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EFFECT OF TEFLON TRANSMITTANCE ON SENSITIVITY OF THERMOLUMINESCENCE DOSIMETER CARDS

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Abstract

Thermoluminescence dosimeter cards purchased by the US Navy in recent years have different radiation sensitivities, e.g., they exhibit a different amount of light per dose unit. Presented tests indicate that the optical transparency of the Teflon encapsulation is partially responsible for the significant variation of the DT-702/PD radiation sensitivity. It was confirmed also that the Teflon transparency is in fact a primary cause of the radiation sensitivity increase in the most recently produced dosimetric cards. This conclusion is based on the correlation found between the calibrated radiation sensitivity of the dosimeter card element and the optical transparency of its Teflon encapsulation. The transparency measurements were performed at the wavelength of 400 nm within a 10 nm spectral interval effectively covering the spectral range of the thermoluminescence. It is anticipated that the experimentally determined correlation will help to approve the acceptance of new thermoluminescence dosimeter cards in the Naval Dosimetry Center inventory as well as improve the production process.

Keywords

calibration; dosimetry; personnel; dosimetry; external; dosimetry; thermoluminescence

INTRODUCTION

SINCE 1998, the US Navy monitors personnel radiation doses with Harshaw model 8840/8841 thermoluminescent dosimeters (TLDs), which are read with Harshaw Model 8800PC readers (Cassata et al. 2002; St. John et al. 2006; Romanyukha et al. 2012). The Harshaw 8841 dosimetric card has four lithium fluoride (LiF:Mg,Cu,P) elements which are Teflon encapsulated on an aluminum substrate. The radiation sensitivity of TLDs is one of the most important parameters of dosimeters. Beneficially, more sensitive dosimeters

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can measure lower doses. But significant variation of the radiation sensitivity in the population of dosimeters could mean that there are significant variations of other properties affecting stability of calibration, dose fading, and uncertainty. In the worst-case scenario, dosimeters with different sensitivity may even require different reading conditions or so-called time-temperature profiles for optimal readout and calibration. That is why the Naval Dosimetry Center (NDC) has established acceptable limits for TLD radiation sensitivity. These acceptable limits of radiation sensitivity are verified during acceptance testing of new dosimeters following the requirements established by the International Organization for Standardization (ISO)/International Electrotechnical Commission (IEC) document standard ISO/IEC 17025 (2005).

In recent years, dosimeters have exhibited quite different radiation sensitivities although they seem to be produced in the same way and from the same components. Variations occur typically because of an uncontrolled change in the materials used to produce them. It was noticed in 2015 that newly purchased dosimeters have a significantly higher radiation sensitivity (>20%) than those that were purchased earlier. The radiation sensitivity of a TLD is defined as dosimeter light output per dose unit. It depends on many factors, including the dosimeter's usage. In 2011, Voss et al. (2011) investigated the effect of multiple read-irradiation cycles on TLD card sensitivity. According to this research, 1,000 heat-irradiation cycles causes about 10% sensitivity loss for card elements 1, 2, and 4 if reader is functioning properly. A typical NDC dosimeter issue period is 90 d. When accounting for shipping and processing time this means that on average TLD cards undergo 3–5 heatings per year. Thus, for a 20-y-old card the number of heatings will be below 100. According to Voss et al. (2011), 100 heatings would cause about 1% of sensitivity loss but not the 20% difference that was seen for new and old TLD cards. Furthermore, according to NDC internal policy, every 2 y every TLD card is the subject of new calibration. All historical calibration data are available for review, and they do not indicate a significant change of sensitivity for the cards that are used for personnel monitoring. TLD light output depends not only on the sensitivity of the TLD elements but also on the transparency of the Teflon used for their encapsulation. The TLD manufacturer stated that the Teflon fabrication process was changed because of a termination of some chemicals and reagents previously used in the production process. The purpose of this work was to investigate if a change in Teflon could be responsible for increased sensitivity of TLDs. The basic techniques involve the comparison of Teflon transparency in the TLD cards calibrated to the same dose.

MATERIALS AND METHODS

The TLD card has four elements (Fig. 1). The TLD element is a disc made of powdered LiF crystal mixed with required dopants. The disc is produced by a hot pressure pelleting process. In this study, 51 standard TLD cards were used. Of these, 26 cards were produced before 2005, and another 25 cards were produced in 2015–2016 after the stated change of the Teflon (Chemours, Wilmington, Delaware, US) fabrication. All cards were properly inspected and calibrated according to the NDC-established procedures. As a result of the calibration, the so-called element correction coefficient or *ecc* was assigned to each of four TLD elements:

$$ecc = (RCF \times D)/L,$$

where RCF ($nC \text{ mrem}^{-1}$) is a reader calibration factor, D is the dose delivered to the element during irradiation ($1 \text{ mSv} = 100 \text{ mrem}$), and L (nC) is the response of the element which is given by the light output and measured with the TLD reader (Romanyukha et al. 2018). Note that the delivered dose is shown in the numerator of eqn (1), and the corresponding TLD-light output is in the denominator. Therefore, TLD cards with higher sensitivities will have smaller ecc values. To measure the transparency of the Teflon encapsulation film, an experimental setup (presented in Fig. 2) was used. The light from an optical radiation source was collected by an optical system and directed to a replaceable aperture. It was placed in front of the TLD card opening so that only the Teflon-film-covered area was illuminated. Three apertures were used in this study: the first one had a collimator hole with a diameter of 1 mm (#1 in Fig. 2), the second aperture also had a collimator hole but with a diameter of 2 mm; while the third one was a ring-type (#2 in Fig. 2). The first and third apertures allowed illumination of only the Teflon while excluding the TLD element whereas use of the second aperture allows illumination of the encapsulated TLD element and remaining Teflon. Similar information was obtained using the first and third apertures, but the ring-type aperture provides a stronger signal due to illumination of a larger surface area and more effective light collection. This was important for higher accuracy of the transparency measurements because the source output drops significantly in the range below 400 nm (Fig. 3).

The amount of detected light that passes through the card depends not only on material transmittance but also on broad-angle light scattering from both Teflon and the LiF:Mg,Cu,P materials. Therefore, measured light output varies with distance L^* between TLD card and spectrometer. The accurate measurements of total transmittance become more complicated. In this study, the measurements were focused on the optical transparency T^* defined as the ratio of detected signal (output) to the reference level (input) at the fixed distance L^* as described below. The reference level can be established as 100% of the light supplied to the TLD element area. For this purpose, one element in the designated TLD card is totally removed (Fig. 1), and thus the light reference level is established. All other TLD card elements are measured intact in a highly reproducible optical geometry. The determined reference level is used for normalization of the light output of all intact TLD cards used in this study because of the high reproducibility of the optic measurements. The reproducibility was achieved by use of a specially designed holder shown in Fig. 2. Positioning uncertainty was estimated as 0.1 mm based on the actual dimensions of the holder and TLD card. This uncertainty has smaller effect for the L direction because $L \gg 0.1 \text{ mm}$. Depending on the aperture used, the transparency of only the Teflon film (excluding the TLD element) or the area including the TLD element covered by Teflon were measured.

Optical measurements were performed by a multichannel spectrometer equipped with a holographic grating and a charge-coupled device (CCD). Simultaneously covered spectral range was available between 200 nm and 850 nm. The maximum of the thermoluminescence emission of the LiF:Mg,Cu,P is about 350 nm (McKeever et al. 1995; Yang et al. 2005). Ideally, the transparency must be measured in the whole spectral range of the

thermoluminescence light. However, the source that was used has very low intensity at 350 nm (Fig. 3). The comparison of the different sets of data obtained between 350 nm and 450 nm showed that the range in the vicinity of 400 nm provides nearly the same qualitative information as a whole range between 350 nm and 450 nm but allows improvement of the signal-to-noise (S/N) ratio for the chosen experimental setup. Therefore, most of the data were collected in the 10 nm spectral interval at 400 nm where the light source output was stronger.

RESULTS AND DISCUSSION

Typical results of the transparency T^* measurements are shown in Fig. 4. As expected, the transparency of Teflon only (solid line) is higher because any TLD element has both Teflon and LiF:Mg,Cu,P material contributing to the loss of optical signal. Quantitative analysis of 204 spectra from 51 TLD cards was performed with a specially designed computer code. It first selects the transparency data in the range between 395 nm and 405 nm and then averages them over 20 pixels of the charge-coupled device and presents results as a table of T^* values vs. the element position and the card serial number. As estimated from multiple measurements of individual TLD elements, the expanded uncertainty of the measured transparency T^* of Teflon film was about 2% ($k = 2$).

The correlation between Teflon transparency T^* and ecc of TLD element is shown in Fig. 5. The sequence of the measurements was designed so that a smaller TLD element number on the x axis of Fig. 5 indicates earlier production of the TLD card. For convenience, both sets of data were normalized so that the average data value for the most recently produced cards would be unity. The ecc values measured for 204 calibrated TLD elements presented in Fig. 5 as open circles indicate significant differences depending on the year of the production. The TLD elements produced before 2005 have significantly higher ecc (lower sensitivity) values than TLD elements produced in 2015–2016. The maximum difference found between any one ecc of old and new TLD card elements was a factor of 2. Corresponding transparency measurements of Teflon film (closed squares in Fig. 5) also demonstrate similar but opposite dependence; e.g., the transparency of the Teflon film in the most recently produced TLDs is higher. Significant scatter of the experimental data in Fig. 5 observed for both ecc and transparency T^* is due to variation of the TLD element property and limited accuracy of the measurements.

It is important, however, to notice that the data scatter observed for transparency measurements is a few times lower than that for ecc . So, the accuracy of transparency measurements is sufficient for comparison to ecc . The increase of ecc values and the opposite drop of transparency for the individual TLD elements are shown in the left part of the Fig. 5 displaying the aged TLD cards. A more obvious conclusion becomes available from the weighted fit of the data sets shown in Fig. 5. This fit is presented by dashed and solid lines related to ecc and Teflon transparency, respectively. The observed trend of the curves including the ecc maxima (the lowest sensitivity) and their coincidence with transparency minimums clearly indicates the correlation of Teflon transparency with ecc values.

Similar variability of transparency was found when the TLD element was illuminated in its central area; i.e., the data obtained represented the optical properties of both the LiF:Mg,Cu,P material and Teflon film. These transparency measurements were done using the same experimental setup except the aperture size; e.g., the diameter of circular aperture was increased to about 2 mm to obtain better signal-to-noise ratio. Because the aperture diameter is limited to the central part of the TLD element, the transparency measured in this configuration accounts for the light loss in both Teflon and LiF:Mg,Cu,P material. Significant reduction of the spectral transparency for the whole TLD element as compared to the transparency of Teflon film (as shown in Fig. 4) supports this assumption. The results of transparency measurements of TLD elements are presented in Fig. 6. Upon weighted fit and normalization of the TLD element transparency data in the same way as above, the correlation of *ecc* and transparency T^* was also observed. Indeed, the normalized transparency values of 204 TLD elements presented in Fig. 6 (bottom curve) indicate similar but even stronger correlation with the *ecc* as compared to one found earlier for Teflon film alone.

Assuming that TLD element transparency is a product of those of Teflon and LiF:Mg,Cu,P material, it is possible to retrieve the contribution of the LiF:Mg,Cu,P material only. Presumably its transparency is affected by both scattering and absorption of thermoluminescence emission inside the material. The transparency was evaluated by dividing the TLD element transparency T^* (solid line in Fig. 6) by that of Teflon only. From these data, it was found that the LiF:Mg,Cu,P material also demonstrates the dependence on the year of element production. The data presented in Fig. 7 indicate up to 15% additional reduction of transparency in TLD cards produced before 2005.

Presented optical measurements of both Teflon and LiF:Mg,Cu,P elements indicate some correlation of *ecc* with transparency of the TLD element. The relative variation of the Teflon transparency was found to be a few times higher as compared to that of the LiF:Mg,Cu,P element. Thus, the improved transparency of Teflon encapsulation material can be considered a primary reason for increasing sensitivity of the most recent TLD cards. The evaluation of the physical properties of Teflon responsible for its transparency is beyond the scope of this paper and may be found elsewhere (White et al. 2016; Lin et al. 2012; Tsai et al. 2008; Galante et al. 2010). There is an indication that light scattering in Teflon film significantly contributes to transparency of the TLD element. Related measurements have established the reduction of transparency with distance L^* to spectrometer input (Fig. 2). Because in all the transparency measurements the source beam was collimated, the dependence found suggests the presence of wide-angle radiation, presumably diffuse light-scattering coming from Teflon film. Light scattering observed in Teflon film is contributing to TLD card transparency and its TL sensitivity. The presence of diffuse scattering in Teflon encapsulation can be observed even visually; e.g., Teflon film looks noticeably milky in the cards produced before 2005 compared to the cards produced in 2015–2016.

CONCLUSION

We developed a method for optical transparency measurements of Teflon-encapsulated TLD elements. The technique allows light output-input ratio measurements for specified areas of

the TLD cards. The light illumination was done by a preconfigured beam. The transparency measurements were available in the spectral range from 200 nm to 850 nm which effectively covers the entire spectrum of the LiF:Mg,Cu,P thermoluminescence emission. The present method provides about 2% ($k = 2$) uncertainty for transparency value measured at the 400 nm wavelength.

Based on the results obtained for 204 calibrated TLD elements, the direct correlation of radiation sensitivity of TLD cards and their Teflon encapsulation transparency has been established. It was found that Teflon transparency is a primary cause of the radiation sensitivity increase in the most recently produced dosimetric cards. Practically this means that transparency of Teflon encapsulation holding the LiF:Mg,Cu,P element in the TLD card may contribute to the value of *ecc* of the TLD element calibrated to the same radiation dose. Variations of both the *ecc* and optical transparency are stronger in the aged TLD cards. Additionally, the transparency of the LiF:Mg,Cu,P material itself may affect the *ecc* although the appearance is different from the related features in Teflon. This effect is also stronger in the aged TLD cards. As follows from this study that a Teflon transparency control, if established at the production site, would significantly improve the *ecc* uniformity for TLD cards.

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Fig. 1.
TLD card. The upper left TLD element was entirely removed for the reference measurements.

Fig. 2.
Experimental setup for transparency measurements.

Fig. 3.
Spectrum of the source radiation.

Fig. 4.
Typical spectral dependence of transparency T^* for Teflon encapsulation film (solid line) and for a whole TLD element (dashed line).

Fig. 5.
Comparison of calibration coefficient ecc (open circles, dashed line) and Teflon transparency T^* (solid squares and solid line). Presented lines use the weighted (10%) fit of the data.

Fig. 6.

Comparison of calibration coefficient ecc (open circles, dashed line), transparency T^* for Teflon film (solid squares and solid line), and a whole TLD element (triangles and dotted line). All lines present the weighted (10%) fit of the data.

Fig. 7.
Relative transparency of the LiF:Mg,Cu,P element.