Radiation Physics and Chemistry 78 (2009) 465–467

Contents lists available at ScienceDirect

Radiation Physics and Chemistry

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

A study of the alanine dosimeter irradiation temperature coefficient from 25 to 80 °C

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ARTICLE INFO

Keywords:
Alanine
Dosimeter
Dosimetry
Electron paramagnetic resonance
Gamma ray
Ionizing radiation
Temperature coefficient

ABSTRACT

The response of high-dose-range chemical dosimeters is dependent on the dosimeter temperature during irradiation. Typically, irradiation temperatures are estimated by measurements, calculations, or some combination of the two. Then using the temperature coefficient for the dosimetry system, the dosimeter response is adjusted or corrected to be consistent with the irradiation temperature for the calibration curve. Consequently, the estimation of irradiation temperature and the response correction via the temperature coefficient are sources of uncertainty in industrial dosimetry. To date, studies of dosimetry system performance at high temperatures have been limited. The maximum irradiation temperature for temperature coefficient studies of commercial alanine dosimeter formulations has not exceeded 50 °C. However, high-energy electron-beam processing can expose dosimeters to temperatures as high as 70 °C. This study aims to examine the temperature coefficient above 50 °C and assess the accuracy of the dosimeter response corrections. The findings reveal small but significant deviations from linearity above 70 °C. The magnitude of this deviation and its implications to dosimetry measurements will be discussed.

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1. Introduction

The influence of irradiation temperature on the response of a dosimeter is an important quantity to assess for radiation metrology. High-dose irradiation treatment will cause a significant rise in the dosimeter temperature. The temperature coefficient, or relative response change per degree, is determined for each dosimetry system so that a dosimeter's response can be adjusted to the irradiation temperature employed for the calibration of the system. Though the magnitude of the temperature coefficient is typically not large (approximately tenths of percent per degree), the lack of a suitable device to accurately measure the irradiation temperature significantly increases the contribution of the temperature correction step to the system measurement uncertainty.

The temperature dependence studies on the alanine dosimetry system can be separated into two eras. The pre-commercial era, summarized by Nagy et al. (2000), studied the influence of irradiation temperature on custom-designed dosimeters manufactured by individual researchers. The work of Nagy et al. (2000) was the first to characterize the temperature coefficient of a mass-produced commercial alanine dosimeter. The dosimeter was a cylindrical alanine pellet manufactured by Bruker BioSpin Corporation (this dosimeter is no longer manufactured or marketed).[1] For this dosimeter type, Nagy et al. (2000) measured a temperature coefficient that varied (with absorbed dose) from 0.14%/K to 0.17%/K. One of the unresolved questions from that work was the large difference in temperature coefficients for the two stereoisomers of alanine, L-α-alanine and DL-α-alanine. Pure L-α-alanine is composed of a single isomer, and DL-α-alanine is a mixture of the L and D stereoisomers. Several years later, Desrosiers et al. (2006) found that the temperature coefficient for the dosimeters prepared with DL-alanine is more than 50% greater than those prepared with L-alanine. For this reason, L-alanine dosimeters are preferred for measurement applications where the irradiation temperatures differ greatly from the calibration irradiation temperature, specifically in regard to minimizing the measurement uncertainty.

The dosimeters currently used for the NIST transfer dosimetry system are distributed by Far West Technology (FWT). To date, the temperature coefficient for this dosimeter remains unpublished. In addition, the performance of this or any commercial dosimeter at irradiation temperatures above 50 °C has yet to be examined.

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[1] Certain commercial equipment, instruments, or materials are identified in this paper to foster understanding. Such identification does not imply recommendation or endorsement by the National Institute of Standards and Technology, nor does it imply that the materials or equipment identified are necessarily the best available for the purpose.

0969-806X/$ - see front matter Published by Elsevier Ltd.
doi:10.1016/j.radphyschem.2009.03.026

This study characterizes the temperature coefficient of the FWT alanine pellet dosimeter up to 80 °C.

2. Experimental

The absorbed doses for this study were delivered by a Gammacell 220 ^{60}Co irradiator (serial number 207; MDS Nordion, Canada) with an activity of 630 TBq (17 kCi,) as of April, 2007. Alanine dosimeters were irradiated in the central uniform-dose position of each source. The calibration method for determining the irradiation geometry dose rate is described in NIST SP250-45 (Humphreys et al., 1998) with the exception of a modification to the calibration scheme described in Desrosiers et al. (2008).

The dosimeters were measured within 24–48 h after irradiation by electron paramagnetic resonance (EPR) spectrometry. A Bruker Biospin ECS106 EPR spectrometer was configured to specifically measure alanine pellet dosimeters (Humphreys et al., 1998). The EPR amplitude is defined here as a ratio of the alanine EPR amplitude of the center resonance and the ruby reference material EPR amplitude (as described in Nagy et al., 2000). The EPR amplitude is normalized by dividing the ratio value by the mass of the individual dosimeter. The resultant value is referred to as the dosimeter response. The alanine EPR recording parameters common to all measurements in this study were: frequency, 9.684 GHz; center field, 345.5 mT; magnetic-field sweep width, 1.0 mT; modulation amplitude, 0.285 mT; time constant, 1.3 s; and sweep time, 21 s.

The specially designed aluminum holder used previously (Nagy et al., 2000) to achieve thermal equilibrium was modified. Tests of that previous design (dosimeters oriented in separate holes of a horizontal plane aluminum disc) when compared to an aluminum sleeve that holds dosimeters in a single vertical stack, demonstrated a higher precision in favor of the sleeve geometry. Six alanine pellets were irradiated at each dose-temperature combination. To pre-equilibrate the dosimeters to a specific temperature, the dosimeters were placed in the aluminum cylinder irradiation assembly and thermally equilibrated for at least 1 h prior to placing them in the Gammacell. Temperature during the irradiation was controlled to ±1 °C by using a high-flow air shower from a TurboJet (FTS Systems) and monitored with a type-T thermocouple. At the conclusion of the irradiation the dosimeters were immediately transferred to the room-temperature environment.

3. Results and discussion

Two absorbed doses were chosen for this study based on previous findings (Nagy et al., 2000) that cited a dependence of the temperature coefficient on absorbed dose. Absorbed doses of 1 and 20 kGy were used as they are representative of a high and low value of the temperature coefficient. For each dose studied, nine groups of pellets (six pellets in each group) were irradiated at an irradiation temperature of (20, 30, 40, 50, 55, 60, 65, 70, 75, 80) °C for the 1 kGy absorbed dose level, and (20, 30, 50, 55, 60, 65, 70, 75, 80) °C for the 20 kGy absorbed dose level. The dosimeter response data for each absorbed dose level was subjected to a linear least-squares regression. A response value at 25 °C was determined from the computed function. The predicted response at 25 °C was used as a reference value used to convert the original response data at each of the irradiation temperatures to relative response values. Relative response at 1 and 20 kGy was plotted versus irradiation temperature and once again subjected to a linear least-squares regression (Fig. 1). For reasons described below, the regression was applied to data measured for irradiation temperatures up to, and including, 70 °C. Measurements made at 75 and 80 °C were not included in the regression, but are shown in Fig. 1 for comparison. The slope of this function is the temperature coefficient that is expressed in the units of %/K.

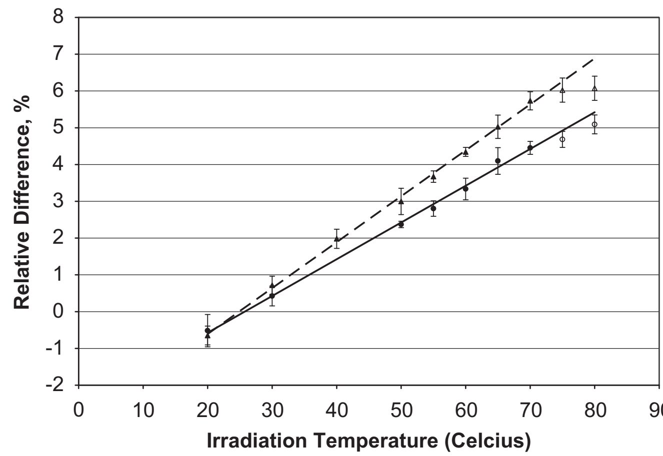

Fig. 1. The difference in alanine dosimeter response relative to the response at 25 °C as a function of the irradiation temperature for 1 kGy (dashed) and 20 kGy (solid) irradiated dosimeters. The open symbols were not used in the linear regression of the data; the function was extrapolated to 80 °C for the purpose of comparison. The error bars represent the dosimeter response standard deviation (1 sigma) for each group of irradiated dosimeters.

The temperature coefficient for the 1 kGy dose level is 0.12%/K (relative uncertainty of 2.4%, $k = 1$) and the temperature coefficient for the 20 kGy dose level is 0.10%/K (relative uncertainty of 1.7%, $k = 1$). Also, as previously observed the temperature coefficient for 20 kGy is less than for 1 kGy and the relative difference is comparable to the previous findings for other alanine dosimeters (Nagy et al., 2000). With these data and that of Nagy et al. (2000), the depression in the temperature coefficient at 20 kGy has now been observed in three different alanine dosimeter types. A common factor in the composition and manufacture of the three systems is the use of L-alanine as the active ingredient. For these dosimeters, the polymer binders, manufacturing methods, and alanine crystal sizes all differ from each other. It is reasonable to assume that the change in temperature coefficient with absorbed dose is an attribute of the L-alanine dosimeter. However, the temperature coefficient difference with absorbed dose is small relative to other sources of uncertainty in irradiation processing and, in practice, the mean of these values, 0.11%/K (relative uncertainty of 2.9%, $k = 1$), is used for all high- and low-dose levels.

The dosimeter relative response plotted versus the irradiation temperature deviates from linearity at 75 and 80 °C for both 1 and 20 kGy (Fig. 1). For an absorbed dose of 1 kGy the measured value at 75 °C is approximately 4% lower than that predicted (by the given linear function) and the measured value at 80 °C is approximately 12% lower. For an absorbed dose of 20 kGy the measured value is approximately 5% and 6% lower than that predicted for 75 and 80 °C, respectively.

4. Conclusions

These data determined that the temperature coefficient of 0.11%/K for the FWT alanine dosimetry system can be used up to 70 °C. Corrections applied to dosimeter responses irradiated above this temperature will depend on the absorbed dose and magnitude of the irradiation temperature. How best to adjust measurements to compensate for irradiation temperatures in excess of 70 °C will depend on the specific process, the uncertainty associated with the irradiation temperature measurement, and the overall uncertainty of the system. These data offer guidance for performing measurement corrections and assessing uncertainties for the FWT alanine dosimetry system at the upper extremes of irradiation temperatures encountered in industrial irradiation processing.

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