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A study of the alanine dosimeter irradiation temperature coefficient in the -77°C to $+50^\circ$C range

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Abstract

Early researchers in alanine dosimetry established that, at least in the 0–50°C temperature range, the amplitude of the electron paramagnetic resonance signal of irradiated alanine grows linearly with irradiation temperature. The irradiation temperature coefficient is derived from the slope of this response–irradiation temperature relationship. However, very little data exists on the linearity of the response below 0°C. Thus, the applicability of the irradiation temperature coefficient determined above 0°C to irradiations conducted below 0°C is uncertain. This work investigates the behavior of the alanine response irradiated using a Cobalt-60 gamma source over the temperature range -77°C to $+50^\circ$C. Since the temperature coefficient is known to be dose dependent [Radiat. Phys. Chem. 57 (2000) 1], a series of dose response studies were conducted over a dose range of 0.5–100 kGy. The study revealed that the temperature response deviates from linearity below -10°C. The implications of these observations to dosimetry will be discussed along with possible chemical mechanisms that would account for these observations.
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Keywords: Alanine; Dosimeters; Dosimetry; Irradiation; Radiation processing; Temperature; Temperature coefficient

1. Introduction

The temperature rise in materials treated with high-intensity industrial ionizing radiation sources is an important influence factor on the dosimeters used to model or monitor the irradiation process. For alanine dosimeters, the temperature during industrial irradiation processing affects the radical production that is reflected in the measured response of the dosimeter. The relationship between the dosimeter's radiation response to absorbed dose and its temperature during irradiation is termed its irradiation temperature coefficient. This temperature coefficient is typically expressed in percent change per degree, and for L-alanine is slightly above[1] $+0.1\%/^\circ$C for temperatures above zero Celsius (Nagy et al., 2000).

In many industrial applications, temperature control during irradiation is unnecessary. Processing typically occurs at ambient temperatures, and dosimeters are characterized under the conditions of use. Consequently, there has been little motivation to characterize the dosimeter's temperature response characteristics in the negative temperature range.

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[1] The exact value is dependent on the formulation of the alanine dosimeter.

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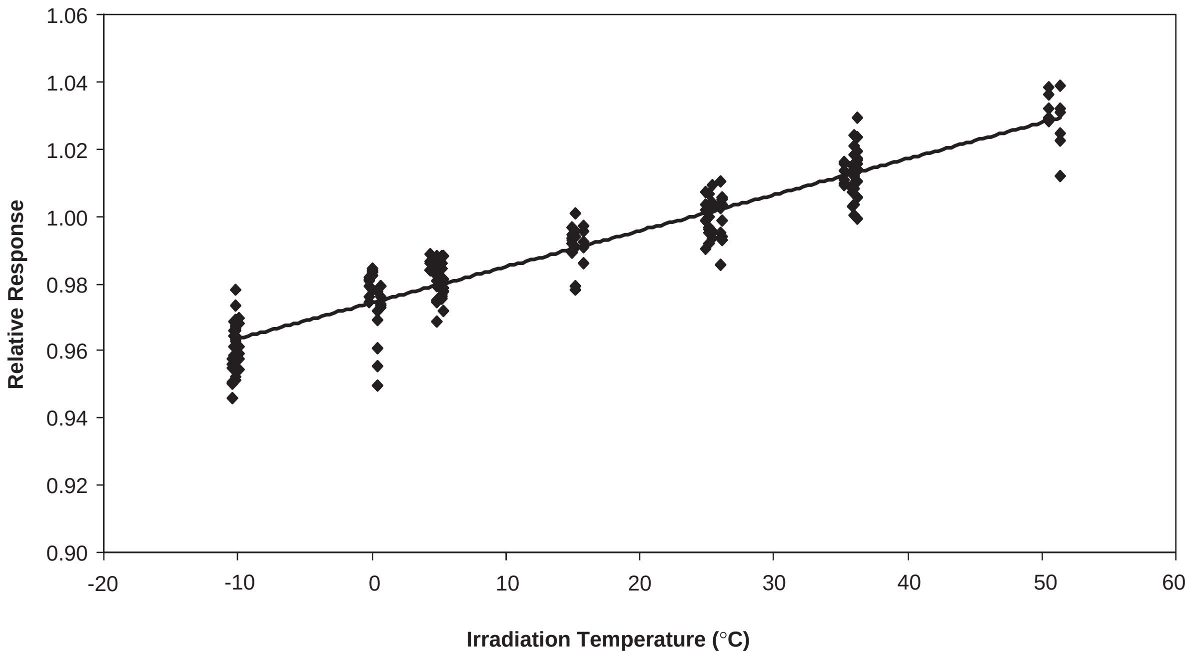

Fig. 1. Dose response relative to that at 25°C for Harwell alanine dosimeters irradiated to absorbed doses of 5, 25, 50, 60, 80 and 100 kGy at fixed temperatures ranging from −10°C to +50°C.

The advent of food irradiation expanded the range of interest to sub-zero temperatures and extended the range over which the coefficient was characterized to approximately −10°C. Recently, an ever-growing number of products are being irradiated that require temperature control at even lower temperatures (e.g., sera used in cell cultures and tissue for implantation), and these applications have fueled the need for investigating the behavior of dosimeters at much lower temperatures.

Many years ago, research in high-energy physics stimulated a limited study on the alanine dosimeter response characteristics at temperatures significantly below zero Celsius. Coninckx et al. (1996) calibrated alanine dosimeters irradiated at 4 and 77 K, temperatures not relevant to industrial applications. Moreover, this work was performed on custom-made alanine dosimeters. Response data for commercially produced alanine dosimeters irradiated at low temperatures are not available.

A key step in launching new irradiation applications at low temperature will be regulatory approval of the process. This requires measurements of absorbed dose made by the industrial processor to be accurate and traceable to a national metrology institute (NMI). As alanine is the reference dosimetry system used by all NMI's in their high-dose calibration services, a characterization of the alanine low temperature dose response was undertaken.

2. Experimental[2]

Commercially available alanine pellet dosimeters from Harwell Dosimeters and Gamma Services were used in these experiments. These dosimeters are similar in dimension, mass and both use L-alanine, however, the binders differ. Harwell uses a paraffin-based binder while Gamma Service uses a non-paraffin (proprietary) binder.

Irradiations were conducted in a Gammacell 220 (Nordion, Canada) Cobalt-60 gamma source with a dose rate of about 18 kGy/h. A system that employed a custom-designed aluminum holder combined with controlled-temperature airflow was used to achieve thermal equilibrium from +50°C to −55°C during irradiation (described in Nagy et al., 2000). For lower temperatures, dry ice was the primary source of temperature control. The dosimeters in the holder assembly were pre-equilibrated to the target temperature prior to irradiation and monitored throughout the irradiation to maintain the target temperature within 1°C throughout the irradiation. The irradiation temperatures used to represent the data in Figs. 1–5 were the computed

[2] 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.

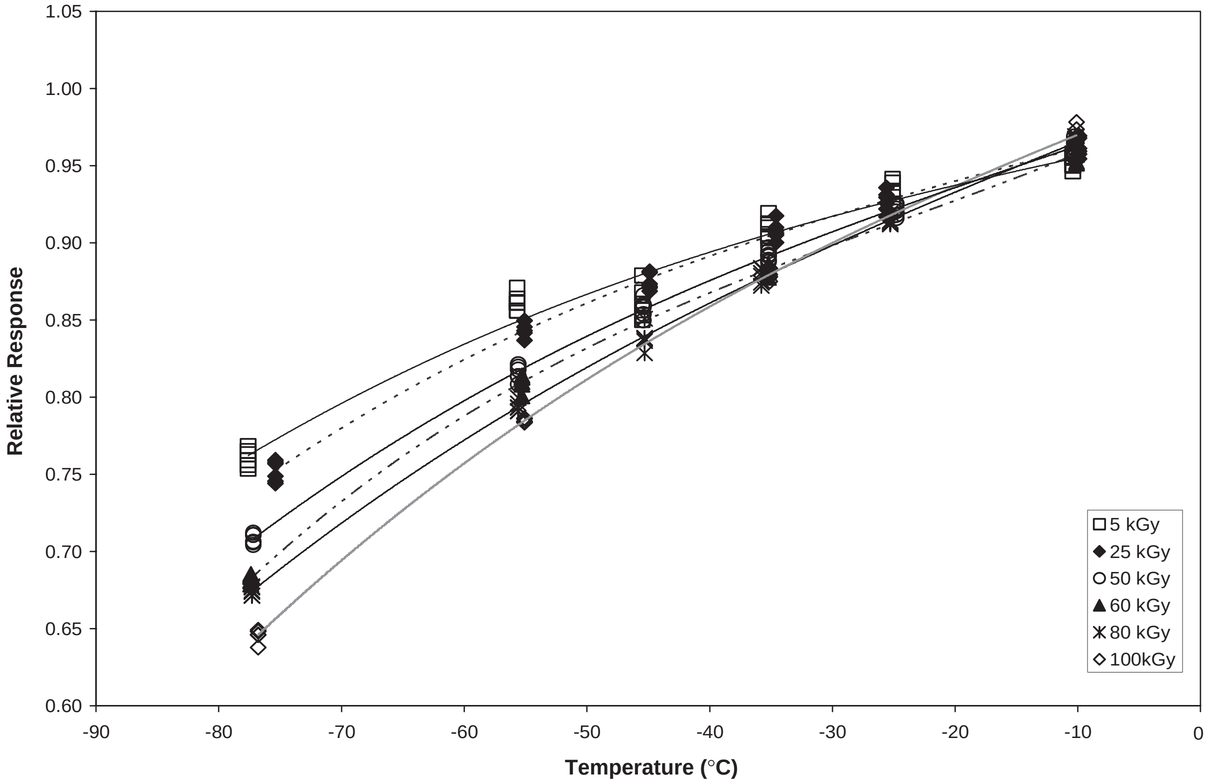

Fig. 2. Dose response relative to that at 25°C for Harwell alanine dosimeters irradiated to absorbed doses of 5, 25, 50, 60, 80 and 100 kGy at fixed temperatures ranging from −77°C to −10°C.

average of the temperatures measured for the duration of the irradiation.

The electron paramagnetic resonance (EPR) signal measurement protocol for ECS106 spectrometer measurements performed at NIST has been described in detail previously (Nagy et al., 2000). The essential features are that each dosimeter is measured at two angles and that these measurements are normalized to dosimeter mass and the spectrometer's internal reference material (ruby crystal). The average of the normalized signals for both angles is used as the dosimeter response.

The EPR signal measurements by Clearant were performed using a Bruker Biospin e-scan spectrometer (microwave power, 0.517 mW; modulation amplitude, 0.101 mT). The signal of each dosimeter was measured, and the signal normalized to the signal of an internal EPR reference material (proprietary) signal to derive an alanine-to-reference ratio. This ratio was further normalized to the dosimeter mass, and this mass-corrected ratio was used as the dosimeter response.

The EPR dosimeter response for dosimeters irradiated to −10°C and higher underwent linear regression and the resultant function was used to compute the EPR response predicted at 25°C. This value served as the reference point from which the relative response for all measurements was calculated. The value for the slope of the relative response plotted versus irradiation temperature is the temperature coefficient. A third-order polynomial was used to fit data that extended below −10°C.

3. Results and discussion

Alanine dosimeters manufactured by Harwell were irradiated to absorbed doses of 5, 25, 50, 60, 80 and 100 kGy at fixed temperatures ranging from −10°C to +50°C. Analysis of the dosimeter relative response (measured on a Bruker e-scan spectrometer) to irradiation temperature at each of the dose levels revealed no significant difference in the derived temperature coefficient. With no discernable difference in the temperature coefficient for each absorbed dose, the data from all doses were combined and plotted as relative response versus temperature (Fig. 1). A linear regression of these data produced a temperature coefficient of +0.11%/°C. This value for the temperature coefficient is consistent with previously reported values for L-alanine pellet dosimeters (Nagy et al., 2000).

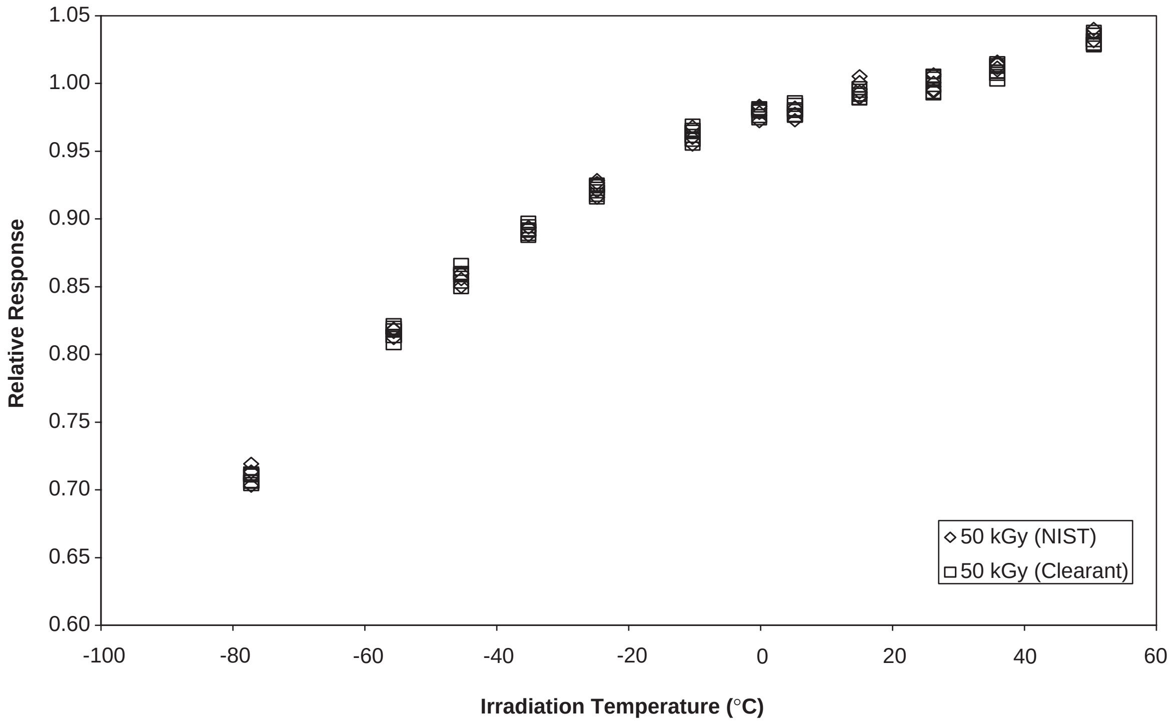

Fig. 3. Dose response relative to that at 25°C for Harwell alanine dosimeters irradiated to an absorbed dose of 50 kGy at fixed temperatures ranging from −77°C to +50°C measured at NIST (ECS106 EPR spectrometer) and Clearant (e-scan EPR spectrometer).

Harwell alanine dosimeters were also irradiated to these same doses but at temperatures in the range −77°C to −10°C. Dosimeter response measurements were adjusted to the same relative value in Fig. 1 and plotted versus irradiation temperature (Fig. 2). A marked deviation from linearity was apparent at each dose level. The deviation from linearity became greater as the temperature decreased. The discrepancy in relative response at −77°C was 10–20% lower than would be predicted from the linear trend in the high temperature region (> −10°C) of the plot. Also, evident from these data is a dependence of the relative response curve on the absorbed dose. The relative response decreases as the dose increases.

In order to correlate dosimeter measurements made on the NIST and Clearant spectrometers, the relative responses for each of the same sets of 5 and 50 kGy dosimeters were measured on the two spectrometers. The measurements for 50 kGy are shown in Fig. 3. Clearly, there is excellent agreement between the data. Similar agreement was observed for the 5 kGy data comparisons (not shown).

The Bruker ECS106 spectrometer was used to study temperature effects on lower doses. It is a more sensitive instrument than the e-scan and can measure with greater precision dosimeters irradiated to doses lower than 5 kGy. Harwell dosimeters irradiated to 0.5, 5, and 50 kGy over the same temperature range were measured on this spectrometer. The dosimeter relative responses measured for all three doses are compared in Fig. 4. As in Fig. 2, the 50 kGy dosimeters showed a greater temperature effect than the 5 kGy dosimeters. However, at 0.5 kGy the trend is reversed.

Since commercial dosimeters are available from other manufacturers, it was important to test whether these observations are unique to the Harwell dosimeter. In addition, binder influences on dosimeter temperature effects have been postulated (Nagy et al., 2000). Gamma Service alanine dosimeters were irradiated over a −55°C to +25°C range for comparison to the Harwell dosimeters previously characterized. Fig. 5 compares the temperature effects on the dose response for both dosimeters. The trends for each system are indistinguishable. The temperature coefficient measured for Gamma Service dosimeters in the −15°C to +25°C range was +0.10%/°C.

4. Conclusion

The temperature coefficients measured for the two most common commercial alanine pellet dosimeters in the temperature range −10°C to +50°C are consistent with published coefficients for other alanine pellet

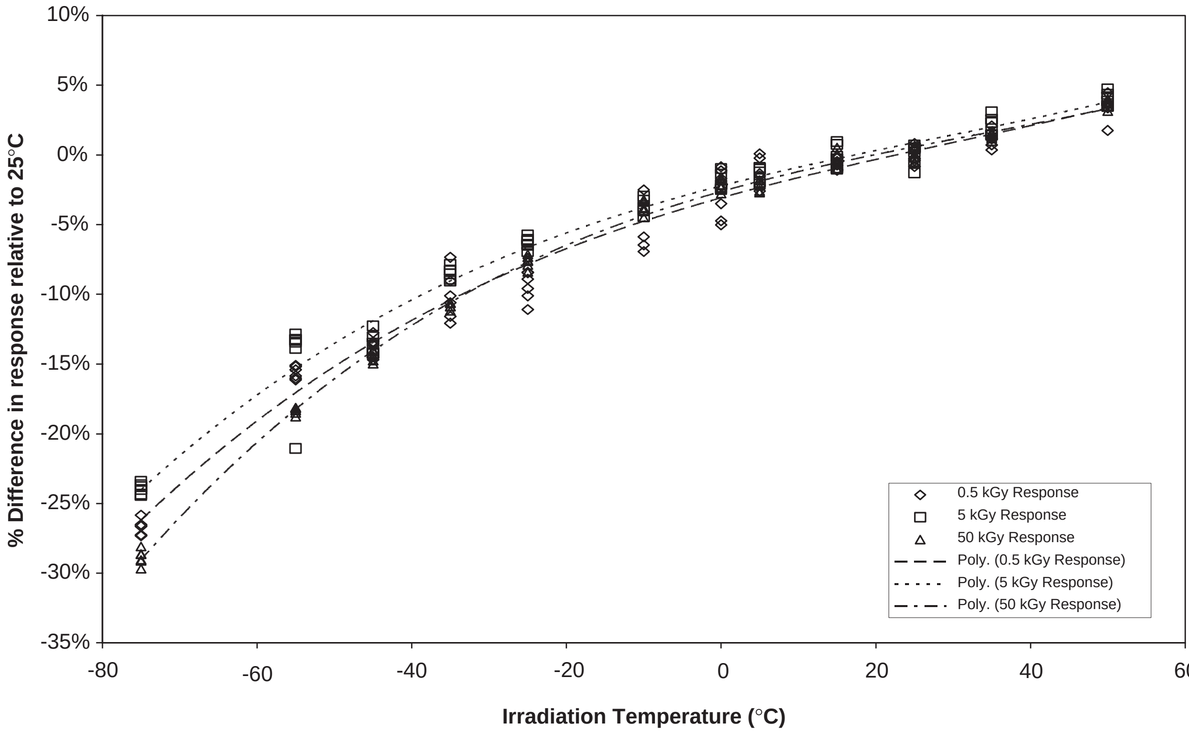

Fig. 4. Relative dose response of Harwell alanine dosimeters irradiated to absorbed doses of 0.5, 5 and 50 kGy at fixed temperatures ranging from −77°C to −10°C (measured at NIST).

dosimeters (Nagy et al., 2000). Extension of the temperature range to −77°C revealed a dosimeter response that is lower than that predicted from the linear relationship above −10°C. The deviation from linearity increased with decreasing temperature and was observed for both commercial dosimeters. Accurate dosimetry requires a temperature-based correction of the dosimeter response to that of the calibration temperature. The observed deviations from linearity are significant and corrections must be applied. Since the alanine dose response over the range studied is non-linear, these observed changes in response would produce greater changes in the computed absorbed dose (or lead to large errors without the appropriate corrections). Subsequent investigations of the temperature effects on alanine film dosimeters as well as alanine dosimeters from other manufacturers that use either DL-alanine or L-alanine crystals exhibit the same effect (Desrosiers and Cooper, unpublished results).

The magnitude of the temperature effect on dosimeter response increased with dose from 5 to 50 kGy (Fig. 2). However, a general recommendation regarding the dose–response relationship as a function of temperature cannot be made since potentially conflicting results were observed in Fig. 4; here, the 0.5 kGy response curve lies between that of 5 and 50 kGy. A dose effect on the temperature coefficient for alanine pellets (from different manufacturers) was previously observed for temperatures above zero (Nagy et al., 2000). However, the dose effect reported by Nagy was not observed for the Harwell and Gamma Service pellets measured here. The dose-dependent effect at low temperature should be examined in greater detail.

The mechanism by which the irradiation temperature affects the alanine dosimeter response to absorbed dose cannot be derived directly from these data. However, a study of what is currently known about the solid state radiation chemistry of crystalline alanine offers a plausible explanation for these observations. Desrosiers et al. (1995) published the first direct evidence for detectable levels of chemically distinct secondary alanine-derived radicals in irradiated alanine crystals. Subsequent spectroscopic analysis studies produced postulated structures and relative yields of these radicals (Malinen et al., 2003, and references therein). The implications of these studies are that the observed alanine spectrum is actually a composite of two or more alanine-derived radicals. The observed changes in the measured alanine signal amplitude likely result from the effect of temperature on the reaction rates for the various pathways available to the reactive chemical intermediates. The net effect of this influence is an

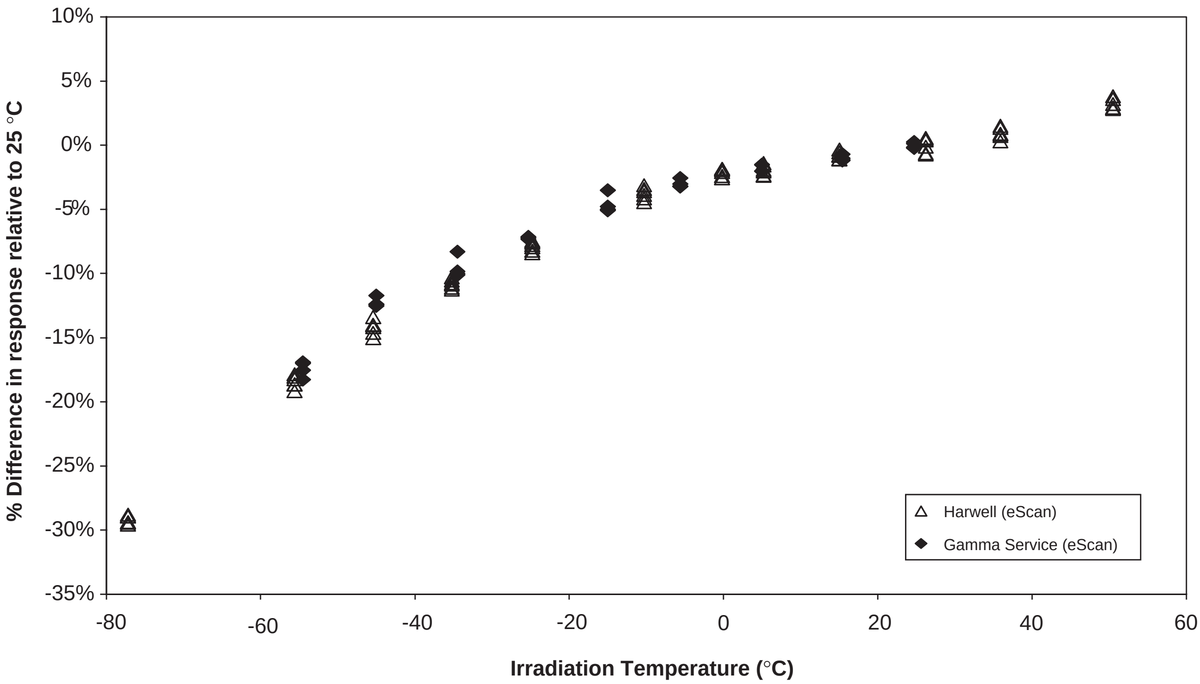

Fig. 5. Relative dose response of Harwell alanine dosimeters irradiated to absorbed doses of 50 kGy at fixed temperatures ranging from −77°C to +50°C (Harwell) and −55°C to +25°C (Gamma Service).

alteration in the relative distributions of the alanine-derived radical species that combine to produce the signal amplitude measured for dosimetry.

The implications for dosimetry are that linear temperature corrections can only be applied in the −10°C to +50°C irradiation temperature range. A non-linear correction must be applied below −10°C. For the NIST measurements between 0.5 and 50 kGy a third-order polynomial[3] would be suggested to apply response corrections. However, it has yet to be determined if this correction is dosimeter batch specific. These data also suggest that, depending on the precision requirements of the dosimetry system employed, a single relationship for this correction may be applied below 50 kGy. However, it is likely that corrections related to absorbed dose should be used above 50 kGy.

[3] A least-squares fit to the data (collectively) in Fig. 4 gave the relationship, $R = (1.3E{-}5)T^3 + (1.2E{-}3)T^2 + (1.5E{-}1)T{-}0.26$, where R is the dosimeter response (in percent relative to 25°C) and T is the irradiation temperature (°C). For this experiment, the difference in the computed percent response at the irradiation temperature to the percent response at 25°C (the calibration temperature) will give the percent correction to apply to the measured response of the irradiated dosimeter (only if the irradiation temperature is below −10°C). For dosimeters irradiated above −10°C, the temperature effect is linear and the temperature coefficient is used to correct dosimeter response to the calibration temperature.

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