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STUDIES ON MICRODOSIMETRIC EFFECTS OF PROTON AND ALPHA PARTICLES INCIDENT ON WATER *)

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UNIVERSITY OF ATHENS

STUDIES ON MICRODOSIMETRIC EFFECTS OF PROTON AND ALPHA PARTICLES INCIDENT ON WATER*

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ABSTRACT. An analytical Monte Carlo simulation code has been developed for nano- and micro-dosimetric calculations in Radiobiological experiments with protons and α-particles irradiation to cell monolayers. For the tracking of charged particles (primaries and secondaries) a semi-empirical single differential (in ejected electron energy) ionisation cross section has been constructed based on photoabsorption, photoionisation and proton impact ionisation data, and has been found to be consistent with available experimental data on both differential and total ionisation cross sections. Detailed distributions of energy deposition and ionisation have been calculated for the considered conditions of irradiation, whereas estimations have been made on the production of the primary chemical species, known to lead to the formation of free radicals.

1.INTRODUCTION

The last years it has been proposed that DNA double strand breaks (DSBs) may play a significant role in the biological effects of ionising radiation . The radiation damage to DNA can arise from direct energy deposition on its chemical constituents (direct action) or can be due to radicals produced by the radiolysis of the surrounding aqueus medium (indirect action) (Chadwick and Leenhouts 1981, Barendsen 1990, Chatterjee and Holley 1990, Frankenberg et al 1990, Stanton et al 1990). Therefore, an understanding of the physical and chemical processes (energy transport and deposition, ionisation distributions or production of primary chemical species) leading to the possible formation of DSBs would be of importance in determining the chain of events responsible for cell damage or indeed for any cell transformation following irradiation. Track structure simulation provides valuable information on the physical and chemical interactions occuring in cellular and subcellular level, as average quantities may provide only poor indications on the frequency distributions of actual depositions.

We have developed a Monte Carlo computer code which simulates the track of 0.3-10 MeV protons as well as that of α-particles (0.8-10 MeV) and the secondary ejected electrons, through small sites (1nm-1μm range), considering water as the biological material. The particle's track modelling, taking into account all the possible interactions with the molecules of the medium, requires accurate differential and total cross-sections. For the ionisation, which is the most probable process in the energy range considered here, the experimental data available not only are very rare, but also include large uncertainties and are inadequate for use

in track structure simulations, since the knowledge of succesive cross-sections as a function of both the incident and ejected particles energy is required. Therefore we have further developed a model for the determination of the single differential ionisation cross-section (SDICS) which combines the classical binary encounter approximation (BEA) with the Bethe theory, and is consistent with experimental data for both differential and total ionisation cross-section (TICS). The model has the advantage of providing the contribution of the molecular subshells to the interaction, a fact which leads to the determination of the primary chemical species formed.

The dosimetric results obtained in this work have been used in the design of an experiment that is in progress, in which DNA solution, and cell monolayers are irradiated by α-particles from an ^{241}Am source.

2. SINGLE DIFFERENTIAL IONISATION CROSS SECTION (SDICS)

2a. Protons and α-particles

The uncertainty of the experimental data for the SDICS in the case of protons and a-particles (0.3 - 2 MeV) with water vapor is reported to be as much as 100% for ejected electrons with energy of 5eV, whereas that uncertainty decreases to 20% for electron energies of 10eV and above (Toburen and Wilson 1977, Toburen et al 1980). These uncertainties produce discrepancies of the order of 30% between the independently determined TICS (experiment by Rudd et al 1985, reported uncertainty 8%) and the TICS calculated from the integration of the experimental SDICS over secondary electron energy. Also it should be noted that, owing to experimental limitations, it is not possible at present to determine experimentally the subshell from which the secondary electron is ejected and subsequently the kind of ion that is formed. It is though a well known fact that these ions play an important role in the indirect action of radiation.

As it was pointed out by many investigators ,the first Born approximation and, also, impulse approximations, play a significant role in the determination of the energy distribution of secondary electrons ejected by fast charged particles from molecules (Inokuti 1971, Kim 1975a, Wilson et al 1984). Slow secondary electrons are mostly produced by glancing collisions in which the dipole interaction dominates. Here, the Born approximation relates the secondary electron spectra to the photoionisation spectra, and as it is well known the leading part of the Born cross-section is proportional to the dipole oscillator strength (Inokuti 1971, Kim

1975a, Berkowitz 1979, Wilson et al 1984, Miller et al 1987). Hard collisions are very important in the ejection of high energy secondary electrons (where quantum effects are less important) and, therefore, semiclassical theories like the binary encounter approximation (BEA) are, in general, adequate for predicting SDICS (Kim 1975a).

Along the lines suggested by Inokuti (1971), Kim (1975a) and Miller et al (1987) on Bethe's theory, based on the first Born approximation, the SDICS for the ejection of secondary electrons with energy between W and W+dW, from the k subshell of a molecular target by a projectile with charge z and velocity u, can be written as:

$$\frac{d\sigma_k}{dW} = \frac{4\pi a_o^2 z^2}{T}\left[A_k(W)\ln\frac{4TR}{(W+I_k)^2} + B_k(W)\right] \qquad (1)$$

where a_0 is the Bohr radius, R is the Rydberg energy, I_k is the ionisation potential of the subshell k, and $T=mu^2/2$ with m being the electron mass. The functions $A_k(W)$ and $B_k(W)$ depend rather on the target molecule than on the incident particle.

The term $A_k(W)$ represents the effect of glancing collisions, and can be written as:

$$A_k(W) = \frac{R^2}{W+I_k}\frac{df_k}{dW} \qquad (2)$$

where df_k/dW is the partial differential optical oscillator strength for the ionisation of subshell k and is related to the photoionisation cross-section $\sigma_{ion.}$ (Tan et al 1978, Wilson et al 1984, Gallagher et al 1988) by the expression:

$$\frac{df_k}{dW} = \left(\frac{df_k}{dE}\right)_{E=W+I_k} = BR_k\frac{df}{dE} = BR_k\frac{mc}{\pi e^2 h}\sigma_{ion.} \qquad (3)$$

where m, e are the electron mass and charge respectively, c is the velocity of light, h is Plank's constant, BR_k is the branching ratio for the subshell k and E is the photon energy. Several experimental data were used for the evaluation of the terms $A_k(W)$. Photoionisation cross-section values were obtained from Katayama et al (1973), Tan et al (1978), Berkowitz (1979), Brion and Thomson (1984b), Haddad and Samson (1986), while for the determination of the partial optical oscillator strengths, branching ratios of Blake and Carver (1967), Truesdale et

al (1982), Brion and Thomson (1984a), Brion and Carnovale (1985), Banna et al (1986), Brion et al (1986) were used. In figure 1, the terms $A_k(W)$ for the 4 outer subshells of water molecule are plotted versus the energy W of the ejected electron, while the term $A_5(W)$ corresponding to the inner subshell (K-electrons) is omitted as it contributes in less than 0.5% .

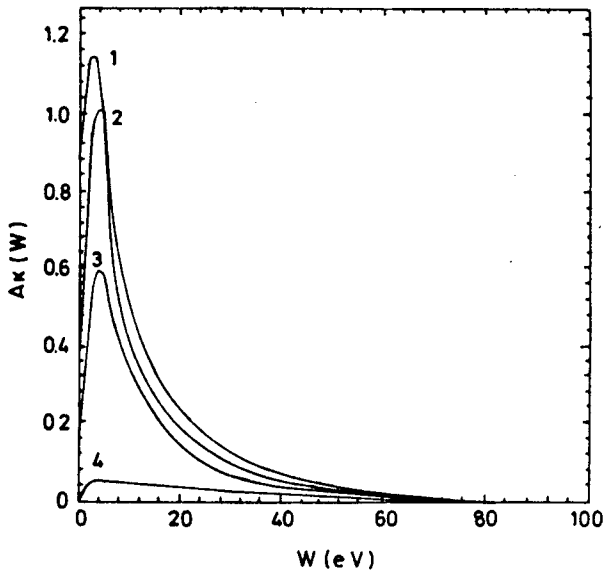

Figure 1. The terms $A_k(W)$, k=1-4, of equation (2) as a function of the energy W of the ejected electron.

The term $B_k(W)$ represents the hard collision component of the secondary electron spectra and is expected to dominate at large secondary electron's energies W. This term has two fundamental properties : a) at large enough T it is independent of T and b) at large W, as indicated by the BEA, it is approximately equal to:

$$\lim_{W\to\infty} B_k(W) = \frac{T}{4\pi a_o^2 z^2}\left(\frac{d\sigma_k}{dW}\right)_{BEA} = n_k R^2 \left[\frac{1}{(W+I_k)^2} + \frac{4}{3}\frac{U_k}{(W+I_k)^3}\right] \qquad (4)$$

where n_k is the number of electrons in the kth subshell of the molecule (for water k=5, n_k=2) and U_k is their average kinetic energy. Values for the average kinetic energies U_k and ionisation potentials I_k were taken from Bolorizadeh and Rudd (1986). In these notations we represented $B_k(W)$ as:

$$B_k(W) = f(W)\, n_k R^2 \left[\frac{1}{(W+I_k)^2} + \frac{4}{3}\frac{U_k}{(W+I_k)^3}\right] \qquad (5)$$

where f(W) is a function suggested to depend on target properties only and to be independent of the molecular subshell (Miller 1989).

The proposed SDICS has the final form:

$$\frac{d\sigma}{dW} = \frac{4\pi a_o^2 z^2}{T}\left\{\sum_{k=1}^{5}\left[A_k(W)\ln\frac{4TR}{(W+I_k)^2} + f(W)\, n_k R^2\left(\frac{1}{(W+I_k)^2} + \frac{4}{3}\frac{U_k}{(W+I_k)^3}\right)\right]\right\} N \qquad (6)$$

where N is a factor that has been introduced in order to take into account the normalisation of the integrated SDICS to the TICS. This representation of the SDICS as a sum of the various subshells contribution to the interaction allows for the determination of the ions formed after the ejection of the secondary electron (see section 3).

Both the function f(W) and the normalisation factor N can be obtained from experimental data. We have used the experimental data given for the SDICS by Toburen and Wilson (1977) and Toburen (1988) at proton energies of 0.3, 0.5, 1 and 1.5 MeV for the estimation of the f(W) according to the equation:

$$f(W) = \frac{\dfrac{T}{4\pi a_o^2 z^2}\left(\dfrac{d\sigma}{dW}\right)_{exp.} - \sum_{k=1}^{5}\left(A_k(W)\ \ln\dfrac{4TR}{(W+I_k)^2}\right)}{\dfrac{T}{4\pi a_o^2 z^2}\left(\dfrac{d\sigma}{dW}\right)_{BEA}} \qquad (7)$$

We found, indeed, that the function f(W) does not show any dependence on the energy of the protons in this energy range. Nevertheless we used an average value for the function f(W), provided from the above four proton's energies, to make up for the experimental uncertainties. The variation of the f(W) as a function of ejected electron's energy W is presented in figure 2. As it was theoretically expected, we notice that f(W) converges to 1 even from as low as 100eV secondary electron energy. Consequently for larger W the function $B_k(W)$ is represented by equation (4).

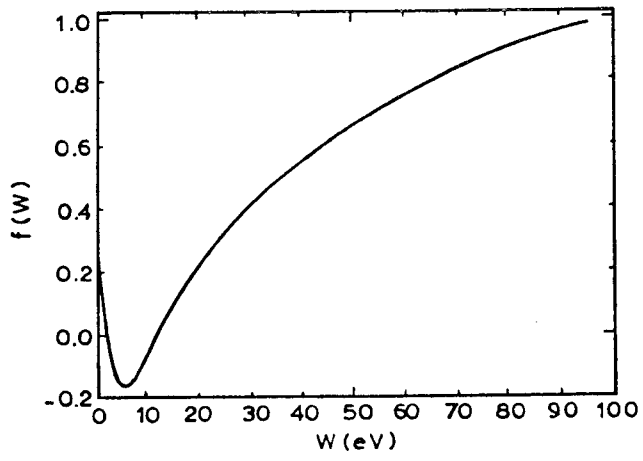

Figure 2 The term f(W) of equation (6) as a function of the ejected electron's energy W.

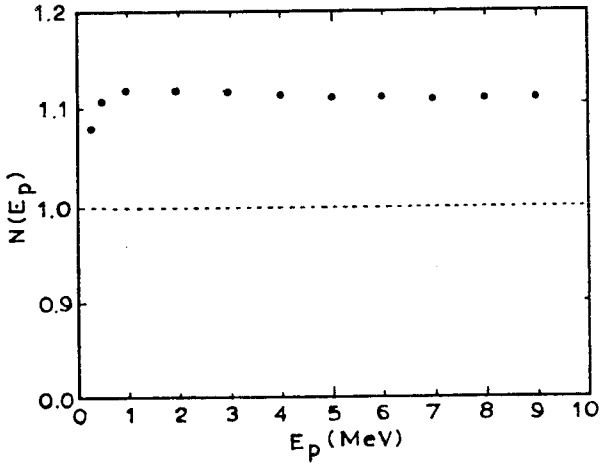

Figure 3 Ratio $N(E_p)$ of the experimental TICS to the integrated model calculations as a function of proton's energy E_p.

The integration of the model SDICS over the ejected electron's energy W, gives the model total ionisation cross-section (TICS). This integration is usefull for normalising the SDICS, because the TICS for proton impact on water are relatively well known since the experimental values obtained by Rudd et al (1985) are reported to have an 8% uncertainty above 0.5 MeV. In figure (3) the ratio $N(E_p)$ of the experimental TICS to the integrated model calculations is presented as a function of proton's energy E_p, and one can observe that the experimental results are larger by a constant factor of 1.1 for proton energy above 0.5 MeV. This ratio represents the normalisation factor N of equation (6). The procedure that was presented here is in accordance with Kim's (1975b) suggestions, claiming that the use of optical data to the SDICS gives the correct "shape" of the distribution while the actual value can be derived from normalisation to the TICS.

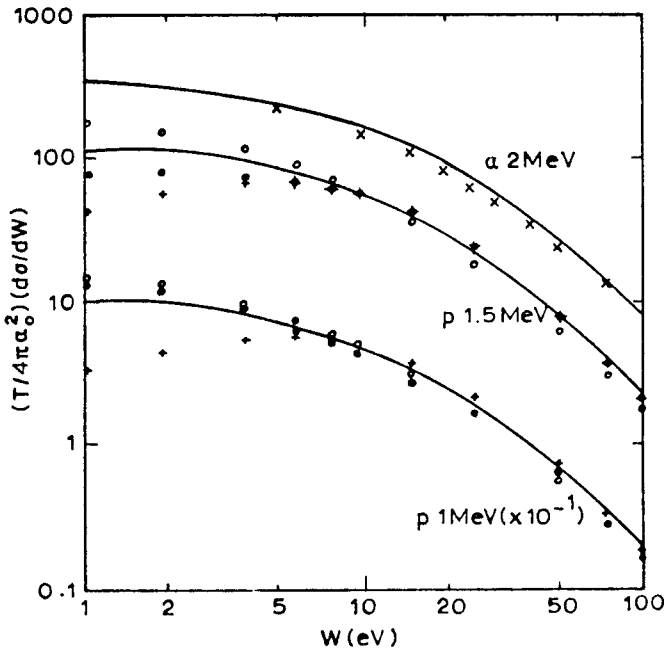

Figure 4 The model SDICS (see text) is presented (solid line) for 1MeV, 1.5MeV protons and 2MeV α-particles, along with data (●, Toburen and Wilson 1977; x, Toburen et al 1980; +, Inokuti et al 1980; o, Rudd 1990).

Some of our results on SDICS are presented in figure 4 in the low energy region (W < 100eV) of the ejected electrons where the majority (>90%) of these electrons can be found and the larger discrepancies are expected. These cross-sections are conveniently multiplied by $T/4\pi a_o^2$ in order to remove the principal dependence on energy. For comparison, our results for 1 and 1.5MeV protons as well as 2MeV α-particles (a simple z^2 scaling on equally velocity's proton data was assumed) are given, along with corresponding experimental data (Toburen and Wilson 1977, Toburen et al 1980) and recent theoretical approximations (Inokuti et al 1987, Rudd 1990). As it can be seen our model reproduces, within experimental uncertainties, the existing experimental data while the calculations of Inokuti et al (1987), based on the empirical model of Wilson et al (1984), seems to underestimate significantly the experimental data at low energies of ejected electrons. A more systematic comparison of our model calculations with the corresponding ones of Rudd (1990) reveals that the latter tends to give higher values at low secondary electron energies. Similarly, Long and Paretzke (1991) have noticed recently that Rudd's model increases sharply at these low

secondary electrons energies.

So far our model can be viewed, simply, as a data extrapolation and interpolation procedure, which is, however, very usefull in transport calculations where SDICS over a broad range of primary and secondary energies are needed. The model enables us to make predictions on SDICS for energies down to 0.3 MeV for protons and 0.8 MeV for α-particles.

2b. Electrons

In the case of the ionisation of water vapor by incident electrons the indistinguishability of the scattered and ejected electrons must be taken into account and the Mott cross section is appropriate in place of the BEA in equations (4), (5) and (6) (Kim 1975a, Bolorizadeh and Rudd 1986). The function f(W) is assumed to be the one used for protons and α-particles, as it depends, mainly, on target properties. The SDICS for incident electrons is then:

$$\frac{d\sigma}{dW} = \frac{4\pi a_o^2 z^2}{T}\left\{\sum_{k=1}^{5}\left[A_k(W)\ln\frac{4TR}{(W+I_k)^2} + f(W)\,n_k R^2\left(\frac{1}{(W+I_k)^2} + \frac{1}{(T-W)^2} - \frac{1}{(W+I_k)(T-W)}\right)\right]\right\}N \quad (8)$$

Figure 5 compares our results on SDICS with the corresponding experimental data for the ionisation of water vapor by 500eV and 1keV electrons (Opal et al 1972, Vroom and Palmer 1977, Bolorizadeh and Rudd 1986). Although there is a good overall agreement, significant differences with Bolorizadeh and Rudd (1986) data are observed when the ejected and scattered electrons have about the same energy and when the detected electrons are close to the primary energy. However, more recent experimental data (Hollman et al 1988), support the predictions of our model, as they indicate that experimental uncertainties cause the mentioned discrepancies.

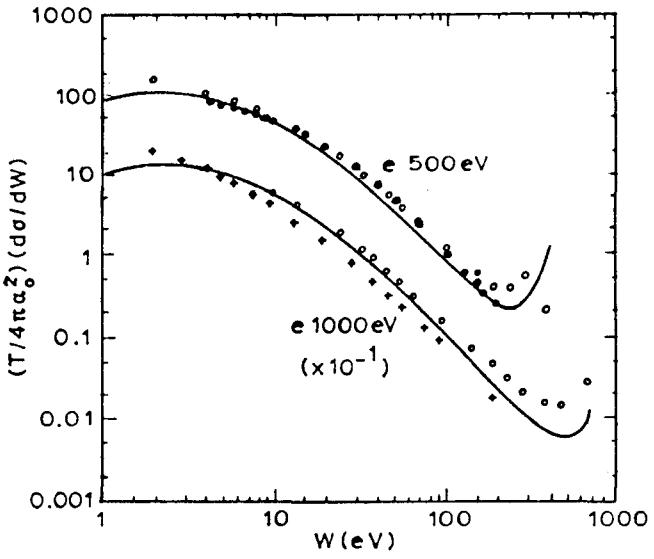

Figure 5 The model SDICS (see text) is presented (solid line) for 500eV (low), and 1keV (upper) electrons in comparison with experimental data (•, Opal et al 1972; ▫, Vroom and Palmer 1977; o, Bolorizadeh and Rudd 1986).

The fact that the experimental data used for comparison were not used in the

optimization procedure allows us to feel that our model can predict SDICS for electron impact on water with reasonable accuracy, for electron energies larger than 100eV, needed in transport calculations. To be consistent with TICS, the data of Schutten et al (1966) were used for normalisation.

3. MONTE CARLO CODE

A Monte Carlo simulation code has been developed for the passage of protons ($E_p > 0.3$ MeV), α-particles ($E_\alpha > 0.8$ MeV) and secondary electrons ($T_e > 100$ eV) through water, based on the derived SDICS. At these energies the difference in the cross-sections for the gaseous and the liquid phases of water is expected to be quite small (<10%) (Thwaites 1981, Long and Paretzke 1991).

A free path between collisions is selected, as usual, from the total interaction (excitation and ionisation) cross-section. The kind of interaction occured (excitation or ionisation) is decided from the relative values of the corresponding cross-sections, considering the excitation total cross-section as 1/3 of the corresponding ionisation one. We assume that if excitation is selected, then 12.4 eV energy is deposited by the particle in the interaction point. These approximations to the excitation process proposed by Berger (1988), are believed not to cause considerable errors in the calculations, as the energy lost from the primary particle due to excitation is less than 5% of the total energy loss. In the case of ionisation interaction, the kinetic energy W of the ejected electron is selected from the SDICS (equation (6) for protons and α-particles). Actually a two dimentional histogram in a convenient W and T grid has been constructed for sampling the SDICS. The energy transferred from the primary particle is then $E = W + I_k$. Thus the primary particle's energy loss E (in a single scattering) depends on the subshell with ionisation potential I_k, from which the electron was ejected. The selection of this subshell is based on the relative contribution of each partial cross-section $d\sigma_k/dW$ to the SDICS, for the known secondary electron's energy W (equation (6)). The primary particle's energy, during the tracking, is reduced by E after each interaction.The kind of ion that is formed in the point of interaction depends on this particular subshell as it was pointed out by Tan et al (1978). The angles of the ejected electron with energy less than 100eV are selected randomly as the low energy secondary electron emission is almost isotropic (Long and Paretzke 1991). For energies beyond this value we accepted the validity of the free-electron scattering.

The secondary ejected electrons history is followed separately by our code. For the results presented in this study the electrons with energy less than 100 eV, which constitute more than 90% of their total number, are assumed to deposit their energy locally, as they are absorbed in less than 3-5 nm distance (Combecher and Kollerbaur 1985, LaVerne et al 1991). In this low electron energy range the relative contribution of the primary chemical species formed are based on the experimental data of Schutten et al (1966). For the ionisation process we used equation (8), while for the excitation and for the elastic scattering the model of Green and Stolarski (1972) and that of Senger et al (1990) were used respectively.

The facilities (geometry, storage and manipulation of data, fitting procedures etc) provided by our photon and electron Monte Carlo code were used in our present code for protons and α-practicles (Angelopoulos et al 1991).

4. RESULTS AND DISCUSSION

The results reported here, are referred to interactions of protons (0.3-10 MeV) and α-particles (0.8-10 MeV) with water layers. The geometry selected corresponds to the one used in Radiobiological experiments for the irradiation of cell monolayers. The particles are assumed to be, in principle, incident perpendicularly to the layers and various physical and chemical parameters are recorded.

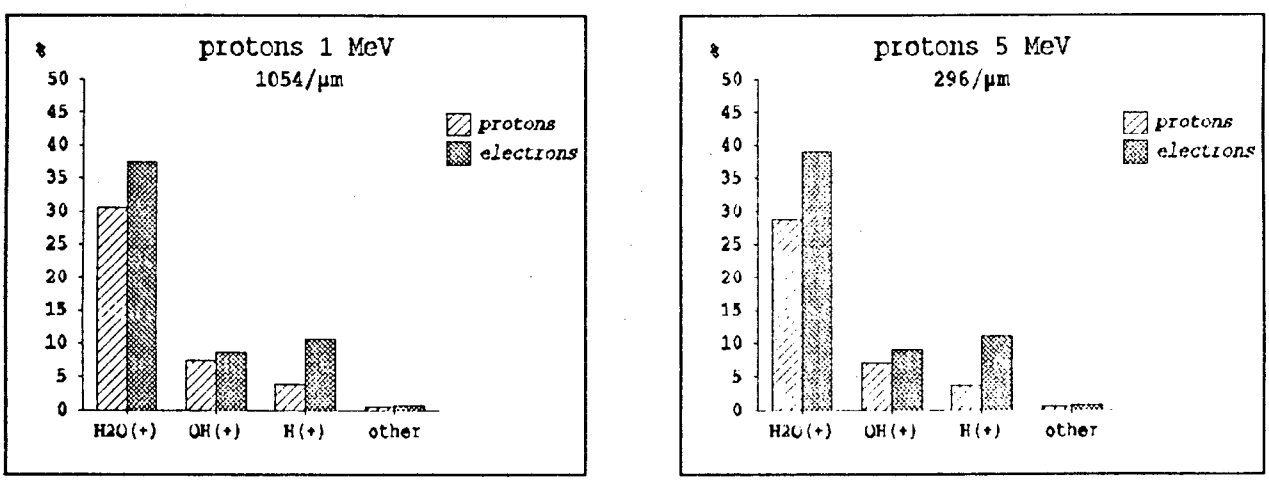

Figure 6. Primary chemical species formed by primary protons and the ejected electrons: 1 MeV protons (left), 5 Mev protons (right)

Preliminary results for the primary chemical species formed by 1MeV and 5MeV incident

protons and the ejected electrons are presented in figure 6. The total ions formed per μm of proton's track are 1054±100 for 1MeV and 296±30 for 5MeV protons. As it can be seen, the branching ratios for the production of H_2O^+, OH^+, H^+ and other (O^+, O^{++} etc), are approximately independent of proton's energy. These ions play an important role in the generation of the hydroxyl radical,

$$H_2O^+ + H_2O \rightarrow H_3O^+ + \cdot OH$$

which contributes to DNA damage (indirect action of radiation). Some unexpected differences can be observed, however in figure 6, between protons and ejected electrons data, which is more pronounced in the case of H^+. We feel that this is a reflection of the use of Schutten's data (1966) for the low energy electrons (see also section 3). Although further work is needed and is currently in progress, our protons results, which are exclusively based on our model calculations, seems to support the calculations of Miller et al (1987).

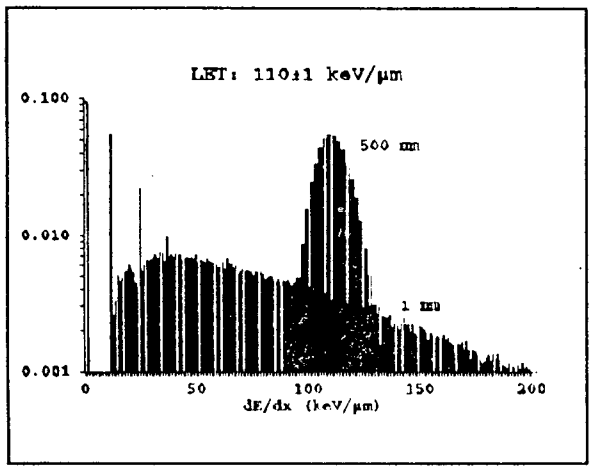

Figure 7 Energy loss distributions (keV/μm) for 4 MeV α-particles for 1 and 500 nm segments. LET value by Thwaites (1981).

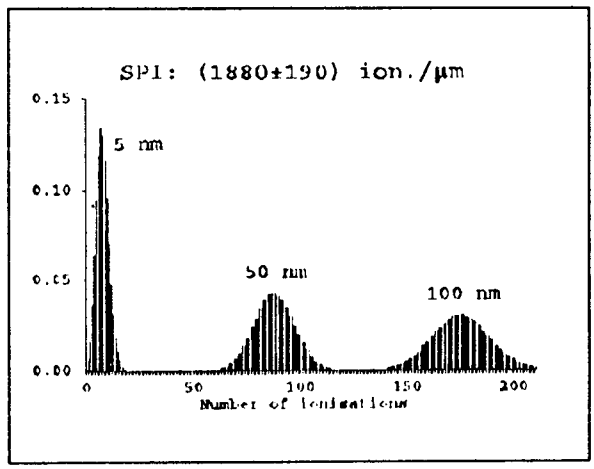

Figure 8 Primary ionisation distributions for 4 MeV α-particles for 5, 50 and 100 nm segments. SPI value by Perris and Zarris (1989).

Both Linear Energy Transfer (LET) and Specific Primary Ionisations (SPI) are extensively used in Radiobiological studies in order to correlate damage induced by ionising radiation in biological systems. As these are average quantities, however, they do not reflect any statistical fluctuations in cellular and subcellular level. Such distributions are presented in figures 7 and 8 for 4 MeV α-particles. In figure 7 the energy loss distribution is presented for 1 and 500 nm particle's track lengths. The distribution for the 1 nm segment is considerably different from the one of 500 nm. For the later the mean as well as the most probable value are well approximated by the corresponding experimental LET values of 110±1 keV/μm and 107±4 keV/μm for vapour and liquid water respectively (Thwaites 1981). The mean value is actually

independent of the segment length but this is not the case with the most probable one value. For the 1 nm distribution the calculated mean value is 110.4 keV/μm with r.m.s. equal to 155.3 keV/μm, while the most probable is zero (about 10% of the α-particles do not interact with the medium at all). The observed discrete energy deposition are due to excitation interactions (12.4 eV energy loss). In figure 8 the distributions of primary ionisations for 4 MeV α-particles are presented for three segments (5, 50 and 100 nm). The corresponding SPI value is 1880 ± 190 ionisations/μm (Perris and Zarris 1989). These figures indicate that average quantities referred to very small volumes may provide only poor indications of the frequency distributions of actual depositions.

In Radiobiological experiments using α-sources, the α-particles can no longer be treated as unidirectional and/or monoenergetic. The energy spectra of common surface α-sources are about symmetrical to the mean value, which can be considered as representative energy of the source for some calculations. For example, this approximation on LET calculations was found to indroduce errors that do not exceed 1-2% for α-energies in the range of 1-5 MeV and spectra spread $\Delta E/E < 30\%$. Absorbed dose calculations, however, based on LET values, introduce much larger errors and the correction of particle's energy after each interaction is required. In figure 9 the ratio of the dose calculations using the energy loss correction to those based on the corresponding LET values (Thwaites 1981) is presented for 2, 3 and 4 MeV α-particles as a function of the particle's pathlength. One can observe that even for a few μm track the error introduced in the calculations without the above mentioned correction could be as large as 35% . Actually, the errors could be much larger since the endpoints presented in figure 9 correspond to 0.7 MeV residual energy.

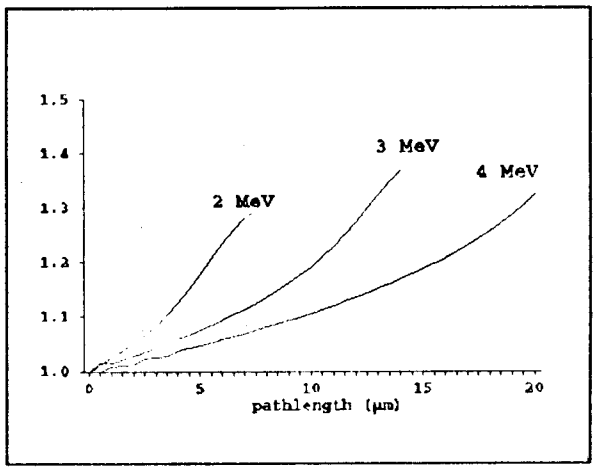

Figure 9 The ratio of the dose calculations with the energy loss correction to those based on LET for 2, 3 and 4 MeV α-particles as a function of the pathlength.

Another factor which also affects dose calculations is the angular distribution of particles incident on sample. The influence of the angle of incidence on a 8μm water-sample on dose calculations is presented in figure 10 for 4MeV α-particles, where the dose relative to perpendicular incidence is given as a function of incidence angle (the energy loss correction was considered in the calculations). In figure 11, the angular distribution of α-particles incident on a 0.4cm radius sample placed on the central axis of a 1.5cm radius surface source is

presented for 1, 5 and 15cm source - sample distance. Figure 11 should be considered in connection to figure 10.

For α-particles irradiation, the source-sample distance is limited by the range of α-particles in air which for practical purposes is less than 2 cm. Measurements and calculations on the energy fluence as well as angular distributions from commonly used α-particles surface sources are largely affected by the accuracy in the determination of this distance. Additionally, the corresponding dosimetric calculations are extremely complicated since the above mentioned corrections for the energy loss and the angular distribution are required.

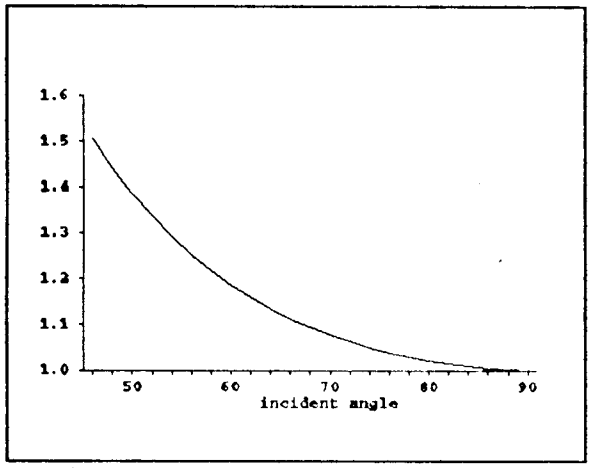

Figure 10 Dose relative to perpenticular incidence as a function of incident angle for 4 MeV α-particles on 8 μm sample.

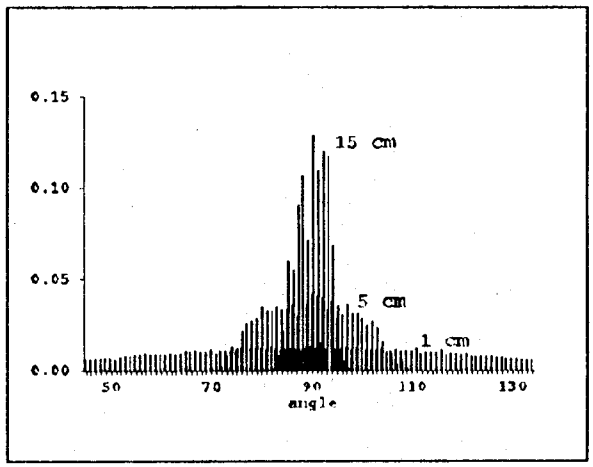

Figure 11 Angular distribution of particles incident on a 0.4 cm radius sample placed on central axis of a surface source (1.5 cm radius) for 1, 5 and 15 cm source-sample distance.

Based on these and similar results (Angelopoulos et al 1990, Zarris et al 1991) an experimental arrangement has been designed and constructed for the irradiation by α-particles (^{241}Am surface source) of DNA solutions and cell monolayers. By creating vacuum of 10^{-1} Torr between the source and the sample (placed on a Myllar foil outside the vacuum) we obtained the advantage to modify at wish the source - sample distance up to 18 cm. This capability allows for the irradiation time to be of the order of minutes instead of less than a second and the uncertainties involved with exposure time (shutter mechanisms) are reduced. Also the system provides a wide range of absorbed dose as these are functions of both distance and time of irradiation.

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