LiF:Mg,Ti TLD response as a function of photon energy for moderately filtered x-ray spectra in the range of 20–250 kVp relative to ^{60}Co

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The response of LiF:Mg,Ti thermoluminescent dosimeters (TLDs) as a function of photon energy was determined using irradiations with moderately filtered x-ray beams in the energy range of 20–250 kVp relative to the response to irradiations with ^{60}Co photons. To determine if the relative light output from LiF:Mg,Ti TLDs per unit air kerma as a function of photon energy can be predicted using calculations such as Monte Carlo (MC) simulations, measurements from the x-ray beam irradiations were compared with MC calculated results, similar to the methodology used by Davis *et al.* [Radiat. Prot. Dosim. **106**, 33–43 (2003)]. TLDs were irradiated in photon beams with well-known air kerma rates using the National Institute of Standards and Technology traceable M-series x-ray beams in the range of 20–250 kVp. For each x-ray beam, several sets of TLDs were irradiated for times corresponding to different air kerma levels to take into account any dose nonlinearity. TLD light output was then compared to that from several sets of TLDs irradiated at similar corresponding air kerma levels using a ^{60}Co irradiator. The MC code MCNP5 was used to account for photon scatter and attenuation in the holder and TLDs and was used to calculate the predicted relative TLD light output per unit air kerma for irradiations with each of the experimentally used photon beams. The measured relative TLD response as a function of photon energy differed by up to 13% from the MC calculations. We conclude that MC calculations do not accurately predict the relative response of TLDs as a function of photon energy, consistent with the conclusions of Davis *et al.* [Radiat. Prot. Dosim. **106**, 33–43 (2003)]. This is likely due to complications in the solid state physics of the thermoluminescence process that are not incorporated into the simulation.
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Key words: thermoluminescence dosimetry, Monte Carlo, energy response, brachytherapy

I. INTRODUCTION

Thermoluminescent dosimeters (TLDs) are commonly used to measure dose from ionizing radiation. In particular, TLDs have been used extensively to measure brachytherapy source parameters such as the dose-rate constant. The dose-rate constant of a brachytherapy seed is defined by the American Association of Physicists in Medicine (AAPM) Task Group 43 to be the ratio of the dose rate at 1 cm along the transverse axis of the source in water to the source air-kerma strength.[1–3] Most dose-rate constant measurements are performed using TLDs placed in a phantom around a source of known air-kerma strength. The light output from the TLDs is then compared to that from TLDs irradiated in a phantom for which the dose to water is known in the absence of the TLD. The reference irradiation is usually from a higher energy source such as ^{60}Co or a 4 or 6 MV linear accelerator, which have well-established dosimetry protocols based on absorbed dose to water. The output from these sources can be calibrated using the AAPM Task Group 51 protocol,[4] or the International Atomic Energy Agency TRS-398 protocol,[5] both of which can provide relatively low uncertainties on the dose to water at a reference depth in phantom. The light output from the TLD per unit dose to water is determined with the reference beam, and the TLD calibration curve is used to determine the dose to water from the TLDs irradiated with the low-energy brachytherapy seed.[1–3] However, LiF TLDs have different photon interaction cross sections than water, so a correction must be applied for the change in TLD light output per unit dose to water as a function of photon energy.[6] This is usually referred to as "energy response" in the literature, and in the context of brachytherapy dosimetry it can also include corrections for volume averaging, detector self-absorption, medium displacement, and conversion of measurements in a plastic phantom to results in liquid water.[1] Since the term "energy response" can have multiple meanings, we will avoid its usage here. The terminology here will refer to the TLD light output per unit of a given quantity, either air kerma in the absence of the TLD, dose to water in a phantom in the absence of the TLD, or absorbed dose in the TLD itself. "TLD response" will be used as a general term to describe the TLD light output per unit of an arbitrary quantity and can be equally applied to results in terms of air kerma, dose to water, or dose to TLD. The TLD response as a function of photon energy is the area of investigation in this work, and because the results are normalized at a reference photon energy (^{60}Co here), it will be referred to as the relative TLD response.

Several studies frequently cited in the brachytherapy literature have investigated the LiF:Mg,Ti TLD energy correction for low-energy brachytherapy sources such as ^{125}I($\sim$28 keV) and ^{103}Pd($\sim$21 keV). Hartmann *et al.*[7] measured a TLD light output per unit dose to tissue of $1.40 \pm 2.8\%$ for 40 kV x rays relative to ^{60}Co photons but provided very few details of their experimental technique. Weaver[8] used an array of ^{125}I seeds to measure a TLD light output per unit dose to water of 1.39 ± 0.03 relative to ^{60}Co photons. A later article by Weaver *et al.*[9] measured a TLD light output per unit dose to water of $1.47 \pm 5\%$ relative to ^{60}Co photons for model 6702 ^{125}I seeds and $1.42 \pm 5\%$ for model 6711 ^{125}I seeds. Meigooni *et al.*[10,11] published two separate articles in 1988 with a measured TLD light output per unit dose to water of $1.41 \pm 3\%$ for 60, 80, and 100 kV x rays relative to 4 MV x rays. Muench *et al.*[12] measured a TLD light output per unit dose to water of 1.41 for 60 kV x rays relative to 4 MV x rays but did not provide an estimated uncertainty on their results. Many of these studies were reviewed in a chapter by Willamson and Rivard from the proceedings of the AAPM 2005 Brachytherapy Physics Summer School.[13] Some of the shortcomings with these studies are that hand calculations were used to determine the conversion from exposure for the low-energy sources free-in-air to absorbed dose to water, and only the articles by Weaver *et al.*[8,9] took into account photon attenuation or scatter effects in the TLDs or supporting apparatus. The results from these studies were all in good agreement with the predicted TLD light output per unit dose to water of 1.41–1.42 calculated using a simple ratio of mass energy-absorption coefficients from LiF to water for 20–30 keV photons relative to ^{60}Co photons.[14] The good agreement between all of these studies led to a common TLD energy correction of $\sim$1.4 being used for most TLD determinations of the dose-rate constant for low energy brachytherapy sources.[1–3]

More recent studies by Das *et al.*[15] and Davis *et al.*[16] have used Monte Carlo (MC) methods to calculate the dose to TLD for different energy photon beams to determine whether the measured TLD light output is directly proportional to the energy deposited (or dose) in the TLD. This direct proportionality is also referred to as "intrinsic linearity."[1,13] If the measured TLD light output per unit dose to TLD is independent of photon energy, then MC calculations could be used reliably to calculate TLD corrections for a wide variety of irradiation conditions based on the calculated dose to TLD. Given the recent influx of new brachytherapy sources, determining if TLD response can be modeled accurately using MC is vital, as some researchers use MC as the sole method for determining the energy correction for TLDs used in brachytherapy seed measurements.[17–22]

The AAPM Task Group 43 Update (TG-43U1) (Ref. 1) report highlights the investigations by Das *et al.*[15] and Davis *et al.*[16] because they give contradictory conclusions regarding the measured TLD light output per unit dose to TLD as a function of photon energy. Das *et al.*[15] concluded that the measured TLD light output per unit dose to TLD is constant as a function of photon energy, while Davis *et al.*[16] found an energy dependent change in TLD response. Figure 1 presents the measured TLD light output per unit calculated dose to TLD normalized to the high-energy calibration source for each of these publications. A constant result at unity would represent no dependence on photon energy for the measured TLD light output per unit dose to TLD. Das *et al.*[15] used both large ($3.2 \times 3.2 \times 0.9$ mm^3) and small ($1 \times 1 \times 1$ mm^3) chips, whereas Davis *et al.*[16] used thin chips ($3.2 \times 3.2 \times 0.4$ mm^3). The TLDs in both investigations were composed of the TLD-100 formula (LiF:Mg,Ti) from Harshaw (now Thermo Electron Corporation, Oakwood Village, OH).[15,16]

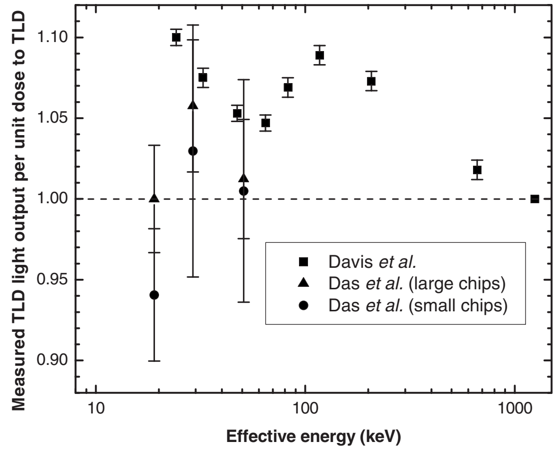

FIG. 1. Results of Das *et al.* (Ref. 15) (● small and ▲ large chips) and Davis *et al.* (Ref. 16) (■) for relative TLD light output per unit dose to TLD as a function of photon energy for LiF:Mg,Ti TLDs.

Das *et al.*[15] measured TLD light output per unit air kerma for irradiations from three lightly filtered x-ray beams ranging from 40 to 125 kVp relative to the light output per unit air kerma for irradiations from a 4 MV x-ray beam. A Monte Carlo photon transport (MCPT) code was used to calculate the dose to TLD per unit air kerma for the irradiations from each of the x-ray beams. The code transported photons only and calculated the collision kerma in the TLD. A cavity theory correction from 0% to 2% was applied to determine the dose to the TLD from the collision kerma. The investigators concluded that there was no significant difference between the measured and calculated relative TLD response as a function of photon energy, and that the TLD light output was directly proportional to the absorbed dose in the active volume of the detector.[15]

Davis *et al.*[16] measured TLD light output per unit air kerma at the National Research Council Canada using the International Organization for Standardization narrow spectrum series (N series) x-ray beams ranging from 30 kV (N30) to 250 kV (N250).[23] The measurements were normalized to the TLD light output per unit air kerma for a ^{60}Co beam. This investigation used the MC code EGSnrc,[24] which transports both photons and electrons through the detector and determines dose in the TLD, eliminating the need for a cavity theory correction. The measured relative TLD light output per unit air kerma as a function of photon energy was compared with the MC calculated relative TLD dose per unit air kerma, and differences of up to 10% were seen, as shown in Fig. 1.[16]

Although the Davis *et al.*[16] investigation has been given as an example, other studies have also reported a difference between the measured relative TLD response as a function of photon energy and the calculated relative TLD response. The first published measurements of TLD light output per unit exposure at different photon energies were done by Cameron *et al.* in 1964.[25] In 1968, Tochilin *et al.*[26] built on Cameron's earlier work and noted that neither set of TLD response data could be explained by the difference in mass energy-absorption coefficients of LiF and air. Budd *et al.*[27] in 1979 reported an average 10% difference between measured and calculated TLD response as a function of photon energy relative to ^{60}Co at energies below 150 keV. This difference falls in the same range as the Davis *et al.*[16] data, but with much larger uncertainties. In 2002, Olko *et al.*[28] also reported a 10% increase in measured versus calculated LiF:Mg,Ti TLD response in an energy range of 20–200 keV relative to ^{137}Cs.

There is no *a priori* reason that the measured light output from LiF:Mg,Ti TLDs for changing photon energy should always be directly proportional to the absorbed dose in the detector volume. The ionization density of the secondary electrons is not constant for photon beams with energies from 1 keV to 20 MeV, and there are well-known ionization density effects with a variety of radiation dosimeters. LiF:Mg,Ti TLDs have a supralinear dose response at high doses (>10 Gy), and the density of the energy deposition in the TLD is thought to play a major role.[29,30] LiF:Mg,Ti TLDs have also been shown to have a much different measured light output to high linear energy transfer (LET) radiation per unit absorbed dose (e.g., alphas, neutrons, and protons) than for low LET radiation (e.g., photons, electrons).[30,31] Another LiF-based TLD, LiF:Mg,Cu,P (TLD-100H), has been shown to have differences of up to 35% between calculated and measured TLD response as a function of photon energy for x-ray beams from 30–250 kV.[16,32] Aside from TLDs, other media also demonstrate changes in response for varying irradiation energies. For example, the average energy required for ionization of dry air, $\overline{W}/e$, is relatively constant over a wide range of photon energies of interest in medical physics, but it departs significantly when the secondary electron energies drop below 1 keV.[33] In addition, in Fricke dosimeters the efficiency of ferric ion production, or $G(\mathrm{Fe}^{3+})$ value, has been shown to have a small dependence on photon energy between ^{60}Co photons and 20 and 30 MV photon beams.[34] None of these effects would be predicted with conventional Monte Carlo simulations that simply calculate the total energy deposited in a dosimeter, without regard for ionization density. For LiF TLDs, several investigators have had success modeling some of these behaviors by taking into account microdosimetric aspects of the thermoluminescence process that are not currently modeled by conventional MC codes.[28,30]

The TG-43U1 report has assigned an uncertainty of 5% on the TLD energy correction.[1] The uncertainty on this correction is the single largest contribution to the TLD measurement uncertainties outlined in TG-43U1. The report suggested that further investigations are necessary to help resolve these discrepancies and help reduce the large uncertainty associated with these corrections.

This work examines the response of LiF:Mg,Ti TLDs to irradiations with moderately filtered x-ray beams in the range of 20–250 kVp (effective energies of 11.5–157 keV) and ^{137}Cs (662 keV) both relative to irradiations with ^{60}Co (1250 keV) photons. The methodology is similar to the one presented by Davis *et al.*[16] The aim of this investigation is to determine if the measured TLD light output per unit absorbed dose to TLD has any dependence on photon energy in the range relevant for brachytherapy dosimetry (20–1250 keV).

II. METHODS AND MATERIALS

Harshaw TLD-100 chips ($3.2\times3.2\times0.9$ mm^3) (Thermo Electron Corporation, Oakwood Village, OH) were used for all measurements. The Cameron method of annealing was used for all the TLDs.[25] The TLDs were first placed in an annealing tray and placed in a 400 °C oven for 1 h. This was followed by 20–30 min cooling to room temperature on an aluminum plate. The TLDs were then placed in an 80 °C oven for 24 h and subsequently cooled to room temperature where they remained for at least 24 h and no more than 3 days before any irradiations took place. The tray used for annealing was made of aluminum and had holes drilled out to hold the individual TLDs in place. After construction, the aluminum tray was heated to 400 °C for at least 6 h. This was done to oxidize the surface and created Al_2O_3, which does not react with the TLDs.

TLDs were read with a Harshaw 5500 automatic TLD reader 24 h after irradiation. The Harshaw 5500 is a hot gas reader with the photomultiplier tube (PMT) aligned to read from the side of the TLD, which has been mechanically lifted from the holding carousel. The time temperature profile for reading each chip starts when the gas is at 50 °C, at which point the gas temperature increases by 15 °C/s to a maximum of 350 °C where it is held constant for 26.7 s, after which the gas is cooled to 50 °C before the next chip is read. The PMT charge is integrated over the temperature ramp-up and plateau regions of the heating profile.

The Harshaw 5500 PMT was found to have a nonlinear over response at high currents. A series of measurements were conducted by the University of Wisconsin Medical Radiation Research Center (UWMRRC) TLD laboratory to characterize the PMT nonlinearity, and a correction algorithm was implemented to correct the integrated charge under the glow curve for every TLD reading. The corrections ranged from 0% to 4.8% for this work depending on the light output of the TLD. A TLD that emitted more light had a higher PMT nonlinearity correction than a TLD that emitted less light. Without correcting for the PMT over response, the TLD data demonstrated a nonlinear dose response in the range from 40–100 cGy, where TLD-100 is expected to be linear.[6] After the correction algorithm was applied, the TLDs demonstrated a linear dose response in this range within $\pm 0.3\%$.

For the measurements, 80 TLDs were chosen based on sensitivity (within 1.5% of the mean reading) and reproducibility (within 1.5%) from an initial batch of 300 TLDs. Chip calibration coefficients were then determined for each of these 80 TLDs to account for their relative response to ^{60}Co irradiation. A chip calibration coefficient (n_{cc}) is the individual chip's light output per unit air kerma when irradiated by ^{60}Co photons. A description of the holder used for the ^{60}Co irradiations is provided in the next section.

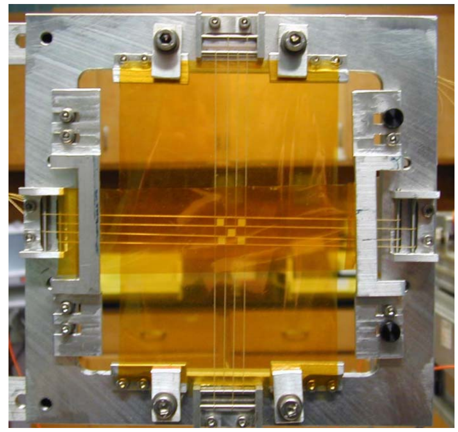

FIG. 2. Specialized holder for x-ray TLD irradiation. The holder consists of two Kapton® sheets (0.025 mm thick) and Kevlar® threads.

II.A. TLD Irradiations

For the ^{60}Co irradiations, the chips were irradiated in a square 10×10 cm^2 poly(methylmethacrylate) (PMMA) holder with a 5.2 mm thick front face and 8.8 mm thick back face. They were held in machined holes approximately 6 mm in diameter. The holder had room for 100 TLDs, with one TLD per hole and horizontal and vertical spacing of 8 mm between the centers of adjacent holes. The holes were 1.1 mm deep, so when a TLD was in place there was a total of about 0.2 mm air gap between the front and back PMMA walls and the faces of the TLD. The front face was thick enough to provide charged particle equilibrium for the ^{60}Co photon beam. The same holder was used for the ^{137}Cs irradiations, but a thinner front plate of 2.9 mm was used to ensure charged particle equilibrium.

The University of Wisconsin Accredited Dosimetry Calibration Laboratory's (UW-ADCL) Theratronics El-Dorado 78 ^{60}Co irradiator was used with a 20×20 cm^2 field size with the front surface of the TLDs at a distance of 100 cm from the source. In one case a 20×20 cm^2 field size at 200 cm was used to obtain a lower air kerma rate. The TLD holder was centered along the central axis of the ^{60}Co beam each time using ceiling and wall-mounted lasers. Irradiation times were determined from air kerma measurements on a reference date using a National Institute of Standards and Technology (NIST) calibrated ionization chamber and were corrected for source decay. The minimum division of programmed irradiation time was 0.01 min, and a shutter error of 0.012 min was factored into the calculated irradiation times, the minimum of which was 0.30 min.

The ^{137}Cs source used for irradiations was the UW-ADCL's Hopewell G10 ^{137}Cs irradiator. The irradiated field size was 10×10 cm^2 with the front surface of the TLDs at 100 cm from the source. Air kerma rates for irradiation were determined on the day of exposure using a NIST calibrated ionization chamber. As it was for the ^{60}Co beam, the TLD holder was centered along the central axis of the ^{137}Cs beam using ceiling and wall-mounted lasers.

A special TLD holder was constructed for the x-ray irradiations and is shown in Fig. 2. The holder was created to allow the TLDs to maintain their position while also being as close as possible to free-in-air conditions. The holder caused minimal attenuation and scatter of the x-ray beam and consisted of a square aluminum frame with a cutout region in the middle measuring 12×12 cm^2, which is larger than the 10×10 cm^2 x-ray field. Kevlar® threads were strung across the square both vertically and horizontally creating nine "holes" for the TLDs to sit in. The threads were approximately 0.25 mm in diameter. Five TLDs were irradiated at the same time and were placed in an "X" pattern to minimize scatter from one TLD to the other. The Kevlar® threads were used to separate and stabilize the TLDs while they were irradiated. Two 0.025 mm thick Kapton® sheets were placed, one in front of the Kevlar® threads and one behind, to keep the TLDs upright during irradiation. The Kapton® was pulled taut so that the front and back sheets touched the surface of the TLDs. As with the other beams, the holder was centered along the central axis of the x-ray beam using ceiling and wall-mounted lasers.

The x-ray device used was an Advanced X-Ray (AXI) constant potential x-ray system with a Gulmay CP 320 generator and a Comet 320 tungsten anode tube. All irradiations with the x-ray unit were performed with the front surface of the TLDs at 100 cm from the source and a field size of 10×10 cm^2. The beam code, first half-value layer (HVL), and the homogeneity coefficient (HC) for the x-ray beams used are presented in Table I. The x-ray beams were carefully matched for first and second HVL with the comparable x-ray beams at NIST, so the NIST beam codes were used here for simplicity.[35] Also presented in Table I are the tabulated beam data from the Gesellschaft für Strahlen und Umweltforschung (GSF) compilation by Seelentag *et al.*,[36] used for the Monte Carlo simulations of the x-ray beams.

Before TLD measurements of a specific x-ray beam were performed, the air kerma rate was measured for that beam using a NIST calibrated ionization chamber. The chambers have a 1.0% uncertainty ($k=2$) in their calibration coefficient from NIST. The TLDs were irradiated to well known "air kerma levels," which means that the irradiation timer was set to deliver a known air kerma at 100 cm if the TLDs and

TABLE I. Beam codes, first HVL, HC, and effective energy of x-ray beams used for experiments. The effective energy is the monoenergetic energy with the same first HVL. Also listed are the tabulated spectra from the GSF compilation (Ref. 36) used as initial spectra for the Monte Carlo simulations. The first HVL and effective energy are compared to that of the experimental x-ray beams.

	UW beams			GSF beams		
Beam codes	1st HVL (mm Al)	HC	Effective energy (keV)	GSF spectrum No.	1st HVL (mm Al)	Effective energy (keV)
M20	0.148	75	11.5	11	0.16	11.8
M30	0.356	65	15.5	21	0.38	15.9
M40	0.728	66	19.8	32	0.71	19.7
M50	1.02	66	22.4	43	1.04	22.6
M60	1.68	66	26.9	49	1.54	26.0
M80	2.96	68	33.5	60	2.76	32.5
M100	4.98	72	42.1	71	5.02	42.2
M120	6.96	78	49.9	80	6.31	47.5
M150	10.2	87	67.0	94	10.2	67.2
M200	14.9	98	99.8	107	14.7	98.6
M250	18.5	98	145	123	18.0	140
L100	2.80	58	32.7	67	2.64	31.9
				68	3.02	33.8
H100	13.4	99	85.9	75	12.9	81.4

holder were not present. This terminology will be used throughout the article. To account for any dose dependent nonlinearity, TLDs were irradiated to four air kerma levels (40, 60, 80, and 100 cGy) using five TLDs for each irradiation. These TLDs were then compared to TLDs irradiated using ^{60}Co the same day. The TLDs irradiated using ^{60}Co were irradiated in sets of five TLDs to three known air kerma levels (44, 66, and 88 cGy). It is not critical that the air kerma irradiation levels for the x-ray beams and ^{60}Co were different as the comparison was made using TLD light output per unit air kerma. All of the photon beams had a field flatness of less than 1% over the central 80% of the field and considerably better flatness over the small area covered by the TLDs. To account for background exposure, four chips were not irradiated and were used as controls. The ^{60}Co chip calibration coefficients for the entire set of TLDs were remeasured once after each set of TLD response measurements to ensure that the chip calibration coefficients had not changed substantially and to maintain a high level of precision.

Beam qualities of H100 (highly filtered) and L100 (lightly filtered) were used to allow comparison of TLD response to the Davis *et al.*[16] N100 data, and to look at differences with beam quality for the same kilovoltage setting of 100 kV on the x-ray tube. Due to the low air kerma rate from the H100 beam, three sets of five TLDs were irradiated to air kerma levels of 4.4, 6.5, and 8.7 cGy. For the ^{60}Co reference for these TLDs, three sets of five TLDs were irradiated at 200 cm from the ^{60}Co source. The air kerma levels were 8.5, 11.5, and 14.5 cGy.

II.B. Monte Carlo calculations

If the light output from the TLD is directly proportional to the absorbed dose in the TLD, and certain conditions outlined below are met, then the TLD light output per unit air kerma as a function of monoenergetic photon energy could be calculated using the ratios of the mass energy-absorption coefficients for the TLD and air. The TLD light output per unit air kerma relative to ^{60}Co photons would be calculated using

$$C_{\mathrm{a}}(E) = \frac{[(\mu_{\mathrm{en}}/\rho)_{\mathrm{TLD}}/(\mu_{\mathrm{en}}/\rho)_{\mathrm{air}}]_E}{[(\mu_{\mathrm{en}}/\rho)_{\mathrm{TLD}}/(\mu_{\mathrm{en}}/\rho)_{\mathrm{air}}]_{^{60}\mathrm{Co}}}, \tag{1}$$

where C_{a} denotes the analytical calculation of the relative TLD light output per unit air kerma as a function of photon energy, and E is the monoenergetic photon energy.[6,16] This calculation can be extended to polyenergetic spectra by using spectrum averaged mass energy-absorption coefficients weighted by the energy fluence, $\overline{\mu_{\mathrm{en}}}/\rho$.

The analytical calculation only holds true under conditions that all the electrons that deposit energy into the TLD are set in motion in the TLD, there is no scatter or attenuation of the photons due to the holder, and there is no attenuation of the photon beam through the TLD.[6] These assumptions are invalid for the work presented here since the finite thickness of the TLD does attenuate the beam and a slight amount of scatter is expected. For these reasons, calculations of dose in the TLDs were performed using MC simulations, and these results will be compared to calculations that use the simple analytical method.

To determine the predicted TLD light output per unit air kerma as a function of photon energy, MC simulations were performed using the MCNP5 code.[37] The mcplib04 (Refs. 38 and 39) and el03 (Ref. 40) libraries were used for photon and electron cross sections. The *F8 tally, which calculates the energy deposition per starting particle, was used to calculate the air kerma and the dose deposited in the TLDs. The *F8 tally uses an energy balance method to determine the energy deposited in a specific geometric volume, known as a "cell." The difference between the amount of kinetic energy entering the cell and the kinetic energy leaving the cell is calculated, with the results normalized by the total number of particle histories.[37] In this way, electrons can deposit energy as they slow down or photons can deposit energy if they are below the predetermined energy cutoff. The MC simulations for the x-ray beam irradiations included the modeled holder with five TLDs in an X pattern, the Kapton® sheets, and the Kevlar® threads.

Following the methodology set forth by Davis *et al.*,[16] the dose to the TLD per unit air kerma $(D_{\mathrm{TLD}}/K_{\mathrm{air}})$ was found using two simulations for each photon beam. The first simulation calculated the dose to the TLD per unit incident fluence with the TLD in the irradiation geometry. The second simulation calculated the air kerma per unit incident fluence in a thin slab (2 μm thick) of air. Electrons were explicitly transported because the electron range was on the order of the size of the TLD for the ^{137}Cs and ^{60}Co beams and thus avoided any cavity theory corrections that otherwise would have been required. The photon energy cutoff was 1 keV for both sets of calculations, and the electron cutoff was 5 keV for the TLD dose calculations and 2 MeV for the air kerma calculations. The high electron cutoff for the air kerma calculations effectively turned off electron transport and meant that kerma was calculated instead of dose. The forced collision variance reduction technique was employed for both the TLD and air kerma simulations to ensure that charged particles were created in the regions of interest.[37] The $D_{\mathrm{TLD}}/K_{\mathrm{air}}$ for each beam was then divided by the same ratio found for the ^{60}Co beam to determine the calculated relative TLD dose per unit air kerma for each energy.

The spectra for the x-ray beams were taken from the Seelentag *et al.* GSF report data,[36] the ^{60}Co spectrum was taken from Mora *et al.*,[41] and the ^{137}Cs spectrum was taken from Seltzer and Bergstrom.[42] The GSF spectra and first HVLs are shown in Table I. Two GSF spectra points are listed for the UW-ADCL's L100 x-ray beam because the HVL fell between two of the listed GSF beams. The composition and densities for air, Kapton®, and PMMA were taken from Berger *et al.*,[43] while the TLD-100 composition and density were from Thermo Electron Corporation (composition by weight of 73.28% Li, 26.70% F, 0.02% Mg, and 0.001% Ti, with a density of 2.64 g/cm^3).[44] The Kevlar® composition was calculated based on the chemical formula of $(C_{14}H_{10}N_2O_2)_n$ from the Kevlar® Technical Guide from DuPont,[45] with a density of 1.44 g/cm^3.

II.C. Data analysis

The raw reading of light output from each TLD was corrected for PMT linearity, background, and individual chip calibration coefficient using

$$R = \frac{M' \cdot k_{\mathrm{lc}} - M_{\mathrm{b}}}{n_{\mathrm{cc}}}, \tag{2}$$

where R is the corrected result (in Gy), M' is the raw integrated charge reading from the PMT (in nC), k_{lc} is the linearity correction applied to the TLD reading from the TLD reader PMT response function, M_{b} is the background reading (in nC) found by reading out nonirradiated control TLDs that were annealed at the same time as the TLDs used for experiments, and n_{cc} is the calibration coefficient unique to each TLD, discussed earlier.

The corrected TLD measurements were divided by the irradiated air kerma level for each TLD, and the results were averaged across all of the air kerma levels for each photon beam. The mean result for a given photon beam, X, was divided by the mean result for the reference TLDs irradiated using the ^{60}Co beam to get

$$M(X) = \frac{\overline{(R/K_{\mathrm{air}})}_X}{\overline{(R/K_{\mathrm{air}})}_{^{60}\mathrm{Co}}}, \tag{3}$$

where $M(X)$ is the measured TLD light output per unit air kerma for photon beam X relative to ^{60}Co, and K_{air} is the irradiated air kerma level (if the TLD and holder were absent).

A similar methodology was used for the analysis of the Monte Carlo simulation results. The MC simulations calculated the dose to the TLD per unit fluence (D_{TLD}/Φ) and the air kerma per unit fluence (K_{air}/Φ), for all x-ray beams used, as well as ^{137}Cs and ^{60}Co. The ratio of these results gave the dose to TLD per unit air kerma $(D_{\mathrm{TLD}}/K_{\mathrm{air}})$. The result for a given photon beam was divided by the results for the ^{60}Co beam to get

$$C_{\mathrm{MC}}(X) = \frac{(D_{\mathrm{TLD}}/K_{\mathrm{air}})_X}{(D_{\mathrm{TLD}}/K_{\mathrm{air}})_{^{60}\mathrm{Co}}}, \tag{4}$$

where $C_{\mathrm{MC}}(X)$ is the Monte Carlo-calculated TLD dose per unit air kerma for photon beam X relative to ^{60}Co.

The ratio of the measured TLD response to the Monte Carlo-calculated TLD response is determined by dividing $M(X)$ by $C_{\mathrm{MC}}(X)$ for each photon beam to get

$$\eta(X) = \frac{M(X)}{C_{\mathrm{MC}}(X)} = \frac{(R/D_{\mathrm{TLD}})_X}{(R/D_{\mathrm{TLD}})_{^{60}\mathrm{Co}}}, \tag{5}$$

where $\eta(X)$ is the measured TLD light output per unit calculated dose to TLD for photon beam X relative to ^{60}Co. If the light output from the TLDs is directly proportional to the absorbed dose to the TLD independent of photon energy, $\eta(X)$ should be unity.

III. RESULTS

The results of the measured TLD light output per unit air kerma and the Monte Carlo-calculated TLD dose per unit air kerma for all of the photon beams relative to ^{60}Co are presented in Table II and plotted in Fig. 3. Also plotted in Fig. 3 is the TLD dose per unit air kerma relative to ^{60}Co calculated with the simple analytical method [Eq. (1)]. The mass energy-absorption coefficients used for the analytical method were based on monoenergetic photon energies and were obtained from the NIST online database.[14] The relative TLD light output per unit air kerma for the H100 and the L100 beams was measured to show the potential differences due to

TABLE II. Measured TLD light output per unit air kerma, $M(X)$, and calculated TLD dose per unit air kerma using MCNP5, $C_{MC}(X)$, both relative to the ^{60}Co irradiations. The respective total combined uncertainty at a coverage factor of $k=1$ is also presented for both sets of data. Also shown is $\eta(X)$, the ratio of measured response to MC calculated response with corresponding percent uncertainty, u, at a coverage factor of $k=1$.

Beam code	Effective E (keV)	$M(X)$ (measured)	$u(M)$ (%)	$C_{MC}(X)$ (MCNP5)	$u(C_{MC})$ (%)	$\eta(X)$ (ratio)	$u(\eta)$ (%)
M20	11.5	0.947	1.4	0.877	6.9	1.079	7.0
M30	15.5	1.151	1.9	1.064	3.0	1.082	3.5
M40	19.8	1.279	1.9	1.165	1.5	1.098	2.4
M50	22.4	1.310	1.8	1.199	1.3	1.093	2.2
M60	26.9	1.372	1.5	1.233	1.2	1.112	1.9
M80	33.5	1.365	1.4	1.231	1.1	1.108	1.7
M100	42.1	1.313	1.4	1.200	1.0	1.094	1.7
M120	49.9	1.279	1.5	1.174	1.0	1.089	1.8
M150	67.0	1.243	1.8	1.115	0.9	1.115	2.0
M200	99.8	1.155	1.3	1.050	0.8	1.099	1.5
M250	145	1.122	1.4	1.010	1.1	1.112	1.8
L100	32.7	1.348	1.5	1.215	1.1	1.109	1.9
H100	88.6	1.199	1.4	1.065	0.9	1.126	1.7
^{137}Cs	662	1.056	1.5	1.018	1.2	1.038	1.9
^{60}Co	1250	1.000	-	1.000	-	1.000	-

differences in spectra shape. The H100 beam has a much narrower spectrum than the M-series beams, while the L100 beam has a broader spectrum with a larger low energy component. The results from these beams agree well with the M-series data when compared using effective energies and seem to show minimal effects from differences in spectra shape (Table II). For clarity, the results from those beams were omitted from the figures so the focus would remain on the M-series beams and because those results slightly perturb the overall trends. This is consistent with NIST Special Publication 250-58,[35] which states that discontinuities can occur when response data from different x-ray beam series are plotted together.

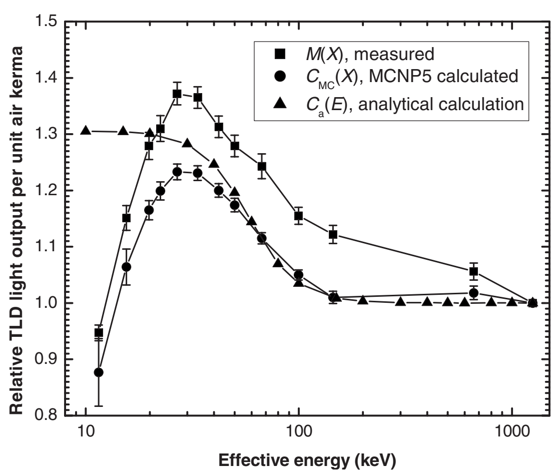

FIG. 3. Measured TLD light output per unit air kerma as a function of photon energy [$M(X)$], as well as calculated dose to TLD per unit air kerma as a function of photon energy for the Monte Carlo [$C_{MC}(X)$] and analytical [$C_a(E)$] methods. All results are normalized to the response to ^{60}Co photons. The decrease in response toward lower photon energies for both $M(X)$ and $C_{MC}(X)$ is due to self-attenuation through the TLD, which the simple analytical method does not take into account. Differences between $M(X)$ and $C_{MC}(X)$ are likely due to solid state physics interactions not being modeled by MCNP5.

At high photon energies, the MC calculated and analytical TLD dose per unit air kerma agree well. However, at energies below approximately 60 keV, the MC and analytical calculations start to diverge. This difference is primarily due to the self-attenuation of the x-ray beam in the TLD, but is also partly due to photon attenuation and scattered photons from the Kapton®, Kevlar® threads, and other TLDs. All photon attenuation and scatter is modeled by the MC simulations, but is not accounted for in the analytical mass energy-absorption coefficient ratio. To verify that this discrepancy at low energies was primarily due to TLD self-attenuation, MC simulations were performed to look at the MC calculated response with varying thickness of TLDs, using monoenergetic 20 keV photons. The simulations were performed with TLD thicknesses of 1, 0.1, and 0.01 mm, and with electron transport turned off so that even the thin TLDs would be considered "large" cavities. The MC calculated TLD dose per unit air kerma was 6.7% lower than the analytical calculation for the 1 mm thick TLD, but this improved to 0.3% lower than the analytical calculation for the 0.1 mm thick TLD and 0.1% higher than the analytical calculation for the 0.01 mm thick TLD. Hence, the deviations seen between the MC calculations and the analytical results for the standard TLD thickness are primarily attributed to the self-attenuation of the beam through the TLD.

The results of the measured TLD light output per unit calculated dose to TLD relative to ^{60}Co [Eq. (5)] are presented in Fig. 4 and Table IV. The TLDs demonstrated an 8%–13% higher measured signal for the x-ray beam irradiations relative to ^{60}Co than was predicted by the MC simula-

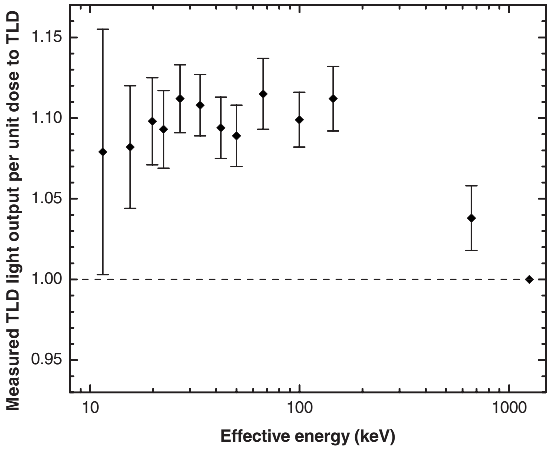

FIG. 4. Ratio of measured TLD light output per unit air kerma to Monte Carlo calculated TLD dose per unit air kerma from Fig. 3. The result is the measured TLD light output per unit calculated dose to TLD, or $\eta(X)$.

tions. For the ^{137}Cs irradiations, the TLDs had a 3.8% higher measured signal relative to ^{60}Co than was predicted by the MC simulations.

III.A. Uncertainty

Sources of uncertainty for the TLD experiments were the TLD reproducibility, air kerma rate determination, TLD positioning, PMT linearity correction, TLD reader stability, and uniformity of the radiation field over the irradiation area. The TLD reproducibility differs for each photon beam as it is the quadratic sum of the standard deviation of the corrected readings [R from Eq. (2)] of the TLDs irradiated at that x-ray beam and the corresponding ^{60}Co readings. The air kerma rate uncertainties were based on the values reported by the UW-ADCL, and stemmed primarily from the uncertainty on the NIST calibration of the reference ionization chambers. The calibration uncertainties of the reference ionization chambers were 0.5% $(k=1)$ for the x-ray beams, 0.75% $(k=1)$ for the ^{137}Cs beam, and 0.7% $(k=1)$ for the ^{60}Co beam. The other measurement uncertainties were constant for each of the photon beams. A sample analysis for the M60 x-ray beam is presented in Table III. Since the measured results were normalized to ^{60}Co, the measurement uncertainty for each beam was added in quadrature with the ^{60}Co measurement uncertainties. Uncertainties for each x-ray beam are listed in Table II with the percent uncertainty expressed with a coverage factor $k=1$.

TABLE III. Example of uncertainty analysis for measured TLD data, using the M60 beam. Values are expressed in percent.

Parameter	Type A		Type B	
TLD reproducibility	0.72			
Air kerma rate determination			0.64	
TLD positioning			0.30	
PMT linearity correction			0.40	
Reader stability			0.20	
Field flatness			0.10	
Quadratic sum	0.72		0.84	
A and B quadratic sum		1.11		
Measured TLD uncertainty			1.11	$\%(k=1)$
			2.21	$\%(k=2)$

Sources of uncertainty for the Monte Carlo simulations include uncertainty in the TLD thickness, TLD positioning, photon spectra, photon cross sections, and statistical uncertainties. Sufficient particles were run with each simulation to reduce the statistical uncertainty for each calculation to less than 0.2%. To determine the uncertainty in the photon cross sections, the methodology proposed in TG-43U1 (Ref. 1) was used. Simulations were performed using the older mcplib03 (Ref. 46) cross section library for photons and consisted of determining $C_{\mathrm{MC}}(X)$ for each beam quality. Most of the differences in photon cross sections between the two libraries are in the photoelectric absorption cross sections, which will tend to affect the lower energy beams. The differences in air kerma results were used to estimate the overall shift in cross sections between the two libraries, and the change in $C_{\mathrm{MC}}(X)$ relative to the overall shift in cross sections was used to determine the sensitivity of $C_{\mathrm{MC}}(X)$ to changes in the photon cross sections. Since $C_{\mathrm{MC}}(X)$ is a ratio of $D_{\mathrm{TLD}}(X)$ and $K_{\mathrm{air}}(X)$, it is expected that the change in $C_{\mathrm{MC}}(X)$ is less than the overall shift in cross sections. The overall uncertainty of photon cross sections in this energy range is 2%,[39] so this uncertainty and the previous results were used to determine the uncertainty in $C_{\mathrm{MC}}(X)$. The highest uncertainty due to cross sections was 1.5%, for the M20 beam. TG-43U1 provides a more detailed explanation about this type of analysis with respect to dose-rate constant uncertainties.[1]

The uncertainties in the spectra were difficult to quantify, because the low energy photon tails of the M-series x-ray beams would tend to have a large impact on the results but the similarity of the tabulated GSF beams to the UW-ADCL beams in this region was not known. In an attempt to determine the impact of very low energy photons, several simulations were repeated using a photon energy cutoff of 10 keV instead of 1 keV. This primarily affected the calculations for the M20–M50 beams by effectively removing the contribution from x-ray beam photons less than 10 keV. As expected, the most pronounced effect was for the M20 beam, with an 11.5% change in calculated relative TLD dose per unit air kerma. This change decreased to 4.5% for the M30 beam, 1.3% for the M40 beam, and 0.8% for the M50 beam. This uncertainty was assumed to be a rectangular distribution and was represented as a type B uncertainty. The choice of 10 keV as the cutoff energy was rather arbitrary, but it did have the intended effect of increasing the calculated uncertainty for the lowest energy beams. An additional 0.5% uncertainty was added to the analysis for each beam to take into account the uncertainties in photon spectra for energies above 10 keV.

The uncertainty due to variations in the TLD thickness was determined by performing simulations with the M20–M80 beams using a TLD thickness 0.02 mm larger than the

TABLE IV. Example of uncertainty analysis for the MCNP5 calculations, using the M60 beam. Values are expressed in percent.

Parameter	Type A		Type B	
Statistical uncertainty	0.12			
TLD thickness			0.05	
TLD positioning			0.30	
Energy cutoff			0.07	
Photon spectrum			0.50	
Cross sections			0.86	
Quadratic sum	0.12		1.05	
A and B quadratic sum		1.05		
MCNP5 uncertainty			1.05	$\%(k=1)$
			2.11	$\%(k=2)$

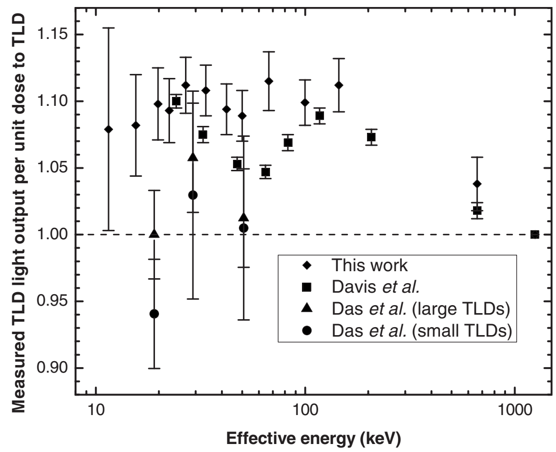

FIG. 5. Comparison of this work with the work of Das *et al.* (Ref. 15) and Davis *et al.* (Ref. 16).

standard thickness. This is consistent with the 2σ variation in measured thickness across 80 TLDs. The calculated difference in $C_{\mathrm{MC}}(X)$ from the standard thickness was 0.7% for the M20 beam and decreased with increasing photon energy. From these simulations the uncertainty was assumed to be a type B Gaussian distribution uncertainty at a coverage of $k=2$. An additional 0.3% uncertainty was added for each of the x-ray beams to account for changes in inter-TLD scatter, since the placement of TLDs in the MC model may not have matched the experiments exactly. A sample of the Monte Carlo uncertainty analysis for the M60 x-ray beam is presented in Table IV. The total combined uncertainties in each of the $C_{\mathrm{MC}}(X)$ values are presented in Table II.

It is possible that the electron contamination from some of the higher energy x-ray beams may have added additional uncertainty in the measurement results. The Kapton® sheet that was used to hold the TLDs in place was only thick enough to provide full CPE conditions for photon energies below about 40 keV, and although the TLD is thick enough to provide full buildup within a very short distance, the possible contamination electrons from the x-ray filter could cause inflated values for the TLD doses. To study this effect, a separate Monte Carlo simulation of the TLD dose was performed with an unfiltered 250 kVp photon source, with photons and electrons transported through the copper/aluminum filter. When electron transport in the air between the filter and the Kapton® sheet was turned off, the depth dose profile in the TLD increased to a maximum at about 75 μm deep, then after the first 75 μm it followed very closely with the kerma profile. When electron transport was turned on between the x-ray filter and the Kapton® sheet, the depth dose profile showed that the TLD dose was 2.4% high in the first 10 μm, but matched the kerma profile past 10 μm. Although the electron spectrum may be different in the first 75 μm than if full buildup conditions were used, we feel that it is unlikely that there was a substantial impact on our results and have not added any additional uncertainty due to this effect.

IV. DISCUSSION

As mentioned in Sec. I, there is no *a priori* reason that the measured TLD light output should be directly proportional to the absorbed dose to the TLD with changing photon energy. The results presented here show that the measured TLD light output was 8%–13% higher for the x-ray beam irradiations relative to ^{60}Co than would be expected based on the Monte Carlo calculations of absorbed dose to TLD. The hypothesis is that there are processes occurring within the TLD at the solid state level that have a dependence on ionization density, which is not taken into account with standard Monte Carlo photon/electron transport codes.

A comparison of the data from this work, Das *et al.*,[15] and Davis *et al.*[16] is shown in Fig. 5. Both this work and Davis *et al.*[15] show the measured TLD light output per unit absorbed dose to TLD is higher than the Monte Carlo-calculated response. In general, the agreement with Davis *et al.*[16] is fairly good, usually within 4%, with a maximum discrepancy of about 6%. The Davis *et al.*[16] data show a "dip" in the response at ~50–60 keV, and although there is a hint that one could be present in the current data (Figs. 4 and 5), it is obscured by the M150 point. The lack of a dip in the current measurements could be that it is obscured in the measurement and calculation uncertainties, but it could also be due to the fact that the current work used x-ray beams with much broader photon energy spectra, which could have the effect of partially "blurring" out this feature. The results in this work appear to be systematically higher than the ones from Davis *et al.*[16] by a few percent (Fig. 5), and the reason for this discrepancy needs to be explored. The Davis *et al.*[16] Monte Carlo simulations used EGSnrc, not MCNP5, but the photon cross section data were very similar between the two codes, and internal benchmark comparisons between the two codes for similar calculations in this energy range have always agreed within 0.5%.

It is possible that some differences in measured TLD response from Davis *et al.*[16] could be attributed to differences in x-ray spectra (narrow spectra versus moderately filtered spectra). However, the results of the H100 and L100 beam comparison suggest that differences in the shape of the photon spectra have only a small effect.

The microdosimetric models by Olko *et al.*[28] suggest that the variation from unity in Fig. 5, seen in this work and by Davis *et al.*,[16] can be explained by differences in the ionization density of electrons generated with the different energy photon beams. The work by Horowitz[29] points to the recombination (i.e., heating) stage as being important for ionization density effects, and these effects are also dependent on the trapping structure in the TLD crystal. For this reason, it is possible that some of the discrepancies between this work and Davis *et al.*[16] could be due to differences in the TLD annealing process, different methods of TLD readout (hot gas versus planchet), or differences in the heating rate and maximum temperature of the TLDs (time temperature profiles). This is only speculation at this point, but if proven true it could be a very important result, since it would make it difficult for TLD measurements at different institutions to have universal validity without similar TLD processing techniques. The importance of listing TLD processing techniques was previously highlighted in a letter to the editor of Physics in Medicine and Biology by Kron *et al.*[47] They recommended a checklist of parameters that should be listed in TLD investigations to facilitate comparisons between different investigators.

The results of this investigation also differ from Das *et al.*[15] The uncertainty in this work is less than that in the Das *et al.*[15] work. Also, Das *et al.*[15] normalized data to 4 MV x rays while this work used ^{60}Co as the reference energy, although there is little expected change in response between ^{60}Co and 4 MV x rays. However, the data of this investigation show that there is a difference between the MC generated TLD response and the measured response as a function of photon energy, whereas Das *et al.*[15] concluded that there was no significant deviation between the measured and MC calculated response. The difference between the MC calculated and the measured response may not have been seen very often in the past because the effect is on the order of magnitude of investigators' uncertainties.[15,25–27] Only by minimizing uncertainties in this study was the effect quantified.

The last important point of discussion is the impact of these results on the dose-rate constants of low energy brachytherapy sources measured with TLDs. The TLD energy correction applied in brachytherapy publications is based on the relative TLD light output per unit dose to water, which was not measured here, so the measurement results from this work cannot be used directly to determine a new energy correction factor. The results from the measured TLD light output per unit dose to TLD as a function of photon energy (Figs. 4 and 5) can be used to predict the difference between the measured energy correction and one calculated using Monte Carlo simulations. Monte Carlo methods for TLDs at 1 cm in water calculate an energy correction in water of about 1.42 for ^{103}Pd or ^{125}I relative to ^{60}Co or 4 or 6 MV photons.[22] The results of this work demonstrate that the actual correction could possibly be 9%−11% higher than that, or in the approximate range of 1.55–1.58. If true, this would have a major impact on measured dose-rate constant results, and in most cases would reduce the generally good agreement that investigators have found between measured and Monte Carlo-calculated dose-rate constants.[1] Williamson and Rivard[13] determined ratios of calculated-to-measured dose rate distributions for ^{103}Pd and ^{125}I seeds across 52 different candidate data sets, and at a radial distance of 1 cm the Monte Carlo calculated results were about 2% less than the measured TLD results on average. If a 9%−11% correction was applied to the measured TLD results, this would mean that the Monte Carlo calculated results would be about 7%−9% higher than the measured TLD results on average. This effect will need to be proven in future work, and if possible it should be measured directly using well characterized ^{125}I and ^{103}Pd sources.

V. CONCLUSIONS

The ratio of measured TLD light output per unit air kerma as a function of photon energy to the MC calculated TLD dose per unit air kerma indicates that the light output of TLD-100 is not directly proportional to the dose to TLD over the range of photon energies investigated. The data indicate that TLD-100 has a measured response that is 8%−13% higher than the MC calculated response in the energy range of 12 keV (effective energy of the M20 beam) to 145 keV (effective energy of the M250 beam) relative to ^{60}Co photons. The data are broadly consistent with the results of Davis *et al.*,[16] although a slightly larger effect was seen here. The implication of this discrepancy is that there may be measurable differences from published TG-43U1 dose-rate constants currently in use. The difference in measured and MC calculated TLD response is likely due to the MC simulations not properly accounting for the solid state physics of the thermoluminescence mechanism. The Monte Carlo simulations do model the self-attenuation of the TLDs, however, which a simple analytical calculation based on a ratio of mass energy-absorption coefficients does not.

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