A new method to verify MCNP-calculated thermal neutron fluence using TLDs and a cadmium sheet

Ahmed I. Khalil, Sangmin Lee, Hyegang Chang, and Sung-Joon Ye* Program in Biomedical Radiation Sciences, Department of Transdisciplinary Studies, Graduate School of Convergence Science and Technology, Seoul National University, Seoul, Korea *sye@snu.ac.kr

Abstract: Thermoluminescent dosimeters are commonly used to obtain neutron dose contribution by subtracting the readout of TLD-700 from TLD-600; however, they carry limited information about the thermal neutron fluence or dose contribution. This study aims to estimate neutron spectrum, thermal neutron fluence and energy deposition in TLDs numerically and experimentally. The two types of TLDs (600 and 700) were grouped into directly exposed to the neutron field of Cf-252, shielded in the front by one cadmium sheet or completely shielded inside a folded cadmium pocket. Each case was simulated and experimentally evaluated. Simulation results showed that a cadmium sheet was sufficient to completely shield direct thermal neutrons but scattered neutron contribution was non-negligible. Therefore, scattered components must be accounted for. The simulation results showed fair agreement with experimental data and were verified using a calibrated ³He neutron measurement probe.

1. Introduction

Thermal neutron dose estimation is important in many fields, especially in BNCT where thermal neutrons may cause scalp necrosis and may have other adverse effects as thermal neutrons do not penetrate to deep seated tumors [1]. BNCT beams are usually monitored using paired ion chambers; however, these do not provide information on thermal neutron fluence or thermal neutron's dose contribution. Instead of the commonly used activation foil method, we propose herein a method for identifying the thermal neutron dose contribution from a Californium-252 source by using pairs of TLD-600, TLD-700, and one Cadmium sheet. The high cross section of 6Li contained in TLD-600 makes them more sensitive to neutrons whereas 7Li that makes up TLD-700 chips makes them insensitive to neutrons due to its low cross section to thermal neutrons. TLDs are grouped and are either directly exposed or placed behind a Cadmium sheet where they are shielded from direct thermal neutrons; this is due to Cadmium's energy cutoff (19,820 barn) to thermal neutrons with energy below 1 eV. We have folded the cadmium sheet to fully enclose another group of TLDs to account for scattered thermal neutrons from walls and ambient air. We can then estimate the thermal neutron contribution by subtracting the obtained neutron dose of the shielded TLDs from the unshielded ones. We have experimentally irradiated the TLDs to obtain only neutron signal. Neutron fluence and ambient dose at the same geometric location can be estimated using Monte Carlo calculations and then verified using a calibrated ³He neutron measurement probe (LB6411).

2. Materials and Methods

2.1 Experimental work

A day prior to the experimental irradiation, a new batch of 80 TLD-600 and 80 TLD-700 were annealed in an electric muffle furnace at 400°C for 1 hour then at 100°C for 2 hours, this is done to eliminate prior residual signal.

A Cf-252 neutron source was used to irradiate a batch containing 60 TLD-600 and 60 TLD-700 chips, the 40 remaining TLDs were left unexposed at a nearby location for background readings. The TLD chips used are supplied from Harshaw Chemical; they were in the form of disks that measure 4.5×0.6 mm. According to the manufacturer, the chips composition was LiF:Mg,Ti, with LiF being the major component per weight ~99%. TLD-600 chips are mainly enriched by ⁶Li (95.6%), and contain small amounts of ⁷Li (4.4%) whereas TLD-700 principally contains ⁷Li (99.99%) and trace amounts of ⁶Li (0.01%) [5]. Despite its minuscule presence in TLD-700, the contribution of 0.01% ⁶Li is non-negligible and adds a small peak that may be visible only from neutron irradiation [6].

60 TLD of each type were divided into 3 groups. Each group consists of 20 TLD-600 and 20 TLD-700 as follows: The first group of TLDs was directly exposed to the neutron beam and thus was exposed to fast and thermalized neutrons (A). The second group was shielded from the front by a 1 mm thick cadmium sheet only (B); this shields it from direct thermal neutrons. The third group was inserted into a pocket shape folded Cadmium; this made it practically shielded from thermal neutrons from all directions, scattered and direct thermal neutrons (C). The contribution of photons is equal in TLD-600 and TLD-700 in a given group, but since TLD-

700 is relatively irresponsive to neutrons while TLD-600 is responsive, then by subtracting the signal of TLD-700 from TLD-600 we can obtain the pure signal from the neutron contribution. Figure 1. shows a simplified geometric setup of the TLDs.



Figure 1. A simplified schematic of the TLD positioning and the Cd sheet

A TLD holder was fabricated using minimal components to not have any effect on the neutron beam, simple cellophane tape was used to hold the TLDs facing the Cf-252 source in a cardboard paper frame. The TLD setup was positioned at 1 meter from the source behind a 50 cm long shadow cone. Irradiation was directed for exactly 48 hours. The californium source has a neutron emission rate of 2.073×10^6 n/s at the calibration date 1.7.2018, and is calculated to be 1.789×10^6 n/s at the start of the irradiation date 23.1.2019. Measurements were taken after the TLD irradiation using a Berthold LB6411 neutron probe to estimate H*(10).

All the TLDs were readout 6 hours after irradiation using a Harshaw 3500 TLD reader, they were individually labelled, and readout following the standard time temperature profile (TTP) for TLD-600. Five outlier TLDs that showed reading deviation of more than 1σ from their averaged group normal distribution value were omitted. Due to TLD-700's non-negligible response to thermal neutrons, we have opted to limit the Region Of Interest (ROI) from channel 16 to channel 66; this was in order to avoid any stray signal caused by thermal neutrons on TLD-700 [6]. Figure 2 shows the averaged TLD-600 glow curve at the mentioned above positions.



2.2 Monte Carlo simulation

A Monte Carlo simulation of the geometry of Pohang University Of Science and Technology (POSTECH)'s neutron irradiation room was modeled using SimpleGeo[2], MCNPX, and then simulated using MCNP6.1 [3]. Since atomic bonds in the moderating polyethylene cone affect thermal neutrons, material treatment was used whenever possible. The shadow cone increased the thermal neutron fluence by thermalizing fast neutrons eminent from the source. Figure 3. (a) Shows a photo of the experimental setup in the calibration room. Figure 3. (b) Shows the geometry of the calibration room as visualized using SimpleGeo.

We have simulated a californium neutron point source with a Watt fission spectrum a = 1.025, b = 2.926 at the real coordinates of the source in POSTECH irradiation room, the source is a commercial Eckert & Ziegler 1 mCi 3036 cylindrical capsule, it is enclosed in an aluminum container. The shadow cone, shield and stage car were all included in the simulation to the nearest measured dimensions, the room measures $830 \times 640 \times 410$ cm with 50 cm thick concrete walls. Neutron spectrum and neutron fluence in n/cm² are obtained using F4 tally, we were able to calculate ambient dose equivalent $H^{*}(10)$ by using ICRP-74 fluence-dose conversion coefficient [4]. Energy deposition in MeV/g was also calculated using F6 tally, and all material cross section data were obtained from BEIR/VII. For this simulation work, we have used 50 Intel Xeon E5-2640 CPU cores and the simulation was continued until tally error was below 5%.



Figure 3. Actual photograph of POSTECH neutron irradiation room (a) and irradiation room as modeled using SimpleGeo (b)

3. Results and Discussion

Simulation results using F4 tally for neutron fluence shows the spectrum using 199 logarithmically separated energy bins in figure 4. To obtain H*(10) we have used MCNP parameter cards to output the F4 in air tally with dose modifier DF4. The calculated ambient dose was $3.585 \ \mu$ Sv/h, which is close to the experimental measurement of $3.460 \ \mu$ Sv/h that was obtained using a Berthold LB6411 neutron probe. The folded cadmium's thermal neutron fluence deviated from the experimental value; This discrepancy may be from the slight difference between simulation and actual dimension of the aluminum material enclosing the source as well as the weakness of the source emission despite the long exposure time.



Figure 4. Neutron spectrum at TLD position using F4 tally in MCNP simulaon

Simulation results using F6 tally (energy deposition) show that a majority of the energy deposit -and thus the dose- in the TLD-600 chips was from thermal neutrons, this is shown in figure 5. By integrating the area under the curve for neutron energies and subtracting the shielded from unshielded TLDs we obtain a result showing that the total energy deposition directly eminent from the source being 48.6% of the total dose contribution, the rest is dose contributed from scatter and fast neutrons, this is in strong agreement with the experimental data. The TLD group inside the folded cadmium pocket shows an energy deposition of 9% that of unshielded TLDs, this corresponds to the epithermal and fast neutron dose contribution. The dose

contribution from neutrons is significantly less in TLD-700 due to its lower cross section.

For the experimental results, the averaged dose read from each TLD-700 group was subtracted from its equally positioned TLD-600 group to obtain the pure neutron signal contribution, when normalized the experimental results prove the simulation results; the experimental results, difference and comparison with simulation are shown in table 1. All values were normalized to the TLD-600 in air position since the experimentally obtained ambient dose equivalent H*(10) was obtained at the same unshielded TLD position.



Figure 5. Energy deposition in MeV/g/history at each TLD position

Table 1. Experimental dose reading ratios compared with
simulated values

	Experimental			
label	TLD-600 signal	TLD-700 signal	<u>Neutron</u> <u>signal</u>	Normalized ¹
Air	8.19	3.85	4.34	1
Cd	5.36	3.13	2.23	0.514
Folded Cd	3.78	3.29	0.49	0.11
signal	Air – Cd (scatter)	2.11	Air - Folded Cd	3.85
	Simulation			
label	MeV/g/n	Normalized ¹	Thermal n/cm²	Normalized ¹
Air	3.56E-05	1	4.31E-07	1
Cd	1.83E-05	0.514	2.01E-07	0.466
Folded Cd	3.19E-06	0.0896	2.32E-08	0.0538
Thermal neutron	Air – Cd	2.30E-7	Air - Folded Cd	4.08E-7

¹Normalized to TLD-600 in air

4. Conclusions

We have established a method to estimate thermal neutron fluence using a pair of TLD-600 and TLD-700, and a cadmium sheet. It was incorporated by means of irradiating the TLDs through a shadow cone using a Cf-252 source and Monte Carlo simulations. The method demonstrated the feasibility to evaluate the thermal neutron dose contribution using the already widely studied and available pair of TLDs specifically TLD-600 and TLD-700. The results showed that in this neutron irradiation setup, the thermal neutron scatter contribution was non-negligible. Therefore, this scatted component must be accounted in estimate of thermal neutron dose. The simulation results matched closely with the experimental values, simulated neutron fluence was further verified by converting it to ambient dose equivalent H*(10) [4] and confirming its value experimentally against a calibrated neutron measurement probe. We concluded that the developed method could sufficiently estimate thermal neutron dose and fluence, instead of a complex and labor-intensive activation foil method.

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