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Berkeley, California

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November 3, 1964

ERRATUM

TO: All recipients of UCRL-11613, UC-37 Instruments
FROM: Technical Information Division
Subject: UCRL-11613, "Development, Testing, and Calibration of the
LRL Thermoluminescent Dosimetry System," Francis M. Miller
and Lowell M. Kleppe, August 24, 1964.

Please make the following corrections on subject report.

Page 3, section B, paragraph two: Richardson's equation should read:

$$I = A(1-r) T^2 \epsilon^{-e\phi/kT} \text{ (r in exponent should be e).}$$

Page 8, paragraph beginning eight line down, add "x rays" to the sentence

"The response of LiF to 100 kVp and 250 kVp x rays was"

Page 14, reference 6. "Lloyd P. Smith . . . pp 8-74 to 7-82 should read:

"pp. 8-74 to 8-82."

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ABSTRACT

The LRL lithium fluoride thermoluminescent dosimetry system is capable of measuring doses of gamma radiation from 10 mr to more than 30 000 r. Calibration curves were obtained for gamma radiation from cobalt and radium, x rays of 100 kVp and 250 kVp, high-energy protons and alpha particles, and for both 5-MeV and thermal neutrons. The problem of reducing variations in the photomultiplier dark-current magnitude and increasing the over-all stability of the system was accomplished by use of an infrared filter, the standardization of the operating procedure, and the cooling of the face of the PM tube with room-temperature dry-nitrogen gas.

This LiF dosimeter is currently being used in a variety of biomedical applications, some of which relate to cancer research and space medicine. Another application is in determining the field pattern of x-ray machines. In the future, personnel monitoring will undoubtedly utilize this dosimeter.

This readout system is being used to study shock- and strain-induced thermoluminescence in CaCO_3 (limestone).

I. INTRODUCTION

In the past 15 years continued research on the thermoluminescent properties of LiF crystals has led to the development of dosimetry systems, useful in the fields of Radiology, Radiation Biology, and Health Physics.^{1,2,3} The LiF crystals used in this experiment were purchased from the Harshaw Chemical Company of Cleveland, Ohio, and are known commercially as TLD-100.⁴ Lithium fluoride crystals containing pure Li⁶ and Li⁷ for special applications are also available under the designation of TLD-600 and TLD-700, respectively.

The development of the LRL thermoluminescent-dosimetry (TLD) system is under the direction of the Health Physics Department and is supervised by H. Wade Patterson, the Department Head. The system was built to provide an instrument capable of readout over a range of six decades — from 10^{-2} r to 10^4 r. The lower doses in the 10 mr to 100 mr range are of particular interest to the Health Physics Department due to the possible applications in the field of low-dose personnel monitoring and for relatively accurate measurement of the cumulative dose that the general public receives due to natural and man-made background radiation. The thermoluminescent response of LiF to ionizing radiation is essentially energy independent over a wide range,⁴ and the powder itself is tissue equivalent. These properties, combined with simplicity and small size of the LiF dosimeters, makes them ideal for personnel dosimetry. A variety of sizes and shapes facilitates implantations in biological specimens and phantoms. The dosimeters are self contained, require no external leads, and do not perturb the radiation field. Although initial cost of a dosimeter loaded with TLD-100 LiF is approximately \$1.70, both the capsule and powder are reusable as long as the total cumulative history of the powder is less than 100 000 r.⁵ The cost of the powder has been lowered from what it was originally and it is very likely that it will be reduced even more as this type of dosimetry comes in to wider use.

II. EQUIPMENT

A. General Description

The detection system includes a photomultiplier (PM) tube, a "bucking" current source (to reduce the effects of dark current, noise, and any other factors that may be inherent in the tube assembly, and to provide a control to minimize the effect of temperature variations in and around the phototube), and a voltage-to-frequency converter to change the PM signal to a frequency resulting in greater accuracy as well as range. Scalers are used to record the results visually. See Fig. 1.

An LRL regulated-power supply with a range of 0.5 kV to 3 kV provides the high voltage for the photomultiplier tube. This unit was found to be adequate for the regulation of the line voltage as the addition of a Sorensen voltage-stabilizer unit caused no measurable improvement in operation of this part of the apparatus.

MUB-4114

Fig. 1. Block diagram of LRL LiF-TLD system.

The phototube used is an EMI 9536-S (EMI Electronics, Ltd., Middlesex, England). The characteristics of this tube show decreased response to infrared radiation and also a low dark current.

A "bucking" current (Gyra Electronics, Model CS-51) is used to reduce the negative dark current of the PM tube. When this is done, the voltage-to-frequency converter "sees" the PM voltage due to the light emitted by the LiF crystals and thus the effect of the dark current of the phototube on the final reading on the scalers is reduced to a level that may be considered negligible for any readings above 1 r. If this dark current is not greatly reduced, a background number would have to be subtracted from each reading taken from the scalers. The subtraction of a background number from the scaler readings is still necessary for doses below 1 r where the scaler readings are not large enough to completely ignore the fluctuating background that had not been bucked out.

The voltage output of the PM tube is directly proportional to the amount of light given off by the LiF crystals when the dark current has been virtually eliminated by the bucking current. This dc voltage is then changed into ac pulses whose frequency is also proportional to the light emitted by the LiF crystals. A voltage-to-frequency converter (Vidar, Model 240) accomplishes this change.

The ac pulses are then passed through a discriminator that eliminates the negative portions of these pulses. This serves to eliminate the possibility of counting any part of the positive bucking current that may exceed the dark current. The counting of both positive and negative components of the pulses would give a scaler reading twice that necessary. Therefore only the positive pulses are counted.

Digitized readout is accomplished by the reading of five digits on a scaler, with additional units recorded on a four-digit register.

B. Operating Characteristics and History of Development

Initially the most notable difficulty with the present system was a persistent increase in the dark current from the PM tube occurring while the samples were heated and read out. The magnitude of the drift was on the order of 50 ± 25 counts/cycle, which was then equivalent to approximately 10 mr/cycle. A fluctuation of this magnitude prohibited the use of the machine for reading doses much below 100 mr with any reasonable accuracy. Therefore certain revisions were necessary before the system could be used for the dose ranges necessary for personal monitoring dosimeters.

The drift of the dark current was felt to be the result of temperature variations of the PM tube caused by infrared radiation from the heater and powder pan; therefore several methods were employed in an attempt to reduce this effect. The importance of temperature on the thermionic current I can be shown by Richardson's⁶ equation $I = A(1-r)T^2 e^{-\frac{\phi}{kT}}$, where $e = 2.718$, the base of the natural logarithm system; k is Boltzmann's constant, 1.380×10^{-16} erg-deg⁻¹; A and ϕ , the electronic work function, are constants depending on the emitting substance and the state of its surface; e is the electronic charge; T is the absolute temperature in °K; and

r is the reflection coefficient. The exponential variation of thermionic dark current with temperature prompts various attempts to control the photocathode temperature. (A 10°C. rise in photocathode temperature above room temperature multiplies the dark current approximately tenfold.)

Various filters and combinations of filters were placed between the heat source and the PM tube. A combination of two CS 1-69 filters for ir absorption and one far-red filter were first placed immediately below the PM tube and were successful in reducing the drift rate to 25 ± 25 counts/cycle. Next the filters were placed in a specially fabricated filter holder at an increased distance from the PM tube, because the filters themselves reached an elevated temperature and subsequently heated the photocathode both by re-radiation and convection. The filter holder allowed for an air space between each of the three filters to cut down on thermal transmission by conduction and convection. This arrangement resulted in a drift rate of 10 ± 25 counts/cycle. However, the sensitivity of the system was reduced by a factor of approximately 2.5 because of the reduction of the light-emitting solid angle "seen" by the PM tube, and by the absorption and reflections caused by the filters. Approximately 30 counts were then equivalent to 10 mr, again indicating difficulty in obtaining reliable readout of doses below 100 mr.

Next the readout system was operated in a refrigerated air-conditioned room. This reduced the drift still further but long periods of time were required for the establishment of thermal equilibrium and the variations in the resulting dark current were often ± 100 counts/cycle with no observable drift. This method of cooling was abandoned because of the resulting instability, which was believed to result from having too great a temperature gradient over the PM assembly.

We then attempted to maintain the PM tube at room temperature by passing room-temperature compressed air over the face of and around the PM tube. The rate of air flow was regulated from approximately one sixth to one fourth cubic foot per minute, as measured by a flow meter. Without LiF powder in the pan, stability was obtained without a detectable drift for periods longer than 1 hour and with a variation in stability ranging from ± 25 to ± 50 counts/cycle, depending on ambient temperature. However, when readouts up to 100 mr were attempted on freshly annealed unexposed powder, and on low-dose powder, variations in the dark current of ± 100 to ± 200 counts/cycle were common; these variations again prohibited the reading of doses of 100 mr or less. These variations were attributed to the effect of oxygen, in the compressed air, on the LiF powder. The effect is apparently a surface effect and is enhanced by the physical nature of the fine crystals.⁷ Therefore, the compressed air was replaced by high-purity dry nitrogen. This gas provided the desired stability and a measured variation of not more than ± 50 counts/cycle from the mean value of the dark current. The nitrogen flow is thought to be adequate for cooling with only a single ir filter in the system. The special filter block was removed and the CS 1-69 filter was placed approximately 1 cm from the PM tube, with the nitrogen passing directly between the face of the PM tube and the filter. A stable dark current was observed with nitrogen flow increased by about 50% over the previous flow rate. The sensitivity of the instrument was increased approximately fivefold, resulting in 145 counts equaling 10 mr, which number is considerably above the cycle-to-cycle variation in the dark current.

At this point the reason for the variation in the dark current was sought and the following explanation was postulated: With the bucking current turned off, the dark current produces approximately 2000 counts/cycle on the scalars. This means that the statistical variation expected in the dark current is approximately $\sqrt{2000}$, or ± 45 counts/cycle. This is in good agreement with the ± 50 counts/cycle now observed. This, then, appears to be a statistical limit below which the readout system cannot accurately read. Attempts to reduce this statistical variation by reducing the magnitude of the dark current fall into two categories. First, a PM tube can be selected for a low dark current. Second, the PM tube can be cooled to reduce the thermal noise.

A trial run was performed by the passing of nitrogen through a short piece of copper tubing immersed in liquid nitrogen. Although a reduction in the magnitude of the dark current was observed, it became evident that an excessively long period of time (an hour or more) would be required to reach temperature stability. Research indicated that lowering the PM temperature to 40°C below the ambient temperature only reduced the dark current to one-half, and that further reduction of the temperature within practical limits did not produce comparable or useful reductions in the dark current.⁸ Even with the dark current cut to one-half its previous value, the reduction in statistical variation is only proportional to $\sqrt{2}$. This fact, combined with the complexity of the cooling apparatus, led to abandonment of further attempts to cool the PM tube below ambient temperature.

III. CALIBRATION PROCEDURE

The accuracy of the powder dispenser was checked with an analytical balance. The mass of 50 LiF samples was determined to the nearest tenth of a milligram. The mean of the 50 determinations was 57.4 mg/sample. All but three of the samples were found to be within $\pm 4\%$ of the mean; the remaining three samples were within $\pm 6\%$ of the mean.

Samples of LiF were exposed to known doses of gamma radiation from calibrated sources of Co^{60} and Ra^{226} . Ten samples were exposed to doses of 1 r and ten to 100 r from Co^{60} . Three samples were exposed to various doses ranging from 10 mr to 10 r from the Ra^{226} source and a similar dose pattern from 30 mr to 30 000 r from Co^{60} . The variation in the readout of the 10 samples of powder with doses of 1 r and 100 r, respectively, was no greater than the variation caused by the powder dispenser. In order to read out doses in excess of 150 r, it is necessary to place one or more optical density filters between the sample and the PM tube in order to prevent the voltage-to-frequency converter from jamming. The calibration curves for the above exposures are shown in Figs. 2 and 3.

The procedure used when reading out samples below 100 mr is slightly different than that for reading out above 100 mr. For doses above 100 mr, the bucking current is adjusted to cancel out the dark current with an empty silver dish on the heater. However, when freshly annealed undosed powder is placed in the dish, an increase in the apparent dark current during the heating cycle corresponds to approximately 10 mr. Therefore, the bucking current is adjusted to eliminate this increase in "background" and a new balance is obtained. Then the doses below 100 mr are read out

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Fig. 2. Calibration curve for LiF readout system without optical filters.

○ - Ra²²⁶ data.
 □ - Co⁶⁰ data.

MUB-4116

Fig. 3. Calibration curves for LiF readout system with optical filters.

Upper line - Co^{60} data with optical filters Nos.1 and 2.
 Lower line - Co^{60} data with optical filters Nos.1, 2, and 3.

with increased accuracy. The cause for the increase in background with undosed powder in the dish is believed to be related to several factors. The increased area of the powder may radiate more infrared to the PM tube. One source indicated that exposure to light results in an increased readout equivalent to approximately 25 mr.⁹ Exposure to natural and man-made background radiation can also dose the powder. However, all of these effects can be successfully eliminated by adjustment of the bucking current.

The response of LiF to 100 kVp and 250 kVp⁷⁻¹⁰⁰ was investigated and the calibration curves are shown in Figs. 4 and 5.

The response of the TLD-100 to 5-MeV neutrons, and to 5-MeV neutrons moderated by 7.5 cm of paraffin was also investigated in order to determine the relative effect of fast vs slow neutrons, and to determine the relative contribution of fast neutrons to the total dose when the powder has been exposed to radiation consisting of both gamma rays and fast neutrons. To a first approximation, the moderated neutron response is 10 times the fast-neutron response. The response of LiF to fast neutrons is a small fraction of the response due to Co⁶⁰ gamma rays.

The calibration curves for high-energy protons and alpha particles are shown in Fig. 6.

IV. SOURCES OF ERROR

The observed variation in readout of various samples exposed to the same dose correlates well with the variation caused by the powder dispenser. A uniform annealing procedure results in no distinguishable differences between batches of powder. It is important that the dosimeter be completely emptied into a clean read out dish, and that the distribution of the powder in the dish be reasonably uniform. Care should be taken in controlling the heating cycle. Excessive heating of the pan above the glow-peak temperature should be avoided because of the resulting infrared contribution to the PM tube. At low dose rates, oxygen should be excluded during readout. All filters in the system should be cleaned periodically due to the evaporation of foreign matter from the heater. All of the above-mentioned items contribute to the readout error in the system. Occasionally an error of 20% or more is observed, even when mid-range doses are read out. It is believed that the statistical combination of all of these sources of error may produce such a variation. It is also possible that foreign matter may have prevented good thermal contact between the heater and the silver dish this prevention resulting in an abnormally low readout. Recycling the powder a second time may be done to release the remaining electrons. A small light leak is usually the cause of an erroneously high reading.*

*Notes concerning the annealing, care, and loading of the LiF and the dosimeter capsules and a detailed procedure for operating the LRL TLD readout system are available from the Health Physics Dept. of LRL.

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Fig. 4. LiF response to 100-kVp x rays, without optical filters.

MUB-4118

Fig. 5. LiF response to 250-kVp x rays, with optical filters Nos. 1 and 2.

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Fig. 6. LiF response to high-energy protons and alpha particles, with optical filters Nos. 1 and 2.

- $\bullet$ - 730 MeV protons.
- $\square$ - 910-MeV alpha particles.

V. APPLICATIONS

The sensitivity of the LiF powder makes it ideal for use in depth-dose determinations. Small capsules may be implanted in various biological specimens in varying patterns. Several groups at LRL are currently utilizing the LiF dosimeters in a variety of biomedical applications, some of which relate to cancer research and space medicine.

The field of a 250-kVp 15-mA x-ray machine was determined at both 100-kVp and 250-kVp with the LiF dosimeters. Sixty dose points in a field of 30-cm radius were determined. This calibration procedure clearly indicated that the field of an x-ray machine can be determined quite accurately at a great many points in a relatively brief time when the LiF is used for dose determinations. Any radiation field can now be determined within a short time interval to a high degree of precision by means of the TLD system.

Personnel monitoring will undoubtedly utilize the properties of LiF. Dose rates from reactors and accelerators may be measured conveniently with LiF capsules, particularly in areas not accessible while the reactor or accelerator is operating.

The readout system is being used to study shock- and strain-induced thermoluminescence in CaCO_3 (limestone).

VI. FUTURE DEVELOPMENT

In the event that other LiF readout systems are to be constructed or the present system is to be modified, certain changes that will improve and simplify the operation are suggested. The control of temperature is an important factor. Nitrogen flow between the PM tube face and the ir filter is a reasonably simple method of controlling photocathode temperature. Ambient-temperature control could be assured by placing the entire system in a temperature-controlled room. A temperature near or somewhat above 25°C may be suitable. A dc heating source would eliminate the main source of 60 cycle ac pickup. A shorter heating cycle would then be possible. However, this may result in a high-intensity glow peak that would cause the voltage-to-frequency converter to jam, unless optical density filters were used to reduce light transmission to the PM tube. Electronic timers would be preferable to the present mechanical timers that sometimes cause large current pulses at the beginning and end of the timer cycle; these pulses result in spurious counts registering on the scalers. It is also possible that an automatic cycling system be incorporated for rapid readout. Advanced scaler units and transistorized high-voltage supplies could be applied advantageously in this system.

VII. SUMMARY

The LRL TLD system successfully covers a dose range exceeding six orders of magnitude. The largest error is in reading doses of approximately 10 mr and is on the order of 35%. Errors of 5% or less are to be expected for reading above 1r. Lithium fluoride is particularly desirable as a dosimeter because of its linear response to a wide range of ionizing radiations, its tissue equivalence, and the various sizes and shapes that are possible for dosimeter capsules.

The temperature-caused drift of the PM tube can be controlled with a relatively simple system that passes nitrogen gas over the infrared filter and the face of the PM tube to carry off excess heat as it builds up.

Although the properties of LiF are not fully understood, reliable dosimetry measurements are now possible. Applications will certainly increase with continued usage and availability of these dosimeters.

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