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DEVELOPMENT OF AN ALANINE DOSIMETER FOR GAMMA DOSIMETRY IN MIXED ENVIRONMENTS²

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ABSTRACT: L- α -Alanine, a nontoxic polycrystalline amino acid, has been investigated for use in high-precision, high-level absorbed-dose measurements in mixed neutron/photon environments such as research and test reactors. The technique is based on the use of electron paramagnetic resonance (EPR) spectroscopy to determine the extent of free radical production in a sample exposed to ionizing radiation, and has been successfully used for photon absorbed-dose measurements at levels exceeding 10^5 Gy with high measurement precision. Application of the technique to mixed environments requires knowledge of the energy-dependent response of the dosimeter for both photons and neutrons. Determination of the dosimeter response to photons is accomplished by irradiations in ^{60}Co and bremsstrahlung sources and by calculations of energy-dependent photon kerma. Neutron response is determined by irradiations in conjunction with $\text{CaF}_2\text{:Mn}$ thermoluminescence dosimeters and by calculations of energy-dependent neutron kerma. Several neutron environments are used, including the ACRR and SPR-III reactors.

KEYWORDS: Alanine, Dosimeter, Mixed Fields, Gamma-Ray, Neutron, Neutron-Gamma Environments, Electron Paramagnetic Resonance, Electron Spin Resonance

Introduction

L- α -Alanine, a nontoxic polycrystalline amino acid, has been used in recent years as a dosimeter for high-precision, high-level absorbed-dose measurements in gamma-radiation fields. The technique is based on the use of electron paramagnetic resonance (EPR) spectroscopy to determine the extent of free radical production in a sample exposed to ionizing radiation. It has been successfully used for gamma-ray absorbed-dose measurements at levels exceeding 10^5 Gy and with a measurement precision of better than 3.5 percent [1]. With carefully controlled conditions, a precision of 1 percent can be obtained.

Because of the wide absorbed-dose range and high precision that can be achieved with this method, there is considerable interest in its possible use in radiation-hardness testing of electronics, both in photon and in neutron environments. Gamma-ray dosimetry for this application has traditionally been done with $\text{CaF}_2\text{:Mn}$ thermoluminescence dosimeters (TLDs) [2] and has therefore been limited to absorbed-dose levels below 5×10^3 Gy, with a measurement precision of 5 to 10 percent. Other applications for which a high-precision dosimeter is needed include facility calibration and absorbed-dose mapping, and characterization of new test configurations.

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Before this technique can be used for mixed-radiation fields, it is necessary to characterize the energy-dependent response of the dosimeter for both photons and neutrons. Determination of the dosimeter response to photons is accomplished by irradiations in ^{60}Co and bremsstrahlung sources and by calculations of energy-dependent photon kerma. Neutron response is determined by irradiations in conjunction with traditional dosimeters, supported by calculations of energy-dependent neutron kerma. Several neutron environments are used, including the Sandia Pulsed Reactor (SPR-III), the central cavity of Sandia's Annular Core Research Reactor (ACRR), and the ACRR's Fueled-Ring External Cavity (FREC) [3].

Equipment

The instrument used for the EPR measurements is a JEOL Ltd. JES-RE1X ESR Spectrometer, equipped with a high-sensitivity cylindrical microwave cavity. Other manufacturers also produce EPR spectrometers that would be suitable for this work. The spectrometer magnets have a diameter of 150 mm and a pole gap of 60 mm, and are capable of a magnetic field strength of 650 mT. The spectrometer operates at a microwave frequency from 8.8 to 9.6 GHz (X-band) and power up to 200 mW. Output of the EPR spectrometer is sent to a personal computer where it is digitized, analyzed and stored. An RS-232 interface in the spectrometer enables the computer both to define and to read spectrometer settings. The software used for the data acquisition and analysis is an adaptation of EPRWare [4] which has been modified to allow for automated data acquisition and analysis. A precision synthetic-quartz tube and positioning rod ensure reproducibility in pellet position within the microwave cavity to within 0.02 mm vertically and 0.1 mm horizontally. There are no contributions to the measured EPR signals from the quartz fixtures.

Dosimeter Preparation

Crystals of L- α -Alanine (purity >99.5%) are ground and sieved to particle sizes smaller than 100 microns to ensure random orientation of the crystals in the pressed pellets. The resulting powder is mixed with a binder of polyethylene and stearic acid (both smaller than 100-micron particle size), in the ratio of 80 percent by weight of alanine, 19 percent by weight of polyethylene, and 1 percent by weight of stearic acid. Cylindrical pellets are formed by pressing 100 mg of the mixture in a hand tableting press, with a resulting pellet diameter of 4.5 mm and height of 5.2 mm. The pellets are sorted by mass to ensure uniformity to within ± 1 mg. Hardening of the pellets is done by annealing in a Pyrex dish at 130°C for 30 minutes, allowing the annealing oven to cool slowly to 85°C and holding for 5 minutes, and finally removing the pellets and allowing them to cool at room temperature. The pellets are inspected for flakes or chips and any unsatisfactory pellets are discarded.

Automation

A large number of dosimeters are involved in a systematic investigation of optimum spectrometer settings, data analysis algorithms, and characterization of dosimeters in various radiation fields. Furthermore, there is a significant amount of tedium involved in the measurement of the EPR spectrum for a pellet, including placement of the pellet in the microwave cavity, acquisition of the EPR spectrum, determination of the signal intensity either by measurement of the signal peak amplitude or by integration of the EPR spectrum, and storage of the spectrum and analysis results. This process can take more than two minutes per sample. As a result, some automation of data acquisition and analysis is essential, although full automation is not possible, or even desirable, during the development stages of a new system. Areas that are candidates for automation include sample handling, instrument control, and data analysis.

A sample changing mechanism has been constructed for automatic handling of up to 50 pellets and is currently undergoing testing and refinement. This sample changer operates under computer control, so that coordination with data acquisition, analysis and storage is possible. Automation of sample handling is essential for unattended operation, but is practical only for a standard sample configuration, i.e., pellet

dimensions and mass. In addition, the pellet design must be consistent with the technique used for sample handling as well as the environments in which the dosimeters will be used. For this reason, the Radiation Metrology Laboratory (RML) has standardized its pellet dimensions without a comprehensive optimization process.

Because the system will be used over a large range of absorbed dose, it is useful to allow the system to determine for itself the most appropriate gain settings for the spectrometer. A preliminary scan of an alanine EPR spectrum is done and the signal checked to determine if it lies within an acceptable range, e.g., 5 to 90 percent of full scale. If the signal falls outside that range, the gain is changed by a factor of 10, and the preliminary scan is repeated. This process is continued until the signal level is acceptable or until an upper or lower gain limit on the spectrometer is reached. A final scan is then recorded. The EPR spectrometer is capable of a range of sweep times. Preliminary scans can be done with sweep times on the order of a few seconds, but the final scans must be long enough that the magnet can respond adequately without distortion of the EPR signal.

Data acquisition and storage were easily automated, because these functions are the same from one sample to the next, differing only in parameters such as sample identification and file name. Data analysis functions common to all samples, such as baseline adjustments and determination of peak amplitude and spectrum integrals, have also been automated. Conversion of EPR signals to absorbed dose, although not difficult, is not as straightforward because it requires comparisons to previous measurements for calibration dosimeters. This aspect of automation has been deferred until the system has been more fully developed.

Measurement Parameters

The measurement parameters shown in Table 1 were found to give acceptable performance for the measurement of EPR signals in the RML alanine pellets. Effects of some parameters such as microwave power level and field modulation width are still being investigated.

Table 1. EPR Measurement Parameters

Microwave Frequency	9.347 GHz
Magnet Center Field	332.5 mT
Magnet Field Sweep Width	± 10 mT
Field Modulation	100 KHz
Field Modulation Width	0.5 mT
Receiver Gain	Variable, 1-1000
Time Constant	0.03 sec
Sweep Time	1 min
Microwave Power	0.8 mW

Typical EPR Spectrum

Figure 1 shows a typical EPR spectrum for irradiated polycrystalline alanine. This curve is the first derivative of the resonance absorption spectrum and is output directly by the EPR spectrometer. There are no contributions to the spectrum from the pellet binder materials. Both the double integral of the EPR spectrum (total absorbance) and the amplitude of the primary peak in the differential spectrum are proportional to the number of free radicals produced in the alanine. Either value may be used as a measure of absorbed dose.

Gamma Response Curve Characteristics

The gamma response curve is determined by irradiation of alanine pellets to known absorbed doses in the Gamma Irradiation Facility (GIF) at Sandia National Laboratories [3]. Irradiations are done at several levels of absorbed dose per decade from 10 to 10^5 Gy, with four pellets used at each level. Following the irradiations, the EPR spectrum for each pellet is recorded, using the automated measurement techniques previously described. In all cases, a single EPR scan for each pellet is used.

Response data are normalized to absorbed dose, receiver gain, field modulation width and square root of the microwave power level. Gamma response data for field modulation widths of 0.5 mT and 0.05 mT are shown in Table 2. Both sets of data are for a microwave power level of 0.8 mW and are obtained from the same pellets. The table entries are the averages of four pellets at each dose point. The data are also shown in Figure 3 for peak-to-peak data and in Figure 4 for integral data. As can be seen from the data, peak-to-peak data are considerably better than integral data at low dose (high gain), both in the linearity of the curve and in the uniformity of response among the four pellets. At high absorbed doses, however, peak-to-peak data suffers from saturation effects that are not as prominent in the integral data. In fact, valid peak-to-peak data are not obtainable for doses above about 1.5×10^4 Gy with the measurement parameters given in Table 1.

Additional decreases in the system sensitivity can be accomplished either by decreasing the field modulation width or by decreasing the microwave power level. Either change results in worsening of the signal-to-noise ratio for both peak-to-peak and integral data and affects significantly the data at low absorbed doses. This is clearly illustrated in Figure 2, which shows an EPR spectrum using a field modulation width of 0.05 mT for an absorbed dose of 10 Gy. Integral data become quite unstable because even small variations in EPR spectrum baseline contribute to the integral. Although peak-to-peak data are not affected by small variations in baseline, instrumentation noise contributes directly to the peak-to-peak value and results in overestimation of the EPR response at low signal-to-noise ratios. However, peak-to-peak data at high absorbed doses become usable. These effects are seen in Figures 3 and 4, which show data for field modulation width of 0.05 and for microwave power level of 0.01 mW. For the purposes of this work, data are taken at a microwave power of 0.8 mW and a field modulation width of 0.5 mT. For measurements at high absorbed doses, the field modulation width is changed rather than microwave power level. The use of integral data appears to offer no advantage over peak-to-peak data in this region. Peak-to-peak data are therefore used for this work.

Figures 3 and 4 show discontinuities in the data that are well outside the standard deviation with which alanine dosimeters can be read. These discontinuities coincide with changes in system gain and are believed

Table 2. Normalized EPR Response for Field Modulation Widths of 0.5 mT and 0.05 mT

ABSORBED DOSE (GY)	FIELD MODULATION WIDTH = 0.5 MT					FIELD MODULATION WIDTH = 0.05 MT				
	RECEIVER GAIN	PEAK-TO-PEAK		INTEGRAL		RECEIVER GAIN	PEAK-TO-PEAK		INTEGRAL	
		RESPONSE	%SD	RESPONSE	%SD		RESPONSE	%SD	RESPONSE	%SD
10	100	6.457E-1	2.64	5.618E+4	17.71	1000	1.074E+0	5.56	4.599E+4	19.03
20	100	6.465E-1	2.19	5.134E+4	21.11	1000	9.622E-1	2.13	6.656E+4	5.80
40	100	6.402E-1	1.17	5.185E+4	3.05	1000	8.842E-1	0.43	5.878E+4	9.21
100	100	6.341E-1	0.45	5.367E+4	0.73	1000	8.744E-1	1.25	6.284E+4	4.13
200	10	6.445E-1	0.73	5.315E+4	2.28	100	8.647E-1	1.10	6.204E+4	1.68
400	10	6.437E-1	0.18	5.269E+4	0.94	100	8.567E-1	0.86	6.158E+4	0.38
1000	10	6.132E-1	0.59	5.040E+4	0.38	100	7.664E-1	0.52	5.845E+4	0.44
2000	1	6.339E-1	0.29	5.125E+4	0.55	10	8.368E-1	0.49	5.930E+4	0.88
4000	1	6.260E-1	0.83	5.082E+4	1.60	10	8.184E-1	0.33	5.826E+4	1.07
10000	1	5.822E-1	0.40	4.824E+4	0.27	10	7.167E-1	0.89	5.505E+4	0.43
20000	1	4.578E-1	0.00	4.445E+4	0.77	1	6.985E-1	1.11	5.191E+4	2.08
40000	1	---	---	3.507E+4	0.76	1	5.805E-1	0.46	4.477E+4	0.85
70000	---	---	---	---	---	1	4.582E-1	0.47	3.739E+4	0.47
100000	---	---	---	---	---	1	3.771E-1	0.74	3.193E+4	1.63

to be the result of system nonlinearities or changes in measurement parameters. They do not affect the results of the current work, but will impact efforts to automate calculations of absorbed dose from EPR response data. This is still being investigated.

Photon Energy Dependence

In order to verify the energy response of the alanine dosimeters for photon irradiations, calculations and measurements of absorbed dose were performed for several known photon environments. These included the GIF ^{60}Co facility, the HERMES-III bremsstrahlung X-ray facility [3] operating at endpoint energies of 9 and 18 MeV, and the SPHINX bremsstrahlung X-ray facility operating at an endpoint energy of 2.5 MeV. Each dosimeter packet consisted of four alanine pellets and four $\text{CaF}_2\text{:Mn}$ thermoluminescence dosimeters (TLDs) placed in cavities within a LuciteTM equilibrator. The thickness of the LuciteTM surrounding the dosimeters is 6 mm for the GIF and SPHINX irradiations, and 30 mm for both the 18-MeV and the 9-MeV HERMES-III irradiations. These thicknesses were chosen to ensure electron equilibrium for the alanine dosimeters in the respective radiation fields. Measured absorbed doses range from approximately 5 Gy to 5000 Gy, depending on the capabilities of the photon sources. Calculations include fully-coupled electron-photon transport and are done with the ADEPT (CEPXS) discrete-ordinates code [5] using a one-dimensional model of the respective dosimeter packets. Figure 5 shows the energy-dependent photon keramas for alanine and $\text{CaF}_2\text{:Mn}$ TLD, derived from the energy-transfer cross sections from Biggs and Lighthill [6]. Figure 6 shows the assumed spectra for the several environments used in this study.

Comparisons between alanine dosimeters and the traditional $\text{CaF}_2\text{:Mn}$ TLDs are shown in Table 3, where calculated and measured ratios of absorbed dose are given for the alanine pellets to the $\text{CaF}_2\text{:Mn}$ TLDs in their respective equilibrators. Estimated uncertainties in the measured ratios, based on uncertainties in the dose measurements and scatter in multiple measurements for a single environment, are on the order of ± 10 -12%. Estimated uncertainties in the calculated dose ratios are on the order of ± 8 -10%, and are due primarily to uncertainties in the assumed photon spectra. Good agreement between calculated and measured values for the GIF is expected because this facility is used for calibration of both the TLDs and the alanine dosimeters. The difference between calculated and measured dose ratios for the GIF facility is consistent with the precision with which the measurements can be made. The differences between the calculated and measured dose ratios for the bremsstrahlung facilities exceed that which might be expected from the uncertainty estimates. The reasons for this are still being investigated, but might include inadequate modeling of source electrons or unidentified modeling errors in the calculation of the photon spectra, especially at low energies.

Table 3. Ratios of Absorbed Dose for Photons

ENVIRONMENT	RATIO, DOSE(ALANINE) TO DOSE(TLD)	
	CALCULATED	MEASURED
GIF (^{60}Co)	1.08	1.12
HERMES-III (18 MeV)	0.90	1.21
HERMES-III (9 MeV)	0.96	1.38
SPHINX (2.5 MeV)	0.80	0.96

Neutron Energy Dependence

Calculations and measurements of absorbed dose were made for several mixed neutron/photon environments, including the central cavity of the SPR-III, the central cavity of the ACRR, and the ACRR FREC fitted with a cadmium and polyethylene liner. This selection of facilities provides a wide range of neutron-to-gamma ratios. Because the alanine sensitivity to neutrons is significantly larger than that for the $\text{CaF}_2\text{:Mn}$ TLDs, there are substantially different ratios of alanine dose to TLD dose for the various environments. The relative effectiveness for neutrons in alanine has been reported in the literature [7]. The neutron response was determined from first principles with track-structure calculations and was in excellent agreement with experimental measurements for a wide range of neutron energies. The energy dependence of the neutron

effectiveness is shown in Figure 7. The relative effectiveness of neutrons in $\text{CaF}_2\text{:Mn}$ TLDs is very low. The energy-dependent efficiency can be modeled as the neutron energy (in MeV) divided by 100 [8]. For a fission neutron spectrum this results in an efficiency of approximately 3% of the neutron absorbed dose. Neutron spectra for the three environments have previously been experimentally determined [9], while photon spectra were assumed to be that for fission gammas. Energy-dependent neutron kerma for the alanine dosimeters and the TLDs were derived from ENDF/B-VI cross sections processed with NJOY [10], and are shown in Figure 8. The larger neutron kerma for alanine at higher energies is mostly due to the (n,p) reaction in hydrogen. At lower neutron energies, the higher neutron kerma is due to the large cross section for the $^{14}\text{N}(\text{n},\gamma)^{15}\text{N}$ reaction. The absorbed dose in the dosimeter attributable to neutrons is obtained by weighting the energy-dependent kerma with the relative effectiveness for the neutrons, and folding that with the neutron spectrum. The total dose is the sum of the photon and effective neutron absorbed doses.

Table 4 shows the calculated and measured ratios of absorbed dose for the alanine pellets to the $\text{CaF}_2\text{:Mn}$ TLDs for the three neutron environments. The table also shows neutron-to-gamma ratios for those environments. Estimated uncertainties in the measured dose ratios are on the order of $\pm 10\text{-}12\%$. Estimated uncertainties in the calculated dose ratios are on the order of $\pm 10\%$. As expected, the ratio of alanine dose to TLD dose decreases as the neutron-

Table 4. Ratios of Absorbed Dose for Neutrons

ENVIRONMENT	NEUTRON TO GAMMA RATIO	RATIO, DOSE(ALANINE) TO DOSE(TLD)	
		CALCULATED	MEASURED
SPR-III Cavity	1.45	3.77	4.28
ACRR Cavity	0.57	1.68	2.13
FREC, Cd/Poly	0.19	1.31	1.42

to-gamma ratio decreases because of the higher neutron response in alanine. The calculated dose ratios have the same general behavior as the measured dose ratios. The calculated ratios all assume a prompt-fission-gamma spectrum, which would be representative of the SPR-III cavity. For this environment, the calculated dose ratio is about 14% lower than the measured ratio. The ACRR pool-type reactor actually has a significantly softer, degraded, photon component. The difference between calculated and measured ratios for the ACRR cavity is about 27%. The FREC Cd/Poly environment has a harder, higher-energy photon component that results from (n, γ) reactions in cadmium by low-energy neutrons. The difference between calculated and measured ratios for the FREC is about 8%. The reasons for the correlation between the dose ratio and photon energy are being investigated. Detailed Monte Carlo calculations are being performed to provide a better quantitative model of the actual photon spectra in these environments and to provide a better foundation for the assigned neutron-to-gamma ratios.

Conclusions and Recommendations

A system has been successfully developed for the use of EPR spectroscopy with L- α -alanine for high-precision absorbed-dose measurements. The system has been sufficiently automated to allow a systematic optimization of measurement parameters. A relatively small effort will be required to extend the application of this system to routine measurements in photon environments.

The calculated and measured alanine-to-TLD dose ratios were in acceptable agreement for the neutron environments, but not for the photon environments. However, the calculations for the neutron environments depend on calculated photon sensitivities. Using the measured alanine-to-TLD dose ratios for the photon environments rather than the calculated ratios would improve further the agreement between calculated and measured ratios for the neutron environments. The issue of photon spectrum sensitivity is unresolved and work is continuing.

The use of alanine for dosimetry in mixed neutron/photon environments is recommended if the experiment requires tissue-equivalent response. However, alanine should not be used for silicon-equivalent dose

measurements (e.g., testing of electronics) because of the very large neutron contribution to the measured dose, which would require large corrections to obtain a silicon dose. Fielding TLDs and alanine together in mixed fields would be quite useful in establishing neutron-to-gamma ratios and in characterizing photon environments.

Investigation of alternate materials for silicon-dose-equivalent measurements in mixed fields should be pursued. For example, the ratio of neutron kerma to photon kerma for Teflon™ is much closer than alanine's to that of silicon. However, the energy-dependence of the neutron relative efficiency has not yet been established for Teflon™.

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Figure 1. Typical Alanine EPR Spectrum

Figure 2. EPR Spectrum Using FMW=0.05 mT, Absorbed Dose = 10 Gy

Figure 3. Normalized EPR Response, Peak-to-Peak Method

Figure 4. Normalized EPR Response, Double-Integral Method

Figure 5. Energy-Dependent Photon Kerma for Alanine and $\text{CaF}_2:\text{Mn}$

Figure 7. Relative Effectiveness for Neutrons in Alanine

Figure 6. Assumed Photon Spectra

Figure 8. Energy-Dependent Neutron Kerma for Alanine and $\text{CaF}_2:\text{Mn}$

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