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Absorbed dose to water determination for kilo-voltage X-rays using alanine/EPR dosimetry systems

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ABSTRACT

Alanine's relative response to kilo-voltage X-rays, compared to ⁶⁰Co reference quality beam, was studied in this work, in order to determine correction factors to be applied to alanine's response when irradiated with low to medium energy X-rays (up to 300 keV).

The relative response to kilo-voltage X-rays of Aerial's alanine dosimeters was determined by three distinct methods: experimental measurements using alanine dosimeters and a calibrated PTW Farmer 30013 ion chamber, Monte Carlo simulations using MCNPX code and finally, analytical calculations based on weighting of X-ray spectra by NIST's published mass energy absorption coefficients. Two sets of X-ray beam qualities, covering high voltages ranging from 50 kV up to 280 kV, were used to study the energy dependence of the alanine dosimeter's response. Obtained results were consistent within 2.1% (average standard deviation at $k = 1$).

1. Introduction

Nowadays, low to medium energy X-ray (up to 300 keV) irradiators start to replace irradiators using radioactive sources like Cesium (¹³⁷Cs) and Cobalt (⁶⁰Co) gamma sources (Dodd and Vetter, 2009) (Tadokoro et al., 2010), mainly in the fields of blood irradiations (Seglam et al., 2011), Sterile Insect Technique (SIT) (Dyck et al., 2005) and food irradiations (Baraki-Golan and Follett, 2017) (Moosekian et al., 2012). This drift towards kilo-voltage X-ray generators is mainly driven by the difficulty of acquiring and transporting radioactive sources, as well as the reassuring radiation safety that provides self-shielded X-ray irradiators to its operators.

Alanine/EPR (Electron Paramagnetic Resonance) dosimetry systems are well used for reference, transfer and routine dosimetry. Their very stable and reproducible measurement insures a low uncertainty on absorbed dose determination. Yet, for low to medium energy X-rays, it has been reported in many studies (Anton and Büermann, 2015) (Waldeland et al., 2010) (Khouri et al., 2015) (Zeng and McCaffrey, 2005) that alanine's response with respect to water is lower than unity, as it is for photon energies higher than 200 keV.

Khouri et al. (Khouri et al., 2015) determined the relative response of alanine dosimeters purchased from Aerial (Illkirch, France). A set of dosimeters was irradiated with 125 kV X-rays. Another set of dosimeters was irradiated with ⁶⁰Co gamma rays (average photon energy of

1.25 MeV). Results showed that the ratio of alanine's response to ⁶⁰Co with respect to X-rays is equal to 1.2, thus, meaning that alanine's response to ⁶⁰Co is 20% higher than alanine's response to 125 kV X-rays.

Anton et Büermann (Anton and Büermann, 2015) carried out Monte Carlo calculations as well as experimental measurements, in order to study alanine's relative response for medium energy X-rays, with tube potentials ranging from 30 to 280 kV. Their work showed that alanine's response drops to 64% for an irradiation at 30 kV, compared to a 100% response in the case of ⁶⁰Co reference beam.

Waldeland et al. (Waldeland et al., 2010) studied the dose to water energy dependence of alanine dosimeters. Monte Carlo simulations and experimental measurements were carried out with tube voltages ranging from 50 to 200 kV. Experimental measurements showed that alanine's response in case of X-ray irradiations with respect to ⁶⁰Co beams varied from 0.68 to 0.9 at 50 kV and 200 kV respectively.

Three different methods are developed in this work to study the alanine dosimeter's relative response to kilo-voltage X-rays, compared to ⁶⁰Co. Absorbed dose to water measurements with alanine dosimeters could be underestimated in case of kilo-voltage X-ray irradiations, if the EPR dosimetry system is calibrated with high energy photons (gamma or X-rays) and electron beams (Waldeland and Malinen, 2011). Thus, the goal of this study is to determine correction factors to be applied to alanine's response when irradiated with kilo-voltage X-rays.

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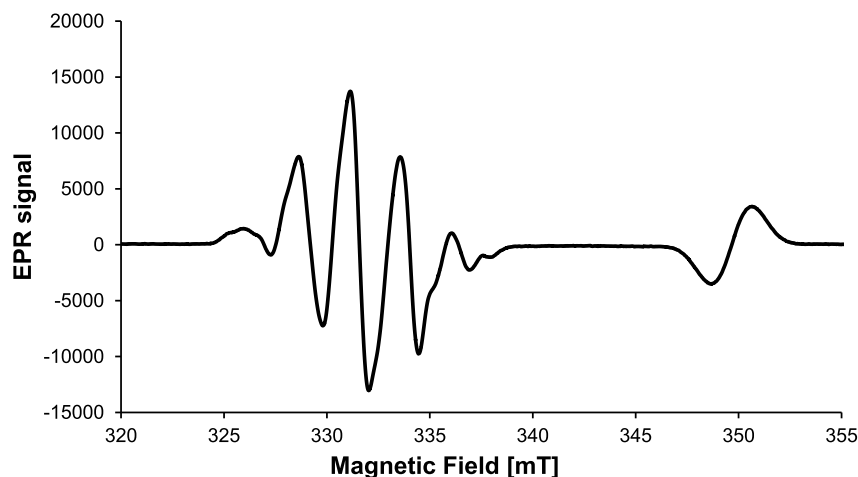


Fig. 1. EPR spectrum of irradiated alanine dosimeter. Resonance peaks between 325 mT and 340 mT are intrinsic to alanine radicals. Singlet resonance peak at 350 mT corresponds to the spectrometer's internal ruby reference.

2. Material and methods

2.1. Alanine dosimeters and EPR spectrometry

Aerial's commercial alanine dosimeters (Lot 09/11) (Kuntz et al., 1998) were used in this work. Alanine pellets have a 4 mm diameter, a thickness of 2.35 mm and an average mass of 36.05 ± 0.05 mg, with a composition of 93% of pure L-alanine and 7% binder.

During irradiations, 5 alanine pellets were placed at the surface of a PMMA (PolyMethylMethacrylate) phantom having 10 cm in diameter and 1.5 cm in thickness. For some beam qualities, an additional 5 mm PMMA plate was added on top of the dosimeters.

EPR readout was performed using a Freiberg Instruments/Magnetech MS5000 spectrometer (Freiberg, Germany) using the following parameters: magnetic field sweep width of 2 mT, sweep time of 5s, modulation amplitude and frequency of 0.7 mT and 100 kHz respectively, microwave power and frequency of 10 mW and 9.253 GHz respectively. Fig. 1 shows the measured EPR spectrum of an irradiated alanine dosimeter. Measured spectra were taken as input in the AerEDE2019 software, developed and commercialized by Aerial (Illkirch, France), in order to correct the dosimeter's response (EPR spectrum's central peak height per mass unit) with irradiation temperature. Absorbed dose to water can be measured by using a calibration curve that correlates the dosimeter's response to a reference absorbed dose to water. In this study, only the dosimeter's response was measured by EPR, in the case of X-ray irradiations.

3. X-ray irradiations

For irradiations carried out at Aerial, a 3 kW high voltage generator powered our BALTEAU-NDT TSD 160/0 X-ray tube, with a maximum applicable high voltage of 100 kV, in order to generate X-rays with a maximum current of 30 mA. The X-ray tube does not have any inherent filtration except the exit window of 0.8 mm of Beryllium.

A second set of irradiations was carried out at the National Physical Laboratory (NPL) at two high voltages of 135 and 280 kV.

Beam qualities used in this study are listed in Table 1. For each beam quality, high voltage and external added filtration are listed, as well as two different beam specifiers: first half value layer (HVL) in aluminum, and the beam's effective energy (E_{eff}) in aluminum. Effective energy is defined as the energy of a mono-energetic beam having the same HVL as its equivalent poly-energetic beam. Values of HVL and E_{eff} were calculated with the SpekCalc software (Poludniowski et al., 2009).

Many codes of practice (IAEA, 2006) (Chair, 2001) (Klevenhagen

et al. 1996) recommend using the first HVL as beam quality specifier, for low energy X-rays. In this study, we used the effective energy as the sole beam quality specifier for the sake of good comparison of already published data. Effective energy can be directly related to the first HVL using the photon attenuation law and NIST's photon attenuation coefficients (Hubbell and Seltzer, 1995).

3.1. Correction factors

Three investigation methods were carried out in order to study the alanine dosimeter's relative response to kilo-voltage X-rays compared to ^{60}Co gamma rays. Based on obtained results, one can calculate correction factors that should be applied to absorbed dose to water measurement performed with alanine in the case of low to medium energy (up to 300 keV) X-ray irradiations. The correction factors can be directly related to the dosimeter's relative response by this equation:

$$k_{\text{corr}}^{Q, Q_0} = \frac{1}{f^{Q, Q_0}}$$

where k_{corr}^{Q, Q_0} is the correction factor to be applied directly to absorbed dose to water measured by alanine dosimeters, f^{Q, Q_0} is the alanine's relative response to a specific X-ray quality (Q) compared to a reference quality (Q_0) such as ^{60}Co . Thus, true absorbed dose to water can be expressed as such:

$$D_w^{Q_0} = k_{\text{corr}}^{Q, Q_0} \times D_w^Q$$

where $D_w^{Q_0}$ is the true absorbed dose to water, relative to a reference beam quality (Q_0), and D_w^Q is the absorbed dose to water measured by alanine dosimeters after irradiation with X-ray beam quality (Q). This method can be applied for irradiations at doses lower than 10 kGy, while at higher doses, the correction factor should be firstly applied to the dosimeter's response, due to the non-linearity of the latter versus absorbed dose to water (Goodman et al., 2017), thus, absorbed dose to water can be then correctly estimated using the EPR system's calibration curve.

3.2. Experimental measurements

Two sets of irradiations were done in order to determine alanine's response per dose to water unit, for a set of X-ray beam qualities, with respect to ^{60}Co reference quality. These irradiations consisted on determining this factor:

Table 1

X-ray beam qualities. Table represents (from left to right) high voltage, added filtration of aluminum, copper, tin, solid water, PMMA, and beam quality specifiers: first HVL and effective energy in aluminum.

Irradiation site	HV [kV]	Al [mm]	Cu [mm]	Sn [mm]	Solid Water [mm]	PMMA [mm]	HVL1 [mm]	E eff Al [keV]
Aerial	50	2.39	0	0	0	5	1.81	27.5
Aerial	70	2.88	0	0	0	5	2.65	31.9
Aerial	90	3.35	0	0	0	5	3.64	36.3
Aerial	100	1.43	0	0	0	5	2.57	31.5
Aerial	100	3.84	0	0	0	5	4.32	39.2
Aerial	100	4.95	0	0	0	5	4.93	41.7
Aerial	90	0.96	0	0	0	0	1.52	25.8
Aerial	100	1.43	0	0	0	0	2.18	29.5
Aerial	100	3.84	0	0	0	0	4.06	38.1
NPL	135	1.2	0.27	0	20	0	9.01	58.9
NPL	280	1	0.26	1.5	20	0	19.6	168

$$f_{exp} = \frac{(r/D_w)_Q}{(r/D_w)_{Q_0}}$$

where f_{exp} is the alanine's relative response measured by experimental setups, r is the alanine dosimeter's response measured by EPR spectrometry, D_w is the absorbed dose to water measured by a calibrated ion chamber, Q is one of the X-ray beam qualities and Q_0 is the ^{60}Co reference quality.

The first set of irradiations was carried out at Aerial with tube voltages ranging from 50 kV up to 100 kV. The absorbed dose to water was measured using a PTW 30013 waterproof Farmer ion chamber, which is calibrated in terms of absorbed dose to water for X-ray radiations qualities, with a traceability to PTB's water calorimetry secondary standard. The measured absorbed dose to water can be expressed as such:

$$D_w = M \cdot N_{D_w} \cdot k_Q \cdot k_{T,P}$$

where D_w is the corrected measurement of the absorbed dose to water, M is the ion chamber reading, N_{D_w} is the dose to water calibration coefficient, k_Q is the beam quality correction factor and $k_{T,P}$ is the air density correction factor.

The irradiation geometry consisted of 5 alanine dosimeters placed in a PMMA holder, as shown in Fig. 2, irradiated with a vertical photon beam. In addition, 1 cm of PMMA was placed below the dosimeters, which were positioned in such a way that the distance between the X-ray tube's focal point and the surface of the dosimeters was equal to 80 cm.

The second set of irradiations, carried out at NPL, consisted of placing 4 alanine dosimeters inside a solid water holder shaped as an ion chamber. This alanine holder was then placed in a 2 cm thick WT1

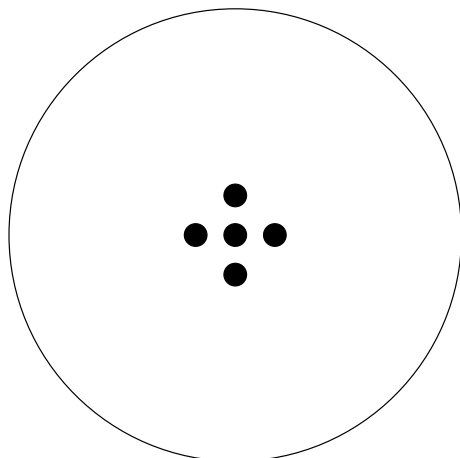


Fig. 2. Top view of the PMMA holder with 5 alanine dosimeters used for irradiations carried out at Aerial.

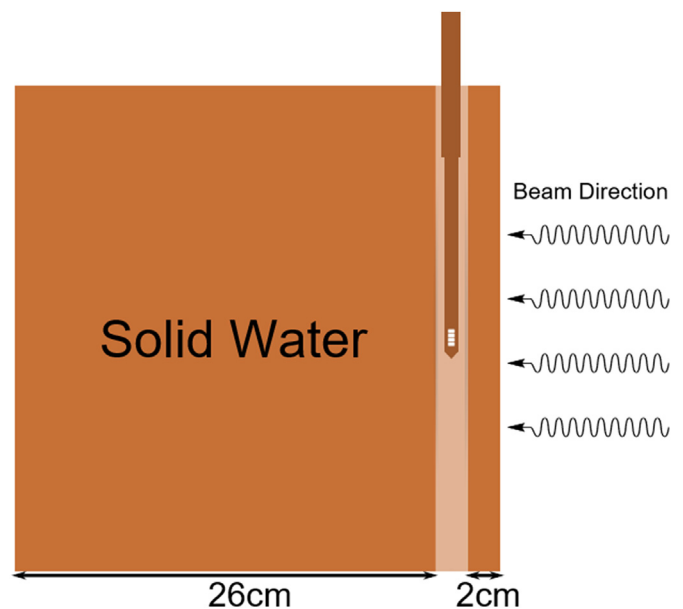


Fig. 3. Side view of the experimental setup using Solid Water phantom for irradiations carried out at NPL.

Solid Water slab. Solid Water slabs were purchased from Phoenix Dosimetry Ltd. Solid Water plates were added between the beam exit window and the dosimeters in order to have the alanine pellets at a water equivalent depth of 2 cm. A total thickness of 26cm of solid water slabs was added behind the dosimeters to ensure full backscatter conditions as shown in Fig. 3. Dose output was measured using NPL's PTW 30012 Farmer ion chamber, which is calibrated in air kerma, with a traceability to NPL's 300 keV free air chamber primary standard. Absorbed dose rate to water was determined using the IPEMB (Institute of Physics and Engineering in Medicine and Biology) code of practice using the in-phantom method for the determination of the absorbed dose to water.

A comparison between the two ion chamber readings (PTW 30013 dose to water calibration, and PTW 30012 air kerma calibration) was performed at NPL. Both chambers were positioned at 2cm water equivalent depth in the phantom showed in Fig. 3 and were irradiated in the same conditions (ambient temperature and pressure). Absorbed dose to water was measured by both chambers after 5 irradiations of 30 s each, at 135 kV and then at 280 kV. Results showed that there is a difference between the two measurements of both instruments. The average ratios of the PTW 30013 readings versus the PTW 30012 readings were 0.938 and 0.985 for irradiations at 135 kV and 280 kV respectively. Thus, in order to compare results obtained after irradiations at NPL, to the results of irradiations performed at Aerial, the absorbed dose to water, used to determine the f_{exp} factor, needs to be

corrected. This correction is needed to ensure that the alanine's response is always compared to a dose unit measured by the same reference instrument, which, in this study, is the PTW 30013 ion chamber.

3.3. Monte Carlo simulations

The MCNPX code (version 2.7.0) (D. B. Pelowitz, 2011) was used to calculate the ratio of the absorbed dose between alanine dosimeter and water. The electron and photon energy cut-offs were set to 1 keV. The true alanine dosimeter composition (93% alanine – 7% binder) was simulated in the code as well as the same experimental irradiation geometries. Cylindrical alanine pellets, placed in PMMA holders inside the irradiation cabinet at 80cm from the X-ray source, were simulated in the case of irradiations carried out at Aerial. Alanine dosimeters placed inside a Solid Water phantom at a depth of 2cm of water equivalent material and placed at 77cm from the X-ray source were simulated to reproduce irradiations carried out at NPL. The calculated factor is:

$$f_{MC} = \frac{D_{dos}}{D_w}$$

where f_{MC} is the alanine's relative response calculated by Monte Carlo Simulations, D_{dos} is the absorbed dose in the dosimeter and D_w is the absorbed dose in water.

Firstly, calculations were done using mono-energetic photon beams covering the range of 1 keV up to 10 MeV. The ratio of absorbed dose (dosimeter to water) was compared to the ratio of the mass energy absorption coefficients of alanine to water based on NIST's data.

Another set of calculations, taking as input X-ray spectra calculated by SpekCalc (see Table 1), were done to determine the dosimeter to water absorbed dose ratio, as a function of the X-ray spectra's effective energies.

3.4. Analytical calculations

A C++ code was developed to analytically calculate the ratio of absorbed dose in the dosimeter with respect to water, taking as entry the energetic fluence distribution given by SpekCalc, and then weighting the distribution by mass energy absorption coefficients published by NIST. The average execution time of the code does not surpass 5 s, for a computer with a central processing unit (CPU) running at a base frequency of 2.3 GHz. The calculated factor was inspired by a factor calculated in (Lessard et al., 2012) for a validation study of a plastic scintillator detector used for photon dosimetry. The calculated factor adapted to this study is:

$$f_w = \frac{\int_0^{E_{max}} \left(\frac{\mu_{en}}{\rho} (E) \right)_{dos} \cdot E \cdot \phi(E) \cdot dE}{\int_0^{E_{max}} \left(\frac{\mu_{en}}{\rho} (E) \right)_{water} \cdot E \cdot \phi(E) \cdot dE} \times \frac{(e^{\mu_{att} \cdot x})_{dos}}{(e^{\mu_{att} \cdot x})_{water}}$$

where f_w is the alanine's relative response determined by analytical calculations, E_{max} is the maximal photon energy of the X-ray spectrum, E is the photon energy at each integration step, $\mu_{en}/\rho(E)$ is the mass energy absorption coefficient at the energy E , $\phi(E)$ is the beam fluence at the energy E , μ_{att} is the average attenuation coefficient given by NIST and x is the dosimeter (or water) thickness.

The first term of the correction factor calculates the energy deposition of an incident photon beam inside the dosimeter and water material, while the second term calculates the beam percentage that is actually being attenuated in the dosimeter and water volume, thus the percentage that is responsible of the energy deposition.

3.5. Reference beam quality correction factor

In most cases, the alanine/EPR dosimetry system is calibrated in

terms of absorbed dose to water for a ^{60}Co reference beam (mean photon energy = 1.25 MeV). In other cases, alternative beam qualities are used as reference beam qualities such as high energy electron or X-ray beams (Araki and Kubo, 2002). Thus, when measuring alanine dosimeters that were irradiated with low to medium energy X-rays, a difference of the dosimeter's response will arise from the difference of the irradiation beam quality.

This difference is taken into account in the case of experimental measurements (see section 3.1) where the dosimeter's response is evaluated in the case of both qualities (X-rays and ^{60}Co gamma rays). However, this reference beam difference is not taken into account with factors determined by Monte Carlo simulations nor by analytical calculations, thus, in case of EPR measurements with a different reference beam quality calibration, a second factor should be added to the ones described in sections 3.2 and 3.3, which is:

$$f_{Q_0} = \left(\frac{D_{dos}}{D_w} \right)_{Q_0}$$

where f_{Q_0} is the alanine to water absorbed dose ratio for the reference beam quality, Q_0 is the reference beam quality, D_{dos} is the absorbed dose in the dosimeter and D_w is the absorbed dose to water.

Therefore, the correction factors become:

$$f_{MC}^{Q, Q_0} = f_{MC} / f_{Q_0}$$

$$f_w^{Q, Q_0} = f_w / f_{Q_0}$$

where f_{MC} and f_w are the alanine's relative responses determined by Monte Carlo simulations and analytical calculations respectively, as described in sections 3.2 and 3.3.

4. Results and discussion

4.1. Experimental measurements

Alanine's response per absorbed dose to water (r/D_w), using X-ray qualities with respect to ^{60}Co reference quality, was measured. Obtained results are plotted in Fig. 4. Results are compared to ones reported by Anton (Anton and Büermann, 2015) and Waldeland (Waldeland et al., 2010). A good agreement is noticed between the three data series over all the energy range. Waldeland's data were studied as a function of effective energy, whereas Anton's data were published as a function of the X-ray spectra's average energy. A study done by (Butler et al., 2016), using Anton's published data, showed a conversion of Anton's used average energies into effective energies, and these are the values that are reported in Fig. 4. Error bars in Fig. 4 represent uncertainties (at $k = 1$). An uncertainty budget of 2.5% ($k = 1$) was calculated in the case of irradiations carried at Aerial, and another budget of 2.4% ($k = 1$) was calculated in the case of irradiations done at the NPL X-ray irradiation facility.

Anton mentioned in his work (Anton and Büermann, 2015) that alanine's relative response drops down to 64% in case of low energy X-ray irradiations, which one can notice in Fig. 4 at an effective energy of 15.9 keV. Waldeland (Waldeland et al., 2010) stated that the experimental relative response of alanine varied from 0.68 up to 0.9 over the studied effective energy range (32 keV up to 99 keV). Results obtained in this work are consistent with Anton and Waldeland's data over the studied effective energy range (25 keV up to 168 keV), yet, one can notice that results obtained in this work are higher than published data. This difference is mainly due to the difference in size and composition of the alanine dosimeters between the three studies, as well as the difference in the used experimental setup.

In Anton's study, the experimentally determined relative response consists of the ratio of the measured dose by a ^{60}Co calibrated alanine/EPR system versus the delivered dose, whereas Waldeland used Monte Carlo simulations as well as experimental measurements in order to

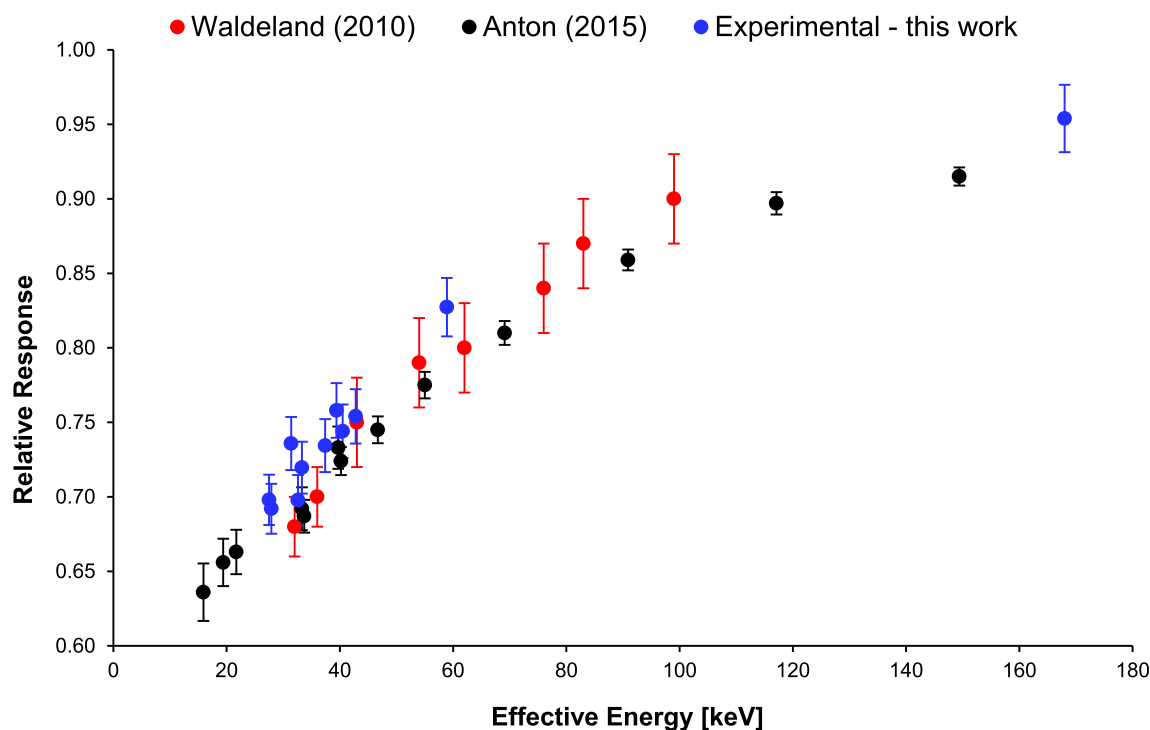


Fig. 4. Aerial's alanine dosimeter response (blue) at different beam qualities, compared to published data (Anton - black) (Waldeland - red). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

estimate the same response presented in section 3.2. Both approaches showed very similar results, as presented in Fig. 4. However, it was reported in (Goodman et al., 2017) that alanine's response is non-linear versus absorbed dose to water for doses higher than 10 kGy, thus, one should be careful to apply the adequate correction factor to the dosimeter's response rather than apply it directly to the measured dose for doses higher than 10 kGy.

We can also note that a convergence tendency is observed in case of effective energies that are higher than 150 keV, yet, true water equivalency (relative response equal to unity) is not observed in the studied effective energy range.

4.2. Monte Carlo simulations

Ratios of the absorbed dose in the dosimeter with respect to absorbed dose to water were calculated using MCNPX simulations. The calculated ratios are represented in Fig. 5, as well as data published by Anton and Waldeland. For this work, presented data correspond to the Monte Carlo factor listed in section 3.4, which takes into account the reference beam quality difference between X-rays and ^{60}Co . The same factor is calculated by Anton and Waldeland using EGSnrc Monte Carlo simulation code (Kawrakow et al., 2020). Error bars represent simulation standard deviations and experimental uncertainties ($k = 1$).

Obtained results are consistent with already published data (Anton and Büermann, 2015) (Waldeland et al., 2010). Plotted values of all three studies represent the same calculated factor. A difference between results is noticed, especially at low effective energies ranging from 15.9 keV to 40 keV, where we can see that results obtained in this study are higher than the ones obtained by Anton and Waldeland. This is due to many factors such as the difference in the used Monte Carlo code between this work (MCNPX) and published data (EGSnrc), the difference of the size and composition of the modelled alanine dosimeters and finally the difference of the irradiation geometry (phantom material, surrounding environment).

4.3. Analytical calculations

The alanine to water dose ratio was calculated analytically based on the weighting of X-ray spectra by mass energy absorption coefficients given by NIST, while taking into account the beam attenuation inside of the dosimeter volume. Fig. 6 shows the comparison of these factors to ones determined in this study by Monte Carlo simulations as well as experimental measurements.

All plotted results in Fig. 6 represent the dosimeter's relative response in case of X-ray irradiations with respect to ^{60}Co reference beam quality. The error bars represent uncertainties ($k = 1$).

Results obtained by analytical calculations and ones obtained by Monte Carlo simulations are in a very good agreement with an average standard deviation of 0.7%, while an average discrepancy of 3.2% is noticed between results obtained by calculations and experimental results. Average standard deviation of the results obtained by the three correction methods is 2.1%.

A clear difference is noticed between results obtained by Monte Carlo simulations and calculations in the first hand, and experimental measurements on the other hand. This difference was also reported in many studies (Anton and Büermann, 2015) (Waldeland et al., 2010) (Zeng and McCaffrey, 2005) (Khouri et al., 2015) (Waldeland and Malinen, 2011), and it arises from the fact that simulations and calculations take into account physical dose deposition only, without taking into account the free radicals creation process that is the base of measurements done by EPR spectrometry. Thus, the ratio of free radical creation yield for X-rays compared to ^{60}Co needs to be evaluated and then integrated in the determination of alanine's relative response by Monte Carlo simulations and analytical calculations.

5. Conclusion

Alanine dosimeter's relative response to kilo-voltage X-rays, compared to ^{60}Co reference beam quality, was studied in this work. The relative response was determined by three different methods. Studied

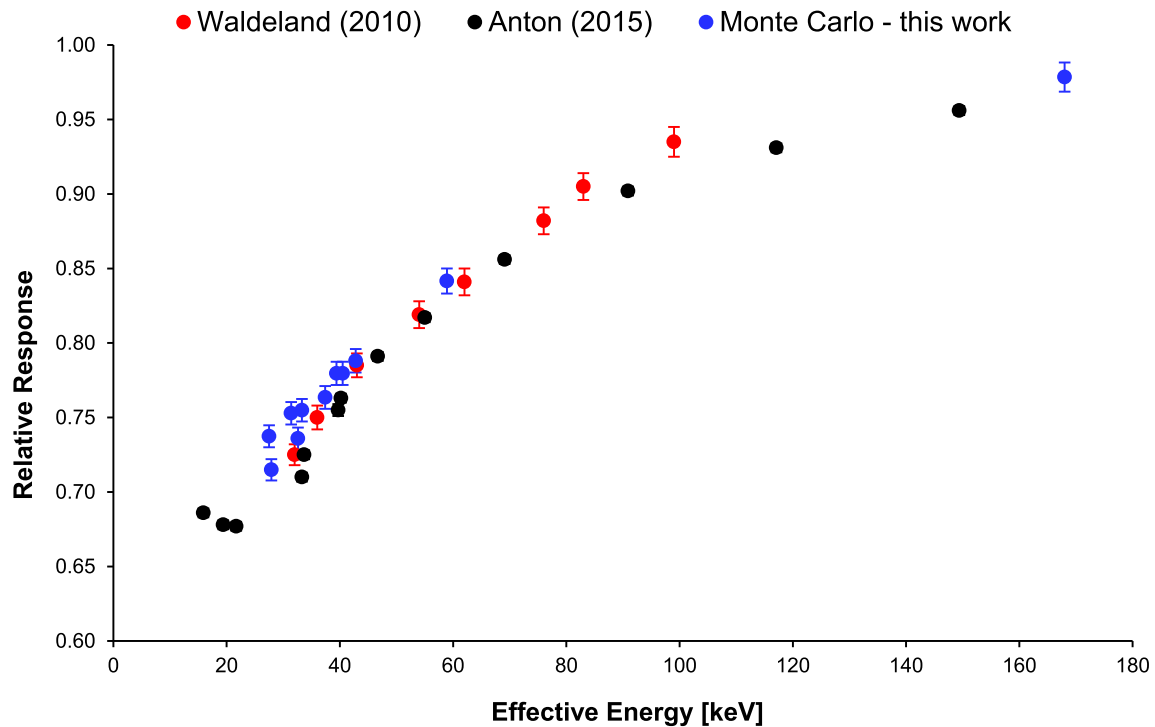


Fig. 5. Alanine to water dose ratio calculated by Monte Carlo simulations at different X-ray qualities with respect to ^{60}Co reference quality.

beam qualities covered the range of 50–280 kV (aluminum HVL: from 1.5 to 19.6 mm – aluminum effective energy: from 25 to 168 keV).

Experimental measurements were performed at Aerial and NPL to study alanine's relative response per dose to water unit for different X-ray beam qualities. Obtained results were in a good agreement with already published data (Anton and Büermann, 2015) (Waldeland and

Malinen, 2011) (Waldeland et al., 2010), yet a slight difference is noticed due to the difference in the dosimeter's composition and irradiation geometry.

Monte Carlo simulations and analytical calculations determined the ratio of the absorbed dose in alanine with respect to water. Results were in a good agreement with ones obtained by Waldeland and Anton, as

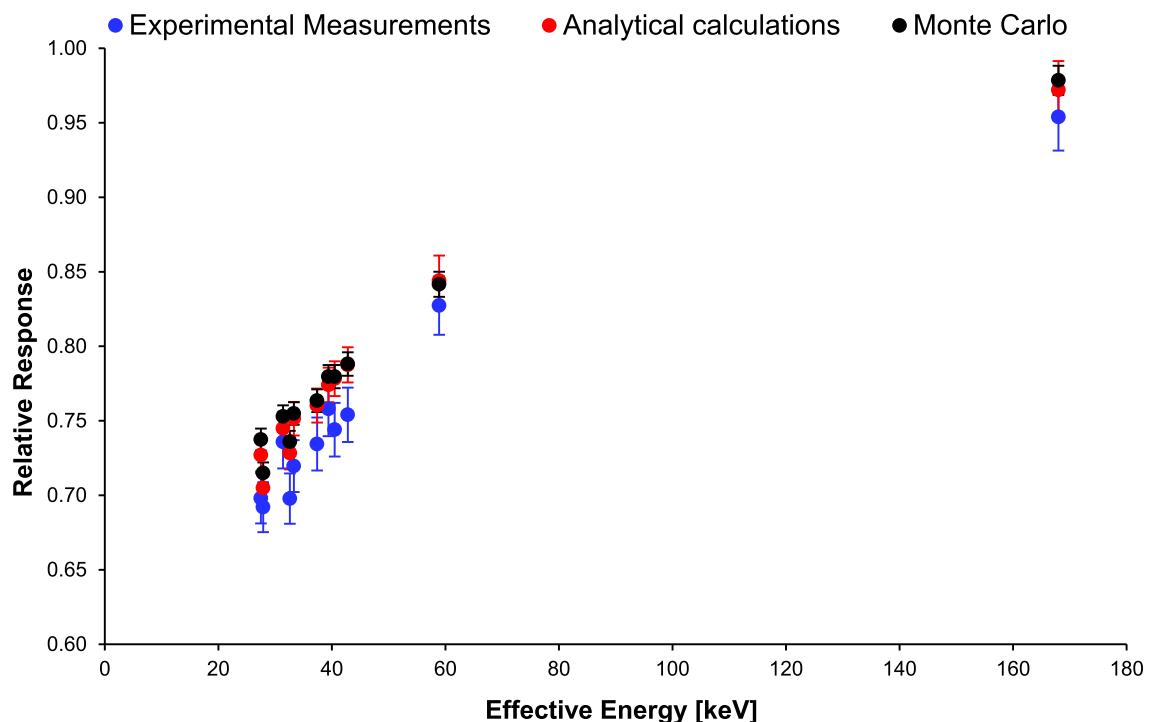


Fig. 6. Comparison of all alanine relative responses obtained in this work for X-ray beam qualities with respect to ^{60}Co reference quality.

well as with results obtained in this study by experimental measurements (average standard deviation of 2.1%, at $k = 1$).

Measuring absorbed dose to water with alanine dosimeters irradiated with kilo-voltage X-rays, using a ^{60}Co calibrated EPR system, requires application of correction factors to the dosimeter's measured EPR response. The three studied methods of determination of alanine's relative response to kilo-voltage X-rays ensure a good estimation of such correction factors.

Analytical calculations seem to be a simple, rapid and trustworthy method to determine correction factors to be applied to alanine's response measured by EPR spectrometry. Yet, the energy dependence of the free radical creation yield (G-value [radicals]/100eV) in alanine needs to be studied in order to integrate this phenomenon in both, simulations and calculations.

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