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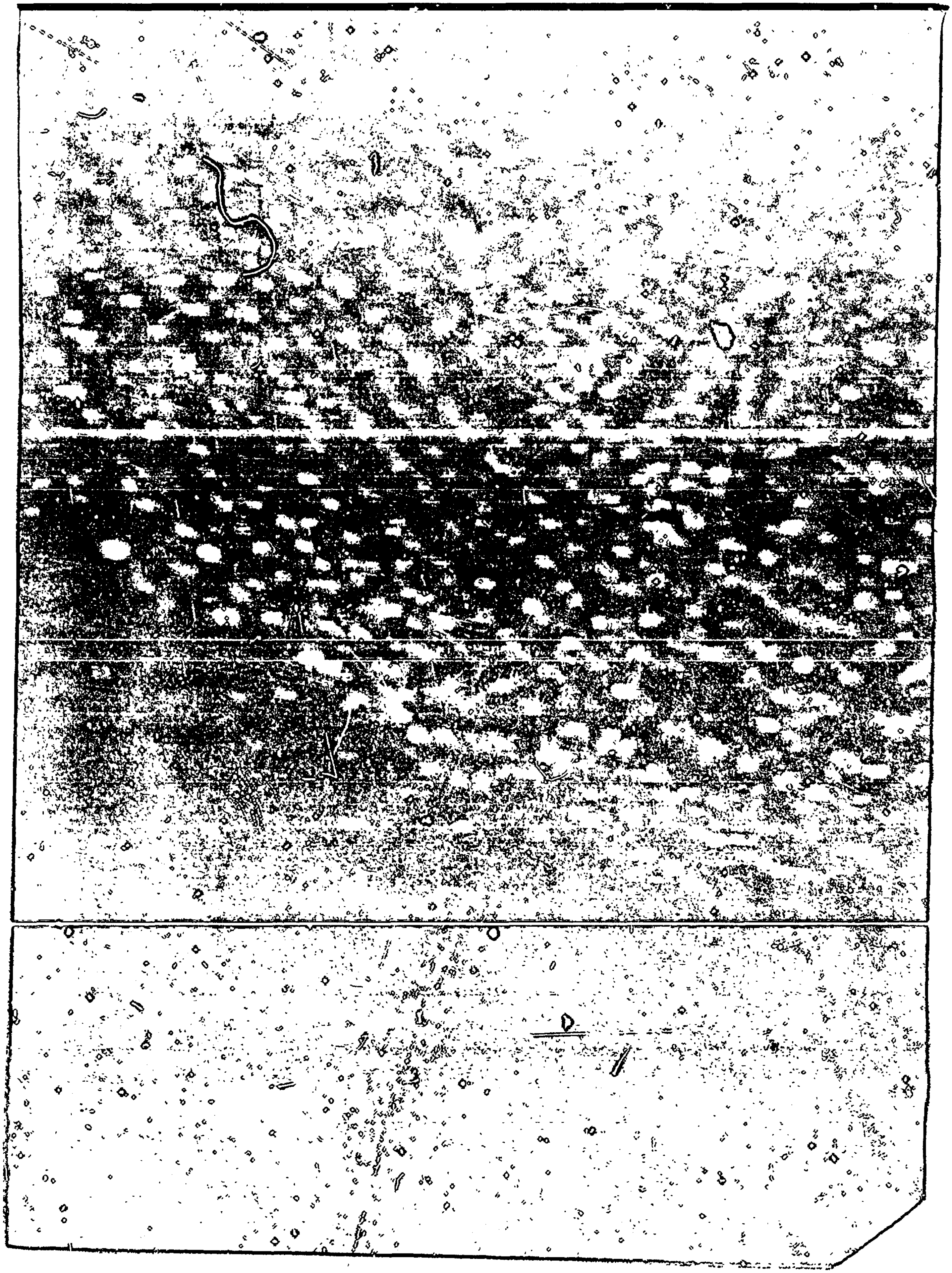

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13. ABSTRACT

Organoleptic testing of radappertized foods has shown that acceptability is highly correlatible with irradiation temperature. In general, irradiation temperatures in the region of -30°C yield the most acceptable product. Thus, a requirement has been generated, particularly in commercial applications, for a dosimeter capable of being used in radiation field mapping and production dose monitoring at these temperatures. Of the various dosimetry systems considered for this application a thermoluminescent system shows the most promise. Samples of the commercially available materials were subjected to a screening and evaluation procedure to determine their applicability. The results of this study are discussed including calibration data for these thermoluminescent materials for electron and gamma irradiation at various temperatures. For example, a sample of TLD-100 in its loose powder form yields values twenty to forty percent lower when irradiated at -196°C than when irradiated at 25°C. Such results show the potential usefulness of thermoluminescent materials but point to the critical dependence of the calibration function on temperature.

A standard procedure for "reading out" thermoluminescent materials is discussed. This procedure involves, among its other features, a ten minute annealing period at 100°C prior to readout. This was found necessary in order to eliminate a number of low energy traps that contribute to spurious results.

Again, for commercial radio-sterilization applications, a suitable total dose range is necessary. The potential applicability of certain high dose (in the megarads region) materials is discussed. For example, our evaluation of Isomet LiF in both its single crystal and loose powder forms will be contrasted with results on this material as reported by Tochilin.

This paper demonstrates the requirement for a low-temperature dosimetry system and evaluates the presently available thermoluminescent materials (both commercially available or those in a developmental stage) with respect to their potential for meeting this requirement.

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TECHNICAL REPORT
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RADIATION DOSIMETRY AT CRYOGENIC TEMPERATURES USING THERMOLUMINESCENCE

by

R.D. Jarrett, J. Halliday and J. Tocci

Project Reference:
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Food Laboratory
U.S. ARMY NATICK LABORATORIES
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FOREWORD

This report describes the evaluation of various thermoluminescence (TL) materials as low temperature dosimeters. The requirement for a low temperature dosimeter results from organoleptic testing of radappertized foods which has shown that acceptability is improved in foods processed in the region of -30°C.

From the results of this report, it can be stated that commercially available TLD materials may be used at low irradiation temperatures provided they are calibrated at the temperatures. It points out, however, the need for additional studies to determine the extent of decrease in TL response as a function of irradiation temperatures.

The work was performed under 1J662713A033, Radiation Services.

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ABSTRACT

Organoleptic testing of radappertized foods has shown that acceptability is highly correlatible with irradiation temperature. In general, irradiation temperatures in the region of -30°C yield the most acceptable product. Thus, a requirement has been generated, particularly in commercial applications, for a dosimeter capable of being used in radiation field mapping and production dose monitoring at these temperatures.

Of the various dosimetry systems considered for this application a thermoluminescent system shows the most promise. Samples of the commercially available materials were subjected to a screening and evaluation procedure to determine their applicability. The results of this study are discussed including calibration data for these thermoluminescent materials for electron and gamma irradiation at various temperatures. For example, a sample of TLD-100 in its loose powder form yields values twenty to forty percent lower when irradiated at -196°C than when irradiated at 25°C. Such results show the potential usefulness of thermoluminescent materials but point to the critical dependence of the calibration function on temperature.

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This paper demonstrates the requirement for a low-temperature dosimetry system and evaluates the presently available thermoluminescent materials (both commercially available or those in a developmental stage) with respect to their potential for meeting this requirement.

General Introduction

In numerous applications of ionizing radiation there exists the need for a dosimetry system with a rather specialized set of properties - the capability of measuring doses in the megarad region and at sub-zero centigrade irradiation temperatures. It is the intent of the present work to explore the applicability of thermoluminescent materials in fulfilling this dual requirement. The development of a satisfactory readout technique and some of the more significant pitfalls encountered are discussed. Several selected thermoluminescent materials are evaluated and discussed relative to their "ambient" and low temperature dose response.

This work was performed using the irradiation facilities of the U.S. Army Natick Laboratories, Natick, Massachusetts, U. S. A. The Irradiation Laboratory has a nine kilowatt electron linear accelerator capable of producing 12 MeV electrons, three Cobalt-60 sources (1.5 megacuries, 35 kilocuries, and 7 kilocuries) and a 200 kilocurie Cesium-137 source. All gamma irradiations were made using the two smaller Cobalt-60 research irradiators which have dose rates of 4.4 kilorads and 0.56 kilorads per second.

Introduction

The basic phenomenon underlying thermoluminescent dosimetry (TLD) is the freeing of trapped electrons by thermal stimulation. A plot of the intensity of the light emitted by the luminescent material as a function of its heat treatment is the familiar "glow" curve. Since the heat treatment involves a temperature-time relationship, the glow curve and hence the results of a thermoluminescent dosimeter reading must be considered relative to the mode of heat treatment. A large number of temperature profiles are possible; three basic types are illustrated in Figure I. The first type (Figure Ia) features a preheat rate from A to B followed by the primary heating rate from B to C. A second type (Figure Ib) involves only the primary heating rate from A to C. A more versatile type (Figure Ic) inserts an annealing plateau between the preheat part of the cycle and the commencement of the primary heating. In all cases C represents the maximum temperature achieved and is usually the point where the integration of the light output is terminated.

The basic commercial instrument used was the Harshaw Model 2000 Analyzer (I). It was operated in the temperature range 40°C to 410°C using various primary heating rates Samples of the thermoluminescent powder were dispensed onto the heating pan and then subjected to the desired temperature profile.

Post Irradiation Annealing

It has been suggested (2) that annealing the material for 10 minutes at 100°C after irradiation and before reading the samples would improve the results. This improvement is based upon the fact that heating at 100°C would greatly reduce or remove the traps below that temperature and give a reliable base line. We evaluated the effectiveness of this annealing procedure by dispensing a sample of irradiated TLD-100 powder into the heating pan of the reader and observing the output signal of the photomultiplier tube as a function of time when the sample was heated to and held at 100°C. It was observed that the signal approached a constant value after 8.5 minutes. These results demonstrate that if the sample is held at 100°C for 10 minutes the base line is stable and any inaccuracy in timing would have a negligible effect.

Based upon the fact that post annealing stabilized the base line we modified our reader to allow post annealing to be performed in the reader as typified by Figure Ic. It is possible to vary the post annealing temperature (B) to any value and the annealing time (B to B^1) to any period. These modifications also allowed to reduce the post anneal time to less than 10 minutes as the samples are now held for an exact time which eliminates the inaccuracies which would be involved in heating the samples in an oven for less than 10 minutes and transferring them to the reader. For most of the results reported we used this procedure with an annealing time of 0.5 minutes. It should be noted that the coincidence meter operates only during the period B^1 thru C. An additional advantage in using this readout procedure is that the temperature of the pan and sample, prior to the start of readout, has less effect on the peak height and integrated charge values. Therefore, we improved the readout reproducibility with this heating procedure.

As is obvious from the preceding description of an optimum readout technique, an integrated peak area as well as peak height (s) is a useful parameter in correlating luminescence with dose level. Based upon the standard error of ten readings at a given dose it was, in fact, determined that the integrated charge (output from the photomultiplier tube) gave the more reproducible results. The standard error using peak height measurements was approximately 5% as contrasted to a value of 2% using integrated charge measurements. The doses used for the evaluation were in excess of ten kilorads.

There are several effects observed incidental to the heat treatment which might lead to instrumental artifacts. These involve mainly the composition and pre-history of the heating pans and the contribution of infrared radiation at the higher temperatures.

During the course of these studies we were supplied with experimental type heating pans from Harshaw. While evaluating the usefulness of these pans at high readout temperatures we observed a rather impressive effect on our readings as a result of pan condition. This effect was observed by making a number of readings using irradiated TLD powder, that had been heated in an oven at 100°C for 10 minutes, in both new and used pans. The readings showed that the condition of the heating pan can affect the peak height values by as much as 35 percent and the integrated charge values by as much as 25 percent. Calibration curves made with the different pans had the same general shape but were displaced by various amounts.

According to the procedure described by Webb (3), the best readout procedure is that which integrates the total area under the glow curve peaks but excludes the rising contribution occurring at high temperatures. This artifact at the end of the glow curve is attributable to the emission of infrared radiation from the phosphor and pan. Hence, the optimum readout conditions for the higher dose levels must be compromised in order to minimize the contribution of this effect.

When quantitative comparisons are made between glow curves, particularly when certain peaks are to be identified and compared, the calibration of the pan temperature indicating device is crucial. A convenient alternative to the use of calibrated thermocouples is a paper thermometer (4). These devices are available in ten degree (centigrade) increments and may be conveniently placed directly on the pan. Gross errors in the pan temperature may be quickly discovered using these thermometers.

TLD Materials Evaluated

Various thermoluminescent materials were evaluated with respect to their fulfillment of part of the dual dose range-irradiation temperature requirement. In addition to regular lithium fluoride (LiF Harshaw TLD-100), the dose response of lithium borate ($Li_2B_4O_7$:Mn), isomet lithium fluoride, strontium fluoride and lead fluoride was studied.

The thermoluminescene powders were dispensed into gelatin capsules (5 x 15 mm) for irradiation. Each capsule contained sufficient powder for seven readings. Exposed powder was dispensed in 28 mg. lots into the built-in heating pan for analyzing the radiation induced thermoluminescence.

The capsules of powder were irradiated to the desired doses in electron equilibrium shields using one of the Cobalt-60 irradiators previously mentioned. Doses were based on chemical dosimetry using the Fricke dosimeter (G value = 15.6).

Most of the low temperature irradiations were conducted at -40°C using cold nitrogen gas to regulate the temperature of the irradiation chamber. The several comparisons made at 196°C were achieved by irradiating the samples immersed in liquid nitrogen.

Lithium Fluoride - TLD - 100

This material is one of the most researched and used thermoluminescent materials available. We therefore initiated our studies using this material and observed many interesting phenomena. Our first temperature response studies showed that the loose powder samples irradiated at -196°C gave results that were twenty to forty percent lower than samples irradiated at 25°C. These studies were over the limited dose range of 10^3 to 5×10^4 rads. To evaluate the temperature response in a temperature range more nearly approaching that to be used in radappertized foods we selected -40°C. It is observed in Figure 2 that in the dose range from 10^3 to 3×10^5 rads the integrated charge values of samples irradiated at -40°C are sixteen to thirty percent lower than similar samples irradiated at 25°C. Above 3×10^5 rads it appears that radiation damage to the crystal is more pronounced in the 25°C samples as evidenced by the reduced response. The peak height values differ quite considerably between the two temperatures as shown in Figure 3.

Lithium Borate

Two commercial forms of lithium borate were evaluated for their usefulness at doses greater than 10^3 rads and at two temperatures 25° and -40°C. They are loose powder and Conrad's teflon disks.

The powdered lithium borate was treated prior to irradiation by baking for 15 minutes at 300°C. The irradiated samples were annealed in an oven at 100°C for 10 minutes and read out with nitrogen gas purging the sample. The response curves of this material (Figure 4) show that it may be used into the megarad range depending upon the accuracy required in the dose measurements. The difference in response between samples irradiated at the two temperatures is approximately $\pm$ 6% of an average value. This is approximately equal to the standard deviation of our peak height readings and three times the standard deviation of our integrated charge readings.

The lithium borate teflon disks show the lowest sensitivity to irradiation temperature of any of the systems we tested, (Figure 5). All the integrated charge values for these disks between 10^3 and 3×10^4 rads were within the normal fluctuation of readings. Above 3×10^4 rads the 25°C samples give more light output per rad than the samples irradiated at -40°C.

Isomet Lithium Fluoride

In our search for a high dose phosphor we obtained a sample of the Isomet LiF suggested by Goldstein (5). This material was reported to be usable in the megarad range and to have a high temperature peak between 400 and 450°C. Tochlin reported (6) reading out 16 mg samples by preheating the sample to 350°C to erase the earlier dominant peak, and then reheating the samples to read out the 450°C peak. We irradiated samples in the megarad range and using Tochlin's procedures tried without success to locate the 450°C peak. We tried using a fast heating rate of approximately 40 C° per second and a slow rate of approximately 7 C° per second. The maximum temperature peak we were able to locate was at approximately 340°C (Figure 6).

It was observed from the glow curves that the height of the 340°C peak was a function of the irradiation dose, while the lower temperature peaks remained relatively constant. Following the reasoning previously stated with regard to annealing the low energy traps, the samples were dispersed onto the heater pan and rapidly heated to 240°C. They were held at this temperature for 0.5 minutes and then heated to a temperature maximum of 390°C at a rate of 5° per second. Figure 7 shows response curves we obtained for samples irradiated at 25° and -40°C, using the peak height values at the 540°C peak and the integrated charge values above 240°C. These curves show that there is considerable effect on the response of this material as a function of the irradiation dose. In the dose range of 10^5 to 3 x 10^6 rads, the samples irradiated at 25°C response is approximately four times that of similar samples irradiated at -40°C. It is noteworthy that using the above procedure this material is a potential high dose phosphor. Our main concern is the availability of similar material in the future.

In an effort to put the phosphor in a form which would be more usable than the loose powder we had Tyco Laboratories (7) grow single crystals .125 x .25 x 1.5 inches. These crystals were made by the continuous growth method (8,9) which allows the dimension of the crystal to be controlled independent of the growth rate. The glow curves for these single crystals might indicate that the concentration of the impurities in the original Isomet crystal are modified by the crystal growth method. It was observed that the main response peak to irradiation dose in these crystals is at 150°C in contrast to peak position at 340°C in the loose powder.

While we were not successful in our initial attempt to incorporate the Isomet material into single crystals that had a megarad dose response, Tyco's growth process shows two potential areas for development. First they are able to control the impurities in the growth crystals under some conditions so that they can conceivably dope the crystals. Secondly, these crystals are very transparent and could be grown in shapes which would allow the crystals to be read spectrophotometerically as well as thermally. These crystals' dose response resembles more the TLD-100 results than the original Isomet powder results Fig. 8.

Strontium and Lead Fluoride

Samples of strontium fluoride and lead fluoride were obtained from Harshaw as materials that might be useful in measuring doses in the megarad range. These samples were irradiated with dose between 0.5 and 7 megarads and evaluated using Harshaw's standard readout procedure of heating to 100°C rapidly and then to temperature maximum using a lower heating rate. The results obtained were negative in that there was no appreciable change in the thermoluminescence as a function of dose.

Conclusion

These studies re-emphasize the importance of careful control of the readout equipment, especially the condition of the heater pan. It may be advisable to maintain a calibrated phosphor that is read periodically to assure that the pan has not degraded to a point that it is giving erroneous readings. In order to reduce the background, calibrations should be made for each phosphor using the optimum set of reader conditions. It has been demonstrated that an improved readout procedure is obtained by holding a sample at an annealing temperature in the reader (without integrating the signal) for a predetermined time and then continuing the heating at a fixed rate while integrating the signal. The reproducibility of the readings is improved in this manner and the contribution from the low energy traps is reduced or eliminated.

Commercially available TLD material may be used at low irradiation temperatures provided they are calibrated at the temperature. More studies need to be conducted to determine the decrease in response as a function of temperature.

Acknowledgements

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References

1. Harshaw Chemical Company, Cleveland, Ohio

2. R. A. Arnold, private comm.

3. G.A. Webb, J. Sci. Instr. 44 (1967)

4. Paper Thermometer Co., 10 Stagg Dr., Natick, Mass. 01760

5. N. Goldstein, E. Tochlin, and W. G. Miller, Health Physics 14, 159-162 (1968)

6. E. Tochlin, private comm.

7. Tyco Corporate Technology Center, Waltham, Mass.

8. H. E. LeBelle, A. I. Mlavski, Material Research Bull. 6, 571 (1971).

9. H. E. LeBelle, ibid., 581 (1971).

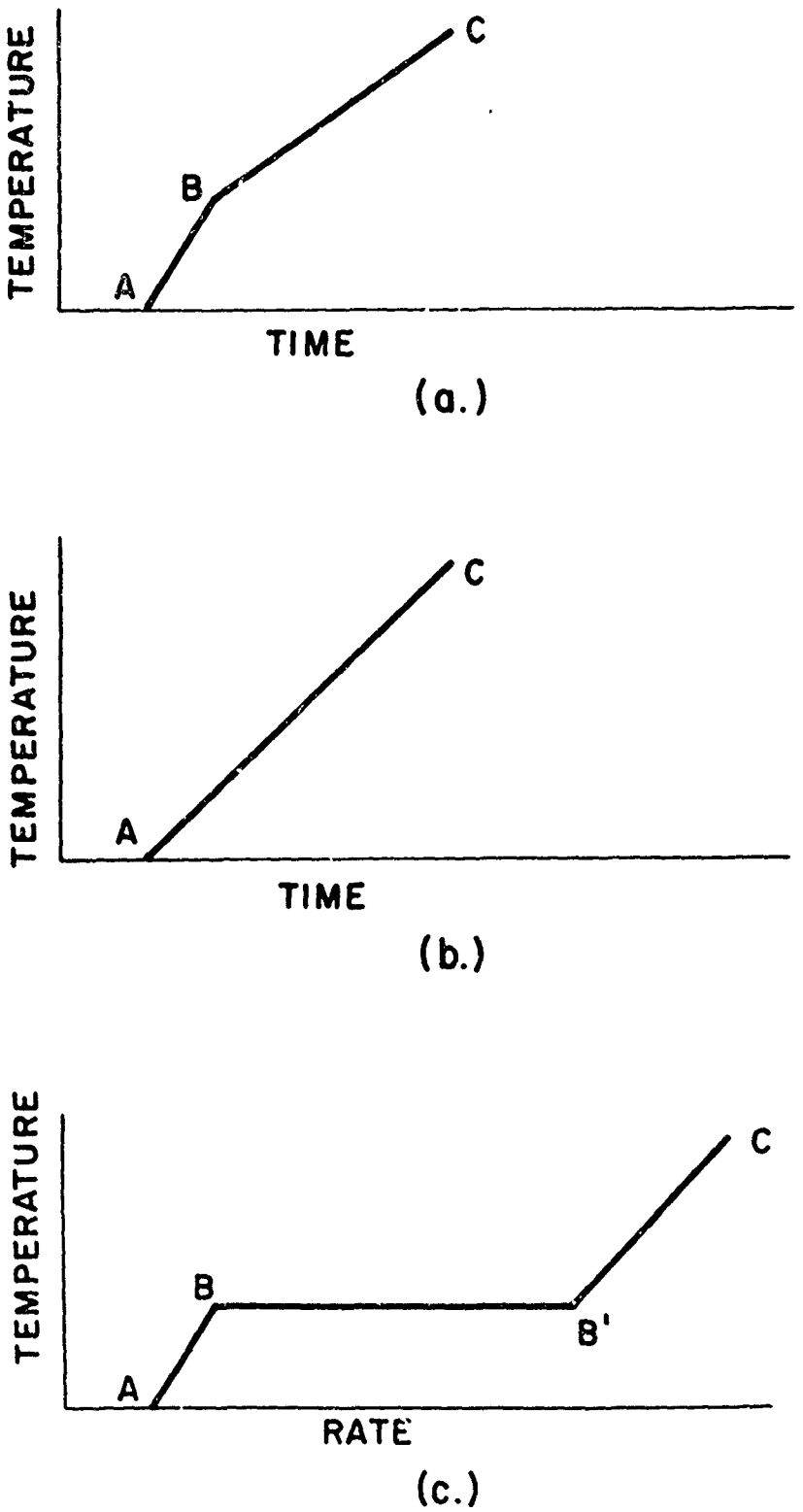

Figure 1. Three basic heating profiles used in evaluating the thermoluminescence properties of materials.

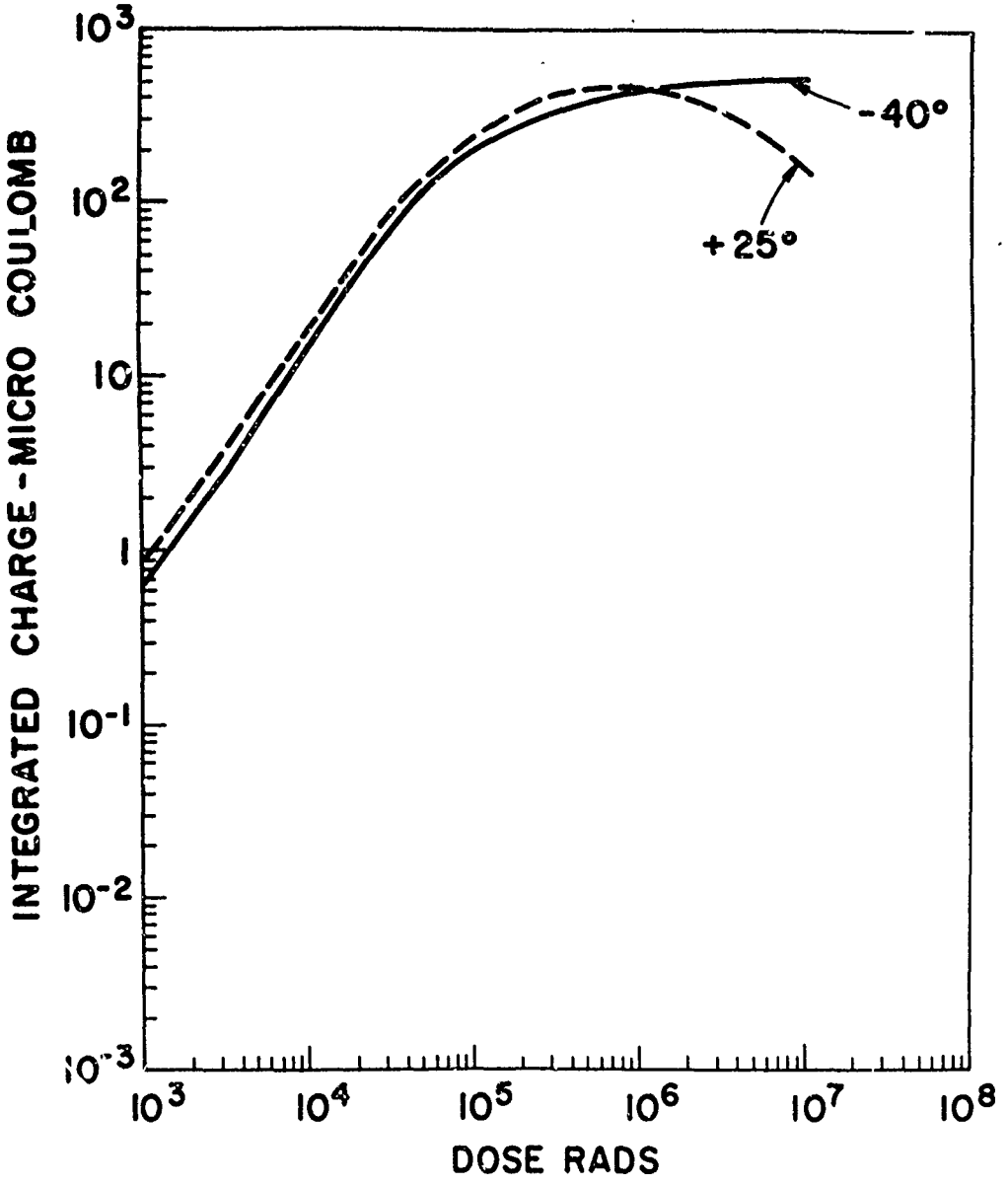

Figure 2. Response curve of LiF-TLD-100 as a function of irradiation temperature.

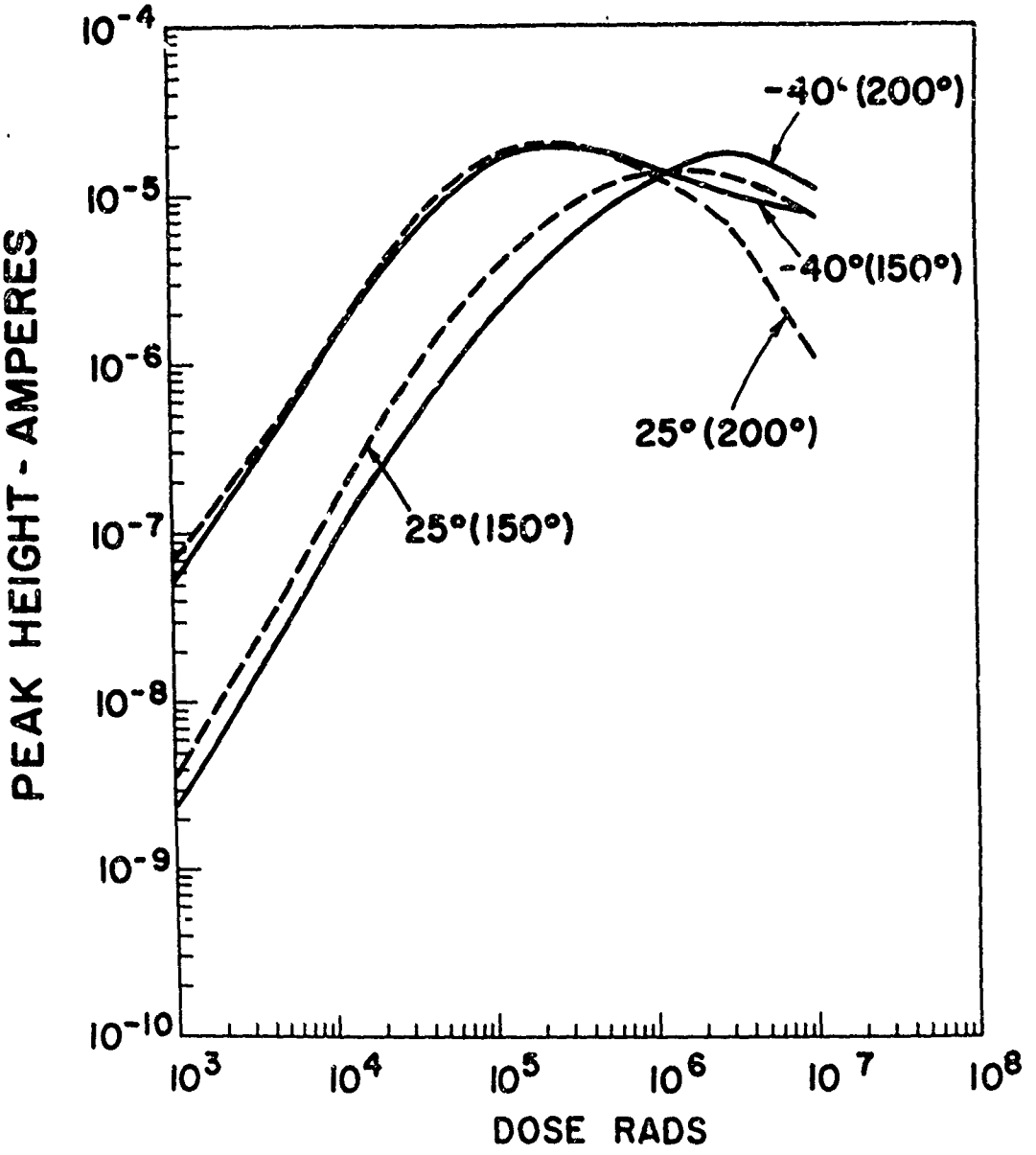

Figure 3. LiF-TLD-100 peak height measurements read at two temperatures (150° and 200°C) as a function of irradiation temperature.

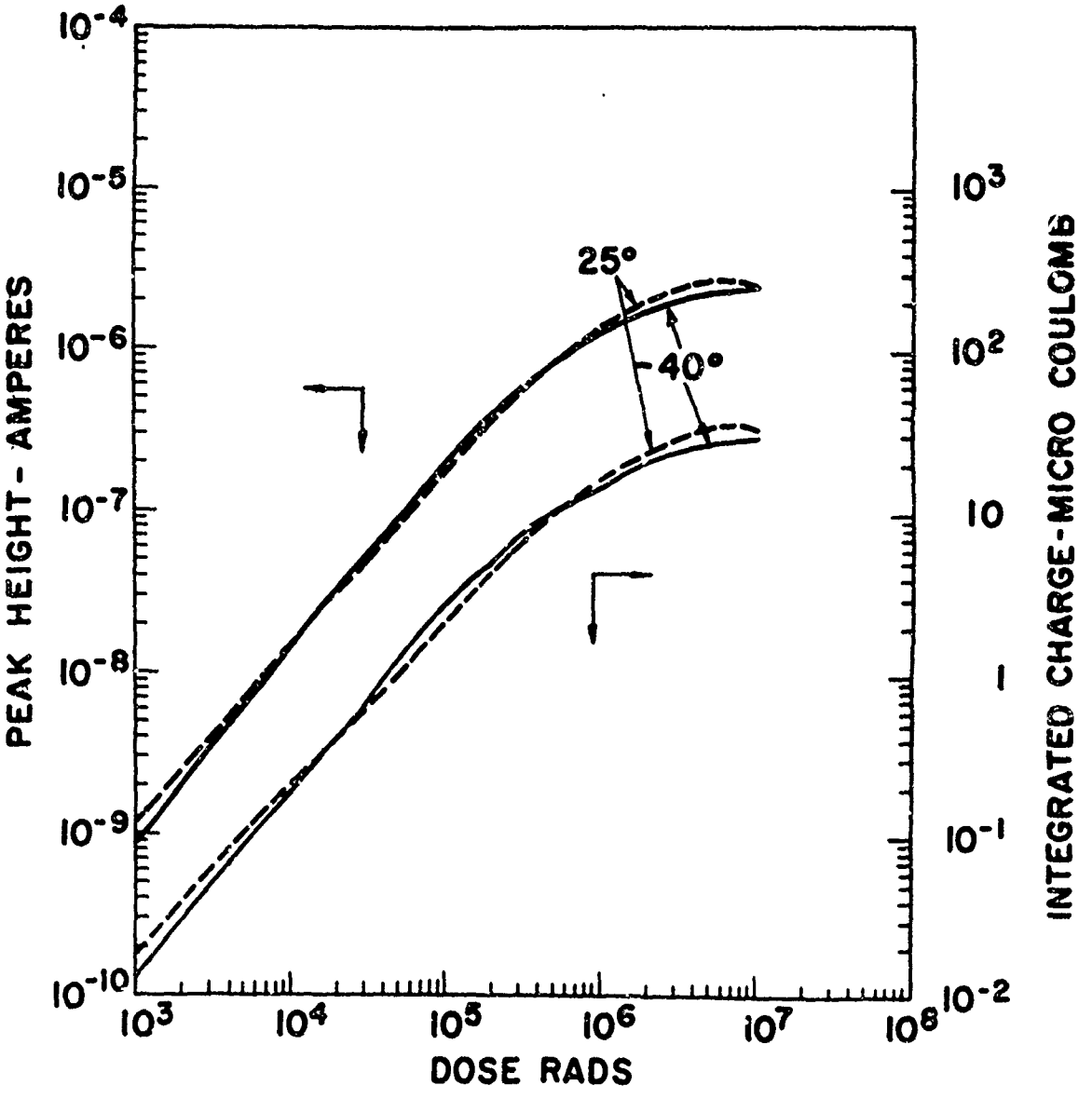

Figure 4. Peak Height and integrated charge values for samples of $Li_7B_4O_7$ irradiate between 10^3 and 10^7 rads at 25° and -40°C.

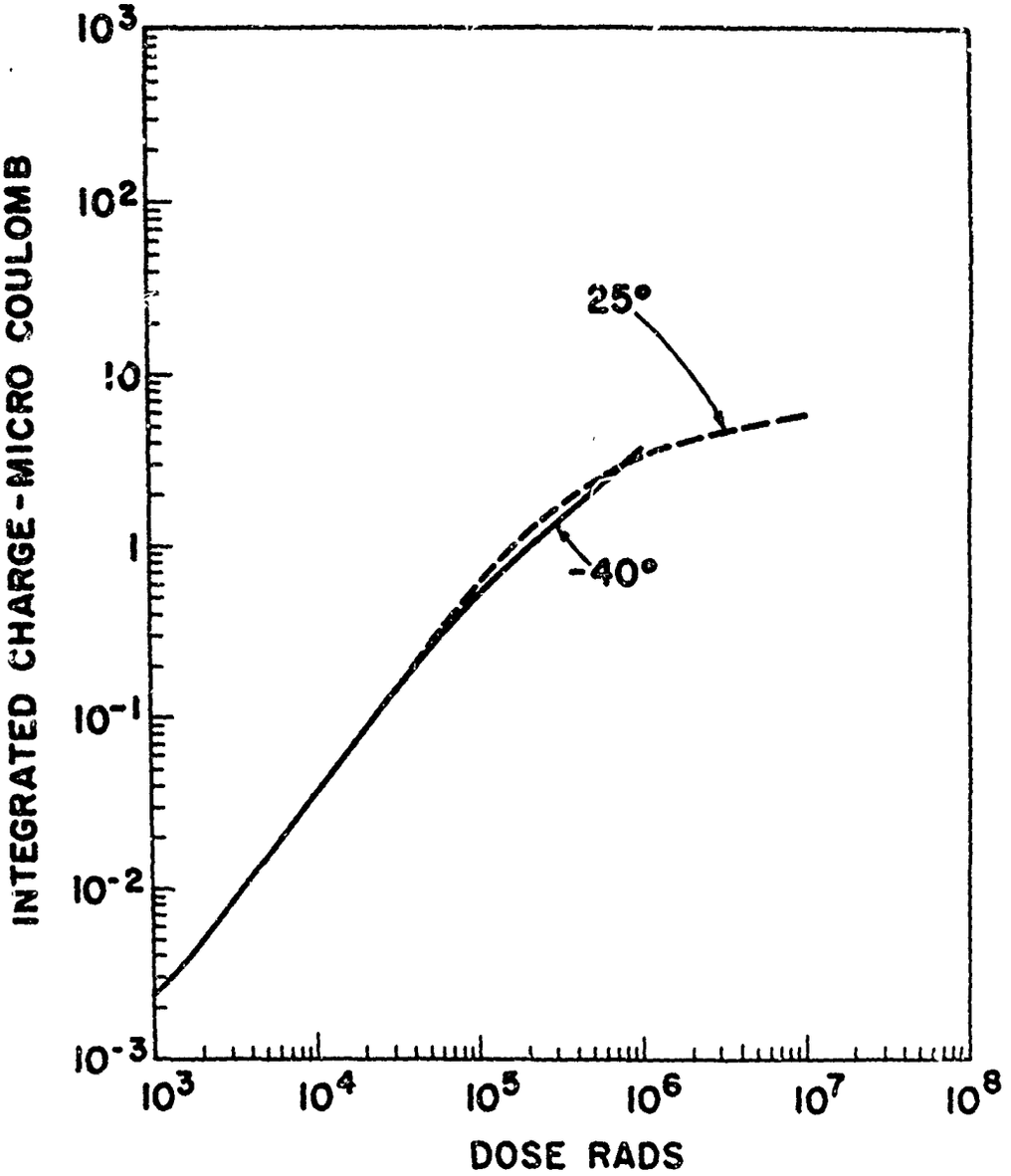

Figure 5. Response curve of teflon disks impregnated with lithium borate when irradiated at 25° and -40°C.

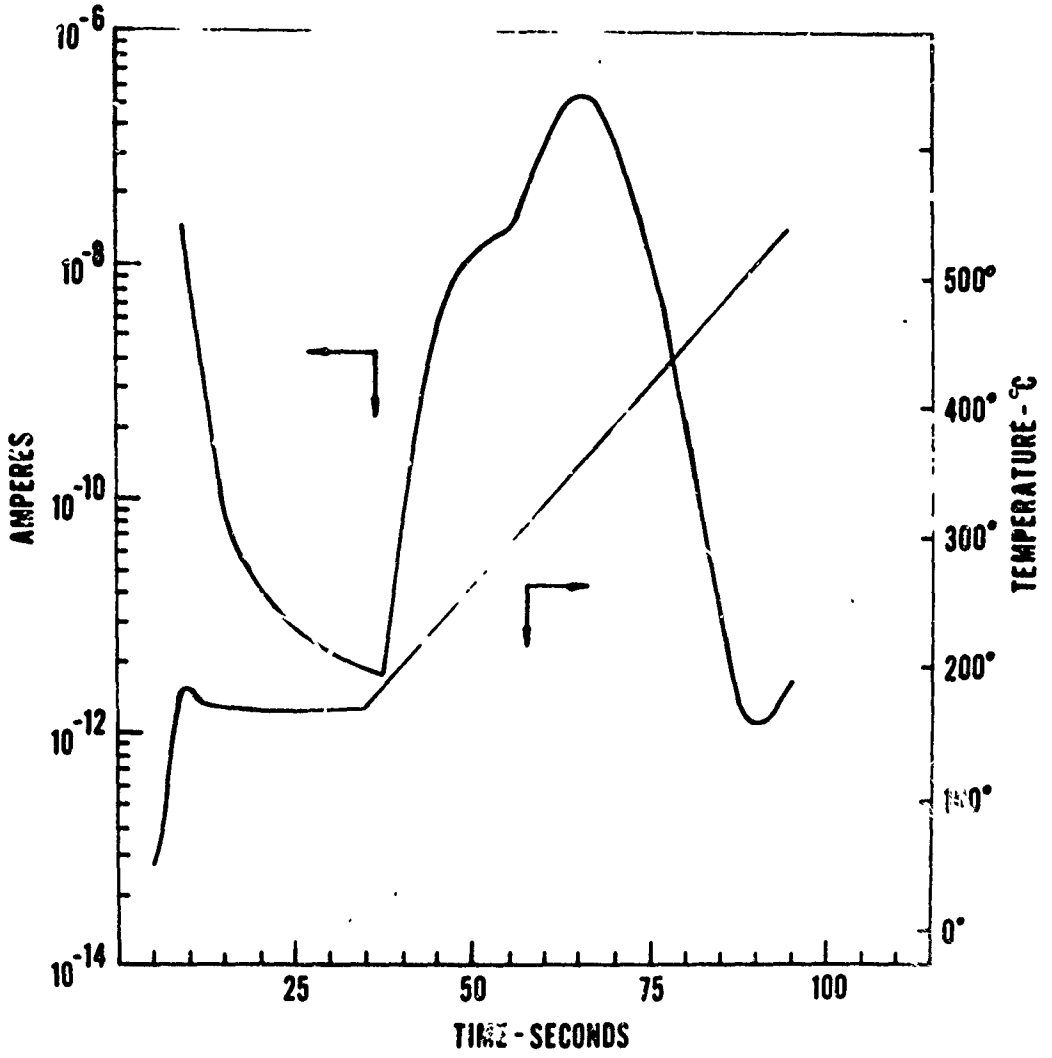

Figure 6. A sample of Isomet LiF loose powder preheated to 170°C and held for 30 seconds, then heated to a temperature maximum of 530°C at a rate of 6 degrees per second.

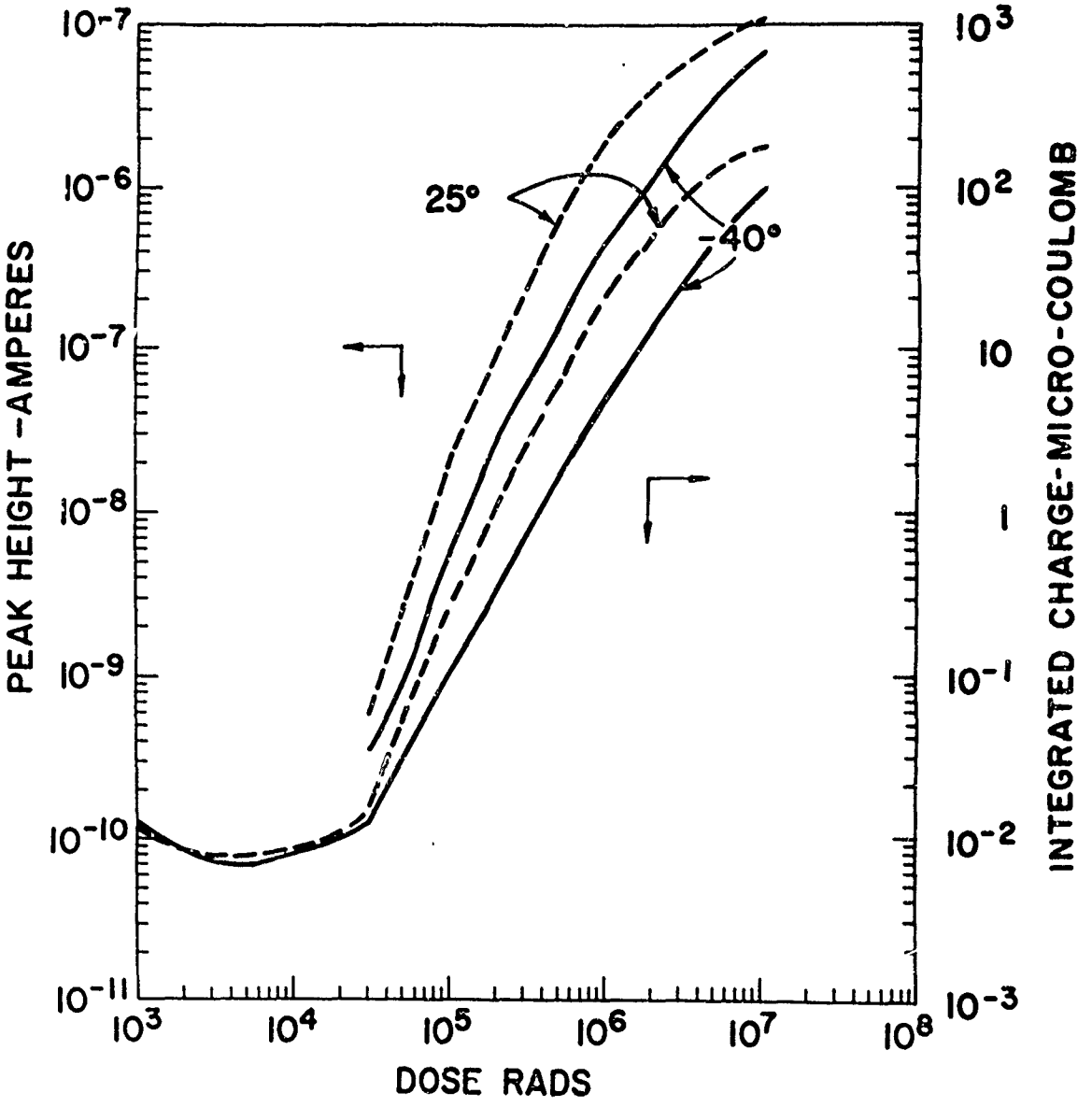

Figure 7. Calibration curves of Isomet LiF loose powder irradiated at -40 and 25°C.

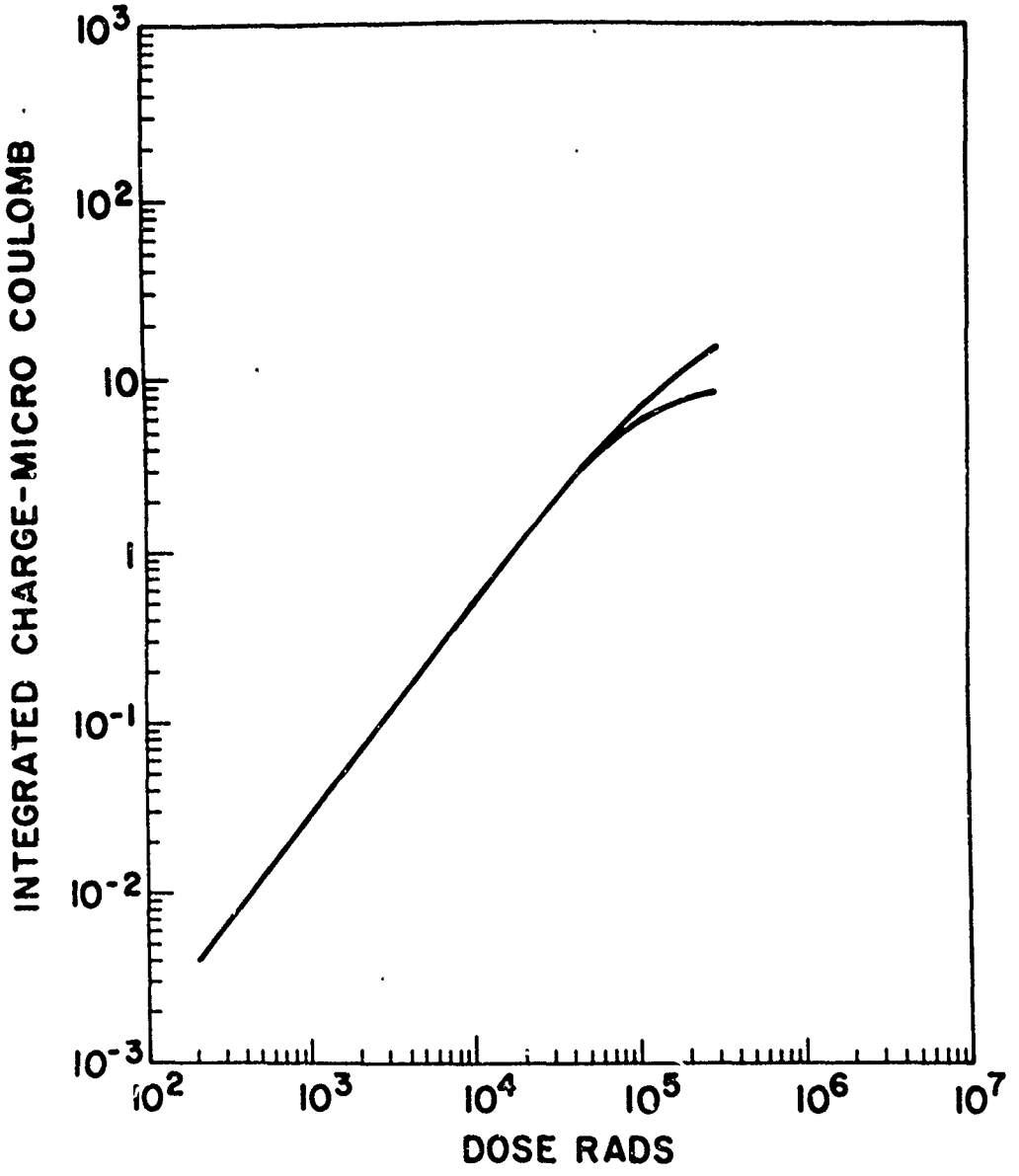

Figure 8. Response curves of Tyco LiF crystal. Lower curve is for a sample which was cycled by irradiating to a dose, then after readout, reirradiating to the next higher dose. Upper curve is the response curve of virgin crystals irradiated to the same doses.