ORP/EERF 73-1

SUITABILITY OF GLASS-ENCAPSULATED CaF_2:Mn THERMOLUMINESCENT DOSIMETERS FOR ENVIRONMENTAL RADIATION SURVEILLANCE

U.S. ENVIRONMENTAL PROTECTION AGENCY

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FOREWORD

The Eastern Environmental Radiation Facility participates in the identification of solutions to problem areas as defined by the Office of Radiation Programs. The Facility provides laboratory capability for evaluation and assessment of radiation sources through environmental studies and surveillance and analysis. The EERF provides technical assistance to the State and local health departments in their radiological health programs and provides special laboratory support for EPA Regional Offices and other federal government agencies as requested.

Charles R. Porter
Acting Director
Eastern Environmental Radiation Facility

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ABSTRACT

The suitability of glass-encapsulated CaF_2:Mn thermoluminescent dosimeters for environmental radiation surveillance was investigated. More than two hundred dosimeters were subjected to extensive laboratory and field tests. Various parameters such as accuracy, precision, sensitivity, self dosing, and fading were investigated.

Selected dosimeters of this type can be used for accurate determination of environmental radiation levels if certain precautions are taken. Such precautions are proper calibration and determination of self-dosing characteristics for individual dosimeters.

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Introduction

The need for a small, accurate dosimeter to measure the natural gamma background radiation around nuclear facilities and to quantitate the increase in exposure due to plant operations has been recognized for some time. The proposed exposure rates in 10 CFR Part 50, Appendix I, require that this dosimeter be sensitive enough to measure increases on the order of 10 mR/yr. Since thermoluminescent dosimetry is rapidly becoming one of the most common techniques for measuring ionizing radiation, it is logical that it be considered for environmental measurements. Glass-encapsulated CaF_2:Mn thermoluminescent dosimeters are presently being used at many nuclear facilities for environmental monitoring purposes. This report, a result of a joint study by the Tennessee Valley Authority (TVA) and the Eastern Environmental Radiation Facility (EERF) of the U. S. Environmental Protection Agency (EPA), investigates the characteristics and suitability of this type dosimeter for environmental radiation measurements.

Physical Characteristics

The dosimeter studied was the EG&G Model TL-15 (figure 1). The thermoluminescent material employed in the dosimeter is CaF_2, manganese activated, and is in the form of two hot-pressed chips which are held on either side of a flat

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heating element. The chips and the heating element are enclosed in a glass envelope which is filled with CO_2 at atmospheric pressure. When in use, the glass envelope is placed in an energy compensating shield made of tin, aluminum, and lead. The shield reduces the over-response of CaF_2:Mn to low energy gamma radiation.

The reader employed was the EG&G Model TL-3 equipped with a low noise photomultiplier (PM) tube (figure 2). Dosimeters are read when a constant current of 6.5 amperes is passed through the heating element of the dosimeter. The heated CaF_2:Mn emits light which is proportional to the magnitude of the exposure. The resulting glow curve which depicts light intensity vs. time or temperature, is plotted on a 5-inch strip chart recorder. The magnitude of the exposure is determined not by integrating the glow curve but by measuring the height of the peak.

Calibration Procedures

Special calibration equipment was constructed that is capable of delivering accurate and reproducible exposure to the dosimeters. The facilities were especially constructed to provide reproducible positioning of dosimeters and to minimize errors introduced by scatter.

At EERF several NBS calibrated radium-226 sources were used to expose the dosimeters. Radium was chosen because its energy spectrum nearly simulates the radiation energies encountered in environmental exposures. Throughout the study the dosimeters were given exposures ranging from 3 mR to 11 mR. This is the range of exposure expected in environmental monitoring situations for periods of a month.

At TVA, a calibrated 0.1 Ci ^{137}Cs source was employed to irradiate the dosimeters. An exposure ring was constructed that positions the dosimeters in a circular ring with the source at the center. Exposures from 1 to 5,000 mR were used in these investigations.

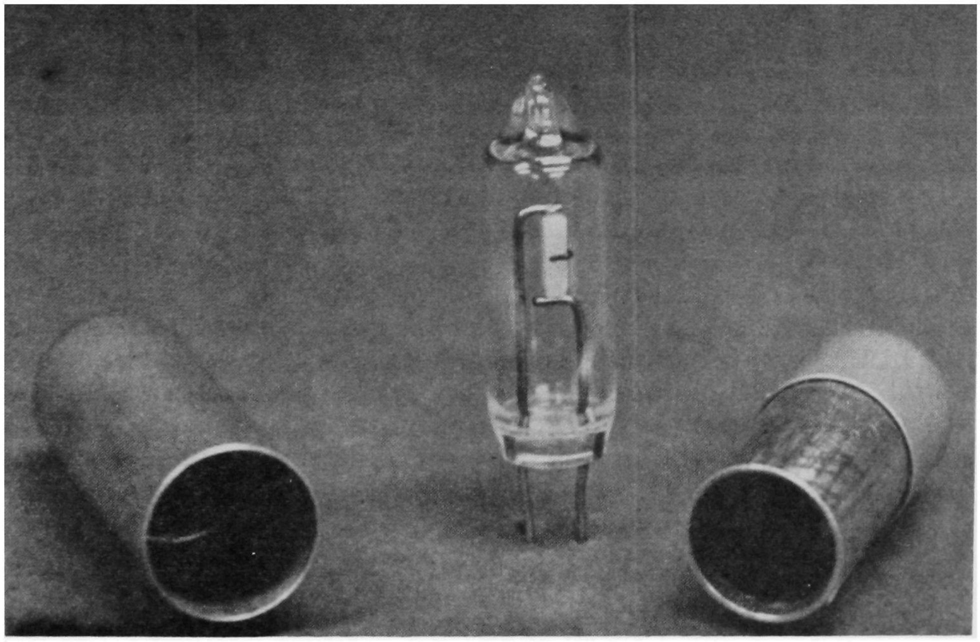

Figure 1. EG&G Model TL-15 Dosimeter

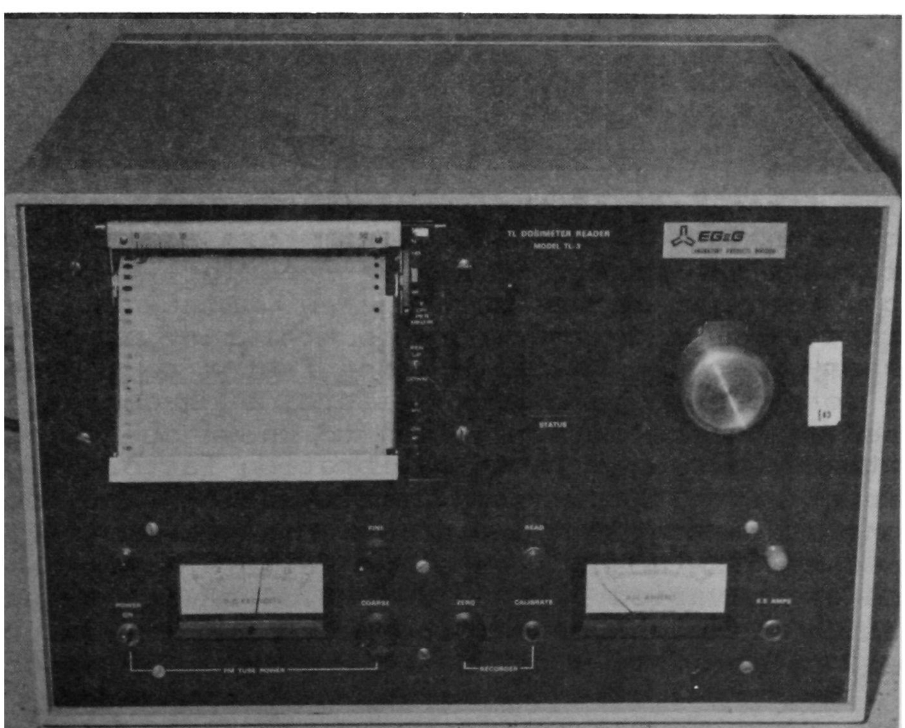

Figure 2. EG&G Model TL-3 Dosimeter Reader

Annealing

Prior to using a dosimeter for making a measurement, it is necessary to anneal the dosimeter to eliminate the stored signal from past exposures. Exposures up to 5 R were made and different annealing techniques were used to determine the most satisfactory. An annealing method was determined to be satisfactory if following an exposure the dosimeter was annealed and read and no detectable reading was obtained. For exposures up to 100 mR, which are possible in environmental surveillance, complete annealing can be obtained by simply putting the dosimeter through an instrument readout cycle as used when making a normal measurement. For exposures higher than 100 mR, the following oven annealing procedures were determined to be satisfactory.

Exposure (mR)	Annealing Procedure
100 - 500	5 minutes at 350^0 C or 2 readout cycles
500 - 3,000	10 minutes at 350^0 C
3,000 - 5,000	15 minutes at 350^0 C

Calibration and Precision

1. Individual Dosimeters

The first part of the investigation was to determine calibration factors for each dosimeter, and from this information the precision and reproducibility for the group of dosimeters was quantitated. Since the reading provided by each dosimeter is in arbitrary units, it is necessary to obtain a calibration factor to convert these units to exposure in mR. Therefore, each dosimeter was exposed a minimum of five times to known quantities of radiation and read so that calibration factors might be obtained. From these data, a mean calibration factor and one sigma standard deviation were calculated for each dosimeter. The mean calibration factors for the dosimeters in the group varied from 0.23 to 0.34 mR/reader unit. This variation dictates that, for precise work, each dosimeter be well calibrated. The standard deviations associated with the measurement of the calibration factors were averaged for the group of 162 dosimeters and was 2.9 percent. This represents the precision or reproducibility of this dosimeter-reader system at typical environmental radiation levels (1 to 50 mR) under laboratory conditions.

The major factors which account for the variations in individual response were found to be the errors in reader adjustments. These adjustments are the strip chart zero, the PM tube high voltage, and the heater current.

The zero adjustment is accomplished by inserting the empty read head into the reader and manipulating the control until a zero reading is obtained on the recorder.

To adjust the high voltage applied to the PM tube a reference light source with an equivalent mR reading is supplied by the manufacturer. The adjustment is made by inserting the light source into the reader and adjusting the high voltage until the equivalent mR reading is obtained on the chart. Considerable differences (20 percent) in the mR equivalence assigned by the manufacturer were found to exist among several light sources. This would prohibit the use of dosimeters on different readers adjusted with different light sources unless the dosimeters were calibrated on each system. With a single reader the ability to reset the high voltage to a known mR equivalent is satisfactory.

The adjustment of heater current to 6.5 amps is accomplished with the aid of a built-in ammeter. Variation in this adjustment will effect a ±3 percent error per 0.1 amp between 6.1 amps and 6.8 amps. Other minor sources of error were due to timing of the calibration exposures and operator error in reading the strip chart recorder.

2. Dosimeters as a Group

To quantitate the variation in response from one dosimeter to another due to the manufacturing process, an average calibration factor for three groups of dosimeters was determined. The three groups were from three production batches--Group F (53 dosimeters), Group G (34 dosimeters), and Group L (75 dosimeters).

The results, summarized in table 1, show the variation in relative response from batch to batch. The variation in relative response of individual dosimeters within each group is indicated by the standard deviation associated with the mean calibration factor for the group. These data illustrate the essentialness of individual calibration factors for each dosimeter.

Table 1

Calibration factors

	Group F (53)	Group G (34)	Group L (75)
Range	.259 - .339	.239 - .313	.230 - .268
Mean ± σ (%)	.280 ± 5.4%	.269 ± 6.7%	.247 ± 3%

Internal Background or Self Dosing

The most significant aspect studied was the internal background or self-dosing properties of the dosimeters. The internal background is due to the irradiation of the thermoluminescent material by the radioactivity in the materials used to construct the dosimeter.

The internal background was determined by placing annealed dosimeters in a storage shield with a known background. This background exposure rate was determined using a calibrated high pressure ionization chamber. The dosimeters were left in the shield for a period of 72 to 120 hours before being read. The reading obtained was a result of a combination of the internal background of the dosimeters plus the natural background in the shield. By subtracting the natural background from the dosimeter reading the internal background was determined. A minimum of five measurements was performed with each individual dosimeter. From these determinations a mean internal background and one sigma standard deviation were calculated for individual dosimeters and for the dosimeters as a group. The results are summarized in table 2. The wide range of internal backgrounds for the various groups indicates that this parameter should be determined for each dosimeter.

Table 2

Internal Backgrounds for Groups of Dosimeters
μR/hr

	Group F	Group G	Group L
Range	7.34 - 33.93	11.10 - 61.00	2.34 - 15.69
Mean ± σ	13.45 ± 5.52	25.37 ± 9.95	3.78 ± 2.58

It is concluded that the primary contributor to the internal backgrounds is ^{40}K in the glass envelopes. A sample of the glass used to construct the envelopes for Groups F and G was examined and found to contain 20 pCi of ^{40}K per gram of glass. Glass for the L Group was found to contain 0.5 pCi of ^{40}K per gram of glass.

NBS Handbook 80 defines the minimum detectable level as three times the standard deviation associated with the background measurement. Therefore, the standard deviation associated with the measurement of the internal background for each dosimeter is important. As an example, assume a typical dosimeter from Group F which has an internal background alone. Therefore, the minimum detectable exposure for a 1-month monitoring period using this dosimeter would be three times 0.44 mR or 1.32 mR, which is below the monthly natural radiation exposure levels.

The average of the individual dosimeter standard deviations from the mean internal background for the dosimeters in each group is given in table 3. An indication of the average minimum detectable exposure for each group is three times the average standard deviation for the group.

Table 3

Minimum Detectable Exposure Rate

	Group F	Group G	Group L
Average σ	0.52 μR/hr	1.33 μR/hr	0.31 μR/hr
M.D.E. (3σ)	1.56 μR/hr	3.99 μR/hr	0.93 μR/hr

Fading as a Function of Temperature

After an exposure, the reading produced by the dosimeter fades with time. An investigation was conducted to quantitate the degree of fading at three environmental temperatures, 18^0 F, 75^0 F, and 130^0 F. It has been observed that essentially all of the fading occurs within the first 24 hours. The maximum fading at the three temperatures is shown in table 4.

Table 4

Fading of Reading with Time

Temperature	Relative Reading with No Time Delay Between Exposure and Readout	Relative Reading with 24-hour Delay Between Exposure and Readout
18^0 F	1.00	0.97
75^0 F	1.00	0.96
130^0 F	1.00	0.95

Since fading is a significant factor, it must be considered for all measurements. Two methods of quantitating the effect are possible. The first is, during dosimeter calibration, to fix the time between exposure and readout to less than one hour. In this manner, the correction factors shown above need to be applied when making field measurements. In contrast, if during dosimeter calibration the time between exposure and readout is fixed at 24 hours or greater, fading and correction factors need not be considered for experimental exposures.

Sensitivity to Light

A dosimeter which has been exposed to radiation will be affected by sunlight and office fluorescent lights, and will cause the dosimeter reading to be less than if it had not been subjected to light. When exposed to sunlight, a dosimeter will lost 40 percent of its reading in 5 minutes and 90 percent in 10 minutes. When exposed to office fluorescent light, a 5 percent reduction is seen in 20 minutes and a 20 percent loss in two hours. Therefore, following an exposure to radiation, the dosimeters must remain in the shield until they are read so as to prevent loss of response.

For dosimeters that have been annealed and not exposed to radiation, sunlight and office fluorescent lights do not cause a buildup of energy within the dosimeter that would produce a reading that could be interpreted as having been an exposure to radiation.

Shock and Vibration Effects

Exposed and unexposed dosimeters were subjected to sudden shock (drop on a hard concrete floor) and steady vibration (60 Hz) and no effect on the anticipated dosimeter readout was seen.

Electromagnetic and Ultrasonic Effects

Exposed dosimeters were subjected to electromagnetic (2450 MHz, 200 mW/cm^2 for 30 minutes) and ultrasonic fields of varying intensity (ultrasonic cleaner for 30 minutes). These studies were performed with the dosimeters inside and outside their protective shields. No effect could be observed with either electromagnetic or ultrasonic fields.

Aging Effects

Preliminary results indicate an aging effect on the dosimeters. This aging effect causes an increase in the calibration factor or a decrease in the sensitivity (less reading for the same exposure). The effect appears to be time-dependent and not a function of use. This was established by exposing and reading a group of new dosimeters six times over a period of three days. These dosimeters showed no change in their calibration factors. Another group which had been exposed and read six times over a period of approximately one year showed an 8 percent increase in calibration factors.

This aging effect might be due to a darkening of the crystal which is observed in the older (over one year) dosimeters. This darkening could cause a decrease in the light output, thereby reducing the response for a given exposure.

Operating Experience

The dosimeters have been used for up to four years to measure the natural background gamma exposure rates in the vicinity of nuclear power plants under construction. The dosimeters are placed on metal stakes so that the exposure rate is monitored one meter from the ground. The stakes are positioned on a 500-foot grid surrounding the plants. Fifty-four dosimeters are used at one site while forty-four are employed at the other. The dosimeters are exposed in the field for three months.

As a result of the work described herein, the exposure rate is calculated by using the following formula:

Formula for the Determination of Exposure Rates in the Environment

$$\frac{(\text{Dosimeter reading})\ (\text{calibration factor})}{(\text{fading factor})\ (\text{hours exposed})} - \text{internal background rate} =$$

$$\text{environmental exposure rate}$$

where the fading factor is 0.96, and the internal background rate is dependent upon the dosimeter.

When dosimeters are used to monitor the levels of radiation around a plant site, experience has shown that at least one year's data are necessary to accurately quantitate the annual exposure levels. This period would allow sufficient time to quantitate any seasonal trends and provide enough information so that an accurate annual average exposure rate might be obtained through appropriate data reduction.

Conclusion

Selected glass-enaapsulated CaF_2 dosimeters can be used for environmental radiation monitoring with reasonable accuracy if certain conditions are met. Determination of internal backgrounds and calibration factors for individual dosimeters is necessary. Fading characteristics should be typical for all the dosimeters in the group. The possibility of an aging effect warrants a reevaluation of calibration factors on an annual basis.

In order to detect an increase of 10 mR/yr above background as proposed in 10 CFR Part 50, Appendix I, it is imperative that measurements be made with great care and accuracy. A monitoring program that utilizes CaF_2 glass-encapsulated dosimeters should be initiated at least one year prior to plant operation, and detailed statistical analysis of these data will be required to determine if the radiation exposure rate prior to plant operation is different than after plant startup.

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ERRATA

The paragraph beginning "NBS Handbook 80 defines..." on page 7, should read:

NBS Handbook 80 defines the minimum detectable level as three times the standard deviation associated with the background measurement. Therefore, the standard deviation associated with the measurement of the internal background for each dosimeter is important. As an example, assume a typical dosimeter from Group F which has an internal background of 9.08 ± 0.58 μR/hr was placed in the field for a one month monitoring period. At the end of the 750-hour monitoring period the dosimeter would have accumulated 6.81 ± 0.44 mR from internal background alone. Therefore, the minimum detectable exposure for a 1-month monitoring period using this dosimeter would be three times 0.44 mR or 1.32 mR, which is below the monthly natural radiation exposure levels.