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MASTER

STATUS AND APPLICATION OF THERMOLUMINESCENT DOSIMETRY SYSTEMS*

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ABSTRACT

Thermoluminescence is broadly defined as the emission of light when a solid is heated to a temperature below that of incandescence. If the amount of light emitted can be related to the absorbed dose (or absorbed dose rate) deposited by ionizing radiation, the material can serve as the basis for a dosimetry system. Since the first report by Becquerel in 1883, a variety of thermoluminescent materials have been found. Recent developments have now made practical systems possible, the most highly developed ones utilizing appropriately "activated" LiF and CaF₂. A few of the useful characteristics

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of such systems include: a very great dynamic range, from a few mr to 10^4 r and higher; usually a very small loss of information before read-out, as low as 5% per year at room temperature; ability to measure extremely high radiation rates, 10^9 r/sec. in some cases; dosimeters can be made very small while retaining fairly good sensitivity, less than 1 r being measured by a 6-mm long by 1-mm diameter device. A wide range of applications has already been made in personnel dosimetry, tissue implantations, environmental studies, and in recoverable satellites. Although there may be a few limitations, thermoluminescent dosimetry systems will undoubtedly become increasingly useful and important dosimetric tools.

DISCUSSION

For many years work has been going on to devise various methods of measuring ionizing radiations of all kinds. One fruitful course of investigation has been in the field of "solid-state" detectors, photographic film and scintillators being perhaps the most notable examples. A variety of phenomena have been used, including: temporary or permanent changes in coloration, electrical conductivity, or light

transmission; light excited by light of shorter or longer wave-length; and thermally excited luminescence. Systems based on this last phenomena will be briefly described.

Thermoluminescence is defined in a general way as the emission of light when a solid is heated to a temperature below that of incandescence. If the amount of light emitted can be related to the energy or dose deposited, the material can serve as the basis for a dosimetry system. Since the first report by Becquerel⁽¹⁾ in 1883, a variety of thermoluminescent materials has been found, among them are CaF_2 , LiF , CaSO_4 , CaCO_3 , SiO_2 , SrSO_4 , and many others.⁽²⁾ Recent developments have now made some practical systems possible.

A thermoluminescent (TL) material has appropriate impurities or activators which provide metastable electron states within the crystal lattice. When ionization takes place as the result of energy absorption from incident radiation, some of the free electrons can become trapped in these states. If the material is now heated to a sufficiently high temperature, trapped electrons are released to return to the ground state with the possible emission of light in the visible and near-visible spectrum. Figure 1 is a schematic representation of this process.

There may be several energy levels of the traps, each requiring a different temperature for a given probability of electron release. Mathematically, the probability of electron escape as a function of the energy barrier, or "trap depth", and of the temperature of the material can be written as follows: (3)

$$p = s \exp (-E/KT),$$

where p = probability of escape per second,

s = proportionality constant of the order of $10^{8 \pm 1} \text{ sec}^{-1}$,

E = energy barrier or trap depth,

T = absolute temperature,

K = Boltzman's constant.

It can easily be seen that for a given temperature the probability of escape increases as the trap depth decreases.

The most useful materials are those whose traps are of such values that the probability of escape at ordinary room temperatures is extremely small (10^{-8} to 10^{-9} sec^{-1}) but approaches the value of 1 before reaching temperatures of incandescence.

The light emitted can be measured using the arrangement shown schematically in Fig. 2. The light intensity plotted

as a function of temperature, or as a function of time as the temperature is being increased, is called a glow curve. Such a curve for a hypothetical thermoluminescent material is shown in Fig. 3. The two peaks indicate the presence of two metastable traps. As the temperature of the material rises the probability of emptying traps increases with a resultant increase in light intensity. The intensity will begin to decrease when the number of unemptied traps becomes sufficiently small. Eventually the intensity will drop to zero as all the traps are emptied, unless there is another higher energy trap that becomes apparent or incandescence is approached.

In general, the total amount of light emitted during the heating process is proportional to the number of electrons released from the traps, excluding some very high temperature effects. The number of electrons placed into traps is proportional to the energy deposited in the material by incident radiation. Hence, the total light out is proportional to the dose received, provided the traps are at high enough energies. The trap indicated by the lower peak in Fig. 3 would loose significant numbers of electrons in a fairly

short time at ordinary room temperature. The peak height can also be used as a measure of absorbed dose. In this case, heating of the material must be done in a reproducible manner, because rate of heating affects the height obtained for a given dose, faster heating rates resulting in peaks of greater amplitude with the maxima shifted to higher temperatures. (4)

All the traps of the thermoluminescent material can be emptied by heating at a high enough temperature for a sufficiently long time. After cooling the material, it is now ready to measure another exposure to radiation.

The relative response versus exposure which is typical of several thermoluminescent materials is shown in Fig. 4. The curve indicates very good sensitivity; some dosimeters can measure considerably less than 1 milliroentgen total exposure. A dynamic range of the order of eight decades is a striking feature of many thermoluminescent dosimeters (TLD). Figure 5 shows the relative response versus energy for three of the more commonly used materials. (4) The response of $\text{CaF}_2:\text{Mn}$ and of CaSO_4 are reminiscent of the energy dependence of film; both respond much like bone. The characteristics of

LiF are reasonably close to those of soft tissue and make it a particularly interesting material for use in dosimetry.

There are several commercial TLD systems now available. However, only the Edgerton, Germeshausen & Grier, Inc. (EG&G) Model 2 Thermoluminescent Dosimeter System and the Controls for Radiation (Con-Rad) TLD System will be described briefly, these being representative of the inherent capabilities of TLD systems in general.

The EG&G Model 2 Dosimeter, shown in Fig. 6, consists of $\text{CaF}_2\text{:Mn}$ coated on a coiled-wire heating element and encapsulated in an evacuated glass envelope.^(5,6) Two gold plated Kovar feed-throughs provide connection to a heating current supply. Because of the over-response of $\text{CaF}_2\text{:Mn}$ to low energies, indicated in Fig. 5, the detector is placed in a case consisting of an aluminum outer shell and a tin-lead energy compensation shield. Figure 7 shows a partially assembled detector and shield assembly and Fig. 8 indicates the energy response of the complete unit.

The Model 2 Reader is shown in Fig. 9. In the lower right hand corner the manner in which the detector is placed in a socket in the reading-head plug can be seen. The plug

is placed in the reading head with electrical connection being made between a set of spades on the plug and a set of contacts in the reading head. The read cycle is initiated by pushing the read button, after which current is passed through the heating coil, the light emitted by the thermoluminescent material is measured by a photomultiplier tube, and the resulting glow curve is drawn on the strip-chart recorder paper. The entire cycle takes approximately 20 seconds.

The peak of the curve is read directly on the scale printed on the paper. Automatic ranging is provided so that full scale can be any decade from 50 mR through 5000 R, depending on how much light is being measured during the read cycle. The resultant full scale is shown at the end of the cycle in the window where the word "empty" appears in Fig. 9. In addition, a pip appears on the right hand side of the chart each time a range change has been made. Some typical traces are shown in Fig. 10. As pointed out earlier, in order to be able to use the peak as the measure of the dose rather than the area under the curve, the material must be heated in a reproducible manner. When this is done it also means that the peak will occur at the same time after

beginning the heating. Thus, the peak of interest will always appear between the closely set lines near the scale figures shown in Fig. 9.

The main specifications of this CaF_2 system, as provided by EG&G,⁽⁶⁾ are listed in Table I.

TABLE I

1. Gamma Dose Range - 5 mR to 5000 R (present limits of reader).
2. Rate Dependence - none to at least 10^9 R/sec.
3. Energy Dependence - $\pm 10\%$ from 60 keV to at least 1.3 MeV (see Fig. 8).
4. Neutron Sensitivity in R/n/cm^2 :
 - a. Thermal - 1.41×10^{-10}
 - b. 5.4 MeV - 0.11×10^{-9}
 - c. 14 MeV - 1.47×10^{-9}
5. Reusability - in excess of 100 readings.
6. Stability - the device may be read-out at any time after exposure with negligible loss of information (less than 5% loss per year when stored in the case and the temperature does not exceed ambient).

7. Percentage Standard Deviation:

a. 100 readings of one dosimeter

1) $5 \text{ mR} - S_x = \pm 4\%$

2) $40 \text{ mR} - S_x = \pm 2\%$

3) $400 \text{ mR} - S_x = \pm 1\%$

b. 10 readings of 10 dosimeters

1) $5 \text{ mR} - S_{\bar{x}} = \pm 4\%$

2) $40 \text{ mR} - S_{\bar{x}} = \pm 2\%$

3) $400 \text{ mR} - S_{\bar{x}} = \pm 1\%$

8. Variance Among Dosimeters - $\pm 10\%$ of average at 90% confidence limit.

9. Accuracy - will depend on the details of calibration by the user. System has been checked at EG&G against a Co^{60} source calibrated by an NBS secondary standard. This results in multiple dosimeter readings within $\pm 15\%$ of reading or 2 mR, whichever is greater.

There is also available from EG&G a microdosimeter which is a sealed glass tube 1 mm in diameter and 6-mm long containing either $\text{CaF}_2:\text{Mn}$ or LiF powder. With a small adapting coil these can be read on the Model 2 Reader. The range covered is from less than 1 R to 10^5 R or greater. Two of

these microdosimeters can be seen on the black patch in the lower left side of Fig. 9.

The Con-Rad TLD system utilizes loose LiF powder,⁽⁷⁾ as shown in Fig. 11. The powder dispenser at the left side of the picture gives a standard charge of 58 mg. The plastic capsules used for individual dosimeters are approximately 5 mm in diameter and 20-mm long. To read the exposure, the powder is placed in a planchet which is then inserted in the reader. A controlled amount of electrical current is then passed through the planchet for a period of about 10 seconds. A photomultiplier tube measures the light emitted and a number proportional to the total light out is read on a digital voltmeter. The entire cycle is approximately 15 seconds long. Manual scale-ranging of two additional decades is provided, but it must be done only after completion of the cycle. Further changes in sensitivity are made by changing the voltage on the photomultiplier tube. Thus, some knowledge of the exposure is desirable.

An interesting feature of a LiF system is that its response to thermal neutrons can be changed considerably by varying the isotopic content of the Li component.⁽⁸⁾ Natural

Li has about 7.5% of Li^6 with a very high cross section for an (n,α) reaction. High-energy ionizing particles are thus generated which can deposit energy in the TLD material.

Con-Rad type N and Harshaw TLD-100 contain the natural isotopic ratio. Con-Rad Type 7 and Harshaw TLD-700 are made from 99.993% Li^7 and have negligible thermal-neutron responses compared with the others.⁽⁴⁾ Harshaw TLD-600 is available and is highly enriched in Li^6 . In addition, if a thermoluminescent material is mixed with a hydrogen-rich material, alcohol for example, there may be an enhanced fast-neutron response.

The main specifications of this LiF system, as provided by Con-Rad,⁽⁹⁾ are listed in Table II.

TABLE II

1. Range - 10 mR to 10^5 R (lower limit 100 mR if N_2 flow during reading not used).
2. Rate Dependence - less than 5% to 3×10^{11} R/hr.
3. Energy Dependence - $\pm 5\%$ 10 keV to 1.3 MeV shielded.

$\pm 40\%$ at 40 keV unshielded.

$\pm 10\%$ at 100 keV to 1.3 MeV unshielded.

4. Neutron Sensitivity - [for Harshaw TLD-100 and TLD-700 only].⁽⁴⁾
 - a. Thermal
 - 1) TLD-100 2.7×10^7 n/cm² equals 1 R gamma-ray exposure.
 - 2) TLD-700 - less than 0.5% of response of TLD-100.
 - b. Fast - TLD-100 & TLD-700, 10^8 n/cm² less than 1 R gamma-ray exposure.
5. Reusability - indefinite for accumulated exposures less than 10^5 R.⁽⁴⁾
6. Stability - less than 5% per year at 20°C.
7. Percentage Standard Deviation
 - a. 10 mR - $\pm 20\%$
 - b. 100 mR - $\pm 5\%$
 - c. 1 R - $\pm 2\%$
8. Variation of weight of TL material dispensed - approximately 1%.
9. Accuracy - will depend on the details of calibration of user. Typical value would be $\pm 5\%$ of precision number below 1R.

Two devices besides standard capsules are available from Con-Rad. One is the Ultra-MicroDosimeter, a 1.5-mm diameter by 10-mm long sealed glass tube containing LiF with

a usable range of 0.1 R to 10^5 R. The other device is the Health Physics-TLD Dosimeter, which is essentially a Con-Rad planchet with LiF powder held in the cup by a permanently attached fine-mesh screen. Its usable range is 0.01 R to 10^5 R. Both dosimeters are reusable.

Some drawbacks exist in the use of thermoluminescent materials and various precautions have to be taken if proper measurements are to be made. For example, when $\text{CaF}_2:\text{Mn}$ is used in air, there is a spurious thermoluminescence equivalent to about 1 R of Co^{60} gamma-radiation, which prevents the measuring of small doses.⁽¹⁰⁾ Enclosing the material in a vacuum or inert gas eliminates the difficulty, but often the wall of the containing vessel disturbs measurements of particle radiation such as electrons and beta-rays. Also, $\text{CaF}_2:\text{Mn}$ is sensitive to ultraviolet light⁽⁶⁾ and should not be exposed to room light for more than 5 or 10 minutes between dedosing by heating and the reading of a subsequent exposure. In the case of LiF when used in loose powder form, a spurious thermoluminescence equivalent to about 1 R of Co^{60} occurs when the powder is read while in an air atmosphere.⁽⁸⁾ It has been found⁽⁹⁾ that nitrogen gas is a specific quencher

of this spurious effect in LiF, so that if the reader enclosure is filled with dry N₂, an exposure as low as 10 mR can be measured. LiF undergoes permanent crystal damage for total accumulated doses greater than about 3×10^4 R, the degree of which depends on the total dose and on the number of exposure and read cycles which have taken place to obtain that total dose.⁽¹¹⁾ Thus, in some instances a record would have to be kept of all exposures of all the powder which is to be reused.

Thermoluminescent dosimeters have already been widely used. The physical characteristics and the cost of such systems compare favorably with film systems of personnel dosimetry. It is reasonable to assume that with further development, considerably better overall dosimetry systems will emerge than is possible with film. Some of the applications other than personnel dosimetry are: environmental studies, including background levels; implantation under the skin of animals left in their natural environment;⁽⁶⁾ and attaching to large insects; deep animal implantation⁽¹²⁾ to measure doses from either external radiation or internal radiation emitters; clinical and radiobiological dosimetry;^(8,13)

and space dosimetry by means of recoverable satellites. (14)

Many characteristics of thermoluminescent materials have yet to be subjected to detailed scrutiny, but their usefulness has already been shown. With further improvements and perhaps new materials, thermoluminescent systems will undoubtedly become increasingly important dosimetric tools.

REFERENCES

1. Becquerel, H., Ann. Chem. Phys., 30, 1-68 (1883).
2. Angino, E. E. and Grogler, N., "Thermoluminescence - Bibliography," TID-3911 (Rev. 1), (1963).
3. Randall, J. T. and Wilkins, M. H. F., "Phosphorescence and Electron Traps." Proc. Roy. Soc. (London), A184:366 (1945).
4. Attix, F. H., "Present Status of Dosimetry by Radiophotoluminescence and Thermoluminescence Methods," NRL Report 6145, (September 1964).
5. Blase, E. F., Lagerquist, R. E., Palmer, R. C., and Rutland, D. F., "A Thermoluminescent Personnel Dosimetry System," Health Physics, 9, 888 (1963).
6. Palmer, R. C., Personal communication.
7. McCall, R. C., Babcock, L. E., and Fix, R. C., "A LiF Thermoluminescent Dosimetry System for Health Physics Applications," Health Physics, 9, 889, (1963).

8. Cameron, J. R., Zimmerman, D., Kenney, G., Buch, R., Bland, R., and Grant, R., "Thermoluminescent Radiation Dosimetry Utilizing LiF," Health Physics, 10, 25-9 (1964).
9. McCall, R. C. and Fix, R. C., Personal communication.
10. Schulman, J. H., Attix, F. H., West, E. J., and Ginther, R. J., "New Thermoluminescent Dosimeter," Review Sci. Instr., 31, 1263-69 (1960).
11. Marrone, M. J. and Attix, F. H., "Damage Effects in $\text{CaF}_2\text{:Mn}$ and LiF Thermoluminescent Dosimeters," Health Physics, 10, 431-36 (1964).
12. Hensley, J. C., Private communication.
13. Malsky, S. J., Amato, C. G., and Roswit, B., "Thermoluminescent Dosimetry," CONF-164-5, from Society of Nuclear Medicine 10th Annual Meeting, Montreal, 1963b.
14. Attix, F. H. and West, E. J., "Thermoluminescent Dosimeters on Recoverable Satellites," NRL Report 5938, (May 1963).

Fig. 1 - Schematic representation of the thermoluminescent process.

Fig. 2 - Schematic diagram of a thermoluminescent dosimeter reader.

Fig. 3 - Glow curve of hypothetical thermoluminescent material.

Fig. 4 - Response vs. exposure typical of several thermoluminescent materials.

Fig. 5 - Dependence of thermoluminescence response per roentgen upon γ -ray quantum energy [after Attix (4)]. All data are normalized to 1.00 at 1.25 MeV (Co^{60} γ -rays). Curves are calculated on the basis of $(\tau + \mu_{\text{en}})/(\mu_{\text{en}})_{\text{air}}$, in cm^2/g , under the assumption of constant response per rad in the phosphor. Circled points were measured with miniature $\text{CaF}_2:\text{Mn}$ dosimeters irradiated by heavily filtered x-rays. Crosses represent the data of Cameron et al. (8), comparing bare LiF (TLD-100) powder with an NBS-calibrated Victoreen 25-R chamber.

Fig. 6 - EG&G thermoluminescent detector.

Fig. 7 - EG&G thermoluminescent detector and energy compensation shield.

Fig. 8 - EG&G thermoluminescent dosimeter relative energy response in shield.

Fig. 9 - EG&G Thermoluminescent Dosimeter Reader, Model 2.

Fig. 10 - EG&G typical chart records.

A. 2.3R -- TWO SCALE CHANGES
(Pips on record in upper margin)

B. 33 MR -- BLACK BODY RADIATION
AFTER PEAK

C. ZERO -- BLACK BODY RADIATION
AFTER DOUBLE LINE

Fig. 11 - Con-Rad TLD System.

33