair of tweezers.

8 in high irradiation chamber was located in a plunger, which took it down in the middle of the source. The plunger was operated by an electric timer. The time of irradiation in seconds, minutes, or hours could be preset on the timer.

Correct dose rate at the time of irradiation was obtained from the chart of dose rates available at the gamma-cell.

4.4. ELECTRON LINEAR ACCELERATOR:

For obtaining very high dose rates of the order of 10^{10} rad/sec the electron linear accelerator (LINAC) at the Argonne National Laboratory was used. The full specifications of the LINAC were as follows:

Energy of electrons: Variable from about 4 to 20 MeV.

Energy Spectrum: About 15% of the total beam with an energy spread of 0.1 MeV. About 60% of the beam with an energy spread of 0.5 MeV. (These data were typical for beam energies of 10 MeV and greater).

Peak Beam Current: Dependent upon the energy of electrons; approximately 180 mA at about 13 MeV.

Pulse Length: Continuously variable from 0.4 μ sec to 5.5 μ sec. Pulse rise and decay time - 0.1 μ sec.

Repetition Rate: Single pulse or pulse chain. Repetition rate continuously variable from 10 Hz to 380 Hz.

Beam Diameter: About 1 cm. Quadrupole focusing available to obtain full intensity beams of a few mm in diameter.

Mounting: Horizontal.

To obtain a trace of the actual pulse of high energy electrons supplied by the LINAC a Faraday cup was mounted immediately behind the dosimeters. It collected the whole beam of electrons. The output of the Faraday cup was given to the oscilloscope on the control panel of the LINAC (Fig. 11). A polaroid camera was mounted on the oscilloscope to obtain photographs of the traces produced by electron beam.

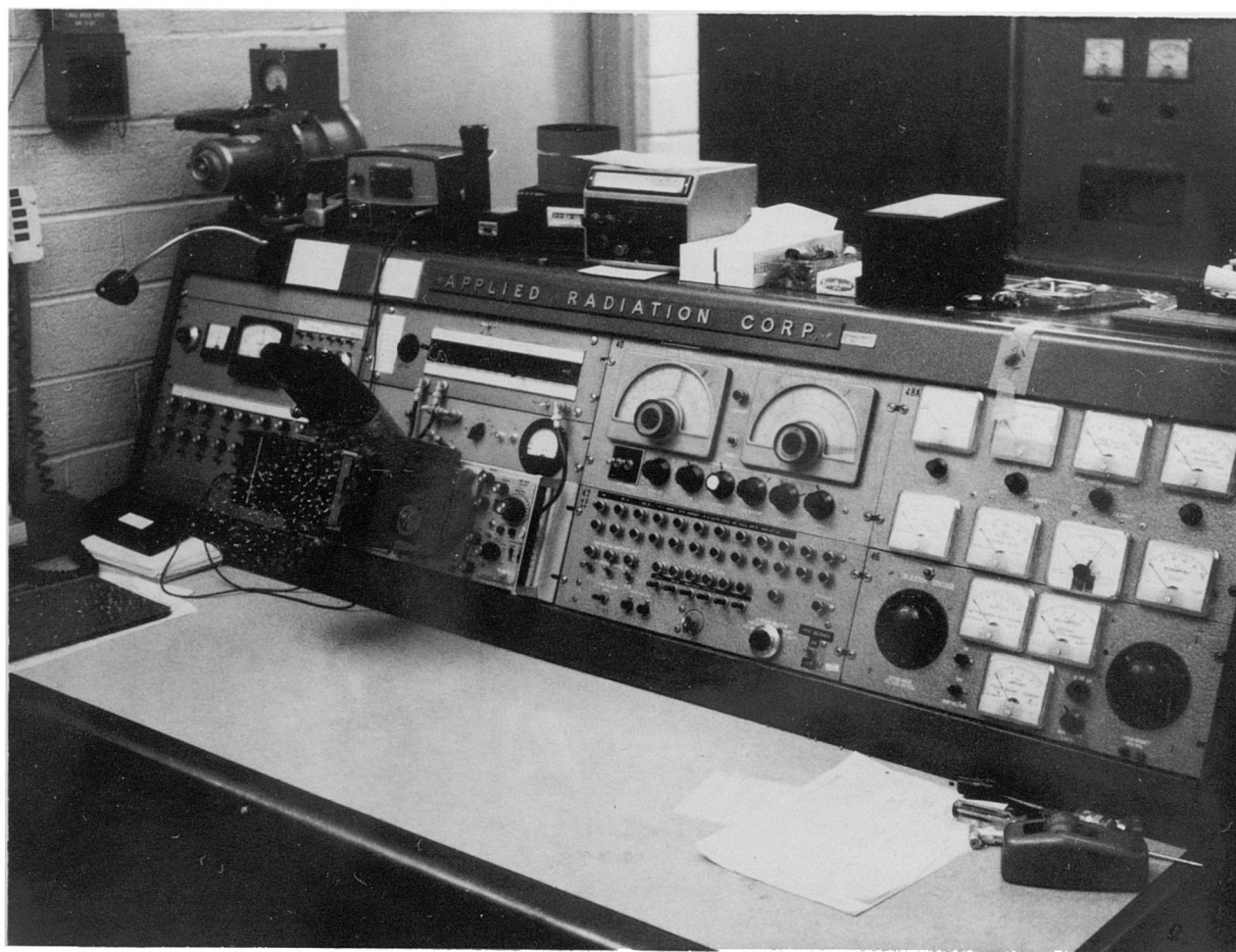


Fig. 11. A view of the control panel of LINAC. Also showing a poloroid camera mounted on the oscilloscope screen.

5. DETERMINATION OF THE CALIBRATION CURVE

5.1. NECESSITY:

The peak height measured on the glow curve plotted by the EG and G reader gave the response of the dosimeters in relative units. This response was supposed to be linear w.r.t. dose given to the dosimeter, if it did not exceed $10^3 R$. A calibration factor was required for the particular set of dosimeters used to convert peak height to the radiation dose. In the dose range above $10^3 R$ and upto about $10^5 R$ the response was supposed to be reproducible but non-linear w.r.t. the applied dose. For using the dosimeters in this high dose range, therefore, a full calibration curve was required to be obtained prior to the actual use of the dosimeters.

5.2. RADIATION FACILITY:

The gamma cell described in section 4.3 was used to irradiate the dosimeters. A polyethylene disc, about 1/2 in. thick, was made to fit exactly in the irradiation chamber of the gamma-cell. Polyethylene was used because it attenuated gamma-rays to the least extent and was convenient to machine and handle. Dosimeters were located in it along the circumference of a 2 in diameter circle concentric with the outer diameter. This ensured that all the dosimeters irradiated at one time received the same amount of dose, and their position did not change in different trials. Correct position of the dosimeters in the irradiation chamber,

such that they received the same amount of dose as if they were all placed at the center of the chamber, was determined from the iso-dose curves for the chamber supplied by the manufacturers. This was 2-3/4 in. above the base of the chamber when the dosimeters were 2 in. away from its axis.

Arrangements of timer in the gamma-cell allowed irradiation of the dosimeters for any amount of pre-set time. But the timer started counting time only after the plunger reached its lowest position. And also the plunger started coming upwards after the pre-set time in the timer was over. Dosimeters were, therefore, likely to absorb certain amount of additional dose during the downward and upward motions of the plunger. This dose was in addition to the dose calculated by the dose rate and the time of irradiation, and should be constant irrespective of the actual time of irradiation.

To determine this additional amount of dose the following mathematical model was used. If the response of the dosimeter due only to this additional dose was represented by R_0 , the dose rate in response units by \dot{R} and the irradiation time by t , then the total response R , of the dosimeter in the linear portion of the calibration curve would be given by:

$$R = R_0 + \dot{R} t \quad (5.2-1)$$

The quantities R_0 and \dot{R} could then be obtained by a least squares analysis of the data in the dose range of $(0-10^3) R$, or about

(0-800) rad in LiF*, in which equation (5.2-1) would hold. The calibration factor in units of rad per unit response could then be obtained by dividing the known value of the dose rate of gamma-cell by the experimental value of \dot{R} .

5.3. EXPERIMENTAL PROCEDURE:

To obtain the calibration curve in the wide range of dose from 0 to about 50 krad the experimental points were chosen to be almost equally distributed on a logarithmic scale. The durations of irradiation required in the gamma-cell for the particular values of doses were calculated. Rounded off values of the durations were taken in order that they might be set conveniently on the timer of the gamma-cell. Dosimeters were annealed at 400°C for 1 hr. before irradiation. Eight dosimeters were used at each experimental point. After irradiating several batches of dosimeters to the pre-determined durations of time all were subjected to a partial annealing at 110°C for 7 min. Dosimeters were laid on an asbestos pad and fed into the furnace for annealing.

5.4. ANALYSIS OF DATA AND RESULTS:

The mean values and the standard deviations of the eight response values at every experimental point were calculated. The data were reported in table 2. Using the first five data points

*Exact conversion factor for converting dose in roentgens to dose in rads absorbed in LiF is 0.8095. For arriving at this number refer to pages 86-88 of ref. (18).

Table 2. Data and results for the calibration curve.

S.No.	Duration of irradiation t, sec.	Estimated dose* D, rad.	Total Response R, units	Total Dose (D+D ₀), rad
1.	3	148.61	51.25±1.28	215.33
2.	6	297.22	78.56±4.05	364.51
3.	9	445.83	109.94±5.53	513.12
4.	12	594.44	141.5±15.43	661.73
5.	16	792.58	193.13±13.61	859.87
6.	30	1486.08	372.38±24.11	1553.37
7.	60	2972.19	607.5±59.7	3039.48
8.	100	4953.65	1263.13±85	5020.94
9.	200	9907.31	3367±207	9974.60
10.	350	17337.79	5775±564	17405.06
11.	600	29721.93	11100±625	29789.20
12.	820	40620.00	14879±682	40687.25

*Estimated dose = (Dose rate in gamma-cell x Duration of irradiation). Dose rate in the gamma-cell at the time of this experiment was 49.54 rad/sec.

and equation (5.2-1) the values of R_0 and \dot{R} were calculated by a least squares analysis. The calibration factor for the dose range from 0 to 800 rad was thus determined to be 4.553 rad per unit response.

The duration of irradiation in the gamma-cell multiplied by the dose rate gave the expected dose given to the dosimeters. The response R_0 (the value obtained by the least squares analysis; 14.78) multiplied by the calibration factor gave the additional dose D_0 . The total dose given by the estimated dose D plus the additional dose D_0 was plotted in Fig. 12 against the total response R to obtain the full calibration curve.

It might be seen from Fig. 12 that the response of TL-21 dosimeters became superlinear w.r.t. dose above a dose value of about 800 rad. One more interesting fact was observed in the calibration curve. The data points in the dose range 3 krad up to 40 krad were analysed by a least squares procedure. The fit obtained was very close to the one drawn through the points. This shows that the response is again almost a linear function of dose in the high dose range, although this line is different from the line corresponding to low dose.

5.5. CALIBRATION AFTER THE MAIN EXPERIMENT:

Dosimeters were being used almost at the limit of their useability, hence it was suspected that they might develop permanent lattice defects which would alter their characteristics altogether. To make sure that this did not take place a calibration curve for the same dosimeters was again obtained after

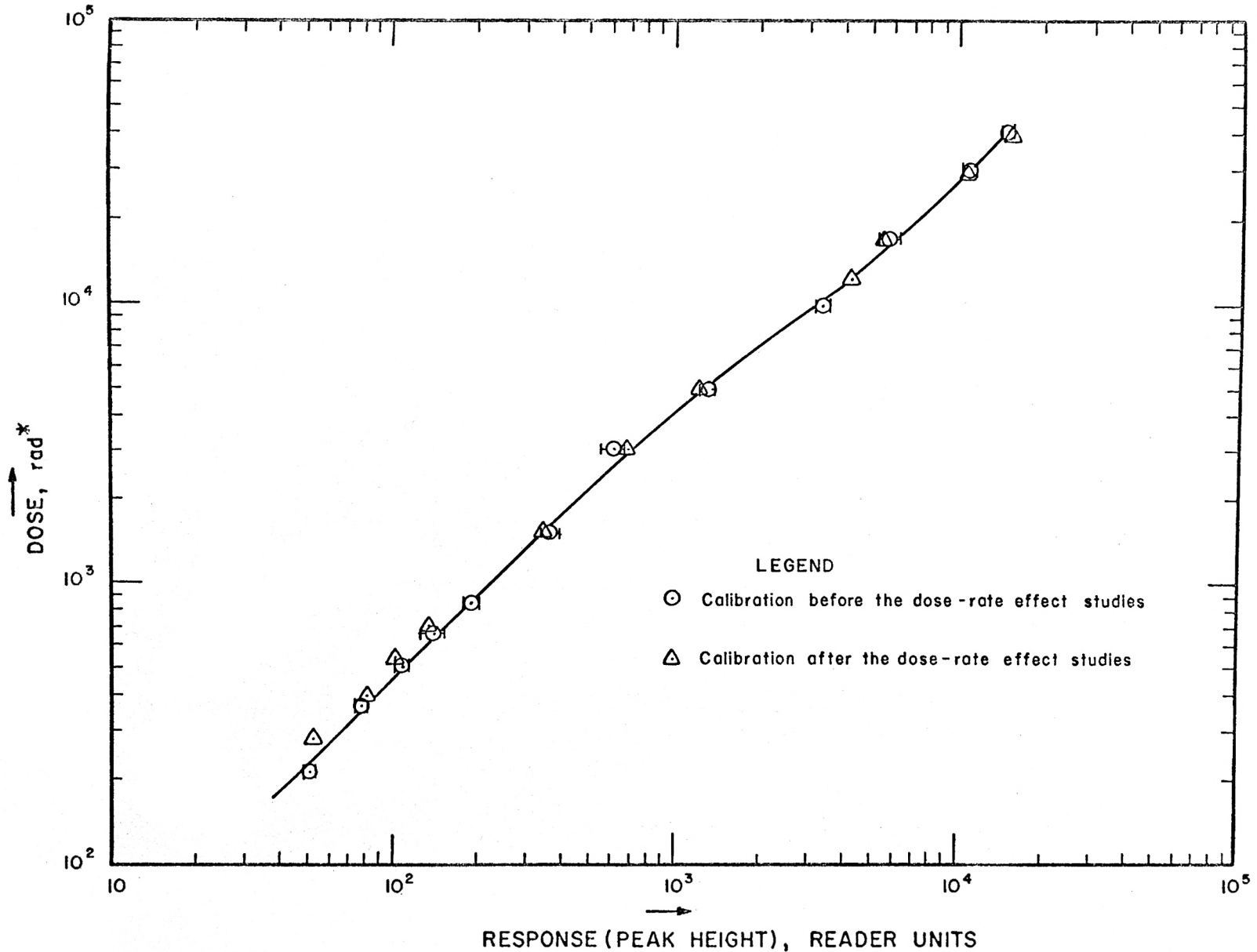


Fig. 12. Calibration curve for the EG and G model TL-21 LiF dosimeters in (0-4x10⁴) rad dose range.

the main experiment (described in the following sections) using the electron linear accelerator was done. The experimental points obtained in this case were also plotted in Fig. 12. The close agreement between these points and the previous calibration curve showed that there were no permanent defects created in the phosphor.

6. DESIGN OF THE EXPERIMENT

6.1. GUIDELINES:

As indicated earlier, one of the main objectives of this work was to investigate whether or not the response of LiF TL dosimeters depended upon the dose rate at which the radiation dose was delivered to them. This could have been predicted straight from theory had the mechanism of TL of LiF been known for sure. Since this was not the case, the phenomenon had to be investigated experimentally. However, guidelines for the design of the experiment were obtained from whatever theory was available.

The only theory which discussed the dose rate dependence of the response was Donnert's theory referred to in sec. 2.2.6. Once again to recall, the theory suggested that the response of a dosimeter would be dependent upon the rate at which the radiation dose was given to them and that this dependence would be characteristic of the total dose given to the dosimeter only if the processes involved in the irradiation were not all of first order kinetically. This would be true, in principle, at any value of the dose rate and any amount of the total dose, but the phenomenon might or might not be observable in an actual experiment. To assure that the phenomenon would be observable it seemed necessary to irradiate the dosimeters at high dose rates and large amounts of total doses. Here again, guidance was obtained from a similar experiment⁽¹⁰⁾ performed to observe the dose rate dependence of film and glass dosimeters. The dose

rates to be used in the experiment were decided to be in the range of 10^9 to 5×10^{10} rad/sec (or, in different units, 1 to 50 krad/ μ sec).

6.2. RADIATION FACILITY:

There were two primary requirements of the radiation facility to be used in this experiment. First, it should provide gamma-radiation or some radiation which is practically equivalent to gamma-radiation. Second, it should be capable of providing radiation dose at a rate in the desired range of 1 to 50 krad/ μ sec. In addition there was one more requirement set on the radiation facility by the type of dosimeters selected for use in the experiment. The model TL-21 dosimeters were good for use in the dose range from a few mR to 10^5 R (or 8.095×10^4 rad (LiF)). Therefore the total dose that could be given to them was to be less than the upper limit of their useful range. For giving a total dose of not exceeding 8×10^4 rad at a dose rate as high as 50 krad/ μ sec. the irradiation time should be no longer than about 1.5 μ sec. It should be actually still smaller for obtaining lower total doses at the highest dose rate aimed at. Therefore, the radiation facility should be capable of giving out radiation in the form of intense, accurate and measureable pulses of 1 μ sec. or less in width.

The high dose rates of interest were not obtainable from any radioisotopes or any other available sources of gamma-radiation. The Electron Linear Accelerator, described in sec. 4.4, available at the Argonne National Laboratory was found to be a suitable

radiation facility for the purpose of this experiment. Full specifications of the LINAC were given in sec. 4.4. The 14 MeV electrons given out by the LINAC with maximum efficiency were considered best substitute for gamma-rays, as the difference in response of the dosimeters to these two types of radiation was seen to be slight (sec. 2.2.1).

Total radiation dose supplied at a time by the LINAC was in the form of a single pulse. The pulse height was measured in units of mA of electron current and the pulse width in units of μsec . The arrangement used to obtain a trace of the pulse shape was described in sec. 4.4. The area under the pulse gave the total charge supplied by the LINAC in that pulse in units of nC (10^{-9} coulombs). The total dose supplied was proportional to the total charge supplied and the dose rate was proportional to the electron beam current. The conversion factor from delivered charge to dose was obtained from ref. 10 to be 0.227 krad/nC. This conversion factor was originally determined by dose measurements with the standard Fricke dosimeter for which the dose and dose-rate dependent response was known.

6.3. STATISTICAL DESIGN OF THE EXPERIMENT:

Statistical design of the experiment consists in setting up approximate values of the controlled variables in the experiment, so as to obtain statistically significant results.

There were two independent controlled variables in our experiment; namely the total dose D and the dose rate θ , and the

mathematical model selected for the analysis of the response R of the dosimeters was given in equation (2.2.7-2). There were in all four constants to be evaluated. Theoretically, measurement of response at four properly chosen combinations of D and θ would suffice for the determination of the four constants. But since the proposed model might not be the true relationship between R, D, and θ , one would be interested in knowing the statistical significance of the fit obtained. This required measurement of response at more than four different combinations of D and θ .

In selection of different combinations of D and θ several factors could be taken into account, such as: good fit over the broadest range of interest of the controlled variables, ease in data analysis, and practical feasibility. Importance of all these factors is discussed in several text books on statistics. Various experimental designs are also suggested for various models based upon these factors. An excellent brief discussion may be found in ref. (19). A standard design suggested for a full second order model is a 3^2 factorial design in the controlled variables. The same design was selected for our incomplete second order model, but this had to be modified because of certain practical limitations faced.

The broadest readily obtainable operating conditions of the accelerator were utilized. It was intended to supply three different dose levels, each at three different pulse widths. This amounted to supplying dose at three different levels and each with three different dose rates. The three pulse widths selected were 0.4, 1.2 and 4 μ sec. The dose levels were set at

30, 15 and 7 krad. These required beam currents of about 40, 20 and 10 mA at 4 μ sec pulse width; 120, 80 and 40 mA at 1.2 μ sec pulse width; and 400, 200 and 100 mA at the pulse width of 0.4 μ sec. Due to some recent modifications (post dating the experiment reported in ref. 10), the accelerator could give a maximum current of only 130 mA. Therefore the two high-dose points at the shortest pulse width could not be obtained. Instead, it was decided to add one high dose point both at the 4 μ sec and the 1.2 μ sec pulse widths, and to get three dose points at the 0.4 μ sec pulse width corresponding to beam currents of 130, 120 and 80 mA. The final experimental design consisting of 11 points is shown in Fig. 13 in the dose-pulse width plane.

From the past experience it was known that the electron beam showed a tendency to meander a little from its target in different trials. There were, therefore, chances of certain dosimeters not being hit by the beam properly. To allow for such losses of dosimeter samples, and to obtain good statistics from the data, it was decided to irradiate 4 sample packs in succession at each experimental point shown in Fig. 13.

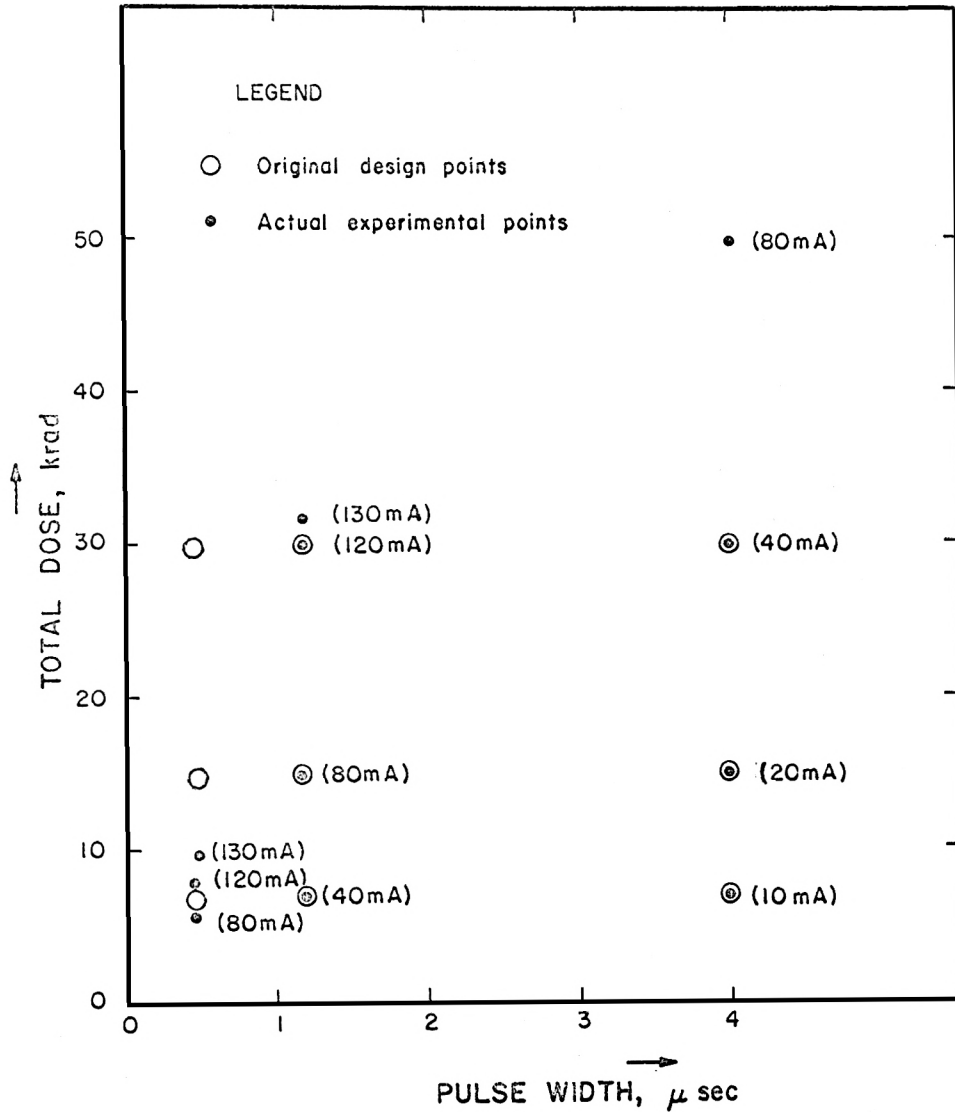


Figure 13. Design of the experiment.
 (Numbers in parantheses near every point represent the beam current).

7. EXPERIMENTAL PROCEDURE

7.1. ELECTRON BEAM MONITORING:

The electron beam given out by the LINAC was 1 cm in diameter. The energy spectrum of the beam was specified as: about 15% of the total beam with an energy spread of 0.1 MeV and about 60% of the beam with an energy spread of 0.5 MeV. The dosimeters were 1.2 cm long, and it was known that the beam does meander a little from the target in different trials. Therefore it was important to know how actually the beam hit the dosimeters, and whether they received the full dose they were expected to receive or not. To accomplish this, high speed x-ray film, Kodak type 580-0, was kept immediately behind the dosimeters. The shape of the beam along with a shadow of the dosimeters was obtained on the x-ray films. (See Fig. 17).

7.2. PREPARATION OF SAMPLES:

Physical size of the dosimeters was 1.4 mm diameter and 1.2 cm long. Since the beam had a roughly circular cross-section of 1 cm diameter, three dosimeters kept side by side could easily be irradiated at a time. Suitable dosimeter packages, convenient to irradiate on the accelerator, were made. These packages consisted of aluminum rings, 5 cm O.D 3 cm I.D and 1/16 in thick, Kodak type 580-0 high speed x-ray film and 3 EG and G model TL-21 Nat LiF thermoluminescent dosimeter which were pre-annealed at 400° for 1 hr. The dosimeters used were from the same batch for which the calibration curve of Fig. 12 was obtained.

Center lines intersecting at 90° were marked on the aluminum rings. The x-ray films were pasted onto the aluminum ring with Duco cement (E.I. DuPont deNemours and company, Wilmington, Delaware), such that the center lines on the x-ray film package coincided with the center lines marked on the rings. Three dosimeters were kept in a thin-walled poly-ethelene bag, in order to keep them clean, and were pasted at the center of the x-ray films using Scotch brand (3M Inc., Saint Paul, Minnesota) tape which was sticky on both sides. Forty nine such packages were made. Serial numbers were marked on all samples with embossing tape at a particular position such that the position of this number on the sample served also as a reference point for describing the positions of dosimeters in the sample. A view of a sample with a scale showing its size could be seen in Fig. 14.

7.3. SAMPLE HOLDING DEVICE:

Alignment of the dosimeter packages along the center line of the electron beam was critical. To facilitate this and to facilitate rapid sample changing on the accelerator a 3-jaw self-centering holding device was used, which may be seen in the irradiation assembly in Fig. 15. The jaws contained co-planar V-grooves in which the aluminum ring used in the sample could be seated. Two levers were provided to expand the jaws which were otherwise held close to the center by a spring. The outer ring of the holding device was provided with a fine ground radial rod which could be easily fixed in any stand on an optical bench.

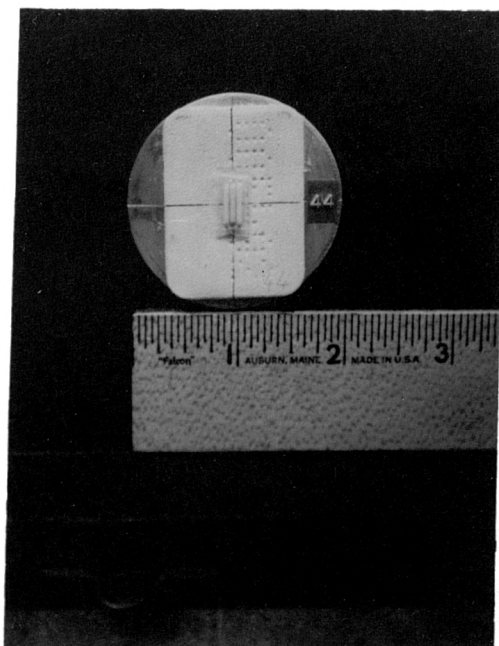


Fig. 14. Dosimeter sample for irradiation on LINAC.

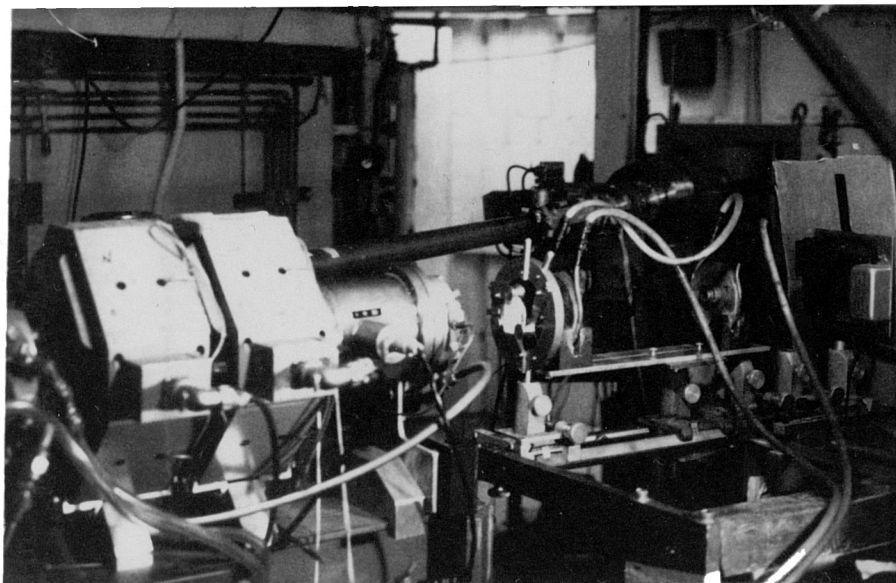


Fig. 15. Irradiation assembly on LINAC.

Once the holding device was aligned with the beam sample changing just amounted to taking out one sample and putting in another in the self-centering jaws without any delay.

7.4. ALIGNMENT OF SAMPLE WITH THE ELECTRON BEAM:

The electron beam was horizontal. An optical bench, provided with the accelerator as a standard accessory, was set horizontal in front of the beam port parallel to the axis of the beam. The holding device described in the preceding section was held in a stand on the optical bench close to the beam exit such that its face was at right angles with the beam axis. The stand was provided with a lateral adjustment screw. A view of the irradiation assembly could be seen in Fig. 15.

A few glass samples were made by fixing (with Duco Cement) ordinary plane-glass pieces on the aluminum rings identical to those used for the dosimeter packages. One of the glass samples was held in the holding device and irradiated with the electron beam for a couple of minutes. The glass turned brown in the portion where it was hit by the beam. The position of this brown spot showed how far the center of the holding device was off from the beam axis. Alignment was corrected using the lateral adjustment screw and by moving the holding device up or down as necessary. Another glass sample was irradiated and the correct alignment was obtained by several such trials.

7.5. IRRADIATION OF SAMPLES:

The irradiation assembly was completed by mounting a Faraday cup immediately behind the holding device. The output of the Faraday cup was fed to the oscilloscope on the control panel of the accelerator on which a polaroid camera was mounted, as described earlier in section 4.4, to take photographs of the pulse-shape traces.

LINAC was operated by licensed operators at the Argonne National Laboratory. First it was set for the maximum current it can deliver (130 mA). The pulse width was set at 0.4 μ sec and the scales on the oscilloscope screen were adjusted to get the pulse shape of a reasonable size. A sample was mounted in the holding device. To keep track of the relative positions of the three dosimeters in the sample, all the samples were mounted in a particular fixed position (which kept the dosimeters vertical and the serial number marked with the embossing tape to the right). The accelerator was pulsed once, and a photograph of the oscilloscope trace of the pulse shape was taken. Sample number, beam current, pulse width and the known scale-calibration factors on the oscilloscope screen were recorded on the photograph. Another fresh sample was mounted in the holding device for another irradiation.

Following the same procedure four dosimeter packages at every setting of the accelerator given by the experimental design in Fig. 13 were irradiated. Forty seven of the total forty nine packages (including some which were spoiled) were irradiated

and two packages #44 and #49 were saved as control samples.

7.6. READ-OUT PROCEDURE:

All the dosimeters were removed from the packages, keeping carefully track of each individual dosimeter. In any package the dosimeter nearest to the embossing tape carrying the serial number was numbered 1, the middle one 2 and the last one 3. The dosimeters were brought back from Argonne National Laboratory to Manhattan, Kansas, the next day and were partially annealed, all simultaneously, at 110°C for 7 min. This partial annealing was carried out about 30 hrs after the irradiation. For partial annealing the dosimeters were set on an asbestos pad and fed into a furnace pre-heated to 110°C.

The high voltage on the dosimeter reader was set as described in section 4.2 in order to obtain the highest dose range upto 50 krad. All the dosimeters were read out approximately 24 hrs after the partial annealing.

7.7. DETERMINATION OF DOSE:

The total dose supplied in a pulse was proportional to the total charge delivered in that pulse. The total charge delivered in various pulses was determined by a planimetric integration of the true shapes of pulses obtained in the photographs. The conversion factor was 0.227 krad (water)/nC (see sec. 6.2) which reduced to 0.188 krad (LiF)/nC. The dose received by a dosimeter would be 0.188 x the charge (nC) only if the dosimeter was well hit by the beam and received the full dose. To check on

this, the x-ray films in all the packages were developed using the standard x-ray film developing techniques.

All the raw data are presented in Appendix B.

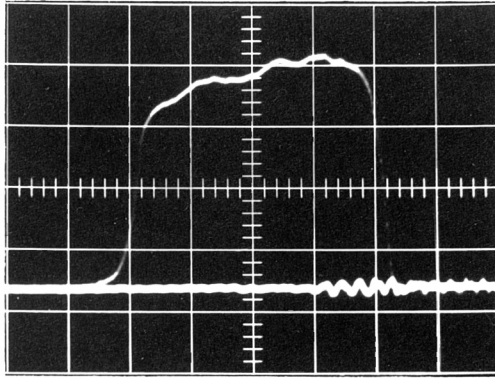
8. ANALYSIS OF DATA

8.1. PULSE SHAPES:

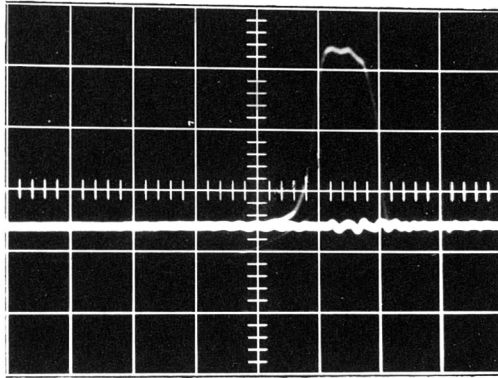
Pulse shapes should have been rectangular, which would assure that the total dose supplied in a pulse was with a constant dose rate. But the pulse rise and decay time in LINAC were specified as 0.1 μ sec, so that true rectangular shapes could not be expected. Typical pulse shapes given by LINAC at the pulse widths of 4.0, 1.2 and 0.4 μ sec were reproduced in Fig. 16. The pulse shapes in the first two cases were reasonably rectangular but the shape in the last case deviated much from being rectangular. In all cases the dose rate values were calculated directly from the current setting, ignoring completely the pulse shapes. The total charge delivered was however obtained from the true pulse shapes.

8.2. SORTING OF THE DOSIMETERS:

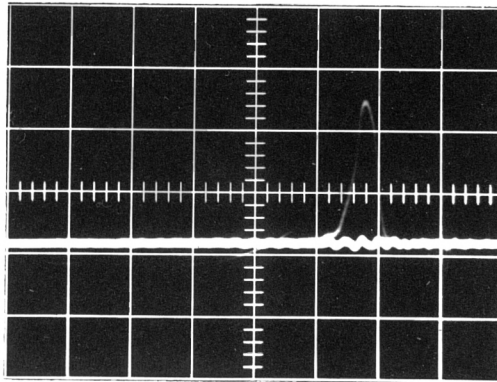
The developed x-ray films revealed three facts: (1) The electron beam in most of the cases did not hit the dosimeters as well as it was supposed to do. The center of the beam was off from the center of the sample due to some probable disturbance in alignment. (2) Position of the beam was not fixed with respect to the center of the sample for all the samples. (3) The size of the projection made by the beam on the film varied with the total charge supplied in the pulse, being largest for the highest amount of charge.



(a) 80 mA, 4.0 μ sec.



(b) 130 mA, 1.2 μ sec.



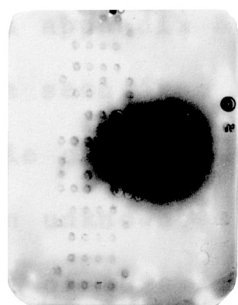
(c) 130 mA, 0.4 μ sec.

Fig. 16. Typical electron beam pulse shapes at three different pulse widths.

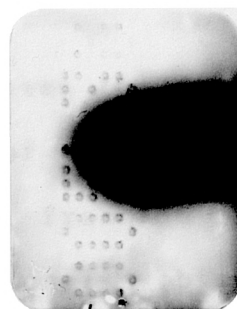
A few of the developed films were reproduced in Fig. 17. The improper irradiation in most of the cases caused the three dosimeters in a sample to receive varying amounts of dose. Obviously the dosimeter nearest the center of the beam received the highest dose and those away from the center received only a fraction of the total dose. This fact was also apparent from the response data obtained.

This was quite unfortunate. It was impossible to say precisely which dosimeter received what fraction of the total dose. It was assumed that the intensity of the beam varied monotonically, from the center to the edge. Dosimeters in various samples whose distance off from the center of the beam was a fixed fraction of the radius of the beam shadow could then be said to have received a fixed fraction of the total dose. On this basis one group of the dosimeters was selected. It consisted of dosimeters which appeared in a majority of cases closest to the center of the beam.

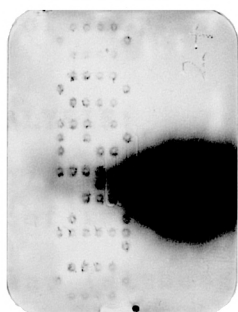
Referring to Fig. 17, the top two pictures are reproductions of the films for samples 3 and 29; the former being an example of a sample receiving a low dose and the latter an example of a sample receiving a high dose. From all such samples dosimeter number 1 (one to the extreme right) was selected in the group. From some samples like for example #40 (bottom right in Fig. 17) dosimeter #2 was selected whereas from samples like #17 (bottom left in Fig. 17) none of the dosimeter could be selected. The electron beam missed the sample miserably in some cases like for example #24 (low dose) and #10 (high dose) (see middle row



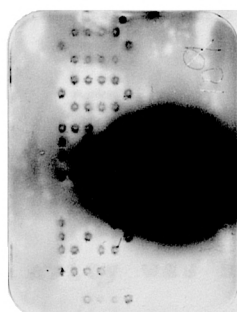
3



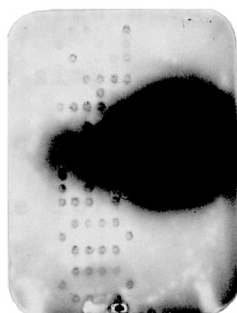
29



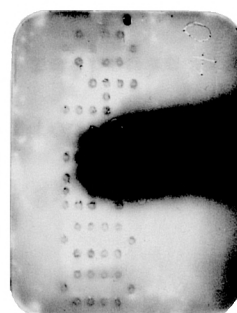
24



10



17



40

Fig. 17. Examples of developed x-ray films showing beam meandering.

in Fig. 17). All such samples were completely discarded. This way out of total 141 dosimeters irradiation only 20 could be picked out. These selected dosimeters were marked with (*) in the table of raw data in appendix B.

It may be seen from Fig. 17 that even the dosimeters picked out did not lie exactly at the center of the beam and hence received only an unknown, although fixed, fraction of the total dose. Because of this limitation the results obtained by the analysis of data were correct only qualitatively. The response surface obtained (see the following sections) showed only the general trend of response with increasing dose and dose rate.

8.3. DATA ANALYSIS:

Response of the model TL-21 dosimeters at low dose rates was a non-linear (although the non-linearity was slight) function of dose at the high dose values as seen in the calibration curve of Fig. 12. This non-linearity effect was to be eliminated when the effect of high dose rates alone was being studied. This could be done simply by converting the response values to response in dose units using the calibration curve. The same thing was done for the response values given by the selected dosimeters.*

*Strictly speaking the response values from 15 MeV. electron irradiation should have been converted to equivalent response to ^{60}Co gamma-rays using the conversion factor given in sec. 2.2.1. before using the calibration curve. But since the calibration curve was linear (line not passing through the origin) in the high dose range and the results looked forward to were only qualitative, this conversion was ignored simply to reduce the number of calculations.

Should there be no effect of high dose rates used in the experiment on the response of the dosimeters, the response in dose units would give the actual dose received by the dosimeters. In such a case data analysis would yield: $R_{01} = 0$, $R_{11} = 0$ and $R_{10} \neq 0$, where the parameters represent the constant coefficients in the mathematical model selected in section 2.2.7. On the other hand if response was affected by the high dose rates the data analysis would yield the coefficients R_{01} and R_{11} different from zero; greater or less than zero depending upon the effect of the high dose rates.

A two-dimensional plot of the data was not possible. The total charge given by the electron beam was fluctuating and no two data points were obtained at any particular fixed dose level. Therefore any simple graphical data analysis was not possible.

The mathematical model selected for data analysis was:

$$R = R_{00} + R_{10}D + R_{01}\theta + R_{11}D\theta$$

The constant coefficients R_{00} , R_{10} , R_{01} and R_{11} were evaluated performing a regression analysis on the data by the "Forward Doolittle technique" described by Hunter in ref. 19. A more general discussion of the technique could be found in ref. 20. The values of D and θ used in the analysis were the total dose and dose rate values which the dosimeters would have received had they been at the center of the beam. Since the actual dose and dose rate received by the dosimeters were constant fractions of the total values, the qualitative nature of the results obtained

were not affected by the higher values of D and θ used. To keep the numbers in reasonable magnitudes the dose values were taken in units of krad and the dose rates in units of krad/ μ sec. Statistical significance of the results was determined by applying F-test on the values of coefficients determined. Results along with the calculated F-ratios and critical F-ratios are presented in table 3. The values of the critical F-ratios were obtained from ref. 21.

8.4. RESULTS:

The data analysis yielded the equation:

$$R = R_{00} + R_{10}D + R_{01}\theta + R_{11}D\theta \quad (8.4)$$

with

$$R_{00} = 1274 \text{ (krad*)}$$

$$R_{10} = 292.6 \text{ (krad* krad}^{-1}\text{)}$$

$$R_{01} = 26.9 \text{ (krad* krad}^{-1}\mu\text{sec)}$$

$$R_{11} = 8.47 \text{ (krad* krad}^{-2}\mu\text{sec)}$$

where krad* is the dose of ^{60}Co gamma-rays delivered at a low dose rate which is equivalent to the response.

The coefficient of θ was seen to be significantly different from zero at the 95% confidence level and that of the interaction term ($D\theta$) at the 80% confidence level. Hence the dose rate dependence of the response of EG and G model TL-21 dosimeters was proved. The results obtained were only of qualitative nature since the actual dose and dose-rates received by the dosimeters were not exactly known on an absolute scale.

8.5. PRESENTATION OF RESULTS:

Equation (8.4) represents a surface in the R-D- θ space. An isometric view of the surface is shown in Fig. 18. The experimental

Table 3. Results of Regression Analysis

Regression Parameter	Value	Calculated F-ratio	Critical F-ratio	Confidence Level
R_{00} (krad*)	1274	--	--	--
R_{10} (krad* krad ⁻¹)	292.6	--	--	--
R_{01} (krad* krad ⁻¹ μsec)	26.9	13.85	4.49	95%
R_{11} (krad* krad ⁻² μsec)	8.47	2.26	1.79 (3.05)	80% (90%)

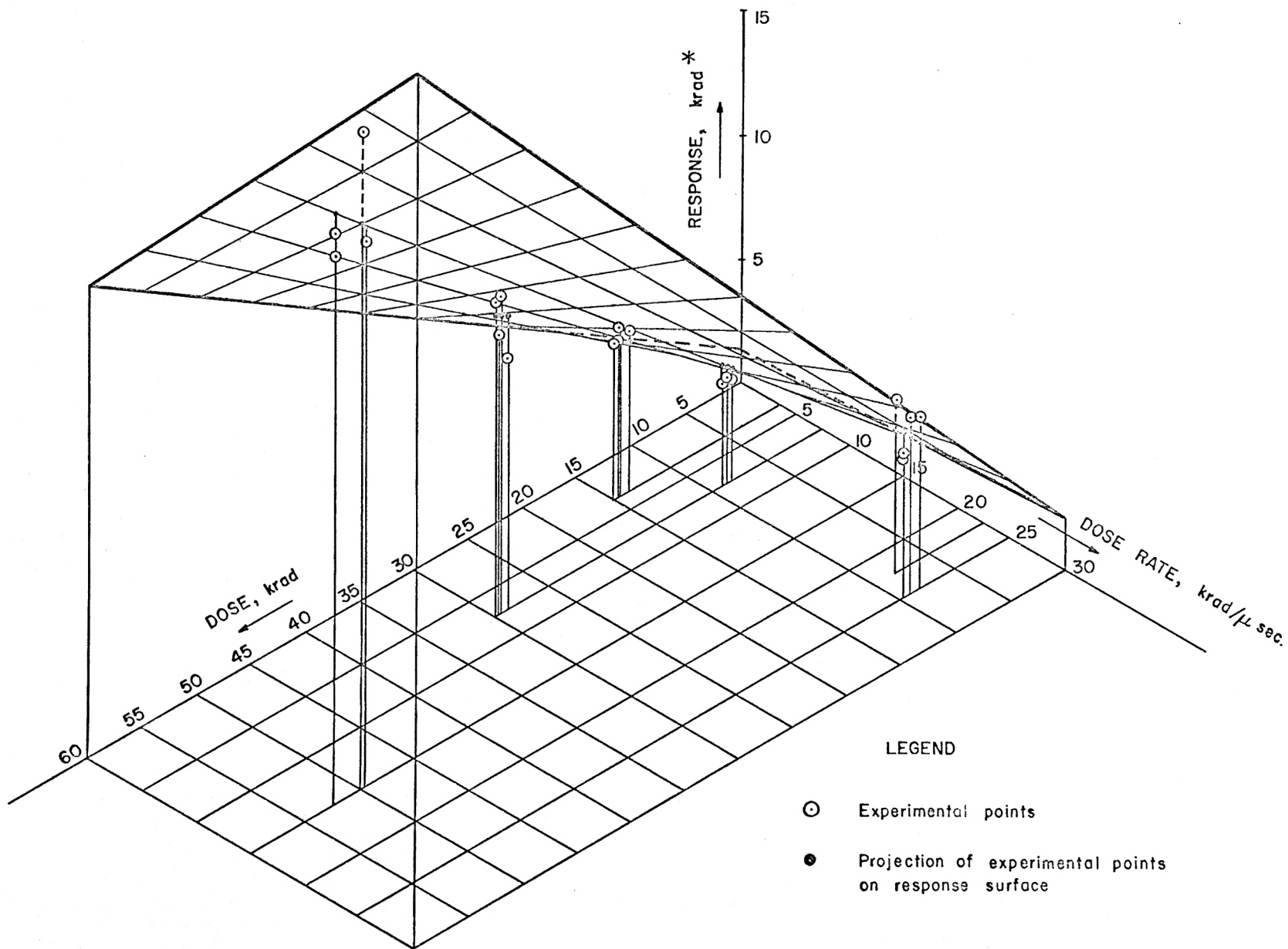


Fig. 18. Response of LiF dosimeters (EG and G model TL-21) to varying dose and dose rate. (Ordinates represent the response converted to equivalent dose of ^{60}Co gamma-rays delivered at low dose rate)

data points have also been located in the space. A majority of the data points lie very close to the surface showing that the fit obtained is very close. Shape of this surface is quite interesting. Surface seems to fold upwards near the origin, but it should be remembered that the surface does not hold good in the low dose and dose rate ranges. One more point worth mentioning is the quantity plotted along the ordinates. It is not the response as obtained directly from the TLD-reader, but is the response converted to dose using the calibration curve determined from the irradiation of dosimeters at low dose-rates by ^{60}Co gamma-rays.

Although the three dimensional plot of the response is its full representation, it is not convenient for quick reference. Its use in practice may be involved in problems such as given the response and dose rate determine the dose. This is clearly not very convenient from the three dimensional plot. Two dimensional plots either in the R- θ plane or R-D plane can be obtained for several fixed values of D or θ respectively. In either case it may be seen from equation (8.4) that the relationships reduce to linear ones. Relationships between R and θ for some fixed values of D, the so-called iso-dose lines, have been plotted in Fig. 19. The types of problem referred to above can be solved using this figure by first locating a point in the plane using the given data and then getting dose values by interpolation between iso-dose lines. Further advantage of this type of plot lies in the fact that it gives a clear picture of the effect of dose-rate alone on the response at several dose levels.

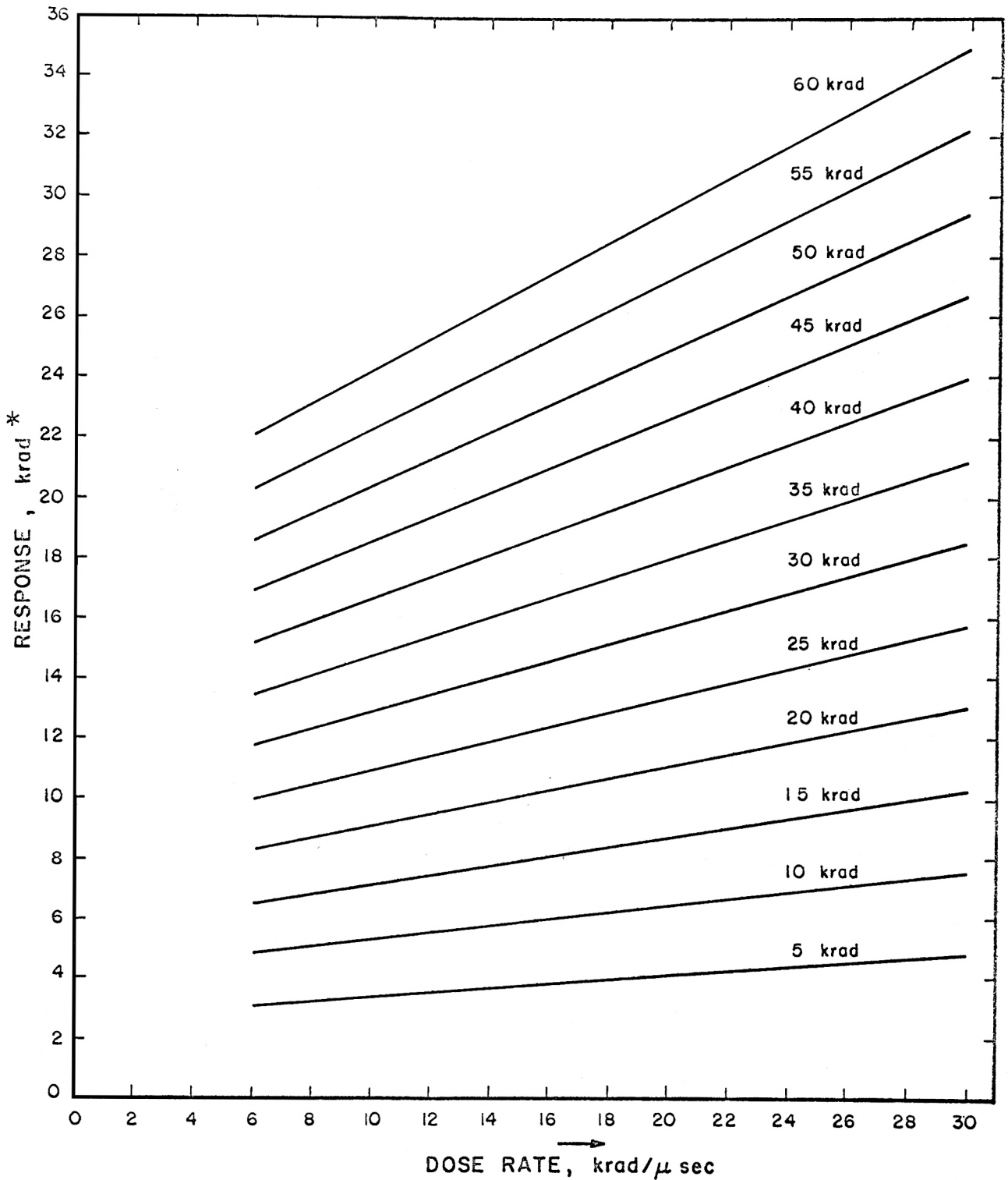


Fig. 19. Response of LiF dosimeters (EG and G model TL-21) to varying dose rate at some fixed dose levels (Iso-dose lines). (Ordinates represent response converted to equivalent ^{60}Co gamma-ray dose delivered at low dose rates).

9. DISCUSSIONS AND CONCLUSIONS

9.1. DISCUSSION OF RESULTS:

The coefficient R_{01} of θ in the response equation has been shown to be significantly different from zero at the 95% confidence level (this is actually true even at the 99% confidence level). This is sufficient to say decisively that the response of EG and G model TL-21 LiF dosimeters is dose rate dependent. Further the coefficient R_{11} of the interaction term ($D\theta$) is also significantly different from zero at the 80% confidence level. Donnert's theory (see Appendix A.) says that if the response is dose-rate dependent, the coefficient R_{11} should also be different from zero. Rather low confidence level is obtained on the significance of this parameter probably because of some insufficient data. Even then 80% confidence level is quite agreeable. Therefore going back from Donnert's theory it may be said that mechanism of irradiation of LiF involves processes of kinetic order other than one.

Only a particular type of LiF dosimeters, namely the EG and G model TL-21, were used in the experiment. But none of the limitations placed on the dosimeters based upon their physical structure and size were surpassed in the experiment. Therefore the qualitative results obtained by this experiment must hold good in general for any kind of LiF dosimeters. The dose levels and dose rates at which the dose rate effect becomes significant might change with the type of dosimeter but the general qualitative features determined in this experiment should not change.

Looking at Fig. 20, wherein some iso-dose lines are shown, two facts are to be noticed:

- (1) the slopes of all the iso-dose lines are positive;
- (2) different iso-dose lines have different slopes, and the slopes increase with increasing dose level; showing that the dose rate dependence is characteristic of the total dose supplied to the dosimeter. This means that the dose rate dependence is also a function of the dose already supplied to the dosimeter.

Referring back to the Cameron's model of thermoluminescence of LiF described briefly in section 3.2.4 if the proposed hypothesis of the creation of traps by radiation were true the following statements can be made in the light of the results of this research:

- (1) the proportionality constant for the creation of traps in LiF is a function of dose rate
- (2) the proportionality constant for the creation of traps in LiF is also a function of the dose already supplied to the dosimeter.

9.2. PREVIOUS INVESTIGATIONS AND RESULTS:

Several investigators have tried to investigate the dose-rate dependence of LiF thermoluminescence dosimeter response. McCall et al. ⁽²²⁾ studied the dose rate effect upto 4×10^3 rad/sec; Karzmark et al. ⁽²³⁾ studied the dose rate effect upto 2×10^8 rad/sec using the scattered electron beam from a 15 MeV linear accelerator and all report dose rate independence

of response. Their results are not in contradiction with our results since the range of dose rate we used was 10^9 upto 5×10^{10} rad/sec, and our conclusions are that LiF dosimeters are affected by dose rate in this range.

9.3. CONCLUSIONS:

There were two main conclusions drawn from these studies.

(1) The response of LiF thermoluminescent dosimeters is an increasing function of the rate at which the radiation dose is supplied to it. For EG and G model TL-21 dosimeters the dose rate dependence is observable in the dose range from approximately 1 krad to 50 krad and in the dose rate range from approximately 1 krad/ μ sec to 50 krad/ μ sec.

(2) The dose rate dependence of LiF dosimeter response is itself an increasing function of the total dose given to the dosimeter.

Another conclusion drawn based on Donnert's theory is:

(3) The kinetic order of the processes involved in the irradiation of LiF phosphor is different from unity.

10. LIMITATIONS OF THE CURRENT INVESTIGATION

Limitations faced in the experiment were mainly from the radiation facility. The LINAC was the only radiation facility within comparatively easy reach that could be used in the experiment, but the practical limitations faced suggest that a more sophisticated radiation facility should be sought for in order to obtain accurate quantitative results.

There were chances of obtaining quantitative results at least for the EG and G model TL-21 dosimeters. All the prerequisites for data analysis, like for example the calibration curve with the radiation delivered at low dose rates and the ratio of the response of LiF to ^{60}Co gamma-rays and to 15 MeV electrons, were available. The beam meandering was the main restriction that made the determination of quantitative results impossible. With the LINAC this problem just seems unavoidable. Only solution that could be suggested for such a problem is to irradiate excessively large number of samples at all the experimental points. Occasional developing of the x-ray films from the already irradiated samples while the experiment is in progress may save several samples from being ruined.

Another limitation faced in way of getting some quantitative results was due to the pulse shapes given by the LINAC. Especially the pulse of 0.4 μsec duration (Fig. 16c) looked more like a triangle than a rectangle. It is important that the dose given in any pulse must be all at a constant dose rate, and this is achieved only when the beam current remains constant over the

entire pulse duration. If this is not achieved and the varying dose rate in a pulse is approximated by some sort of average constant dose rate, the quantitative results obtained will not be accurate. The best way to get away with this kind of problem on LINAC is to avoid using any pulses shorter than 1 μ sec. duration. This will cut down one lower dose level that was used in this experiment.

11. SCOPE FOR FUTURE WORK

There is vast scope of work to be done in the field of LiF thermoluminescence dosimetry, whose practical value seems to be great.

Further work that may be done in the specific direction of studying the dose-rate dependence obviously includes getting some quantitative results which may be of practical use. The tasks to be faced in this particular work will be: making available a radiation facility that is more sophisticated than LINAC, and selection of some LiF dosimeters whose higher limit of usability is sufficiently large.

In the general field of thermoluminescence of LiF the main problem of the determination of the detailed mechanisms of irradiation and readout still remains unsolved. The complex shape of the glow curve obtained from LiF and the interesting effects of pre-irradiation annealing studied by Zimmerman et al.⁽¹¹⁾ suggest that the mechanisms involved in the thermoluminescence of LiF are far more complicated than those for other phosphors like for example CaF_2 . Without the knowledge of the actual mechanism of thermoluminescence it is unsafe to extrapolate any of the experimental facts.

Some of the previous investigators like Bonfiglioli et al.⁽⁷⁾ and May and Partridge⁽²⁴⁾ have tried to assess the mechanisms of thermoluminescence of other phosphors like alkali halides. Their experiments may be repeated with LiF and in addition new experiments may be designed for studying the mechanism of thermoluminescence of LiF.

In the field of practical applications of LiF also much work remains to be done. Dosimetry in the mixed fields of neutron and gamma radiation is one problem the answer to which probably lies in LiF thermoluminescence dosimetry. Neutron dosimetry using LiF has been studied by Cameron et al.⁽⁴⁾ and Endres⁽¹⁶⁾ but much research work is still required for the development of a complete dosimetry system which can be used in the mixed fields of neutron and gamma-radiation.

ACKNOWLEDGEMENTS

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13. LITERATURE CITED

1. Spurny, Z., "Thermoluminescent dosimetry," At. En. Rev. Vol. 3, No. 2, 61 (1965).
2. "TL Dosimeter system, Models TL-3B and TL-3C", Operation Manual, #S-316-MN, EG and G Santa Barbara Division (1966).
3. Bräunlich, P. and Scharmann, A., "Dosimetry with the aid of LiF," Nukleonik 4, 65 (1962).
4. Cameron, J. R., Zimmerman, D. W. Kenney, G., Buch, R., Bland, R., and Grant, R., "Thermoluminescent radiation dosimetry utilizing LiF," Health Phys. 10, 25 (1964).
5. Schulman, J. H., "Survey of luminescence dosimetry," CONF-650637, AEC symposium series 8, 7 (1967).
6. Randall, J. T. and Wilkins, M. H. F., "Phosphorescence and electron traps," Pro Royal Sco. of London A 184, 366 (1945).
7. Bonfiglioli, G., Brovotto, P. and Cortese, C., "Thermoluminescence and F. Centers," Phys. Rev. 114, 951 (1959).
8. Hill, J. J. and Schwed, P., "Experimental study of the mechanism of thermoluminescence in irradiated NaCl," J. Chem. Phys. 23, 652 (1955).
9. Cameron, J. R., Zimmerman, D. W. and Bland, R. W., "Thermoluminescence Vs. roentgens in LiF: a proposed mathematical model," CONF-650637, AEC symposium series 8, 47 (1967).
10. Donnert, H. J., Klein, N. and Sasse, R. A., "The response of selected gamma radiation detectors to radiation delivered at very high dose rates," Proceedings of the Army Science Conference, U. S. Military Academy, West Point, New York (1964) (classified).
11. Zimmerman, D. W., Rhyner, C. R. and Cameron, J. R., "Thermal annealing effects on the thermoluminescence of LiF," Health Phys. 12, 525 (1966).
12. Morehead, F. F. and Daniels, F., "Thermoluminescence and coloration of LiF produced by α -particles, electrons, gamma-rays and neutrons," J. Chem. Phys. 27, 1318 (1957).
13. Pinkerton, A. P. and Laughlin, J. S., "Energy dependence of LiF dosimeters at high electron energies," Phys. Med. Biol. 11, 129 (1966).

14. Crosby, E. H., Admond, P. R. and Shalek, R. J., "Energy dependence of electrons at high energies," Phys. Med. Biol. 11, 131 (1966).
15. Karzmark, C. J., Fowler, J. F. and White, J. T., "Problems of reader design and measurement error in LiF thermoluminescent dosimetry," Int. J. App. Rad. Isotop. 17, 161 (1966).
16. Endres, G. W. R., "Thermoluminescent dosimetry studies at Hanford," AEC report BNWL-SA-23.
17. Dean, P. N. and Larkins, J. H., "Thermoluminescent dosimetry with activated LiF," AEC report LAMS-3034, 205 (1963).
18. Spinks, J. W. T. and Woods, R. J., "An introduction to radiation chemistry," John Wiley & Sons, Inc., New York (1964).
19. Hunter, J. S. "Response Surface Methodology," NDL-SP-1, Nuclear Defense Laboratory, Army Chemical center, Maryland (1962).
20. Bennett, C. A. and Franklin, N. L., "Statistical Analysis in Chemistry and the Chemical Industry," John Wiley & Sons, New York (1954).
21. Fryer, H. C., "Concepts and methods of experimental statistics," Allyn and Bacon, Inc., (1966).
22. McCall, R. C., Babcock, L. E. and Fix, R. C., "The LiF thermoluminescent dosimeter system for radiation research", Rad. Res. 19, 200 (1963).
23. Karzmark, C. J., White, J. and Fowler, J. F., "LiF thermoluminescence dosimetry," Phys. Med. Biol. 9, #3, 273 (1964).
24. May, C. E. and Partridge, J. A., "Thermoluminescent kinetics of α -irradiated alkali halides," J. Chem. Phys, 40, 1401 (1964).

APPENDIX A*

DOSE RATE EFFECT VS. REACTION KINETICS

Following is the reproduction of a qualitative theory developed by Donnert to analyse the dose rate effect on the response of dosimeters.

Any physical quantity which is a suitable measure of observable changes in a dosimeter can be used to characterize the response R of the dosimeter to ionizing radiation. This response R is most certainly a function of absorbed dose D , i.e., $R = R(D)$. This function, defined for $0 \leq D < \infty$, can be represented by converging power series expansions as

$$R = R(D) = \sum_{i=0}^{\infty} R_i D^i \quad (\text{A-1})$$

fitting the response curve by a polynomial where the expansion coefficients R_i are dose-independent, that is $\partial R_i / \partial D \equiv 0$ for $i \in \{0, 1, 2, \dots\}$. The expansion coefficients R_i are, however, functions of several other variables, which include a set of certain fixed parameters characterizing the bulk material properties of the dosimeter as well as the specific physical quantity chosen to represent the response R ; they also depend on the type of radiation quanta and their energy spectrum, which are here considered to be fixed parameters.

*Reproduced from ref. (10), which is classified. The theory presented in this appendix is, however, unclassified.

Most important, the expansion coefficients R_1 may also be functions of dose-rate $\theta = dD/dt$, which is a significant variable in the current investigation. This important fact can be inferred as a necessary consequence from a simple argument in reaction kinetics, considering the entirety of all processes in the dosimeter caused by the absorption of ionizing radiation. It suffices to examine this thermodynamic system for a quasi-stationary condition, which is realized by irradiation at some constant dose-rate θ , so that $d\theta/dt \equiv 0$. Detailed balancing of reaction rates yields a functional relationship between the rate of change in response, given by dR/dt , and the constant dose-rate θ , which can be represented as

$$\frac{dR}{dt} = F(D, \theta) \equiv [A(D) + f(D, \theta)]\theta \quad (\text{A-2})$$

The rate of change in response, that is dR/dt , will depend on several other variables as does the response R itself; most important, however, one must generally assume that dR/dt may be a function of the already prevailing response R of the dosimeter and, hence, of the adsorbed dose D previously received. In formula (A-2) linear and nonlinear terms in θ have been separated; thus, $A(D)$ is independent of θ , i.e., $\partial A/\partial \theta \equiv 0$. The quantity $f(D, \theta) \equiv 0$ must satisfy one of the two mutually exclusive conditions: either $f(D, \theta) \equiv 0$ (condition I) must hold, or both $f(D, \theta) \neq 0$ and $\partial f/\partial \theta \neq 0$ (condition II) must be met. In terms of an idempotent physical interpretation, condition I holds if and only if all processes in the system are first order reactions.

One can also represent dR/dt by a converging power series, which follows by differentiation* of the series (A-1) as

$$\frac{dR}{dt} \equiv \frac{\partial R}{\partial D} \frac{dD}{dt} = \left[\sum_{i=1}^{\infty} i R_i D^{i-1} \right] \theta \quad (\text{A-3})$$

The two expressions (A-2) and (A-3) for dR/dt must be identical. Consequently, the expansion coefficients R_i for $i \in \{1, 2, \dots\}$ are independent of θ if and only if condition I holds; furthermore, $\partial R/\partial D \equiv (dR/dt)/\theta = A(D)$ is independent of θ and the "integration constant" R_0 in a physically meaningful solution of this partial differential equation must also be independent of θ to satisfy the physical boundary condition for an unirradiated dosimeter, for which the response, given by $[R(D)]_{D=0}$, is certainly independent of θ . In this case, $\partial R_i/\partial \theta \equiv 0$ is satisfied for $i \in \{0, 1, 2, \dots\}$. By differentiation of the expression (A-1), which yields $\partial R/\partial \theta \equiv 0$, one corroborates readily that the response R is, in principle, not a function of dose-rate θ if and only if a dosimeter system can be characterized by condition I.

From a practical point of view, this necessary and sufficient condition can be relaxed to a less rigorous form. If all processes in a dosimeter system are only reactions of kinetically first order, the rigorous condition I is replaced by the weaker condition $A(D) \gg f(D, \theta)$; although the response R is then,

* In this theory all operations indicated for infinite series can be proven as mathematically rigorous.

in principle, a function of dose-rate θ , as implied by condition II, actual observation of the phenomenon is precluded because the dose-rate effect is not of sufficient magnitude to be practically significant.

For a dosimeter system governed by condition II the set of expansion coefficients R_i for $\{i\} \equiv \{1,2,\dots\}$ cannot be independent of the dose-rate θ ; in this case $\partial R_i / \partial \theta \neq 0$ must hold for $\{i\} \equiv \{1,2,\dots\}$. Differentiation of the expression (A-1) yields now $\partial R / \partial \theta \neq 0$; this implies that the response $R = R(D,\theta)$ is, indeed, a function of two significant parameters, absorbed dose D as well as dose-rate θ . This phenomenon should be actually observable unless $A(D) \gg f(D,\theta)$, which precludes that the dose-rate effect might be of negligible magnitude.

A further conclusion can be inferred for dosimeter systems exhibiting a dose-rate dependent response $R = R(D,\theta)$ which, necessarily, must satisfy $\partial R / \partial \theta \neq 0$. In order to assure that the dosimeter response $R = R(D,\theta)$ is, in fact, a function of absorbed dose D , it is necessary and sufficient that $\partial R / \partial D \neq 0$ holds. One infers readily from formulae (A-2) and (A-3) that

$$\frac{\partial R}{\partial D} = A(D) + f(D,\theta) \neq 0 \quad (\text{A-4})$$

where, pursuant to condition II, $f(D,\theta) \neq 0$ and $\partial f / \partial \theta \neq 0$ must hold; this ascertains, obviously, that the premise $\partial R / \partial D \neq 0$ is satisfied. Differentiation of equation (A-4) yields $\partial / \partial \theta (\partial R / \partial D) = \partial f / \partial \theta \neq 0$. For physically reasonable functions $R = R(D,\theta)$, one may certainly assume the two differentiations as interchangeable,

hence,

$$\frac{\partial}{\partial D} \left[\frac{\partial R}{\partial \theta} \right] = \frac{\partial}{\partial \theta} \left[\frac{\partial R}{\partial D} \right] = \frac{\partial f}{\partial \theta} \neq 0 \quad (\text{A-5})$$

This is a necessary and sufficient condition for the existence of interaction between the two variables D and θ , that is $R = R(D, \theta) \neq R'(D) + R''(\theta)$. One must, therefore, conclude that any dose-rate effect in a dosimetry system is, at least in principle, a dose-dependent phenomenon. This does, however, not necessarily imply that this phenomenon is of observable magnitude.

Simplest mathematical equation for response R which will satisfy condition (A-5) will be

$$R = R_{00} + R_{10}D + R_{01}\theta + R_{11}D\theta \quad (\text{A-6})$$

with a non-zero R_{11} coefficient.

APPENDIX B

Table 4. Data from high dose and high dose rate irradiation on Electron Linear Accelerator

Pulse Width μsec.	Beam Current mA.	Total Charge nC	Total Dose krad	Dose rate krad/μsec	Package No.	Dosimeter No.	Response (Glow peak height)	Equivalent dose from ⁶⁰ Co γ-rays
0.4	130	52.43	9.86	24.44	3	1*	1540	5500
						2	1300	
						3	730	
0.4	130	47.66	8.96	24.44	4	1*	2070	7010
						2	1410	
						3	900	
0.4	130	42.9	8.07	24.44	5	1*	1990	6820
						2	1470	
						3	790	
0.4	130	52.43	9.86	24.44	6	1*	1570	5800
						2	1170	
						3	730	
1.2	130	152.52	28.68	24.44	7	1	8000	
						2	5010	
						3	3170	
1.2	130	152.52	28.67	24.44	8	1	3900	
						2	2480	
						3	1290	

Table 4 (continued)

1.2	130	157.29	29.57	24.44	9	1	2190	
						2	920	
						3	560	
1.2	130	162.06	30.47	24.44	10	1	3460	
						2	1800	
						3	1210	
0.4	120	47.66	8.96	22.56	11	1	1410	
						2	920	
						3	700	
0.4	120	47.66	8.96	22.56	12	1	1680	
						2	1230	
						3	680	
0.4	120	42.9	8.07	22.56	13	1*	2030	6900
						2	1590	
						3	1130	
0.4	120	47.66	8.96	22.56	14	1	1500	
						2	1110	
						3	670	
1.2	120	138.23	25.99	22.56	15	1	3290	
						2	1780	
						3	1010	
1.2	120	138.23	25.99	22.56	16	1	3180	
						2	2500	
						3	1610	

Table 4 (continued)

1.2	120	138.23	25.99	22.56	17	1	2320
						2	1500
						3	900
1.2	120	133.46	25.09	22.56	18	1	3100
						2	1710
						3	1030
0.4	80	39.08	7.35	15.04	19	1	500
						2	406
						3	247
0.4	80	41.94	7.88	15.04	20	1	950
						2	710
						3	500
0.4	80	40.03	7.53	15.04	21	1	500
						2	381
						3	260
0.4	80	38.13	7.17	15.04	22	1	650
						2	500
						3	461
1.2	80	89.61	16.83	15.04	23	1	1850
						2	1480
						3	880
1.2	80	99.13	18.64	15.04	24	1	890
						2	500
						3	320

Table 4. (continued)

1.2	80	95.32	17.92	15.04	25	1	840	
						2	500	
						3	371	
1.2	80	97.23	18.28	15.04	26	1	880	
						2	505	
						3	380	
4.0	80	263.1	49.46	15.04	28	1*	9700	26300
						2	8400	
						3	5700	
4.0	80	278.35	52.33	15.04	29	1*	8400	22900
						2	7000	
						3	5000	
4.0	80	278.35	52.33	15.04	30	1*	8100	22000
						2	5600	
						3	3190	
4.0	80	259.20	48.74	15.04	31	1*	8000	21800
						2	7000	
						3	5000	
1.2	40	45.76	8.6	7.52	32	1*	1040	4200
						2	990	
						3	820	
1.2	40	43.85	8.24	7.52	33	1*	1000	4100
						2	830	
						3	830	

Table 4 (continued)

1.2	40	51.48	9.67	7.52	34	1	730	
						2	500	
						3	253	
1.2	40	47.66	8.96	7.52	35	1*	1000	4100
						2	900	
						3	770	
4.0	40	154.43	29.03	7.52	36	1*	4370	12700
						2	2310	
						3	1000	
4.0	40	158.24	29.75	7.52	37	1*	4330	12600
						2	3520	
						3	2420	
4.0	40	156.33	29.39	7.52	38	1*	3750	11200
						2	3490	
						3	1760	
4.0	40	151.66	28.51	7.52	39	1*	3370	10200
						2	2180	
						3	880	
4.0	20	72.76	13.68	3.76	40	1	1900	6400
						2*	1810	
						3	1350	
4.0	20	80.57	15.15	3.76	41	1	1800	
						2*	1790	6300
						3	1610	

Table 4 (continued)

4.0	20	78.99	14.85	3.76	42	1*	1990	6820
						2	1790	
						3	960	
4.0	20	78.99	14.85	3.76	43	1*	1760	6250
						2	1300	
						3	880	
-	-	-	-	-	44†	1	0	
						2	0	
						3	0	
4.0	10	31.2	5.87	1.88	45	1	454	
						2	500	
						3	500	
4.0	10	35.54	6.68	1.88	46	1	505	
						2	530	
						3	620	
4.0	10	31.2	5.87	1.88	47	1	500	
						2	503	
						3	503	
4.0	10	36.13	6.79	1.88	48	1	530	
						2	540	
						3	485	
-	-	-	-	-	49†	1	0	
						2	0	
						3	0	

†Control samples

*Dosimeter selected for data analysis

EFFECTS OF VERY HIGH DOSE RATES ON THE
RESPONSE OF LiF THERMOLUMINESCENT DOSIMETERS

by

MOHAMMED KAISERUDDIN

B. E., Osmania University, India, 1965

AN ABSTRACT OF A MASTER'S THESIS

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ABSTRACT

Theory of thermoluminescence and the thermoluminescent dosimetry was reviewed. Based upon experimental results the pre-irradiation procedure considered suitable was heating the dosimeters at 400°C for 1 hr. Partial annealing after irradiation at 110°C for 7 min was found experimentally to help in removing the fading effects on dosimeter response, and hence was employed. A calibration curve for the EG and G model TL-21 LiF thermoluminescent dosimeters in the dose range 0-4 x 10⁴ rad of ⁶⁰Co gamma-rays at low dose rate was obtained.

To obtain dose rates of the order of 10¹⁰ rad/sec the Electron Linear Accelerator at the Argonne National Laboratory was used. 14 MeV electron beam was used to irradiate the dosimeters. Total doses of the order of 10⁴ rad were supplied in single pulses of widths of the order of μsec.

Data was analysed by a regression procedure. The conclusions drawn were: (1) the response of LiF dosimeters is dependent upon the dose rate of delivered radiation in the dose rate range of approximately 10⁹ - 3 x 10¹⁰ rad/sec and in the dose range of about 6 - 50 krad; (2) the dose rate effect is a dose dependent phenomenon; and (3) the kinetic order of the processes involved in the irradiation of LiF is other than unity.