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An experimental and Monte Carlo investigation of the energy dependence of alanine/EPR dosimetry: II. Clinical electron beams

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Abstract

The energy dependence of alanine/EPR dosimetry for 8, 12, 18 and 22 MeV clinical electron beams was investigated by experiment and by Monte Carlo simulations. Alanine pellets in a waterproof holder were irradiated in a water phantom using an Elekta Precise linear accelerator. The dose rates at the reference point were determined following the TG-51 protocol using an NACP-02 parallel-plate chamber calibrated in a ⁶⁰Co beam. The EPR spectra of irradiated pellets were measured using a Bruker EMX 081 EPR spectrometer. Experimentally, we found no significant change in alanine/EPR response to absorbed dose-to-water over the energy range 8-22 MeV at an uncertainty level of 0.6%. However, the response for high-energy electrons is about $1.3 (\pm 1.1)\%$ lower than for ⁶⁰Co. The EGSnrc Monte Carlo system was used to calculate the ratio of absorbed dose-to-alanine to absorbed dose-to-water and it was shown that there is 1.3 (± 0.2)% reduction in this ratio from the ⁶⁰Co beam to the electron beams, which confirms the experimental results. Alanine/EPR response per unit absorbed dose-to-alanine was also investigated and it is the same for high-energy electrons and 60 Co γ -rays.

1. Introduction

Alanine/EPR has been used in radiation processing for over 20 years at high dose levels in food irradiation, radiation processing and medical sterilization, mainly for γ -ray irradiations (Regulla 2000). The principle is based on a measurement of the radiation-induced free radicals in alanine dosimeters by means of electron paramagnetic resonance (EPR) (Raymond 1968). The measurement accuracy of this system is generally good due to the low dependence of

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the alanine response on irradiation variables, such as energy, dose rate, temperature and the high precision capabilities of EPR spectrometers. In recent years, alanine/EPR dosimetry has been applied to lower, clinical doses (Nagy *et al* 2002, Fainstein *et al* 2000, Hayes *et al* 1999, Ruckerbauer *et al* 1996, Sharpe *et al* 1996). The measurement precision for EPR signals can reach 1% (1σ) or better for doses higher than 10 Gy.

The response of the alanine/EPR system to MV photon beams has been reported in our recent paper (Zeng *et al* 2004, referred to as paper I) and in other studies (Sharpe 2003, Bergstrand *et al* 2003, Olsen *et al* 1990). All these studies show that alanine response to MV linac photon beams is not energy dependent within 1%, but it is about 0.6% lower than for 60 Co beams.

There are a few published studies focusing on the use of alanine in radiation therapy with electron beams. In De Angelis et al's (2000) work, alanine was applied in electron arc therapy for treatment planning verification and the alanine-determined doses were in very good agreement with the ion-chamber-determined doses. Their investigation also shows no change in the response per unit absorbed dose-to-water of alanine pellets (alanine/polyethylene) in the energy range 4-16 MeV. Ciesielski et al (1993) studied the energy dependence of an agar-alanine phantom (alanine crystals embedded in an agar matrix) by using Burlin's cavity theory and they estimated a variation of less than 2% from 150 keV to 20 MeV. Olsen et al (1990) experimentally measured the response per unit absorbed dose-to-water of alanine pellets (alanine/polyvinyl pyrrolidone) for electron beam energies between 4 and 16 MeV and observed no variation within 0.6%. Onori et al (1990) also reported a flat energy response for electrons in the energy range 10-28 MeV and the same calibration factor with ⁶⁰Co by using Fricke dosimetry as the reference. Their dosimeter material was alanine/paraffin. For calibration purposes, Sharpe and Burns (1995) compared the relative response of alanine/paraffin dosimeters to high-energy electrons and ⁶⁰Co together with Fricke and dichromate dosimeters and concluded that the response of alanine dosimeters is not significantly different for ⁶⁰Co and high-energy electrons.

With the potential for more alanine applications with clinical electrons, it is valuable to study the energy dependence of alanine with a high measurement and calculation accuracy. In the present work, we investigated four clinical electron energies from 8 MeV to 22 MeV and measured the alanine response per unit absorbed dose-to-water for each beam relative to the reference ⁶⁰Co beam. We carried out Monte Carlo calculations of the absorbed dose-to-alanine per unit absorbed dose-to-water. From measurements and Monte Carlo simulations we derived the alanine/EPR response per unit absorbed dose-to-alanine for electron beams and for ⁶⁰Co γ -rays. The effects of the dosimeter holder on the energy dependence and calibration correction factors were estimated.

2. Methods

The energy dependence of alanine/EPR dosimetry is defined as the variation of alanine/EPR response per unit absorbed dose-to-water versus beam energy. We used the same method as described in paper I to investigate the energy dependence. The alanine/EPR response per unit absorbed dose-to-water for a beam quality Q was derived by the slope of the calibration curve, denoted as $(Slope)_Q$ in paper I. For this investigation of the energy dependence of electron beams, we compared the values of $(Slope)_Q$ by normalizing them to the 12 MeV reference beam. For the investigation of calibration factors relative to ${}^{60}Co \gamma$ -rays, the $(Slope)_Q$ of electron beams were normalized to that of ${}^{60}Co$.

Equation (5) in paper I was applied to calculate the yield of free radicals per unit absorbed dose-to-alanine for electron beams relative to ⁶⁰Co. The relative yield was obtained in terms of

Table 1. Values used for k'_{R50} and reference depths in water, d_{ref} , derived from R_{50} , following the TG-51 protocol and using a 10 cm × 10 cm applicator and SSD = 100 cm. The thickness of the PMMA entrance window of the water phantom was included. For the dose determination, the value of $N_{D,w,C0}$ for the NACP chamber is 16.28 cGy/nc and the value of k_{ecal} is 0.883.

Energy (MeV)	8	12	18	22
<i>R</i> ₅₀ (cm)	3.18	4.73	7.13	8.98
$d_{\rm ref}$ (cm)	1.81	2.74	4.18	5.29
k'_{R50}	1.038	1.022	1.003	0.992

 $\frac{(\text{Slope})_Q}{(\text{Slope})_{Co}} / \frac{(D_{\text{alanine}}/D_{\text{water}})_Q}{(D_{\text{alanine}}/D_{\text{water}})_{Co}}$, with D_{alanine} , and D_{water} being the calculated absorbed dose-to-alanine and the absorbed dose-to-water, respectively.

3. Experiment

3.1. Material and instruments

Alanine dosimeters in the form of pellets were obtained from Gamma-service Produktbestrahlung GmbH, Germany, Lot T020604. The composition was stated to be 96% L-alpha-alanine and 4% other binder material (mostly polyvinyl pyrrolidone). Electron beams were delivered by an Elekta Prec*i*se medical linac, which offers five nominal energies at 4, 8, 12, 18 and 22 MeV. ⁶⁰Co γ -rays were from an AECL Eldorado-6 therapy unit. Spectrum acquisition of irradiated pellets was performed using a Bruker EMX 081 EPR spectrometer.

3.2. Irradiations

Four electron beams were used for the investigation—8, 12, 18 and 22 MeV. Measurements for the 4 MeV beam were not carried out due to the increased uncertainty in positioning and the significant dose gradient effect in the alanine dosimeters at this energy. The irradiations were carried out in a water phantom with a horizontal beam configuration. The SSD was 100 cm and the field size, shaped by a standard applicator, was $10 \text{ cm} \times 10 \text{ cm}$ at the entrance window of the water phantom. The thickness of this entrance window was taken into account when determining the reference depth for each energy. The reference depths for the four electron beams were defined by using the AAPM's TG-51 protocol (Almond *et al* 1999) and are listed in table 1. The linac was operated at its maximum dose rate, which gives approximately 5 Gy min⁻¹ at the reference depth.

A parallel-plate NACP-02 chamber, calibrated in terms of absorbed dose-to-water in a ⁶⁰Co beam, was used to determine the dose at the reference point, following the TG-51 protocol (except that a revised value of the photon–electron conversion factor, $k_{ecal} = 0.883$, from Mainegra-Hing *et al* (2002), was used). TG-51 prefers that parallel-plate chambers not be directly calibrated in a ⁶⁰Co beam, but be cross-calibrated against a cylindrical chamber in a high-energy electron beam. Both methods were investigated (using a NE2571 cylindrical chamber) and it was found that the dose determined using the cross-calibration route was less than 0.3% lower than using the direct calibration of the NACP chamber (which implies that the new value of k_{ecal} leads to more consistent dose estimates than the previous value). Since the two methods agree with each other, the direct method (i.e. calibration of the NACP chamber in ⁶⁰Co) was chosen as it simplifies the uncertainty analysis (see below).

For the electron measurements, depth-ionization curves were obtained prior to the irradiations to determine R_{50} , the depth at which the absorbed dose falls to 50% of its

	Energy (MeV)				
	8	12	18	22	
Components of uncertainty		Type A			Type B
Chamber reading	0.09	0.09	0.09	0.09	0.08
Repeatability (linac drift)	0.08	0.03	0.10	0.03	
Run length variation	0.10	0.10	0.10	0.10	
Polarity correction	0.05	0.05	0.05	0.05	
Recombination correction					1.10
Beam uniformity	0.02	0.02	0.02	0.02	
$N_{D,W,Co}$ (repeatability)	0.05	0.05	0.05	0.05	
k'_{B50}					0.40
k _{ecal}					0.98
Combined uncertainty	0.17	0.15	0.18	0.15	0.99 ^a
Overall uncertainty in dose relative to ⁶⁰ Co ^a	1.00	1.00	1.00	1.00	
Overall uncertainty in relative e-doses ^b	0.45	0.45	0.46	0.45	

Table 2. Standard uncertainties (%) in the determinations of absorbed dose-to-water in electron beams relative to the dose in the ⁶⁰Co beam using an NACP chamber calibrated in the ⁶⁰Co beam.

^a Includes uncertainty in the product $k'_{R50} k_{ecal}$ which is equivalent to the uncertainty on k_{ecal} alone bacause of cancellation of factors.

^b Includes the uncertainty in k'_{R50} .

maximum, and thereby to derive the reference depth, $d_{\text{ref}} = 0.6R_{50} - 0.1$ cm. McEwen and Ross (2003) have shown that for this linac, the electron energy is stable from day to day to better than the 1% level (in terms of R_{50}), which can lead to a maximum error of 0.2% in the dose (due to incorrect positioning and assignment of the k_Q factor). A second NACP chamber was used at each energy as a check on the performance of the calibrated chamber. The ratio of NACP readings was constant at the $\pm 0.1\%$ level and showed no trend with energy. All chamber readings in electron beams were corrected for recombination and polarity as well as temperature and pressure. The recombination correction was around 0.5% and the polarity correction was about 0.05% at all energies.

In TG-51, the dose measured by the NACP parallel-plate chamber is given by

$$D_w = M \cdot N_{D,w,\text{Co}} \cdot k'_{R50} \cdot k_{\text{ecal}},\tag{1}$$

where *M* is the chamber measurement, $N_{D,w,Co}$ is the calibration coefficient in ⁶⁰Co determined by water calorimetry, k'_{R50} is the electron quality conversion factor and k_{ecal} is the photon– electron conversion factor which converts the absorbed dose calibration coefficient in a ⁶⁰Co beam to that in a high-energy electron beam. The values of factors used in the present work are presented in table 1.

We estimated the uncertainty of the assigned electron beam doses by following the procedures in the IAEA Code of Practice, TRS-398 (2000), i.e. we have estimated the uncertainty on each of the factors that appear in equation (1), including the individual factors needed to calculate k'_{R50} and k_{ecal} . Our values are based on our best estimates using the most updated data and are presented in table 2. There are two overall uncertainties of interest in this work. When we are comparing the relative alanine response in electron beams, we are only sensitive to the uncertainty in k'_{R50} since the factor k_{ecal} is common in all the dose determinations. In contrast, when we are interested in the alanine response in electron beams relative to that in ⁶⁰Co, we are interested in the product of $k'_{R50} k_{ecal}$, but when evaluating the uncertainty on the product, it is basically the same as the uncertainty on k_{ecal} itself due to



Reference depth

Figure 1. Positioning of alanine dosimeters in the water phantom relative to the beam direction. Six pellets (5 mm diameter, 3 mm in height) were placed symmetrically about the central axis in a disc-like holder (diameter of 31 mm), which was made of Virtual WaterTM ($\rho = 1.03 \text{ g cm}^{-3}$). Pellets were placed side by side (2 mm separation) and centred 7 mm inside the holder rim. The thickness of the holder's upstream wall was 1.8 mm. The geometrical centre of pellets was positioned at the reference depth for each electron energy with the SSD = 100 cm and a field size 10 cm × 10 cm. The beam direction was perpendicular to the flat surface of alanine pellets.

cancellation of factors in the product. These uncertainties for use in different situations are presented in table 2.

For the alanine irradiations, six pellets were placed side by side in a holder with the flat surface of the pellets facing the beam direction (figure 1). The holder was made of Virtual WaterTM with a density of 1.03 g cm⁻³. This material is designed to be water equivalent in electron beams and measurements have confirmed this (McEwen and Niven 2003). For the purposes of this investigation, the 1.8 mm front wall of the holder was taken to be 1.8 mm of water. The disc-shaped design of the holder was chosen so that the alanine pellets sampled the same part of the electron beam as the parallel-plate ion chamber. Also, the similarity in the shapes of the chamber and holder meant that any systematic error in the positioning would not have an effect on the investigation. The geometrical centre of the alanine pellet was taken to be the effective point of measurement and was positioned at the reference depth. For all beams it was found that at the reference depth, the depth–dose curve was either flat or linear at the 0.1% level and therefore no dose gradient correction was required. A small correction for radial non-uniformity was required to correct for the different sizes and positions of the NACP chamber and alanine pellets. This correction was largest at 8 MeV where it was 0.15%.

The linac monitor was used to transfer the dose from the ion chamber to the alanine. For each set of alanine irradiations at one energy, the monitor was calibrated before and afterwards using the ion chamber and any drift in calibration was taken into account. The maximum drift observed during these measurements was 0.1%.

For irradiations in ⁶⁰Co, the same dosimeter holder was used and the beam direction was also perpendicular to the flat surface of the dosimeters. The dose rate at the reference depth of 5 cm of water plus 3 mm of PMMA entrance window in the 10 cm \times 10 cm field has been previously determined using the primary standard sealed water calorimeter at the National Research Council of Canada (NRC) (Seuntjens *et al* 1999). NRC-calibrated ion chambers are used regularly to monitor the output from the source with a precision of $\pm 0.05\%$. The dose rate at the time of the alanine irradiations was about 0.5 Gy min⁻¹.



Figure 2. Ratios of mass collision stopping powers for the pellet material (4% polyvinyl pyrrolidone) to pure alanine. Density-effect corrections for the pellet material were calculated using the ESTAR code (Berger *et al* 1996) and assuming either the bulk density of the pellet (1.22 g cm^{-3} , triangles) or the crystalline density of the alanine in the pellet (1.42 g cm^{-3} , circles). In this work, we use the crystalline density values since experiments with graphite indicate that one should use the crystalline data.

The alanine pellets were irradiated to 4 dose points, 20, 30, 40 and 50 Gy in each beam of interest. The irradiation temperature was maintained at 21 ± 1 °C.

3.3. EPR measurements and uncertainty estimation

A Bruker EMX 081 spectrometer was used to measure the EPR signals of the irradiated pellets. The characteristics of this alanine/EPR dosimetry system, the measurement methods and the uncertainty estimation have been described in paper I. The uncertainty of each slope value was calculated by including both the EPR measurement uncertainties and the dose determination uncertainties. Table 2 shows that the type B uncertainties in the determination of the absorbed dose-to-water for electron beams relative to that of ⁶⁰Co are higher than the type A uncertainties, therefore, for the energy dependence investigations of electrons, we used the 12 MeV electron beam as the reference, in order to avoid the high type B uncertainties in k_{ecal} . However, when comparing results in ⁶⁰Co and electron beams, i.e. for the alanine response relative to ⁶⁰Co, the type B uncertainties in k_{ecal} were included.

4. Monte Carlo simulations

The Monte Carlo simulations were done using the EGSnrc user-code DOSRZnrc (Kawrakow and Rogers 2000, Rogers *et al* 2001). The user codes in EGSnrc require a geometry with cylindrical symmetry, so a simplification of the dosimetry holder was made, with a single pellet located at the centre of a cylindrical disc-like holder.

We used the NIST ESTAR code (Berger *et al* 1996) to calculate the density-effect correction for the mass collision stopping power of the alanine pellet. The pellet manufacturing procedure introduces voids between alanine crystals, so the bulk density of our pellets is 1.22 g cm^{-2} , much less than the crystalline density of 1.42 g cm^{-3} . The difference in collision stopping power when the bulk density-effect correction is used compared to when the crystalline density effect is used is shown in figure 2. For the energies of interest in this study



Figure 3. Ratios of mass energy absorption coefficients, μ_{en}/ρ , of alanine to water and of dosimeter material to water obtained from MassEn program (http://dax.northgate.utah.edu/MassEn.html) based on NIST cross sections (Hubbell and Seltzer 1995) and ratios of restricted mass collision stopping powers, $dT/\rho dx$ as used in the present calculations. Solid lines are for the ratios of pure alanine to water while the dotted lines are for pellet material (4% polyvinyl pyrrolidone) to water.

the difference is of the order of 0.7-0.9%. A previous study of the stopping powers for graphite indicated that the stopping powers calculated using the crystalline density-effect correction are consistent with the measured stopping powers, while the stopping powers using bulk density-effect correction induce an overestimate of about 1% for MeV electrons (MacPherson 1998, MacPherson *et al* 1998). Therefore, in this study, we applied the crystalline density-effect correction for alanine pellets in the MC simulations while using the correct bulk density for the pellet.

PEGS4 was used for cross section data in the calculations with AE = 0.521 MeV and AP = 0.001 MeV. The ratios of restricted mass collision stopping powers, $dT/\rho dx$, and mass energy absorption coefficients, μ_{en}/ρ , of alanine or dosimeter to water are plotted in figure 3. The spectra of electrons from the NRC Elekta accelerator plotted in figure 4 were generated by using the BEAM Monte Carlo code (Rogers *et al* 1995).

5. Results and discussion

5.1. Energy dependence of alanine response for electrons

The ratios of the alanine/EPR response per unit absorbed dose-to-water in electron beams normalized to that for a 12 MeV beam are listed in table 3. No significant variation in the ratios was found at an uncertainty level of 0.6%, indicating that the alanine/EPR response per unit absorbed dose-to-water does not depend on the energy of the electron beam. The lack of energy dependence for electron beams found in this study is consistent with the previous studies of De Angelis *et al* (2000), Olsen *et al* (1990), Onori *et al* (1990) and Sharpe and Burns (1995), although the dosimeter compositions used in those studies are different.

5.2. Electron beam response relative to ⁶⁰Co response

Table 3 also gives the alanine/EPR response per unit absorbed dose-to-water in electron beams relative to that in a 60 Co beam. On average, the response for electrons is about 1.6% (±1.1%)



Figure 4. Spectra at the central axis of 8, 12, 18, 22 MeV electron beams from the Elekta precise medical linac generated by BEAM Monte Carlo code (Rogers *et al* 1995). Field size $10 \text{ cm} \times 10 \text{ cm}$ on the phantom surface with SSD 100 cm.

Table 3. Energy dependence of alanine/EPR response per unit absorbed dose-to-water. For the relative response in electron beams, $(Slope)_Q/(Slope)_{12 \text{ MeV}}$, the uncertainty was calculated by taking account the statistical uncertainty of linear fitting, including the EPR measurement uncertainties, as well as the uncertainties in absorbed dose-to-water determinations for 8, 18, 22 MeV beams relative to the 12 MeV beams. For the response relative to the ⁶⁰Co beams, (Slope)_Q/(Slope)_{Co}, the uncertainties of the dose determination relative to ⁶⁰Co in table 2 were used.

Beam energy (MeV)	$(\text{Slope})_Q/$ $(\text{Slope})_{12 \text{ MeV}}$	Uncertainty (%)	$(\text{Slope})_Q/$ $(\text{Slope})_{Co}$	Uncertainty (%)
8	1.001	0.6	0.981	1.0
12	1.000	0.3	0.981	1.1
18	1.009	0.8	0.989	1.1
22	1.004	0.8	0.985	1.1

less than that for ⁶⁰Co. The Monte Carlo simulations showed that the dosimeter holder caused an average reduction of 0.3% in alanine response for electrons due to the fluence change induced by the Virtual Water. There is no impact for ⁶⁰Co. When the effect of the dosimeter holder is included, the correction factor for electrons is 1.3% (\pm 1.1%). Onori *et al* and Sharpe and Burns also compared electrons to ⁶⁰Co and concluded that the calibration factors are the same. However, the error bars in electron dosimetry were in the same magnitude as the correction factor of 1.3%, which may have masked the differences.

5.3. Radiation yield for electrons and $^{60}Co \gamma$ -rays

Monte Carlo simulations were carried out to calculate the ratio of absorbed dose-to-alanine to absorbed dose-to-water under the experimental geometries. The relative radiation yield in electron beams to that in a ⁶⁰Co beam was derived using equation (5) in paper I. Radiation yield relates to the free radical production per unit absorbed energy in the dosimeter. Table 4 displays the results. All data are unity, well within the uncertainty of 1.1%, implying that the



Figure 5. A comparison of measured and Monte Carlo calculated values of the alanine response per unit absorbed dose-to-water for electrons relative to that of 60 Co. The measurement data are corrected for the effect of the dosimeter holder. The dotted line drawn at 0.987 on the *y*-axis indicates the average lower response of 1.3% for electrons relative to 60 Co. Note that the error bars on the Monte Carlo calculations are statistical only.

Table 4. The radiation yield for electrons relative to 60 Co γ -rays. The relative radiation yield is unity within uncertainty, indicating that the radiation yield of free radicals in alanine is the same for 60 Co γ -rays and for high-energy electrons.

Beams (MeV)	Relative radiation yield to ⁶⁰ Co	Uncertainty (%)
8	1.000	1.1
12	0.994	1.1
18	1.004	1.1
22	0.999	1.1

radiation yield of free radicals in alanine is the same for ⁶⁰Co γ -ray beams and for high-energy electron beams.

5.4. Discussion

The most probable electron energy at the reference depth for the four beams investigated is between 4 MeV and 10 MeV. In figure 3, the ratio of stopping power of dosimeter to water is flat within 0.2% over this energy range, so very little energy dependence for electron beams is expected.

For the comparison of alanine response in electron and 60 Co beams, the larger uncertainty in relative ion chamber dosimetry between 60 Co and electron beams limits the conclusions that can be drawn from the analysis. The differences in response observed in our experiment are barely significant, although the fact that all the points in figure 5 are less than unity would suggest a real effect. In the future, when a primary standard for electron beams is established at NRC, we could re-analyse these results with lower experimental uncertainties. However, the Monte Carlo simulations predict that if the radiation yield of alanine is the same for electrons and 60 Co (i.e. the alanine EPR response per unit absorbed dose in the alanine is independent of beam quality) there is about a 1.3% (±0.2%) reduction in alanine response per unit absorbed dose-to-water in electron versus ⁶⁰Co beams. The comparison of measurement and Monte Carlo prediction is shown in figure 5. We offset the calculated and measured data to separate the error bars. The level of agreement is encouraging. The difference in response may be interpreted as the gap between the alanine to water stopping power ratio for high-energy electrons and the alanine to water mass energy absorption ratio for ⁶⁰Co, shown in figure 3.

Alanine dosimeters are available in different compositions—a mixture of alanine and a binding agent. The most commonly used bonding agents are polyvinyl pyrrolidone (PVP), paraffin wax, polyethylene and polystyrene. We carried out Monte Carlo calculations and found that changing bonding agents does not change the lack of energy dependence of alanine dosimeters for MeV electron beams. This would explain why researchers using different compositions of dosimeter report very similar results. However, the paraffin and polyethylene bonding agents may affect the relative response in ⁶⁰Co and electron beams. For example, our calculations predict that 20% paraffin or polyethylene (a very high proportion of binder) may decrease the calibration correction factor from 1.3% to about 1.0%.

6. Conclusions

In this study, we investigated the energy dependence of alanine/EPR dosimetry for clinical electron beams. It was found that alanine/EPR response per unit absorbed dose-to-water at the standard reference depth does not depend on electron beam energy, but it is about 1.3% lower than that for ⁶⁰Co γ -rays. Furthermore, within the experimental and calculation uncertainties, the radiation yield, based on dose-to-alanine, is the same for clinical electron beams as that for ⁶⁰Co γ -rays.

Note added in proof. Since the acceptance of this manuscript, a report of a similar study, 'An experimental investigation of the electron energy dependence of the EPR alanine dosimetry system' by Bergstrand *et al*, has been published in *Radiation Measurements* **39** (2005) 21–8.

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