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The microdosimetric one-hit detector model for calculating the relative efficiency of the alanine pellet dosimeter in low energy x-ray beams

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Abstract

The alanine pellet dosimeter is a widely used reference dosimeter in both medical and industrial dosimetry across a wide range of beam qualities. A challenge when using alanine in low energy x-ray beams is its strong energy dependence; a significant decline is observed in the alanine response per dose-to-water relative to irradiations in a cobalt-60 reference field. This decrease is caused by the physical difference in alanine to water dose ratios combined with a radiochemical decrease in the intrinsic detector efficiency but is difficult to characterize mainly due to experimental uncertainties.

Here we have applied a microdosimetric one-hit detector model to characterize the intrinsic detector efficiency of the alanine pellet dosimeter in low energy x-ray beams. Microdosimetric distributions were estimated from track structure calculations using the Geant4-DNA Monte Carlo software, where literature data was used to determine free model parameters.

The model was applied to two sets of x-ray spectra with low (40 kV to 170 kV) and medium (100 kV to 300 kV) tube potential, covering a wide range of beam qualities. A relative detector efficiency of 0.937 was obtained for the low energy set with variations between −1.0% and 1.5%, whereas the efficiency varied between approximately 0.925 and 0.985 for the medium energy set, with a strong correlation to the half-value layer of the beam.

It is concluded, that the tube potential and half-value layer of an x-ray beam is not sufficient characterization to uniquely determine the relative efficiency of an alanine pellet dosimeter. However, the variation in relative efficiency with respect to the half-value layer is small.

1 Introduction

Kilo-voltage (kV) x-rays have extensive applications in radiotherapy, radiation processing, small animal irradiation, and blood irradiation. Irradiation of blood product for prevention of graft-versus-host disease is performed on roughly 10% of all utilized blood product (Sullivan et al., 2007). Traditionally it has been carried out using Cs-137 irradiators, however in recent years a demand for replacement of radionuclide irradiators with x-ray emitters have been introduced (Dodd and Vetter, 2009). Dosimetry protocols recommend water based dosimetry using ion chambers (Aukett et al., 1996; IAEA, 2001); however for applications like blood irradiation, where blood bags are typically irradiated from two or more directions in a closed canister, ion chamber measurements are impractical due to the geometry of the irradiation cavity or the complex x-ray fields. Here both

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placement of the ion chamber and determination of beam quality through half-value layer (HVL) measurements are difficult. In these cases the use of the alanine/electron paramagnetic resonance (EPR) dosimeter may prove more practical.

The alanine/EPR dosimetry system consists of L-α-alanine, in the form of pellets or films, which produce stable free radicals when irradiated. The concentration of stable free radicals produced is proportional to dose to the dosimeter for a wide range of beam qualities (Olsen et al., 1990; Sharpe and Duane, 2003; Bergstrand et al., 2003; Zeng et al., 2004; Zeng and McCaffrey, 2005; Anton et al., 2008) and is stable with a signal loss of few percent over years (Sleptchonok et al., 2000). An EPR spectrometer is used to measure the EPR response, here referring to the peak-to-peak amplitude of the first derivative of the EPR absorption spectrum normalized to the individual pellet mass.

An important characteristic for ionizing radiation detectors is the photon energy response. The relative response \( F_{Q, Q_0} \) after a dose of photons of quality \( Q \), normalized to the response for reference quality \( Q_0 \) (typically cobalt-60 or cesium-137 gamma-rays) is defined as

\[
F_{Q, Q_0} = \frac{(R/D_w)_Q}{(R/D_w)_{Q_0}} = G_{Q, Q_0} \cdot H_{Q, Q_0},
\]

where \( R \) is the detector response and \( D_w \) is the dose to water. \( H_{Q, Q_0} \) is the ratio of dose to detector material \( D_{\text{dos}} \) to \( D_w \) in the photon quality \( Q \) relative to the reference quality \( Q_0 \)

\[
H_{Q, Q_0} = \frac{(D_{\text{dos}}/D_w)_Q}{(D_{\text{dos}}/D_w)_{Q_0}},
\]

and \( G_{Q, Q_0} \) is the relative detector efficiency

\[
G_{Q, Q_0} = \frac{(R/D_{\text{dos}})_Q}{(R/D_{\text{dos}})_{Q_0}}.
\]

The EPR response of the alanine pellet dosimeter irradiated at low energy x-ray qualities relative to cobalt-60 is energy dependent. Recently efforts has been made in characterizing this intrinsic energy dependence from experiments (Zeng and McCaffrey, 2005; Waldeiland et al., 2010; Anton and Büermann, 2015; Khoury et al., 2015; Hjørringgaard et al., 2020). All experimental characterizations of the relative efficiency are carried out in well defined x-ray fields. It is not obvious how the transfer from reference conditions to non-reference conditions, such as small self-shielded x-ray emitters, affects the relative efficiency of the alanine pellet dosimeter. Direct measurements of the relative efficiency in these kind of geometries are difficult at best, and other approaches for determining the relative efficiency are desired.

Olko (2002) investigated the energy dependence with a microdosimetric one-hit detector model. The focus was on different thermoluminescence detector materials, but alanine was included in the analysis. The one-hit detector model refers to types of detectors showing a linear response at low doses and saturating exponentially for higher doses as

\[
R(D) = R_0 \left[ 1 - \exp \left( -\frac{D}{D_0} \right) \right],
\]

where \( R(D) \) is the response at dose \( D \), \( R_0 \) is the saturation response, and \( D_0 \) is the dose at which 63% of saturation response is obtained. This characteristic response curve is observed in alanine dosimeters with a characteristic dose \( D_0 \) in the order of 100 kGy (as later illustrated in Figure 7).
The objective of the present study is to determine the beam quality dependence of the relative
detector efficiency of the alanine pellet dosimeter in kV x-ray fields. This is achieved by applying a
microdosimetric one-hit detector model to a wide range of x-ray beam qualities.

2 Background

2.1 Microdosimetry

Charged particle tracks containing information on spatial positions – and sizes – of energy deposi-
tions in a medium can be obtained by Monte Carlo calculations. Several MC codes for track structure
calculations are available for a selection of beam parameters and materials, e.g. Geant4-DNA (Incerti
et al., 2010b), PHITS (Sato et al., 2018) etc. In microdosimetry a target volume in the material is con-
sidered. An ionizing particle passing through the target volume, producing at least one ionization
in the target volume, is called a single event. Single events leading to a production of detector signal
(stable free radical formation in alanine) is called a hit. Since the transfer of energy occurs as discrete
events (ionizations and excitations) the energy deposited in the target volumes is not uniform but
constitutes a characteristic microdosimetric distribution.

The energy $\varepsilon$ deposited in a target volume for a single event is related to the number of ioniza-
tions $j$ within the target volume by $\varepsilon = j \cdot W$, where $W$ is the mean energy required to produce an ion
pair in the material. The energy deposited normalized to the mass $m$ of the target volume is the
specific energy $z = \varepsilon / m$ in the target volume – the microdosimetric analogue of dose. The stochastic
nature of ionizations and the related specific energy motivates the consideration of the frequency
distributions $f_1(z)$. For a frequency distribution normalized to one event ($\int_0^{\infty} f_1(z) \, dz = 1$) the first
moment (mean specific energy) is

$$z_F = \int_0^{\infty} z f_1(z) \, dz. \quad (5)$$

The subscript 1 refers to a single event. Microdosimetric distributions of ionizations are defined in
an analogous way. Single event distributions are independent of the dose, but do depend on track
and target volume characteristics, such as size and shape.

2.2 The microdosimetric one-hit detector model

The microdosimetric one-hit detector model is based on the multi-hit model which can describe
inactivation of microorganisms. In multi-hit theory it is assumed that the detector contains a type
of target. The target can tolerate a certain amount of hits, however after $n$ hits the target is affected
(e.g. cell death, radical formation, trapping of electron in TLD, etc.). It is assumed that the hits occur
independently of each other and thus can be described by Poisson statistics. The probability of no
effect occurring $S$ as a function of dose $D$ is then (Kellerer, 1987)

$$S(D) = \sum_{\nu=0}^{n-1} \frac{e^{-aD}(aD)^{\nu}}{\nu!}. \quad (6)$$
This is the probability that \( n \) hits does not lead to an effect in the target.

One-hit detectors are a special case of Equation (6) where the probability of no effect occurring is purely exponential \( (n = 1, \text{see Figure 7}) \) so that

\[
S(D) = e^{-\alpha D},
\]  

(7)

where \( \alpha \) is a saturation parameter. In the one-hit model this probability can be expressed in terms of microdosimetric quantities as \( \text{(Zaider, 1990; Olko, 2002, 2006)} \)

\[
S(D) = \exp \left[ -\frac{D}{\bar{D}_F} \int_0^\infty (1 - e^{-\alpha z}) f_1(z) \mathrm{d}z \right].
\]  

(8)

Here \( \bar{D}_F \) is the average dose deposited in the target volume by single events, the ratio \( \frac{D}{\bar{D}_F} \) is thus the average number of events occurring in the target volume after irradiation with dose \( D \). The function \( r(z) = 1 - \exp(-\alpha z) \) represents the probability of an effect occurring after irradiation of specific energy \( z \), and is called the response function for a one-hit detector. The integral is then the average probability that the effect takes place in the target volume given a frequency distribution of specific energy \( f_1(z) \).

The normalized detector response \( R \) after irradiation with dose \( D \) is the complement probability of the probability of no effect occurring

\[
R(D) = 1 - S(D)
\]  

(9)

\[
= 1 - \exp \left[ -\frac{D}{\bar{D}_F} \int_0^\infty (1 - e^{-\alpha z}) f_1(z) \mathrm{d}z \right].
\]  

(10)

By setting the characteristic dose \( D_0 \) equal to

\[
D_0 = \frac{\bar{D}_F}{\int_0^\infty (1 - e^{-\alpha z}) f_1(z) \mathrm{d}z},
\]  

(11)

the detector response of Equation (10) can be simplified as

\[
R(D) = 1 - \exp \left( -\frac{D}{D_0} \right),
\]  

(12)

which is the characteristic response function for one-hit detectors (see Figure 7).

For low doses \( D \ll \bar{D}_F \) Equation (10) reduces to

\[
R(D) = \frac{D}{\bar{D}_F} \int_0^\infty (1 - e^{-\alpha z}) f_1(z) \mathrm{d}z,
\]  

(13)

and since the relative efficiency (see Equation (1)) is

\[
G_{Q,Q_0} = \frac{(R/D)_{Q_0}}{(R/D)_Q},
\]  

(14)

the relative efficiency can be calculated with the microdosimetric one-hit detector model as

\[
G_{Q,Q_0} = \frac{1}{\bar{D}_F} \frac{\int_0^\infty (1 - e^{-\alpha z}) f_1^Q(z) \mathrm{d}z}{\int_0^\infty (1 - e^{-\alpha z}) f_1^{Q_0}(z) \mathrm{d}z}. 
\]  

(15)
The microdosimetric one-hit detector model depends on two free parameters, the saturation parameter $\alpha$ and the target diameter $d$ (assuming spherical volume target). The latter does not appear directly in Equation (15), but the frequency distribution of specific energy is dependent on this parameter.

3 Materials and Methods

The general procedure for implementing the microdosimetric one-hit detector model is outlined below:

1. A set of monoenergetic electron tracks in water with energy $E$ ranging from 1 keV to 1400 keV is produced. Positions of ionization produced by both the primary electrons and the produced secondaries are scored.

2. Microdosimetric frequency distributions of ionizations for the individual electron energies $f_1 (j, E)$ are calculated according to the method described by Kellerer and Chmelevsky (1975), Kellerer et al. (1985), and Rossi and Zaider (1996).

3. Energy distribution of initial Compton- and photoelectrons produced in, or entering, the detector region for a specific primary photon spectrum is scored. Only the electrons produced directly by the primary photon beam, or entering the detector from a different region, is scored in order to avoid double counting in regard to Step 1.

4. Microdosimetric frequency distribution for the primary photon spectra is calculated by folding the monoenergetic electron frequency distributions with the energy distribution of initial secondary electrons.

5. The relative detector efficiency for the primary photon spectra at the irradiation conditions used to obtain the secondary electron spectra is calculated according to Equation (15).

Steps 1 and 3 are performed using MC calculation, while steps 2, 4, and 5 are obtained from post-processing of the MC calculated results. An overview of the MC calculations is given in Table 1.

This general procedure will be applied in different contexts. First, to estimate the free parameters of the model, $\alpha$ and $d$, based on literature values of the relative detector efficiency obtained from Waldeland et al. (2010). The determined model parameters will then be validated by comparison of model predictions with experimental determination of the relative detector efficiency presented in the present study. Finally, the model is applied to a variety of constructed x-ray fields to investigate the general dependence of the detector efficiency to different beam characteristics.

1 The initial energy distribution of secondary electrons will henceforth be referred to as the secondary electron spectrum.
Table 1: Summary of the Monte Carlo simulation parameters based on the guidelines by the AAPM TG-268 (Sechopoulos et al., 2018). MD refer to MC calculations of microdosimetric distributions and SES refer to MC calculations of secondary electron spectra.

| Item name                      | Description                                                                 | References                        |
|--------------------------------|-----------------------------------------------------------------------------|-----------------------------------|
| Code, version                  | Geant4 v.10.5                                                               | Agostinelli et al. (2003)          |
| Validation                     | Comparison of model calculated relative efficiency with literature data.    |                                   |
| Source description             | **MD**: Monoenergetic electrons (1 keV - 1400 keV) initialized in the center of a water box. | Waldeland et al. (2010)           |
|                                | **SES**: Source based on individual experimental geometries.                |                                   |
| Transport parameters           | **MD**: Geant4-DNA physics processes. Specifically G4DNABornIonisationModel for ionizations. | Incerti et al. (2010a)            |
|                                | **SES**: G4EmPenelopePhysics low-energy electromagnetic models.             |                                   |
| Variance reduction technique (VRT) | No VRT was applied.                                                        |                                   |
| Scored quantities              | **MD**: (x, y, z)-coordinates of ionizations in water.                      |                                   |
|                                | **SES**: Energy distribution of Compton- and photoelectrons produced in or entering alanine pellet. |                                   |
| No. histories                  | **MD**: Varying significantly to obtain desired number of ionizations.      |                                   |
|                                | **SES**: Typically $5 \times 10^7$ primary photons.                         |                                   |
| Statistical methods            | **MD**: Electron tracks are calculated until $\sim 2 \times 10^5$ ionization positions are scored. |                                   |
|                                | **SES**: The batch method was used to evaluate statistical uncertainty on energy distribution. |                                   |
| Post-processing                | Microdosimetric frequency distributions for primary photon spectra are calculated by weighting of the monoenergetic frequency distributions of the secondary electron spectra (see Equation (18)). |                                   |
3.1 Alanine pellet dosimeters

Since different alanine pellet dosimeters are commercially available, pellets of different size and composition are used throughout the literature. Two different versions of the alanine pellet dosimeter are used for calculations in the present work, and a brief description is given here.

For calculations where data from Waldeland et al. (2010) is used, the alanine pellets investigated in their work are considered. These pellets consist of 96% alanine and 4% unspecified binder, with a height of 3 mm and diameter 4.8 mm. For the calculations, the binder was assumed to be paraffin wax and a bulk density of 1.2 g cm\(^{-3}\) was used.

For calculations concerning irradiations in a cobalt-60 Gammacell, alanine pellets obtained from Harwell Dosimeters were considered. These pellets have height 2.7 mm and diameter 4.8 mm, with a composition of 91% alanine and 9% paraffin wax. Calculations for these dosimeters were performed using a dosimeter material composition with bulk density 1.23 g cm\(^{-3}\).

3.2 Microdosimetric distributions for monoenergetic electrons

A set of monoenergetic electron tracks in water was calculated using the Geant4-DNA MC code (Incerti et al., 2010b,a; Bernal et al., 2015; Incerti et al., 2018). The energies range from 1 keV to 1400 keV in steps of 1 keV. The electron tracks consist of \((x, y, z)\)-coordinates for all ionizations produced by the primary electron and the subsequent secondaries. Water (or water vapor scaled to unity density) is typically used for track structure calculations because of the lack of appropriate cross section data for other materials at very low energies.

The number of electron tracks at each energy was chosen such that the total number of ionizations produced was of the order \(2 \times 10^5\), however each individual track was analyzed independently.

The frequency distribution of ionizations for the individual electron energies \(f_1(j, E)\) for target diameters ranging from 5 nm to 30 nm was calculated using the weighted sampling procedure described in Kellerer and Chmelevsky (1975), Kellerer et al. (1985), and Rossi and Zaider (1996). By this method the dose distribution of ionizations \(d_1(j)\) is obtained, and the frequency distribution is calculated according to

\[
    f_1(j) = \frac{z^{-1}d_1(j)}{\int_0^\infty z^{-1}d_1(j) \, dj}.
\] (16)

The frequency distribution of specific energy \(f_1(z, E)\) was obtained by multiplying the number of ionizations \(j\) by a chosen \(W\) value of 30 eV per ion pair, identical to what was used by Olko (2002), to obtain the deposited energy \(\varepsilon\), and normalizing this to the mass of the target volume. In total the specific energy is calculated by

\[
    z = \frac{6Wj}{\rho_{\text{target}}\pi d^3},
\] (17)

where \(\rho_{\text{target}}\) is the density of the target material, which for the calculation in water is 1.0 g cm\(^{-3}\).

Examples of the obtained frequency distribution of specific energy for monoenergetic electrons in water is shown in Figure 1. Here the distribution is shown for four individual electron energies,
Figure 1: Microdosimetric single-event dose distribution of specific energy for 1 keV, 10 keV, 100 keV and 200 keV electrons in water, using target diameter $d = 10\,\text{nm}$.

1 keV, 10 keV, 100 keV, and 200 keV, with target diameter $d = 10\,\text{nm}$. All distributions are normalized. It is evident that increasing the initial electron energy results in a lower mean number of ionizations in the target volumes, as well as a convergence towards a specific distribution. For low electron energies the produced ionizations are more localized resulting in a larger mean number of ionizations (and thus mean specific energy) in the target volumes. The microdosimetric distributions for a complex photon field is therefore very sensitive to the fraction of low to high energy secondary electrons produced in the detector.

3.3 Microdosimetric distributions for photon spectra

To illustrate the process of calculating the microdosimetric distributions from primary photon spectra the following section will be based on a cobalt-60 reference beam. The microdosimetric distributions for the reference beam is required (according to Equation (15)) for calculation of the relative efficiency. The reference beam quality is a Nordion GC220 Gammacell located at Risø High Dose Reference Laboratory (HDRL). The spectral distribution of photons at the central position in the Gammacell was calculated using the FLURZnrc usercode of the EGSnrc MC software (Kawrakow et al., 2017), based on published information on material and geometry of the Gammacell (Hefne, 2000; Rodrigues et al., 2009).

To obtain the microdosimetric distributions, first the initial energy distribution of secondary electrons produced in the detector material by primary photons must be calculated. The secondary electron spectra were calculated using the Geant4 MC toolkit (Agostinelli et al., 2003). Electrons produced by Compton and photoelectric interactions in alanine by the primary photons were included...
in the secondary electron spectrum, as well as secondary electrons produced in the surrounding material that enter the alanine pellet. Both the normalized primary gamma spectrum at the central region of the HDRL cobalt-60 Gammacell and the normalized secondary electron spectrum produced in an alanine pellet placed in the central region obtained by calculation are shown in Figure 2. For the calculated secondary electron spectra in the alanine pellet dosimeter approximately 14% are generated with energy $\leq 10$ keV. The discontinuity of the secondary electron spectrum at $\approx 0.96$ MeV and 1.12 MeV correspond to the maximum energy of generated Compton electrons (which is the dominant photon interaction process at the 1 MeV region) for the two cobalt-60 peaks.

![Figure 2: Primary photon energy spectrum at central region of the HDRL cobalt-60 Gammacell (blue) and secondary electron energy spectrum produced in an alanine pellet irradiated in the central position (red) calculated using the Geant4 MC toolkit. Both spectra are normalized to unit area under the curve.](image)

The microdosimetric dose distribution of specific energy for the photon spectra were then calculated by folding the monoenergetic electron frequency distributions (see Figure 1) over the secondary electron spectra $\Phi(E)$ by

$$d_1^{Q_0}(z) = \frac{\int d_1(z, E)\Phi^{Q_0}(E)E \, dE}{\int \Phi^{Q_0}(E)E \, dE}. \quad (18)$$

Here the superscript $Q_0$ imply that the equation is valid for the reference cobalt-60 quality, however the same equation is applicable for an arbitrary photon beam quality $Q$. The resulting dose distribution of specific energy is shown in Figure 3 for different target volumes. The shape of the microdosimetric distributions is governed by the ionization density as well as the size of the target volume. For an increase in target size more ionizations may occur within the target volume, however since the target volume increase the specific energy will typically be shifted towards lower
Figure 3: Microdosimetric single-event dose distribution of specific energy for alanine pellet in the HDRL cobalt-60 Gammacell. Different distributions are shown, illustrating the dependence of the microdosimetric distributions on target volume. The vertical dashed lines represent the mean specific energy of each distribution.

The same process for calculating the microdosimetric distribution of specific energy described in this section for the cobalt-60 reference beam is applied for all x-ray and gamma spectra analyzed in the present study.

4 Results

4.1 Fixing free parameters of the microdosimetric one-hit detector model

Literature values of the relative efficiency of the alanine pellet dosimeter are used to determine the value of the two free parameters of the microdosimetric one-hit detector model $\alpha$ and $d$. The data used for the analysis is obtained from Waldeland et al. (2010). In their work they used spectra of the x-ray beam qualities calculated using SpekCalc (Poludniowski and Evans, 2007; Poludniowski, 2007). These spectra are reproduced here using the detailed information about their input for SpekCalc. A list of the beam modalities from Waldeland et al. (2010) is shown in Table 2. For the calculation of x-ray spectra the input voltage was adjusted (on average roughly 14 %) to make the calculated HVL match the measured HVL as done in Waldeland et al. (2010).

Other literature values of the relative efficiency of the alanine pellet dosimeter in this energy range exist, however available information about the beam qualities are insufficient for reproduction using SpekCalc.
Table 2: List of beam modalities produced based on the work of Waldeland et al. (2010).

| Potential [kV] | $E_{\text{eff}}$ [keV] | Filtration [mm] | HVL [mm] | $G_{Q,Q_0}$ |
|----------------|------------------------|-----------------|-----------|--------------|
|                | Nominal Al Cu Al Cu    |                 |           |              |
| 50             | 32                     | 4.2             | 2.6       | 0.93 ± 0.04  |
| 70             | 36                     | 4.0             | 3.4       | 0.92 ± 0.04  |
| 100            | 43                     | 4.4             | 5.0       | 0.93 ± 0.04  |
| 120            | 54                     | 7.0             | -         | 0.39 0.94 ± 0.04 |
| 135            | 62                     | 10.5            | -         | 0.58 0.94 ± 0.04 |
| 150            | 76                     | 4.0             | 0.53      | 0.94 0.94 ± 0.04 |
| 180            | 83                     | 6.0             | 0.53      | 1.16 0.95 ± 0.04 |
| 200            | 99                     | 6.0             | 0.99      | 1.73 0.97 ± 0.04 |

4.1.1 Secondary electron spectra

The secondary electron spectrum was calculated for all beam modalities listed in Table 2 based on the source, material, and geometrical information available in Waldeland et al. (2010). The irradiation geometry was implemented in Geant4 with simulation parameters given in Table 1. The alanine pellet dosimeter was placed at 2 cm depth in a water phantom at 50 cm source to surface distance. The calculated energy distribution of Compton- and photoelectrons produced in, or entering, the alanine dosimeter volume is shown in Figure 4 for the 50 kV and 200 kV beam modalities in Table 2. It is evident that the secondary electron spectra for the 50 kV beam is heavily dominated by low energy electrons (e.g. electron energies below 10 keV) compared to the 200 kV spectrum. The fraction of initial secondary electrons with energies below 10 keV to the total number of electrons produced decreases from 74 % for the 50 kV beam to 49 % for the 200 kV beam. The plateau of zero initial secondary electrons observed between 10 keV and 20 keV for the 50 kV beam is an effect of the...
4.1.2 Microdosimetric frequency distribution

The microdosimetric frequency distribution of specific energy is calculated for target diameters in the range 5 nm to 30 nm according to Equation (18). Figure 5 show the single-event dose distribution of specific energy for the 50 kV and 200 kV spectra from Waldeland et al. (2010), using target diameter 10 nm. The single-event dose distribution of specific energy for the cobalt-60 reference shown in Figure 3 is included in Figure 5 for reference. The distinction between the frequency distribution of specific energy for the 50 kV and 200 kV beams are subtle. Greater secondary electron energies result in less localized ionizations in the individual electron tracks and thus a frequency distribution shifted towards lower specific energies – or less localized dose deposition.

4.1.3 Model-fit to litterature data

The free parameters of the model $\alpha$ and $d$ are determined by comparison of calculated and literature values for the relative dosimeter efficiency $G_{Q,Q_0}$. The microdosimetric frequency distributions calculated in Section 4.1.2 is used to calculate the relative efficiency for a wide range of saturation
parameters according to Equation (15). The relative efficiency obtained by use of different combi-
nations of model parameters is then compared to the literature values related to the x-ray spectra
used for calculation of the microdosimetric frequency distributions (see Table 2). The optimal set
of model parameters is determined by minimizing the relative least squares of the calculated and
experimental values
\[
M = \sum_{Q=1}^{N} \left( \frac{G_{Q,Q_0}^{\text{exp}} - G_{Q,Q_0}^{\text{calc}}}{G_{Q,Q_0}^{\text{exp}}} \right)^2,
\]
where the sum is over all beam qualities in the set.

The obtained values of the parameter \(M\) as a function of the saturation parameter \(\alpha\) is shown in
Figure 6. The optimal saturation parameter, for each target diameter, is then obtained by locating the
minimum of \(M\) with respect to \(\alpha\). From Figure 6 it appears that target diameters in the range 10 nm
to 30 nm are all able to reproduce the literature values of the relative efficiency reasonably well with
the right choice of saturation parameter. Since no clear correlation between target diameter and
the level of agreement is apparent, an additional constraint on the choice of model parameters is
introduced.

4.1.4 Linearity index of cobalt-60 reference

As an additional constraint on the choice of free parameters, the predictive ability of the model is
evaluated by comparison of experimental and model values for response per dose in the reference
cobalt-60 field \(Q_0\). The response of alanine in the reference cobalt-60 field \(Q_0\) is calculated according
to Equation (10), using sets of model parameters determined from Figure 6. The linearity index
The response per dose at each dose point normalized to a specific dose point, is calculated by

\[
f(D) = \frac{(R/D)_{\text{dose}}}{(R/D)_{\text{ref. dose}}}.
\]  

(20)

Here the reference dose point is chosen to be \(D_w = 1\) kGy, corresponding to \(D_{\text{ala}} = 0.97\) kGy.

Figure 7 show the calculated response and linearity curves for the free parameter sets obtained in Figure 6 as well as the response curve for pellets irradiated in a cobalt-60 Gammacell, normalized to the saturation response. The relative difference between model and experimental values calculated by

\[
\text{Rel. diff.} = \frac{X_{\text{mod}} - X_{\text{exp}}}{X_{\text{exp}}},
\]

where \(X\) is the physical quantity investigated. The comparison of model and experimental data in

![Figure 7: Comparison of model and experimental values for dosimeter response (left) and linearity index (right) curves. The relative differences of model and experimental values are shown below with the dashed line representing a 5% difference.](image)

Figure 7 clearly suggest a parameter set of \(d = 10\) nm in water, which has a corresponding saturation parameter \(\alpha = 2.86 \times 10^{-5}\) G\(\text{y}^{-1}\), for calculating the dosimeter properties in the reference cobalt-60 field. Since this set of model parameters display best agreement for a wider range of beam qualities it is adopted for further use in the model.

This comparison of experimental response curves normalized to saturation response with response curves calculated using the microdosimetric one-hit detector model represents an assumption that the saturation of response is only due to a saturation in stable free radical production at high doses. This assumption implies that the EPR-readout, in this case the peak-to-peak height in the first derivative of the EPR-spectrum, is proportional to the concentration of stable free radicals, which is however not the case. The measured response curves also includes a saturation intrinsic to the spectrometer. For instance different values of the characteristic dose can be measured for differ-
ent values of the microwave power used in the EPR-spectrometer (Wieser and Girzikowsky, 1996; Malinen et al., 2003). Here the comparison is used to pick out a set of model parameters, from a list of parameter sets which all, according to Figure 6, reproduce literature values at kV x-ray energies reasonably well.

4.2 Uncertainty considerations

In the following section the considerations and handling of uncertainties for the model is described.

4.2.1 Monte Carlo calculations

For MC calculations of radiation transport uncertainties typically include components from translation from laboratory conditions to MC geometry and materials, cross-section data, input physics, and statistical effects. In the present study the relative detector efficiency is calculated according to Equation (15), where $\Sigma_f$ and $f_1(z)$ are obtained through MC calculations. Since Equation (15) is expressed as a ratio with these parameters appearing in both the numerator and denominator, and these parameters are obtained using the same MC code and input physics, it is assumed that the contribution to the final uncertainty of the relative efficiency is negligible compared the contribution from the applied experimental data.

4.2.2 Literature values

A significant contribution to the overall uncertainty comes from the use of literature data based on measurements to fix the free parameters of the model (see Table 2). The effect of experimental uncertainties on the determined set model parameters is examined by performing a bootstrap analysis. This analysis was performed keeping the target diameter fixed at $d = 10$ nm and scoring the corresponding optimal saturation parameter for $1 \times 10^5$ random samples, with replacement and including uncertainties, of the data points.

The distribution of optimal saturation parameters obtained from this analysis was best described by a log-normal distribution. By fitting a log-normal distribution to the collection of optimal saturation parameters an upper and lower limit, defined by the 2.5% and 97.5% percentile corresponding to two standard deviations for a normal distributed parameter, was estimated. The saturation parameter obtained by this analysis was $\alpha = (2.86^{+3.11}_{-1.65}) \times 10^{-5} \text{ Gy}^{-1}$.

4.3 Characterization of the alanine pellet dosimeter

The parameter values obtained in Section 4.1 can now be applied for relevant primary x-ray spectra in order to calculate the relative efficiency of the alanine dosimeter in that particular beam quality. In the following section this has been done in different ways to explore the alanine dosimeter efficiency dependence on several parameters.
The calculation of secondary electron spectra was performed using the same geometry, and alanine pellet size and composition, as was used for fixing the model parameters in Section 4.1, based on the information from Waldeeland et al. (2010).

### 4.3.1 Monoenergetic photons

The secondary electron spectrum was calculated with the Geant4 MC toolkit for a range of monoenergetic primary photons. Using Equation (15) together with the free parameter values $d = 10 \text{ nm}$ and $\alpha = 2.86 \times 10^{-5} \text{ Gy}^{-1}$ and the calculated secondary electron spectra for monoenergetic photons in alanine, the relative efficiency is obtained. The top part of Figure 8 show the calculated relative efficiency for monoenergetic primary photons, as well as the result of changing the set of model parameters by $\pm 2\sigma_{\alpha}$.

![Figure 8](image)

**Figure 8:** Top: Relative efficiency $G_{Q,0}$, as a function of monoenergetic photon energy $E_{\text{mono}}$, calculated with the microdosimetric one-hit detector model for monoenergetic primary photons (solid line). The upper and lower bound (dashed lines) is determined by the method described in Section 4.2. Data points represent literature values of the relative efficiency versus effective energy $E_{\text{eff}}$.

Bottom: The mean specific energy for target diameter $d = 10 \text{ nm}$.

The bottom part of Figure 8 show the corresponding mean specific energy calculated according to Equation (5) using target diameter $d = 10 \text{ nm}$. The mean specific energy is a measure for how localized the dose deposition is. It is worth noting that the local minimum and maximum apparent in the energy dependence of the relative efficiency – at 20 keV and 50 keV respectively – directly corresponds to the opposite extrema in the mean specific energy. The bump in the curve for mean specific energy occurs as the fraction of secondary electrons produced by Compton scattering increases, since the low energy electrons produced through Compton scattering deposit their energy more locally.
The capabilities of the model for calculating the relative efficiency of the alanine pellet dosimeter in low energy x-ray fields was tested by comparison with literature data. In Figure 8 the relative efficiency measured in Zeng and McCaffrey (2005), Waldeland et al. (2010), Anton and Büermann (2015), and Hjørringgaard et al. (2020) is presented as a function of the respective x-ray beams. The data on relative efficiency from Anton and Büermann (2015) is obtained by taking the ratio of relative response to MC calculated dose ratios listed in Table 6 and Table 7 of their paper.

By visual inspection of Figure 8 good agreement is observed between model calculated relative efficiency for monoenergetic x-rays and experimental data characterized by the effective energy of the x-ray beam. The overall trend with local maximum and minimum around 20 keV and 50 keV appear to be present in the experimental data as well.

Statistical examination of the model calculations was based on the null hypothesis that the relative efficiency is unity over the energy range covered by literature data. This choice of null hypothesis is based on the fact that the relative efficiency converges to unity for higher energies. Due to the small sample size in the literature data, the analysis was performed using bootstrapping including the experimental uncertainties. For each set of resampled data the mean residual of both the unity model of the null hypothesis and the microdosimetric model to the bootstrapped sample was scored. The difference between the two models was determined to be statistically significant, and it is thereby assumed that the microdosimetric model is a better representation of the experimental data.

It is of course not obvious that effective energy as a beam qualifier is sufficient classification of beams to determine the relative efficiency. As such a direct comparison of relative efficiency for monoenergetic photons to effective energy of composite x-ray fields may not be optimal. Lack of knowledge about the spectral distribution for the literature data does however make this comparison the most reasonable. How the relative efficiency depends on other beam characteristics than just the effective energy can now be investigated using the microdosimetric one-hit detector model.

4.3.2 X-ray beam characteristics

Differences in x-ray tube geometry – filtration material and thickness, target material and angle, etc. – entails a wide range in HVL values for the same tube potential. From a survey on the status of clinical x-ray dosimetry in North American clinics the variation in HVL for x-ray tubes with tube potential 10 kV to 300 kV is obtained (see Figure 2 of Ma et al., 2001). This variation gives an indication of the x-ray beam quality range for which the dosimeter properties should be characterized.

Based on the model parameters determined in this study, the general dependence of the relative efficiency of the alanine pellet dosimeter on different x-ray beam qualifiers is investigated. A set of primary x-ray spectra was generated using the SpekCalc software. The x-ray spectra was of varying high voltage (HV), 40 kV to 300 kV, and HVL, the latter obtained by varying the external filtration.
Options used for all calculated spectra include filtration of 1.0 mm beryllium and 1000 mm of air, and anode angle 30°. The variation in HVL is obtained by changing the external aluminum or copper filtration. The range in input parameters for the calculation of x-ray spectra was chosen such that the generated spectra cover the range of beam qualities listed in Ma et al. (2001).

The relative detector efficiency for each beam quality was calculated in the same manner as described in previous sections. Figure 9a show the calculated relative efficiency calculated for the set of x-ray spectra with tube potential 40 kV to 170 kV and HVL ≈ 0.2 mm Al to 14 mm Al.

**Figure 9:** Relative efficiency of the alanine dosimeter (color bar) calculated using the microdosimetric one-hit detector model with parameters \( d = 10 \text{ nm} \) and \( \alpha = 2.86 \times 10^{-5} \text{ Gy}^{-1} \). Primary photon spectra were obtained using SpekCalc (see text for details).

The general dependence of the relative efficiency on HV and HVL shown in Figure 9a appear to follow the approximation in Figure 8. The range of HVLs displayed correspond to effective energies in the approximate range 15 keV to 90 keV corresponding to the minimum valley in Figure 8. The relative detector efficiency for x-ray qualities in this range was found to vary between roughly 0.925 and 0.985.

Figure 9b shows the calculated relative efficiency calculated for the set of x-ray spectra with tube potential 100 kV to 300 kV and HVL ≈ 0.2 mm Cu to 5 mm Cu. Note that the range of the colorbar in Figure 9b is different to Figure 9a. The HVL in the set of spectra corresponds to effective energies in the range 45 keV to 200 keV. The general tendency is a increase in the relative detector efficiency with increasing HVL in this beam quality region, in accordance with Figure 8. For this set of x-ray beam qualities the variation in relative efficiency for a specified tube potential reaches several percent, varying between roughly 0.925 and 0.985.

### 5 Discussion

In the present study the microdosimetric one-hit detector model was applied to the alanine pellet dosimeter to calculate the relative efficiency in low energy x-ray beams with respect to a cobalt-60 reference field. Literature values obtained from Waldeland et al. (2010) were used to fix the free parameters of the model, the target diameter \( d \) and the saturation parameter \( \alpha \). Using only
the minimization of relative least squares shown in Figure 6 was not sufficient to conclusively
determine an optimal parameter set, therefore an additional constraint concerning the prediction
of response curves for pellets irradiated in a Cobalt-60 Gammacell was included. The optimal
set of model parameters determined in this manner was obtained to be \( d = 10 \text{ nm} \) in water and
\( \alpha = 2.86 \times 10^{-5} \text{ Gy}^{-1} \). A different value for the mean energy required to produce an ion pair \( W \)
chosen for the analysis might result in different sets of model parameters. Olko (2002) arrived at a
target diameter of \( d_{\text{ala}} = 6 \text{ nm} \) corresponding to 8–9 nm in water, converting by density ratio.

As discussed by Olko (2002) this order of target diameter is much larger than the size of an
alanine molecule implying the saturation effect at high doses are not due to a lack of unionized
alanine molecules. Olko (2002) argue that other effects of ionizing radiation – cross linking, coiling of
chains – can trap the free radicals, preventing recombination at normal dose ranges. At high doses,
or high Linear Energy Transfer (LET), these structures are destroyed, leading to recombination and
a reduced detector efficiency. In this case the target size is interpreted as the effective range of
recombination of the free radicals.

The comparison between calculated response, and linearity, curves with measured response
curves for alanine pellet dosimeters irradiated in a cobalt-60 Gammacell include an underlying
assumption that the measured EPR response (peak-to-peak height of the first derivative of the EPR
spectrum) is directly proportional to the concentration of stable free radicals. This assumption im-
plies that the observed saturation in the EPR response is only due to saturation in the production
of stable free radicals. Effects of the readout procedure, such as choice of microwave power and
modulation amplitude, may influence the relative spectrometer sensitivity for low and high radical
concentrations, and thus affect the dose level at which saturation occurs. This effect would need
to be characterized for consistency. In the present study the comparison is applied to pick a set of
model parameters from a list of model parameter sets which all reproduce the kV x-ray literature
data for relative detector response reasonably well.

The agreement between model calculated relative efficiency of the alanine pellet dosimeter for
monoenergetic photons in the energy range 5 keV to 1000 keV and experimental data was assessed
by both visual inspection and by performing a bootstrapping analysis. This analysis showed the
difference between the model calculations and the null hypothesis of a unity relative efficiency was
statistically significant. The use of effective energy as the single beam qualifier may however be an
oversimplification as shown in Section 4.3.2.

Applying the microdosimetric one-hit detector model for a set of monoenergetic primary photon
beams showed a similar anomalous dependency of the relative detector efficiency on photon energy.
The energy dependence showed a local extremum at 25 keV and 50 keV. The characteristic anom-
alous shape occurs as with increasing photon energy the fraction of secondary electrons produced
by Compton effect increases. The same anomalous shape is obtained by Olko (1999, 2002), however
they report local maximum and minimum at 40 keV and 80 keV respectively. This difference may be
due to the difference in applied model parameters as well as the choice of alanine detector material.
Olko (2002) use a mixture of 90% alanine and 10% paraffin wax in an unspecified geometry for
calculating the secondary electron spectra, whereas a mixture of a mixture of 96% alanine and 4% paraffin wax has been used in the present study.

The microdosimetric one-hit detector model has further been applied to two sets of generated x-ray spectra covering a wide range of x-ray beam qualities used in clinics. The low energy set was generated with HV in the range 40 kV to 170 kV and HVL from 0.2 mm Al to 14 mm Al, and the medium energy set consist of HVs from 100 kV to 300 kV with HVL from 0.2 mm Cu to 5 mm Cu. This study was performed to explore the usefulness of different beam qualifiers for characterization of the relative detector efficiency. The relative detector efficiency in the low energy set varies within $-1.0\%$ and $1.5\%$ of the average value of $G_{Q,Q_0} = 0.937$ for the investigated beam quality range. This indicates that the tube potential may be sufficient for practical use in choosing a literature value of the relative detector efficiency to apply to measurements. However further investigation of the influence of e.g. phantom material, alanine pellet position in phantom, etc. should be explored in detail. For the medium energy set of generated x-ray spectra a significant dependence of the relative detector efficiency on the HVL was observed. For HVL $>3$ mm Cu the relative detector efficiency appear to be independent on the x-ray tube potential. The reason for this is probably that the fraction of low energy electrons generated in the detector by these hard filtered beams is quite low for all the investigated tube potentials making variations between spectra insignificant.

Several aspects of the model calculations which may impact the final model prediction of the relative efficiency have not been considered in the present study. MC calculation at very low energies should be interpreted with reservations regarding interaction cross-sections. Different ionization cross-section models for very low energies are available in the Geant4-DNA MC toolkit (Bernal et al., 2015). The effect on the model predicted relative efficiency of the alanine pellet dosimeter from using different low energy ionization cross-section models has not been explored in the present study. Only cross sections for water are available for the calculation of the microdosimetric distributions, which is a major limitation in the application of this model.

The recombination of free radicals has been shown to be dependent on the beam quality (Hansen and Olsen, 1989). For heavy charged particles high LET beams show significantly greater fading compared to lower LET beams. The same effect could be present for x-rays, where low energy x-rays have greater LET (by secondaries) relative to high energy x-rays. This effect may be of importance when assessing the detector response, but have not been investigated here.

6 Conclusion

The microdosimetric one-hit detector model was applied to calculate the relative efficiency of the alanine pellet dosimeter. The free parameters of the model was determined by comparison of model calculations with literature data (Waldeland et al., 2010) on the relative efficiency.

The model was then applied to a set of monoenergetic photon beams, showing a characteristic relationship between the relative efficiency and monoenergetic photon energy. The relative efficiency was found to be overall increasing with photon energy, but with a local maximum and minimum at
Lastly, the model was applied to a wide set of x-ray beam qualities. The results showed that the effective energy (or HVL) of an x-ray beam is not necessarily sufficient uniquely to determine the relative efficiency of the alanine pellet dosimeter.

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Highlights:

- The relative efficiency of the alanine pellet dosimeter was examined using a microdosimetric model.
- Model parameters were established using literature data for relevant beam qualities.
- Results include a characterization of the relative efficiency for a wide range of beam qualities.
Declaration of interests

☒ The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

☐ The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: