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The IRS Thermoluminescent Dosimetry System

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March, 1993

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Abstract

We have established a system consisting of a reader and two annealing ovens for measuring absorbed dose using thermoluminescent detectors (TLDs). This report describes the principal characteristics of the system as well as the results of measurements using LiF and CaF₂:Mn TLDs. The reader is a Victoreen model 2800M using an electrically heated planchet and a variable gain photomultiplier for integrating the light output from the TLD. We have carried out measurements which tests the performance of the reader from its detection limit to saturation. The TLDs are annealed in conventional, small volume laboratory ovens which have been equipped with auxiliary temperature controllers. We describe those characteristics of the anneal cycle which have an important impact on TLD performance. We have measured the supralinearity of LiF TLDs up to an absorbed dose of about 20 Gy, and have studied the effect of accumulated dose on the sensitivity. We report results of measurements of absorbed dose due to ⁶⁰Co γ -rays covering almost six decades (30 μ Gy to 24 Gy) using LiF. Typically, the standard uncertainty on the mean of a set of five measurements is less than 0.5%. Finally, we give some preliminary results obtained using CaF₂:Mn TLDs.

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1 Introduction

Thermoluminescent dosimeters (TLDs) are widely used for the measurement of absorbed dose. They have many useful features, such as small size, wide dynamic range and dose-rate independence. They also have a number of disadvantages including a complicated anneal cycle to erase the past radiation history, nonlinear response for high absorbed doses, loss of sensitivity with accumulated dose and variations in the sensitivity depending on the details of the anneal cycle. Although there are many materials which exhibit thermoluminescence, one of the most extensively studied is LiF. Its radiation absorption characteristics are similar to those of water, making it well suited for applications in personnel monitoring, radiation biology and radiation therapy. A second material which has received fairly wide application is $\text{CaF}_2\text{:Mn}$. Since its effective atomic number (16.2) is twice that of LiF, it is not so well suited for applications where tissue equivalence is an advantage. However, it is more sensitive than LiF, less susceptible to radiation damage and its response is more nearly linear for large absorbed doses.

In order to have available within our group the ability to use TLDs for various dosimetric purposes, we have purchased a Victoreen model 2800M TLD reader. Because of the importance of the anneal cycle in establishing TLD sensitivity, we have carefully characterized both a high and a low temperature oven for use in annealing the TLDs. Section 2 describes the main characteristics of the reader while section 3 gives the thermal characteristics of the annealing ovens. In section 4 we give the results of our measurements of some of the characteristics of LiF as a thermoluminescent material, including its nonlinear response for large absorbed doses. Section 5 gives the details of the operating procedure we have developed

for use with LiF. We have used LiF TLDs to measure absorbed dose over a wide range and we report on these results in section 6. Finally, in section 7 we report on some preliminary results regarding the use of $\text{CaF}_2\text{:Mn}$ as a thermoluminescent material.

2 The TLD Reader

The Victoreen model 2800M TLD reader is a manual unit with a circular drawer fitted with three different chambers positioned 90° apart. The photomultiplier is mounted vertically above the drawer so that its photocathode views the chamber which is at the top. The first chamber contains the heated planchet in which TLD chips are placed. The second chamber contains a stable reference light consisting of a scintillator and a radioactive source. The final chamber is for reading bulbs containing TLD powder.

The electronics are configured to integrate the current produced by the photomultiplier for a fixed time period. Charges from a few pC to a maximum of $45\text{ }\mu\text{C}$ can be measured, permitting the measurement of absorbed dose over several orders of magnitude. The dynamic range can be extended by several more orders of magnitude by varying the gain of the photomultiplier. It is also possible to have neutral density filters installed in front of the photomultiplier if very large light outputs are to be measured. However, these must be installed at the factory.

Figure 1 shows how the integrated current¹ from the reference light changes with the high voltage applied to the photomultiplier. The minimum voltage that can be applied is 400 V, while the gain saturates at about 1300 V. Over this voltage range the gain has changed by

¹Note that the $45\text{ }\mu\text{C}$ limit can be exceeded when reading the reference light.

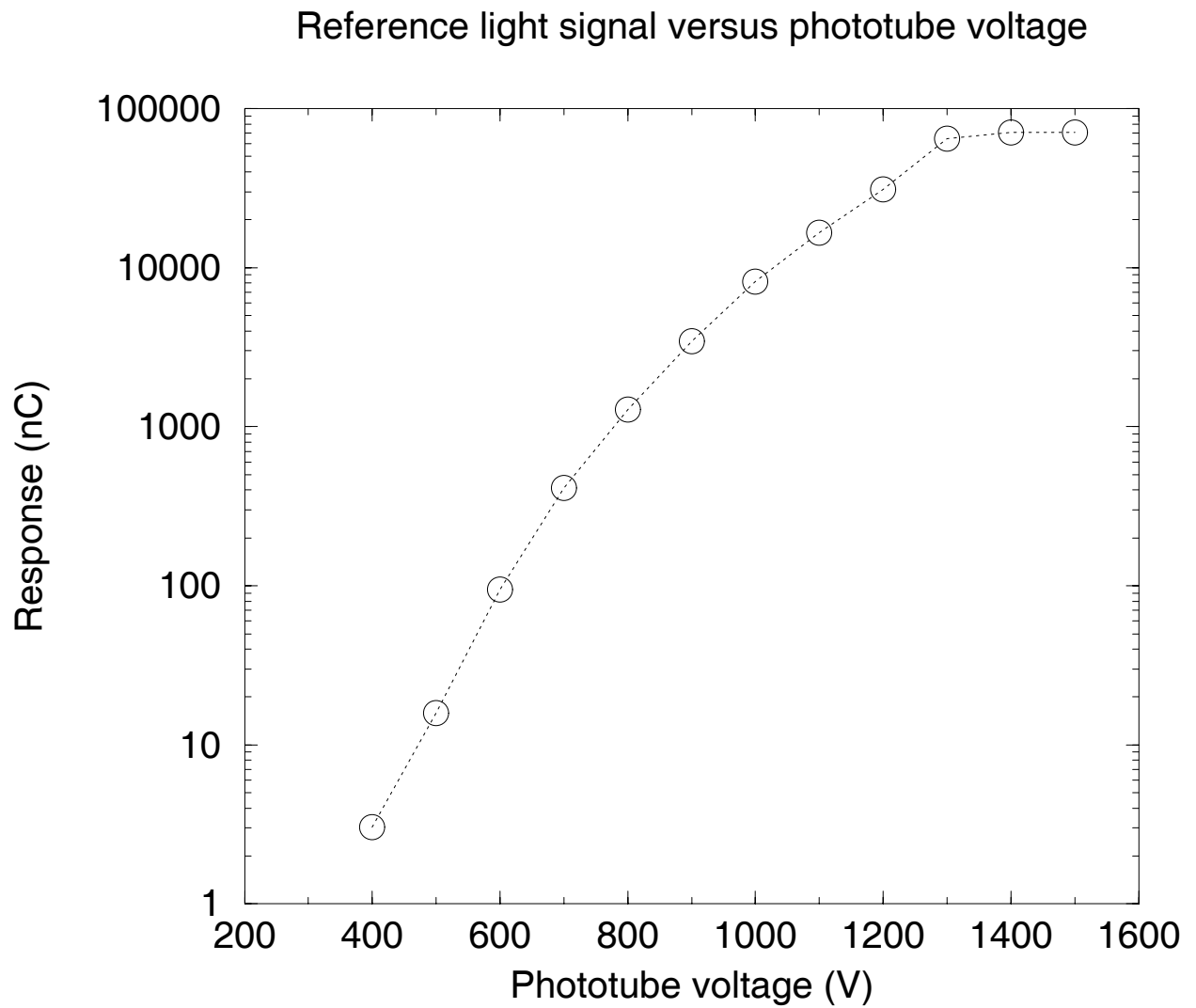


Figure 1: Measured photomultiplier signal as a function of the voltage on the base when the reference light is in position. The current has been integrated for 39 seconds to get the response in nanocoulombs. The maximum photomultiplier gain is achieved at about 1300 V.

more than four orders of magnitude.

Figure 2 shows how the background signal from the heated planchet changes with the photomultiplier gain. These data were obtained for the heating cycle we use for reading LiF TLDs (see table 1) so that the maximum planchet temperature was 240°C. At low gain the background is almost constant, suggesting that it is dominated by the background signal from the photomultiplier itself.

Figure 3 shows the signal to noise ratio when the signal is the reference light output. The data in figure 3 were obtained by taking the ratio of the data in figure 1 to those in figure 2. In practice, one chooses an operating voltage which gives a reasonable output signal from the TLDs so that signal-to-noise only becomes a limitation near the detection limit. Figure 3 is somewhat different than the equivalent figure shown by Kasper *et al* (1992). Their graph shows the signal to noise ratio increasing up to about 800 V after which it decreases as the high voltage is increased. On the other hand, for our reader the signal to noise ratio increases monotonically with voltage until the maximum gain is reached at about 1300 V.

The heating cycle to be used in measuring the TLD glow curve is characterized by four parameters. The first is the preheat temperature, T_p , and is the temperature the planchet should achieve as quickly as possible. The second is the heating rate, dT/dt , and is the rate at which the temperature should rise to its maximum value. The third parameter is the maximum temperature the planchet should achieve, T_m , and the final parameter is the total readout time, t_r . The reader displays the glow curve on a CRT using a maximum of 450 channels, each one representing 0.1 seconds. The glow curve can be smoothed by changing a parameter referred to as the time constant. This parameter does not affect the value of the integrated current, and we have found that a setting of 10 gives a satisfactory display.

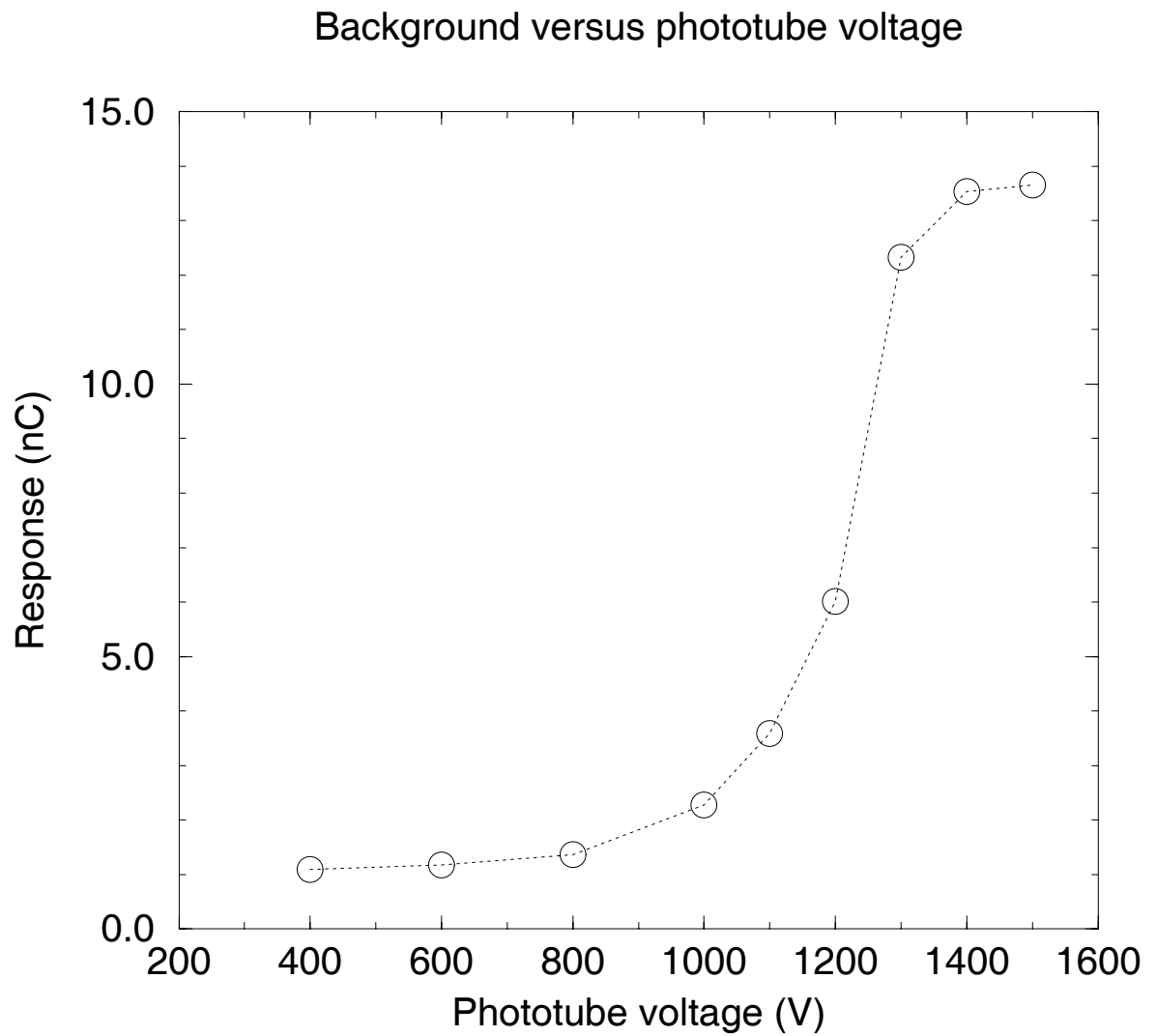


Figure 2: Measured background signal from the heated planchet as a function of the photomultiplier voltage. The LiF heating cycle was used (see table 1), with 240°C being the maximum planchet temperature.

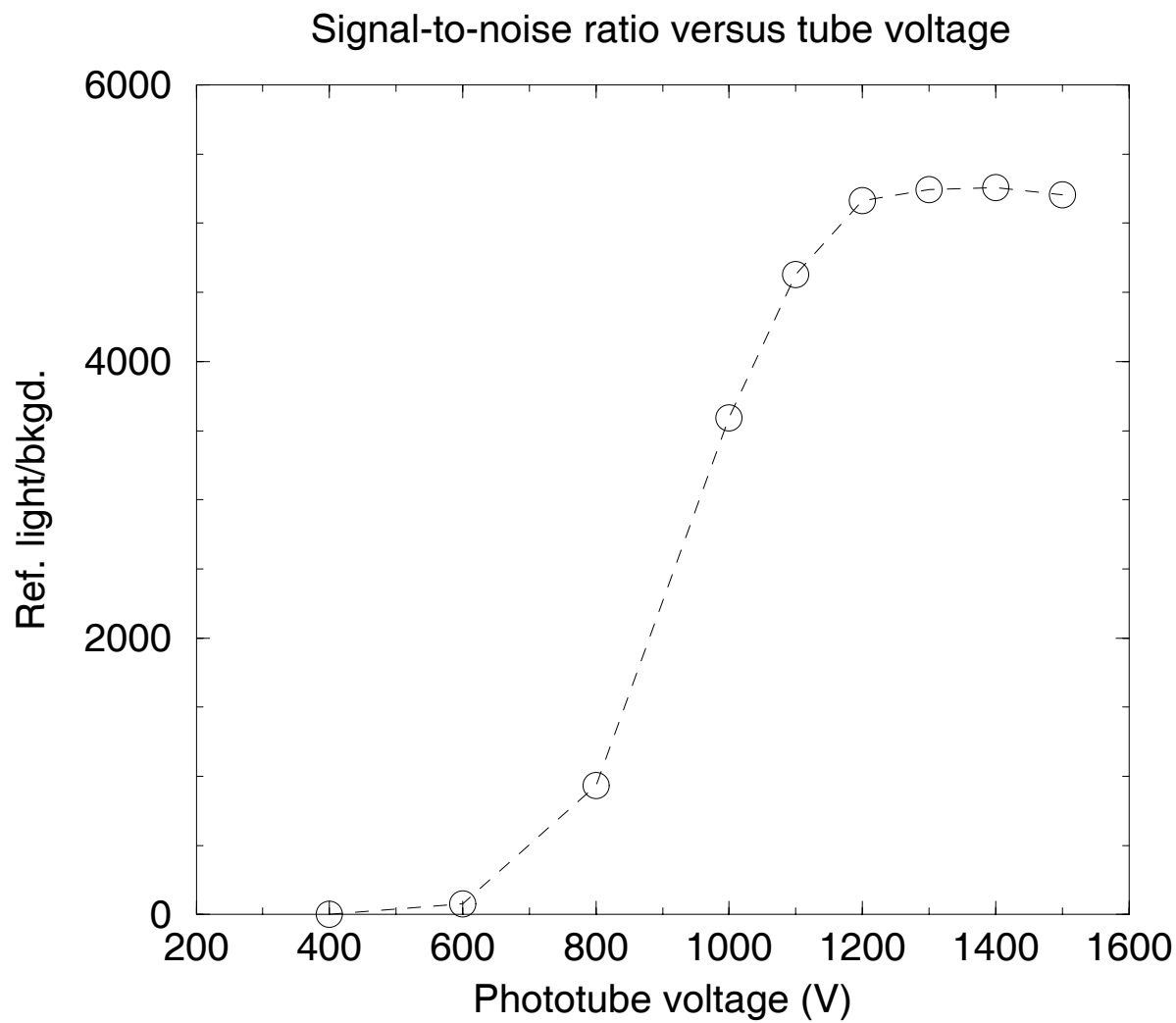


Figure 3: Signal-to-noise ratio calculated using the data shown in figures 1 and 2.

A region of interest can be selected from the total number of channels and the integrated current over this region will be displayed on the CRT. The parameters which we have used for reading LiF and CaF₂:Mn TLDs are given in table 1.

TLD material	T_p (°C)	dT/dt (°C/s)	T_m (°C)	t_r (s)	ROI (channels)
LiF	100	5	240	45	41-430
CaF ₂ :Mn	200	10	400	45	40-350

Table 1: Reader settings used for generating LiF and CaF₂:Mn glow curves. The parameters are: T_p - preheat temperature; dT/dt - heating rate from the preheat temperature to the maximum; T_m - maximum temperature during readout; t_r - total readout time; ROI - region of interest, or portion of the 450 channels over which the phototube current is integrated. The time constant associated with the display of the glow curve was set to 10.

We use the inlet provided with the reader to maintain a flow of nitrogen over the planchet whenever TLDs are being read. Measurements indicated that the planchet background is slightly smaller with nitrogen present. Only a few measurements were carried out to compare the TLD response with and without nitrogen. These results indicated that the response was very similar whether or not nitrogen was present. After the read cycle is completed, the drawer is not opened until the planchet temperature has fallen below 40°C.

3 The Annealing Cycle

In order to recover the original TLD sensitivity after irradiation and readout it is necessary to anneal the TLDs. Unfortunately, the TLD sensitivity is dependent on the details of the

anneal cycle, so good control of the anneal cycle is important. Various annealing cycles have been proposed over the years, but there are two fairly widely accepted procedures (Driscoll *et al* (1986), Horowitz (1990)). We have chosen the one which consists of a high temperature anneal at 400°C for one hour followed by a low temperature anneal at 100°C for two hours. The TLDs are rapidly cooled from 400°C to room temperature by placing them on a large brass block immediately after removal from the high temperature oven. The TLDs are placed in quartz holders for annealing.

On attempting to use LiF TLDs to measure absorbed doses in the range of 10 Gy we found an enhancement in the TLD sensitivity, even though the standard anneal procedure had been followed. We discovered that the problem was because the TLDs in the high temperature oven were not at 400°C even though the probe which controlled the oven temperature indicated that the air temperature was 400°C. The TLDs (in their quartz holder) were placed on a large steel plate resting on the bottom of the oven. Using thermocouple probes we found, under some circumstances, that the temperature of the steel plate was at least 50°C below the air temperature.

After the oven is turned on it is important to wait for the plate to come to thermal equilibrium. Figure 4 shows the approach to thermal equilibrium in the high temperature oven from a cold start. The air temperature reaches 400°C after about 20 minutes but at this time the temperature of the plate is only about 200°C. It takes about two hours for the steel plate to reach the air temperature.

We also encountered temperature nonuniformities in our low temperature oven. In this case the heating element is on the bottom of the oven and we found that the plate on which the TLDs were placed was hotter than the air temperature. We added a fan to provide

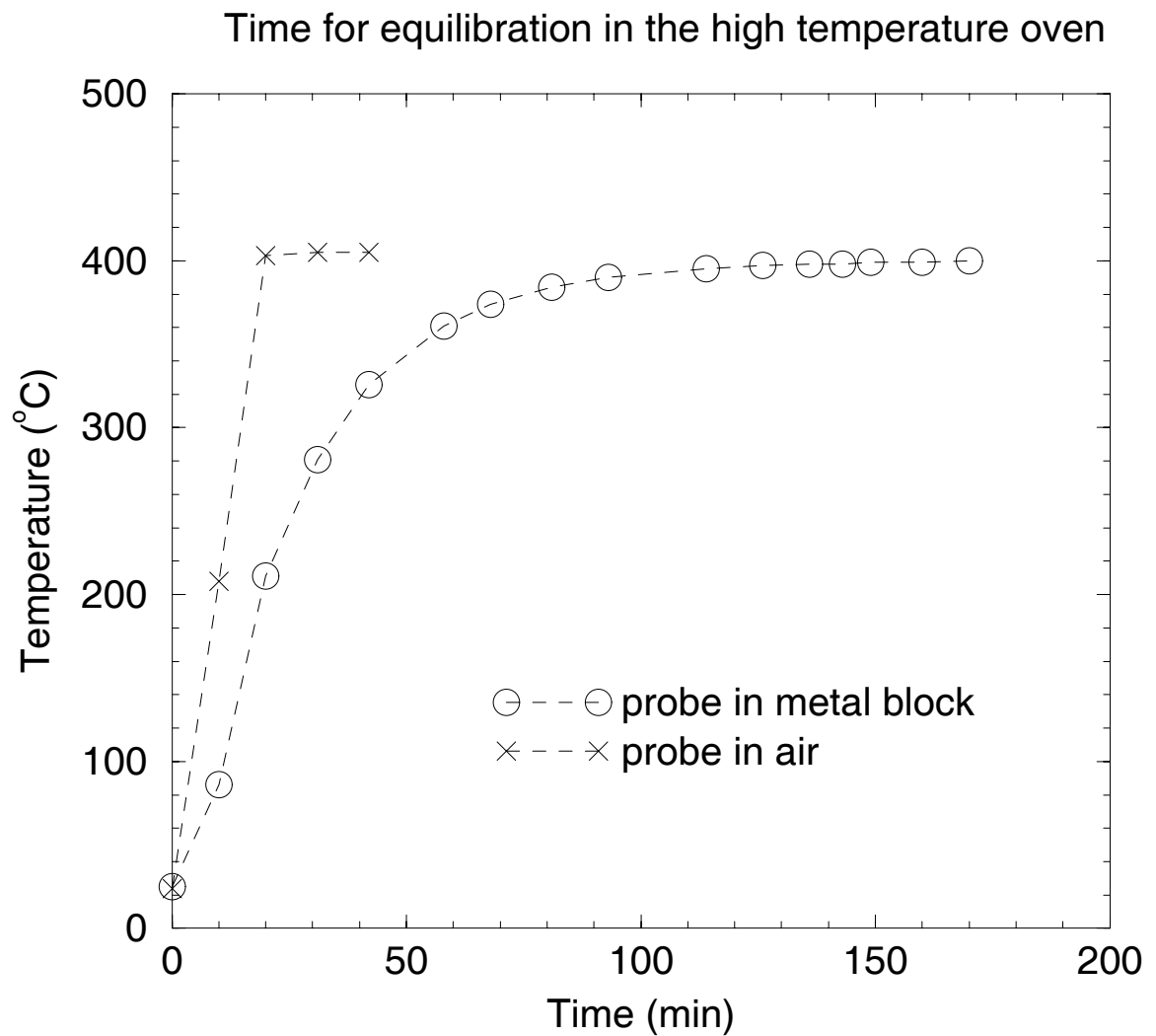


Figure 4: Approach to thermal equilibrium in the high temperature (400°C) oven. The probe which is used in the control loop is mounted in air, and shows that the air temperature has stabilized after about 20 minutes. However, the large steel block on which the TLDs are placed requires about 2 hours to reach thermal equilibrium.

convection with the result that the plate temperature is close to the air temperature. Figure 5 shows the approach to thermal equilibrium in the low temperature oven from a cold start. The air is at 100°C after about 10 minutes but it takes about 60 minutes for the temperature of the plate to stabilize. Both the high and low temperature ovens are now equipped with thermocouple probes attached to the plates on which the TLDs are placed. This way, the temperature in the immediate vicinity of the TLDs can be checked before they are placed inside.

Another important parameter in the anneal cycle is the cooling rate after the TLDs are removed from the high temperature oven. Upon removal from the high temperature oven we immediately place the TLDs (in their quartz holders) on a large brass block at room temperature. Figure 6 shows the cooling rate as measured using a small thermocouple probe.

Horowitz (1990) has recently examined the effects of various aspects of the anneal cycle on LiF TLD sensitivity. His data show that the sensitivity, if glow peaks 4 and 5 are read, is independent of the cooling rate above about $100^{\circ}\text{C}/\text{minute}$. Figure 6 shows that our initial cooling rate is about $400^{\circ}\text{C}/\text{minute}$, well above the minimum recommended by Horowitz.

4 Characteristics of LiF TLDs

This section describes various measurements which were carried out to characterize LiF as a TLD material. Since LiF has been extensively studied, all of the characteristics which we have examined have also been discussed in the literature. However, there is often considerable variation in the results reported by different investigators. In some cases the differences are

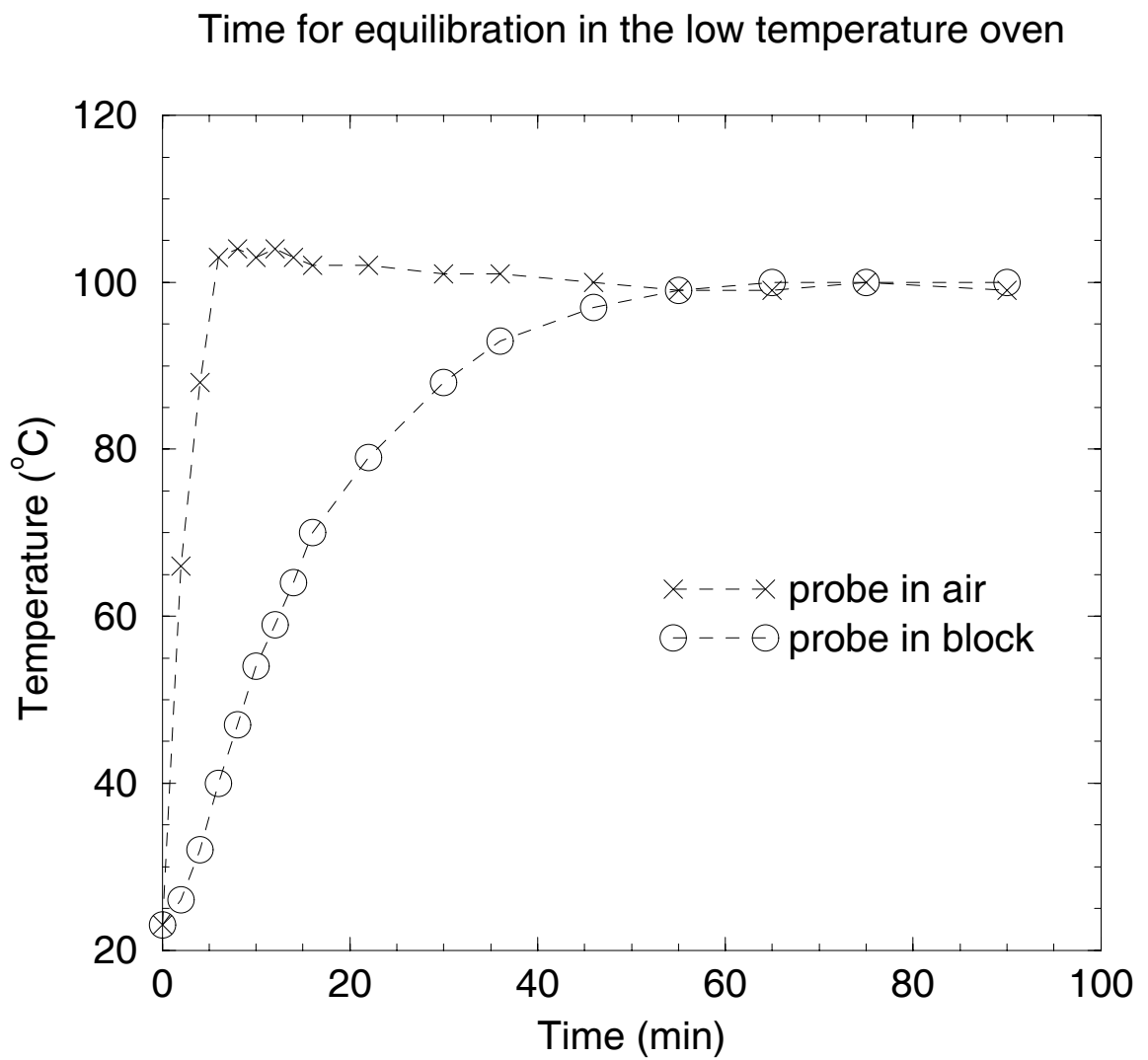


Figure 5: Same as figure 4 but for the low temperature (100°C) oven. In this case, the block on which the TLDs are placed reaches thermal equilibrium after about one hour.

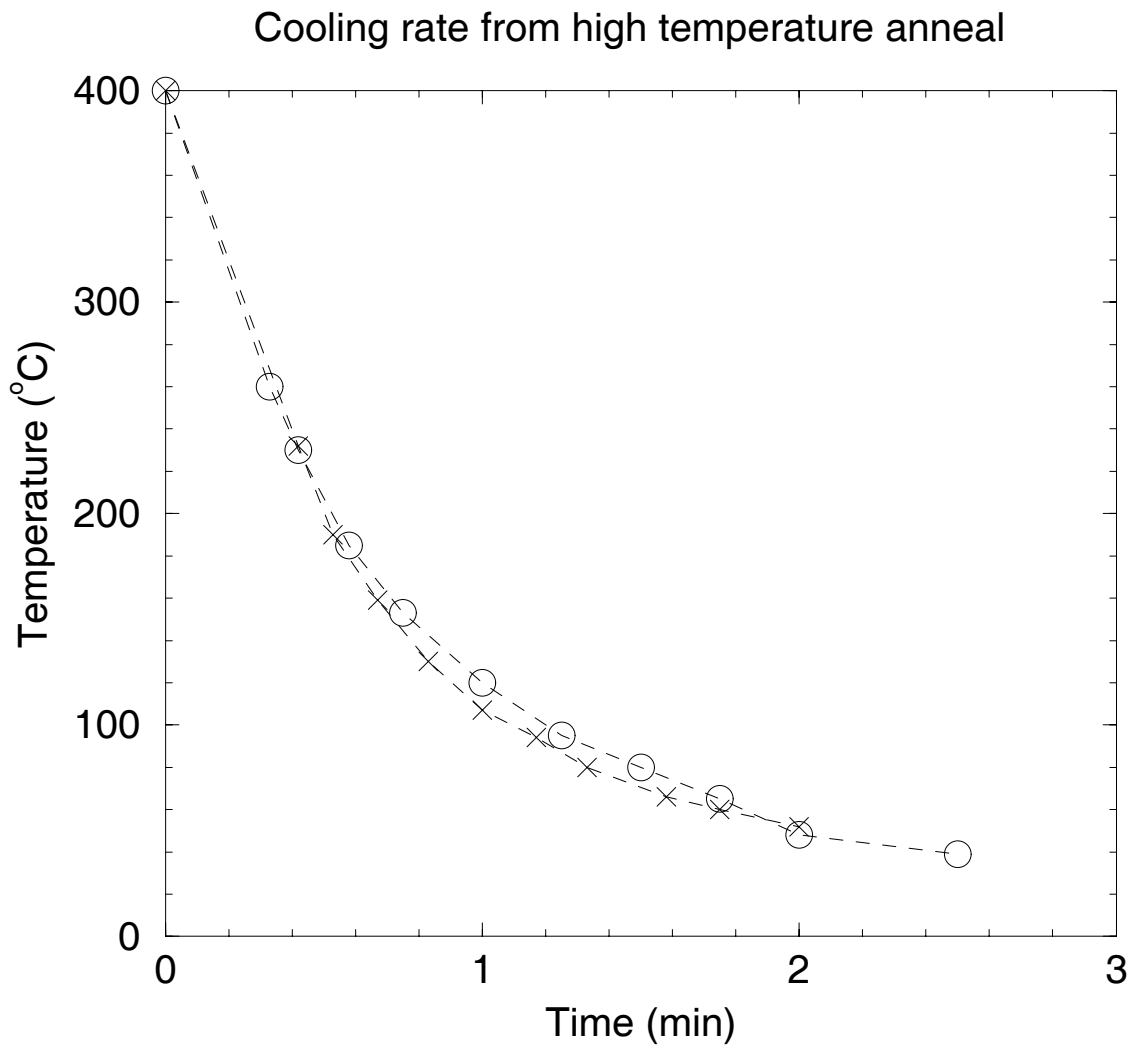


Figure 6: Approximate cooling rate of the TLDs after they are removed from the high temperature oven and placed on a large brass block. The two data sets represented by different symbols correspond to two measurements of the cooling rate.

not important, but in the case of the supralinearity, it is important to have measurements which accurately characterize our system.

All of the irradiations were carried out using ^{60}Co γ -rays. The TLDs were irradiated in a Lucite phantom to guarantee charged particle equilibrium. The phantom consisted of two Lucite plates, 5.8 cm by 7.6 cm. In one plate, which was 7 mm thick, twenty five circular holes were drilled. Each hole was just large enough to hold one TLD chip. The second plate, which was 6 mm thick, was held on top of the first with nylon screws. Various dose rates were obtained by using the several exposure-calibrated measurement geometries on both the low and high intensity ^{60}Co sources. The dose to the LiF chip, D_{LiF} , was related to the exposure, X , using the relation given by Attix (1986)

$$D_{LiF} = 0.802X, \quad (1)$$

where D_{LiF} is in cGy and X is in R.

4.1 Sensitivity versus Mass

The LiF chips used in this work have dimensions of 3.2 mm by 3.2 mm by 0.89 mm, and a mass of about 24 mg. The spread in the masses for a set of 25 chips is about 4%, but figure 7 shows that variations in the chip sensitivity (about 17%) are not correlated with variations in the mass.

4.2 Sensitivity versus High Temperature Anneal

As pointed out in section 3 an important early objective of this work was to determine why the TLDs showed an enhanced sensitivity after receiving an absorbed dose of several

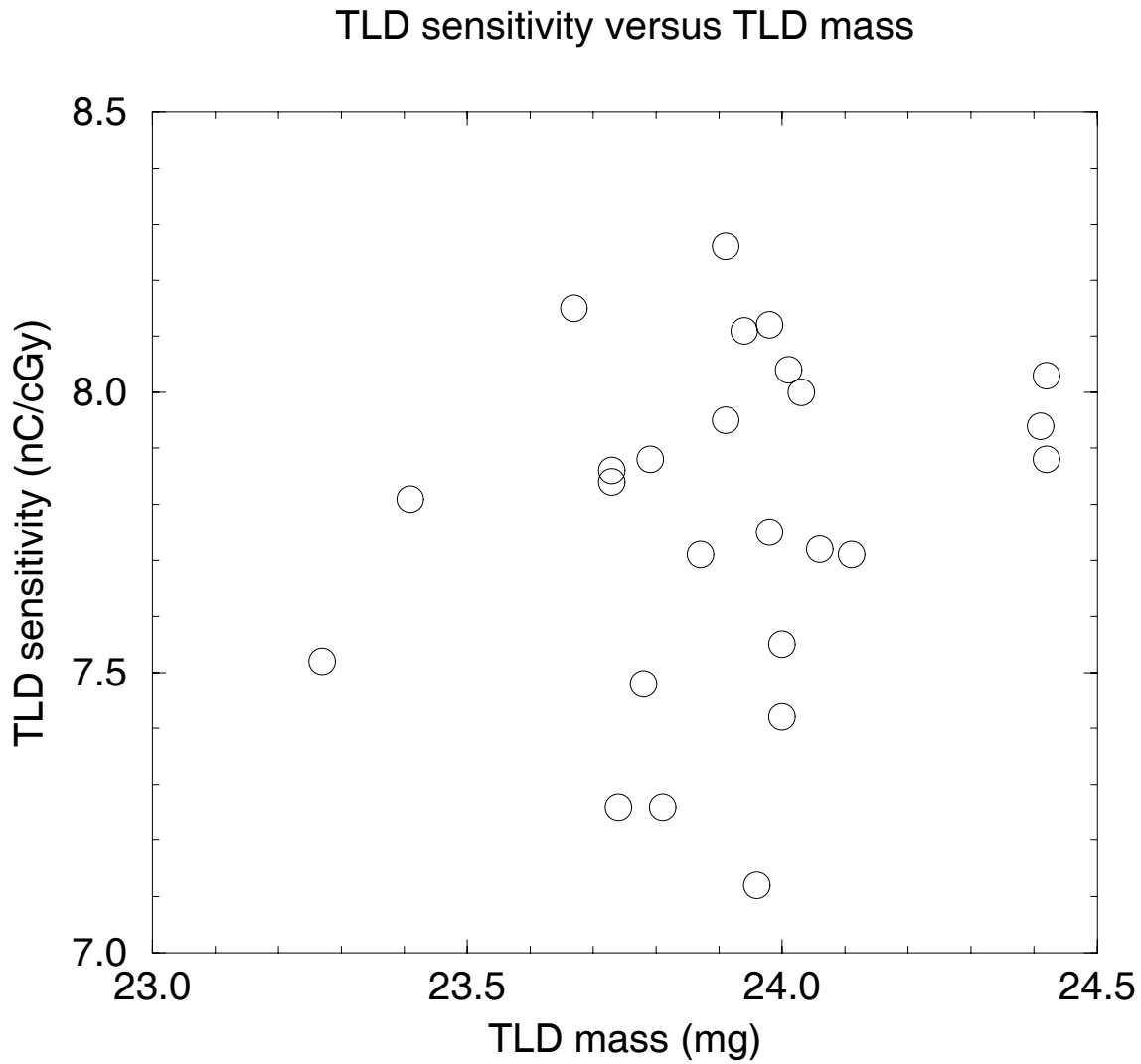


Figure 7: The sensitivity of LiF chips versus the mass of the chip. The observed variation in sensitivity is not correlated with the mass of the chip. The measured sensitivity corresponds to that obtained when the photomultiplier gain is set so that the reference light output is 100 nC.

Gy. The problem turned out to be poor temperature control in the high temperature oven, resulting in the TLDs being annealed at a temperature significantly below 400°C. Once good temperature control had been achieved, more detailed measurements were carried out to investigate the effect of the high temperature anneal on TLD sensitivity.

First the sensitivity was measured as a function of temperature for TLDs which received only a low dose (about 6.8 cGy). Figure 8 shows that for small absorbed doses the sensitivity begins to decrease rapidly above about 450°C and by 550°C it is down by 26%. However, for temperatures below about 400°C the sensitivity is constant. Next, the sensitivity was measured for TLDs which received larger absorbed doses. Batches of TLDs were first annealed using the standard anneal procedure, and their sensitivity measured using an absorbed dose of about 6.7 cGy. They were then given an absorbed dose of either 8 Gy or 24 Gy, read out, and annealed using the standard procedure except that the temperature of the high temperature oven was varied. Their sensitivity was again measured using an absorbed dose of about 6.7 cGy, and any change in sensitivity noted. The results are plotted in figure 9 and show a large enhancement in the sensitivity once the temperature of the high temperature oven is much below 380°C. This graph shows the importance of good temperature control for the high temperature oven, and it also shows that if large doses are to be measured, the temperature must not be much less than 400°C.

4.3 Sensitivity versus Anneal Cycles

Ogunleye *et al* (1987) have studied the effect of repeated anneal cycles on the sensitivity of LiF. They find a decrease of sensitivity which is approximately linear with the number of

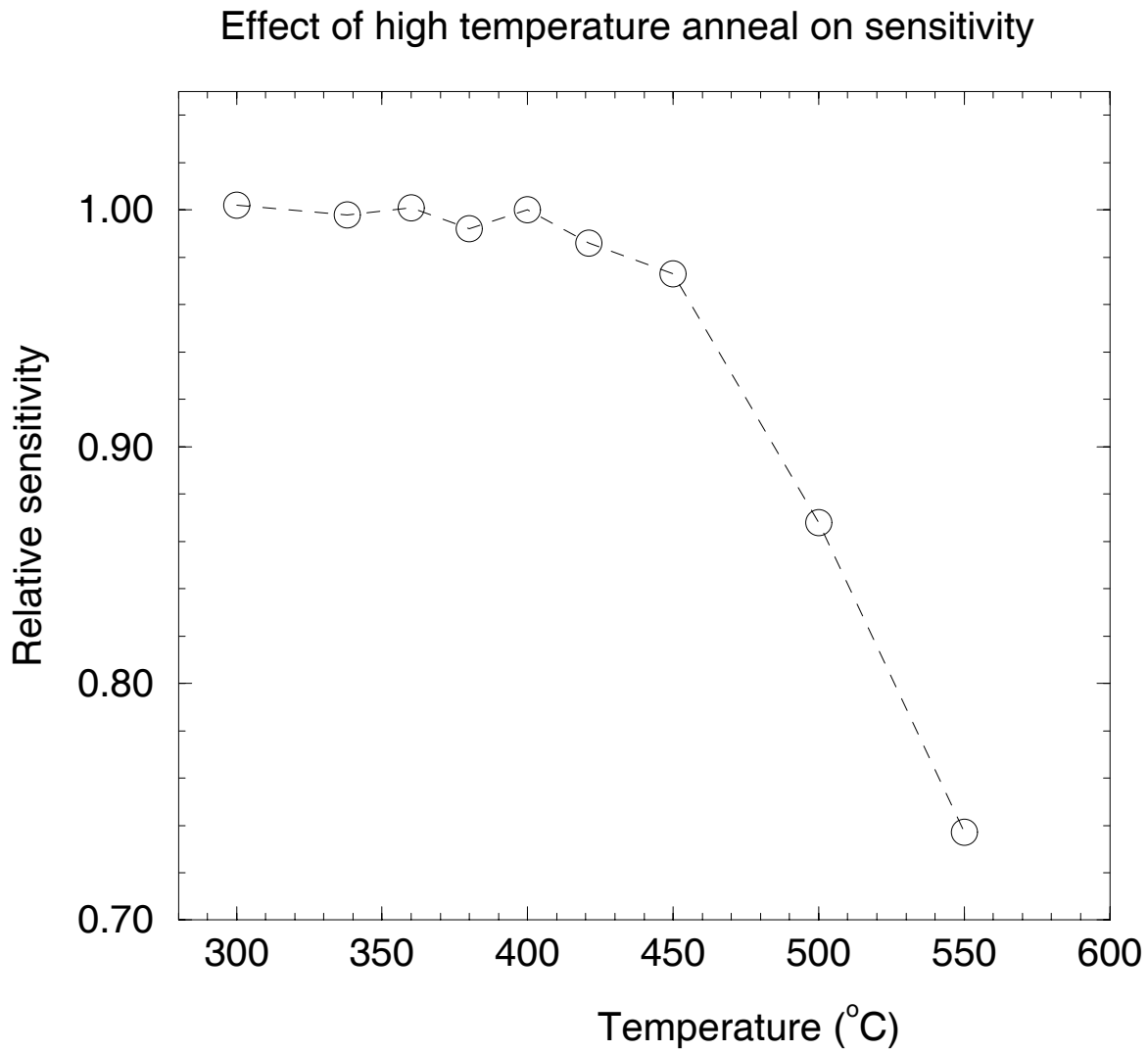


Figure 8: Effect of the high temperature anneal on the LiF sensitivity for TLDs which received only low doses. The sensitivity was calculated from the response obtained when the TLDs received an absorbed dose of about 6.8 cGy.

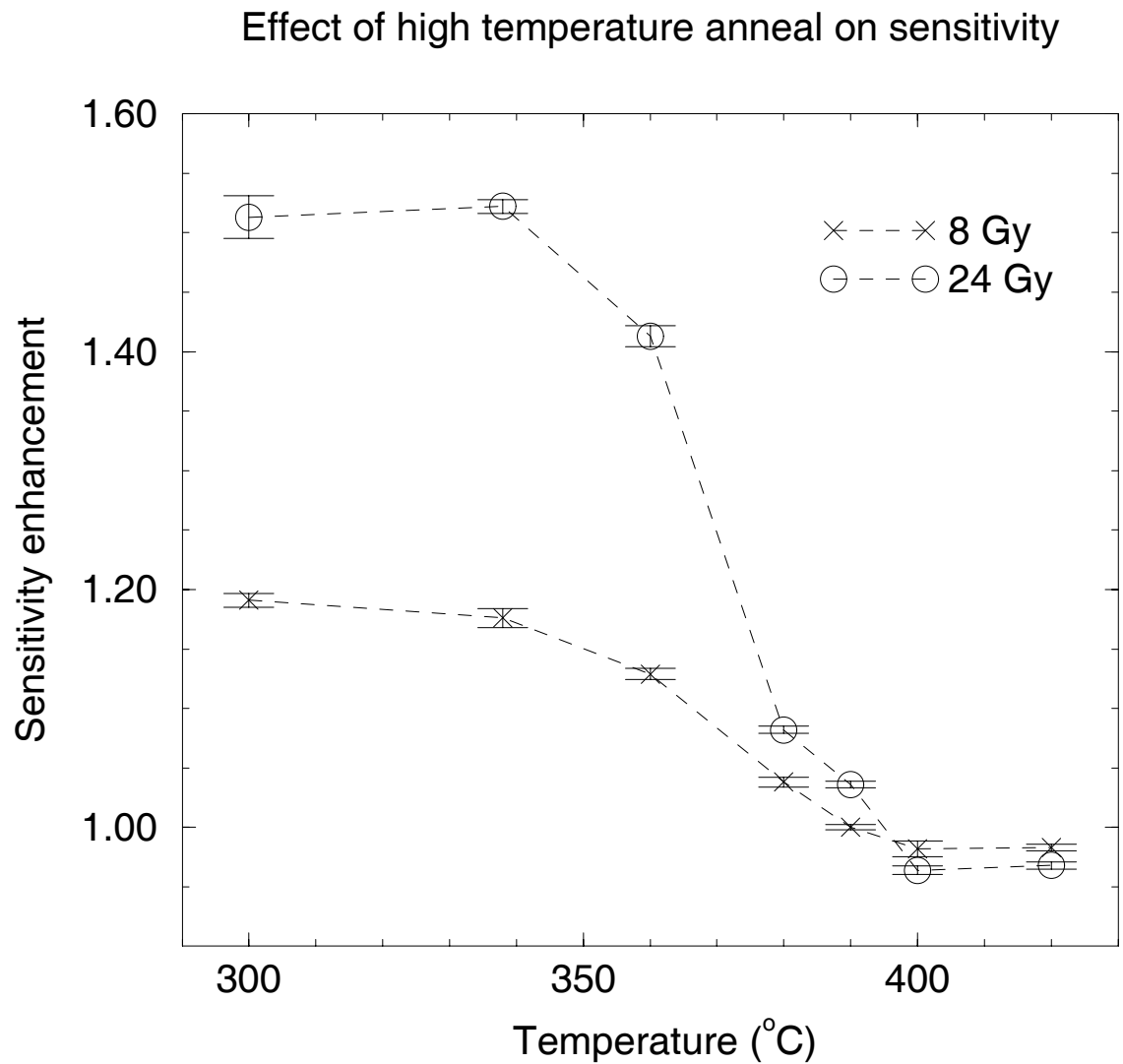


Figure 9: Data showing the effect of the high temperature anneal on LiF sensitivity for TLDs which received high doses. The reference sensitivity was obtained using the standard anneal cycle and a calibrating absorbed dose of 6.8 cGy. The sensitivity was again measured after the TLDs received a dose of either 8 or 24 Gy and were annealed at a temperature other than 400°C.

cycles and which amounts to about 15% after 100 cycles. We have not studied systematically the effect over a large number of anneal cycles, but figure 10 shows how the mean sensitivity of ten TLDs changed over eight cycles. The dashed line is a least squares fit to the data of a straight line, and is given by

$$S = -4.04(\pm 1.45)N + 2476(\pm 7), \quad (2)$$

where S is the sensitivity, N is the number of the anneal cycle and the numbers in parentheses are the standard uncertainties. Equation 2 predicts a loss of sensitivity of $0.16(\pm 0.06)\%$ per anneal cycle, in good agreement with the result of Ogunleye *et al.*

Various parameters associated with the anneal cycle affect the TLD sensitivity, with the cooling rate from the high temperature anneal being one of the most important. The scatter of the data in figure 10 about the straight line gives an indication of the extent to which uncontrolled variations in our anneal cycle introduces changes in the sensitivity. The maximum deviation from the line is about 0.5%, which indicates that changes in the TLD sensitivity due to variations in the anneal cycle are not likely to exceed 1%.

4.4 Supralinearity

It is well known that the response of LiF is nonlinear for high absorbed doses. Figures 11 and 12 show the response enhancement measured with our TLDs. In figure 11 the dose is plotted on a logarithmic scale to show how the supralinearity sets in at lower doses. In figure 12 the same data are plotted on a linear scale to better show the behaviour at high doses. The nonlinearity amounts to about 50% for a dose of 22 Gy. The dashed line passing through the data in figures 11 and 12 is a least-squares fit to the data of a function of the

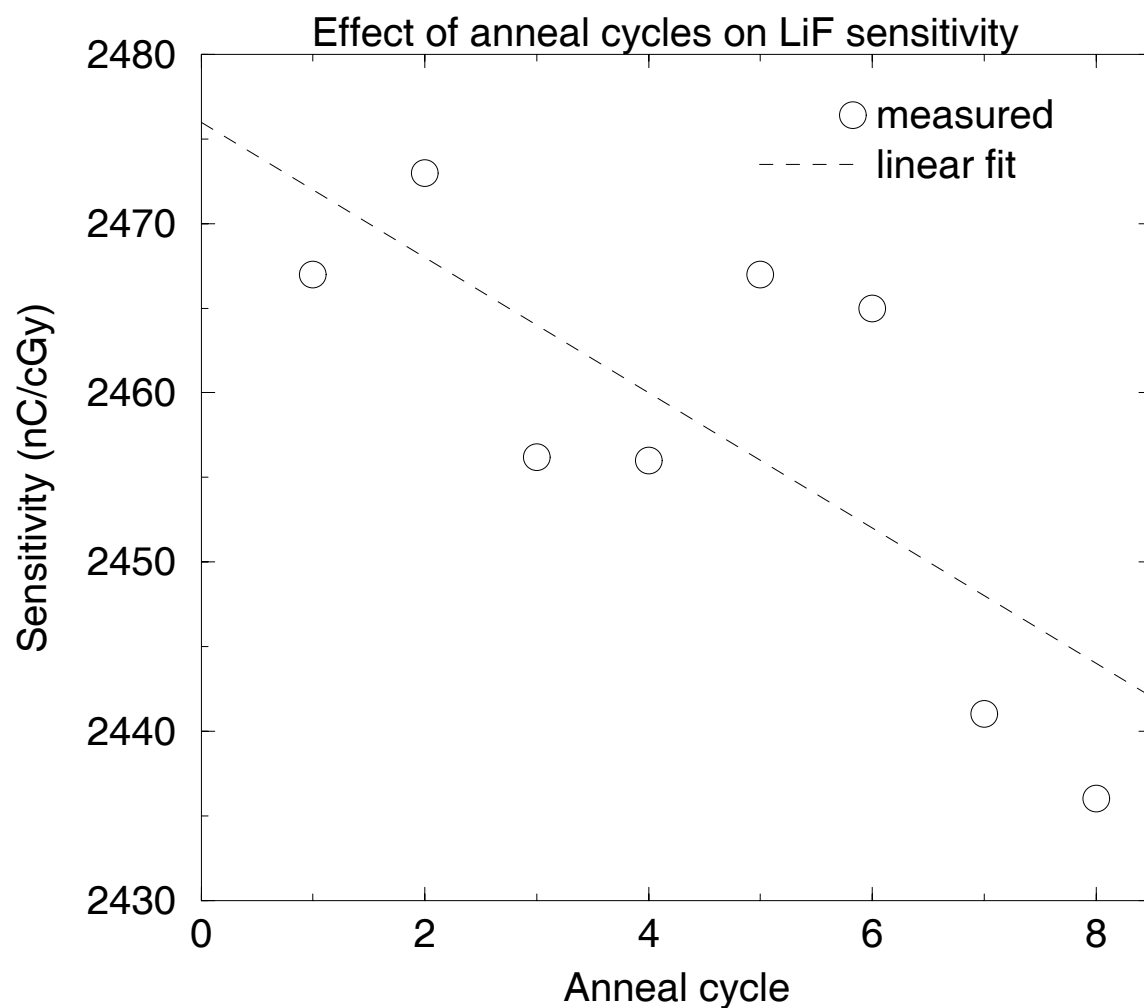


Figure 10: Effect of repeated anneal cycles on LiF sensitivity. The mean sensitivity of ten TLDs was followed for eight anneal cycles. The absorbed dose delivered per cycle was less than 1 cGy. The photomultiplier high voltage was set to 1200 V.

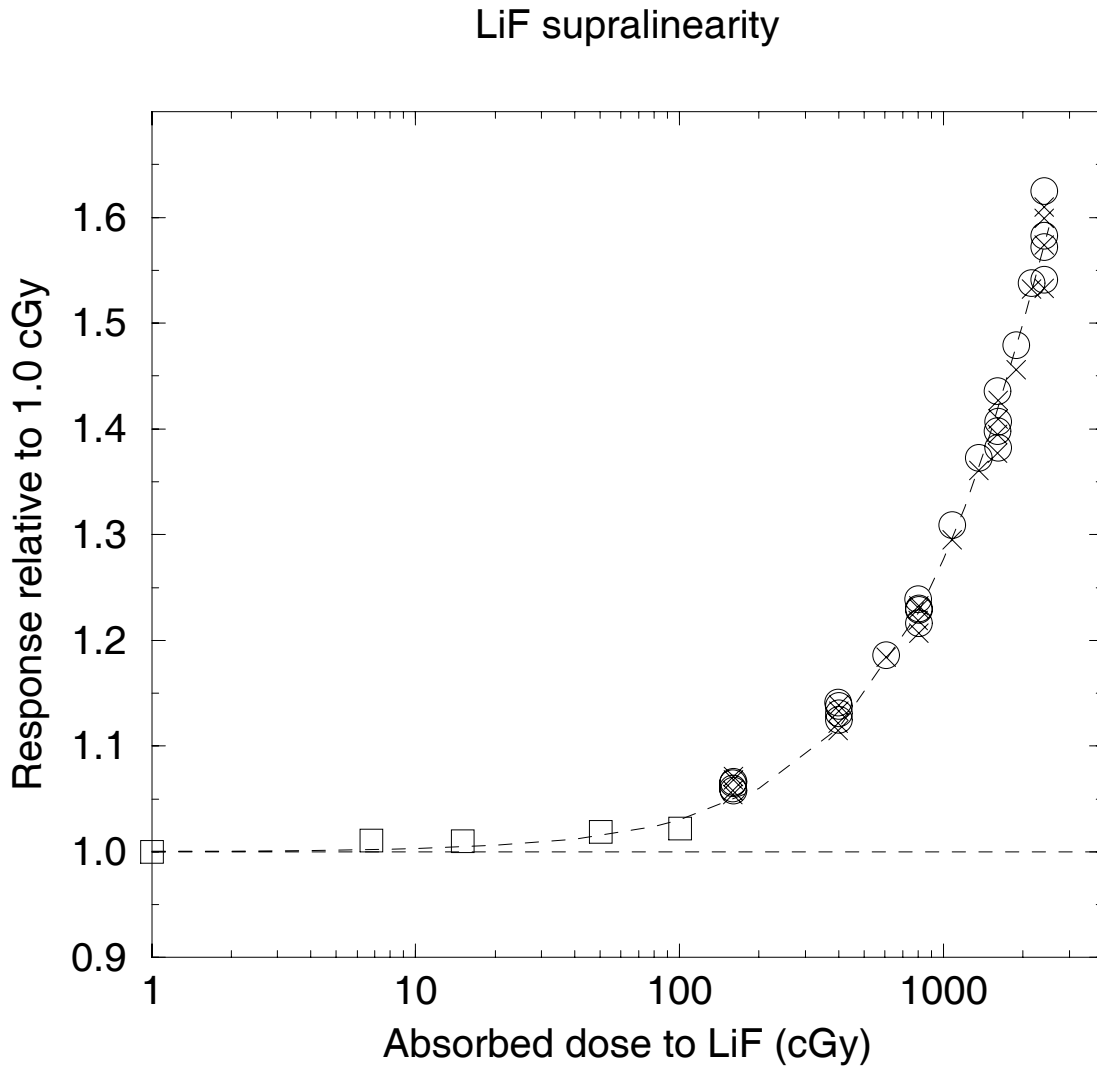


Figure 11: Measured response enhancement of LiF TLDs to high absorbed doses. The enhancement is measured relative to the response observed for a dose of 1.0 cGy. Three sets, each containing 25 TLDs, were used for the measurements and the data for each set are represented by a different symbol. Each datum point is the mean of four TLD readings. The dashed line through the data is a least-squares fit to a second-order polynomial (see text).

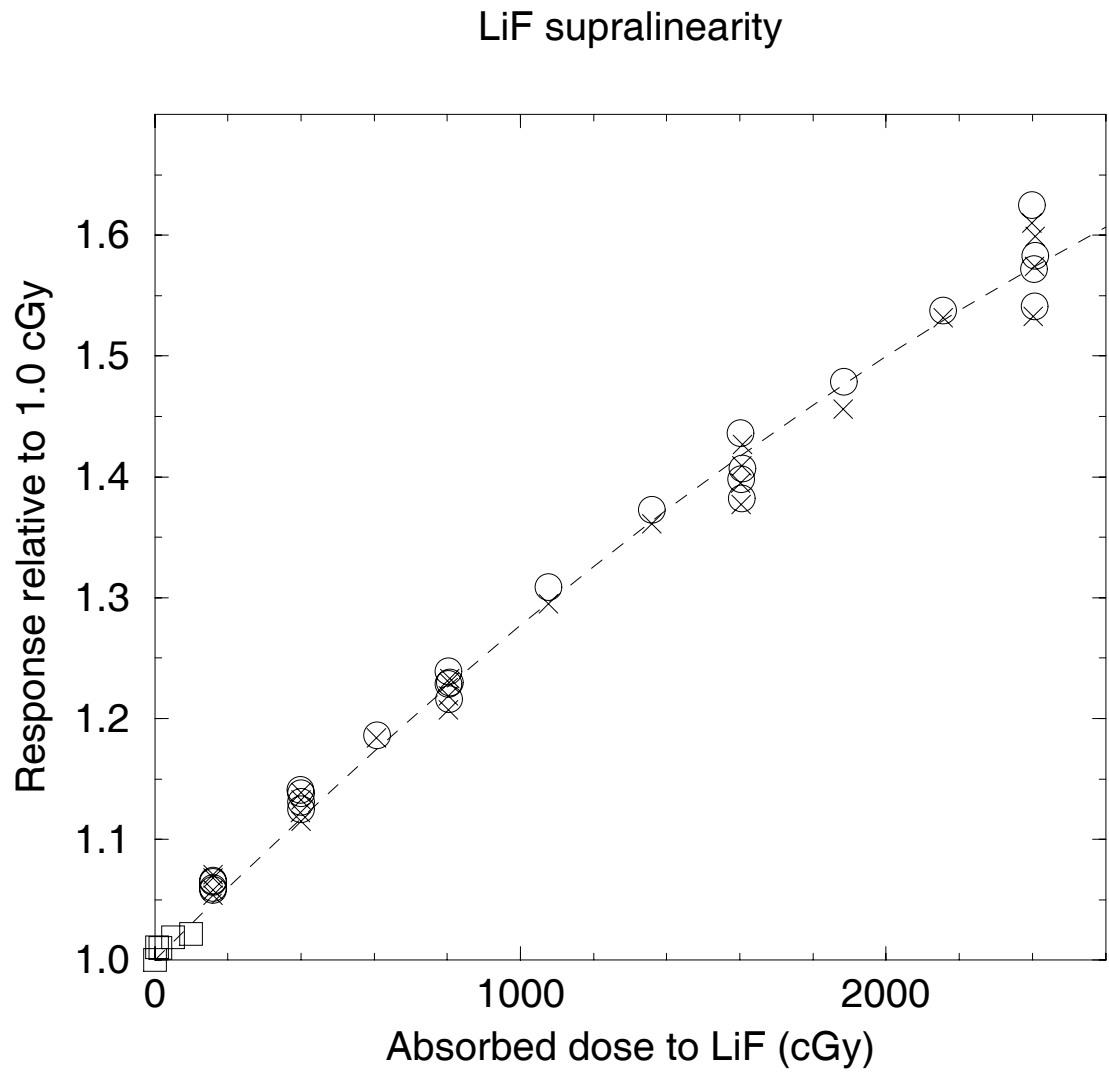


Figure 12: Same as figure 11 except that the absorbed dose scale is linear.

form

$$e_r(D) = 1.00 + aD + bD^2, \quad (3)$$

where $e_r(D)$ is the response enhancement, D is the absorbed dose to LiF in cGy and a and b are free parameters. Fitting the data gave $a = 3.044(\pm 0.072) \cdot 10^{-4}$ and $b = -2.73(\pm 0.35) \cdot 10^{-8}$ where the standard uncertainties on a and b are given within parentheses.

There is an interesting relationship between the supralinearity and the maximum enhancement in sensitivity as the annealing temperature is lowered (figure 9). For both 8 and 24 Gy, the measured enhancement is numerically very similar to the measured supralinearity for the same absorbed dose.

4.5 Sensitivity versus Accumulated Dose

Marrone and Attix (1964) have measured how the sensitivity of LiF changes for absorbed doses up to 10 MGy. They observed that the loss of sensitivity was more severe if the dose was delivered in several fractions rather than in one fraction. They speculate that the high temperature anneal tends to fix (i.e., make permanent) the damage. Our measurements are summarized in figure 13 for absorbed doses up to 100 Gy. This figure shows a larger loss of sensitivity if the absorbed dose is delivered in several fractions, consistent with the observation of Marrone and Attix.

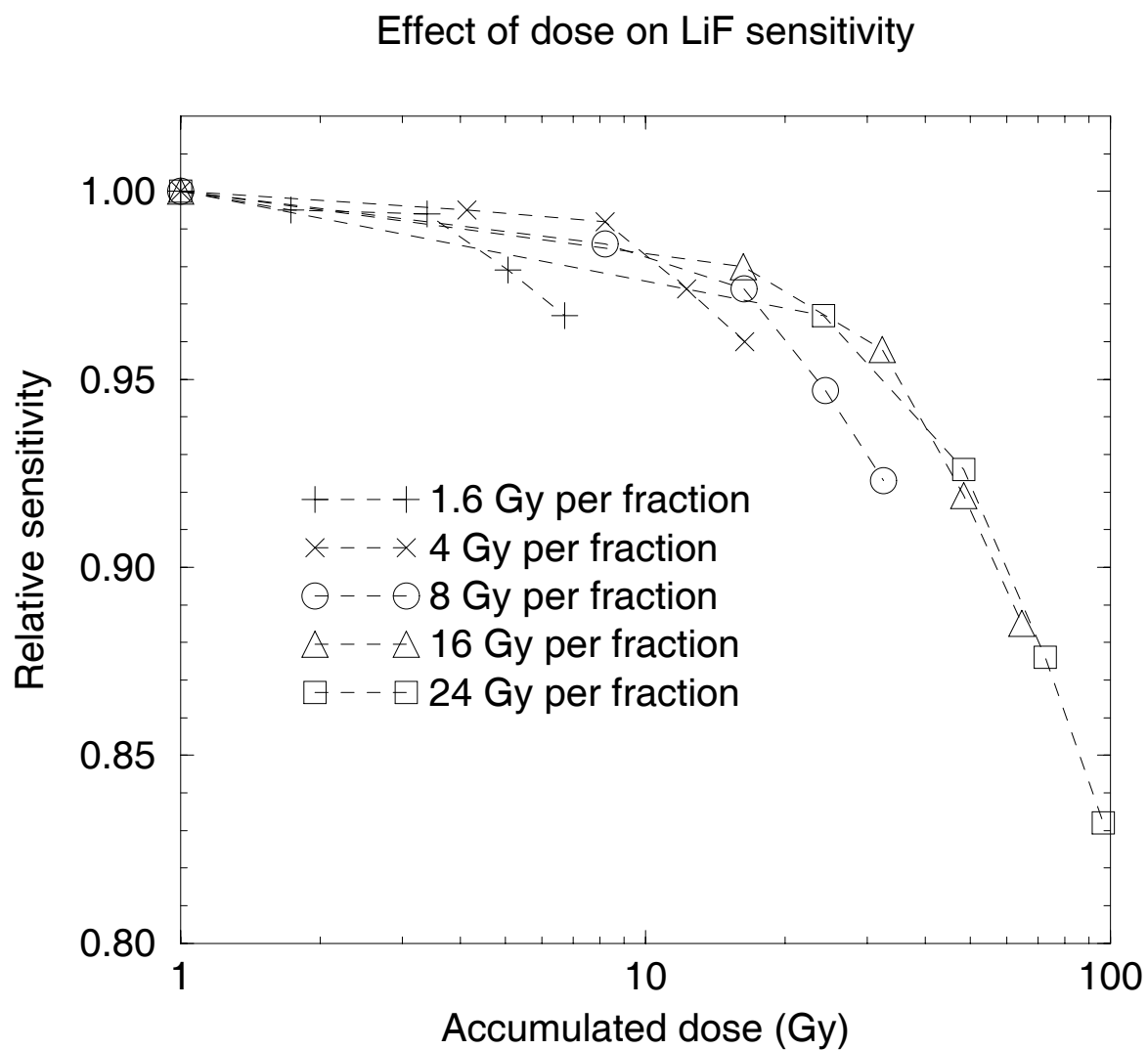


Figure 13: Effect of accumulated dose on the sensitivity of LiF chips. Between each fraction the TLDs were taken through the standard anneal cycle.

4.6 Sensitized LiF

It is well known that the sensitivity of LiF can be increased by a factor of five or more by irradiating it to a high dose and annealing it at a low temperature (Horowitz (1984b)). Another interesting feature of sensitized LiF is that its response is reported to be almost linear with absorbed dose well into the region where normal LiF shows large supralinear effects. This could be a useful advantage for measurements of absorbed dose in the range of 1 Gy to 20 Gy.

We irradiated TLDs to about 1 kGy and then annealed them at 280°C for one hour followed by 100°C for two hours. The sensitivity was measured as a function of absorbed dose for doses from 0.1 Gy to 20 Gy. Figure 14 shows the sensitivity, normalized to the sensitivity at 0.1 Gy, as a function of absorbed dose. For normal LiF, by 20 Gy the sensitivity has increased by about 50% over its value for low doses (figure 12). In contrast, for sensitized LiF the sensitivity has decreased, but by only about 3% at 20 Gy. The dashed line in figure 14 is a least squares fit of a straight line to the measured data, and is given by

$$e_r(D) = 1.001(\pm 0.003) - 1.47(\pm 0.21) \times 10^{-5}D, \quad (4)$$

where $e_r(D)$ denotes the relative sensitivity (in analogy with equation 3), D is the dose to LiF in cGy and the numbers in parentheses are the standard uncertainties.

Jones (1980) has reported success in using sensitized LiF for use in personnel and environmental dosimetry. The fact that the sensitivity is approximately independent of absorbed dose is a definite advantage for high dose measurements. However, additional work is required to demonstrate that there are no other serious disadvantages introduced through sensitization.

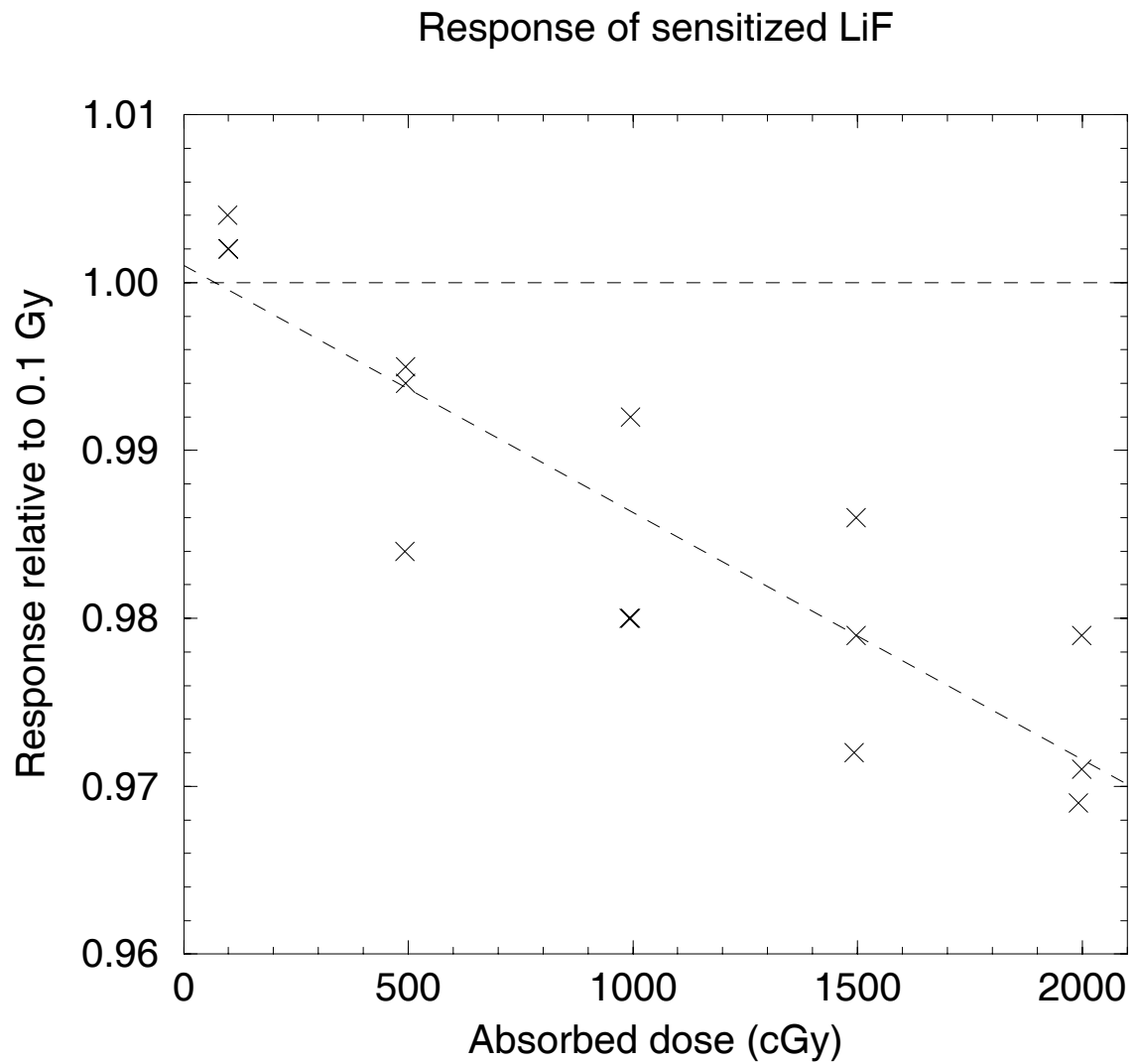


Figure 14: Sensitivity of sensitized LiF as a function of absorbed dose. The sensitivity at a given dose has been normalized to its value at 0.1 Gy. Each datum point corresponds to a single TLD measurement. The dashed line is a linear fit to the data.

5 The IRS Protocol for LiF

Section 4 shows that there are a large number of factors which influence TLD response. Even after many years of use, there is still debate as to the best technique to achieve optimum precision. Nevertheless, a useful guideline is that whatever protocol is adopted, it should be followed closely. We describe in some detail how we calibrate LiF TLDs and how we use them to measure an unknown absorbed dose.

We store and anneal the TLDs in quartz holders. Each holder has 25 circular depressions cut in its base, each large enough to accommodate one TLD. A quartz lid is placed over the base to protect the TLDs from the environment and to keep them in place.

In some early work we used aluminum holders. Although they seemed to work well, we changed to quartz because the literature accompanying Harshaw TLDs recommends against the use of aluminum. An important consideration is the mass of the holder if metal is to be used. Measurements on our high temperature oven show that it can take a long time for metal slabs to achieve thermal equilibrium at 400°C, possibly perturbing the anneal cycle. At one time we were transferring our quartz holders to and from the ovens on a metal base. We have abandoned this practice to reduce the equilibration times.

Immediately after the TLDs are removed from the 400°C, one-hour anneal they are placed on a large brass block, the lid is removed from the quartz holder and they are allowed to cool to room temperature. They are then transferred to the low temperature oven and annealed for two hours at 100°C. Upon removal from the low temperature oven they are again cooled on the brass block.

The TLDs are transferred to some appropriate holder for irradiation. We use vacuum

tweezers for moving the TLDs so that mechanical contact is minimized. For calibration purposes using ^{60}Co γ -rays the TLDs are placed in a small Lucite holder as described in section 4. Depending on the radiation levels being read, due attention must be paid to the absorbed dose contributed by naturally occurring background radiation. This amounts to about $2.5\ \mu\text{Gy}$ per day, so that after a few days it is well above the detection limit of the reader.

After irradiation, the TLDs are annealed for 12 minutes at 100°C . This step is generally recommended to eliminate the low energy traps which are subject to fading.

The reader cycle used for LiF has been described in Section 2. A nitrogen flow is maintained in the vicinity of the planchet so that the heating occurs in an inert environment. The planchet area is much larger than the area of the TLD chips we use. We position the TLDs at the centre of the planchet, but according to Victoreen the measured response is insensitive to the placement of the TLD. The TLD is allowed to cool in the inert environment to less than 40°C before it is removed and placed back in its quartz holder.

The light source built into the reader is used to check its overall performance but we do not use it to correct the measured TLD response. This is because the light output is known to be temperature dependent (Piesch (1981)) and is therefore influenced by the number and rate of read cycles. The background is also slightly dependent on the read cycle history, so for optimum precision it is best to bracket every five or so TLD readings with readings of the background. A background reading with a freshly annealed, unirradiated TLD in the planchet is about 10% greater than with the planchet empty. Unless the background is a significant fraction of the measured response, it is therefore adequate to use the empty planchet to estimate the background.

Because of uncontrolled variations in the anneal cycle it is difficult to recover the same TLD sensitivity after repeated anneal cycles. Horowitz (1990) has attempted to define an anneal cycle which minimizes variations in the sensitivity. However, we believe a more robust approach is to calibrate a subset of a group of TLDs all of which have gone through the same anneal cycle. We now describe in more detail how this is done, and how one corrects for supralinearity effects.

Suppose a set of n TLDs has been taken through the standard anneal cycle. To calibrate them, the set is then irradiated with a known absorbed dose in the dose region where the response is known to be linear (see figure 11). Let R_i be the reading of the i^{th} TLD, where R_i has been corrected for background and optionally normalized to some value of the reference light signal. Then the sensitivity, S_i , of each TLD can be calculated using

$$S_i = R_i/D_{LiF} \quad (i = 1, n), \quad (5)$$

where D_{LiF} is the calibration dose delivered to the set.

The same set of TLDs is now to be used to measure unknown doses. The TLDs are again taken through the standard anneal cycle. Rather than assuming that the sensitivity has not changed, we make the more relaxed assumption that the sensitivity of all the TLDs will have changed in the same way as a result of the anneal. We choose a subset of the larger set and use it to establish how the sensitivity of the whole set has changed.

Let there be m TLDs in the calibration set. The sensitivity of these TLDs will be given by

$$S'_i = R'_i/D'_{LiF} \quad (i = 1, m), \quad (6)$$

where the prime is used to distinguish this secondary calibration from the calibration of the

whole set as represented by equation 5. The ratio of the sensitivities, S'_i/S_i , will be denoted by ϵ_i and referred to as the TLD efficiency. A mean value, $\bar{\epsilon}$, can be calculated for the m TLDs, and we then assume that

$$S'_i = \bar{\epsilon}S_i \quad (i = m + 1, n). \quad (7)$$

If the j^{th} TLD is used to measure an unknown dose (in the linear response region) the dose will be given by

$$D_j = R'_j/S'_j = R'_j/(\bar{\epsilon}S_j). \quad (8)$$

Equation 8 can be generalized to include the effects of supralinearity by writing

$$e_r(D_j)D_j = R'_j/(\bar{\epsilon}S_j), \quad (9)$$

where $e_r(D_j)$ is the response enhancement shown in figures 11 and 12 and characterized empirically by equation 3. Equation 9 can be solved for D_j by iteration. The steps outlined in equations 5 through 9 can be implemented conveniently using a computer-based spreadsheet such as Excel by Microsoft.

In writing equation 5 it was assumed that the TLDs were calibrated in a dose range where they respond linearly. If this is not convenient, equation 5 can be generalized as

$$S_i = R_i/(e_r(D_{LiF})D_{LiF}). \quad (10)$$

If equation 10 is used to calculate the sensitivity, the result should be independent of the calibrating absorbed dose, D_{LiF} .

The TLD sensitivity, S_i , depends not only on the characteristics of the TLD, but also on the settings used with the reader, most notably the high voltage. The high voltage must

be adjusted so that the reading, R_i , is large enough to give adequate precision, but not so large as to saturate the reader. Table 2 gives the operating conditions for three values of the high voltage. These three settings are adequate to cover the entire dose range from the detection limit to about 30 Gy using standard LiF chips.

High voltage (V)	Approximate dose range (Gy)	Typical reference light output (nC)	Typical dose for calibration (Gy)	Typical response at calibration (nC)
600	1-30	90	0.07	50
900	0.01-1	3400	0.01	250
1200	3×10^{-5} -0.1 [†]	30,000	0.007	1500

[†] At the lower limit, the reader background equals the TLD response

Table 2: Three settings of the photomultiplier high voltage are adequate to cover the absorbed dose range from the detection limit to 30 Gy when using standard LiF chips. This table gives the dose range covered by each voltage setting, the approximate output from the reference light, a suitable absorbed dose for calibrating the TLDs, and the approximate TLD output at the calibration dose.

6 Absorbed Dose Measurements using LiF

6.1 Standard LiF

Figure 15 shows the background produced by an empty planchet for the standard LiF read cycle, and a high voltage of 1200 V. Figures 16 and 17 show typical LiF glow curves for

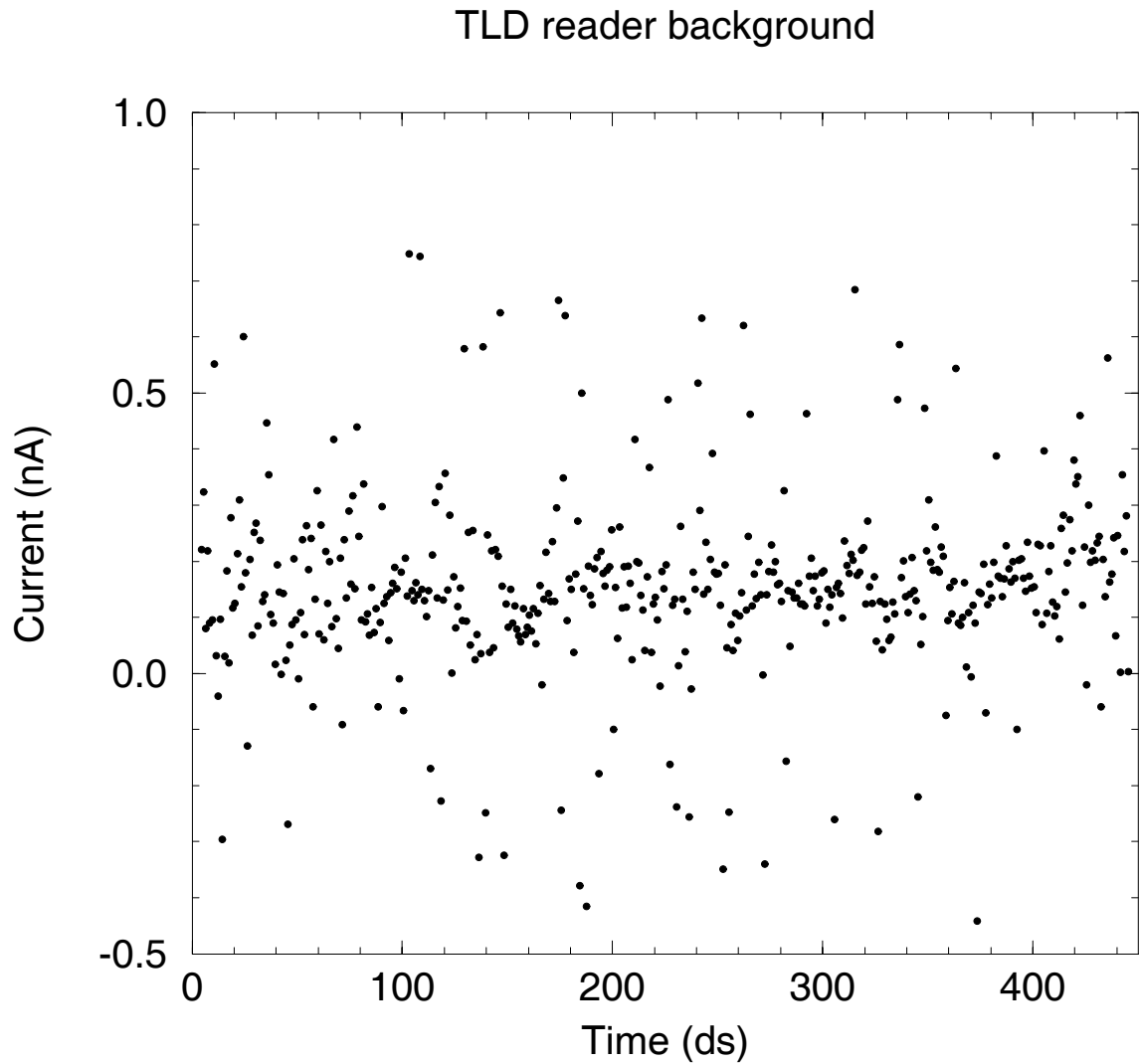


Figure 15: Background current during the standard LiF read cycle, and a high voltage of 1200 V applied to the photomultiplier. Each channel on the time axis corresponds to 0.1 seconds. The data were read out using the RS-232 port on the reader and a running average over ten channels was applied. This gives approximately the same smoothing as applied to the glow curve displayed on the reader.

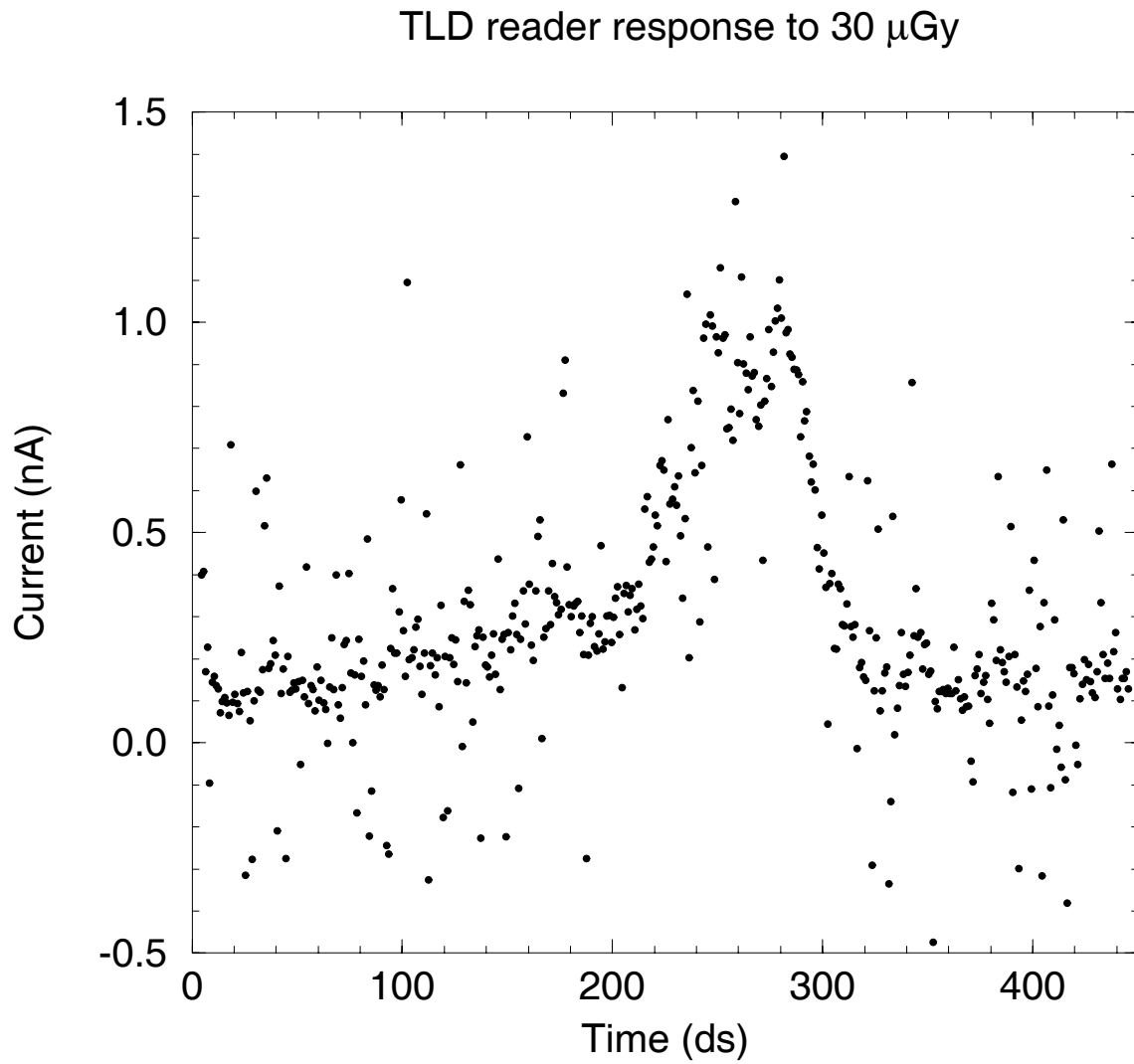


Figure 16: Glow curve obtained from a LiF TLD which had received an absorbed dose of about 30 μGy . The standard LiF readout cycle was used, and the high voltage on the photomultiplier was 1200 V. The data were read out using the RS-232 port on the reader and a running average over ten channels was applied. This gives approximately the same smoothing as applied to the glow curve displayed on the reader.

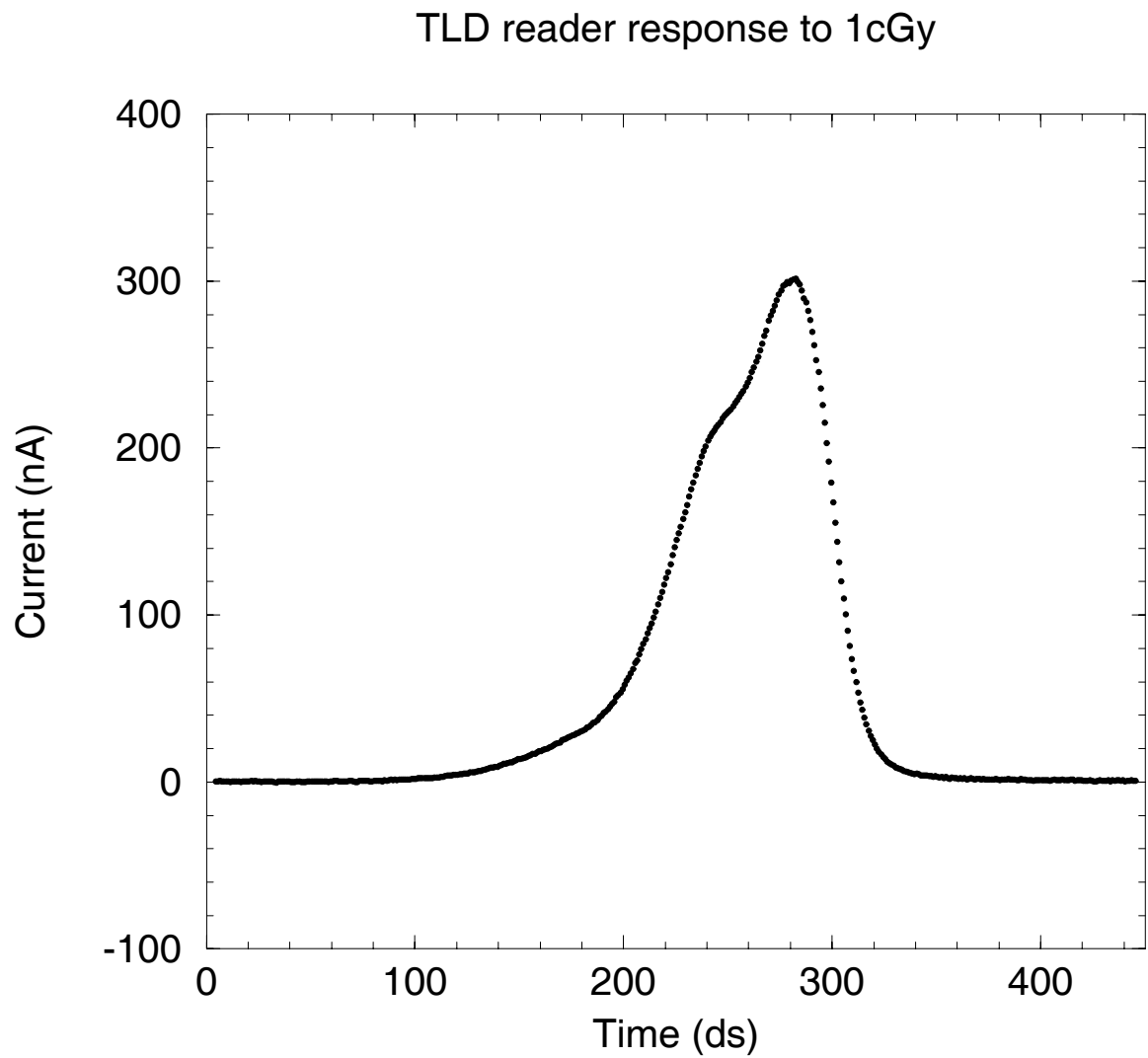


Figure 17: Same as figure 16 but for an absorbed dose of about 1 cGy.

absorbed doses of 30 μGy and 1 cGy respectively. These were also obtained at 1200 V, which gives about maximum sensitivity for the reader. The glow curves were transferred from the reader using its digital output port, and have been smoothed with approximately the same time constant which is applied to the glow curve as displayed on the reader itself. The reader integrates the current over the pre-defined region of interest, and this charge, with the background subtracted, gives the TLD response.

The data in figure 18 are meant to give a general indication of the performance of the TLD system over the absorbed dose range from the detection limit to about 20 Gy, which is as high as we characterized the supralinearity of LiF. All of the data were acquired and processed according to the protocol outlined in section 5. From about 0.5 cGy to 20 Gy the standard uncertainty on the mean of five TLDs is about 0.3%. Below 0.5 cGy the uncertainty becomes greater as the background becomes a larger fraction of the signal and by 30 μGy (figure 16) the background is approximately equal to the signal.

6.2 Sensitized LiF

We have also used sensitized LiF (see section 4.6) to measure absorbed doses in the range 500 to 2000 cGy. The same readout cycle was used as for standard LiF, and the high voltage was set to 550 V. The background reading with unirradiated sensitized chips was only slightly higher than with normal chips. However, there was evidence that the sensitized TLDs were considerably more sensitive to light. As well, repeated anneal cycles (280°C for one hour followed by 100°C for two hours) led to a decrease of sensitivity of about 1% per cycle.

Figure 19 gives the results of absorbed dose measurements carried out using sensitized

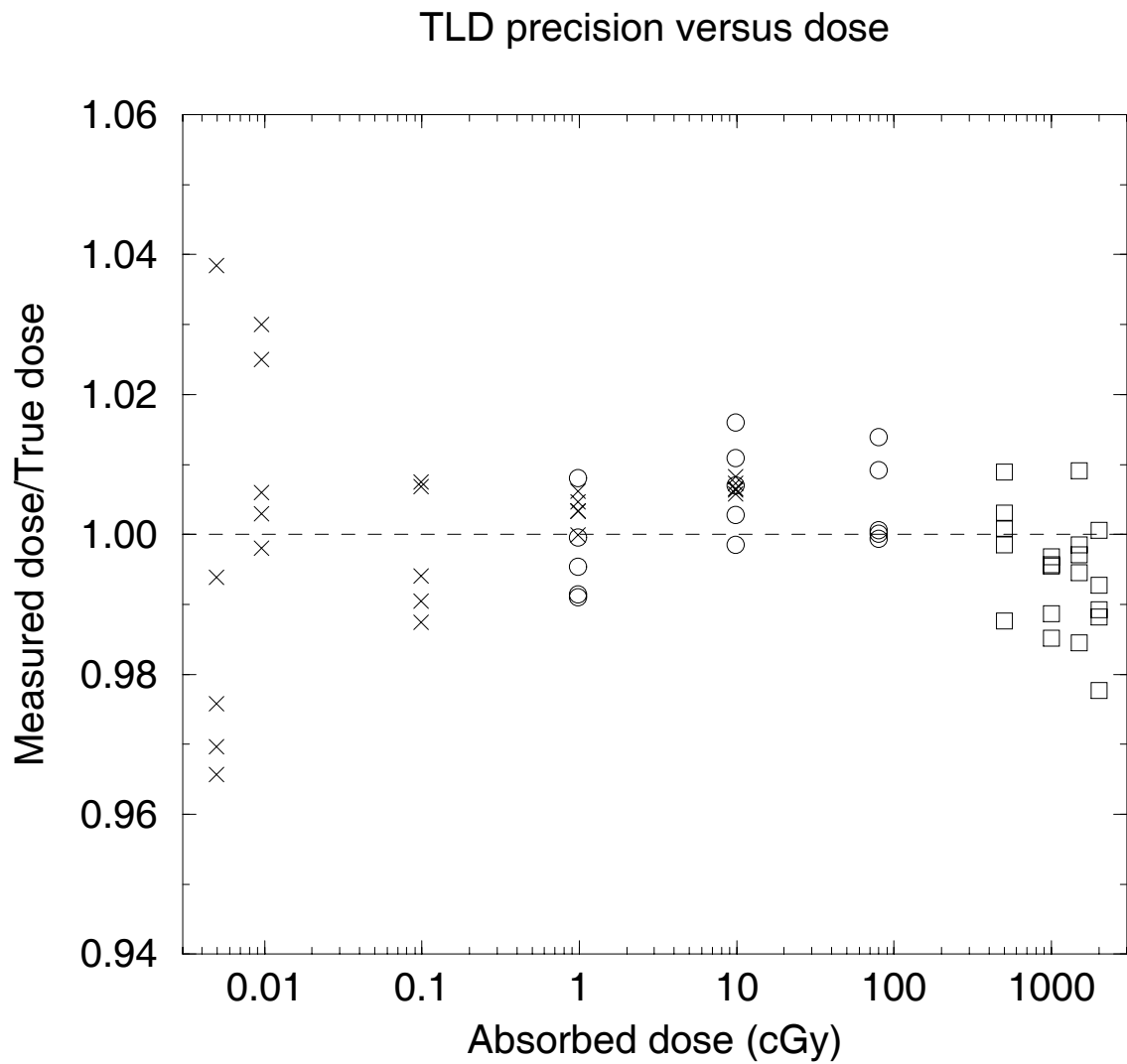


Figure 18: Data showing how the precision of the absorbed dose measured using LiF TLDs depends on the absorbed dose delivered to the TLD. All of the irradiations were performed using ^{60}Co γ -rays. Typically five TLDs were irradiated and read for each value of the absorbed dose. The symbols identify the reader high voltage: \times - 1200 V; \circ - 900 V; \square - 600 V.

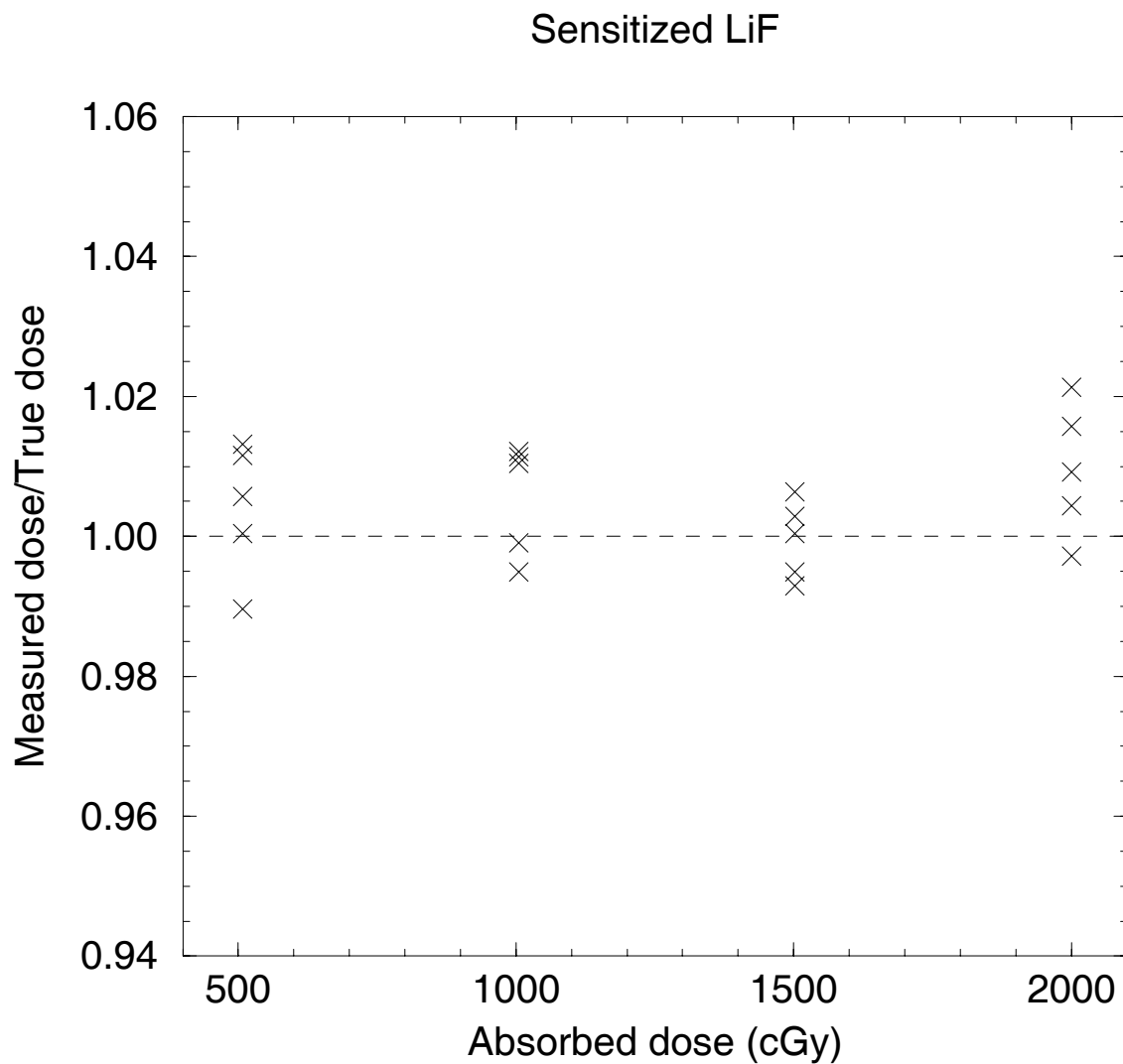


Figure 19: Data showing the precision to be expected when using sensitized LiF to measure large absorbed doses. The irradiations were done using ^{60}Co γ -rays. Each datum point corresponds to the reading from a single TLD.

LiF. The data were processed using the procedure described in section 5 but with the non-linearity correction appropriate for sensitized LiF (equation 4). Although not as extensive as the data given in figure 18, these data indicate that about the same precision can be obtained with either normal or sensitized LiF.

7 Characteristics of $\text{CaF}_2\text{:Mn}$ TLDs

Although LiF TLDs can be used to measure the absorbed dose over a wide range they are less well suited for measuring large doses (greater than a few Gy) than small. This is because the thermoluminescent response becomes nonlinear for large doses (figure 11) and the TLDs are susceptible to radiation damage (figure 13). Although less widely studied than LiF, $\text{CaF}_2\text{:Mn}$ has been proposed as a TLD material since it is thought to have a more linear response and to be less sensitive to radiation damage. For example, the manual which accompanies the Victoreen model 2800M reader (Victoreen (1989)) states that $\text{CaF}_2\text{:Mn}$ is linear from about $10\text{ }\mu\text{Gy}$ to 1000 Gy . Gorbics *et al* (1973) have also studied $\text{CaF}_2\text{:Mn}$ and their measurements of the nonlinearity are less conclusive. They find some supralinearity, which depends on the phosphor configuration and the method of readout. Marrone and Attix (1963) have studied the effects of radiation damage in both LiF and $\text{CaF}_2\text{:Mn}$. They conclude that 100 Gy is enough to significantly damage LiF, while $100 \times 10^4\text{ Gy}$ is required to cause about the same loss of sensitivity for $\text{CaF}_2\text{:Mn}$. On the negative side, $\text{CaF}_2\text{:Mn}$ is not so well suited for applications where approximate water equivalence is important since the effective atomic number of $\text{CaF}_2\text{:Mn}$ (16.3) is almost twice that of LiF (8.2).

We have carried out some preliminary measurements on $\text{CaF}_2\text{:Mn}$ with a view to de-

termining if it might be a suitable replacement for LiF for measuring absorbed dose in the range of 1 Gy to 20 Gy. In particular, we have studied the linearity of its response and its fading characteristics. Our measurements indicate that the problems associated with $\text{CaF}_2\text{:Mn}$ outweigh any advantages, at least in the dose range up to 20 Gy.

As for LiF, various annealing procedures have been proposed for $\text{CaF}_2\text{:Mn}$. In their survey article, Driscoll *et al* (1986) recommend thirty to sixty minutes at a temperature between 450 and 500°C. For $\text{CaF}_2\text{:Dy}$, Burgkhardt *et al* (1977) follow a high temperature anneal at 400°C for one hour by a low temperature anneal at 100°C for two hours. For the measurements reported here, the $\text{CaF}_2\text{:Mn}$ TLDs have been annealed for one hour at 500°C, followed by two hours at 100°C. The TLDs were rapidly cooled from 500°C to room temperature using the same procedure as was applied to the LiF TLDs. Before readout, irradiated TLDs were annealed at 100°C for 12 minutes.

Glow curves for absorbed doses of 0.85 Gy and 20.5 Gy are shown in figures 20 and 21 respectively. With the reader high voltage set to 500 V, absorbed doses from 0.1 to 25 Gy can be measured.

We noted a loss of sensitivity with $\text{CaF}_2\text{:Mn}$ TLDs after repeated anneal cycles. After a single dose of 0.85 Gy and 20.5 Gy the sensitivity decreased by 3% and 5% respectively. We have not investigated separately the effects of the annealing temperature and absorbed dose on the sensitivity although it has been reported that $\text{CaF}_2\text{:Mn}$ is very insensitive to radiation damage. We have noticed a change in the color of the $\text{CaF}_2\text{:Mn}$ TLDs after use, so it may be that the loss of sensitivity is simply due to decreased transparency.

Figure 22 shows our measurements of the supralinear response of $\text{CaF}_2\text{:Mn}$. The TLD response per unit absorbed dose was measured from 0.5 Gy to 23 Gy, and the values nor-

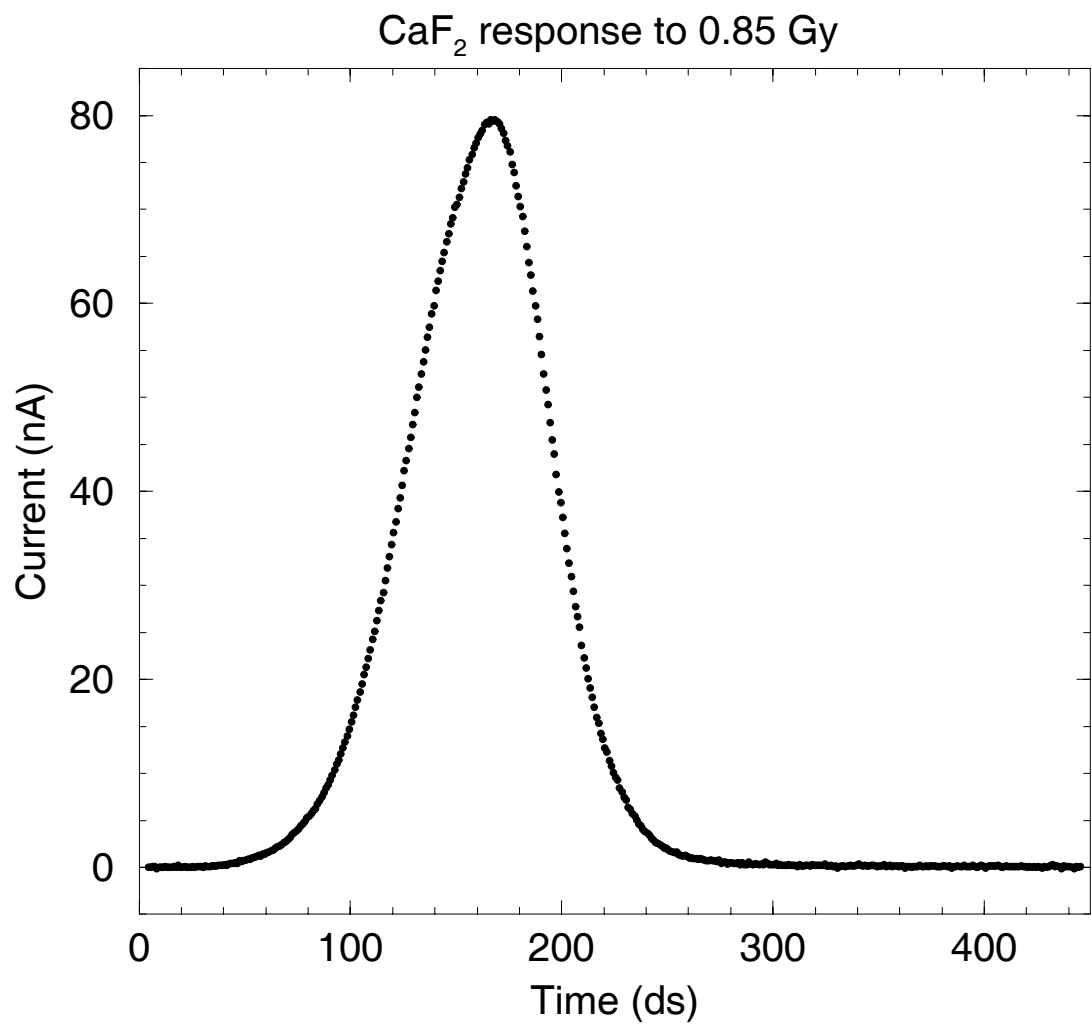


Figure 20: Glow curve obtained from a CaF_2 :Mn TLD which had received an absorbed dose of 0.85 Gy. The readout cycle given in table 1 was used, and the photomultiplier high voltage was 500 V. The data were obtained and processed as described in the caption for figure 16.

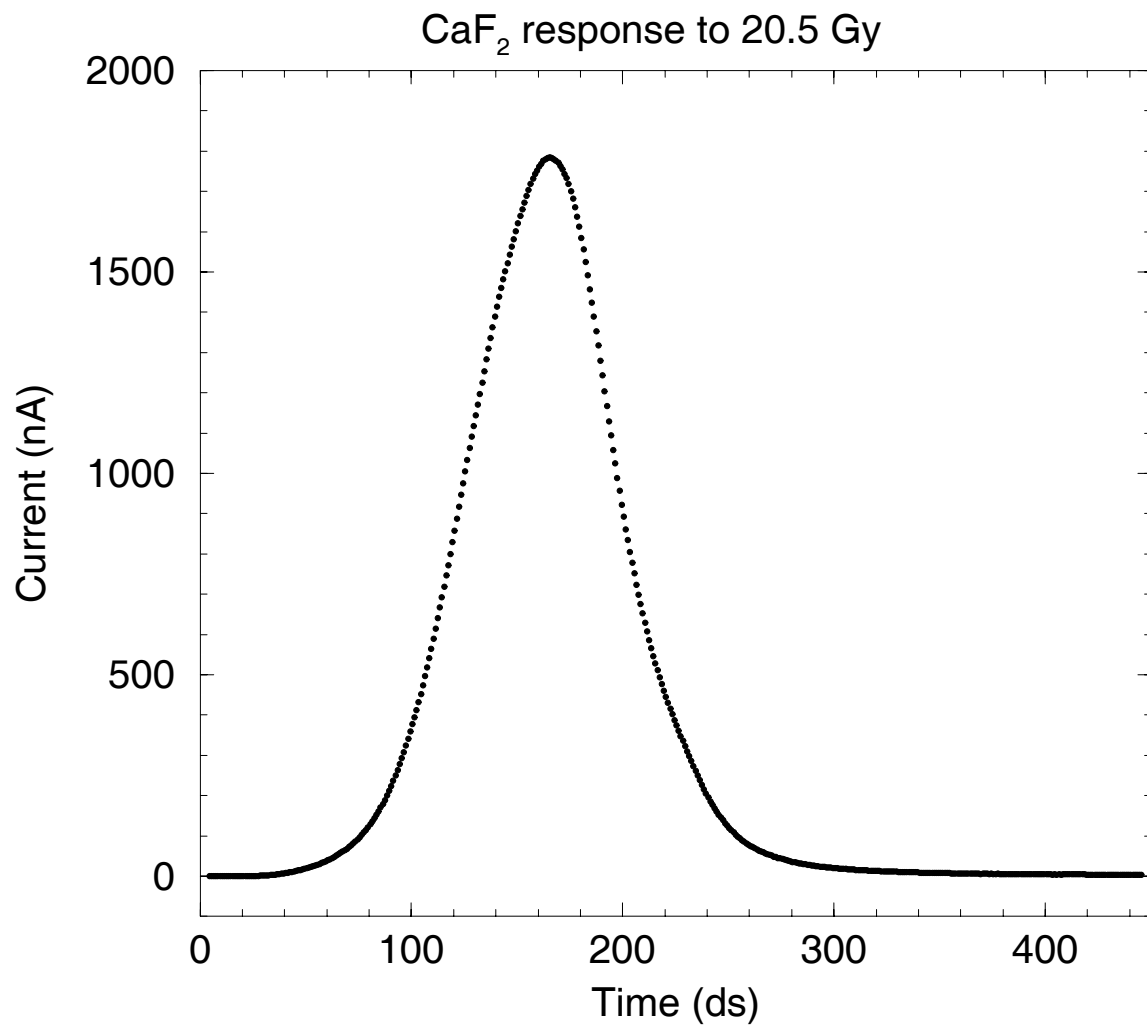


Figure 21: Same as figure 20, but for an absorbed dose of 20.5 Gy.

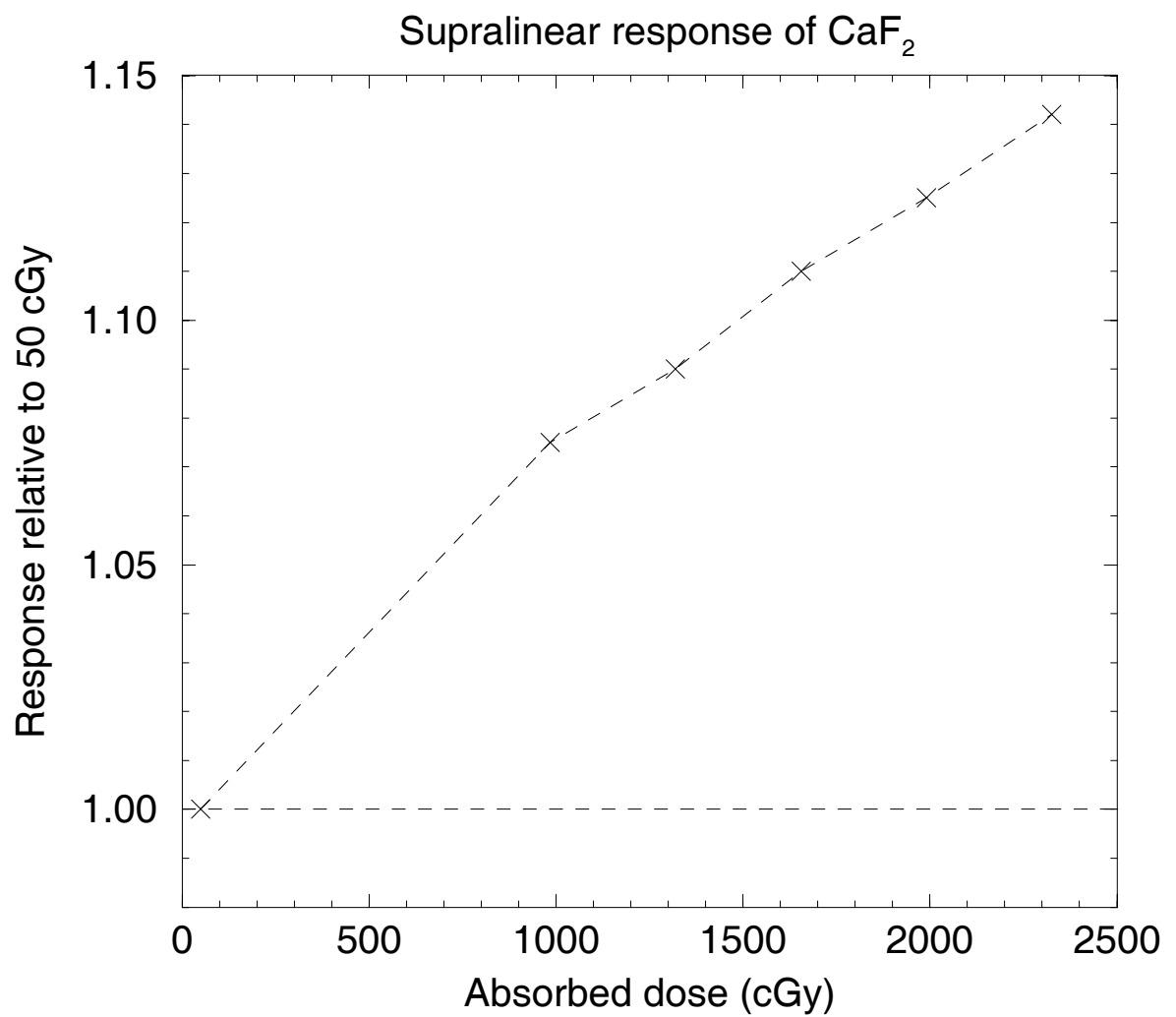


Figure 22: Measured supralinear response of $\text{CaF}_2\text{:Mn}$. The TLD response per unit absorbed dose was measured for several absorbed doses from 0.5 Gy to 23 Gy. The graph shows the response per unit dose relative to the value for 0.5 Gy.

malized to the result for 0.5 Gy. Figure 22 shows that the response of $\text{CaF}_2\text{:Mn}$ is not linear with dose, the deviation from linearity amounting to about 14% at 23 Gy. This enhancement of the response is considerably larger than that measured by Gorbics *et al* (1973). For $\text{CaF}_2\text{:Mn}$ chips they find that the response per unit dose has a maximum at about 30 Gy, where the enhancement is about 5% over the value at low doses. However, they also show that the degree of supralinearity depends on the phosphor configuration.

The final characteristic of $\text{CaF}_2\text{:Mn}$ that we studied was the degree of fading of the thermoluminescent signal after irradiation. The results obtained for two absorbed doses are shown in figure 23. We see an initial rapid fading of 4 to 6% in the first few hours after irradiation, but the light output appears to stabilize after about 25 hours. This result is in general agreement with other measurements reported in the literature and summarized by Horowitz (1984a).

8 Conclusions

Using the Victoreen model 2800M reader and separate low and high temperature annealing ovens, we have established a protocol for using LiF TLDs for the measurement of absorbed dose. Tables 1 and 2 summarize the settings to be used with the reader to cover the dose range from 30 μGy to 30 Gy.

We have studied various characteristics of LiF TLDs and have measured the supralinearity up to 24 Gy. We have shown how to use a subset of TLDs from a larger set, all of which have been through the same anneal cycle, to eliminate the effects of annealing on TLD sensitivity. Using this protocol, figure 18 indicates the precision that can be expected over the dose range

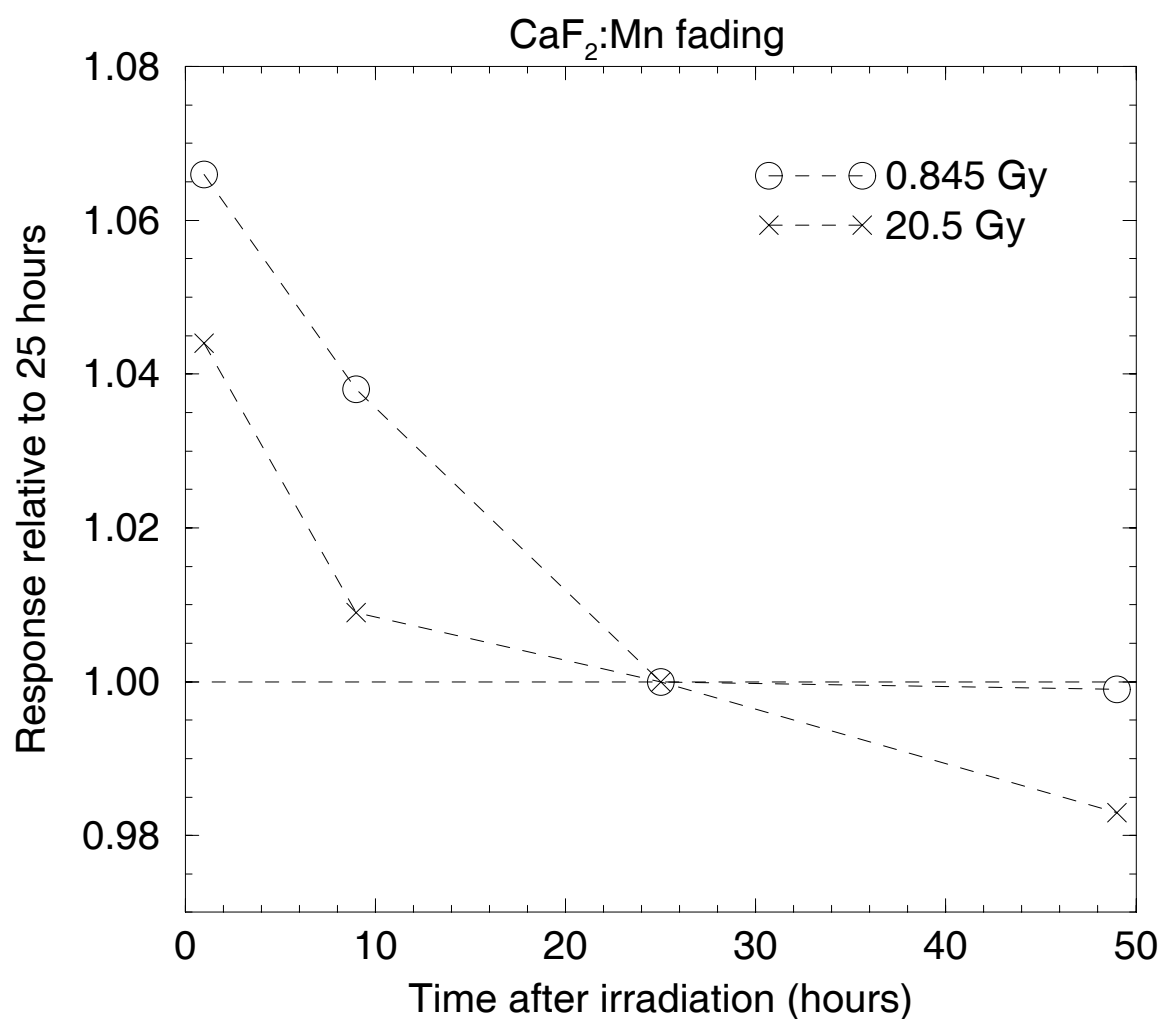


Figure 23: Measured change in the light output from CaF₂:Mn TLDs as a function of the time after irradiation. The results have been normalized to the response obtained 25 hours after the irradiation. Each datum point is the mean of three TLD readings.

from 30 μ Gy to 24 Gy. As a rule of thumb, the standard uncertainty on a dose measurement which is based on the mean of five TLD readings should be less than 0.5%.

We have also examined the possibility of using $\text{CaF}_2\text{:Mn}$ as a thermoluminescent material. Despite some reports to the contrary, we find that its response is not linear with dose, although the degree of supralinearity is significantly less than for LiF. Furthermore, it suffers from fading during the first several hours after irradiation. Given that its effective atomic number is significantly higher than for LiF, we do not see $\text{CaF}_2\text{:Mn}$ replacing LiF for applications where approximate water equivalence is important.

9 Acknowledgements

The effort to establish a thermoluminescent dosimetry system for IRS was begun by Miroslav Lieskovsky who worked on contract to NRC from January to March, 1992. He showed that the high temperature anneal was the cause of the changes in sensitivity which had been observed. Unfortunately, he completed his contract and took up permanent employment before it was realized that the basic problem was poor temperature control in the high temperature oven.

David Marchington provided valuable technical assistance, such as adding a fan to the low temperature oven and constructing various Lucite and aluminum phantoms. Leo Heistek installed temperature controllers on both ovens thus permitting the oven temperature to be controlled to better than 1°C. Ken Shortt and Norman Klassen, as well as other members of the IRS group, provided advice and assistance as required.

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