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Microdosimetric properties of ionizing electrons in water: a test of the PENELOPE code system

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Abstract

The ability to simulate the tortuous path of very low-energy electrons in condensed matter is important for a variety of applications in radiobiology. Event-by-event Monte Carlo codes such as OREC, MOCA and PITS represent the preferred method of computing distributions of microdosimetric quantities. However, event-by-event Monte Carlo is computationally expensive, and the cross sections needed to transport simulations to this level of detail are usually only available for water. In the recently developed PENELOPE code system, 'hard' electron and positron interactions are simulated in a detailed way while 'soft' interactions are treated using multiple scattering theory. Using this mixed simulation algorithm, electrons and positrons can be transported down to energies as low as 100 eV. To our knowledge, PENELOPE is the first widely available, general purpose Monte Carlo code system capable of transporting electrons and positrons in arbitrary media down to such low energies. The ability to transport electrons and positrons to such low energies opens up the possibility of using a general purpose Monte Carlo code system for microdosimetry.

This paper presents the results of a code intercomparison study designed to test the applicability of the PENELOPE code system for microdosimetry applications. For sites comparable in size to a mammalian cell or cell nucleus, single-event distributions, site-hit probabilities and the frequency-mean specific energy per event are in reasonable agreement with those predicted using event-by-event Monte Carlo. Site-hit probabilities and the mean specific energy per event can be estimated to within about 1–10% of those predicted using event-by-event Monte Carlo. However, for some combinations of site size and source-target geometry, site-hit probabilities and the mean specific energy per event may only agree to within 25–60%. The most problematic source-target geometry is one in which the emitted electrons are very close to the tally site (e.g., a point source on the surface of a cell). Although event-by-event Monte Carlo will continue to be the method of choice for microdosimetry, PENELOPE

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is a useful, computationally efficient tool for some classes of microdosimetry problem. PENELOPE may prove particularly useful for applications that involve radiation transport through materials other than water or for applications that are too computationally intensive for event-by-event Monte Carlo, such as *in vivo* microdosimetry of spatially complex distributions of radioisotopes inside the human body.

1. Introduction

PENELOPE, an acronym for PENetration and Energy LOss of Positrons and Electrons, is a general purpose Monte Carlo code to simulate the behaviour of ionizing electrons and photons in arbitrary materials composed of elements with atomic numbers from 1 to 92 (Baró *et al* 1995, Salvat *et al* 1996, Sempau *et al* 1997). PENELOPE implements a so-called mixed simulation algorithm (Baró *et al* 1995, Salvat *et al* 1996) in which 'hard' electron and positron interactions are simulated in a detailed way while 'soft' interactions are treated using multiple scattering theory (Berger and Wang 1988). For higher energy electrons and photons, benchmarking activities by Sempau *et al* (1997) demonstrate that PENELOPE compares favourably with the EGS4 (Nelson *et al* 1985), ETRAN (Seltzer 1986) and the ITS3 (Halbleib *et al* 1992) Monte Carlo code systems. However, PENELOPE is capable of transporting electrons and positrons down to energies as low as 100 eV whereas EGS4, ETRAN, ITS3 and MCNP (Briesmeister 1997) are generally limited to electron and positron kinetic energies above 1–20 keV. To our knowledge, PENELOPE is the first widely available, general purpose Monte Carlo code system capable of transporting electrons and positrons in arbitrary media down to such low energies[3].

The ability to simulate the tortuous path of very low-energy electrons in condensed matter is important for a variety of applications in radiation chemistry and radiobiology (see Nikjoo *et al* 1994, 1998). In cellular and sub-cellular-sized regions of matter, the stochastic nature of the energy transfer events produced by ionizing radiation are often characterized in terms of microdosimetric quantities such as the frequency-mean specific energy per (radiation) event $\bar{z}_F$ (ICRU 1983, Rossi and Zaider 1996). For the most part, microdosimetry has been confined to the realm of event-by-event Monte Carlo codes such as MOCA (Wilson and Paretzke 1981, Paretzke 1987, Wilson *et al* 1988), OREC (Turner *et al* 1983, 1988) and PITS (Wilson *et al* 1994, Wilson and Nikjoo 1999). However, event-by-event Monte Carlo is very computation intensive. Also, the cross sections needed to perform simulations at this level of detail are usually only available for water. On the other hand, PENELOPE offers some of the simulation efficiency of condensed-history Monte Carlo along with an event-by-event physics model for hard collisions that may be comparable to purely analog Monte Carlo code systems. This paper presents the results of a Monte Carlo code intercomparison study designed to test the applicability of the PENELOPE code system for microdosimetry applications. The scope of the study includes the comparison of single-event distributions, the frequency-mean specific energy per event and hit probabilities for sites from 10 nm to 10 μm in diameter and primary (source) electron energies in the range 1–100 keV.

[3] Although PENELOPE can transport electrons and positrons down to energies as low as 100 eV, the cross sections and physics below about 500 eV are more uncertain than those at higher energies because relevant refinements in the differential cross sections are neglected. Ongoing work by the PENELOPE team could have a positive impact on low-energy, charged-particle transport in the near future. (Personal communications from F Salvat and J Sempau, September 17, 2001.)

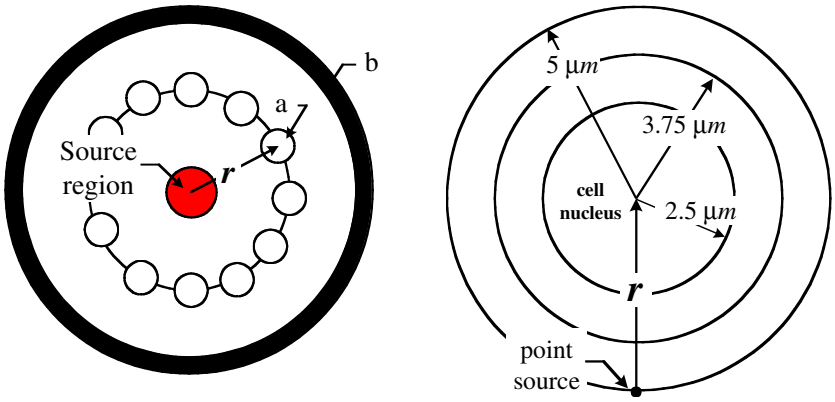

Figure 1. Schematic illustrating the annular tally regions and spherical sites used to tabulate dosimetric and microdosimetric quantities. *Left panel:* Source and tally sites used in the PNNL-developed PENELOPE application. (a) Microdosimetric quantities are tabulated in ten spherical sites located distance r from the centre of the source region. (b) The absorbed dose and other dosimetric quantities are also tallied in annular regions located at various distances from the source. *Right panel:* The source and tally regions used to model a radiolabelled monoclonal antibody on the surface of a mammalian cell (Bolch and Kim 1994). For the PENELOPE simulations shown in figure 3 and 4, the source region is 0 μm in diameter (point source) and r equals 5 μm (surface of cell) or 5.1 μm (0.1 μm away from cell surface). Separate PENELOPE simulations are used for the 2.5, 3.75 and 5 μm tally-site sizes.

2. Methods

The physics model and cross sections implemented in the PENELOPE code system have been described in detail by others (Baró *et al* 1995, Salvat *et al* 1996, Sempau *et al* 1997). The crucial component of the PENELOPE code system is a set of Fortran 77 routines that can be used to perform 'near analog' electron–photon simulations with little prior knowledge of electron and photon transport physics. The intricacies of sampling from the relevant cross section tables and temporary storage of secondary particles created during the transport of a primary (source) particle are automatically and, for the most part, transparently handled within the PENELOPE transport kernel. A complete PENELOPE application consists of the PENELOPE transport kernel and a set of user-supplied routines to record ('tally') the location of energy transfer events, control the overall evolution of particle tracks (e.g. initial creation of source particles) and determine the geometry of the materials used in the simulation.

The Pacific Northwest National Laboratory (PNNL) has developed a PENELOPE application[4] to calculate dosimetric and microdosimetric quantities for a spherical source region composed of one material and surrounded by an infinite medium of the same or a different material. The initial location of the primary (emitted) electrons is determined by random sampling from within the source region, and dosimetric and microdosimetric quantities are tallied in the regions indicated in figure 1. All the results reported in this paper are for a point isotropic electron source in an infinite medium of water at a density of 1.0 g cm^{-3}. Because the diameter of the source region is zero, random sampling of the initial source particle location is turned off. The level of detail included in the simulation of electron transport processes can be controlled in PENELOPE using several parameters (see Salvat *et al* 1996). Except where explicitly noted otherwise, PENELOPE simulation parameters were selected to

[4] All the results reported in this paper are based on the standard (default) PENELOPE physics model and cross sections (Salvat *et al* 1996).

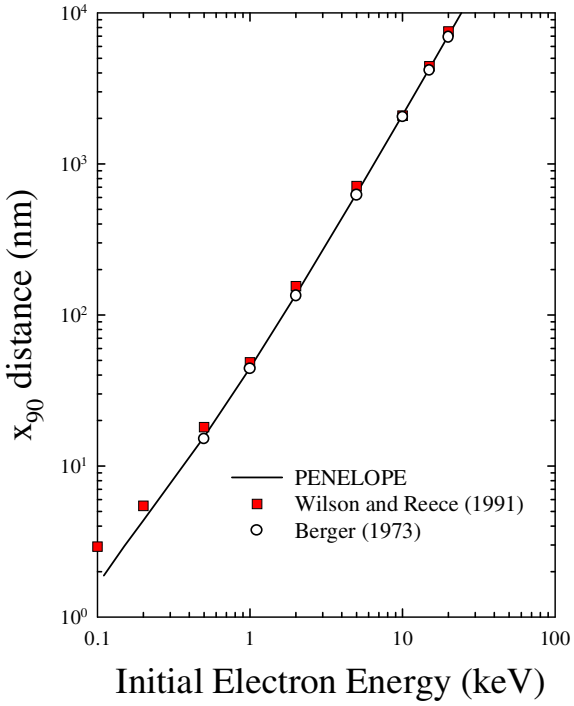

Figure 2. Distance through water in which 90% of the primary electron's kinetic energy is absorbed. The Berger results (1973) are based on multiple scattering theory. The Wilson and Reece results (1991) are based on simulations performed using the MOCA event-by-event Monte Carlo code system (Wilson and Paretzke 1981, Paretzke 1987, Wilson *et al* 1988).

give a detailed treatment of electron elastic and inelastic collisions ($C_1 = 0.00$, $C_2 = 0.01$, $W_{CC} = 100$ eV, $W_{CR} = 100$ eV). Electrons and positrons with kinetic energies below 100 eV and photons with energies less than 1 keV are assumed to deposit all their energy locally. The maximum allowed mean free path length between hard events, $HFP_{\max}$, was set to a value of 10^{308} cm; thus the distance between hard collisions is always determined by the C_1, C_2, W_{CC} and W_{CR} simulation control parameters.

3. Results and discussion

Figure 2 shows a graph of the distance, x_{90}, away from the source region in which ninety percent of the primary electron's energy is absorbed. The x_{90} distances for water computed using PENELOPE are within 3% of those reported by Berger (1973). For electrons with energies greater than 10 keV, the PENELOPE results are 1–6% lower than those obtained with the MOCA event-by-event Monte Carlo radiation transport code system (Wilson and Reece 1991). For lower energy electrons, the difference between the PENELOPE and MOCA x_{90} distances tends to increase as the initial kinetic energy of the electron decreases. For 1 keV electrons, the x_{90} distances differ by about 9%. For 110 eV electrons (close to the minimum energy electron PENELOPE can transport), the MOCA and PENELOPE x_{90} distances differ by 70% (3.2 nm MOCA and 1.9 nm PENELOPE). The agreement in the x_{90} distances is reasonable for electrons with energies greater than 1–10 keV. The increasingly large difference between

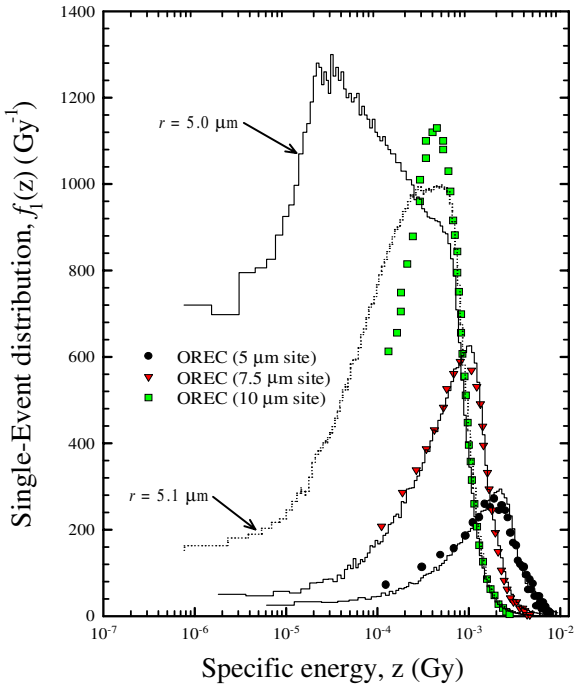

Figure 3. Single-event distribution for a point-isotropic source of 100 keV electrons located on the surface of a 10 μm site ($r = 5$ μm; refer to figure 1, right panel). The centres of 5 and 7.5 μm sites are located 5 μm away from the source as shown in figure 1, right panel. *Lines:* 500 CPU minute ($\sim 4 \times 10^5$ tracks) PENELOPE simulation on a 500 MHz Intel® Pentium® III Xeon computer. Simulation parameters are $C_1 = 0.00$, $C_2 = 0.01$, $W_{CC} = 100$ eV and $W_{CR} = 100$ eV. *Dotted line:* Same as solid lines except the 10 μm tally site is located slightly farther away from the source ($r = 5.1$ μm instead of 5.0 μm). *Filled symbols:* 10 000 simulated OREC tracks (Bolch and Kim 1994).

the x_{90} distances for lower energy electrons indicates that the small-scale pattern of energy deposits predicted by MOCA and PENELOPE are quite different.

In radiobiology, statistical fluctuations in the radiation field (dose) experienced by a cell or critical regions within a cell, are often of interest. The mean chord length (ICRU 1983) associated with the volumes of some critical cellular targets (e.g. the cell nucleus) is of the order of 1–10 μm. For a sphere of diameter d, the mean chord length is $2d/3$ (ICRU 1983). Figure 3 shows the single-event distribution (ICRU 1983), $f_1(z)$, for sites 5, 7.5, and 10 μm in diameter located 5 μm away from a point-isotropic source of 100 keV electrons (refer to figure 1, right panel). The 10 μm site represents the outer boundary of the cell and the 5 μm diameter site represents the nucleus. This source-target geometry is intended to represent a radiolabelled monoclonal antibody on the surface of a mammalian cell (Bolch and Kim 1994).

As the diameter of the site increases, the width of the peak in the single-event distributions decreases and shifts towards lower specific-energy events. For the 5 and 7.5 μm sites, the PENELOPE and OREC single-event distributions are in good agreement. For the 10 μm site, the shapes of the single-event distributions are significantly different for specific energies less than about 0.6 mGy (the energy imparted is 1.96 keV). However, the first moment of the single-event distributions (i.e. the frequency-mean specific energy per event $\bar{z}_F$) is about the same. Bolch and Kim (1994) report a $\bar{z}_F$ of 3.2 and 0.68 mGy for the 5 and 10 μm sites,

respectively, compared to 3.24 and 0.615 mGy for PENELOPE. For 1 MeV electrons, $\bar{z}_F$ is 1.2 mGy for the 5 μm site and 0.26 mGy for the 10 μm site (Bolch and Kim 1994). The PENELOPE-calculated $\bar{z}_F$'s are within 6% of these values.

The low specific-energy events shown in figure 3 correspond to electrons that pass through a small portion of the tally volume and undergo relatively few elastic or inelastic collisions. Radiative transfer events (i.e. emission of bremsstrahlung radiation) are very rare for 100 keV electrons in water. If the 10 μm tally volume is moved slightly farther away from the source, the number of short track segments passing through the site decreases and the shape of the single-event distribution becomes more consistent with the one predicted by OREC (figure 3, dotted lines). Because the PENELOPE physics model is optimized for macro-scale dosimetry (sites with characteristic dimensions hundreds or more micrometres in length), the discrepancies seen in the PENELOPE single-event distribution for a point source located at the surface of 10 μm site are not entirely unexpected. Although PENELOPE uses relatively detailed physics models to simulate hard events (large angle and large energy-transfer events), soft interactions are incorporated into the model using multiple scattering theory and a refined continuous slowing down approximation (CSDA) model that accounts for energy straggling (Salvat *et al* 1996).

The reliability of a PENELOPE simulation ultimately rests on the accuracy of the differential cross section (DCS) model and the condition that for each microdosimetric event a statistically significant number of soft and hard collisions occur in the tally volume. For the sake of computational efficiency, PENELOPE uses a DCS model that has a physically reasonable shape, can be rapidly sampled by Monte Carlo methods and gives the correct (asymptotic) behaviour for track segments formed by at least 15 or 20 hard collisions (Salvat *et al* 1996). As the number of hard collisions in the tally volume decreases, the approximate DCSs used in PENELOPE can give rise to spurious, low specific-energy events, as can be easily seen in figure 3 for the 10 μm site.

The number of hard collisions occurring in a track segment can be controlled to some extent by varying the W_{CC}, W_{CR}, C_1, and C_1 simulation parameters. W_{CC} determines the cutoff energy for hard inelastic collisions and W_{CR} specifies the cutoff energy for hard radiative transfer events. The C_1 and C_2 simulation parameters determine the distance between hard elastic collisions. A value of $C_1 = 0$ forces PENELOPE to use the detailed (analog) model for all elastic scattering interactions. The PENELOPE manual (Salvat *et al* 1996) suggests that the following simulation parameters are 'safe' for most applications: $C_1 = 0.001$, $C_2 = 0.01$, $W_{CC} = W_{CR} = 0.01E_0$. Here, E_0 is the initial kinetic energy of the primary electron or positron. The maximum allowed value for C_1 is 0.2, and the C_2 parameter has a maximum allowed value of 0.1 (Salvat *et al* 1996). For cutoff energies W_{CC} and W_{CR} greater than or equal to E_0, the CSDA approximation is always used to transport electrons and positrons.

Because microdosimetric quantities are sensitive to the small-scale spatial distribution of the energy deposits created along a track, some combinations of simulation parameters in the allowed range are inappropriate for microdosimetry applications. For example, figure 4 shows the single-event distributions calculated with the simulation parameters $C_1 = 0.2$, $C_2 = 0.1$, $W_{CC} = 100\,\text{eV}$ and $W_{CR} = 100\,\text{eV}$. The event distributions for the 5 and 7.5 μm diameter sites deviate significantly from the OREC distributions, although the shape of the distribution from the 10 μm site is similar. Based on our experience, the constraints $0 \leqslant C_1 \leqslant 0.02$, $0 < C_2 \leqslant 0.02$, $100\,\text{eV} \leqslant W_{CC} \leqslant 500\,\text{eV}$ and $100\,\text{eV} \leqslant W_{CR} \leqslant 500\,\text{eV}$ are recommended when using PENELOPE for microdosimetry applications. Values of W_{CC} and W_{CR} below 100 eV can be used but they produce non-physical oscillations in the low specific-energy region of the single-event distribution (not shown).

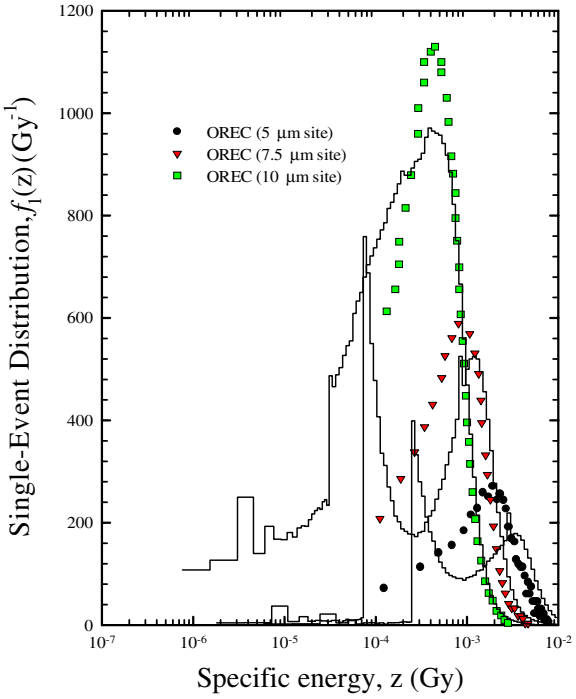

Figure 4. Single-event distribution for a point-isotropic source of 100 keV electrons located on the surface of a 10 μm site ($r = 5$ μm in figure 1, right panel). The centres of 5 and 7.5 μm sites are located 5 μm away from the source. *Solid lines:* 720 CPU minute ($\sim 8 \times 10^5$ tracks) PENELOPE simulation on a 500 MHz Intel® Pentium® III Xeon computer. Simulation parameters are $C_1 = 0.2$, $C_2 = 0.1$, $W_{CC} = 100$ eV and $W_{CR} = 100$ eV. These values were selected to illustrate the non-physical behaviour of PENELOPE for some combinations of simulation parameters (see the recommended range of simulation parameters and discussion in the main text). *Filled symbols:* 10 000 simulated OREC tracks (Bolch and Kim 1994).

The data shown in figures 3 and 4 suggest that the PENELOPE physics model, which is fast and accurate for macro-scale dosimetry, can also be applied—with care—to the estimation of microdosimetric quantities and event distributions in sites comparable in size to a mammalian cell or cell nucleus. To further probe the limitations and applicability the PENELOPE code system for microdosimetry purposes, we conducted an intercomparison of PITS (Wilson *et al* 1994, Wilson and Nikjoo 1999) and PENELOPE calculations of $\bar{z}_F$ and the site hit probability, p_{hit}, for a wider range site sizes and electron energies. A site 'hit' occurs when the primary source particle, or one of the spatially correlated particles produced by the primary, imparts at least some energy to the region of interest. Table 1 summarizes the results of the intercomparison. These data indicate that PENELOPE tends to overestimate $\bar{z}_F$ and underestimate p_{hit}. These trends in $\bar{z}_F$ and p_{hit} are mainly due to the approximate DCS models used to account for elastic and inelastic scattering processes in PENELOPE.

For primary electron energies greater than about 5 keV and site sizes in the range from 1 to 10 μm, PENELOPE overestimates $\bar{z}_F$ by factors ranging from a few percent up to a maximum of 25%. The differences in site hit probabilities for this range of energies and site sizes are generally of the order of 4 or 5% but may be as large as 25–60%. Low-energy electrons close to the tally site appear to be particularly problematic (e.g. the results for the 10 keV electron source on the surface of a 5 μm site). For 1–5 keV electrons and very small site sizes (i.e.

Table 1. Summary of frequency-mean specific energies and hit probabilities for a range of primary electron energies and site sites. Radial distance from the source is the same as r in figure 1 (left panel). Estimates of the hit probabilities and mean specific energy are generally accurate to one or two significant digits. For illustrative purposes only, all results are reported to three significant digits. The numbers in parentheses are the percent difference between the PITS and PENELOPE results.

Electron energy (keV)	Site diameter (μm)	Radial distance from source (μm)	Mean specific energy (Gy)		Probability site hit	
			PITS	PENELOPE	PITS	PENELOPE
1	0.01	0.05	35 200	55 900 (−58.8)	0.00148	0.000632 (57.3)
5	0.05	0.025	231	306 (−32.3)	0.415	0.360 (13.2)
5	0.05	0.05	281	372 (−32.5)	0.0642	0.0595 (7.4)
5	0.05	0.1	292	399 (−36.5)	0.0163	0.0154 (5.5)
5	0.05	0.15	316	425 (−34.6)	0.007 71	0.007 38 (4.2)
5	0.05	0.2	333	456 (−36.9)	0.004 73	0.004 49 (5.0)
5	0.05	0.25	358	489 (−36.6)	0.003 24	0.003 07 (5.1)
5	0.05	0.3	391	526 (−34.6)	0.002 45	0.002 24 (8.6)
10	5.0	5.0	0.006 66	0.006 50 (2.5)	0.002 70	0.001 02 (62.3)
10	10.0	5.0	0.001 94	0.002 09 (−7.8)	0.640	0.627 (2.1)
25	1.0	5.0	0.295	0.340 (−15.2)	0.003 54	0.003 50 (1.0)
25	1.0	6.0	0.323	0.381 (−17.9)	0.002 66	0.002 52 (5.2)
25	1.0	7.0	0.367	0.429 (−17.0)	0.001 90	0.001 81 (4.5)
25	1.0	8.0	0.393	0.480 (−22.3)	0.001 37	0.001 25 (8.7)
25	1.0	9.0	0.446	0.544 (−22.0)	0.000 975	0.000 797 (18.2)
25	1.0	10.0	0.484	0.604 (−24.9)	0.000 608	0.000 449 (26.2)
95	5.0	5.0	0.003 30	0.003 34 (−1.3)	0.0712	0.0688 (3.4)
95	10.0	5.0	0.000 609	0.000 637 (−4.7)	0.483	0.497 (−2.9)

10–50 nm), PENELOPE overestimates $\bar{z}_F$ by 30–60% and underestimates p_{hit} by factors of the order of 5–60%.

4. Conclusions

The results of the intercomparison suggest that PENELOPE can be used to estimate microdosimetric quantities to a reasonable level of accuracy for some site sizes and energies of interest in radiobiology. For site sizes comparable to a mammalian cell or cell nucleus (5–10 μm), single-event distributions are consistent with those predicted by the more detailed OREC Monte Carlo code system. For sites with diameter in the range 1–10 μm and electron energies in the range 5–100 keV, the frequency-mean specific energy per event can be estimated to within 1–25%. The accuracy of these estimates tends to decrease as the site size and electron energy decrease. Site hit probabilities can be estimated to within 5–60% for site diameters from 10 nm to 10 μm. The most problematic source-target geometry is one in which the emitted electrons are very close to the tally site (e.g. a point source on the surface of a cell).

A potential advantage of the PENELOPE code system over event-by-event Monte Carlo codes, such as OREC and PITS, is that it may be possible to estimate microdosimetric quantities in materials other than water. Also, the mixed simulation algorithm used in PENELOPE is quite likely more computationally efficient than those used in purely analogue Monte Carlo codes. The ability to transport electrons or photons through higher atomic number materials and then estimate microdosimetric quantities in cell-sized regions of tissue may be useful for the dosimetry of low-LET single-cell (microbeam) irradiators under development

at PNNL and elsewhere (Miller *et al* 2000, Wilson *et al* 2001). PENELOPE may also prove useful for the estimation of microdosimetric quantities for complex spatial distributions of radioisotopes inside the human body (e.g. radiolabelled monoclonal antibodies, brachytherapy and other nuclear medicine applications). The estimation of microdosimetric quantities for cell-sized sites is also potentially important for the assessment of health risks associated with the inhalation of metal tritides (Strom *et al* 2001) or other inhaled or ingested radioactive materials.

Recent studies indicate that the so-called bystander cells (cells that do not experience a radiation event) may still contribute to the observed radiation response of a collection of cells (Mothersill *et al* 2001, Mothersill and Seymour 2001, Lewis and Mayhugh 2001, Sawant *et al* 2001a, 2001b). Bystander effects may produce supralinear dose-response effects (Brenner *et al* 2001) in the low dose regime most relevant to health protection. The ability to partition a population of cells into those that are hit and those that are not is essential for the effort to develop mechanism-based models of collective (hit and non-hit) radiation response. Microdosimetry provides a convenient formalism to partition a collection of cells into those that are hit and those that are not. In a uniformly irradiated region of matter, the probability that a cell experiences one or more radiation events is (ICRU 1983) $1-\exp(-D/\bar{z}_F)$, where D is the absorbed dose and $\bar{z}_F$ is evaluated in a cell-sized tally volume.

Although event-by-event Monte Carlo will continue to be the method of choice for microdosimetry, PENELOPE is a useful, computationally efficient tool for some classes of microdosimetry problem. PENELOPE may prove particularly useful for applications that involve radiation transport through materials other than water or for applications that are too computationally intensive for event-by-event Monte Carlo, such as *in vivo* microdosimetry of spatially complex distributions of radioisotopes inside the human body.

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