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Characterization of a new alanine film dosimeter: relative humidity and post-irradiation stability

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Abstract

The alanine dosimetry system is now preferred by all primary and secondary calibration laboratories for their high-dose ionizing radiation calibration services. Alanine dosimeters in the form of cylindrical pellets have been available in commercial quantities for several years. Despite their high quality, the physical dimensions of the dosimeter were not suitable for many electron-beam processing dosimetry applications. Alanine in the form of a thin-film would be more appropriate for electron-beam applications. However, a reliable manufacturer/supplier for high-quality alanine film dosimeters was lacking. To fill this void Bruker BioSpin and Kodak have teamed up to successfully develop an alanine film dosimeter that appears to have excellent dose measurement properties in a highly robust form. Described here are the characterization studies for this new dosimeter that assess the effects of relative humidity and the post-irradiation temporal stability of the alanine film dosimeter.

Keywords: Alanine; Dosimetry; Electron beam; Electron paramagnetic resonance; Gamma rays; Relative humidity

1. Significance

In calibration laboratories, the relative humidity (RH) is either controlled or very limited in range. However, the range of environments those dosimeters are exposed to can vary widely from one geographic region to another. Preferably, a dosimeter should either be insensitive to its environment or respond in a predictable manner to environmental influences ensuring consistent and comparable results between facilities in different locations. Post-irradiation stability, sometimes referred to as "fade characteristic" is also an important attribute. If the post-irradiation response of the dosimeter changes significantly over time, the time-dependent dosimeter measurements require additional controls to ensure readings are taken at the correct interval. This lack of flexibility can have detrimental effects to facility scheduling where necessary shutdowns for maintenance or source replenishment may make delays in analysis unavoidable.

2. Testing parameters

The Bruker e-scan EPR spectrometer is designed to record the alanine dosimeter's radiation response relative to a manufacturer-installed EPR "marker" material to compensate for the moisture content of the dosimeter. The Bruker e-scan dosimeter holders (probes) contain the reference marker material. During the

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measurement process, the spectral sweep is set to include both the alanine and marker EPR signals. It is the ratio of the alanine signal amplitude to marker signal amplitude that is the quantity used to determine absorbed dose.

EPR measurement parameters for National Institute of Standards and Technology (NISTs) EMS 104 film measurements were a microwave power of 1.99 mW and modulation amplitude of 2.9 G (0.29 mT). EPR parameters for STERIS' e-scan EPR spectrometer were a microwave power of 0.99 mW and modulation amplitude of 2.0 G (0.20 mT).

NIST irradiations were conducted in a Nordion GammaCell 220 with a rate of 18 kGy/h. STERIS employed two sources, a GammaCell 220 (8 kGy/h) and a 5 MeV electron-beam accelerator (10 kGy/s) in order to test the new film under a wide range of conditions.

3. Relative humidity: testing and results

The first set of tests was performed on the Kodak[1] alanine dosimeter film lot B0247. The dosimeter is composed of an alanine-polymer layer ($\sim 4.5 \times 0.4\,cm^2$) deposited on a 14.5-cm polymer support. The support serves as a handle for easy introduction and removal of the dosimeter into the Bruker e-scan spectrometer holder. Located on the handle of each dosimeter is a unique number and barcode. The number allows the user to identify the dosimeter and its associated lot identification code. The barcode offers several advantages in an industrial environment that include its use to track the dosimeter within the facility as well as enabling it to be used in an on-line calibration service in development at NIST (Desrosiers et al., 2002).

Several groups of these alanine dosimeters were conditioned at the NIST in RH environmental chambers controlled to 0%, 33%, 44%, 57%, 75% and 94% RH according to previously described procedures (Sleptchonok et al., 2000). After the dosimeters were conditioned for several days, they were then hermetically sealed in sachets. After setting aside conditioned dosimeters for gamma irradiation at NIST, several sachets of dosimeters equilibrated at 0%, 33%, 75%, and 94% RH were shipped to STERIS Isomedix for irradiation and post-irradiation analysis. The STERIS-irradiated dosimeters were irradiated in the sealed sachets to a dose of 20 kGy. The EPR spectrum was recorded and analyzed with one of two Bruker e-scan spectrometers (designated as 119 and 123) at 10-min intervals for a minimum period of 4000 min ($\cong$67 h) and a maximum period of 5000 min ($\cong$83 h). After the measurements were complete, the dosimeter was re-sealed and stored in a temperature-controlled environment with an average temperature of 25°C. When the dosimeter readings were completed for each of the conditioned dosimeters these same dosimeters were then re-read using the same testing parameters. NIST-irradiated dosimeters were gamma-irradiated in sealed sachets to a dose of 5.0 kGy (1.5% expanded uncertainty at a 95% confidence level). This measurement protocol compensates for environmental changes in that if the moisture content of the dosimeter changes during the measurement period, the alanine signal and the marker signal changes are equivalent. This approach has the net effect of eliminating environmental influences on the measurement; the ratio remains constant even if the moisture content changes during the measurement session (Sleptchonok et al., 2000).

All four RH-conditioned dosimeters irradiated at STERIS exhibited a change in alanine-to-marker signal ratio that ranged from −1.5% to +0.5% (see Figs. 1 and 2 for representative data) relative to the dosimeter's final response value. The test at 94% RH was repeated three times since this is an extreme or worst-case condition.

NIST studies included dosimeters conditioned at relative humidities not distributed to STERIS. These measurements were performed on a Bruker EMS104 EPR spectrometer. Since this spectrometer does not have an internal EPR reference material, only the alanine signal amplitudes were measured. Because of this the signal amplitude measurements could be influenced by the moisture content of the dosimeter. The dosimeters were removed from the constant RH environment, sealed in sachets and gamma-irradiated to 5.0 kGy in the sealed sachets. The dosimeters were measured within 1 h of removal from the gamma irradiator. The data are shown in Fig. 3. The large range of EPR signal amplitudes measured for 0% RH dosimeters may reflect a distribution in the location of water in the alanine-containing active layer of the film. Some films may have varying degrees of water bound such that it is difficult to remove by desiccation. These films would yield smaller EPR signal amplitudes than films purged of water. This signal reduction arises from the effect of moisture on the Q of the spectrometer's microwave resonator. This effect would not be observed in the e-scan spectrometer since the internal reference material experiences an identical effect that is the basis for the use of signal ratios as the measurand.

4. Time-dependent stability (fade): testing and results

Alanine dosimeters from lot B0247 were also used to determine the stability of the film dosimeters

[1] 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.

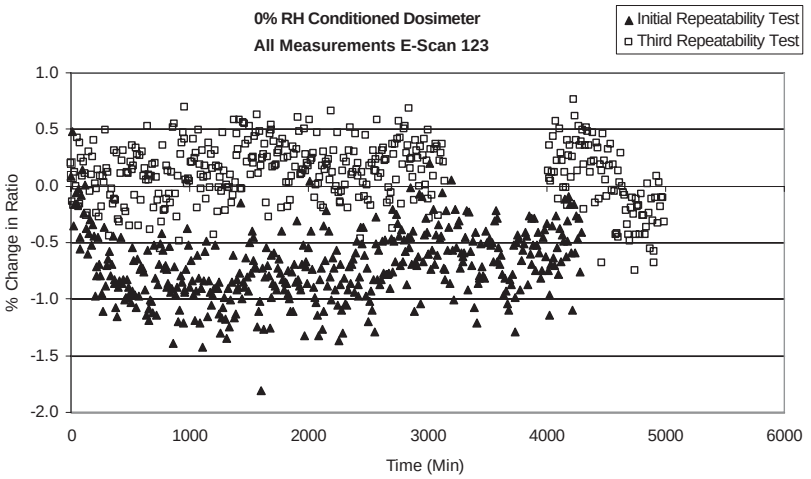

Fig. 1. The percent change in the response ratio for a 20 kGy irradiated alanine film pre-equilibrated to 0% RH.

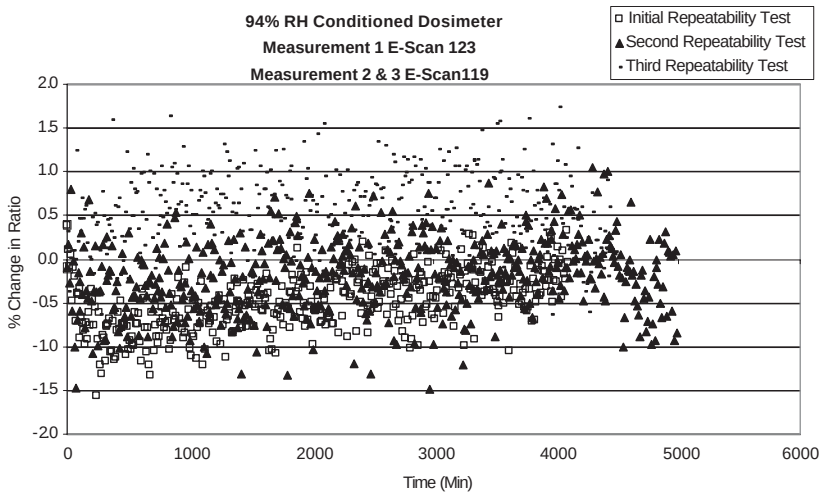

Fig. 2. The percent change in the response ratio for a 20 kGy irradiated alanine film pre-equilibrated to 94% RH.

immediately following irradiation and for a period thereafter. Irradiations with gamma and e-beam sources were performed for comparison. The Gammacell was used to test moderate dose-rate irradiations whereas the electron beam accelerator was used to test high dose-rate conditions.

An alanine film dosimeter was irradiated to a dose of 20 kGy in the electron beam accelerator and read every 10 min for approximately 80 h using the e-scan spectrometer. Another alanine film dosimeter was gamma-irradiated to an absorbed dose of 20 kGy and immediately after removal from the source measured every 10 min for approximately 110 h using the e-scan EPR spectrometer. The electron-beam irradiated dosimeter experienced an overall change in response of 1.5% that ranged from −1.5% to 0.0% of the initial value (see Fig. 4). The gamma-irradiated dosimeter experienced an overall change in response of 1.5% that ranged from –0.5% to 1.0% of the initial value (see Fig. 5). The line in Figs. 4 and 5 is a 10-point moving average. This was chosen as a simple means of identifying any drift in the ratio within the random scatter of data points.

The STERIS Gammacell 220 was used to irradiate a set of calibration dosimeters in the dose range of 5.0–50.0 kGy. After the irradiations one dosimeter from each dose point was measured and then again one month later to determine if they had experienced any fade and/or development characteristics. Each dosimeter was within ±2% of its calibration dose. The mean alanine/marker ratio was calculated and the difference between the calibration dose and the measured dose was 0.006%

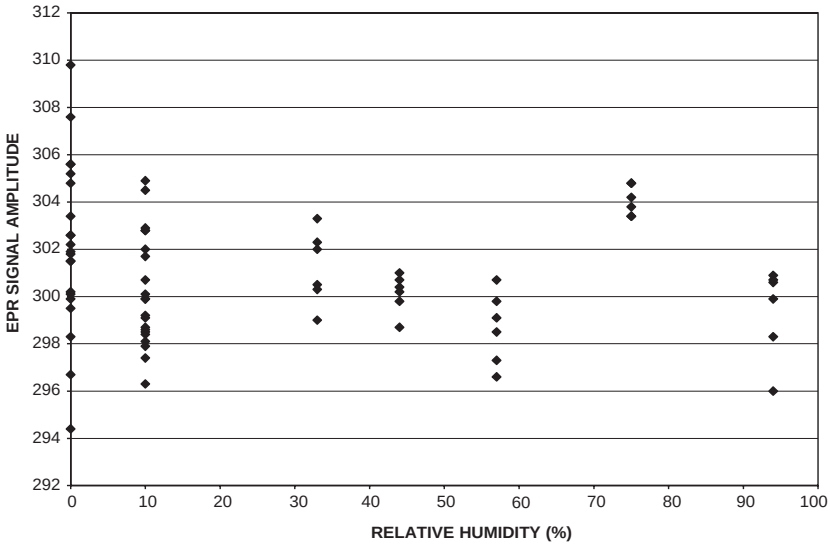

Fig. 3. Comparison of the EPR amplitudes of 5.0 kGy gamma-irradiated alanine film dosimeters pre-equilibrated at several relative humidities ranging from 0% RH to 94% RH.

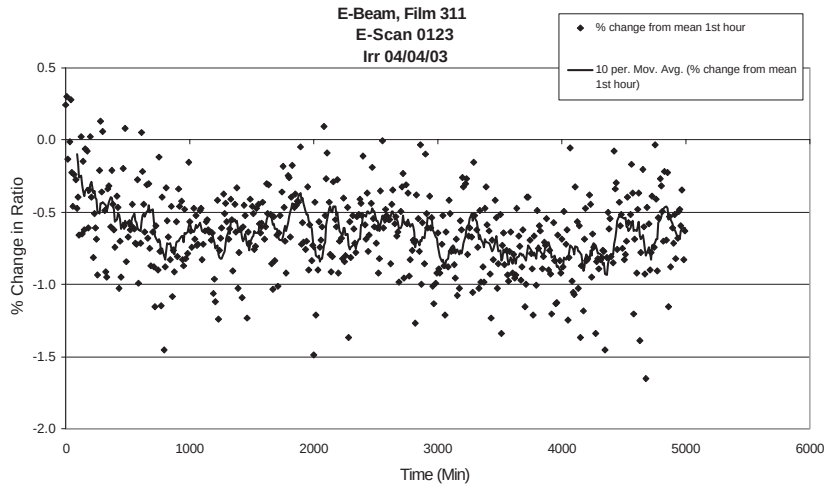

Fig. 4. The percent change in the response ratio for a 20 kGy electron-beam irradiated alanine film.

for the first set of readings and 0.007% for the second set of readings.

5. Discussion of results

From this experiment, it was concluded that the response of the Kodak alanine film dosimeters is relatively insensitive to the effects of humidity over a broad range of RH. While the existing industry standard calls for packaging to protect the dosimeter from the effects of the humidity in the environment, these data suggest that this requirement is unnecessary for the Kodak alanine dosimeter, although packaging may be advisable if the possibility of damage exists during dosimeter handling in the industrial process.

During the time-dependence measurements, all dosimeters were stored in a controlled environment with a temperature in the range of 15.0–25.0°C. These tests indicate that the Kodak alanine film dosimeters are subject to little if any fade or development characteristics in controlled environments.

The alanine dosimetry system has long been heralded as superior to all other dosimetry systems used in industrial processing. However, since the alanine system use has been limited to primary and secondary calibration laboratories, the transferability of this system to the potentially harsh conditions at the

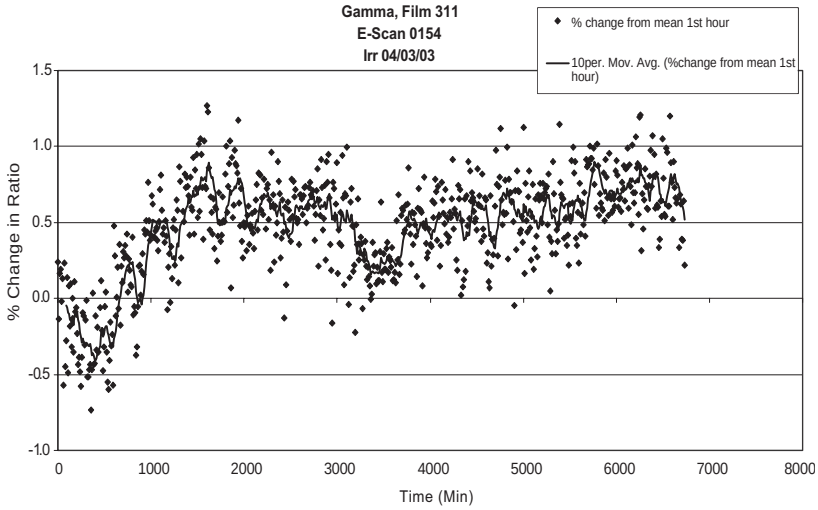

Fig. 5. The percent change in the response ratio for a 20 kGy gamma-ray irradiated alanine film.

industrial level was unclear. The interoperability between gamma and electron-beam irradiated dosimeters is important in establishing traceability to national standards. These data demonstrate that the superior attributes of the alanine system are transferable to industry. The combined advances in dosimeter manufacturing and spectrometer engineering offer the radiation processing industry a significant improvement in its measurement capabilities that should translate to cost savings and improvements in quality.

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