PRACTICAL CONSIDERATIONS FOR TLD-400/700-BASED GAMMA RAY DOSIMETRY FOR BNCT APPLICATIONS IN A HIGH THERMAL NEUTRON FLUENCE

Steven W. Martsolf,* Jennifer E. Johnson,* Christopher E. D. Vostmyer,* Barry D. Albertson,† and Stephen E. Binney*

Abstract—**Operating experience with thermoluminescent dosimeters used in a boron neutron capture therapy research project is reported. In particular, certain facets of the use of thermoluminescent dosimeters for gamma ray dose measurements in the presence of a high thermal neutron fluence are discussed, including a comparison of TLD-400 and TLD-700 for gamma ray dosimetry, annealing procedures, and the effects of neutrons (^{56}Mn activation) on TLD-400. The TLD-400 were observed to have a thermal neutron sensitivity (due to ^{56}Mn beta decay) of 1.5×10^{-13} Gy per n cm^{-2}. An algorithm was developed to correct for the ^{56}Mn beta decay thermal neutron-induced effects on TLD-400 by using a two-stage thermoluminescent readout for the thermoluminescent dosimeter chips.**

Health Phys. 69(6):966–970; 1995

Key words: thermoluminescence dosimetry; dosimetry, gamma; radiation, low level; neutron detection

INTRODUCTION

THERMOLUMINESCENT DOSIMETERS (TLD) have become a common dosimetric tool for a variety of low-level radiation measurements. Often the application involved is environmental dose measurement or personnel dose assessment. However, when TLDs are employed to measure gamma ray doses in a radiation field that has a high thermal neutron dose component, corrections must be made for the neutron-induced dose. ^{7}LiF (TLD-700)‡ and CaF_2 (TLD-400)‡ TLD have been used to monitor the gamma ray fields associated with the boron neutron capture therapy (BNCT) research program at Oregon State University (OSU). This program involved *in vitro* thermal neutron irradiation of ^{10}B-loaded rat pituitary and other types of endocrine-related tumor cells in the thermal column of the Oregon State TRIGA Reactor (OSTR). Details of the TLD dosimetry for this project are discussed below. Although this discussion is directed toward BNCT studies, it is also applicable to other gamma ray dose measurements in a high thermal neutron fluence.

In the spring of 1991, OSU and Oregon Health Sciences University (OHSU) researchers began a collaborative investigation to determine the efficacy of BNCT directed against pituitary and other endocrine-related tumors. The scientific basis for BNCT has been well documented (Slatkin 1991; Barth et al. 1992). The OSU/OHSU research was aimed at investigating a unique method of cell targeting via endocrine cell receptor-mediated ^{10}B delivery mechanisms (Albertson et al. 1993).

Early decisions were made regarding the types of dosimetry to be considered for characterizing the thermal column neutron and gamma radiation fields. As part of this decision making process, various discussions were held with individuals at U.S. DOE-Idaho, EG&G-Idaho, Naval Research Laboratory, U.S. DOE Environmental Measurements Laboratory, and Harshaw/Bicron. Irradiation samples were sufficiently small so as to preclude the consideration of other types of dosimetry besides small TLD chips. In addition, OSU Radiation Center personnel have had a successful history with and currently employ LiF (TLD-100)‡ TLD for their environmental background gamma ray measurement program.

Based upon the plethora of published LiF TLD experience, their known and generally favorable characteristics, and the local active and successful TLD-100 program, it was decided to first test the efficacy of the TLD-700 for measuring the direct and indirect gamma ray contributions of the relatively high-level mixed radiation field present during the BNCT experiments. Thin gold (bare and cadmium-covered) foils were used to measure the thermal and epithermal neutron contributions, while ^{10}B-shielded indium foils were used to measure the fast neutron component. Ultimately, as described below, it was decided to employ only TLD-400 chips for gamma ray dose measurements in these experiments.

* Department of Nuclear Engineering, Oregon State University, Corvallis, OR 97331-5902; † Department of Medicine, Division of Endocrinology, Diabetes, and Clinical Nutrition, Oregon Health Sciences University, 3181 S.W. Sam Jackson Park Road, Portland, OR 97201-3098.

‡ TLD100: LiF:Mg, Ti (92.5% ^{7}Li); TLD-400: CaF_2:Mn; TLD-700: LiF:Mg, Ti (99.99% ^{7}Li); 3.17 mm × 3.17 mm × 0.9 mm, Harshaw/Bicron, 6801 Cochran Road, Solon, OH 44139.

(*Manuscript received* 26 *October* 1994; *revised manuscript received* 5 *May* 1995, *accepted* 4 *August* 1995)

0017-9078/95/$3.00/0

MATERIALS AND METHODS

The OSTR is a 1.1 MW TRIGA Mark II[§] research reactor. The reactor is light water cooled and moderated and has FLIP fuel (70% enriched ^{235}U, 8.5 wt% uranium, mixed with $ZrH_{1.6}$). For these studies it was operated at steady state powers of 100 kW to 1 MW.

The irradiation facility used for the BNCT work was the thermal column. The standard configuration for the thermal column is a stack of graphite about 120 cm by 120 cm in cross section and 43 cm (non-removable) plus 127 cm (removable) long. The BNCT irradiation facility (Fig. 1) involves the replacement of three adjacent 10 cm × 10 cm × 127 cm graphite stringers with a bismuth shield, a bismuth sample holder, and a graphite plug to reduce the gamma ray dose rate while maintaining a sufficiently high thermal neutron flux (Albertson et al. 1993). The bismuth shielding consists of 21.7 cm in the inner direction (toward the reactor core), 11.6 cm in the outer direction, a minimum of 6 cm on the sides, 2.5 cm on the top, and 3.1 cm on the bottom. The bismuth shielding is followed by a 40 cm long graphite plug. In this configuration, at the sample location (23 cm from the inner end of the removable stringers), the thermal neutron flux at 1 MW is 5.2×10^{10} n cm^{-2} s^{-1}, the epithermal neutron flux is 9.2×10^{8} n cm^{-2} s^{-1} Δu^{-1}, the fast neutron flux is 1.5×10^{8} n cm^{-2} s^{-1}, and the gamma ray dose rate is 0.011 Gy s^{-1}.

The major equipment used to carry out the initial BNCT TLD gamma dosimetry program included the TLD-400 and TLD-700 chips, a TLD reader,[‖] an annealing oven and tray, and a 2.4 m Bq (65 Ci) ^{60}Co irradiator.[¶]

A defined TLD protocol was established to ensure effective, controlled, and repeatable employment of the TLD. Briefly, this protocol includes storage and handling, inspection (including cleaning, if necessary), annealing, sensitivity batch checking, development of calibration curves, packaging, irradiation, and TLD readout. Key elements of this protocol for the TLD-400 (ultimately the TLD of choice for these BNCT studies) include the following:

- Pre-irradiation oven-anneal in an anodized aluminum annealing tray at 400 °C for 60 min followed by a 45 s quenching of the tray and TLD to room temperature.
- Initial and periodic sensitivity batch checks at 2 Gy with a ^{60}Co source (TLD outside ±5% of the mean are discarded).
- All measurements (batch checks, calibrations, and reactor irradiations) made with triplicate samples.

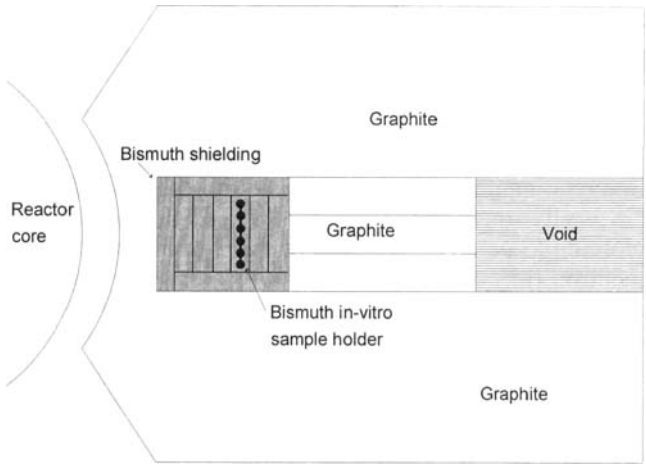

Fig. 1. Oregon State University Triga reactor thermal column with boron neutron capture therapy experimental facility in place.

- Triplicate samples packaged and sealed in thin, clear, low-density plastic envelopes facilitating individual TLD serialization and history tracking.
- TLD calibration curve with ^{60}Co source (0 to 5 Gy).
- TLD glow curve integration from 100 °C to 350 °C with a 7.06 °C s^{-1} linear temperature ramp and 50.2 s total readout.
- Development of new calibration curves when indicated by periodic batch check results.

Calibration of specific TLD batches is performed by exposure to ^{60}Co gamma rays. Triplicate TLD samples are irradiated to 1, 2, 3, 4, and 5 Gy. Non-irradiated TLD triplicate samples are used as a background measurement. Subsequent to TLD readout, a quadratic (nearly linear) calibration curve is developed relating absorbed dose (Gy) to thermoluminescence (TL in μC).

Also, periodic sensitivity checks for given TLD batches are conducted. Typically nine or more randomly selected TLDs from a given batch are irradiated at 2 Gy. If their average TL response differs by more than 5% from the previous calibration curve, then the entire batch is sensitivity checked at 2 Gy, TLDs outside ±5% of the average are discarded, and a new calibration curve is generated.

RESULTS AND DISCUSSION

The literature is replete with information regarding TLD employment in low-level single and mixed radiation fields and, conversely, relatively less with specific information about the use of TLD for gamma ray dose measurements in high thermal neutron fluences [see, for example, Horowitz et al. (1977) and Horowitz (1984)]. This work confirms Horowitz's CaF_2:Mn work, but goes beyond by introducing the need and technique for correcting for the induced TL from the ^{56}Mn beta decay up to the precise time of TLD readout. Note that the thermal neutron fluences reported here are about two orders of magnitude greater than in Horowitz's experiments. Con-

[§] General Atomics, P. O. Box 85608, San Diego, CA 92186-9784.
[‖] Harshaw Model 2000-A Thermoluminescent Detector, Model 2000-B Automatic Integrating Picoammeter, Harshaw Chemical Co. (now Harshaw/Bicron, 6801 Cochran Road, Solon, OH 44139).
[¶] Model R60124, U.S. Nuclear Systems Corp., P. O. Box 208, Burbank, CA.

sequently, the following information may be pertinent to a TLD program for measurements of this type.

Selection of TLD-400 vs. TLD-700

Both TLD-700 and TLD-400 chips were evaluated to compare their efficacies in measuring the direct and indirect gamma ray components in the mixed field. Typically these TLDs were exposed in the thermal column to gamma ray doses in the 1 to 3 Gy range and neutron fluences of the order of 10^{12} to 10^{13} n cm^{-2}. It was quickly recognized that TLD-700 chips yielded TL two to three times that expected for the predicted gamma ray doses. It was suspected that the additional TL was due to ^{6}Li(n,α) reactions in the small quantities of ^{6}Li contaminant within the TLD-700 chip when exposed to the relatively large thermal neutron fluences. Although the early reported ^{6}Li content of TLD-700 chips was about 0.01% (Cameron et al. 1968), it was determined from the manufacturer‡ that the TLD-700 chips used at OSU came from a batch that was ~0.04% ^{6}Li, which is typical of TLD-700 chips currently available for purchase (Horowitz 1984). A series of comparative experiments irradiating TLD-700 and TLD-400 chips, both bare and encapsulated in enriched $^6Li_2CO_3$ (92.39 wt% ^{6}Li), was performed. The responses from the ^{6}Li-shielded TLD-400 and TLD-700 chips were within 3% of each other. These results confirmed the significant over-response of the unshielded TLD-700 chips to thermal neutrons. Thus, the TLD-400 was selected as the gamma ray dosimeter of choice for the BNCT irradiations.

Annealing procedures

The initial annealing procedure for both TLD-400 and TLD-700 chips consisted of a 1-h pre-irradiation anneal at 400°C, followed by 100°C for 2 h and a post-irradiation/pre-read anneal at 100°C for 10 min. This protocol was followed exactly until further investigation could determine its necessity. After several experiments, it became evident that these recommended annealing procedures were not necessary for gamma ray doses above 1 Gy. Cursory experiments confirmed that for high-level gamma ray doses (i.e., 1 to 3 Gy), any pre-annealing beyond the anneal imposed by the TLD readout instrument was unnecessary. However, to ensure reproducibility and uniformity and to mitigate the potential of using TLDs which were either previously exposed and not read out or were unintentionally exposed to unknown significant TL-inducing sources, a modified oven pre-irradiation annealing protocol (400°C for 1 h) was invoked for all TLD-400 and TLD-700 chips.

A series of pre-read anneal experiments demonstrated no measurable difference for the TLD-700 at high dose (2 Gy ^{60}Co) with or without pre-read anneal and confirmed the published and "known" lack of pre-read anneal requirement for the CaF_2:Mn phosphor. With respect to the TLD-700, after post-irradiation pre-read anneal delays of up to 14 d, the additional low temperature peak TL (few nC) could not be distinguished from the total TL (tens of μC).

Effects of neutrons on TLD-400 chips

Perhaps the most significant practical consideration encountered has been the effect of the thermal neutron activation of manganese when the TLD-400 chips are subjected to thermal neutron fluences on the order of 10^{12} to 10^{13} n cm^{-2}. At these thermal neutron fluences, the ^{56}Mn activity in these TLD produces an associated prompt gamma ray and subsequent beta dose in the TLD chip during and after reactor irradiation (Horowitz et al. 1977). This TL is an additional TL in excess of that produced by the reactor gamma rays during the reactor irradiation. This additional TL would otherwise be attributed to reactor gamma rays if corrections were not made for the thermal neutron contribution. The neutron-induced ^{56}Mn prompt gamma ray contribution is not accounted for in this work.

Horowitz et al. (1977) state that "the thermal neutron induced dose *is not necessarily negligible* compared to the gamma induced dose in these [TLD-400] dosimeters in mixed n-γ radiation fields" (italics added). A series of TLD-400 exposures, conducted in the thermal column at various reactor thermal neutron fluences, indicated that Horowitz's comment can be an understatement in some cases. The TL from these TLDs was observed to increase as a function of the delay time after irradiation before TLD readout (see Fig. 2) and can be characterized quite accurately by a $(1-e^{-\lambda t_1})$ response, where λ is the decay constant for ^{56}Mn and t_1 is the readout time after the end of irradiation. Table 1 indicates the degree of fit to a curve of the form

$$TL = TL_\gamma + TL_n(1 - e^{-\lambda t_1}), \quad (1)$$

where TL_γ is the gamma ray-induced response (produced directly by reactor gamma radiation during irradiation) and TL_n is the equilibrium neutron-induced response from ^{56}Mn beta decay [representative of the additional TL accumulated during irradiation and an "infinitely"

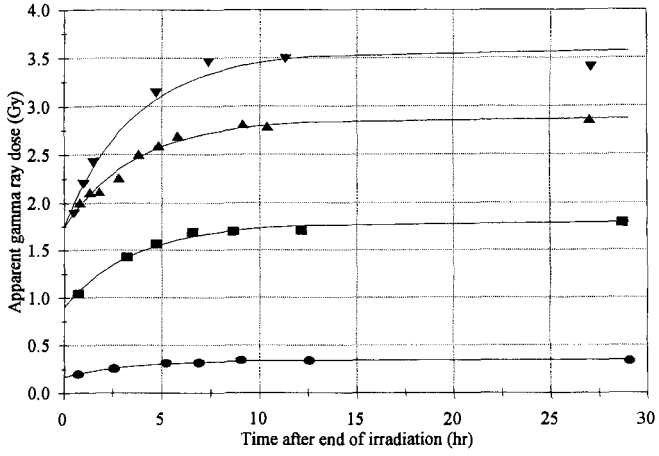

Fig. 2. Sensitivity of TLD-400 chips to thermal neutron fluence (● 1.1 × 10^{12} n cm^{-2}; ■ 6.3 × 10^{12} n cm^{-2}; ▲ 9.2 × 10^{12} n cm^{-2}; ▼ 1.1 × 10^{13} n cm^{-2}). The increase in apparent gamma ray dose above the value at time zero is due to the dose deposited in the TLD from thermal neutron-induced ^{56}Mn decay.

Table 1. Relative effects of gamma rays and neutrons on TLD-400 chips.

Thermal neutron fluence (n cm^{-2})	Gamma ray-induced TL (μC)	Neutron-induced TL (μC)	Correlation coefficient
1.1×10^{12}	1.56	1.47	0.9827
6.3×10^{12}	7.94	7.90	0.9849
9.2×10^{12}	15.43	10.36	0.9677
1.1×10^{13}	15.42	16.86	0.9761

long, i.e., say, greater than five ^{56}Mn half lives (~13 h) post-irradiation period]. The mean thermal neutron-induced beta decay response was $1.5 \pm 0.2 \times 10^{-13}$ Gy per n cm^{-2}, where the absorbed dose was determined from the ^{60}Co TLD calibration curve. This value is consistent with previous measurements (1.0 to 1.3 $\times$ 10^{-13}, Reddy et al. 1969; 1.4×10^{-13}, Horowitz et al. 1977). However, the presently reported value is only due to the beta decay-induced component, whereas the values reported in Horowitz et al. (1977) are total neutron-induced values (about 80% of which is from beta decay).

The manganese content of the OSU TLD-400 chips was determined by neutron activation analysis. Five chips were randomly chosen from a single lot. Their manganese content was 2.44 ± 0.11 (1σ) weight percent.

A more meaningful comparison with the thermal neutron sensitivity results of previous authors is the beta decay contribution only on a per manganese mass basis. In this case, the OSU value is 6.1×10^{-14} Gy per n cm^{-2} per % manganese, and Horowitz's value (corrected to a thermal neutron spectrum) is 5.6×10^{-14} Gy per n cm^{-2} per % manganese (Horowitz et al. 1977).

For the mixed radiation field associated with the OSU BNCT experiments (reactor gamma ray dose to thermal neutron fluence ratio of 2.1×10^{-13} Gy per n cm^{-2}), TL_γ and TL_n are generally comparable in magnitude (see Table 1). Thus for even a 1-h delay in reading out TLD exposed to this mixed field, an approximately 24% increase in the total recovered TL results. Without correction, this additional TL would otherwise be attributed to the reactor gamma ray dose. Thus, readout of the TLD-400 chips within one-half hour (the shortest time practical for the OSU BNCT experiments) after the end of irradiation was developed as a protocol for these irradiations. Without correction, this results in no more than a 13% increase in the reported reactor gamma ray dose. However, as described below, an algorithm has now been developed to correct all TLD-400 responses for the ^{56}Mn beta decay neutron-induced contribution. This method does not require irradiation of a second TLD. All information can be obtained from a dual count of a single TLD. Furthermore, it does not depend on knowing the fraction of beta energy deposited in the chip nor on a dose per disintegration factor.

Consider N_{55} atoms of ^{55}Mn originally in a TLD-400 chip. The activity of the ^{56}Mn during the reactor irradiation is

$$A_{\text{irr}}(t) = R(1 - e^{-\lambda t}), \tag{2}$$

where the absorption rate $R = N_{55} \int \sigma_{a55}(E)\, dE$. At time t after the end of irradiation, the ^{56}Mn activity is

$$A_{\text{post}}(t) = A_{\text{irr}}(t_0)\, e^{-\lambda t}, \tag{3}$$

where t_0 is the irradiation time.

The TLD dose D_1 is read out as soon as possible (at time t_1) after the end of irradiation. D_1 is the sum of the gamma ray-induced dose D_γ and the neutron-induced ^{56}Mn beta decay dose up to time t_1, i.e.,

$$D_1 = D_\gamma + D_n(t_1). \tag{4}$$

If it is assumed that a fraction f of the beta particle energy emitted in the ^{56}Mn decay is deposited in the TLD chip, then the dose contributed by the neutron-induced ^{56}Mn beta decay and subsequent decay up to time t_1 after the end of irradiation is

$$\begin{aligned} D_n(t_1) &= fK \int_0^{t_0} A_{\text{irr}}(t)dt + fK \int_0^{t_1} A_{\text{post}}(t)dt \\ &= \frac{fKR}{\lambda}[\lambda t_0 - e^{-\lambda t_1}(1 - e^{-\lambda t_0})], \end{aligned} \tag{5}$$

where K is a conversion constant (dose per disintegration).

The same TLD chip can subsequently be read out at an "infinite" time after the end of irradiation, e.g., the next day, to give a dose $D_n(>t_1)$, which is given as the difference of eqn (5) evaluated at $t \to \infty$ and $D_n(t_1)$, i.e.,

$$D_n(>t_1) = fKR\, t_0 - D_n(t_1). \tag{6}$$

$D_n(>t_1)$ represents the total neutron-induced ^{56}Mn beta decay dose after time t_1.

Substitution of eqn (6) into eqn (5) allows the elimination of K, f, and R. Then,

$$D_n(t_1) = D_n(>t_1)\left[\frac{\lambda t_0\, e^{\lambda t_1}}{1 - e^{-\lambda t_0}} - 1\right]. \tag{7}$$

Substitution of eqn (7) into eqn (4) permits the gamma ray-induced dose to be determined in terms of the two measured quantities D_1 and $D_n(>t_1)$ and independent of f and K, namely

$$D_\gamma = D_1 - D_n(>t_1)\left[\frac{\lambda t_0\, e^{\lambda t_1}}{1 - e^{-\lambda t_0}} - 1\right]. \tag{8}$$

CONCLUSIONS

Recent experience with a TLD-based gamma ray dosimetry program for BNCT has brought to light several issues surrounding the use of TLD-400 and TLD-700 in a high intensity, mixed gamma ray/thermal neutron radiation field. Most significant was the effect of neutron-induced manganese activation upon the total TL of exposed TLD-400 chips, since under certain conditions this effect can approximately double the TL output if the TLD are not read out until after several ^{56}Mn half lives. A mathematical model has been presented to correct for the neutron-induced ^{56}Mn beta decay contri-

bution to the TL in TLD-400 chips. This model does not require any supplemental measurements by other dosimeters. Although the fraction of the ^{56}Mn beta energy that is deposited in the TLD chip is difficult to calculate for a given TLD chip geometry (Horowitz et al. 1977), the technique described above is independent of the size of the TLD chip, since it does not depend on the fraction of the beta energy which is deposited in the chip. The alternative of using TLD-700 chips is unattractive because of their relatively high ^{6}Li content, i.e., 0.034 to 0.087% (Ayyangar et al. 1968).

Acknowledgments—This work was supported in part under Special Research Subcontract No. C91-103307 by the INEL BNCT Research Program, Idaho Falls, ID.

REFERENCES

Albertson, B. D.; Millan, M. A.; Binney, S. E.; Willeke, G. B.; Martsolf, S. W.; Johnson, J. E.; Loriaux, D. L. The use of boron neutron capture for the treatment of pituitary tumors. In: Barth, R. F.; Soloway, A. H., eds. Proceedings of the 5th International Symposium on Neutron Capture Therapy. New York: Plenum Publishing; 623–628; 1993.

Ayyangar, K.; Reddy, A. R.; Brownell, G. L. Some studies on thermoluminescence from LiF and other materials exposed to neutrons and other radiation. In: Proceedings of the 2nd International Conference on Luminescence Dosimetry. Springfield, VA: U.S. AEC; U.S. AEC Report CONF-680920; 525–542; 1968.

Barth, R. F.; Soloway, A. H.; Fairchild, R. G.; Brugger, R. M. Boron neutron capture therapy for cancer. Cancer 70:2995–3007; 1992.

Cameron, J. R.; Suntharalingam, N.; Kenney, G. N. Thermoluminescent dosimetry. Madison, WI: The University of Wisconsin Press; 1968.

Horowitz, Y. S.; Ben Shahar, B.; Dubi, A.; Pinto, H. Thermoluminescence in CaF_2:Dy and CaF_2:Mn induced by monoenergetic, parallel beam, 81.0 meV diffracted neutrons. Phys. Med. Biol. 22:500–510; 1977.

Horowitz, Y. S. Thermoluminescence and thermoluminescent dosimetry, Vol. I. Boca Raton, FL: CRC Press; 1984.

Reddy, A. R.; Ayyangar, K.; Brownell, G. L. Thermoluminescence response of LiF to reactor neutrons. Radiat. Res. 40:552–562; 1969.

Slatkin, D. N. A history of boron neutron capture therapy of brain tumors. Brain 114:1609–1629; 1991.

■ ■