CHARACTERISTICS OF TEFLON-LiF
THERMOLUMINESCENT DOS IMETERS

G. W. R. Endres

JANUARY 1968

AEC RESEARCH &
DEVELOPMENT REPORT

BATTELLE

NORTHWEST

BATTELLE MEMORIAL INSTITUTE

PACIFIC NORTHWEST LABORATORY

3000 STEVENS DRIVE, P. O. BOX 999, RICHLAND, WASHINGTON 99352

LEGAL NOTICE

This report was prepared as an account of Government sponsored work. Neither the United States, nor the Commission, nor any person acting on behalf of the Commission:

A. Makes any warranty or representation, expressed or implied, with respect to the accuracy, completeness, or usefulness of the information contained in this report, or that the use of any information, apparatus, method, or process disclosed in this report may not infringe privately owned rights; or

B. Assumes any liabilities with respect to the use of, or for damages resulting from the use of any information, apparatus, method, or process disclosed in this report.

As used in the above, "person acting on behalf of the Commission" includes any employee or contractor of the Commission, or employee of such contractor, to the extent that such employee or contractor of the Commission, or employee of such contractor prepares, disseminates, or provides access to, any information pursuant to his employment or contract with the Commission, or his employment with such contractor.

PACIFIC NORTHWEST LABORATORY

RICHLAND, WASHINGTON

operated by

BATTELLE MEMORIAL INSTITUTE

for the

UNITED STATES ATOMIC ENERGY COMMISSION UNDER CONTRACT AT(45-1)-1830

PRINTED BY/ FOR THE U. S. ATOMIC ENERGY COMMISSION

UNCLASSIFIED

3 3679 00061 2913

BNWL-741

CHARACTERISTICS OF TEFLON-LiF
THERMOLUMINESCENT DOSIMETERS (a)

by

G. W. R. Endres (b)

Battelle Memorial Institute
Pacific Northwest Laboratory
Richland, Washington

January, 1968

Paper presented at the Symposium on Personnel Radiation Dosimetry held in Chicago, Illinois, January 30-31 and February 1, 1967.

- (a) This paper is based on work performed under United States Atomic Energy Commission Contract AT(45-1)-1830.
- (b) Senior Development Engineer, Environmental and Radiological Sciences Department, Pacific Northwest Laboratory, operated by Battelle Memorial Institute for the United States Atomic Energy Commission, Richland, Washington.

TABLE OF CONTENTS

LIST OF FIGURES	iii
LIST OF TABLES	iii
INTRODUCTION AND THEORY	1
CHARACTERISTICS	2
CONCLUSIONS	17
ACKNOWLEDGEMENTS	18
REFERENCES	19
DISTRIBUTION	20

LIST OF FIGURES

1	Finger Ring Dosimetry	3
2	Energy Response	5
3	Exposure Response, 1/2" Teflon Discs, 3/8" Hole in Planchet	7
4	Exposure Response, Oblong Discs - 3/8" Diameter Hole	8
5	Exposure Response Curve Background Corrected	9
6	Exposure Response Curve Background Corrected	10
7	Uranium Beta Exposure Response	11
8	Response vs Time After Exposure	14

LIST OF TABLES

I	U β Exposures - Bare and Shielded	13
II	TLD Disc Exposures	16

CHARACTERISTICS OF TEFLON-LiF
THERMOLUMINESCENT DOSIMETERS

INTRODUCTION AND THEORY

The mechanisms of thermoluminescence have been known for many years and studied by a number of people.^(1, 2) Many substances such as CaSO_4 , CaF_2 , and LiF , exhibit the property of thermoluminescence, in fact, most solid state materials thermoluminesce to some extent. In many respects, the most suitable material to use for thermoluminescent dosimetry is the phosphor LiF (lithium fluoride) activated with a small amount of manganese impurity.

When a thermoluminescent phosphor is exposed to ionizing radiation, many of the freed electrons (or holes) become trapped at lattice imperfections caused by the impurities in the crystalline solid. In certain phosphors they may remain trapped almost indefinitely if the phosphor is not stored at too high a temperature. If the temperature is raised, the probability of escape from the trap sites is increased and the electrons (or holes) are released to recombine later with opposite charge carriers with emission of light during the process. The plot of light output as a function of temperature is called a glow curve; any peaks in the curve are called glow peaks.

The total amount of light emitted during the heating process is proportional to the number of electrons released from the traps. The total number of electrons in the traps is proportional to the amount of radiation exposure. The light sum during part or all the glow curve, or

the height of a glow peak can be used as a measure of radiation exposure. After readout, the phosphor is essentially returned to its original condition and is ready to be used again.

So far all the thermoluminescent studies conducted at Battelle-Northwest Laboratories have used the phosphor LiF. The program was originally directed toward the development of an improved personnel neutron dosimeter.⁽³⁾ Since the phosphor is sensitive only to charged particles and only indirectly sensitive to neutrons, a hydrogenous material (ethyl alcohol) must be added to the phosphor used to detect neutrons. Then any gamma sensitivity must be subtracted from the phosphor response to obtain the neutron exposure. This process required two dosimeters and an accurate knowledge of the properties of the phosphor when exposed to gamma, x-ray radiation, and neutrons.

Considerable amount of time and effort has been spent measuring these properties of LiF. For such studies we used a TLD reader purchased from Controls for Radiation. This reader is one of the very first units sold by Conrad and has been modified several times to improve its performance. The instrument is designed for use with LiF phosphor in either the granular form or in teflon matrix.

CHARACTERISTICS

Of concern here are the characteristics of the phosphors in a teflon matrix. See Figure 1. This work was done during 1966 and 1967, after the phosphor in teflon became available. The properties of the

Figure 1
Finger Ring Dosimetry

teflon-LiF are similar to those of the loose phosphor except, of course, the handling of the dosimeters is much more convenient since there is no powder to measure out or spill.

The energy response of the teflon-LiF in the form of 1/2" dia. discs 0.015 inches thick, was measured using gamma and x-ray sources available in the Calibrations Facility.⁽⁴⁾ In each exposure, 5 discs were exposed to 1 R of gamma or x-ray radiation and then readout in the prescribed manner using a nitrogen flush. The nitrogen atmosphere minimizes spurious light emission caused by oxygen present in an atmosphere. The phenomenon of the spurious emission is not well understood, but readout in N₂ has now become common practice. The results of the energy response measurements given in Figure 2 show an increase in sensitivity of the teflon-LiF below 100 keV rising to a maximum between 20 and 25 keV. The error limits shown are 95% confidence limits based on 5 measurements.

The teflon-LiF material is available in forms other than 1/2" discs. Most of the studies were conducted with the 1/2" discs as a matter of convenience. Rods with dimensions of 1 mm in diameter by 6 mm long are also commonly available. Except for lower sensitivity, the characteristics of the rods are similar to those of the discs.

The exposure response of the teflon-LiF is linear within the errors of measurement up to exposures to 10 R radium gamma. Other experimenters have shown a linear response up to 10³ R. Above 10³ R the response of the phosphor is superlinear. The reasons for the superlinear response are not

Figure 2
Energy Response

well understood. The response was measured for the 1/2" dia. disc and for oblong discs cut to fit the new personnel hand dosimeter. See Figures 3 and 4. Both types of discs were readout with a heater planchet having a 3/8" hole in the retainer screen. The screen is used to hold the discs in good thermal contact with the heating surface of the planchet. The minimum detectable dose is about 50 mR with the earlier readout equipment and geometry.

After modification of the reader to improve the operating characteristics, a new set of calibration curves were determined. At the same time, new 1/2" discs with more Li^7F in them were obtained. The exposure response is still linear from 25 mR (the lower detection limit) to 20 R. With the new reader the sensitivity is higher so that the system as a whole is about five times as sensitive as before. The new sensitivity of the system is shown in Figures 5 and 6.

The sensitivity of the teflon- LiF to beta radiation was examined by a set of exposures up to 1000 mrad of uranium beta. The exposures were made with the discs on a slab of natural uranium. Again, the response was linear, but was only about half the response of the discs to gamma radiation. This difference in response leads to difficulty in interpreting hand dosimeter measurements when a combined γ and β exposure occurred. The uranium β calibration curve is shown in Figure 7.

In an attempt to devise a system to more accurately evaluate mixed β and γ exposures, several experiments were conducted with the improved reader

Figure 3
Exposure Response, 1/2" Teflon Discs, 3/8" Hole in Planchet

Figure 4
Exposure Response, Oblong Discs - 3/8" Diameter Hole

Figure 5
Exposure Response Curve
Background Corrected

Figure 6
Exposure Response Curve
Background Corrected

Figure 7
Uranium Beta Exposure Response

using absorbers and the 1/2" teflon discs stacked on top of each other. Results of the experiments using aluminum absorbers indicate that the minimum detection limit is about 200 mrad of uranium beta radiation. This technique utilizes the difference reading between shielded and unshielded discs as a measure of the beta exposure. In this case a piece of aluminum 0.030 inches thick was used as the shield. The results are shown in Table I.

A series of exposures to uranium beta and 16 keV and 52 keV x-ray radiation was conducted to see if a degree of separation between beta and γ exposure could be accomplished by stacking three discs on top of each other. The discs themselves are not as effective in shielding the beta radiation as the 0.030" aluminum shield and thus, do not provide an adequate difference reading. At 200 mrad exposures the difference between the disc closest to the source and the disc farthest away was about 20 per cent. The difference using the aluminum absorber was about 50 per cent for a 200 mrad exposure.

The decay of the light output as a function of time after exposure was investigated using sets of discs exposed to 500 mR radium gamma radiation. A decay of approximately 10 per cent was observed when the discs were readout 10 days after exposure. These results are shown in Figure 8. The readings were normalized with a standard light source to help eliminate any gain changes in the electronic system. After a 10-day waiting period, there is a slight increase in light output. These effects have been noted by other experimenters and were discussed at the International Conference on Luminescence Dosimetry at Stanford in June, 1965.

TABLE I

U β Exposures - Bare and Shielded

	<u>Bare</u>	<u>.030" al</u>
1 rad	516 $\pm$ 79	212 $\pm$ 24
500 mrad	344 $\pm$ 17	150 $\pm$ 18
200 mrad	222 $\pm$ 2	151 $\pm$ 12
100 mrad	150 $\pm$ 11	137 $\pm$ 25
50 mrad	139 $\pm$ 35	153 $\pm$ 15
25 mrad	136 $\pm$ 7	133 $\pm$ 10
Background	121 $\pm$ 5

Figure 8
Response vs Time After Exposure

The discs may be used after an annealing procedure is completed. The initial annealing before first use is conducted by the manufacturer by heating the discs at 300° C for 16 hours, then at 80° C for 6 hours. Before reuse of the dosimeters, they are annealed at 300° C for 15 minutes followed by 80° C for two hours. The sensitivity of the discs used in the preceding experiments was measured after the second annealing and was found to be (on the average) about 20% less than the sensitivity before annealing. Later annealing studies have shown that a better annealing formula is two hours at 300° C and two hours at 80° C with slow cool to room temperature.

A special study was conducted using the discs through 10 cycles without annealing between each use to determine the characteristics of the discs without normal annealing. Sets of six discs were exposed to 300 mR of 17 keV and 52 keV x-rays and radium γ radiation each day for 10 days. The discs were readout each day but were not annealed before the next exposure. After the first day, the readout held constant to within about 10 per cent except for the last day when the average was high about 20 per cent. Table II is a summary of these data. These data indicate that the standard annealing may not be needed when working with low exposures. The readout cycle seems to provide the necessary annealing, but it is best to use a standard annealing before each use. Although the manufacturer says the sensitivity of the discs may vary by as much as $\pm 30\%$ after annealing, other experimenters⁽⁵⁾ have devised an annealing procedure that does not affect the sensitivity of the phosphor.

TABLE II

TLD Disc Exposures

<u>Day</u>	300 mR <u>17 keV</u>	300 mR <u>52 keV</u>	300 mR <u>Ra γ</u>
1	94	87	88
2	128	111	118
3	122	110	102
4	127	112	108
5	129	111	105
6	124	112	104
7	120	107	101
8	133	120	109
9	130	118	109
10	143	133	123

The numbers are an average of six readings.

Nevertheless, the sensitivity of each batch or group of discs should be measured after each annealing. The dosimeter may be used as many as 20 times or more.

For the group of discs used in these studies, the background reading including photomultiplier dark current and spurious thermoluminescent effects, is about 25 per cent of the 200 mR signal. It appears necessary to read an unirradiated disc in any measurement series to determine the background.

The teflon-LiF is essentially impervious to mechanical disturbance. They can be dropped, hammered or otherwise abused with negligible results. They do, however, become slightly activated when exposed for long periods of time to normal laboratory lighting (fluorescent). The discs are normally kept in a light-tight box and covered during exposure. Of course, the discs will accumulate the natural background radiation also.

The statistical uncertainties for all the exposures in this study were calculated using five measurements for each data point and 95% confidence. The uncertainties vary from 1 to 8 per cent and average about 5 per cent except for exposures close to the detection limit. The uncertainty on individual measurements is between 10 and 20% except close to the detection limit.

CONCLUSIONS

For use in finger rings to measure exposure to the extremities, TLD discs offer many advantages over the present film devices. The discs require a minimum of handling, do not need processing before readout, and

may be loaded in normal room light without inducing spurious readings. The relatively flat energy response eliminates the necessity for making large corrections when working with low energy radiation sources. Generally, the measurements are more accurate than those obtained using film. We recommend the use of the new oblong finger rings developed by L. F. Kocher of the Biophysical Sciences Section because of the better comfort to the wearer.

ACKNOWLEDGEMENTS

The author wishes to thank Mr. D. L. Haggard who readout many of the exposed discs and helped plot the results. We are also indebted to Mr. H. A. Meloeny of the Calibration Facility where the calibrated exposures were conducted.

REFERENCES

1. F. H. Attix. Present Status of Dosimetry by Radiophotoluminescence and Thermoluminescence Methods, NRL-6145. 1964.
2. Luminescence Dosimetry Proceedings of International Symposium on Luminescence Dosimetry, Stanford, California, June, 1965. Published 1967, 8th AEC Symposium Series.
3. Dosimetry Technology Staff, BNWL-339.
4. H. V. Larson, I. T. Myers, W. C. Roesch. Nucleonics, Vol. 13, 11, p. 100, November, 1955.
5. F. V. Cipperly. A General Summary of Personnel Monitoring Systems Utilized at the National Reactor Testing Station, ID-12056, September, 1966.

DISTRIBUTION

Number
of Copies

5	<u>Atomic Energy Commission, RL</u> C. L. Robinson A. Brunstad R. K. Sharp J. R. Yesberger Technical Library
1	<u>Atlantic Richfield Hanford Company</u> G. E. Backman
2	<u>Douglas United Nuclear, Inc.</u> C. D. Corbit File
2	<u>Atomic Energy Commission, Chicago</u> G. H. Lee Patent Group
1	<u>United States Testing</u> W. V. Baumgartner
60	<u>Battelle-Northwest</u> L. A. Braby P. E. Bramson L. W. Brackenbush R. O. Budd J. P. Corley F. T. Cross G. W. R. Endres (25) W. L. Fisher D. M. Fleming P. C. Friend J. J. Fuquay W. A. Glass A. J. Haverfield J. F. Honstead

Number
of Copies

Battelle-Northwest (contd.)

W. P. Howell
R. L. Kathren
A. R. Keene
L. F. Kocher
H. V. Larson
D. McConnon
I. C. Nelson
C. E. Newton, Jr.
L. L. Nichols
K. G. Pailthorp
H. E. Palmer
F. L. Rising
N. M. Robinson
R. H. Scott
J. M. Selby
W. E. Wilson
N. G. Wittenbrock
Technical File (5)