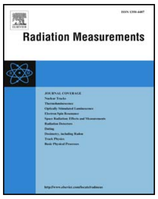
Radiation Measurements 108 (2018) 20–25

Contents lists available at ScienceDirect

Radiation Measurements

journal homepage: www.elsevier.com/locate/radmeas



Fading time and irradiation dose estimation from thermoluminescent dosemeters using glow curve deconvolution

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ARTICLE INFO

Keywords:
TL-DOS
Thermoluminescence
Glow curve deconvolution
Fading time estimation
Irradiation dose estimation

ABSTRACT

Thermoluminescence dosemeters are commonly used in various fields of dosimetry. During the elapsed time between irradiation and readout, dosemeters are affected by thermal fading. As a result the measured signal is reduced resulting in an underestimation of the irradiation dose. This is a challenging issue, especially in personal dosimetry. Different techniques have been developed in the past to overcome the influence of fading. These techniques often come along with a loss of information which reduces the accuracy of the irradiation dose estimation. In this work, a method is developed which is based on glow curve deconvolution and which results in an irradiation dose estimation with no fading time dependence. The method also gives an estimation of the fading time.

1. Introduction

Thermoluminescence dosemeters (TLDs) are commonly used in different fields of application such as environmental or personal dosimetry. An estimate of the irradiation dose is the amount of thermoluminescence (TL) light emitted by the dosemeter during heating. The loss of TL signal with elapsed time between irradiation and readout is a well known characteristic of TLDs and is referred to as fading or post-irradiation annealing. Refs. (Horowitz, 1990; Harvey et al., 2010) give an overview on the subject of fading for different TL materials.

For individual dose monitoring the effect of fading is a challenging issue as the measurement of a reduced signal results in an underestimation of the irradiation dose. If the fading characteristics of the used TL material as well as the fading time are known, the number of measured photons and with this the calculated irradiation dose can be corrected for this effect (Doremus and Higgins, 1994; Furetta et al., 1999). In most cases, however, the irradiation date is unknown and thus techniques have been developed to overcome the effect of fading. Most approaches restrict the irradiation dose estimation to high-temperature peaks of the measured glow curves with larger half-lifes. One possible realization of this approach is to apply a pre-heating procedure before the actual measurement, described, e.g., in Refs. (Walbersloh and Busch, 2015; Lee et al., 2015). During this procedure the low-temperature peaks with shorter half-lifes are erased so that the remaining signal is independent of the elapsed time between irradiation and readout. Other possibilities are to read out the whole glow curve and to either identify the high temperature peaks with a glow curve deconvolution (GCD) or to set a suitable region of interest as described, e.g., in Ref. (Weinsteinet al, 2003).

A disadvantage of these methods is the reduction of the overall signal strength. Furthermore, the pre-heating technique erases parts of the glow curve and thus removes potential information about the nature of the irradiation. Therefore, several efforts have been undertaken in the past to calculate the fading time from the glow curve itself, see Refs. (Moscovitch, 1986; Furetta and Azorín, 1989; Budzanowskiet al, 1999).

In this paper, we present a method with which the irradiation dose can be estimated independently of the fading time and which overcomes the problem of information loss. It is based on a fit of the glow curve with a glow peak model, so that the individual glow peaks can be separated, which is possible if the number of single components as well as their approximately temperatures are known. The respective signal strengths are used to construct an estimator for the fading time, which in turn is used to correct the number of measured photons and with this the calculated irradiation dose.

This paper is structured as follows: In Sections 2–6 the materials and methods are presented. The fading time estimation and the fading time independent irradiation dose estimation are discussed in Section 7 and 8. Section 9 concludes the paper.

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https://doi.org/10.1016/j.radmeas.2017.11.002
Received 2 August 2017; Received in revised form 29 September 2017; Accepted 9 November 2017

2. The TL-DOS system

The dosemeters used in this study belong to the TL-DOS system which was developed at the individual monitoring service of the Materialprüfungsamt Nordrhein-Westfalen (MPA NRW) in Dortmund, Germany. The sensitive material used for the dosemeters is a thin layer of LiF:Mg,Ti which is hot-sintered on an aluminum substrate plate mounted inside an aluminum code ring for protection and identification. The dosemeters are produced and tested at the MPA NRW. The investigations presented in this paper are done in the context of the MPA's personal routine dosimetry service, for which approximately 1,10,000 film dosemeters are currently evaluated per month. An important requirement for the usage of TL dosemeters in such a service is a short heating and readout time.

During the readout process, the dosemeters are heated to 573K using a constant-temperature heating cartridge resulting in exponential heating. This leads to a full glow curve in about 10 s measured with a photomultiplier tube and recorded with a sampling frequency of 10 ms. The measurement chamber is flushed with nitrogen during the readout to limit the high-temperature background.

The setup does not allow for the simultaneous measurement of the temperature of the dosemeters during the readout procedure. Since each dosemeter has a slightly different heat transfer due to contact heating and fabrication tolerances, a technique was developed to convert each glow curve from the time into the temperature domain based on an individual heating function. An investigation with a temperature-sensitive setup shows that this conversion leads to a deviation between the estimated and the real temperature of the dosemeter of less than 1% in the region where the glow peaks occur. See Ref. (Theinert et al,) for more details of this technique.

For more details on the dosemeters and the complete readout system see Refs. (Walbersloh and Busch, 2015; Theinert et al,).

3. Measurements and data sets

For the fading studies and the batch calibration presented in this paper, a total of 800 newly produced dosemeters are used. Furthermore, a set of 22 dosemeters is used as a reference sample, as recommended in Ref. (Horowitz, 1990). In comparison to the newly produced dosemeters for the fading studies the dosemeters of the reference sample are already used many times and have a well known response. All dosemeters are annealed at a temperature of 673 K and then cooled down to room temperature, each within 15 s. The dosemeters are each irradiated once with a^{137}Cs gamma source within 2 h after their annealing and then stored in an isolated box to guarantee constant temperature and humidity conditions for all detectors. The lightproof box avoids any optical annealing or stimulation. The dosemeters are annealed and irradiated at various times (depending on the fading or calibration studies) and then all detectors are read out successively in a short time interval. All irradiation doses investigated in this paper are given in units of $H_p(10)$.

For the fading studies, a total of 1600 measurements are performed. The dosemeters are grouped into batches of 40 pieces. For each fading time, four sets of detectors are irradiated with 0.5 mSv, 1 mSv, 5 mSv, and 10 mSv, respectively, and then stored for 30 min, 4 h, 1 d, 2 d, 4 d, 10 d, 16 d, 22 d, 32 d and 41 d before they are read out.

For calibration purposes, a total of 450 measurements are performed. The dosemeters are grouped into batches of 50 and are irradiated with nine different irradiation doses ranging from 0.05 mSv to 15 mSv. The fading time of all calibration measurements lies between 25 min and 50 min, and the pre-irradiation fading time is 1 h at most.

For the reference studies, a total of 700 measurements is performed. The reference dosemeters are each irradiated with 5 mSv. They are read out between the different sets of fading and calibration measurements to investigate possible sensitivity changes in the TL reader.

4. Glow curve model and deconvolution

The glow curve of LiF:Mg,Ti dosemeters shows five significant glow peaks for temperatures up to 573 K, denoted P_1 to P_5, and each has a different half-life ranging from a few minutes up to several years, see e.g. Ref. (Harvey et al., 2010) and references therein. Glow peak P_1 is decreased completely in most practical applications due to its short half-life. The glow curve model used in this paper is based on a superposition of the four glow peaks P_2 to P_5. Each single glow peak can be described with Equation (1) given by Randall-Wilkins (Randall and Wilkins, 1945),

$$I(T) = s \cdot n_0 \cdot e^{-\frac{E}{kT}} \cdot \exp\left(-\frac{s}{\beta}\int_{T_0}^{T} e^{-\frac{E}{kT'}} dT'\right), \tag{1}$$

where I is the intensity of a single thermoluminescence glow peak and E is its activation energy, s is the so-called frequency factor, n_0 is the initial concentration of trapped carriers and k is the Boltzmann constant. T is the absolute temperature, T_0 is the initial temperature and β describes the heating rate.

Equation (1) is based on the assumption of linear heating. For the exponential heating used here, the heating rate β can be described as

$$\beta = \frac{dT}{dt} = \alpha(T_g - T), \tag{2}$$

where α is the exponential heating factor and T_g is the temperature of the constant-temperature heating cartridge. Equation (1) then becomes

$$I(T) = s \cdot n_0 \cdot e^{-\frac{E}{kT}} \cdot \exp\left(-\frac{s}{\alpha}\int_{T_0}^{T} \frac{e^{-\frac{E}{kT'}}}{T_g - T'} dT'\right). \tag{3}$$

To guarantee an efficient glow curve deconvolution, Equation (3) is transformed, based on the calculations presented in Ref. (Kitiset al, 2006), from $I(T, n_0, s, E)$ to $I(T, I_m, T_m, E)$ with the parameters I_m as the maximal intensity of the glow peak and T_m as the temperature at the glow peak maximum. Thus, the shape of a single glow peak is characterized by three parameters, namely I_m, T_m and E.

In addition to the signal from the four glow peaks, the measured glow curve also comprises of a background induced, among other contributions, by black body radiation of the heater and the detector, see Ref. (van Dijk and Busscher, 2002) for a discussion. Empirical investigations show the background of the TL-DOS system, I_{bg}, can be parameterized as

$$I_{bg}(T) = a + b \cdot T + c \cdot e^{d \cdot T}. \tag{4}$$

The equation is inspired by the model of Ref. (Kitis et al., 2012), although a linear term is added to improve the agreement with the data. A contribution of high-temperature glow peaks ($\geq$ 573 K) from previous irradiation is not considered because of the high annealing temperature of 673 K.

For the GCD, all glow curve fits are performed with a custom-made software package written in Python using the open-source package scipy (Jones et al., 2001). A pre-fit, based on a simplified model introduced in Ref. (Kitis et al., 1998), is performed to stabilize the fit and to estimate start-values for I_m, T_m and E. The pre-fit itself uses the peak temperatures measured with the temperature-sensitive setup as initial values.

As an example for a fit to the data, Fig. 1 shows a measured glow curve and its GCD. No significant deviation between the data and the fitted approximation is observed.

Only for less than 1.4% of more than 2700 the fit algorithm could not find the optimal parameters and fail or the result of the fit have a high reduced χ^2 (greater than 10). Those glow curves are not considered in the following studies.