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Absorbed-dose/dose-rate dependence studies for the alanine-EPR dosimetry system

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ABSTRACT

Over the course of the last decade, routine monitoring of the alanine dosimetry system revealed a small but significant observation that, after examination, led to the characterization of a previously unknown absorbed-dose-dependent, dose-rate effect for the alanine system. The newly discovered rate effect is of potential concern for electron-beam dosimetry, since electron-beam dosimetry typically derives its traceability to national standards through comparisons to gamma-ray calibrations of the dosimetry system. The largest discrepancy in source dose rates is between gamma-ray sources and electron-beam accelerators. Investigating the influence of temperature on the alanine rate effect is an important first step in preparation for a comparison study between electron-beam and gamma-ray dosimetry. Here, new data is presented on the influence of irradiation temperature (from −40 to +50 °C) on the dose-rate effect measured at 50 kGy.

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1. Introduction

Check standards are used by the NIST Ionizing Radiation Division to monitor the performance of the alanine dosimetry system that is central to its high-dose transfer dosimetry service. These measurements are performed to confirm the operational readiness of the calibration curve. Deviations from the expected check-standard values can result from a wide range of sources that include both manufacturing abnormalities in a dosimeter and spectrometer-related changes. Recently, check-standard measurement deviations unveiled a previously unknown rate effect for the alanine dosimetry system (Desrosiers et al., 2008). This rate-effect study characterized a complex relation between the radiation chemistry of crystalline alanine and the applied dose rate that was also dependent on the absorbed dose. That the rate effect only becomes significant above 5 kGy likely contributed to it only recently being discovered despite decades of research in alanine dosimetry. It was learned that the effect is intrinsic to alanine and is not dependent on the chemical form or manufacturing formulation of the alanine dosimeter. The study postulated that the production of one (or more) of the radiation-induced alanine radicals is dependent on the dose rate.

Typically, high-dose-rate calibration sources maintained by National Measurement Institutes (NMIs) are calibrated against low-dose-rate sources that are used to realize the gray with a primary standard (e.g., calorimeter). If a dose rate is established by comparison of dosimeters irradiated in a high-rate gamma calibration source to dosimeters calibrated in a low-rate source at absorbed doses in the kGy range, errors could be introduced in the dose rate for the high-rate calibration source. If this calibration source is in turn used for service work, a dose-dependent error would be further transferred to industrial customers. Even if the scenario cited above is not relevant for a specific NMI, two additional considerations remain. First, caution would be advised to avoid using alanine dosimeters at high doses (>5 kGy) for transfer calibrations of low-dose-rate ($<2\,\mathrm{Gy\,s^{-1}}$) sources; second, to avoid using a gamma source with a low-dose-rate ($<2\,\mathrm{Gy\,s^{-1}}$) to calibrate alanine dosimeters.

The range of dose rates used in the previous study (0.1–5 $\mathrm{Gy\,s^{-1}}$, Desrosiers et al., 2008) is typical for gamma-ray sources. However, electron-beam irradiators are being used for many industrial applications. This raises the question: do rate effects exist for the much higher dose rates ($10^4\,\mathrm{Gy\,s^{-1}}$ and higher) employed in electron-beam dosimetry? The electron and gamma irradiation technologies are so vastly different that a subtle rate effect could be masked by larger effects (e.g., irradiation temperature). Electron-beam irradiations are typically accompanied by significant increases in temperature, whereas gamma-ray irradiations used to calibrate the dosimetry system are typically temperature-controlled at ambient or moderate temperatures. This work presents the results of measurements that are fundamental to the electron-beam dosimetry investigation.

Previously published 1995-through-2007 source-comparison measurements (Desrosiers et al., 2008) compared the response ratios of dosimeters irradiated (to the same dose) with gamma sources of dose rates lower than $1.4\,\mathrm{Gy\,s^{-1}}$ (these include the Pool,

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GC45, and GC232) to dosimeters irradiated concurrently with gamma sources of dose rates greater than 3.5 Gy s^{-1}. The absorbed doses for these measurements were grouped as low dose (1 kGy) or high dose (predominately 25 kGy, but one 20 kGy and one 30 kGy ratio is included). The 1 kGy ratios of low-rate to high-rate measurements remain clustered about unity consistently from 1995 through 2007 regardless of the gamma source used. The 20–30 kGy measurement ratios consistently indicate a response 1–2% lower than that from the 1 kGy measurements regardless of the gamma source used. A linear regression of previously published data (Desrosiers et al., 2008) for the 20–30 kGy ratio data suggests that maximum effect (computed at zero dose rate) should be approximately 2%. The regression intercepts unity at a dose rate of approximately 4 Gy s^{-1}. The regression of the 20–30 kGy ratio data becomes significant (intercepts 0.99 of the y-axis) at a dose rate of approximately 2 Gy s^{-1}. Measurements to date suggest that the effect is not linear with absorbed dose rate, so the linear approximations would represent conservative estimates. A more comprehensive study of high- and low-dose dosimeter response ratios between 1 and 4 Gy s^{-1} is needed to assess the dose-rate effect transition. In the interim, these data will guide the assignment of uncertainties to this effect for calibrations of sources in this dose-rate range.

Alanine as a dosimeter for the high dose rates produced by high-energy industrial electron-beam accelerators is worthy of investigation, because the dosimetry system is typically calibrated by comparison to much lower dose-rate gamma-ray sources. The differences between electron-beam and gamma-ray irradiation processing will present several challenges to the investigation. One of these challenges, the influence of irradiation temperature, can be controlled for gamma-ray dosimetry in a laboratory facility but is uncontrolled for high-dose electron-beam irradiations. In practice, the dosimeter response is corrected for differences between the average irradiation temperature of the field dosimeter and the irradiation temperature used for the dosimetry system calibration. This dosimeter response adjustment is made using a previously determined temperature coefficient. However, applying these corrections in studies designed to search for a dose-rate effect between electron-beam and gamma-ray irradiators without first investigating the influence of temperature on the rate effect itself may have unintended consequences. If, as postulated by Desrosiers et al. (2008), the rate effect is a result of changes in the free radical distribution, then the irradiation temperature could potentially further influence the reaction rates that govern the yield of the different free radicals. In turn, the altered free radical distribution could (positively or negatively) alter the net electron paramagnetic resonance (EPR) spectral amplitude that manifests itself as the observed dose-rate effect. Here, the term *net spectral amplitude* is used to convey the concept that the EPR amplitude used as a measure of the dosimeter response is actually a composite of EPR amplitudes from at least two, likely three, and possibly more alanine free radicals (Wieser et al., 1993; Desrosiers et al., 1995; Malinen et al., 2003).

2. Experimental

The absorbed doses for this study were delivered with either of two Gammacell 220 ^{60}Co gamma-ray sources (Nordion, Canada).[1] The Gammacell activities were: 108 TBq (2.9 kCi, serial number GC232); and 531 TBq (14 kCi, serial number GC207), all as of August 1, 2008. Alanine dosimeters were irradiated in the center position geometry of each source. The irradiation geometry and its calibration are described in NIST SP250-45 (Humphreys et al., 1998) with the exception of a modification to the calibration scheme described by Desrosiers et al. (2008). Temperature during the irradiation was controlled by using a high-flow air shower from a TurboJet (FTS Systems) and monitored with a type-T thermocouple.

The dosimeters were measured by electron paramagnetic resonance spectrometry. A Bruker Biospin ECS106 EPR spectrometer was configured to specifically measure alanine pellet dosimeters (Humphreys et al., 1998). The EPR amplitude is 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 alanine/ruby EPR amplitude ratio 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.

Alanine dosimeter response ratios were determined from dosimeters irradiated in tandem in the GC207 and GC232 Gammacell 220 ^{60}Co irradiators. The tandem irradiations were timed such that they would terminate at approximately the same time and permit the post-irradiation readout times to be equivalent. Dosimeters were stored under ambient laboratory conditions for 24 h before measurement.

3. Results and discussion

A set of experiments was undertaken to examine the effect of irradiation temperature on the dose-dependent dose-rate effect previously observed (Desrosiers et al., 2008). Response ratios for dosimeters irradiated at high dose (50 kGy) and low dose (1 kGy) were determined by comparing dosimeter responses of equivalent dose that were achieved with different dose rates. The Gammacells GC207 (3.1 Gy s^{-1}) and GC232 (0.62 Gy s^{-1}) were used for these irradiations. At each dose level in each Gammacell, an average irradiation temperature was maintained at each of the following: −40, −10, 0, +10, +24, +38, or +50 °C. The dosimeter response at each dose level and temperature was used to compute

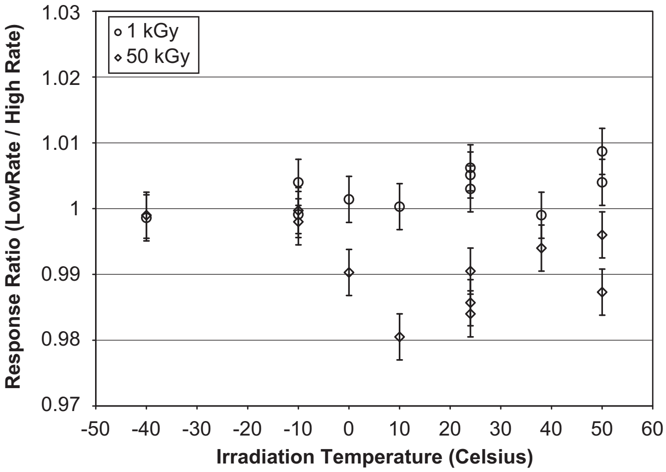

Fig. 1. Plot of the alanine dosimeter response ratio (low rate, 0.62 Gy s^{-1}, over high rate, 3.1 Gy s^{-1}) versus the irradiation temperature in degrees Celsius for 1 kGy (circles) and 50 kGy (diamonds). The error bars represent the standard uncertainty ($k = 1$).

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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.

response ratios in terms of the dosimeter response for the low-dose-rate source divided by the dosimeter response for the high-dose-rate source. These data are summarized in Fig. 1. The nonequivalence of the response ratios between 1 and 50 kGy at +24 °C was comparable to the previous findings (Desrosiers et al., 2008). The results obtained for the higher temperatures, +38 and +50 °C, though within the range of the +24 °C data, suggested that there may be a trend towards a weaker dose-rate effect at elevated temperatures. The dose-rate effects at 0 and +10 °C are consistent with the +24 °C data. However, a distinct change in the dose-rate effect occurred at the irradiation temperatures of −10 and −40 °C. There was no measurable rate effect at 50 kGy at either temperature.

4. Conclusions

The initial concern that the elevated temperatures common to high-dose electron-beam irradiations would negatively impact the ability to explore the possibility of dose-rate effects under these conditions has been alleviated. The data suggest that there may be a trend towards a reduction in the rate effect with increasing irradiation temperature. This is a positive result as the uncertainty in the electron-beam irradiation temperature would not negatively influence the precision of the alanine dose assessment. The finding that there is no dose-rate effect for irradiation temperatures of −10 and −40 °C is of particular interest to dosimetry for electron-beam processing of food and medical products. Measurements to determine the rate effect temperature profiles for absorbed doses between 5 and 100 kGy are planned.

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