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A STUDY OF THE IRRADIATION TEMPERATURE COEFFICIENT FOR L-ALANINE AND DL-ALANINE DOSEMETERS

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Alanine dosimetry is now well established both as a reference and routine dosemeter for industrial irradiation processing. Accurate dosimetry under the relatively harsh conditions of industrial processing requires a characterisation of the parameters that influence the dosemeter response. The temperature of the dosemeter during irradiation is a difficult quantity to measure so that the accuracy of the temperature coefficient that governs the dosemeter response becomes a critical factor. Numerous publications have reported temperature coefficients for several types of alanine dosemeters. The observed differences in the measured values were commonly attributed to the differences in the polymer binder or the experimental design of the measurement. However, the data demonstrated a consistent difference in the temperature coefficients between L-alanine and DL-alanine. Since there were no commonalities in the dosemeter composition or the measurement methods applied, a clear conclusion is not possible. To resolve this issue, the two isomeric forms of alanine dosemeters were prepared and irradiated in an identical manner. The results indicated that the DL-alanine temperature coefficient is more than 50% higher than the L-alanine temperature coefficient.

INTRODUCTION

A correction for the average temperature experienced by a dosemeter during irradiation with electrons and photons improves the accuracy of the dose measurement. The relationship between the dosemeter's radiation response to the absorbed dose and its temperature during irradiation is termed the irradiation temperature coefficient. This temperature coefficient is typically expressed in percentage change per degree. The temperature rise in dosemeters irradiated with high-intensity ionising radiation sources can be appreciable; however, the temperature during irradiation is often difficult or impractical to be measured directly. In the absence of a direct measurement, an estimation of the irradiation temperature is often employed to make this correction for the computation of the absorbed dose. Since this estimate includes unavoidable significant errors, the magnitude of the temperature coefficient is the next consideration in any efforts to minimise the measurement uncertainty.

In 2000, a compilation of all the published temperature coefficients was tabulated.(1) The observed differences were attributed to several factors that include the polymer binder type and concentration; manufacturing parameters; experimental design of the temperature-controlled irradiations; and computations from limited data. One influence not explored was the isomeric composition of alanine. NIST's high-precision temperature-controlled irradiation apparatus and the dosemeter manufacturing technology of the China Institute of Atomic Energy (CIAE) was joined in a collaborative effort to examine dosemeters prepared with L-α-alanine, composed solely of the L isomer, and DL-α-alanine, a mixture of the L and D isomers.

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MATERIALS AND METHODS

The preparation of alanine dosemeters was carried out at CIAE. (The mention of commercial products throughout this paper does not imply recommendation or endorsement by the National Institute of Standards and Technology, nor does it imply that products identified are necessarily the best available for this purpose.) Two different dosemeter types were prepared identically from uniform mixtures of either L-alanine (Beijing Chemical Reagent Company, BR) or DL-alanine (Beijing Chemical Reagent Company, BR) with pure paraffin (melting point range from 54 to 56°C; Shanghai Huashen, BR). Each dosemeter had a mass of ~60 mg with 95% alanine and 5% paraffin by weight. The protocol for manufacturing alanine dosemeters can be summarised as follows:

- Grinding the polycrystalline alanine.
- Sieving the ground alanine to select a particle size range from 50 to 125 μm.
- Cutting the paraffin to small diameter granules.
- Uniform mixing of the alanine and the paraffin through a two-step procedure of grinding and heating (61°C) several times.
- Pressing the mixture into a mold to form cylindrical pellets of 4.8 mm in diameter and 3 mm in height.

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Irradiations were conducted in a Gammacell 220 (Nordion, Canada) ^{60}Co gamma source (dose rate ≈ 15 kGy h^{-1}). The dosemeters were placed in a custom-designed aluminium holder surrounded by a controlled-temperature airflow that was capable of achieving thermal equilibrium from −80 to +100°C during irradiation[1]. The holder accommodated six dosemeters, in which three dosemeters of each type were spaced equally in the holder and irradiated simultaneously to 10 kGy. The dosemeters in the holder assembly were pre-equilibrated to the target temperature prior to each irradiation. The irradiation temperature was held at the target temperature (within ±1°C) throughout the irradiation period.

The electron paramagnetic resonance (EPR) signal measurement protocol for the Bruker ECS106 spectrometer measurements was described in detail previously[1]. Essentially, each dosemeter was measured at two angles and these measurements were normalised to the dosemeter mass and the EPR signal amplitude of the spectrometer's internal reference material (ruby crystal). The average of the normalised signal amplitudes for both angles was used as the dosemeter response.

The response for the dosemeters irradiated from −10 to +50°C underwent linear regression and the resultant function was used to compute the predicted EPR response at 25°C. This value served as the reference point from which the relative response for each measurement was calculated. The value for the slope of the relative response plotted versus the irradiation temperature is the temperature coefficient.

RESULTS AND DISCUSSION

A graphical display of the published temperature coefficients plotted against the absorbed dose at which they were measured is shown in Figure 1. The data were taken from Nagy[1] and references therein, with the exception of one data point (0.11% per K at 50 kGy)[2]. The temperature coefficients selected for this graph were limited to the data that identified the isomeric form of the alanine and to the measurements made in a temperature range that approximated the range used in this study (−10 to +50°C). The L-alanine response with temperature was shown to be nonlinear below −10°C[2].

There is a distinct difference between the DL-alanine and the L-alanine temperature coefficients. The mean DL-alanine temperature coefficient, 0.24% per K is 57% higher than the mean L-alanine temperature coefficient, 0.15% per K. Despite this obvious difference, it has never been conclusively determined if this difference can be attributable to the isomeric form of the alanine. Potential influences from dosemeter composition and/or dimensions along with the experimental design of the measurements were too dissimilar among the studies. Qualitatively, the data in Figure 1 suggest that the

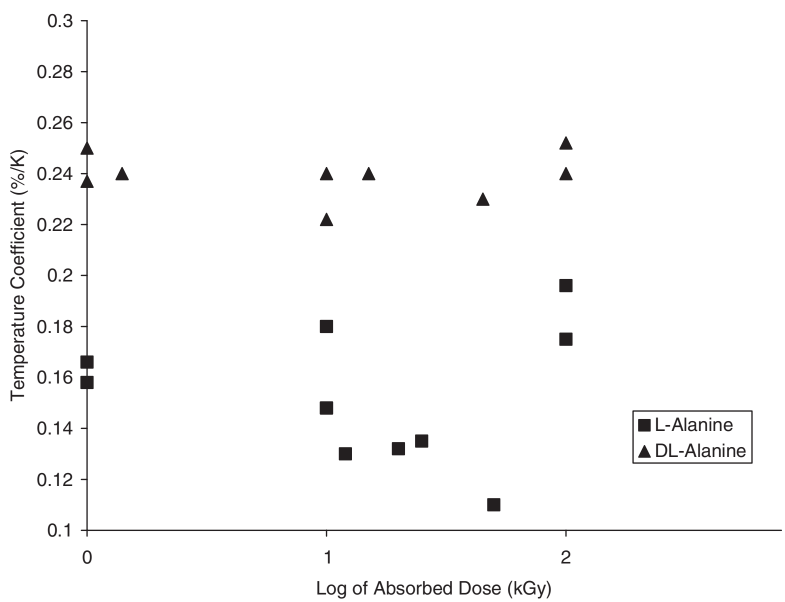

Figure 1. A graph of published temperature coefficients for DL-alanine and L-alanine dosemeters vs. the absorbed dose in kGy.

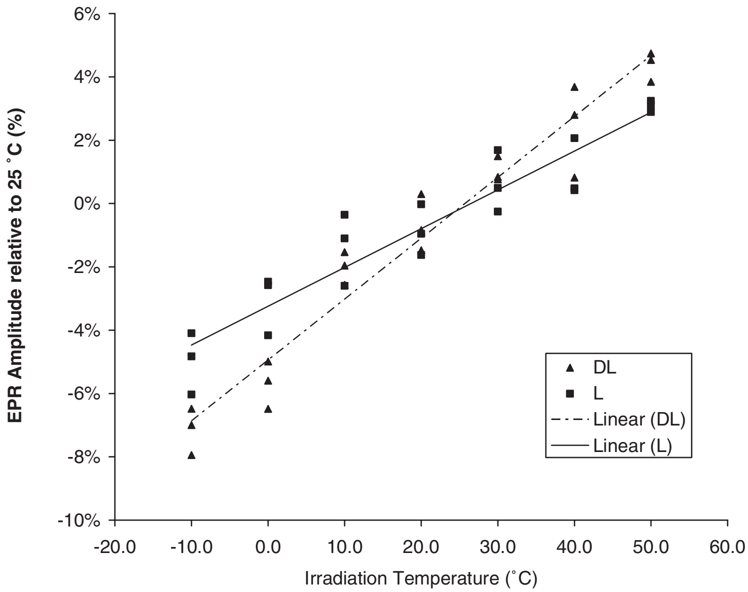

Figure 2. The percentage difference in EPR response, relative to the predicted response at 25°C, for DL- alanine and L-alanine dosemeters vs. the irradiation temperature in °C.

magnitude of these effects is relatively small. The standard deviation of the DL-alanine temperature coefficients is ~4%. These measurements were made on dosemeters of similar composition and dimensions from three different research groups. The standard deviation of the L-alanine temperature coefficients is 16%; these data were compiled from five dosemeter types, four different research groups, and a broad range of dosemeter dimensions (from films to 1 cm pellets).

As described above in Materials and Methods section, the two dosemeter types were collocated and irradiated simultaneously from −10 to +50°C. The percentage difference in response at a specific temperature relative to the predicted response at 25°C is plotted in Figure 2. The slope of these data for each dosemeter type is the temperature coefficient. From these data, the DL-alanine temperature coefficient was determined to be 0.19% per K while that of the L-alanine was determined to be 0.12% per K. Both these coefficients are 20% lower than the mean coefficients for the respective isomeric alanine dosemeter forms extracted from Figure 1. This difference may be attributable to the experimental design of the measurement system. Interestingly, the DL-alanine temperature coefficient is once again 57% higher than the L-alanine temperature coefficient.

As mentioned above, the L-alanine response with temperature was shown to be nonlinear below −10°C[2]. To determine if this effect can be observed in DL-alanine, two temperatures below −10°C were selected (−30 and −77°C). It was found that the DL-alanine dosemeter response deviated from linearity below −10°C in a manner analogous to the L-alanine dosemeters. The magnitude of the deviation was consistent with the previous study on low-temperature effects on the alanine dosemeter response.

CONCLUSION

These data present conclusive evidence that the temperature coefficient for the dosemeters prepared with DL-alanine is more than 50% higher than those prepared with L-alanine. Therefore, L-alanine dosemeters are preferred for measurement applications where the irradiation temperatures differ greatly from the calibration irradiation temperature, especially if minimising the measurement uncertainty is a concern.

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