Comment

Comment on ‘Development of a primary standard for absorbed dose from unsealed radionuclide solutions’

C K Ross

1005 Southover Lane, Victoria, BC, V8Y 3C3, Canada

E-mail: carlkross@gmail.com

Received 23 February 2017, revised 3 June 2017
Accepted for publication 12 June 2017
Published 27 July 2017

Abstract
The authors of a recent paper (Billas et al 2016 Metrologia 53 1259–71) describe the development of a primary standard for the absorbed dose in an aqueous solution containing the radionuclide $^{90}$Y. By positioning a thin-window extrapolation chamber near the surface of the solution they determine the absorbed dose in the air cavity. Using Monte Carlo calculations and knowledge of the $^{90}$Y $\beta$-ray spectrum they relate the absorbed dose in air to that in the aqueous solution. I point out that there is no need to develop a new standard for this problem because knowledge of the activity of the radionuclide, combined with its decay characteristics, is adequate to establish a primary standard for the absorbed dose in water. Furthermore, the uncertainty using this approach is significantly smaller than that achieved using the authors’ technique. Results from Monte Carlo calculations are reported that: (1) determine the minimum phantom size required to establish charged particle equilibrium; (2) show that the authors have neglected bremsstrahlung losses from the phantom; (3) show why the authors’ results are very sensitive to the air gap between the solution and the window of the ionization chamber. Finally, I consider the problem of calibrating secondary detectors against the primary standard.

Keywords: absorbed dose to water, radionuclide solution, MIRD, ionization chamber, PENELOPE

(Some figures may appear in colour only in the online journal)

1. Introduction

Radionuclides play an important role in medicine for both diagnostic and therapeutic purposes. In general, some estimate of the absorbed dose in tissue much be established and is often referred to as the medical internal radionuclide dose or MIRD [1]. The radiation dose assessment resource (RADAR) [2] has been established as an on-line resource to help in the estimate of the MIRD. Basically, the MIRD is estimated from the known decay data of the radionuclides of interest.

Billas et al [3] review the general problem of measuring the absorbed dose due to distributed radionuclides in biological systems. They point out that the detector systems that have been used in the past often lead to large uncertainties. They then simplify the problem to considering a uniform distribution of $^{90}$Y in an aqueous solution and seek to establish the absorbed dose at a point removed from any boundaries so that charged particle equilibrium applies.

The approach they adopt is to place an ionization chamber close to the surface of the $^{90}$Y solution so that it responds to the electrons emitted from the surface. They use the ionization...
chamber to establish the absorbed dose in air and use Monte Carlo calculations to relate this result to the absorbed dose in water when charged particle equilibrium applies.

I will show that the equilibrium absorbed dose can be established if the activity of the solution is known and that the result is traceable to the SI. No reference to any other measurement of the absorbed dose is required, thus meeting the requirements for a primary standard [4]. In addition, this approach leads to a smaller uncertainty than achieved by the authors using an ionization chamber.

I will comment on some details of the authors’ work and provide clarification as to why their result is very sensitive to the air gap between the surface of the solution and the window of the ionization chamber. Finally, I will consider the problem of calibrating dosimeters against the primary standard. Suitable calibrated dosimeters can then be used to establish the absorbed dose when charged particle equilibrium does not apply.

2. Analysis

With a probability of 99.98 %, $^{90}$Y decays with the emission of a $\beta$-ray to the ground state of $^{90}$Zr. The decay is referred to as a first-forbidden unique transition [5, 6] and has an end-point energy of 2278.7(16) keV, where common notation is used to indicate the standard uncertainty. The $\beta$-ray spectrum can be calculated and has also been measured, permitting the results to be compared. Mougeot [7] has developed software to calculate the spectrum using different assumptions. The differences between the spectra are small and a typical spectrum is shown in figure 1.

Billas et al state that the MIRD result is not traceable to a primary standard of absorbed dose. However, I will show that, under conditions of charged particle equilibrium, the MIRD result is indeed traceable to the SI and thus can be considered to be a primary measurement. If we use $A$ to denote the activity per unit mass and $E$ to denote the average energy of the $\beta$-ray spectrum, then the absorbed dose rate in water, $\bar{D}_w$, is given by

$$\bar{D}_w = 1.602 \cdot 10^{-16} A E k_{brem}.$$  

where $\bar{D}_w$ will have units of Gy s$^{-1}$ if $A$ and $E$ are in units of Bq kg$^{-1}$ and keV, respectively. The correction factor, $k_{brem}$, corrects for energy carried away by bremsstrahlung radiation. The activity of $^{90}$Y can be established using primary standards based on liquid scintillation spectrometry or 4$\pi$/3 proportional counting. Zimmerman and Ratel [8] report on a comparison carried out between eight national laboratories. Agreement between the laboratories was excellent and the typical uncertainty reported was 0.3 %.

The $\beta$-ray spectrum of $^{90}$Y can be measured or calculated. The calculated spectrum requires knowledge of the end-point energy, which has an uncertainty of 0.07 % and is traceable to the SI. Mougeot [5] has compared the measured and calculated spectra and calculated the average energy in each case. Differences between the average energies are less than 0.5 %. For the spectrum based on a measured shape factor, Mougeot\(^1\) obtains a mean energy of 924.9 keV and we take 4.4 keV as the standard uncertainty. Combining in quadrature the uncertainties of $A$ and $E$ leads to a standard uncertainty for $\bar{D}_w$ of about 0.6 %.

The radiation yield for 930 keV electrons is about 0.36 % so $k_{brem}$ can be expected to be between 0.996 and unity but a more exact estimate will be discussed later.

The approach adopted by Billas et al is to place a thin-window ionization chamber close to the surface of the aqueous solution containing $^{90}$Y. By using an extrapolation chamber with a well-defined air volume they establish the absorbed dose rate in air, $\bar{D}_{air}$, due to the radiation emitted from the surface of the radionuclide solution. They then use Monte Carlo calculations to relate $\bar{D}_{air}$ to $\bar{D}_w$, the absorbed dose rate in water at a point in the solution far removed from any surfaces, and define $k_{MC}$ as $k_{MC} = \bar{D}_w/\bar{D}_{air}$. They write that

$$\bar{D}_w = \bar{D}_{air} \cdot k_{MC},$$

and claim that a measurement of $\bar{D}_{air}$ using the extrapolation chamber leads to a primary standard for $\bar{D}_w$ with an uncertainty of about 1.6 %.

A primary standard should permit the calibration of secondary detectors but it is not clear if the water phantom developed by Billas et al could be used for that purpose. The continuous slowing down approximation (CSDA) gives 1.1 cm for the range of 2.3 MeV electrons in water [9] but their water phantom is only 1.62 cm deep. Although this is likely deep enough to provide an equilibrium electron spectrum from the surface there may be no location where charged particle equilibrium prevails. In order to test this possibility, a cylindrical water phantom was constructed, 4 cm in radius and 2.8 cm deep and the absorbed dose was calculated as a function of depth using the PENELOPE [10] Monte Carlo code and its user code penEASY [11]. The $\beta$-ray source spectrum was submitted to PENELOPE using 7 keV energy bins.

\(^1\) Private communication.
The absorbed dose was tallied in 2 mm bins along the cylindrical axis and in a radial bin 2 cm in radius. The results are presented in figure 2 and show that equilibrium is established at a depth of about 0.9 cm. Thus, a phantom with depth of 1.62 cm is not adequate to provide a position where secondary dosimeters could be calibrated. A depth of at least 2.8 cm would provide a region of about 1 cm where the absorbed dose could be considered constant.

Figure 2 also provides an estimate for $k_{\text{brem}}$. The Monte Carlo calculation was repeated but with photon transport disabled and the photon energy deposited where it was created. These data points lie above the full simulation by about 0.39% thus giving a value for $k_{\text{brem}}$ of 0.996. This result can be expected to have some dependence on the phantom size.

From the description given by Billas et al., they do not account for photon loss when calculating their MIRD value. The mean value of the $\beta$-ray spectrum used for the Monte Carlo calculations is 924.9(44) keV and this gives the MIRD result, ignoring bremsstrahlung loss, shown in figure 2. Note that this value differs by 0.4% from the true absorbed dose.

The authors calculate $k_{\text{MC}}$ using Monte Carlo techniques and it has a value of approximately 2.5. The reason the factor is so large can be understood from the fact that their ionization chamber is responding to electrons emitted from only one surface of the phantom while at depth electrons are contributing from two (imaginary) surfaces. The authors’ state that ‘The MC-calculated correction factor was a dose ratio and so does not depend directly on the absolute values calculated’. Although this statement might apply if one were calculating nearby points on a depth-dose curve it is not relevant in this case. As the authors point out, the absorbed dose at depth is not strongly dependent on radiation transport although energy carried away by photons needs to be accounted for. On the other hand, the calculation of $D_{\text{air}}$ depends on the detailed transport of electrons through the phantom surface, the air gap and the ionization chamber.

The authors show that $k_{\text{MC}}$ is quite sensitive to the air gap between the surface of the phantom and the ionization chamber window. This sensitivity leads to the largest contribution to their uncertainty budget and amounts to 1.15%. They state that the sensitivity is due to several factors including ‘divergence of the electric field inside the extrapolation chamber collecting volume …’. They do not describe any calculations of the electric field and the radiation transport calculations do not include any such effect. However, it is interesting to determine why the ionization chamber response changes by about 2% per mm of air gap.

A simplified model of their apparatus was constructed. The dimensions were similar to those described by Billas et al. but all materials were simulated as either air or water. Radiation transport was carried out using PENELQOPE and penEasy. The source spectrum was as shown in figure 1 but emission was confined to a 45°cone in the forward direction in order to reduce computation time. The low-energy cutoff for radiation transport was set to 10 keV in the water phantom but was reduced to 1 keV for transport in air.

The energy deposited in the air cavity was tallied and was found to decrease by 1.6(2)% when the air gap was changed from 2 mm to 3 mm. This result compares reasonably well with the value of 2% obtained by Billas et al. The spectrum of electrons striking the window of the ionization chamber is shown in figure 1. The spectra for the different gaps are indistinguishable on this scale. The total number of electrons striking the window differ by only 0.67(6)% for the two gaps, with there being fewer electrons for the larger gap. Figure 3 shows the ratio of the spectra for the two gaps and it is clear that low energy electrons are preferentially removed as the air gap increases. Because the stopping power increases rapidly
as the electron energy decreases, their removal from the spectrum leads to less energy deposition in the ionization chamber than would be expected from the relatively small change in the total number of electrons.

One of the main purposes of a primary standard is to permit the calibration of secondary detector systems. If a small detector were placed near the centre of the absorbed dose distribution depicted in figure 2, then a calibration factor, \( N_{D_w} \), can be obtained as

\[
N_{D_w} = \frac{D_w}{R_d}, \tag{3}
\]

where \( R_d \) is the detector reading per unit time in response to the absorbed dose rate, \( D_w \). Because the detector displaces solution containing radionuclide, its response will decrease as the size of the detector increases. It order to estimate this effect, a volume was isolated near the centre of a water-filled cylindrical phantom, 1.5 cm in radius and 3 cm thick. This volume, representing an idealized detector, was taken to be a cylinder, 2.5 mm in radius, containing water but no \(^{90}\)Y. The absorbed dose was tallied in this volume for several thicknesses and compared to the absorbed dose under conditions of charged particle equilibrium. The results are shown in figure 4 and indicate that the detector should not be much thicker than 1 mm if its response is to be at least 75% of maximum. If the detector’s calibration factor is to be compared to that obtained in other radiation fields, such as \(^{60}\)Co γ-rays, its response must be corrected by data similar to that shown in figure 4.

EBT3 radiochromic film (GafChromic EBT3; International Specialty Products, Wayne, NJ) may be a useful secondary dosimeter for applications in nuclear medicine. It has recently found application for \( \alpha \)-particle dosimetry [12] but it had to be modified because of the short range of the \( \alpha \) particles. No modification should be required for \(^{90}\)Y dosimetry. It consists of a sensitive layer, 28 µm thick, sandwiched between two protective polyester layers, each 125 µm thick. The model described above does not apply in this case because the protective films do not participate in the detector response. The model was refined to simulate the structure of the film, again taking all materials to be water-equivalent. The absorbed dose to the sensitive layer was found to be 89% of the unperturbed absorbed dose.

3. Conclusions

Billas et al claim to have developed the first absorbed dose standard for an aqueous solution containing \(^{90}\)Y. I argue that the estimation of absorbed dose based on \(^{90}\)Y decay data already meets the requirements for a primary measurement. In fact, the uncertainty of this approach is less by about a factor of 2.5 than that obtained using the extrapolation chamber. The authors seem to have overlooked the correction for bremsstrahlung loss which amounts to an effect of about 0.4%. On several occasions, the authors describe their technique as a ‘direct measurement’ of the absorbed dose in water. No measurement is carried out at the point in the phantom where the absorbed dose is desired and a detailed Monte Carlo calculation is required to relate the measured quantity to the absorbed dose in water. It is not appropriate to describe this as a ‘direct measurement’. In fact, it may be more productive to consider their work as a valuable test of recent Monte Carlo codes and the associated stopping power data rather than a new standard.

Acknowledgments

Dr Josep Sempau of the Universitat Politècnica de Catalunya helped with the PENEOPE and penEasy Monte Carlo software. Dr Xavier Mougeot of the Laboratoire National Henri Becquerel provided useful information and advice on the \( \beta \)-ray spectrum of \(^{90}\)Y.

ORCID

C K Ross ∗ https://orcid.org/0000-0002-6608-3975

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