Thermoluminescent Neutron Dosimetry

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A literature review and the present status of thermoluminescent neutron dosimetry are presented. The theory on thermoluminescent dosimetry is given in detail. The applications of thermoluminescence phenomenon in thermal neutron dosimetry, in fast neutron dosimetry, and in mixed field dosimetry are discussed. Some experimental results are included. The prospective use of thermoluminescent dosimeters in other nuclear detection or measurement is also given.

INTRODUCTION

It is now agreed, in general, that thermoluminescence is caused by the thermally activated release of electrons trapped at irregular points in the crystal.⁽¹⁾ The process of raising a material temperature above ambient to cause a detectable emission of light below the incandescent temperature is called thermoluminescence. This phenomenon has long been known; nevertheless, the application of thermoluminescence principle to dosimetry is still a new field to investigate.

Lithium fluoride (LiF) was one of the first thermoluminescent materials investigated for possible application to dosimetry by Daniels,⁽²⁾ but was abandoned after being found unsatisfactory in a number of respects. There seemed to be no appreciable interest in thermoluminescence dosimetry until 1960. Then Cameron⁽³⁾ saw the possibilities in thermoluminescence dosimetry and has carried on in his laboratory a program with improved apparatus and basic research. His success in demonstrating the possibilities of using LiF in gamma dosimetry has led to widespread interest in many laboratories. Although much information has been published on gamma dosimetry, there is very little in the literature concerning thermoluminescent neutron dosi-

metry.

This review, therefore, is intended to provide the up-to-date literature, and to give a deep insight into the development of thermoluminescent neutron dosimetry.

GENERAL REVIEW

1. *Historical background.* The early history of thermoluminescence can be traced back to the year 1663 when the Irish chemist and physicist, Boyle,⁽⁴⁾ investigating the properties of a diamond, noticed the thermoluminescence phenomenon for the first time. In the middle of the 19th century after the discovery of radioactivity, French physicist Becquerel⁽⁵⁾ laid down the foundation for a real treatment of thermoluminescence in radium-irradiated fluoride. During the first 30 years of this century, many papers were published on several properties of thermoluminescence. The book by Prizibram⁽⁴⁾ and by Pringsheim⁽⁶⁾ are recognized as the best sources for studying the historical development of thermoluminescence.

2. *Literature review on theory.* The German physicist Urbach⁽⁷⁾ was a pioneer in developing the theoretical interpretation of the thermoluminescent phenomenon. Randall and Wilkins,⁽⁸⁾ Hill and Schwed,⁽⁹⁾ Bonfiglioli et al.,⁽¹⁰⁾

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Halperin et al.,⁽¹¹⁾ May and Partridge,⁽¹²⁾ have improved Urbach's theory and experimental technique. Seitz' model in interpreting luminescence can also be applied to the case of thermoluminescence. A bibliography on thermoluminescence has been compiled by Angino et al.⁽¹⁴⁾

3. *Literature review on dosimetry.* In addition to LiF, other materials such as CaSO_4 -Mn and CaF_2 have been used as thermoluminescent dosimeters.^{(15),(16)} Scientists in Germany, USSR, Belgium, Japan as well as the USA have been studying the prospective thermoluminescent dosimeters in the last 15 years. A bibliography on thermoluminescence dosimetry has been compiled by Spurny.⁽¹⁷⁾

THEORY

Thermoluminescence, in fact, is a special case of phosphorescence. Seitz' luminescence model⁽¹³⁾ can interpret the mechanism of this phenomenon fairly satisfactorily. To apply Seitz' model, one should change the time variable t to a temperature variable T by using the following relation

$$dT = \beta dt \dots\dots\dots (1)$$

where β is the linear heating rate.

The escape rate of the electron from the trap can be expressed in two different ways mathematically:

$$dn/dt = -n \exp(-E/KT) \dots\dots\dots (2)$$

and

$$dn_i/dt = -p_i n_i N \exp(-E/KT) \dots\dots (3)$$

where

s = frequency factor describing the frequency of electron movement in traps, which is less than the vibrational frequency of the crystal lattice

E = energy barrier or trap depth

K = Boltzmann's constant

T = absolute temperature

n = number of trapped electrons

n_i = number of L_i -centers per unit volume still empty at time t

p_i = the probability factor for the transition to the L_i -centers

N = number of electrons per unit volume at time t lying on the F-centers.

Integrating Equations (2) and (3),

one obtains

$$n = n_0 \exp \left[- \int \frac{s}{\beta} \exp(-E/KT) dT \right] \dots\dots\dots (4)$$

$$n_i = n_{i0} \exp$$

$$\left[- \int \frac{p_i}{\beta} N \exp(-E/KT) dT \right] \dots\dots (5)$$

Since

$$I = dn/dt \dots\dots\dots (6)$$

then

$$I = -n_0 s \exp(-E/KT) \exp$$

$$\left[- \int \frac{s}{\beta} \exp(-E/KT) dT \right] \dots\dots (7)$$

$$I_i = -n_{i0} p_i N \exp(-E/KT) \exp$$

$$\left[- \int \frac{p_i}{\beta} N \exp(-E/KT) dT \right] \dots\dots (8)$$

where

n_{i0} = initial concentration of L_i -centers

n_0 = initial number of trapped electrons

I = light intensity

I_i = light intensity of the i th glow curve peak.

The glow curve is the plot of light intensity versus temperature or time. **Figure 1** shows the glow curves obtained from two different LiF specimens exposed to neutron radiation.

Figure 1. The Glow Curves of type 7 LiF (top) and type N LiF (bottom) exposed to neutron radiation

The difference between Equations (2) and (3) is due to the different interpretations on the kinetic order of thermoluminescence; however, both equations are based on the assumption that electrons in traps have a Maxwellian distribution of thermal energy. Equation (2) is called first-order kinetics proposed by Randall and Wilkins,⁽⁸⁾ while Equation (3) is called second-order kinetics proposed by Bonfiglioli et al.¹⁰ These two equations are the fundamental equations in dealing with

thermoluminescence kinetics. Equations (7) and (8), on the other hand, are the fundamental equations in dealing with thermoluminescence dosimetry. The principal factor in Equation (7) is n_0 and that in Equation (8) is n_{i0} . n_0 is the number of ionized activators in the thermoluminescent dosimeter, and is proportional to the absorbed dose. According to the second-order kinetics, n_{i0} is the concentration of L_i -centers at the beginning of the i th glow curve peak, and is proportional to the absorbed dose also. The value of n_0 is given by the area included between a glow curve and the thermal background curve, while the value of n_{i0} is given by the area of the i th peak excluding the thermal background. The total light emitted, over part or all of the glow curve area, or the height of one or more of the glow curve peaks, may serve as a measure of the absorbed dose in the thermoluminescence dosimeter. A detailed mathematical derivation of Equations (1) to (8) has been given by Weng.⁽¹⁸⁾

In addition to the mathematical expressions mentioned above, some schematic presentations in interpreting thermoluminescence theory and dosimetry can be seen in the work of Seitz,⁽¹³⁾ Augenstine et al.,⁽¹⁹⁾ and Attix.⁽²⁰⁾

DOSIMETER READER

The principle of the thermoluminescent dosimeter reader is as follows:

The light is emitted when an irradiated thermoluminescent dosimeter is heated in proximity to photomultiplier tube. The resulting signal from the photomultiplier tube is integrated by suitable electronic circuitry, and then measured and displayed on a digital voltmeter. If the glow curves are needed for examining, one can add a sensitive electrometer and a recording chart. The resulting

signal from the photomultiplier tube is transmitted to the electrometer and is recorded on the chart as the glow curve with the time scale as the abscissa. If the temperature scale instead of time scale is needed, the thermocouple system should be used.

In the United States, commercial readers are available from the following companies:

1. Controls for Radiations, Inc., Cambridge, Massachusetts
2. Madison Research and Development Laboratories Inc., Middletown, Wisconsin
3. Edgerton, Germeshausen & Grier, Inc., Goleta, California
4. The Harshaw Chemical Company, Cleveland, Ohio

These readers vary both in prices and functions. As a whole, the price is fairly expensive as compared to other types of nuclear instruments.

The calibration of the reader is normally carried out against gamma radiation, say, cobalt-60 or cesium-137 source. The readings, therefore, are in terms of gamma doses. The reason of using gamma source for calibration lies in the fact that thermoluminescent dosimeter is gamma energy independent, and gamma sources are available in most laboratories. Furthermore, thermoluminescent dosimeter is very sensitive to gamma radiation. In order to convert the gamma equivalent dose to neutron dose, another calibration should be taken. This additional calibration is very complicated since thermoluminescent dosimeter is neutron energy dependent.^{(18),(21),(22)} In short, one should measure the neutron flux first. There is a certain relation between neutron flux and neutron dose as long as the neutron energy is known. The reader is then calibrated against the neutron source with known flux and energy; in other words, the

reader is calibrated against the neutron source with known quantity of dose. The relation between gamma equivalent dose and neutron dose can thus be established. The detail of reader calibration against neutron and gamma sources has been prepared at Texas A&M University.^{(18),(21)} Karzmark et al.⁽²³⁾ have published a paper on thermoluminescent reader in detail.

THERMAL NEUTRON DOSIMETRY

Among the three thermoluminescent dosimeters; namely, LiF, CaSO₄-Mn, and CaF₂, LiF has been proved the most successful neutron dosimeter. The thermal neutron cross section of Li is 71 barns, and that of Li⁶ is 950 barns.⁽²⁵⁾

Commercial LiF dosimeters are available from the Harshaw Chemical Company and Controls for Radiation, Inc. The main products of the Harshaw Chemical Company are TLD-100 LiF with natural abundance of Li, TLD-600 LiF with 95.62% enrichment in Li⁶, and TLD-700 LiF with 99.91% enrichment in Li⁷. For Controls for Radiations, Inc., the main products are type N LiF and type 7 LiF; the former is similar to TLD-100, and the latter is similar to TLD-700.

The most important interaction of thermal neutrons with LiF is capture through the reaction $\text{Li}^6 (n, \alpha) \text{H}^3$ with the combined energy of 4.8 Mev of its products, the ionization is dominated by the alpha and H³.

The thermoluminescent responses of Con-Rad type N LiF have been measured.⁽¹⁸⁾ Several LiF dosimeters were irradiated along the central length of the thermal column in a 100-Kw swimming pool reactor at Texas A & M. The gamma background was reduced to the minimum amount by inserting a 6-inch lead shield between the core face and the end

of thermal column. The result of this experiment is shown in **Figure 2**.⁽¹⁸⁾ From Figure

2 one can see the response of LiF to thermal neutrons is similar to that of gammas.

Figure 2. Light Output versus Thermal Neutron Fluence

In practical use, one pair of two different LiF dosimeters must be used in thermal neutron dosimetry. One of them is LiF with natural abundance in Li or with high enrichment in Li^6 ; the other is enriched in Li^7 . For instance, a pair of type N and type 7, a pair of TLD-100 and TLD-700, or a pair of TLD-600 and TLD-700, will accomplish the work. This is due to the fact that the thermal neutron cross sections of type N and type 7 (TLD-100 and TLD-700, TLD-600 and TLD-700) are considerably different. Therefore the difference in readings from the paired dosimeters will provide the gamma and neutron dose respectively. According to Simpson's work,⁽²⁶⁾ the paired dosimeter technique essentially determines the gamma dose. Type N or TLD-100 LiF is merely an auxiliary thermal neutron dosimeter when thermal neutron dose is desired, since type 7 or TLD-700 has

little response to thermal neutron. Nevertheless, the paired dosimeters are still considered a very useful technique to determine thermal neutron and gamma doses.

Cautions must be taken that thermal neutrons are strongly absorbed in outer layer of Li^6 so that the thick LiF samples are not uniformly irradiated throughout.⁽²¹⁾ The half value layer of LiF for thermal neutrons is 0.338cm assuming the average density of LiF power is 1.25gm/cm^3 .

FAST NEUTRON DOSIMETRY

The application of thermoluminescence phenomenon to fast neutron dosimetry is not as successful as thermal neutron dosimetry. This is mainly due to the small activation cross section of LiF at higher neutron energies.

Alcohol may enhance the sensitivity of LiF

response to fast neutrons; however, it may reduce the sensitivity at low dose. This is due to the absorption of the gamma-rays by alcohol. Fast neutrons are always accompanied by gamma-rays, partly because neutron sources also emit gamma-rays and partly because neutron interactions with matter produce gamma-rays.

In fast neutron dosimetry, ionization is essentially due to the recoil protons from the alcohol surrounding the LiF powder; in fact, the LiF phosphor becomes a proton source. The absorption factor may be expressed as the ratio of LiF response with the proton source material to the response without the proton source material. A detailed study of the absorption factor has been carried out by Endres.⁽²⁷⁾

Several organic compounds have been investigated to increase the fast neutron response of LiF; among those are: ethyl alcohol ($\text{CH}_3\text{CH}_2\text{OH}$), methanol (CH_3OH), carbon tetrachloride (CCl_4), and heptane or iso-octane ($((\text{CH}_3)_2\text{CH}(\text{CH}_2)_4\text{CH}_3)$) which has 5% more hydrogen atoms per unit volume than ethyl alcohol. No significant improvement in results have been noted.

LiF is neutron energy dependent. Investigations have been made at the following neutron energies:

0.26, 0.56, 0.84, 1.27, 1.96, 2.67, 2.80, 4.50, 5.30, and 14.10 Mev by Wingate et al.⁽²¹⁾
2.00, 2.80, 4.20, 5.74, 6.20, and 14.00 Mev by Weng⁽¹⁸⁾

All of them have reached the same conclusion.

In the energy range from thermal to 1.5 Mev, Kastner et al.⁽²²⁾ have made a careful study. They found a marked increase in light output about 0.25 Mev. This is attributed to the $\text{Li}^6 (n, \alpha) \text{H}^3$ reaction. In this region,

therefore, LiF dosimeter must be carried out very carefully so there will be no distortion due to the presence of moderated neutrons in the region of the 0.25 Mev resonance.

MIXED FIELD DOSIMETRY

The prominent aspect of thermoluminescent dosimetry is its capability to separate the radiation from a mixed field where fast neutrons, thermal neutrons, and gamma-rays all exist. For a practical measurement, a set of 3 dosimeters; for example, type N (TLD-100), type 7 (TLD-700), type 7 plus alcohol (TLD-700 plus alcohol), will provide the result. Type 7 is a measure of gamma dose only based on the fact that its activation cross section for neutrons is very small and thus the response due to neutrons can be neglected. The dosimeter with alcohol has a response to fast neutrons and gammas, thus the fast neutron dose can be determined from the difference in response between these two dosimeters. As previously mentioned, the paired dosimeters will determine the thermal neutron dose.

Based on this assumption that type 7 LiF has no direct response to neutrons, the mixed field in the reactor core at Texas A&M has been measured. The result was not as expected due to the absorption of gamma-rays by alcohol. The fast neutron dose can only be determined when the absorption factor of alcohol has been well measured in advance.

OTHER PROSPECTIVE USES

1. *Gamma heating measurement in a critical assembly.* The principle lies in the fact that gamma dose and gamma heating have some similarity in nature, in that both of them are related to the energy absorbed in the medium concerned. The possibility of discriminating

the various types of radiation in a mixed field by using thermoluminescent neutron dosimeters makes this prospective technique in measuring gamma heating more promising than ever, since the presence of neutrons has no appreciable perturbation. The new development would be much easier to use than the direct calorimetric measurement of temperature changes which required painstaking care and introduces many possible systematic errors. Johnson and Stanford⁽²⁸⁾ have published a paper on this particular topic.

2. *Neutron image storage.* Photographic methods have long been used for detecting thermal neutron images; however, they lack the ability to integrate neutron image information in the presence of interfering gamma radiation for long periods of time. The ability to discriminate the neutron radiation from gamma radiation makes thermoluminescent neutron dosimeter a prospective neutron image detector. The advantages of thermoluminescent image detectors are the ability to accumulate thermal neutron image information for extended period without saturation or reciprocity-law-failure response problems, and equal or better relative neutron-gamma responses than other available direct detection methods. Kastner et al.⁽²⁹⁾ have investigated this work at Argonne National Laboratory.

CONCLUSIONS

The thermoluminescent neutron dosimeter is a very promising tool in dosimetry work. Lithium fluoride phosphor is the dosimeter

of choice because of its large thermal neutron cross section, long dose storage, inertness to environment, and tissue equivalence. Even in the low dose level, LiF dosimeter is preferred.⁽³⁰⁾ However, some obstacles must be overcome :

1. LiF is extremely sensitive to minute impurity concentrations and thus it is difficult to produce LiF consistently with the same properties.

2. The reading device is relatively complicated and expensive.

3. LiF undergoes permanent crystal damage for accumulating exposures, the degree of which depends on the total dose, on the number of exposure, and on the read cycles which have taken place to obtain that total dose.⁽³¹⁾

The practical use of neutron radiation in industry, biology, medicine, and in research has been increasing so rapidly that the need of neutron dosimetry is increasing immensely. The improvement of present thermoluminescent neutron dosimeter system will meet this urgent need in the near future.

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