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Molecular scale track structure simulations in liquid water using the Geant4-DNA Monte-Carlo processes

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ABSTRACT

This paper presents a study of energy deposits induced by ionising particles in liquid water at the molecular scale. Particles track structures were generated using the Geant4-DNA processes of the Geant4 Monte-Carlo toolkit. These processes cover electrons (0.025 eV–1 MeV), protons (1 keV–100 MeV), hydrogen atoms (1 keV–100 MeV) and alpha particles (10 keV–40 MeV) including their different charge states. Electron ranges and lineal energies for protons were calculated in nanometric and micrometric volumes.

1. Introduction

Track structure simulation codes have become increasingly important in recent years, especially those dedicated to nanometric- or molecular-scale applications. Such codes remain the only tools able to reproduce in detail energy deposits in small biological structures, such as the cell nucleus or even the DNA molecule and its surroundings. The availability of a suitable toolkit with which users can reproduce ionising tracks through a biological medium in a flexible manner that suits their particular applications is becoming particularly important for radiobiological studies. Several rather advanced dedicated codes are leaders in this field (Friedland et al., 2008; Nikjoo et al., 1998), taking into account the physico-chemical and chemical phases, including the simulation of free radicals and chemical species diffusion and interactions in irradiated liquid water. Some of these codes include the detailed spatial structure of the DNA molecule, so that the damaging effects on the DNA strands and structure modifications can be studied. A review can be found in a recent paper by Nikjoo et al. (2006). However, these "home built" codes are not easily accessible and are usually designed for specific applications, which means that new users may find it difficult to access and adapt to their special cases, especially if the authors are not at hand. The main advantage of extending the Geant4 toolkit (Agostinelli et al., 2003; Allison et al., 2006) to handle micro- and nano-dosimetric simulations is that Geant4 offers a common platform, freely available to all users. Developments are implemented on a common architecture basis, taking into consideration possible future extensions. Furthermore, new users in the field can benefit from detailed user guide reports, a user forum, tutorials and training sessions regularly organized by the Geant4 collaboration [http://www.geant4.org].

Using the processes provided by the low energy electromagnetic package available in Geant4, the energy cut-off can be as low as 250 eV for electromagnetic interactions. However, this value is still too high for nanometric applications. To lower this cut-off value, the low energy electromagnetic package has recently been extended, offering to the Geant4 users new processes down to the very low energy scale. The previously released Geant4-DNA version (Chauvie et al., 2006, 2007) has been refined and new cross-sections were added, extending the codes capabilities to higher energies for electrons and protons. All needed physical interactions were studied and the corresponding available cross-sections were reviewed in the literature; most were based on the plane-wave born approximation (PWBA) for inelastic interactions and on semi-empirical models for the low energies where the PWBA fails. The Geant4-DNA processes cover electrons (0.025 eV–1 MeV),

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protons (1 keV–100 MeV), hydrogen (1 keV–100 MeV) and alpha particles (10 keV–40 MeV) and their charge states. Finally, electron ranges were calculated for the available energy domain down till 0.025 eV. Protons lineal energies were compared with TRIOL code results in nanometric volumes and to experimental data for micrometric targets.

2. Methodology

2.1. Protons cross-sections

For inelastic collisions, ionisation and excitation in our case, the first born approximation (FBA) discussed in Landau and Lifshitz (1970) and Bethe (1930, 1933) was used when the proton's kinetic energy is above 500 keV, which corresponds to a much higher speed than the orbital electrons of the target in their excited or ionised state. The energy loss function (ELF), also called the 'Bethe surface' characterizing the target was thoroughly studied by Emfietzoglou for water in its 3 different phases. The only available experimental ELF of water molecules in the optical limit range ($q=0$) were reported by Heller et al. (1974) and Hayashi et al. (1998). From those data, calculation details of the ELF for positive momentum transfer range ($q>0$) have been published by Emfietzoglou et al. 2003c, 2005a,b, 2006a,b, 2007 and 2005b). *5 ionisation* states and *5 excitation* states for water molecules in the liquid state were considered using 3 different parameter sets for the calculations. The first and the second sets were adapted by Emfietzoglou to fit to the experimental data of Heller and Hayashi, respectively, while the third set was the one adopted by Dingfelder et al. (1998), which was also based on Heller's data. Although two of these sets are based on the same experimental values (Heller's), they show differences for the partial shell ionisation and excitation cross-sections, however the total ionisation cross-section and the resulting stopping power are very similar in both cases. Unfortunately, as far as we know, excepting the experiments of Paretzke (1988) on water vapour (also see Fig. 6 in Dingfelder et al. 2000), there are no other experimental data to confirm one of these two parameters sets by showing the excitation–ionisation ratio or even the interaction ratios related to the contribution of each shell. Since excitation energy loss in this case contributes no more than ~5% to the total energy loss and the final total stopping cross-sections agree well with the ICRU report 49 values as verified by Dingfelder et al. (2000), we consider these values to be sufficiently correct for Monte-Carlo simulations at the molecular scale.

We obtain thus, three different cross-section sets covering the same energy range for fast protons. Users can choose which set to use by selecting the corresponding cross-section file name in the process class. In the future, a specific method will be introduced into the Geant4 code in order to facilitate switching between cross-section sets in any user application without interfering with the Geant4 source code files.

When the incident projectile velocity becomes comparable to the velocity of the target's orbital electrons, the FBA becomes inapplicable. For this reason, for protons with kinetic energy below ~500 keV the semi-empirical model proposed by Rudd et al. (1985, 1992) (see also Dingfelder et al. 2000) was implemented for ionisation, and the Miller and Green (1973) method fitted to Yousif et al. (1986) data for excitation.

For charge transfer, although the data sets are not all in perfect agreement, they are relatively sufficient to describe this kind of interactions (Toburen, 1998; Dagnac et al., 1970; Berkner et al., 1970). For electron capture and hydrogen stripping we used the analytical fit expressions described in Dingfelder et al. (2000), as they provide an accurate fit to most of the available experimental data.

With decreasing energy of the ion, nuclear interactions and elastic collisions become more frequent. For protons in water, nuclear energy loss is reported to be about 5% at 5 keV and reaches 24% at 1 keV (ICRU report 49, 1993). Ions elastic scattering can be handled by the multiple scattering model of Urban especially adapted for the standard Geant4 processes ("G4hMultipleScattering" class), it is detailed in Urban (2006) and in the Geant4 physics reference manual available on the Geant4 official web site. The model is based on the Lewis (1950) multiple scattering model taking into account angular deflection and spatial displacement of the particle, without the usual small-angle approximation, giving relatively good results for small and large angles according to Lewis (1950). Since the standard processes, including multiple scattering, are recommended for energies above 1 keV in Geant4, this energy restriction is also adopted for the Geant4-DNA ions processes. Actually it is hard to find reliable data for ions below 1 keV in liquid water. In this case, limiting the processes usage to above this energy limit can be a solution when the target is small and the energy of the ion does not go below 1 keV within the target dimensions. Beside, usually in radiobiology and other applications like space radiation for example, interest is focused on high energy particle tracks crossing small dimensions cell structures and not on simulating stopping particles like in medical radiation applications.

2.2. Secondary electron direction

The same approach is used for fast and slow protons to calculate the ejection angle of the electrons produced. For hard collisions where the secondary electron energy exceeds 100 eV we used energy and momentum conservation as described by Emfietzoglou et al. (2000) based on Rudd et al. (1992), and the polar angle is given with respect to the incident particle movement direction as follows:

$$\theta = \cos^{-1}\left(\sqrt{\frac{W}{W_{\max}}}\right) \quad \text{where } W_{\max} = 4T \tag{1}$$

For secondary electrons with energies below 100 eV the previous formulae is no longer valid and electrons are generated in an isotropic manner with $\theta \in [0,\pi]$.

2.3. Alpha particle cross-sections

For alpha particles a speed scaling procedure with a scaling factor is possible using protons already calculated cross-sections for ionisation and excitation (Dingfelder et al., 2006). The scaling factor is an effective charge that takes into account the screening effect of the nuclear charge caused by the boundary electrons of the incident particle and thought it depends on the energy transfer during the collision. Different effective charges were implemented for He^0, He^+ and He^{++} projectiles (Dingfelder et al., 2005). Charge transfer processes were calculated using analytical fit by Dingfelder et al. (2005).

2.4. Electron cross-sections

2.4.1. Inelastic collisions

For ionisations and excitations we followed the dielectric formalism as described by Emfietzoglou and Moscovitch (2002a), Emfietzoglou (2002b), Emfietzoglou et al. (2003a), Emfietzoglou (2003b) and Emfietzoglou and Nikjoo (2005c) Nevertheless, only for the K-shell, were the kinetic energy of the electrons in that