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Potassium dithionate EPR dosimetry for determination of absorbed dose and LET
distributions in different radiation qualities.

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Abstract

With an increasing interest in using protons and light ions for radiation therapy there is a need for possibilities to simultaneously determine both absorbed dose (D) and linear energy transfer, LET, (L_{Δ}). Potassium dithionate ($K_2S_2O_6$) tablets were irradiated in a conventional 6 MV linear accelerator photon beam and a N^{7+} beam ($E = 33.5$ MeV/u) respectively. The EPR spectrum of irradiated potassium dithionate is a narrow doublet consisting of two signals, R_1 and R_2 , with different microwave power saturation properties. On the basis of identification in related substances by EPR and ENDOR, these two signals are assigned to two non-equivalent SO_3^- -radicals. Our experiments showed that the ratios of these two lines (R_1/R_2) were clearly connected to beam LET. Irrespective of the mechanistic details this investigation suggests a new method for measurement of absorbed dose and beam LET by using potassium dithionate EPR dosimetry.

Keywords: EPR dosimetry, linear energy transfer, LET, charged particles.

1. Introduction

Radiation induced radicals are often very stable in crystalline solid materials. These radicals can be detected and quantified by means of electron paramagnetic resonance, EPR. The signal intensity is proportional to the concentration of paramagnetic species (at least up to kGy doses) and is therefore also proportional to the absorbed dose. Polycrystalline alanine has been used for EPR dosimetry since more than 25 years (Regulla and Deffner 1982) but suffers from low sensitivity for routine applications in radiation therapy. A number of more sensitive dosimeter materials (e. g. salts of formic acid and dithionic acid salts) has therefore been proposed and some of them has been tested for measurements of low doses with enhanced precision (Lund et al. 2002, Gustafsson et al. 2004). The currently most promising material is lithium formate monohydrate (Gustafsson et al. 2008a, Antonovic et al. 2009, Adolfsson et al. 2010, Waldeland et al. 2010a and Waldeland et al. 2010b). It is two to six times more sensitive than alanine depending on the read-out procedure and has a simple, single - lined, EPR spectrum. Dithionates are another proposed type of EPR dosimeter materials that are even more sensitive than lithium formate monohydrate and with remarkable narrow EPR line widths and stable EPR signals due to the irradiation induced cleavage of the S-S bond of the $S_2O_6^{2-}$ ion (Bogushevich and Ugolev 2000, Lund et al. 2002, Danilczuk et al. 2008, Gustafsson et al. 2008b). The radical yield can in some cases be further enhanced by transition metal dopants (Gustafsson et al. 2005). The far most intense absorption line has been identified as due to SO_3^- anion radicals in some cases stabilized at two inequivalent sites (Baran et al. 2006, 2007, 2008). This is similar to the radical formation observed in irradiated caesium dithionate (Mahgoub et al. 1983, 1994).

Several EPR studies have shown that the yield of stable paramagnetic species, for a constant absorbed dose, depends on the linear energy transfer (LET) of the radiation. The yield of radicals and thus the EPR signal intensity is decreasing with increasing LET (e. g. Hansen and Olsen 1989, Olsson et al. 2000, Waldeland et al. 2010b). For determinations of absorbed dose in high-LET radiation fields using EPR dosimetry, the LET-dependence must be taken

into account. For accurate dose determinations the calibration of the dosimeters must either be performed in beams with the same LET as the beam in which the dosimeters are going to be used, or otherwise the signal intensity in the dosimeters must be multiplied with a correction factor to compensate for the differences in radical yield depending on differences in LET between irradiation of calibration tablets and dosimeters.

Ciesielski and Wielopolski (1994) and Ciesielski et al. (1998) have shown that the shape of the EPR spectrum of irradiated alanine depends on the beam quality. In addition to an increased peak to peak height with increasing absorbed dose, they observed a radiation quality dependent change in the relative ratio of two peaks in the alanine spectrum. ENDOR studies have shown that the alanine spectrum consists of the signals from at least three radicals (Sagstuen et al. (1997)). The increased temperature expected in the heavy charged particle tracks (Norman (1967)) might induce changes in the relative amounts of radicals in irradiated alanine, detected as slight changes in the relative ratio of EPR lines in the alanine spectra. However, as pointed out by Malinen et al. (2006), the underlying mechanisms need to be further studied.

Heavy charged particles ionize the irradiated matter more densely along the particle tracks, probably resulting in an increased local radical concentration. It is therefore expected that irradiation with heavy particles also has an influence on the phase memory time, T_m . For purely homogeneously broadened EPR signals this is expected to be seen as increased EPR line widths for samples irradiated with heavy particles as compared to photon irradiated samples. However, it should be noted that changes in T_m may not significantly change the line widths of EPR signals broadened by unresolved hyperfine couplings, i.e. inhomogeneous broadened EPR signals. In literature it has been reported that lithium formate irradiated with neutrons (Malinen et al. 2006) and protons and nitrogen ions (Waldeland et al. 2010b) have a slightly increased line width as compared to samples irradiated with photons. Rakvin et al. (2007) observed a change in T_m using pulsed EPR methods in alanine irradiated with

different ions as compared to photon irradiated samples. Ciesielski and Wielopolski (1994) did however not observe an increased line width after irradiation with heavy charged particles (but they observed an increased line width for high doses above 10 kGy regardless of radiation quality). They concluded that there was no substantial increase in local radical concentration in alanine irradiated with heavy particles as compared to low-LET-irradiation. Recently Marrale et al. (2009) used pulsed EPR methods to measure radical distributions in ammonium tartrate exposed to radiation with different LET. They found experimental evidence that the radical distributions are indeed dependent on radiation LET. Several investigators (Ciesielski and Wielopolski (1994), Ciesielski et al. (1998), Malinen et al. (2006)) report changes in the power saturation properties of material irradiated with high LET as compared to photon irradiated samples indicating that high-LET irradiation may alter the crystal lattice and therefore the spin-lattice relaxation time T_1 .

Potassium dithionate dosimeters irradiated by heavy charged particles (C^{6+} and N^{7+}) have recently been studied using EPR imaging (Gustafsson et al. (2008b)). It was shown that the EPR spectrum of potassium dithionate is a narrow doublet (total width approximately 0.5 mT) consisting of two signals, R_1 and R_2 with different microwave power saturation properties. In addition to the possibility to use the peak-to-peak amplitude ($R_1 + R_2$) for measurements of absorbed dose, there were also indications that the relative ratio of the two signals, R_1 and R_2 , depended on beam LET. The aim of the present investigation is to further study this relation between the relative ratio of the two signals, R_1 and R_2 and different beam LET. As shown below, this ratio could indeed be quantified and the presented work therefore suggests a new method for measurement of absorbed dose and beam LET by using potassium dithionate EPR dosimetry.

2. Materials and methods

2.1. Chemicals and sample preparation

Potassium dithionate was synthesised according to the standard literature (Brauer 1954) at the Department of Materials Chemistry, Uppsala University, Sweden. Solid household paraffin (100 % by weight, Haugen-Gruppen AB) was used as a binder.

Polycrystalline potassium dithionate was crushed in a mortar and sieved to grain size interval $180 < d < 500 \mu\text{m}$ using an Endecotts MINOR test sieve shaker. The material was heated with 10 % paraffin until the paraffin had melted. The temperature was always well below the melting point for potassium dithionate. The mixture consisting of potassium dithionate and liquefied paraffin was thoroughly mixed while cooling to room temperature. Melting and mixing was repeated three times in order to produce a homogeneous mixture. Finally the mixture was kept in room temperature until the paraffin had solidified completely and was thoroughly mixed a last time. Tablets (diameter 4.5 mm, length 5 mm, weight 170 mg) were pressed using a manual tablet press. The mass difference between the tablets was less than $\pm 2 \text{ mg}$.

2.2. Irradiations

Photon irradiations were performed using a Varian Clinac C/D 6 MV linear accelerator. Doses were measured as absorbed dose in water using an ionisation chamber of type RK 83-05 traceable to the primary standard dosimetry laboratory BIPM in Paris. Tablets were irradiated in a $15 \text{ cm} \cdot 15 \text{ cm}$ radiation field at 5 cm depth in a PMMA slab phantom using a dose rate of 3 Gy/min. The uncertainty in all delivered doses was estimated to be 1.0 % of the dose (coverage factor $k = 1.96$). Charged particle irradiations were performed using a $^{14}\text{N}^{7+}$ - particle beam accelerated by the Gustaf Werner Cyclotron located at the The Svedberg Laboratory, TSL, Uppsala University, Sweden. The beam energy was calculated by determining the depth-dose distribution measured stepwise by an ionisation chamber submerged at increasing depths in a water tank (resolution in the measured depth-dose distribution was estimated to $5 \mu\text{m}$). The maximum absorbed dose was measured at 3.05

mm depth in water and the beam energy could therefore be calculated to $E \approx 33.5$ MeV/u or (including rest mass of nucleus) $E \approx 469$ MeV. Tablets irradiated with the $^{14}\text{N}^{7+}$ - particle beam were placed in a custom made sample holder made of PMMA and irradiated with the flat side facing the beam as indicated in figure 1. The beam width, 20 mm, was much larger than the diameter of the tablet ensuring a homogenous irradiation of the flat tablet side facing the beam.

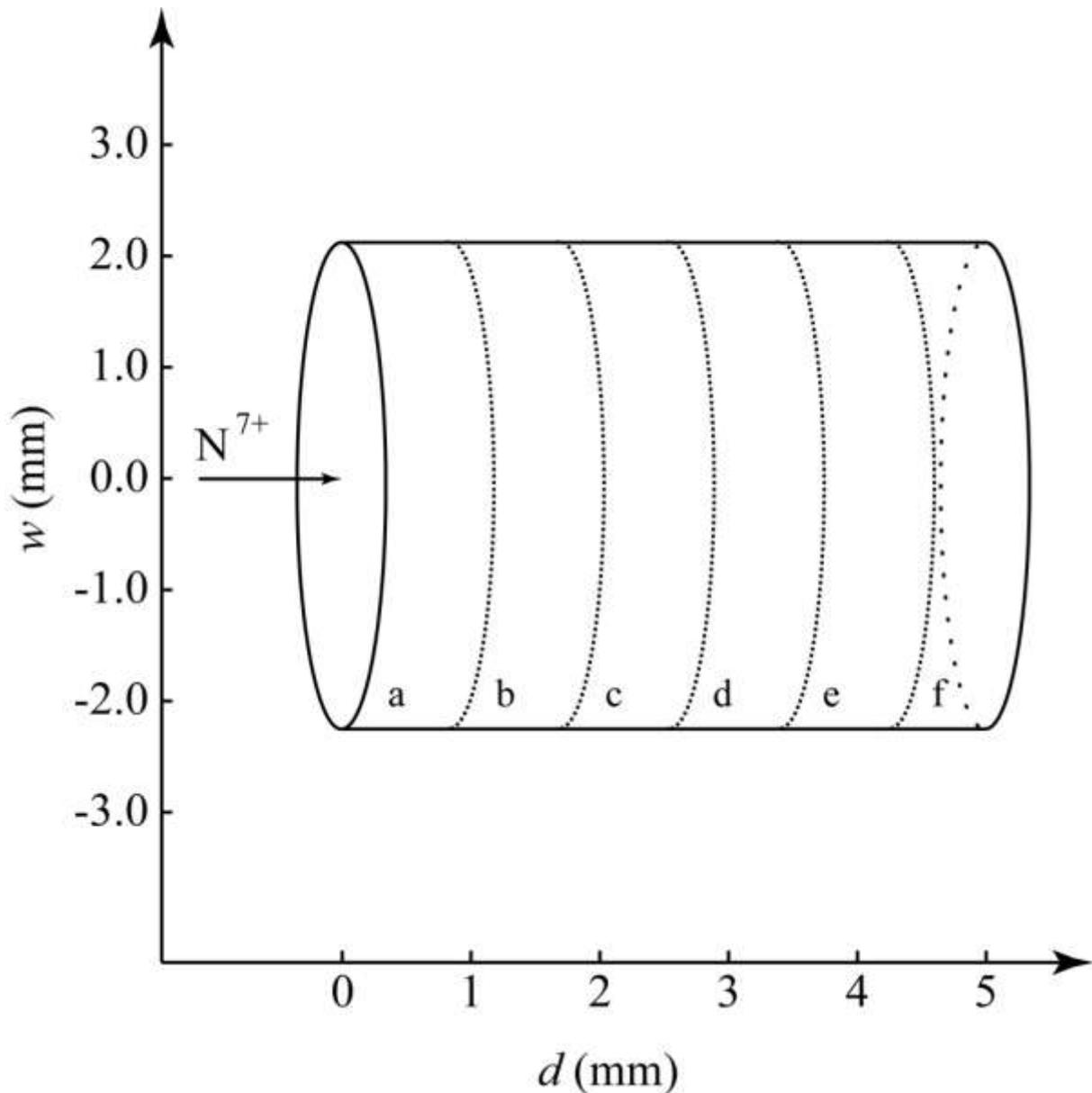


Figure 1. Potassium dithionate tablets were irradiated with the flat side facing the beam. The figure also shows preparation of slices a to f at various tablet depths.

2.3. Estimation of the mass collision stopping power in photon irradiated potassium dithionate

The mass collision stopping power, $\frac{S_{coll}}{\rho}$, is related to the linear energy transfer, L_{Δ} , by a certain cut-off value Δ introduced to compensate for escaping δ -particles in a small object or thin foil (ICRU 1998). In the following of this article Δ was chosen to equal the maximum particle energy, $\Delta = E_{max}$, so that the linear energy transfer would be equal to the mass collision stopping power, $L_{\Delta} = \frac{S_{coll}}{\rho}$ for simplified calculations. Mass collision stopping

power ($\frac{S_{coll}}{\rho}$) in potassium dithionate irradiated with 1.75 MeV photons (mean energy of the 6 MV linear accelerator) can be estimated to $\frac{S_{coll}}{\rho} \approx 1.57 \text{ MeV} \cdot \text{cm}^2 \cdot \text{g}^{-1}$ using tabulated values (ICRU 1984).

2.4. Estimation of mass collision stopping power in N^{7+} irradiated potassium dithionate

The distribution of the absorbed dose and mass collision stopping power in potassium dithionate dosimeters following irradiation with the described N^{7+} -particle beam was simulated by means of the Monte Carlo code SRIM 2008 (Ziegler and Biersack 2008). The distribution of the mass collision stopping power in water was also simulated using the same method for comparison. Simulations are shown in figure 2. The figure also shows how the tablets were sliced for preparation of samples at various tablet depths (further discussed in section 2.5). Monte Carlo simulations of the particle range in water ($d = 3.03 \text{ mm}$) agreed well with measurement ($d = 3.05 \text{ mm}$) suggesting that the simulation protocol was reliable. Simulation of the particle range in potassium dithionate gave $d = 1.85 \text{ mm}$ which agrees well with the particle range measured in potassium dithionate using EPR imaging (EPRI) (Gustafsson et al. 2008b). Mean absorbed dose, $\overline{D_{PDT}}$ (Gy), and mean mass collision

stopping power, $\frac{\overline{S_{coll}}}{\rho}$ ($\text{MeV} \cdot \text{cm}^2 \cdot \text{g}^{-1}$), were calculated for each tablet slice (see section 2.5 and figure 1) for all N^{7+} irradiated tablets. Calculations gave $\overline{D_{PDT}} = 0$ Gy for all tablet slices d, e and f (see section 2.5 and figure 1) and these slices are therefore omitted in the following.

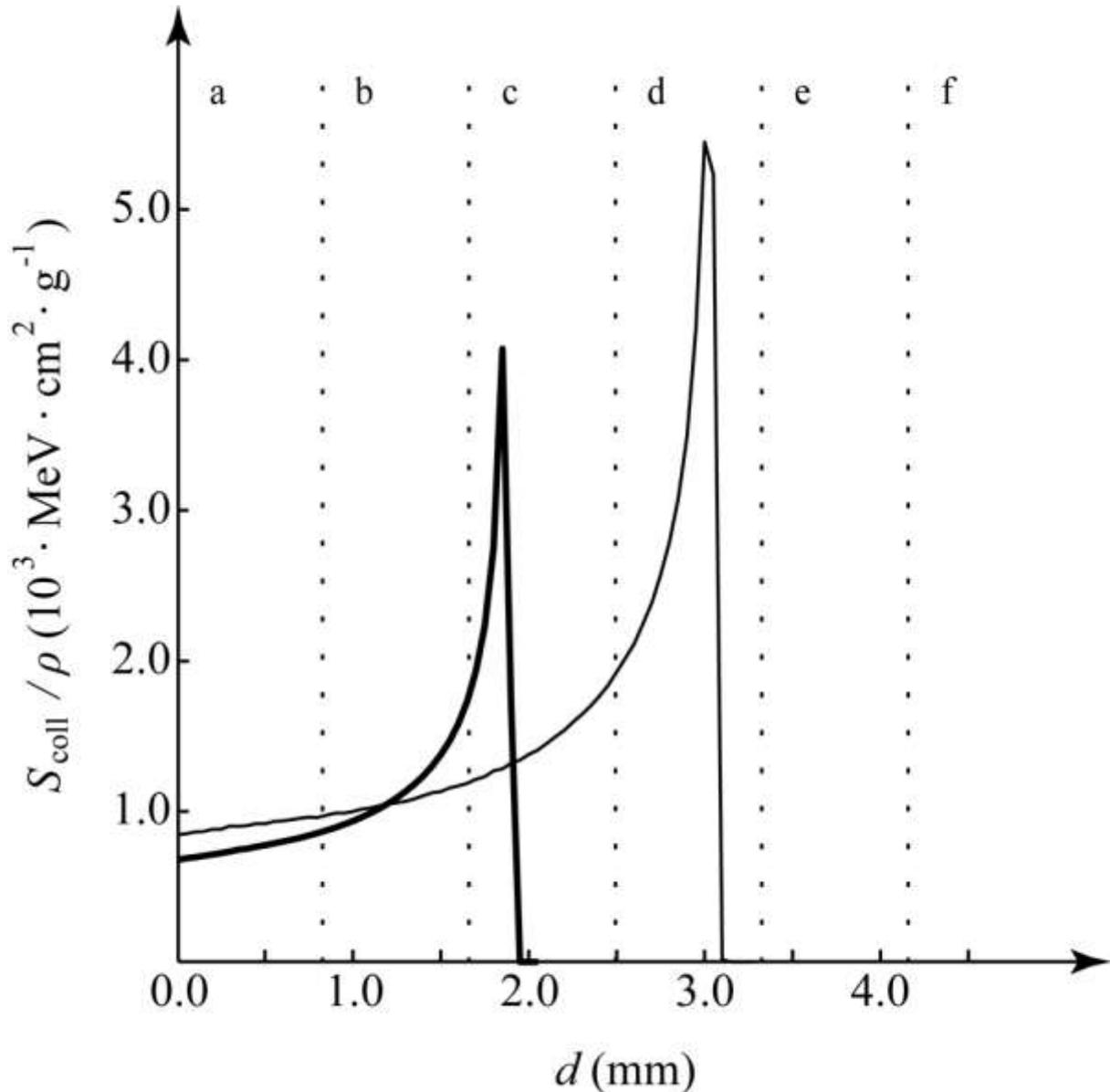


Figure 2. Monte Carlo simulation of the mass collision stopping power as a function of penetration depth in water (thin line) and potassium dithionate (bold line) following irradiation with a N^{7+} particle beam ($E = 469$ MeV). Dashed lines indicate tablet slices a - f.

2.5. EPR measurements

EPR measurements were performed using a BRUKER E580 ELEXSYS EPR spectrometer equipped with the standard cavity 4102 ST. All measurements were performed at 295 K

using 0.1 mT modulation amplitude, 100 kHz modulation frequency, 20 ms time constant, 5*42 seconds sweep, 1024 measurement points distributed over 3 mT. The microwave power was varied in the range of 0.1 mW to 200 mW.

Tablets which had been irradiated with the N^{7+} particle beam were sliced into six pieces as indicated in figure 1. The uncertainty in tablet slicing was estimated to ± 0.2 mm. Slices thus covered were: a: 0.0 mm - (0.8 \pm 0.2) mm, b: (0.8 \pm 0.2) mm - (1.7 \pm 0.2) mm, c: (1.7 \pm 0.2) mm - (2.5 \pm 0.2) mm, d: (2.5 \pm 0.2) mm - (3.3 \pm 0.2) mm, e: (3.3 \pm 0.2) mm - (4.2 \pm 0.2) mm and f: (4.2 \pm 0.2) mm to 5.0 mm. Slices were gently crushed in a mortar and transferred to EPR sample tubes (Heraeus suprasil, outer diameter 3 mm). The mass of each sample was 13 ± 1 mg. Photon irradiated tablets were measured with the whole tablet inserted in a WILMAD EPR sample tube Q-5M-6M-O-200m-FB (inner diameter 5 mm, flat bottom).

The intensities of peaks R_1 and R_2 were measured in arbitrary units as indicated in figure 3.

3. Results

3.1. EPR spectrum of potassium dithionate

Typical spectra of photon irradiated potassium dithionate and potassium dithionate irradiated in N^{7+} are shown in figure 3 (4 mW applied microwave power). No EPR signal was detected in potassium dithionate prior to irradiation.

The relative intensity of R_1 and R_2 varied with microwave power. R_1 reached its maximum intensity at 2 mW and R_2 reached its maximum at 8 mW. An applied microwave power of 4 mW was chosen for all further experiments as an acceptable compromise between microwave saturation properties and measurement sensitivity.

EPR spectra from three tablet slices from regions of increasing depths in a N^{7+} irradiated potassium dithionate tablet are presented in figure 4 (the signal intensities are normalised to equal R_1 intensities by division with the measured R_1 in each spectrum to allow for simple comparison of EPR spectral shape in the figure).

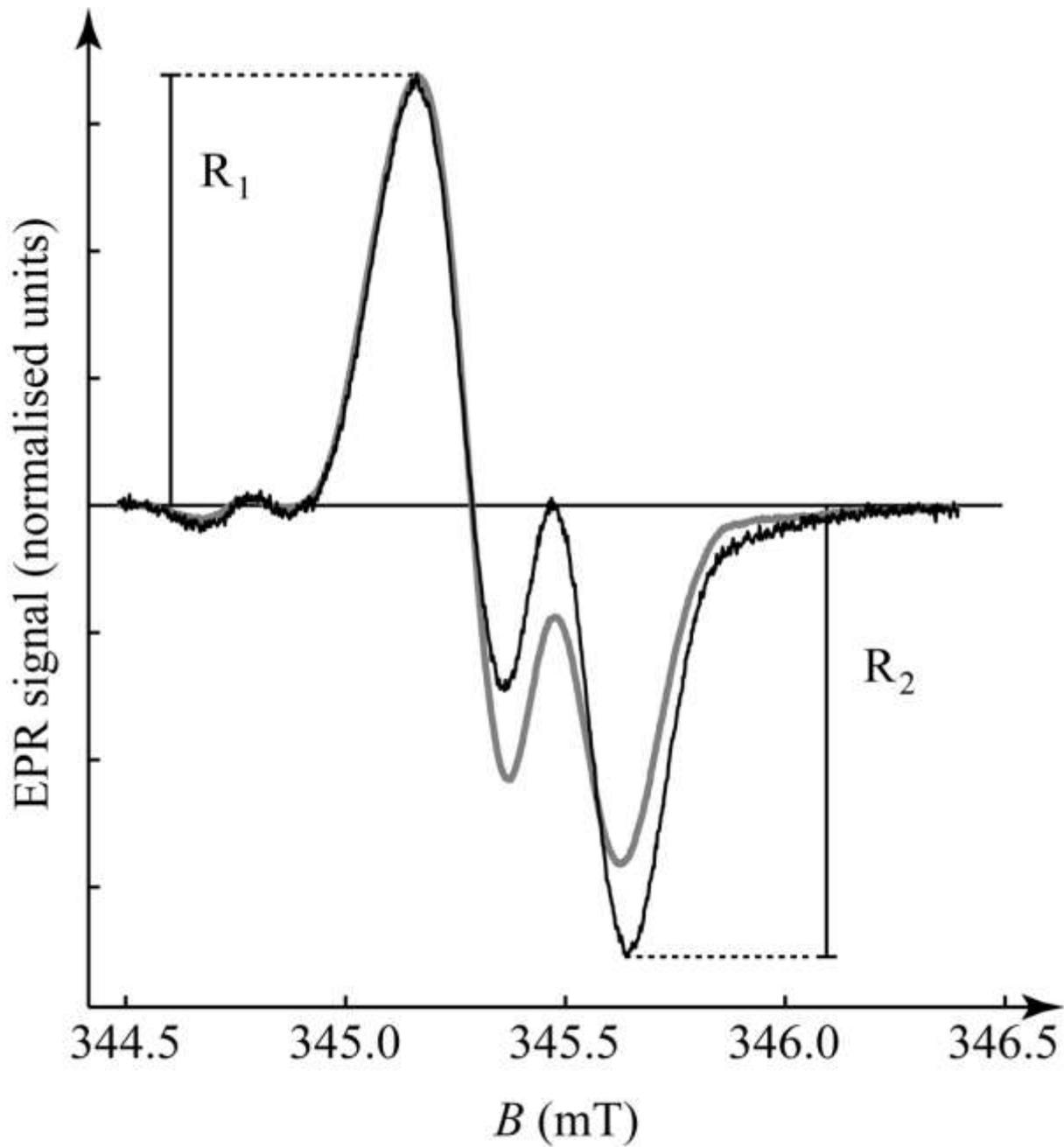


Figure 3. EPR spectrum of potassium dithionate irradiated with N^{6+} ions (black line) and photon irradiated potassium dithionate (bold grey line). Measurements were performed using an applied microwave power of 4 mW. The EPR spectra are normalised to equal R_1 intensities. Notice the differences in EPR spectral shape for the two samples.

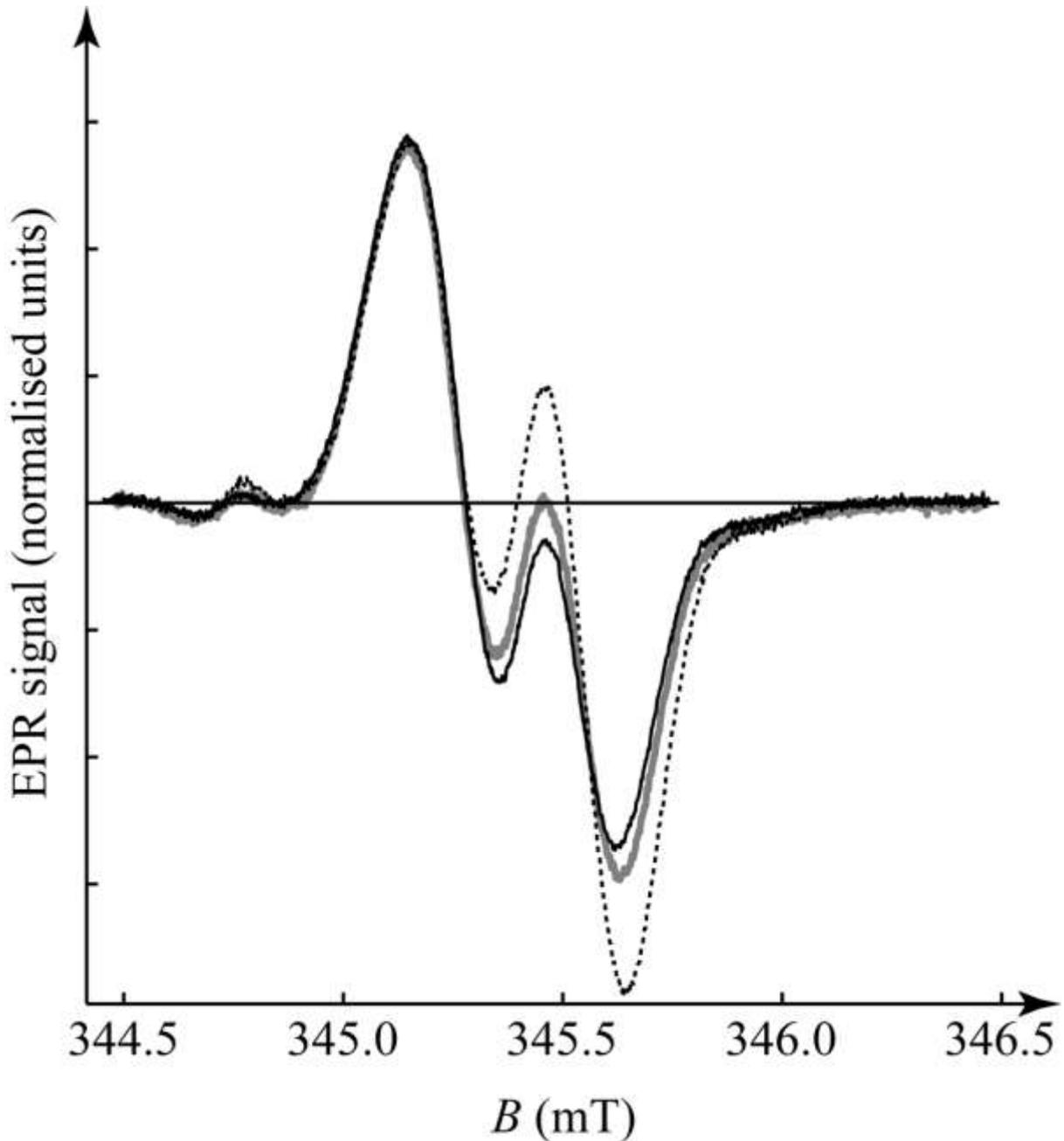


Figure 4. EPR spectra (4 mW applied microwave power) from three tablet slices at regions of increasing depths in a N^{7+} irradiated potassium dithionate tablet. Spectra are normalised to equal R_1 intensities. Black line - slice a ($0.0 \text{ mm} - (0.8 \pm 0.2) \text{ mm}$), bold grey line - slice b ($(0.8 \pm 0.2) \text{ mm} - (1.7 \pm 0.2) \text{ mm}$), dashed black line - slice c ($(1.7 \pm 0.2) \text{ mm} - (2.5 \pm 0.2) \text{ mm}$). Slices d, e and f gave no EPR signal and are therefore omitted in the figure.

3.2. R_1 / R_2 ratio for different beam qualities (different beam LET)

The intensities of peaks R_1 and R_2 (in arbitrary units as indicated in figure 3) and the total mass normalised EPR signal intensity (I_w^{norm}) were measured for all photon irradiated tablets and for all slices of N^{7+} - irradiated tablets. Figure 5 shows measured R_1/R_2 -ratio as a

function of calculated mean mass collision stopping power, $\frac{\overline{S_{coll}}}{\rho}$ ($\text{MeV} \cdot \text{cm}^2 \cdot \text{g}^{-1}$) for all samples. The line indicates an exponential decay ($y = a + b \cdot e^{-c \cdot x}$) fitted to the measured data using the least squares method. Mean values ($\overline{R_1/R_2}$) and the range in the measured R_1/R_2 ratio in potassium dithionate irradiated with photons and for increasing depth in N^{7+} - irradiated samples are displayed in table I.

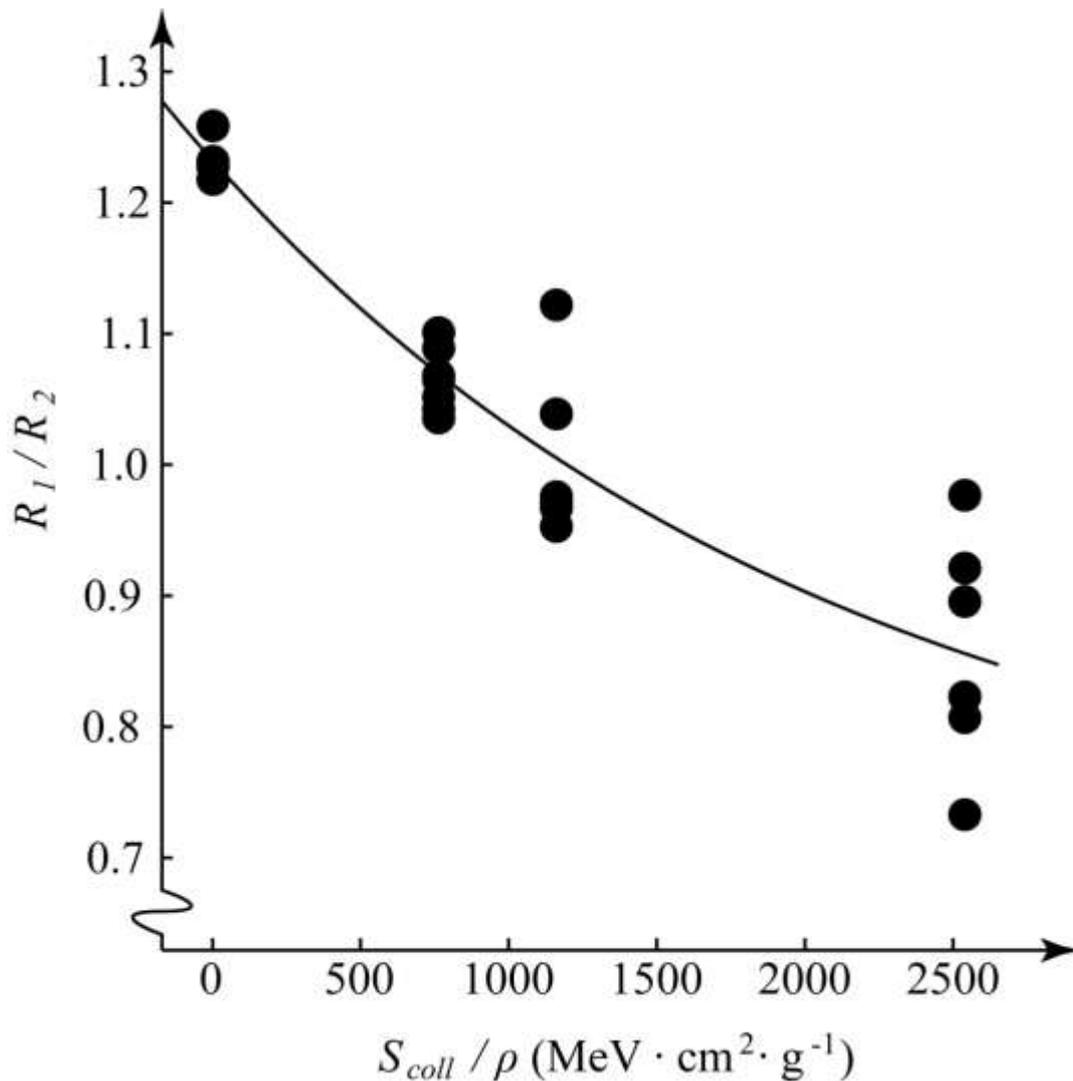


Figure 5. Measured R_1/R_2 -ratio as a function of calculated mean mass collision stopping power, $\frac{\overline{S_{coll}}}{\rho}$ ($\text{MeV} \cdot \text{cm}^2 \cdot \text{g}^{-1}$) for all samples. The line indicates an exponential decay ($y = a + b \cdot e^{-c \cdot x}$) fitted to the measured data using the least squares method (y - measured R_1/R_2 -ratio, x - calculated mean mass collision stopping power, $\frac{\overline{S_{coll}}}{\rho}$ ($\text{MeV} \cdot \text{cm}^2 \cdot \text{g}^{-1}$), a - constant (dimension-less), b- constant (dimension-less) and c - constant ($\text{MeV}^{-1} \cdot \text{cm}^{-2} \cdot \text{g}$)).

TABLE I

Mean values ($\overline{R_1/R_2}$) and the range in the measured R_1/R_2 ratio in potassium dithionate irradiated with photons and for increasing depth in N^{7+} - irradiated samples.

Beam quality	Slice	$\overline{R_1/R_2}$	Range in measured R_1/R_2
6 MV	-	1.24	1.22 - 1.26
N^{7+}	a	1.07	1.04 - 1.10
N^{7+}	b	1.00	0.95 - 1.12
N^{7+}	c	0.86	0.73 - 0.98

4. Discussion

The doublet in the EPR spectra of irradiated potassium dithionate is attributed to two signals with different microwave power saturation properties as shown earlier (Gustafsson et al 2008b). The two signals are assigned to two non-equivalent SO_3^- -radicals due to the irradiation induced cleavage of the S-S bond with similarities to the radical formation observed in irradiated caesium dithionate (Mahgoub et al. 1983, 1994) and irradiated barium dithionate (Baran et al. 2006, 2007). The two SO_3^- radicals in $\text{K}_2\text{S}_2\text{O}_6$ are probably located in different sites in the crystal lattice, and it is not unreasonable that they therefore have different microwave power saturation properties with R_1 being more easily saturated than R_2 . Similar saturation properties were observed for the two signals present in photon irradiated barium dithionate by Baran et al. (2006).

The LET from particle beams is increasing with increasing penetration depth as indicated in figure 2. As shown in figure 3 - 5 and table I there was a clear dependence of the R_1/R_2 - ratio on the beam quality. This result was therefore in accordance with previous measurements by EPR imaging (Gustafsson et al. 2008b). The measured data could be reasonably modelled by an exponential dependence between the R_1/R_2 - ratio in the potassium dithionate EPR spectrum and beam quality, indicating a possible relation. The relatively large spread in the measured R_1/R_2 - ratios could be explained by the comparatively large uncertainty (± 0.2 mm) in the tablet depths covered by each slice. This uncertainty in the tablet depths covered by each slice give large variations in the mass collision stopping power as seen in figure 2 and will therefore give uncertainties in both the absorbed dose in each slice as well as uncertainties in the LET (L_Δ).

Without using sophisticated methods like those described by Malinen et al. (2006) and Waldeland et al. (2010b) differences in line width in EPR spectra from samples irradiated with different beam qualities were not detectable. This might be explained by the fact that the increase in line width due to higher ionisation density is small.

CONCLUSIONS

The current study indicates that the intensity ratio R_1 / R_2 of two signals in EPR spectra of potassium dithionate can be used to estimate the beam quality (LET) in addition to determinations of absorbed dose using the peak-to-peak amplitude ($R_1 + R_2$). Potassium dithionate may therefore be a promising EPR dosimeter material for measurements of absorbed doses in high-LET radiation fields. The ratio of R_1 / R_2 could also be used for the calculation of a correction factor to compensate for the differences in signal intensity which are expected if irradiation of calibration tablets and dosimeters are performed in beams with different LET. The aim of further investigations will be to deduce a calibration table from the LET dependent spectrum changes described in the present work. This will allow high precision measurements of absorbed dose in high-LET beams without the need for calibrating dosimeters in exactly the same beam LET using potassium dithionate EPR dosimetry. Important for future measurements will also be to thoroughly study the long term stability of the radiation induced EPR signal including the stability of the R_1/R_2 - ratio with respect to time.

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