Energy deposition model based on electron scattering cross section data from water molecules

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Abstract. A complete set of electrons scattering cross sections by water molecules over a broad energy range, from the meV to the MeV ranges, is presented in this study. These data have been obtained by combining experiments and calculations and cover most relevant processes, both elastic and inelastic, which can take place in the considered energy range. A new Monte Carlo simulation programme has been developed using as input parameter these cross sectional data as well as experimental energy loss spectra. The simulation procedure has been applied to obtain electron tracks and energy deposition plots in water when irradiated by a Ru-106 plaque as those used for brachytherapy of ocular tumours. Finally, the low energy electron tracks provided by the present model have been compared with those obtained with other codes available in the literature.

1. Introduction

Important medical applications of radiation for therapy, diagnostics or patient protection require energy deposition models with increasing resolution. When molecular details are important, as it is the case of DNA damage studies, spatial resolution should be within the order of magnitude of nanometre. For this level of description atomic and molecular properties of the target need to be taken into account. High energy radiation produces abundant secondary electrons which are the main responsible of the energy transfer map and the radiation damage. Pioneering studies of H. Huels, L. Sanche and co-workers [1-3] show that even sub-ionising electrons could produce damage, in terms of strand breaks and molecular dissociations, more efficiently than traditional ways based on ionisation of the medium. Extensive experimental [4-16] and theoretical [17-21] studies have been carried out in recent years to corroborate and quantify these early evidences. However, most medical procedures are based on radiation dose. This means that the magnitude of reference is the energy absorbed per mass unit and therefore only a macroscopic description of the procedure can be achieved. Medical protocols, or codes of practice [22], recommend using ionization chambers calibrated in terms of absorbed dose to water to characterise the radiation beams for radiotherapy. Using these devices for dose assignment implies that the equivalence charge-energy is a well known constant, which does not apply for low energy, and obviously ignore any effect due to subionising radiation. In order to increase the accuracy of these devices by improving their spatial resolution, in the last few years a great effort has been paid to develop small ionisation based detectors as proportional counters. Dose measurement procedures based on reduced area detectors have been developed.
in the field of microdosimetry [23]. These techniques increased notably the definition of energy transfer maps but its magnitude of reference is still the absorbed dose, no molecular effects are generally included in such models. For this reason, using energy deposition simulation methods as a complement of dose measurements is strongly recommended for radiation planning in electron, photon and ion beam radiotherapy [24-25]. Numerous Monte Carlo simulation codes have been developed for this purpose, i.e. PARTRAC [26-27], MCNPX [28], PENELLOPE [29-31], GEANT-4 [32-33], etc... Most of these codes are sophisticated programmes managing radiation interaction data and empirical laws applied to representative radiation particles (electron, photons, ions) interacting with a great variety of materials. However, input parameters and mathematical relations used by these codes are generally valid for high energy particles, principally for energies above 100 eV. Most of them are based on the first Born approximation which, as we showed in previous studies [34-37], overestimates the interaction probabilities by about 30% at 10 keV even in favourable cases as electron scattered by light molecules (N₂, O₂, CH₄, CO₂).

These considerations motivated the present study. We are presenting here a new Monte Carlo simulation programme for electron interactions with molecular media from high incident energies, in the MeV range, slowing down to a few eV by successive collisions with the atoms and molecules constituting the medium. The input parameters for this programme are the electron scattering cross sections, both elastic and inelastic, which has been previously measured or calculated depending on the considered process and energy range. To illustrate this report, we have chosen a medium formed by water vapour. Water is one of the main constituent of living tissue so it is strongly relevant for medical applications. As biomolecular systems are normally surrounded by liquid water, present results will be generalised to liquid conditions by changing the target density. At this stage, no physical properties of liquids, as collective effects, are included in the present model and they will be the subject of further studies.

2. Cross sectional data
In order to define the trajectories of each particle related to the radiation beam, interaction probabilities, or cross sections, for all the accessible interaction processes are in principle needed. When a collision event takes place an amount of the incident energy can be transferred to the target as internal energy (inelastic scattering) or not, simply transferring linear momentum (elastic scattering). In both cases, the energy and direction of emerging particles should be known. In other words, we need to know both differential and integral cross section for all the possible collisions, elastic and inelastic, that can take place in the considered energy range. In order to organise data requirements, the energy range considered in this study has been divided into three regions: a high energy region from 10 keV up to several MeV, an intermediate-high energy region from 10 eV to 10 keV and finally a low energy region for energies below 10 eV. As far as the electron molecule interactions are concerned, this study has been mainly focused on obtaining data at intermediate and high energies. So we are firstly describing the experiments and calculations which have been carried out in this energy range and then going to the high and low energy domains, respectively.

2.1 Intermediate-high energies (10-10000 eV):

2.1.1 Experiment: Measurements have been devoted to obtain total electron scattering cross sections (TCS), ionisation cross sections (ICS) and electron energy loss (EEL) spectra for incident energies between 50 and 5000 eV. Details on the experimental techniques as well as a complete discussion of the results can be found in Ref. [38], thus only a brief summary is given here: TCS have been measured with a transmission beam technique [39] which also allows measurements of energy loss spectra for small scattering angles from 0 to 10 deg. In order to record EEL spectra for higher scattering angles, the scattering chamber was replaced by an effusive molecular beam which allowed scattering angle selection by rotating the electron gun with respect to the energy spectrometer. Ionisation cross sections in this range were derived from the simultaneous measurement of electron and ion currents by applying a synchronised pulsing system to extract the ions from the collision chamber. Present experimental results for TCS and ICS, with estimated errors of 5% and 7% respectively, are shown in Figure 1. Following the discussion of Ref. [38], some previous experimental results [40-42] are also shown in this figure for comparison. A complementary discussion of data on electron scattering from water molecules can be found in Refs. [43,44]. Present ionisation cross section results are also shown in Figure 1 together with previous measurements of Ref. [45]. Finally, by averaging energy loss spectra for these energies as a function of the scattering angle, a
unique energy loss distribution function with statistical uncertainties within 15% has been derived (see Figure 2).

2.1.2 Theory: Calculations were carried out with the optical potential procedure described in previous papers [46-48]. Basically, it is an independent atom representation which includes screening corrections [49] to derive molecular differential and integral elastic as well as integral inelastic cross sections. As an example, total cross sections calculated with this procedure are shown in Figure 1. Following the suggestion of Ref. [50] these calculations include an approximate additional term to take into account rotational excitations. As shown in this figure the agreement between the present TCS measurements and calculation is perfect between 10 and 5000 eV.

Figure 1. Total electron scattering cross sections: ▲, present measurements; ◇ experimental data of Ref. [40]; ▼ experimental data from Ref. [41]; ●, measurements of Ref. [42]; ——, present calculation; — —, theoretical data from Ref. [51]. Ionisation cross sections: △, present measurements; ×, experimental results from Ref. [45].

Figure 2. Energy loss distribution function for electrons in water vapour from 0 to 100 eV.
2.2 Low energy region (<10 eV)

Below 10 eV the situation is not so ideal. Our calculations, including the rotational excitation term agree reasonably with low energy R-matrix calculations from Ref. [51]. However, both differ from representative experimental data by a factor of 2 below 5 eV. The origin of this discrepancy has been discussed previously [38] and it seems to be connected to the target rotational distribution and the acceptance angle of detectors used in the experiments. Since this point has not been completely clarify at the moment, we will take the experimental results of [40] and [42] as recommendable low energy data for our model.

2.3 High energy region (>10000 eV)

For really high incident energy, molecules can be represented as a sum of atoms and both incident and scattered electron can be considered, in framework of the first Born approximation [52, 53], as plane waves. Under these assumptions integral elastic and inelastic cross section calculations can be represented by simple energy dependent formulae [54]. In order to check the validity of this approximation, Figure 3 is a Fano plot (TCS multiplied by energy versus the logarithm of energy) where the convergence of our measurements and calculations with those approximate values for energies above 10 eV is shown. Due to systematic errors discussed elsewhere [34, 35, 54], previous measurements [41] and calculations [55] diverge from this asymptotic behaviour.

![Figure 3. Fano plot (total cross section multiplied by electron energy as a function of energy in a logarithmic scale): •, present experimental data; ○, experimental data from Ref. [41]; ---, present optical potential calculation; —, calculation of Ref. [55]. -- Born-Bethe calculation.](image-url)
Figure 4. Electron scattering integral cross sections from H$_2$O: —, Optical potential total cross sections; ·--·, Optical potential integral inelastic cross sections; ---, Optical potential integral elastic cross sections; ——, Born-Bethe total cross sections; ·--·, Born-Bethe integral inelastic cross sections; ---, Born-Bethe integral elastic cross sections.

Figure 5. Differential cross section for electron scattering from water molecules for energies above 1 keV as a function of the momentum transfer: +, optical potential calculations; ——, Analytical fit for high-energy extrapolation.
predicted by the Born approximation. Integral cross sections calculated by this method from 1 keV to 3.5 MeV are plotted in Figure 4 together with the model potential calculations described in the previous section. As shown in this figure, at 10 keV there is an excellent agreement between both calculations. In order to obtain differential cross section for high energy electrons, Figure 5 shows a plot of all the differential cross section data we calculated with the above model potential method for energies between 1 and 10 keV as a function of the momentum transfer. As predicted by the Born approximation, these differential values follow a simple exponential dependence on the momentum transfer which allows easily the extrapolation of data toward higher energies. For energies above 500 eV the energy los distribution function of Figure 6 has been used. This function includes excitation and ionization of the atomic oxygen inner shell.

As a combination of data presented in the above three sub-sections, a complete set of electron scattering cross sections has been obtained to be used as input parameters of the simulation procedure described in section IV. Total electron scattering cross sections are experimental with total uncertainties of about 5%. Integral elastic cross sections are calculated with numerical uncertainties within 10%. Integral inelastic cross sections are the result of subtracting the elastic ones from the total scattering cross section with an estimated error of 12%. The ionisation cross sections used were experimental with uncertainties of about 7%. Subtracting the ionisation from the integral inelastic cross section, the remainder curve presents two maxima which can be attributed to electronic excitation and neutral dissociation processes. Identifying the excitation with the lowest energy maximum allows a deconvolution of both components. The estimated error for these remainder inelastic channels is about 25%. Further studies would be needed in order to reduce their assigned uncertainty.

3. Source emission spectra
For monoenergetic electron beams, as those produced by accelerators we would have all required data to feed our simulation procedure. However, interesting brachytherapy applications in ophthalmology are using beta emitters as radiation sources [56] since 1966. Properties of Ru-106 radionuclide for high-dose local irradiation were described by Freundlich [57]. Ocular melanoma is currently treated with Ru-106 plaque brachytherapy and extensive statistical studies has been carried out in order to correlate dosage.
with patient evolution [58, 59] as well as comparisons with other radiotherapy techniques [60, 61]. As these studies show, critical out of target region and optic nerve irradiations can affect to visual acuity [62]. This is therefore a clear example in which high accuracy energy deposition models are required. As the ocular medium can easily be assimilated to water, it constitutes an excellent system to check the reliability of the present model. Geometrical aspects of some Ru-106 plaques used in brachytherapy are shown in Figure 7.

3.1 Beta emission spectra: Ru-106 decays to Rd-106, with 373.59 day half-life period, by beta emission of 39.4 keV maximum energy. This quickly decays (30 second half-life period) by 5 different beta emission ways to Pd-106, which is stable, with a maximum energy of 3.541 MeV. In these conditions, the Ru-106 decay series reaches secular equilibrium and its electron emission spectrum is a combination of continuum intensity distributions which extends from relatively low energies up to 3.541 MeV. To cover such broad energy range, a telescopic mounting of three silicon (SiPAD) detectors, of 1500 micron thick and 50x50 mm$^2$ surface each, has been installed and calibrated with standard beta and alpha sources. By software analysis of coincidence spectra from each detector, the whole electron emission spectrum can be reconstructed (see Figure 8).

3.2 Photon emission spectra: As a consequence of the Ru106/Rh106 beta decay there is a subsequent gamma and X photon emission which requires to be considered by the model. Standard calibrated solid state spectrometers have been used to determine the energy and intensity of emitted photons. Representative spectra are shown in Figure 9 for (a) X ray emission and (b) gamma ray emission.
4. Monte Carlo simulation procedure

As we have already mentioned, the final goal of this study is to develop a model to simulate electron tracks in water to provide information about the energy deposition and the molecular interactions taking place during the energy degradation procedure. The simulation programme we developed for that was based on the GEANT-4 code [32]. We only used from GEANT-4 the general Monte Carlo routines available on its tool kit but all the physics related to electron molecule interactions constitute a new C++ programme, developed by us, which uses as input parameter the electron scattering cross sections we previously measured or calculated and the experimental electron energy loss spectra. Technical details on this programme can be found in previous publications [63, 64]. Photons are also included in the procedure but according to available photon interaction library [65], their main effect is to produce high energy secondary electrons which enter directly in the electron simulation process.

Track simulation of electrons in water emitted by a plane plaque of Ru-106, with the initial energy distribution given by the spectrum of Figure 8, is shown in Figure 10. We assumed the target density corresponding to liquid water but using the cross sectional data of isolated water molecules. Therefore, collective liquid properties are not taken into account in this model and will be the subject of further investigations. An energy deposition diagram as a function of the penetration of the electron beam in water is given in Figure 11.

In order to show details of the information provide by the model, Figures 12 and 13 represent single tracks for 2 keV electrons in water vapour (200 Torr pressure). Figure 12 shows the energy degradation procedure by successive collisions with target molecules. Different interactions taking place along the tracks are shown in Figure 13.

Figure 9. Photon emission spectra of Ru-106 plaque: (a), X-ray spectrum; (b), gamma ray spectrum.
**Figure 10.** Track simulation of electrons emitted by Ru-106 plaque: Left, tridimensional plot; right, vertical section.

**Figure 11.** Energy deposition of Ru-106 plaque emission in liquid water as a function of penetration depth (X).
Figure 12. Modeling 2 keV electron tracks in 200 Torr of water vapour. Energy degradation procedure by successive collisions is represented by the colour code shown on the left.

Figure 13. Modeling 2 keV electron tracks in 200 Torr of water vapour. Colliding processes are shown: •, Auger electron generation; *, ionisation; ●, electronic excitation; ●, neutral dissociation. Finally, a comparison between the present simulation and that obtained with the PENelope code [29-31] is shown in Figure 14. Both simulations predict a maximum electron penetration in the medium of about 2 mm and therefore the absorbed dose in a 2x2x2 mm³ volume would be the same. However, single electron tracks and energy deposition events given by each model are completely different. The number of
interactions predicted by PENELOPE is less than the present one being higher the energy transferred in each individual collision. Also scattering angles given by PENELOPE simulation tend to be higher than ours. These differences can be explained looking at the energy loss distribution function (see Figure 15). Our energy loss distribution function is a continuous line with a threshold at 6.5 eV which follows the observed electron energy loss spectra in water while that of PENELOPE is an arbitrary delta function set placed on strategic energies to give the conventional value of the electron stopping power in water [66]. This could justify the macroscopic agreement between both models being so different their microscopic descriptions.

Figure 14. Simulation of 5 keV electron tracks in 1 atm of water vapor. Left, present Monte Carlo code; right, PENELOPE code.

Figure 15. Comparison of (0-100 eV) energy loss distribution functions: —, present simulation code; —, PENELOPE code.
5. Conclusions
A comprehensive set of electron interaction cross sections both elastic and inelastic with water molecules has been provided in this study by combining experimental results and theoretical calculations, covering a broad range from the MeV down to the meV domains. Consistency of data has been proved for energies above 10 eV. Below this value, important discrepancies appear between theory and experiment which will require further studies in order to be clarified. Using these cross sectional data and experimental energy loss spectra as input parameters, a simulation Monte Carlo program has been developed providing detail of electron tracks, energy deposition and interaction processes at the molecular levels. While macroscopic results of these simulations, in terms of absorbed dose, are in agreement with those given by available simulation programs they give a completely different microscopic description.

Finally the model has been applied to determine the energy deposition in water of Ru-106 beta emitter nuclides (Ru-106/Rd-106) which are commonly used in brachytherapy. The efficiency of these models in medical practice should be checked.

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