Impact of the $I$-value of diamond on the energy deposition in different beam qualities

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Abstract

Diamond detectors are increasingly employed in dosimetry. Their response has been investigated by means of Monte Carlo (MC) methods, but there is no consensus on what mass density $\rho$, mean excitation energy $I$ and number of conduction electrons per atom $n_{ce}$ to use in the simulations. The ambiguity occurs due to its seeming similarity with graphite (both are carbon allotropes). Except for the difference in $\rho$ between crystalline graphite (2.265 g cm$^{-3}$) and diamond (3.515 g cm$^{-3}$), their dielectric properties are assumed to be identical. This is incorrect, and the two materials should be distinguished: ($\rho$ = 2.265 g cm$^{-3}$, $I$ = 81.0 eV, $n_{ce}$ = 1) for graphite and ($\rho$ = 3.515 g cm$^{-3}$, $I$ = 88.5 eV, $n_{ce}$ = 0) for diamond. Simulations done with the MC code PENELOPE show that the energy imparted in diamond decreases by up to 1% with respect to 'pseudo-diamond' ($\rho$ = 3.515 g cm$^{-3}$, $I$ = 81.0 eV, $n_{ce}$ = 0) depending on the beam quality and cavity thickness. The energy imparted changed the most in cavities that are small compared with the range of electrons. The difference in the density-effect term relative to graphite was the smallest for diamond owing to an interplay effect that $\rho$, $I$ and $n_{ce}$ have on this term, in contrast to pseudo-diamond media when either $\rho$ or $I$ alone were adjusted. The study also presents a parameterized density-effect correction function for diamond that may be used by MC codes like EGSnrc. The ESTAR program assumes that $n_{ce}$ = 2 for all carbon-based materials, hence it delivers an erroneous density-effect correction term for graphite and diamond. Despite the small changes of the energy imparted in diamond simulated with two different $I$ values and expected close-to-negligible deviation from the published small-field output correction data, it is important to pay attention to material properties and model the medium faithfully.

1. Introduction

Clinical dosimetry in radiation therapy was traditionally based on cavity theory where the absorbed dose to the detector cavity medium is converted to absorbed dose to water by means of the ratio of mass electronic stopping powers of water to detector cavity medium (Bragg–Gray and Spencer–Attix cavity theories), the ratio of mass energy-absorption coefficients (large-cavity theory), or both (Burlin cavity theory) (Andreò et al 2017). These formalisms rely on certain assumptions that may introduce systematic uncertainties if not fulfilled (e.g. when the radiation field is perturbed by the presence of the detector cavity). Nowadays computational power is no longer a problem, and the physics models underlying general-purpose Monte Carlo (MC) radiation transport codes are accurate enough and have been benchmarked even for low-energy electron transport (Sempau et al 2003, Ali and Rogers 2008). Hence, the absorbed dose energy response of dosimeters and the perturbation of radiation fluence caused by extra-cameral components and non-water-equivalence of the detector cavity itself can be determined...
The reliability of MC simulations depends on the accuracy of the cross-section data underlying radiation transport, the simulation parameters that control radiation transport, as well as modeling of detector and experimental setup geometry, and material properties. The material properties have either pre-defined data (for instance, adapted from Berger et al. 2017) or are declared manually. User-selected input parameters are, typically, chemical composition, mass density $\rho$ and mean excitation energy (the $I$-value). Modeling of the most common detectors with cavities made of, for instance, air, silicon, or lithium fluoride, is rather straightforward because their values of $\rho$ and $I$ are well established. However, this is not the case for diamond detectors, which have gained interest and are being employed in high-energy photon- and electron beam dosimetry (Laub and Crilly 2014, Ralston et al. 2014, Di Venanzio et al. 2015, De Coste et al. 2017) as well as in proton (Gomà et al. 2016) and carbon (Marsolat et al. 2016, Rossomme et al. 2016) beam dosimetry. They have also been used in kilovoltage (kV) photon beams (Dadmore et al. 2018, Kaveckyte et al. 2020) and in brachytherapy (BT) (Kaveckyte et al. 2018, Rossi et al. 2019). The suitability of diamond detectors in very-high-energy and high-dose-rate FLASH radiotherapy dosimetry is inconclusive. The detectors were reported to exhibit large dose-rate dependence in a very-high-energy electron beam (Favaudon et al. 2019) but not in a proton beam (Patriarca et al. 2018).

A problem in the MC modeling of diamond arises because of its seeming similarity to graphite. Both materials are allotropes of carbon, i.e. they have the same atomic number ($Z = 6$), but differ in their crystalline structure, mass density ($\rho = 2.265$ g cm$^{-3}$ for crystalline graphite, $\rho = 3.515$ g cm$^{-3}$ for diamond) and dielectric response function (DRF) (graphite is a conductor whereas diamond is an insulator). Within the dielectric formalism, the $I$-value can be deduced knowing the optical energy loss function (OELF) of the medium, i.e. the imaginary part of $-1$ divided by the DRF (Fano 1963). Thus, it is expected that the $I$-value of diamond differs from the mean excitation energy of graphite ($I = 81.0$ eV). By modeling the OELF of diamond, which is shifted towards higher energy losses compared to that of graphite, Fernández-Varea et al. (2021) concluded that $I = (88.5 \pm 2.0)$ eV for diamond.

In spite of the growing application of diamond detectors for dosimetry, the International Commission on Radiation units and Measurements (ICRU) has not made any distinction between graphite and diamond regarding the mass density, the $I$-value and the number of conduction electrons per atom, $n_{ce}$, presumably due to the limited use of diamond detectors in the past. On the other hand, the data sets for graphite were updated several times because it is the typical material of the wall and entrance window of cylindrical and plane-parallel air ionization chambers, respectively, and the electrode material in both types of ionization chambers employed for reference dosimetry in radiation therapy. Graphite is also used in calorimetry as a primary standard. Recently, the reference data for graphite in the ICRU Report 37 (ICRU 1984) were updated in the ICRU Report 90 (ICRU 2016). Specifically, the crystalline mass density of 2.265 g cm$^{-3}$ together with the $I$-value of 81.0 eV should be used to calculate the mass electronic stopping power of light and heavy charged particles with, respectively, the Bethe and Bethe–Bloch formulas and $n_{ce}$ was reduced from 2 to 1.

MC simulations of diamond detector response are necessary for the calculation of small-field high-energy photon beam output correction factors where cavity theories break down. Since the corrections are around a few percent (De Coste et al. 2017 and references therein), a high accuracy of detector modeling is essential. Additionally, the use of MC-calculated beam quality correction factors allows for non-relative BT dosimetry where the detector calibration beam quality (generally $^{60}$Co or 6 MV photon beams) is very different from the user’s beam quality. Most MC studies do account for the dissimilar mass densities of crystalline graphite and diamond (Mobit et al. 1997, Scott et al. 2012, Bouchard et al. 2015, Andreo and Bennakhloff 2017, Fenwick et al. 2018, Kaveckyte et al. 2020). The choice of the $I$-value varies and only a couple of investigations have employed $I = 87.6$ eV (Górka et al. 2006, 2008, Marsolat et al. 2015). Other articles do not specify the $I$-value at all, but it is reasonable to assume that it was the same as that recommended for graphite (78 eV or 81.0 eV depending on the date of publication). The number of conduction electrons is not specified either and is likely to correspond to that of graphite (2 or 1 depending on the date of publication). Moreover, some authors use the mass density of crystalline graphite in the calculation of the mass electronic stopping power of diamond but the actual mass density of diamond in MC simulations (Mobit et al. 1997, Andreo and Bennakhloff 2017). This can further lead to the misinterpretation of the simulation results, especially given an ongoing discussion on what causes the over-response of diamond compared to water (Scott et al. 2012, Bouchard et al. 2015, Andreo and Bennakhloff 2017, Andreo 2018a, Fenwick et al. 2018).

Therefore, given the increasing popularity of synthetic diamond dosimeters, the purpose of the present work was to highlight and clarify which properties of diamond should be adopted in MC simulations and for the calculation of mass electronic stopping powers. Besides, we investigated the implications of a higher $I$-value for diamond on energy deposition in this material. Since it is anticipated that the change depends on the size of the diamond cavity and the beam quality, several realistic scenarios were tested; namely, 25–180 kV therapeutic...
x-ray beams, BT sources (photons sources $^{125}$I, $^{169}$Yb and $^{192}$Ir, and a $\beta$-source $^{106}$Ru), $^{60}$Co calibration beam, 6 MV photon beams from a linear accelerator (large and small fields) and a 100 MeV electron beam.

2. Background

The relativistic Bethe formula for the mass electronic stopping power $S_d(E)/\rho$ of swift light charged particles (electrons and positrons) quantifies their average energy loss per unit mass thickness in the medium as a function of kinetic energy $E$ (Bethe 1932). Following the notation in ICRU Reports 37 and 90, the equation can be written as

$$S_d(E)/\rho = 2\pi r_e^2 m_e c^2 Z A^2 1/\beta^2 \left[ 2 \ln \left( \frac{E}{M} \right) + \ln \left( 1 + \frac{\tau}{2} \right) + F^\pm(\tau) - \delta \right], \quad (1)$$

where $r_e$ is the classical electron radius, $m_e c^2$ is the electron rest energy, $m_\infty$ is the atomic mass unit, $\beta = v/c$ is the ratio of the incident particle velocity to the speed of light, $\tau = E/(m_e c^2)$, $Z$ and $A$ are the atomic and mass numbers of the atoms that make up the medium, respectively, and $I$ is the mean excitation energy of the medium. The analytical functions $F^\pm(\tau)$ solely depend on the incident particle type and its energy (ICRU 1984, 2016). The density-effect correction term $\delta$ (Fano 1963, Sternheimer et al 1984 and references therein) accounts for the polarization of the atoms (or molecules) in a dense medium when atoms farther from the charged-particle track experience a weaker Coulomb field exerted on them. Hence, the energy losses of a charged particle traversing a dense medium are smaller than those in a gas with the same composition. At ultra-relativistic energies ($\beta\rightarrow1$) the $\delta$-term dominates in equation (1) and approaches the asymptotic form

$$\delta(\beta) \rightarrow 2 \ln \left( \frac{1}{\sqrt{1 - \beta^2}} \frac{\Omega_p}{I} \right) - 1, \quad (2)$$

where $\Omega_p$ is the plasma energy of the medium; it is calculated as

$$\Omega_p = (28.8159 \text{ eV}) \frac{Z}{A} \frac{\rho}{g \text{ cm}^{-3}}. \quad (3)$$

Therefore, in the asymptotic limit, when equation (2) is plugged in equation (1), the mass electronic stopping power no longer depends on the $I$-value.

The only term in equation (1) whose evaluation is not straightforward is the density-effect correction. In principle, $\delta$ ought to be computed from a set of two coupled equations that involve the full DRF and an integral of the associated OELF of the considered medium (Fano 1963, Inokuti and Smith 1982). Unfortunately, for most materials the information on the DRF is scarce or fragmentary. To circumvent this shortcoming, Sternheimer developed a simplified model (see Sternheimer et al 1982, 1983, 1984, and references therein) where the DRF and the corresponding OELF are replaced with a sum of Dirac delta functions (‘oscillators’), one per atomic (sub)shell. The position and weight (‘strength’) of each delta function are related to the (sub)shell binding energy and occupation number, respectively. In the case of a conductor, an oscillator is added with a null binding energy and a strength equal to the number of conduction electrons per atom, $n_c$. Sternheimer’s method has a free parameter that is fixed by imposing that the mean excitation energy resulting from the simplified OELF reproduces a preselected $I$-value. It is worth mentioning that the tables of $S_d/\rho$ and $\delta$ in ICRU Reports 37 and 90 were calculated from equation (1) in conjunction with Sternheimer’s method. On the other hand, Fano (1963) introduced a slight refinement in the simplified calculation of $\delta$ where the said coupled equations are first written in terms of only the OELF, which is then expressed as a sum of Dirac delta functions. This strategy is implemented e.g. in the PENELOE MC code (Salvat 2019).

When either the Sternheimer or the Fano simplified calculation scheme of the $\delta$-term is adopted, equation (1) can be rewritten as (see Andreo and Bennakhlef 2017)

$$S_d(E)/\rho \propto \frac{Z}{A} \frac{1}{\beta^2} [\xi(\beta; I) + F^\pm(\beta) - \delta(\beta; \rho, I, n_c)]. \quad (4)$$

The non-trivial material-dependent terms in equation (4) are $\xi$ and $\delta$, and their contribution to $S_d/\rho$ depends on the kinetic energy of the particle. The reduction in $S_d/\rho$ caused by the $\delta$-term depends not only on the mass density of the medium as the name suggests, but also on its DRF. While the dependence on the $I$-value has been emphasized by Andreo and Bennakhlef (2017), the role of $n_c$ is still overlooked.

To facilitate the practical application of the numerical values of $\delta$, the following fitting formula has been extensively used (Sternheimer et al 1984 and references therein)
### 3. Methods

#### 3.1. Density-effect correction term

Since there is no consensus on which parameter values \((\rho, I, n_c)\) to pick for diamond, various sets have been considered here for the calculation of \(S_{\text{dil}}/\rho:\)

1. \(\delta_g \equiv \delta(\beta; 2.265 \text{ g cm}^{-3}, 81.0 \text{ eV}, 1);\) this set is recommended to calculate \(\delta\) and \(S_{\text{dil}}/\rho\) for graphite in ICRU Report 90 (ICRU 2016).
2. \(\delta_{\text{pd1}} \equiv \delta(\beta; 3.515 \text{ g cm}^{-3}, 81.0 \text{ eV}, 1);\) these values are probably used in most modern MC simulations for diamond, and they account for the difference in mass density between crystalline graphite and diamond but not in dielectric properties.
3. \(\delta_{\text{pd2}} \equiv \delta(\beta; 3.515 \text{ g cm}^{-3}, 88.5 \text{ eV}, 1);\) this parameter set does not make the distinction that diamond is, unlike graphite, an insulator.
4. \(\delta_{\text{d}} \equiv \delta(\beta; 3.515 \text{ g cm}^{-3}, 88.5 \text{ eV}, 0);\) this set models diamond more faithfully than the previous ones.

The density-effect correction terms \(\delta_g, \delta_{\text{pd1}}, \delta_{\text{pd2}}\) and \(\delta_{\text{d}}\) were calculated with Sternheimer’s formalism (Sternheimer et al 1984 and references therein). For comparison, \(\delta_g\) and \(\delta_{\text{d}}\) were also computed with Fano’s simplified method (Fano 1963).

The adjustable parameters for the fitting function \(\delta(X)\) of diamond \((\delta_{\text{d}})\) were determined with the Levenberg–Marquardt iterative algorithm. The iterations were discontinued when the \(\chi^2\) tolerance value reached \(10^{-8}\). The goodness of fit was evaluated by comparing the residuals with the typical values in Sternheimer et al (1984).

#### 3.2. MC simulations of energy deposition

The general-purpose MC code system PENETOPE (version 2018) (Salvat 2019) and the main program penEasy (version 2019-09-21) (Sempau et al 2011) were utilized to score the energy imparted in a diamond cavity by several beam qualities. PENETOPE implements detailed simulation of photon interactions and a mixed (class II) electron transport algorithm. The geometry input files were created defining bodies delimited by quadric surfaces, which were coded according to the PENGEOm conventions and syntax.

The following beam qualities were investigated:

1. Kilovoltage photon beams: 25, 50, 100 and 180 kV corresponding to the CCRI x-ray beam qualities (Kessler and Burns 2018) (table 1). The diamond cavity was located in the middle of a cylindrical water volume with a thickness of 2 mm and a 2 mm radius to create build-up for electrons. The source-to-diamond distances were 50 cm for the 25 and 50 kV beams, and 100 cm for the 100 and 180 kV beams corresponding to detector calibration conditions in kV beams. The x-ray spectra were generated with the SpekCalc software (Poludniowski et al 2009). The program was fed with the tube potential, added filtration material and thickness, and half-value-layer value of each kV beam quality.
II. BT sources: $^{106}$Ru (average $\beta$ energy 1.4 MeV), $^{125}$I (average photon energy 28 keV), $^{169}$Yb (average photon energy 93 keV with major emissions at 50 and 200 keV), $^{192}$Ir source (average photon energy 350 keV). The sources were approximated as being point-like and placed at the center of a $20 \times 20 \times 20$ cm$^3$ water phantom to mimic BT measurement conditions. The source-to-diamond distances were chosen as the reference points used in the corresponding BT beam qualities, i.e., 2 mm for $^{106}$Ru (Thomson et al. 2019), 10 mm for $^{125}$I, $^{169}$Yb and $^{192}$Ir sources (Rivard et al. 2004). The decay data for the $^{125}$I and $^{192}$Ir radioactive sources were taken from Rivard et al. (2004) and Borg and Rogers (1999), respectively, whereas those of $^{169}$Yb and $^{106}$Ru were generated with PENNUC (García-Toraño et al. 2019).

III. 60Co calibration beam (two major gamma emissions at 1.17 and 1.33 MeV). The detector calibration conditions of the TRS-398 protocol (Andreou et al. 2000) were followed, i.e., the diamond cavity was at 5 cm reference depth in a $30 \times 30 \times 30$ cm$^3$ water phantom, the source-to-surface distance was 95 cm and the field size was $10 \times 10$ cm$^2$ at the reference depth.

IV. 6 MV photon beam. The simulation geometry was modeled complying with the small-field output factor measurement conditions (Palmans et al. 2017), i.e., the diamond cavity was at 10 cm depth in a $30 \times 30 \times 30$ cm$^3$ water phantom. The source-to-surface distance was 100 cm, and the field sizes were $10 \times 10$ cm$^2$ and $0.5 \times 0.5$ cm$^2$ at the surface to simulate large and small fields, respectively. The photon energy spectra for these field sizes were extracted from the IAEA phase space files of a Varian Clinac iX 6 MV beam for the corresponding field sizes (Hedin et al. 2010). The bin width was 30 keV. The information about photon incidence angle and position was lost but this was assumed to have a negligible impact on the results because we compared ratios of energy imparted in the diamond cavity.

V. 100 MeV very-high-energy electron beam. The diamond cavity was at a depth of 10 cm in a $30 \times 30 \times 30$ cm$^3$ water phantom. The electron beam was defined as a point source at 100 cm distance from the water surface and the field size was $2 \times 2$ cm$^2$ at the surface, which is similar to the existing pre-clinical dosimetry measurements (Subiel et al. 2017, Favaudon et al. 2019).

In all cases, the scoring volume was a bare cylinder of diamond with a diameter of 2.2 mm. Four thicknesses of the cylinder were considered, namely 1, 3, 10 and 30 $\mu$m. In particular, 2.2 mm diameter and 1 $\mu$m thickness match the nominal dimensions of the active volume of the only commercially available synthetic diamond detector, microDiamond (PTW, Freiburg, Germany). The other thicknesses were included to assess the dependence of the energy imparted in the cavity on its thickness. The cross-section files for the ‘pseudo-diamond’ and diamond media were created manually running PENELOPE’s material.exe pre-processor, setting $\rho = 3.515$ g cm$^{-3}$ in both cases and the $I$-value to either 81.0 or 88.5 eV. The oscillator strength ($f_{\text{os}}$) and energy ($W_{\text{cb}}$) of the plasmon were set to 0 because diamond is an insulator, i.e., $n_{\text{ee}} = 0$.

Regarding the simulation parameters, the electron and positron cutoff energies were 1 keV in the scoring volume and neighboring regions and 10 keV in other regions. The cutoff energy for photon transport was set to 1 keV everywhere. The threshold energy losses for detailed inelastic collisions ($W_{\text{CC}}$) and hard radiative interactions ($W_{\text{CR}}$) were 1 keV, the elastic collision parameters ($C_1$ and $C_2$) were 0.05 in the scoring volume and neighboring regions within the secondary electron range and 0.1 in other regions. To boost the simulation efficiency, an auxiliary sphere was created around the scoring volume for every case, and electron transport was discontinued outside it. The radius of the sphere was chosen depending on electron energy so that electrons with the maximum energy produced outside the sphere would not be able to reach the scoring volume. The radiation yield of the terminated electrons was kept below 0.2%. The statistical expanded uncertainties ($k = 2$) were less than or equal to 0.5%.

4. Results and discussion

4.1. Density-effect correction term

Figure 1(a) shows the $\delta$-term calculated for graphite ($\delta_\text{g}$), pseudo-diamond media ($\delta_\text{pd1}$ and $\delta_\text{pd2}$), and diamond ($\delta_\text{d}$) with Sternheimer’s approach (Sternheimer et al. 1982, 1983, 1984, ICRU 1984). This approximate method avoids the need of knowing the full OELF (which is lacking for many materials) and it has been exploited in ICRU Reports 37 and 49 (ICRU 1984, 1993) as well as in the new ICRU Report 90 (ICRU 2016). A table with the $\delta$-term for diamond is provided in the supplementary data (available online at stacks.iop.org/PMB/66/125004/mmedia). Figure 1(a) highlights the marked difference between graphite and diamond, which is largest at the lowest energies where the $\delta$-term of diamond goes to zero. As pointed out by Fernández-Varea et al. (2021), diamond is an insulator and its $\delta$-term approaches zero at low energies faster compared to pseudo-diamond which is treated as graphite, i.e. having $n_{\text{ee}} = 1$. The departure at low energies is brought about by the large band gap (5.5 eV) in the DRF of diamond whereas it is zero for crystalline graphite.
Figure 1. (a) Density-effect correction term for graphite ($\delta_g$), pseudo-diamond ($\delta_{pd1}$ and $\delta_{pd2}$) and diamond ($\delta_d$) as a function of electron kinetic energy. (b) Percentage difference in the density-effect correction term of pseudo-diamond and diamond ($\delta_{x}$, $x = pd1$, pd2, d) with respect to graphite ($\delta_g$); the inset depicts the same information but for $E < 1$ MeV.

Figure 1(b) displays the percentage difference in the $\delta$-term between the pseudo-diamond media ($\delta_{pd1}$ and $\delta_{pd2}$) and diamond ($\delta_d$) with respect to graphite ($\delta_g$). The deviation is largest when the crystalline graphite mass density is replaced with that of diamond leaving the other parameters unchanged ($\delta_{pd1}$). When both $\rho$ and $I$ are modified, the discrepancy is smaller as a consequence of an interplay effect that these parameters have on the $\delta$-term. The variation in $n_{ec}$ for the same ($\rho$, I) set, i.e. $\delta_{pd2}$ and $\delta_d$, affects only the low and medium energies. The latter discrepancy occurs due to the rearrangement of the distribution of oscillators. Although the differences between the $\delta$-terms are substantial below 1 MeV, the contribution of $\delta$ to the mass electronic stopping power of light charged particles in diamond is less than 5% in this energy interval.

The deviation reduces to a few percent in the high-energy region and is the smallest for pseudo-diamond pd2 and diamond. This behavior can be understood from equations (2) and (3) where the ratio $\sqrt{\rho / I}$ is the relevant quantity. Then, increasing simultaneously $\rho$ and $I$ yields a smaller discrepancy as compared to a change in $\rho$ alone ($\delta_{pd1}$).

4.2. Differences between the $\delta$-term calculation methods and implementations in MC codes

For graphite and diamond, the $\delta$-terms calculated using the methods by Fano (1963) and Sternheimer et al. (1984) differ by up to a factor of six for $E < 0.2$ MeV, where $\delta$ is much smaller than $\xi$ and $F^\pm$, see equation (4). The two methods agree completely at energies above 1 MeV. ICRU Reports 37 and 90 resort to Sternheimer’s method for the key reference data calculations, whereas Fano’s method is implemented in the PENELOPE MC code to evaluate the $\delta$-term. Another widely-used MC radiation transport code is EGSnrc (Kawrakow et al. 2013),
whose pre-processor PEGS4 (Nelson et al 1985) generates the material data. The default approach there employs parameterized $\delta$-term functions, equation (5), tabulated for a pre-defined material list which, unfortunately, does not include diamond. The user is also allowed to enter the parameter values manually. If a user-defined input data is not entered, the fitting parameters are determined from a generic formula based on the $I$-value and the physical state (solid/liquid or gaseous) of the medium (Sternheimer and Peierls 1971).

The adjustable parameters obtained for the fitting function $\delta(X)$, equation (5), of diamond ($\delta_d$) were $\{X_0 = -0.0701, X_1 = 2.4236, a = 0.1426, m = 3.3284\}$ (recall that $\delta(X) = 0$ if $X < X_0$ because $n_{ce} = 0$). The maximum difference between the numerically evaluated and parameterized $\delta$-term functions, $|\Delta \delta|_{\text{max}}$, was 0.039 in the intermediate region $X_0 < X < X_1$ and 0.004 in the asymptotic region $X_1 < X$. In the former case, it would translate to a relative error of 1% in the $\delta$-term, and in the latter case the relative error is negligible. That is, the fit reproduces the behavior of the $\delta$-term accurately where its contribution to $S_{el}/\rho$ is highest. For comparison, Sternheimer et al (1982, 1983, 1984) aimed at $|\Delta \delta|_{\text{max}} < 0.1$ and an average value $\langle|\Delta \delta|\rangle$ of 0.03–0.05 in the intermediate region, and $|\Delta \delta|_{\text{max}} < 0.015$ in the asymptotic region. Our results are well within these limits and, moreover, $\langle|\Delta \delta|\rangle = 0.013$ in the intermediate region. The determined parameters may be used to generate the $\delta$-term of diamond for the EGSnrc simulations or to evaluate the water-to-diamond stopping power ratio $[S_{el}/\rho]_w$ employed in dosimetry. Note that, owing to the nature of the $\delta(X)$ function, the adjustable parameters are highly interdependent and other combinations of their values would lead to comparable density-effect corrections.

The fit function $\delta(X)$ for diamond with generic parameters $\{X_0 = 0.2, X_1 = 2.0, a = 0.3, m = 3.0\}$ from Sternheimer and Peierls (1971) has $|\Delta \delta|_{\text{max}} = 0.280$ in the $X_0 < X < X_1$ interval and 0.007 when $X_1 < X$. The difference is largest up to 5 MeV, where the contribution of the $\delta$-term to $S_{el}/\rho$ is about 12%, and it rapidly decreases to 1% above 15 MeV.

Although the discrepancies between the discussed fit functions and the numerical density-effect corrections are largest at low electron kinetic energies where the $\delta$-term is negligible compared to the other terms in equation (1), $\delta(X)$ with the parameters presented here gives a more accurate and consistent description of energy deposition in diamond.

The ESTAR program (Berger et al 2017) can also be utilized to compute the $\delta$-term for diamond. The calculation is carried out by means of Sternheimer’s method. The user can select the values of $\rho$ and $I$, but not the value of $n_{ce}$. In fact, the program imposes that $n_{ce} = 2$ whenever $Z = 6$, as in the case of graphite (ICRU 1984). Consequently, the $\delta$-term generated by ESTAR yields unphysical results for diamond with $(\rho = 3.515$ g cm$^{-3}$, $I = 88.5$ eV) and crystalline graphite with $(\rho = 2.265$ g cm$^{-3}$, $I = 81.0$ eV).

4.3. Mass electronic stopping power

Based on the findings by Fernández-Varea et al (2021) and those of section 4.1, figure 2 shows the percentage change in $S_{el}/\rho$ for electrons for pseudo-diamonds and diamond with respect to graphite. The mass electronic stopping power decreases for all $(\rho, I, n_{ce})$ sets, and the magnitude depends on the term dominating in equation (4). Below 0.1 MeV, the contribution of the $\delta$-term to $S_{el}/\rho$ is smaller than 0.3%, and $\xi(\beta; I)$ dominates in equation (4) at these energies. This is also apparent in figure 2 comparing graphite with pseudo-diamond pd1 (red dotted curve) where a variation in $\rho$ exclusively does not entail any difference in $S_{el}/\rho$ below around 0.1 MeV. When the $I$-value is modified, the impact is non-negligible at these electron kinetic energies, and $S_{el}/\rho$ is reduced by up to 3%.

Above 0.1 MeV the percentage change in $S_{el}/\rho$ is affected by $\xi(\beta; I)$ and $\delta(\beta; \rho, I, n_{ce})$. The influence of the mass density alone in the $\delta$-term, and subsequently $S_{el}/\rho$, is visible comparing graphite and pseudo-diamond pd1 (red dotted curve). When both $\rho$ and $I$ are set to those of diamond, the relative variation in $S_{el}/\rho$ further increases to 1.5% (green dotted–dashed curve). The role of the number of conduction electrons per atom $n_{ce}$ amounts to a 0.5% variation in $S_{el}/\rho$ which is seen comparing diamond (blue dashed curve) and pseudo-diamond (green dotted–dashed curve). Note that when $n_{ce} = 0$, the $\delta$-term calculated with Sternheimer’s method drops rapidly to zero at 0.17 MeV, thus a sharp structure appears in the curve pertaining to diamond. Above ~ 30 MeV, the percentage reduction in $S_{el}/\rho$ is around 1.7% for all media when compared with graphite. This trend arises from the asymptotic behavior of the $\delta$-term, see equations (2) and (3). In this energy region, the only non-trivial medium-dependent quantity in $S_{el}/\rho$ is the plasma energy $\Omega_p$, which just depends on $\rho$ and is the same for the compared sets of pseudo-diamond and diamond media.

Although MC simulation has largely replaced cavity theories due to its higher accuracy, the mass electronic stopping power is still convenient to evaluate the absorbed dose energy dependence of the medium as a function of electron kinetic energy, or to determine the absorbed dose to water. In these cases, attention must be paid on how $S_{el}/\rho$ is calculated. Figure 3 displays water-to-medium ratios of mass electronic stopping powers $[S_{el}(E)/\rho]_w$ for graphite and diamond calculated with the relativistic Bethe formula, equation (1), for electrons. As explained above, the two ratios differ and there is no reason to use graphite properties for the calculation of
for diamond (see Mobit et al 1997, Andreo and Bennmakhlof 2017). On the contrary, such an approach would yield an incorrect determination of the absorbed dose to water when the Bragg–Gray, Spencer–Attix or intermediate-cavity theories are employed. Furthermore, the use of different $(\rho, I, n_{ee})$ sets for diamond in MC simulations and $S_{el}/\rho$ calculations does not give a coherent picture of electron energy deposition in diamond.

Figure 2. Percentage change in the mass electronic stopping power of pseudo-diamonds (3.515 g cm$^{-3}, 81.0$ eV, 1) and (3.515 g cm$^{-3}, 88.5$ eV, 1), and diamond (3.515 g cm$^{-3}, 88.5$ eV, 0) with respect to graphite (2.265 g cm$^{-3}, 81.0$ eV, 1) as a function of electron kinetic energy. The curves were calculated with the relativistic Bethe formula for electrons.

Figure 3. Water-to-medium ratios of mass electronic stopping powers $S_{el}(E)/\rho_{\text{med}}$ for graphite (2.265 g cm$^{-3}, 81.0$ eV, 1) and diamond (3.515 g cm$^{-3}, 88.5$ eV, 0) as a function of electron kinetic energy. The solid curves were calculated with the relativistic Bethe formula for electrons and the dashed curve was obtained from the material.exe pre-processor of PENELOPE.
not hinge on the stopping powers, hence they do not affect the quantities scored in MC simulations (only the $\delta$-term is needed from equation (1)). However, the comparison underlines the importance of knowing not only the medium’s properties but also how $S_{\text{el}}/\rho$ is calculated. A table with the mass electronic stopping powers of electrons and positrons in diamond can be downloaded from the supplementary data.

### 4.4. Energy deposition in diamond

The MC-calculated percentage change in the energy imparted in diamond cavities of various thicknesses when the $I$-value was increased from 81.0 to 88.5 eV is summarized in table 2 for the analyzed beam qualities.

#### 4.4.1. Kilovoltage photon beams

When the diamond cavity thickness was 1 μm, the energy imparted was approximately 1% lower for the diamond with $I = 88.5$ eV for all kV beams. The energy imparted in the 3 μm thick cavity decreased by 0.3%–0.5% for the 25 and 50 kV beams, and by 0.7% for the 100 and 180 kV beams. When the cavity thickness increased to 10 and 30 μm, there was no significant modification in energy imparted. This stems from the fact that charged-particle equilibrium exists for such cavities and secondary electrons were stopped within the cavity. The average fluence-weighted electron energy was 10, 16, 24 and 29 keV for the 25, 50, 100 and 180 kV beams, respectively, when scored in a 1 μm thick diamond cavity. This would correspond to continuous-slowing-down approximation (CSDA) electron ranges from 0.8 to 5 μm in diamond. Then, the 10 and 30 μm thick cavities can be approximated as large and, contrary to the Bragg–Gray and intermediate-cavity theories (1 and 3 μm thick cavities), the mass electronic stopping power is no longer adequate to describe energy deposition in the medium.

#### 4.4.2. BT sources

Similarly to kV x-ray beams, the variation in energy imparted depends on the cavity dimensions when the $I$-value increases. For the $^{125}$I radioactive source, the energy imparted in a 1 μm thick cavity was 1.3% lower for $I = 88.5$ eV, and 0.6% lower for a 3 μm thick cavity. No significant change in energy imparted with respect to the $I$-value occurred for 10 and 30 μm thick cavities. The average fluence-weighted electron energy in the diamond cavity at 10 mm distance from the $^{125}$I source is approximately 20 keV. This corresponds to a CSDA range of approximately 3 μm in diamond. Electrons are therefore stopped in thicker cavities so that the $I$-value does not play a role. A 3 μm thick cavity, on the other hand, acts as an intermediate one. Similar results were obtained for the $^{169}$Yb and $^{192}$Ir sources. For the $^{106}$Ru source, which is a β-emitter, the energy imparted decreased by approximately 1% when the $I$-value was increased to 88.5 eV for all cavity thicknesses.

#### 4.4.3. $^{60}$Co beam

The energy imparted in a diamond cavity with $I = 88.5$ eV was close-to 1% lower compared to a pseudo-diamond medium with $I = 81.0$ eV. As expected, it did not depend on the cavity thickness because the fraction of photon interactions inside the cavity itself is small and the CSDA range of the electrons is longer than the cavity thickness. The difference was of the same magnitude as for a 6 MV photon beam due to similar electron fluence spectra. Thus, if a diamond detector is calibrated in a $^{60}$Co beam and operated for measurements in a 6 MV large photon field, the choice of $I$-value would not affect the beam quality correction factors determined from either MC calculations or mass electronic stopping power ratios.

#### 4.4.4. 6 MV photon beam

For a large 10 × 10 cm$^2$ field, the energy imparted in a 30 μm thick diamond cavity decreased by 0.9% for $I = 88.5$ eV. The corresponding decrease was 0.7% for a 0.5 × 0.5 cm$^2$ small field. Since the variation is similar, the output correction factors employed nowadays to account for diamond detector response in small-field high-energy photon dosimetry are expected to be the same if the new $I$-value for diamond were adopted. However, a

### Table 2. Percentage change in energy imparted in cylindrical diamond cavities with $r = 1.1$ mm and the indicated thicknesses when the $I$-value was increased from 81.0 to 88.5 eV ($\rho = 3.515$ g cm$^{-3}$, $n_{\text{el}} = 0$). The statistical expanded uncertainties were less than or equal to 0.5% ($k = 2$).

| Beam quality | $1$ μm | $3$ μm | $10$ μm | $30$ μm |
|--------------|--------|--------|--------|--------|
| 25–180 kV $^{125}$I, $^{192}$Ir | $-0.7\% \text{ to } -1.2\%$ | $-0.3\% \text{ to } -0.7\%$ | negligible | negligible |
| $^{106}$Ru | $-1.1\% \text{ to } -1.7\%$ | $-0.6\% \text{ to } -1.1\%$ | negligible | negligible |
| $^{60}$Co | $-1.2\%$ | $-1.0\%$ | $-0.7\%$ | $-0.8\%$ |
| 6 MV, 0.5 × 0.5 cm$^2$ | $-1.0\%$ | | | |
| 6 MV, 10 × 10 cm$^2$ | | | | |
| 100 MeV, 2 × 2 cm$^2$ | | | | Negligible |
complete detector should be simulated to obtain clinically relevant values. The change in energy imparted in the two cases was statistically the same because of the similarity of electron energy fluence distributions in the diamond cavity, see figure 4.

Since the photon scatter fraction is smaller in a small field, the corresponding electron fluence spectrum is shifted towards higher energies with respect to a large-field case. However, the most probable energy of the differential ema distribution, which describes energy transfer from primary charged particles (i.e. secondary electrons) to secondary charged particles, is 1 MeV (Andreo and Benmakhlouf 2017). The two electron fluence spectra coincide at this energy yielding a similar change in energy imparted for a given I-value independently of the field size. Nevertheless, the use of the I-value corresponding to diamond rather than graphite is important when the energy imparted in the diamond cavity is not evaluated as a ratio, or when the electron fluence differential in energy differ between the compared cases.

It should be noted that the output correction factors tabulated in the TRS-483 Report (Palmans et al 2017) are for a natural diamond detector, which is obsolete, and for a 1 μm thick commercially available synthetic diamond dosimeter. A 30 μm thick cavity was selected to increase simulation efficiency in the present study, but the results would be valid for a thinner cavity as well because volume averaging in the beam incidence direction, i.e. perpendicular to the detector active volume surface, is negligible.

4.4.5. 100 MeV electron beam
The energy imparted in a 30 μm thick diamond cavity did not depend on the I-value. The δ-term values do differ between pseudo-diamond and diamond (see figure 1), but at these high energies the δ-term has reached its asymptotic behavior (see equation (2) and figure 4) where $S_{el}/\rho$ becomes independent of the I-value. Although the radiation yield for 100 MeV electrons in water is approximately 32%, the fraction of photons interacting in the diamond cavity is sufficiently low, and the negligible variation in energy imparted can be explained by this property of $S_{el}/\rho$.

5. Conclusions
Graphite and diamond do not share the same physical properties, in particular the mass density and the DRF, and different sets of (\(\rho, I, n_e\)) values affect the energy deposition in the medium depending on beam quality and cavity thickness via the I-value directly and also via the density-effect term. Apart from the well-known change in the mass density, the I-value and the number of conduction electrons should also be adjusted for the most faithful description of diamond. Furthermore, it is important to use the same set of diamond material parameters in the MC simulations and for the calculation of the mass electronic stopping power. Otherwise, the
interpretation of MC-calculated energy imparted in diamond and the comparison with $S_d/\rho$ calculated for crystalline graphite will be inconsistent (e.g. for the evaluation of water-equivalence of diamond). Different combinations of currently used ($\rho, I, n_{ce})$ values influence the density-effect term the most at low electron kinetic energies where the density-effect contribution to electron energy loss is low, but the discrepancies remain appreciable at electron kinetic energies above 10 MeV where diamond detectors have potential for use. Therefore, the study presents a parameterized density-effect correction function that can be employed in MC codes like EGSnrc instead of the one delivered by the ESTAR program. ESTAR assumes that the number of conduction electrons is 2 for all carbon-based materials, which is incorrect for diamond as well as crystalline graphite.

The analyzed scenarios with several beam qualities show that the percentage change in $S_d/\rho$ explains the variation in energy imparted in diamond depending on the cavity size and electron CSDA range. If the electron range is short compared to the dimensions of the diamond cavity, electrons are assumed to be absorbed on the spot, and $[S_d/\rho]_e$ ceases to be useful. For instance, in the ideal large-cavity limit, energy deposition in the medium is described by the mass energy-absorption coefficients and the change in $\rho$ and $I$ of the medium does not play a role as long as secondary electrons are stopped within the cavity and charged-particle equilibrium between the cavity and the surrounding medium exists. If the electron range exceeds the dimensions of the cavity (Bragg–Gray cavity theory), or in the intermediate case where electrons coming from outside and those produced inside (Burlin cavity theory) deposit energy, $[S_d/\rho]_e$ can be used but attention has to be paid to which ($\rho, I, n_{ce})$ values are selected and how $S_d/\rho$ is calculated.

In practice, diamond detectors are mainly operated in high-energy small-field photon beams, and the current output correction factors calculated using pseudo-diamond material properties are not expected to vary significantly if diamond were modeled with a higher $I$-value and zero conduction electrons. While the differences in energy imparted are small in the investigated beam qualities, modeling diamond to the best of knowledge is important for the sake of consistency and to better understand energy deposition in a wide interval of electron kinetic energies.

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