

Radial dose distribution and effective delta ray radius (Penumbra radius): Determination for some ions passing through water

E.M. Awad^{a,*}, M. Abu-Shady^b

^a Physics Department, Faculty of Science, Menoufia University, Shebin El-Koom, Menoufia 32511, Egypt

^b Mathematics and Computer Science Department, Faculty of Science, Menoufia University, Shebin El-Koom, Menoufia 32511, Egypt

ARTICLE INFO

Keywords:

Radial dose
Penumbra radius
Effective δ -ray range
Heavy ions
Bragg peak
LET

ABSTRACT

An analytical equation for calculating radial dose of heavy ions in water is introduced by Awad et al. (Applied Radiation and Isotopes, 142 (2018) 135–142). It is simple alternative to Monte Carlo code and is a promising equation, however, still needs refinement. Refinement was added through adjusting radial dose integration upper limit which gives the effective δ -ray range, r_{max} and ion's penumbra radius in water as well. Radial dose distributions for 85 ions forming fifteen energy groups from 0.25 to 24 MeV/n were studied. By employing the effective δ -ray range, it was possible to get more consistent radial dose distribution in comparison to experimental and Monte Carlo simulation data. The corresponding LET values of those ions were estimated and compared with SRIM program. Penumbra radii for 85 ions were determined. Good description for the penumbra radii was obtained using a proposed new equation which fits experimental data as well.

1. Introduction

Radial dose distribution around the ion trajectory $D(r, R)$ is the energy deposited per unit mass at distance r normal to the ion pass. It is of principal importance in track structure theory and is significant for many applications in biology, hadron therapy as well as electronic devices. It can be used for calculating the inactivation cross section of bacteria and enzymes due to heavy ions [1], for estimating ionization quenching in organic plastic scintillator [2], for determine the detection threshold of SSNTDs [3,4], for expressing the track etchability of some irradiated crystal [5]. Radial dose can also be used to estimate microscopic energy deposition [6] and the number of clustered DNA damage due to irradiation [7,8]. Radial dose is utilized for radiation transport software [9,10], treatment planning for heavy ion radiotherapy, space radiation risk assessment and estimating the cell survival rate for heavy particle cancer therapy [11–13] and predict the cell surviving rate and the relative biological effectiveness (RBE) of the ions [14]. Radial dose can be used to determine lateral distribution of charge due to ion imping in silicon memory cells (penumbra radius) which induce soft error and miscalculation in processing unit causing memory cell upset [15].

Many theoretical, semi-empirical analytical models were proposed to explain the interaction of swift heavy ions with matter. The amorphous track model by Katz and co-workers in 60s of the last century

[16–18] and its reduction model, the local effect model (LEM), by Scholz and Kraft [19,20] were the pioneer models of such subject. A simple analytical model of ion track structure based on classical collision dynamics was introduced by Kiefer and Straaten [21]. Meanwhile, in radiation chemistry a model of heavy particle tracks has been proposed in which [22,23] a dense core region of enormous ionization density is surrounded by a penumbra region where energy deposition occurs mainly in ionization events by energetic secondary electrons released by primary particles. Moribayashi [24] proposing a fitting function that able to calculate the radial dose independent on the incident ion. It is worth noting that radial dose models had many modifications and improvements during the last few decades, however, still need further refinements [25–30]. For more information, please refer to these references and references therein.

Monte Carlo (MC) codes for track structure simulations are widely used to study the biological effects as well as physical, physico-chemical, and chemical stages of the interaction of ionizing radiation with liquid water. MC codes provide detailed information on microscopic energy deposition in the medium [31–34]. In a calculation of a particle track, the primary particle and its secondary particles are followed in their passage through matter by considering all possible interactions until the particle's energy is totally dissipated. Each consecutive interaction is generated by a random sampling procedure. Most of these codes are dealing with electrons excitation and ionization cross

* Corresponding author.

E-mail address: elsayed_awad@science.menoufia.edu.eg (E.M. Awad).

sections. However, another Monte Carlo approach was given in RITR-ACKS program [35,36]. The physical and physico-chemical stages of the radiolysis of water due to heavy ions were simulated. The ion is followed on an event-by-event basis, calculating all ionization and excitation events produced and recording the position of the generated radiolytic species and the energy and direction of the secondary electrons.

Geant4-DNA [9,10,37] is considered as a most powerful MC simulations tool in the present days. It describes, step by step, electromagnetic interactions of particles with the surrounding media where physical interaction processes for electrons such as elastic scattering (7.4 eV–1 MeV), electronic excitation (9 eV–1 MeV), ionization (11 eV–1 MeV), vibrational excitation (2 eV–100 eV), and molecular attachment (4 eV–13 eV) can be simulated. Therefore, Geant-4 DNA provides detailed information regarding interaction of fast ions with matter with the inclusion of all primary excitation and ionization events accompanying the passage of these ions through matter. The radial dose distribution obtained by the present work will be compared to radial dose distribution data obtained by Geant-4 DNA data.

The aim of the present work is to extend the applicability of Awad et al. [38] code for track structure to calculate the effective δ -ray radius, r_{max} . How r_{max} was deduced for every heavy ion in water medium will be explained. The effect of r_{max} on radial dose for 85 ions of different energy groups will be presented. The ions linear energy transfer, LET in water will be estimated from radial dose and compared to SRIM [39] data. The penumbra radii of the different ions that were deduced from effective δ -ray radius were compared with previous equations as well as with experimental data.

2. Methodology

A semi-empirical analytical model based on electronic radiation damage is introduced by Awad et al. [38] for calculating the radial dose distribution $D(r, R)$. $D(r, R)$ is assumed to be the energy deposited per unit mass in a short cylinder whose axis is parallel to the path of the ion. In this approach, the empirical electron range-energy formula [40] was considered where the electron range, R in cm is given as:

$$R(T) = a_1 \left(\frac{1}{a_2} \ln \left(1 + a_2 \frac{T}{mc^2} \right) - \frac{a_3 \frac{T}{mc^2}}{1 + a_4 \left(\frac{T}{mc^2} \right)^{a_5}} \right) \quad (1)$$

$$a_1 = \frac{b_1 A_T}{\rho Z_T^{b_2}}, a_2 = b_3 Z_T, a_3 = b_4 - b_5 Z_T, a_4 = b_6 - b_7 Z_T, a_5 = \frac{b_8}{Z_T^{b_9}},$$

where Z_T and A_T are the atomic and mass number of the target, respectively and b_i ($i = 1, 2, \dots, 9$) are constants independent of absorber material (Table 1).

To calculate the radial dose, two fundamental quantities must be determined. For detail steps and more information, please refer to [38]. The number of liberated electrons in ion-medium interaction and the energy carried by each electron as a function of the radial distance, r .

Table 1

Values of the constants b_i and d_i used for electron range-energy equation, Eq. (1) and its inverse relation of energy-range equation, Eq. (2).

i	b_i	d_i
1	0.2335 ± 0.0091	$(2.98 \pm 0.3) \times 10^3$
2	1.209 ± 0.015	6.14 ± 0.29
3	$(1.78 \pm 0.36) \times 10^{-4}$	1.026 ± 0.02
4	0.9891 ± 0.001	$(2.57 \pm 0.12) \times 10^2$
5	$(3.01 \pm 0.35) \times 10^{-4}$	0.34 ± 0.19
6	1.468 ± 0.09	$(1.47 \pm 0.19) \times 10^3$
7	$(1.18 \pm 0.097) \times 10^{-2}$	0.692 ± 0.039
8	1.232 ± 0.067	0.905 ± 0.031
9	0.109 ± 0.017	0.1874 ± 0.0086

Therefore, the electron energy, T in MeV as a function of its position must be determined. To obtain this, the inverse of the energy-range of Eq. (1) must be implemented [40] as follows:

$$T(R) = 0.511 c_1 \left(e^{\frac{R}{c_1} \left(c_2 + \frac{c_3}{1 + c_4 R^{c_5}} \right)} - 1 \right) \quad (2)$$

where c_i ($i = 1, 2, \dots, 5$) are constant for a given target (Z_T, A_T) and d_i ($i = 1, 2, \dots, 9$) are another constants independent of absorber material and are given in Table 1 as:

$$c_1 = \frac{d_1}{Z_T}, c_2 = \frac{d_2 Z_T^{d_3}}{A_T}, c_3 = d_4 - d_5 Z_T, c_4 = \frac{d_6}{Z_T^{d_7}}, c_5 = \frac{d_8}{Z_T^{d_9}}.$$

Eqs. (1) and (2) are important for defining the necessary constants.

The maximum transferred energy to delta rays is given by:

$$T_{max} = \frac{2mc^2\beta^2}{1 - \beta^2} \quad (3)$$

β is the ion velocity in medium relative to velocity of light in vacuum. T_{max} can also be estimated [41] as follows:

$$T_{max} = \frac{2\tau(\tau + 2)m_e c^2}{1 + 2(\tau + 1)\left(\frac{m_e}{M}\right) + \left(\frac{m_e}{M}\right)^2}, \quad (4)$$

where τ is the projectile ratio of kinetic energy to rest mass energy and M is the mass of the projectile and $m_e c^2$ is the electron mass energy and E is the kinetic energy of the primary particle. It must be noted that Eqs. (3) and (4) give the same maximum energy transferred to the liberated electrons.

Once the numbers of liberated δ -electrons are found and their energies are assigned, the radial dose can be estimated. It must be remembered that the secondary electrons are assumed to originate from each point on the ion trajectory and the local distributed dose at each radius from the ion trajectory (the radial dose), $D(r, R)$ is therefore as follows:

$$D(r, R) = \frac{-CZ^{*2}}{\rho 2\pi r c_1 \beta^2} \int_{R_{min}}^{R_{max}} \frac{e^{X_4 X_3 (X_2 - X_1)} e^{\left(\frac{R-r}{c_1} X_1\right)}}{(e^{X_4} - 1)^2} dR, \quad (5)$$

$$X_1 = c_2 + \frac{c_3}{1 + c_4 (R - r)^{c_5}} \quad (6)$$

$$X_2 = \frac{c_3 c_4 (R - r)^{c_5}}{(1 + c_4 (R - r)^{c_5})^2} \quad (7)$$

$$X_3 = \frac{c_2}{c_1} + \frac{\frac{c_3}{c_4} (1 + c_4 R^{c_5}) - \frac{c_3 c_4 c_5}{c_1} R^{c_5}}{(1 + c_4 R^{c_5})^2} \quad (8)$$

$$X_4 = \frac{R}{c_1} \left(c_2 + \frac{c_3}{1 + c_4 R^{c_5}} \right) \quad (9)$$

where C is a constant and for water (Zhang et al., 1985) equals

$$C = \frac{2\pi N e^4}{mc^2} = 8.5 \frac{keV}{mm} = 1.369 \times 10^{-7} \frac{erg}{cm} = 1.369 \times 10^{-14} \frac{J}{cm}. \quad (10)$$

For calculating the radial dose, the ion parameters of Z^* (the effective charge number of the ion) which was calculated according to Barkas' formula as $Z^* = Z(1 - e^{-1.25\beta Z^{-\frac{2}{3}}})$ and β were incorporated in the calculation as well as the target parameter of $\rho = 1 \frac{gm}{cm^3}$ for water medium.

An algorithm was constructed to calculate the integration in Eq. (5) numerically by using the Mid-Point Method of integration. For a given radial distance r slightly less than R_{min} the above integration was estimated between R_{min} and R_{max} . Since, the two integration limits R_{min} and R_{max} are significant, R_{min} was kept constant at 0.2 nm. As explained in the previous work [38], R_{max} was deduced from Eqs. (3)–(4) and substitute back to Eq. (1). But it was found that calculated radial dose using full δ -ray range does not give accurate dose profile. Therefore, the

objective of the current work is to find and propose a newly effective δ -ray range. This effective δ -ray range was determined by the try and error method. This try and error approach was controlled by the next two Eqs. (11) and (12) or steps.

The total radial dose distributed over the 2π around the ion trajectory at all radial distances was determined by carrying out the following double integration.

$$D = 2\pi \int_{R_{min}}^{R_{max}} \int_{r=0.2}^{r=r_{max} \geq R_{min}} rD(r, R) dr dR. \quad (11)$$

The two integration limits were carried out in such a way that the down limit for the second integration is R_{min} and the upper limit is R_{max} (are the maximum δ -ray range obtained from Eq. (3) or Eq. (4)). For the first integration, the lower limit is taken as $r = 0.2$ nm and the upper limit of this integration is a part from R_{max} and is taken as $r = r_{max} \geq R_{min}$. The total dose is then determined. This total radial dose is related to the linear energy transfer, LET of the ion at a given energy through the following relation:

$$D = 1.602 \times 10^{-9} \times LET \left(\frac{keV}{\mu m} \right) \times fluence \left(\frac{electron}{cm^2} \right) \times \frac{1}{\rho \left(\frac{g}{cm^3} \right)} \quad (12)$$

The LET of the ion is then determined. By comparing the obtained LET with the tabulated- LET (LET_{SRIM}) of the ions from SRIM program [39], one can judge how accurate the deduced integration limits are. Otherwise, the process carried out for Eqs. (11) and (12) will be repeated many times and every time the suggested integration limit of $r = r_{max}$ will slightly adjusted (increased or decreased) until LET calculated by the present approach comes equal or closer to LET_{SRIM} data. Therefore, the looking for integration limits is determined. This step is used to determine the ion's LET as well as the newly suggested $r = r_{max}$ which is considered as the ion's penumbra radius.

For radial dose profile determination, the new integration limits of Eq. (5) are then assigned in such a way that $R_{min} = 0.2$ nm and R_{max} will become the newly suggested effective δ -ray range $r = r_{max}$. Then r increased step-by-step smoothly with 1 nm long and the corresponding R_{min} was adjusted accordingly. The above radial dose integration was carried out at every radial distance, r i.e. at $r = 1$ nm then at $r = 2$ nm and so on with 1 nm step till the end of the effective δ -ray range reached (an extensive work). The integration limits were determined for the 85 ions under study before calculating the radial dose distribution for any ion. A FORTRAN algorithm was constructed to facilitate the above integrations numerically by using the Mid-Point Method of integration.

3. Penumbra radius

Ion track formation and interaction of swift heavy ions with the target molecules is a complicated process and passes through physical, physicochemical and chemical stages [42]. All these processes are time dependent where physical stage has time, $t < 10^{-15}$ s, physicochemical stage has $10^{-12} \leq t \leq 10^{-9}$ s and chemical stage occurs in time $t > 10^{-8}$ s. For more details about these processes, please refer to the following recent article [42] and references there in. However, spatial distribution of ionization energy generated by an energetic swift heavy ions can simply described as highly dense ionization core surrounding by a region called penumbra [16,23]. Penumbra is the region where secondary electrons released by the primary particle deposit their energy. It shows how far the secondary electrons reach its effect around the ion trajectory and the damage it may cause for biological as well as electronic systems. The outer border (radius) of penumbra [22] is given by

$$r_{max} = 0.768E - 1.925\sqrt{E} + 1.257[\mu m] \quad (13)$$

Another penumbra radius formula [21] is given as

$$r_{max} = 6.16 \times 10^{-2} E^{1.7} [\mu m] \quad (14)$$

where, E is the kinetic energy of the projectile [MeV/nucleon]. The penumbra radii obtained by the present work ($r = r_{max}$) are compared with the data obtained by those equations.

4. Experimental and Geant-4 DNA data

Radial dose distribution was measured experimentally by tissue-equivalent gas chamber. A large ionization chamber consists of an aluminum cylinder and a copper central wire serves as an ion collector. A probe is attached to rotate into various radial positions from the center to almost the chamber wall. A faraday cup was used to determine the beam intensity and ionization current was measured using a vibrating read electrometer where ionization was converted to energy deposited. The experimental data [43–46] for 1 MeV proton, 3 MeV alpha, 24 MeV Carbon and 41.1 MeV Oxygen were collected and used for comparison with present calculations.

Geant4-DNA toolkit data for p, α -particles, C and O ions of different energies were obtained [10] and compared with current calculations. In Monte Carlo simulation, these ions were shot into liquid water and energy depositions around the particle track were scored in concentric cylindrical shells around the incident particle track. Each shell has a thickness of 1 nm. The validation of the current approach was achieved through comparing the present calculations with the available experimental as well as Geant-4 DNA data. Table 2 compile the fifteen groups of 85 ions of different E/n where the experimental data are defined as (*) and Monte Carlo Data are defined as (**).

5. Results

5.1. δ -ray effective range, r_{max}

Integration limits play an important role in radial dose estimation especially the upper limit as explained in the Methodology section. The effective ranges for 85 ions representing 15 groups were determined. It is worth knowing that, the present work estimated the radial dose from R_{min} equals 0.2 nm corresponding to δ -ray of energy ≈ 3 eV and the maximum δ -ray range, R_{max} equals 44500 nm corresponding to ≈ 53.5 keV. Therefore, the upper energy limit for the incident ions was found in the present study at 24 MeV/n. The ratio between the effective δ -ray range, r_{max} (Eqs. (11) and (12)) and maximum δ -ray range of the liberated δ -electrons, R_{max} (Eqs. (1) and (3)) ($R_{ratio} = \frac{r_{max}}{R_{max}}$) were determined for the different energy groups and are illustrated as a function of the group velocity, β in Fig. 1.

One can observe that there are three different R_{ratio} regions, one before 1 MeV/n ($\beta = 0.046$), one between 1 and 15 MeV/n and one from 15 MeV/n and above. At energy equal or less than 1 MeV/n, the

Table 2

Radial dose distribution for fifteen groups of 85 ions of different E/n were studied, some of them has experimental data (*) and others has Geant-4 Monte Carlo Data (**) for the sake of comparison.

E/n (MeV/n)	β	Studied bands of ions
0.25	0.02316	p, α^{***} , C, O, I*
0.50	0.03275	p, d*, α , C, O, Br*, I*
0.75	0.04010	p, α^{***} , C, O, Fe
1.00	0.046	p *** , d*, α , C, O, Ne, Fe
2.00	0.06542	p*, α , C *** , O, Ne, Fe
2.40	0.072	p, α , C, O*, Ne, Fe
2.57	0.074	p, α , C, O *** , Ne, Fe
3.00	0.080	p*, α , C, O, Ne, Fe
4.00	0.092376	p, α , C, O, Ne, Fe
6.00	0.112956	p, α , C, O, Ne, Fe
8.10	0.131023	p, α , C, O, Ne*, Fe
10.0	0.145361	p ** , α^{**} , C ** , O ** , Fe **
15.0	0.1773	p, α , O, Fe
20.0	0.204	p, α , O, Fe
24.0	0.222721	p, α , C, O, Ne, Fe

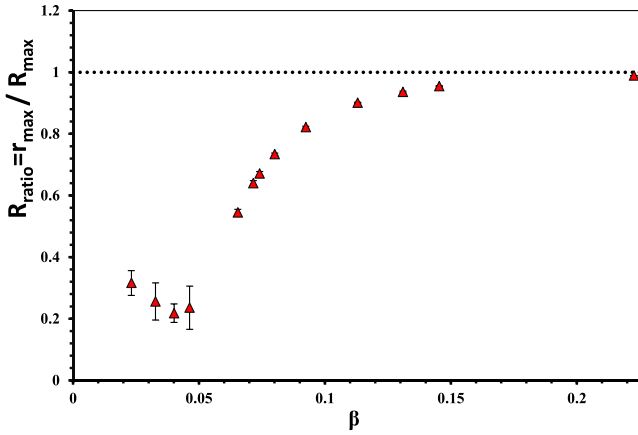


Fig. 1. The variation in range ratio, $R_{ratio} = \frac{r_{max}}{R_{max}}$ as a function of the ion's relative velocity, β for the different energy groups.

effective radii for those ions are about constant and their average is $\approx \frac{1}{3}R_{max}$. For ions of energy greater than 1 MeV, the effective radii increase by increasing the ion energy. At energy equal and greater than 15 MeV/n, R_{ratio} becomes closer to 1 and the effective radius, r_{max} will almost equal full δ -ray range, R_{max} .

The validity of the current calculations as well as accurate integration limits determination are guaranteed by the fact that the radial dose integration over all radial distances around the ion path must yield the ion's electronic LET (Eqs. (11) and (12)). Fig. 2 shows the ratio between the calculated linear energy transfers to the tabulated electronic LET ones, $LET_{ratio} = \frac{LET}{LET_{SRIM}}$ for the fifteen groups under study. Please note that the vertical dashed line at 1 MeV/n and the horizontal dashed line at 1 of y-axis are to guide the eye only. One can observe that, there are two different trends for the calculated linear energy transfer. A trend at ion's energy equal or less than 1 MeV/n in which the calculated LET is less than the electronic LET_{SRIM} by 30%. It seems that for low energy ions not only the electronic processes are affecting the deposited energy, instead many other processes may be taken place, see discussions. For ions of energy greater than 1 MeV/n, the LET_{ratio} is ≈ 1 and hence the calculated LET s by the present approach are about equal the electronic LET_{SRIM} . Therefore, the present approach gives good expectation for the ion's LET in this region. One has to say that, this LET_{ratio} study is the most significant step in such studies.

5.2. Radial dose

Once the desired effective δ -ray range, r_{max} is determined, the

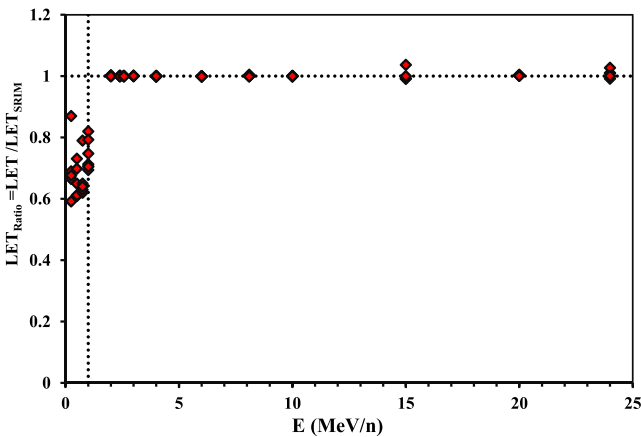


Fig. 2. The relative LET ratio, $LET_{ratio} = \frac{LET}{LET_{SRIM}}$ of the different ions and groups as a function of the ion's kinetic energy, E (MeV/n).

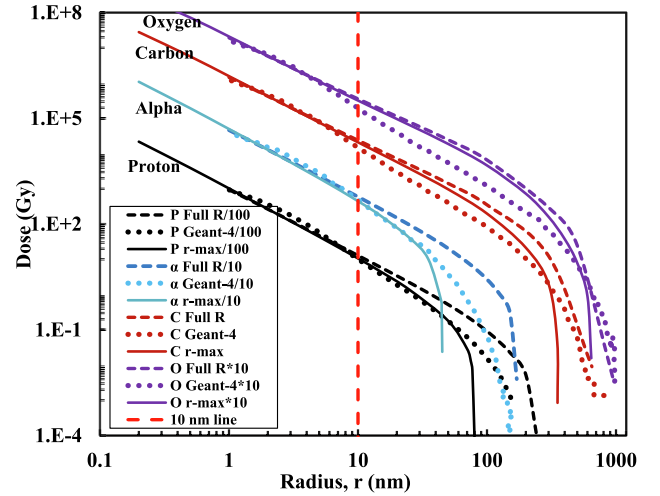


Fig. 3. Calculated radial dose for 1 MeV p divided by 100, 0.75 MeV/n α divided by 10, 2 MeV/n C and 2.569 MeV/n O multiplied by 10 using the full range, R_{max} and the effective δ -ray range, r_{max} compared to Geant-4 DNA simulation data at the different radial distances, r .

integration limits of Eq. (5) are assigned and the radial dose can be calculated at every position, r from the ion trajectory. Implementing the new integration limits on radial dose estimations has improved the dose profile. Fig. 3 shows the improvement in radial dose profile of the some ions by using current approach (using r_{max}) in comparison with profiles of using the full δ -ray range as well as Geant-4 DNA Monte Carlo data. To distinguish between data, data for 1 MeV p (Full, effective and Geant-4) are divided by 100, data for 0.75 MeV/n alpha are divided by 10, data for 2 MeV/n C are kept the same while data for 2.569 O are multiplied by 10. Good improvements were obtained especially for proton and alpha where exact matching between current radial dose and Geant-4 DNA radial dose data at almost all radial distances was observed. Reasonable improvements were found for C and O profile and C data is better because it becomes closer to Geant-4 data but no exact matching at larger radii were observed.

Radial dose profile using r_{max} for a big data set of 85 ions divided into 15 groups were studied. The radial dose profile for some of these groups will be shown and the other will not for similarity only. The group that has experimental and/or Geant-4 DNA data will be presented in this context. The calculated radial dose profiles for p, d, α , C, O, I ions at energy equal 0.5, 1 and 2 MeV/n as a function of the radial distance, r are presented in Fig. 4(a–c). The experimental data for d as well as I are dropped in Fig. 4-a. Experimental data for p, d and p Geant-4 data are given in Fig. 4-b. While experimental data for p, C and C Geant-4 are given in Fig. 4-c for the sake of comparison. Excellent dose profile and good matching between present work, experimental as well as Monte Carlo data were observed. Similarly, the dose profile for p, α , C, O, Ne and Fe as a function of radial distance, r at energies 2.4, 3 and 8.1 MeV/n are compiled in Fig. 5(a–c). Experimental data for O, P and Ne are added to the figures.

$D(r, R) \times r^2$ as a function of radial distance, r discerns the differences between the different dose profile obtained by current approach, experimental and Geant-4 data. Fig. 6(a–c) shows that calculated dose profile, experimental and Geant-4 data are following the $\frac{1}{r^2}$ behavior in agreement with previous studies [11,21,47–49]. Therefore, the spatial distribution of the energy deposited is not homogeneous, but each individual ion deposits its energy with an approximately $\frac{1}{r^2}$ dependence on the radial distance from the trajectory.

Radial dose profile for ions p, α , C, O and Fe at energy 10 MeV/n in comparison with Geant-4 DNA MC simulation data are presented in Fig. 7-a. The two dotted vertical lines at $r = 1, 4$ nm are to guide the eyes only. To improve the comparison between the two model, the

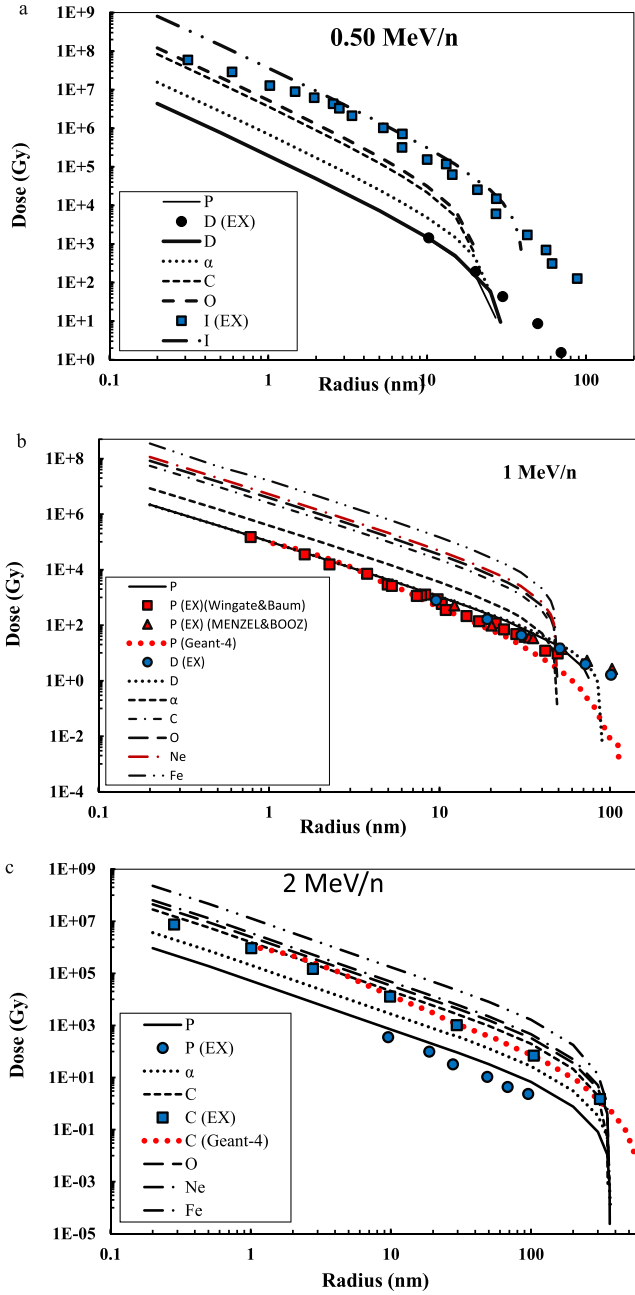


Fig. 4. (a-c) a- The calculated radial dose distributions for p, d, α , C, O, I and I (EX) for the group energy 0.5 MeV/n. b- The calculated radial dose distributions for p, p(EX), p(Geant-4), d, d(EX), α , C, O, Ne and Fe for the group energy 1 MeV/n. c- The calculated radial dose distributions for p, p(EX), α , C, C(EX), C(Geant-4), O, Ne and Fe for the group energy 2 MeV/n, respectively.

values of $D(r, R) \times r^2$ for these ions as a function of the radial distance, r are given in Fig. 7-b. Monte Carlo data show better $\frac{1}{r^2}$ behavior than the present approach at such high energy. One can observe that in the region of $r > 1$ nm, the radial dose is slightly underestimated while in the region between $r = 1-4$ nm the present calculations and Monte Carlo simulation are almost similar. After 5 nm of radial distance, the deviation between the two data increases. At radial distance equal 100 nm the current radial dose gives larger dose than Geant-4 data.

5.3. Radial dose applications

The energy deposition radially around the ion trajectory and the corresponding distribution of dose around the ions can be considered as

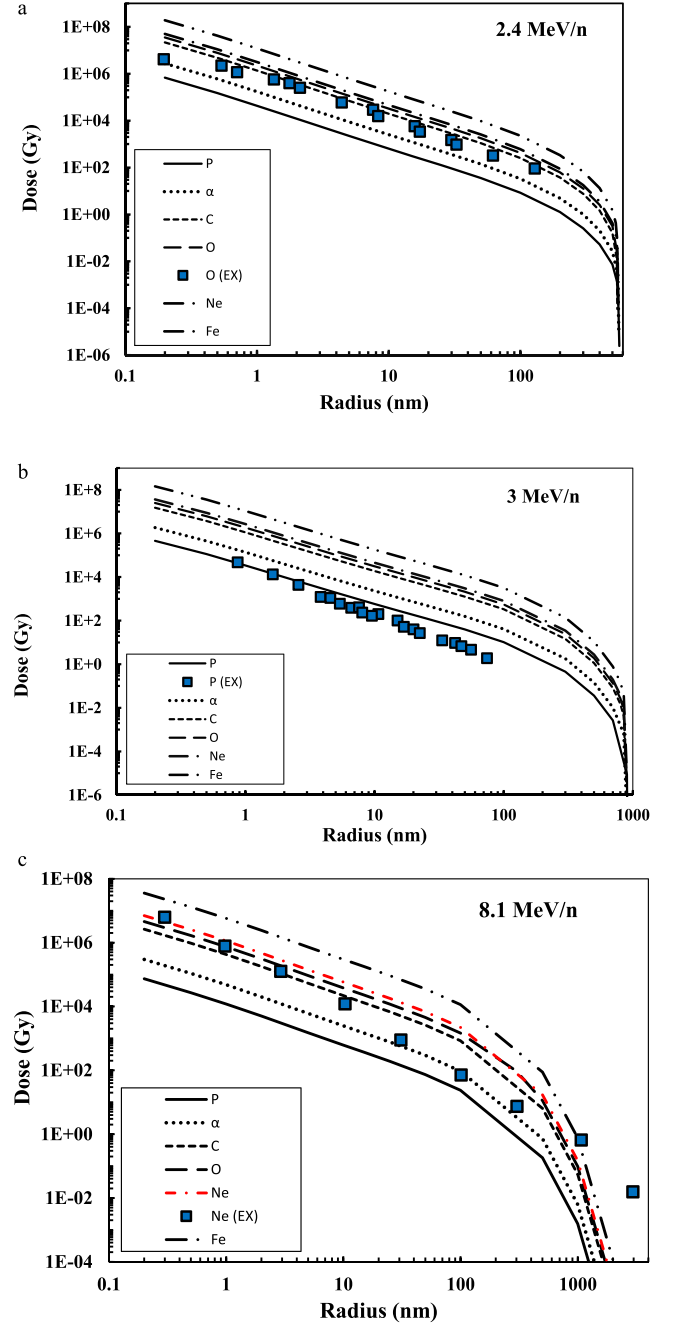


Fig. 5. (a-c) a- The calculated radial dose distributions for p, α , C, O, O(EX), Ne and Fe for the group energy 2.4 MeV/n. b- The calculated radial dose distributions for p, p(EX), α , C, O, Ne and Fe for the group energy 3 MeV/n. c- The calculated radial dose distributions for p, α , C, O, Ne, Ne(EX) and Fe for the group energy 8.1 MeV/n, respectively.

a fundamental quantity and can be used to deduce some important information about the incident ion in the medium. From radial dose one can determine the stopping power and/or the linear energy transfer (LET) of the incident ion as well as determine how far from the ion trajectory the energy of the liberated electrons are effectively delivered (the ion's penumbra radius).

5.3.1. Ion LET

Estimating the total radial dose distributed over the 2π around the ion trajectory at all radial distances was determined by the double integration of Eq. (11). The corresponding ion LET is thus directly

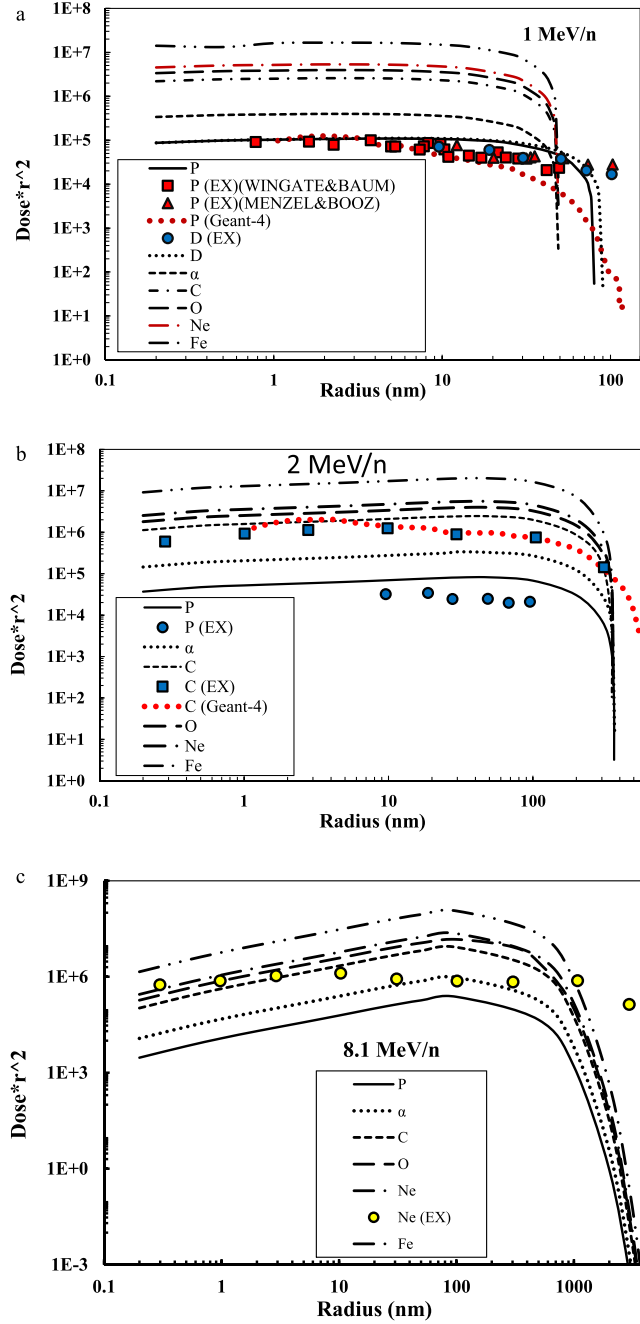


Fig. 6. (a-c) a- The $\text{Dose} \times r^2$ for p, p(EX), p(Geant-4), d, d(EX), α , C, O, Ne and Fe as a function of the radial distance, r for the group energy 1 MeV/n. b- The $\text{Dose} \times r^2$ for p, p(EX), α , C, C(EX), C(Geant-4), O, Ne and Fe as a function of the radial distance, r for the group energy 2 MeV/n. c- The $\text{Dose} \times r^2$ for p, α , C, O, Ne, Ne(EX) and Fe as a function of the radial distance, r for the group energy 8.1 MeV/n, respectively.

deduced from Eq. (12). The LET (KeV/ μm) of the six ions under investigation (p, α , O, C, Ne and Fe) at the energy range from 0.25 to 24 MeV/n in comparison with SRIM as well as Benton-Henke range-energy Table [50] are shown in Fig. 8 (a-b) using logarithmic scale. For better illustration, two of these ions (O and Fe) are showing as example in Fig. 9(a, b) using linear scale. Investigating these figures one can observe that LET as a function of the ion's energy can be split into two separate parts; before the Bragg peak (Bethe-Bloch) and after the Bragg peak (Thin Down) [51,52]. In the Bethe-Bloch region, the model is able to determine the ion's LET exactly like SRIM and Benton-Henke data but in the Thin Down region the accuracy of determine ion's LET is not the

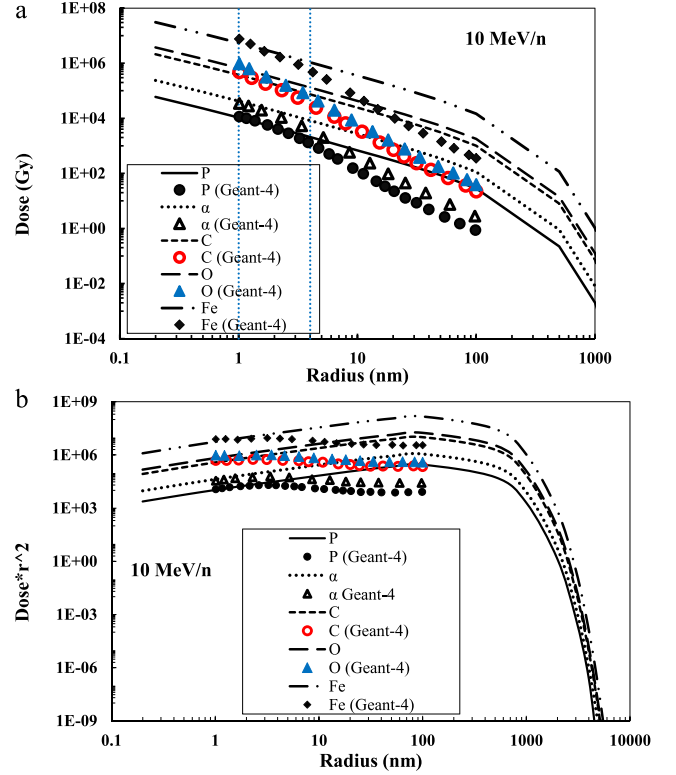


Fig. 7. (a,b) a- Radial dose distribution p, α , C, O, and Fe compared to the corresponding Geant-4 DNA data. b- The $\text{Dose} \times r^2$ for p, α , C, O, and Fe compared to the corresponding Geant-4 DNA data for the group energy 10 MeV/n, respectively.

same. Despite the fact that the present work is compared to the electronic stopping power of SRIM data, the discrepancies between the data, as expected, was observed. This discrepancy will be addressed in the discussion chapter.

5.3.2. Penumbra radius

After adjusting the integration limits as was explained, the upper integration limit or the effective δ -ray range, r_{max} can be determined. This r_{max} is considered as the ion's penumbra radius. The average penumbra radius at the studied energy from 0.25 to 24 MeV/n was calculated and presented in Fig. 10. The statistical error in each point is smaller than the circle symbol given in the figure. It was found that ion's penumbra radius; r_{max} depends on the ion's kinetic energy, E (energy/n) through the following polynomial equation

$$r_{max} = 0.068E^2 + 0.2215E - 0.2339 \text{ (}\mu\text{m)}, \quad R^2 = 0.9999 \quad (15)$$

Comparing the obtained penumbra radius with penumbra radii obtained by Kiefer and Straaten [21] (Eq. (13)) and by Chatterjee and Schaefer (Eq. (14)) [22] one found that they all are in the same order of magnitude and the current approach gives larger penumbra radius than the other equations do. The lateral distribution of the charge generated in silicon by 15 MeV oxygen ion was determined experimentally [15] and it was found equal 300–400 nm in agreement with the present model as shown in Fig. 10.

6. Discussion

Effective δ -ray range, r_{max} increases by increasing the ion's energy and r_{max} is always less than R_{max} (Fig. 1). However, ions of $E \leq 1$ MeV/n are having $r_{max} \approx \frac{1}{3}R_{max}$. The fewer energy ions are producing less δ -ray energy. These liberated electrons of less energy may suffer larger scattering and resistance with surrounding electron matrix producing

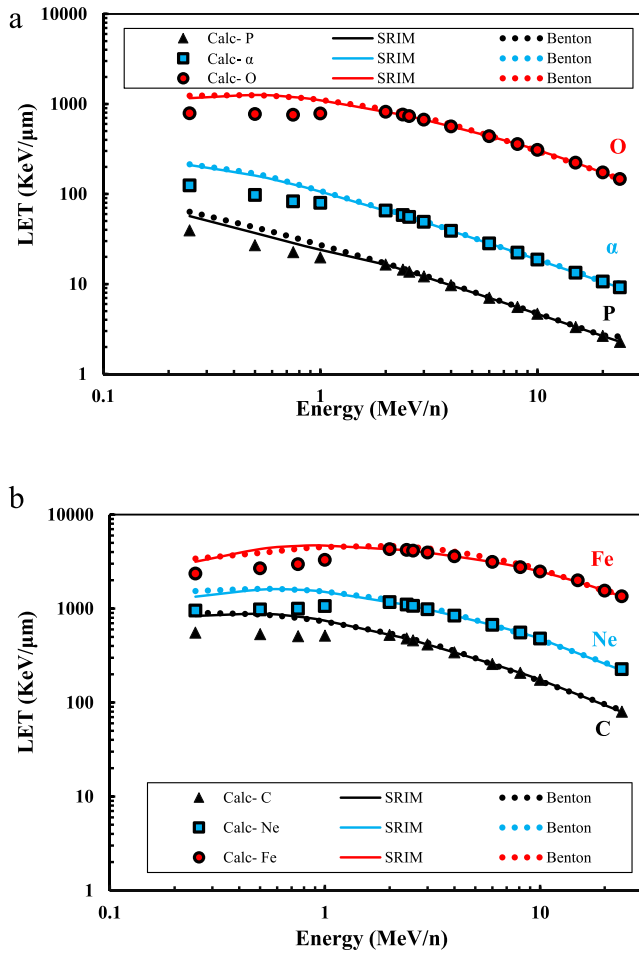


Fig. 8. (a,b) LET calculated by the present approach for (p, α and O) and (C, Ne and Fe), respectively at the different ion's energy compared with the corresponding ones obtained by Benton-Henke energy table and SRIM code.

less effective δ -ray range. This may explain the smaller effective δ -ray range in this region. In general, the reduction in δ -ray range could be accepted due to the stochastic nature of δ -electron motion in the surrounding medium [36].

The predicted radial dose distribution has been improved using the new proposed δ -ray range, r_{max} . Modified radial dose profile by taking into account the effective δ -ray range has improved the obtained radial dose for 85 ions of different energies. The results of the present approach are compared against experimental and Geant-4 DNA data. Global agreement was found between the different data at all radial distances as illustrated in Figs. (3)–(7). At the ion's core and at smaller radii of less than or equal 5 nm good matching and coincident between the present calculations, experimental and Geant-4 DNA data was obtained. In the core region a huge number of ionized electrons interacting with each other and with the surrounding electrons where most of the ionizations occur in a small cylinder close to the ion path. In addition to this, slow secondary electrons may be trapped by track potential which formed by the electric field near the incident ion path [53,54] creating high doses. The present approach succeeded to predict such high dose in this region.

But at high energy ions > 8 MeV/n and at higher radial distance > 50 nm, the present approach overestimates the radial dose and the $\frac{1}{r^2}$ behavior is not exactly fulfill. The interaction of high-energy ions is characterized by almost pure electronic excitation and ionization of the target atoms. The primary ionization and excitation processes and the following electron cascade have stochastic nature. Therefore, the assumption that all δ -electrons are liberated normal (with $\approx 90^\circ$) to the

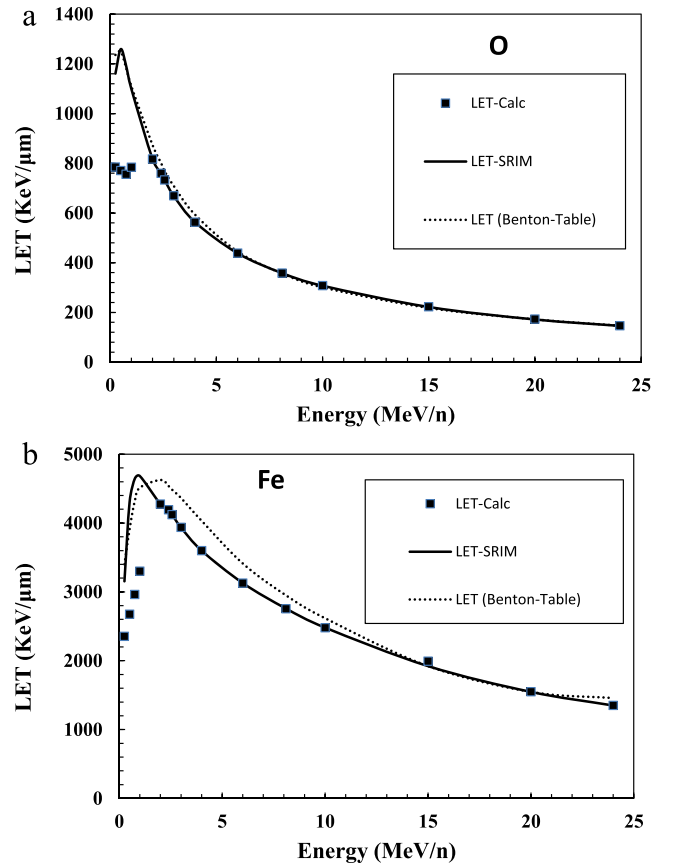


Fig. 9. (a,b) Bragg's peak for O and Fe ions where LET as a function of Energy (MeV/n) are given, respectively. The model exactly fits the data in the Bethe-Bloch region however the accuracy in the Thin Down region is not the same.

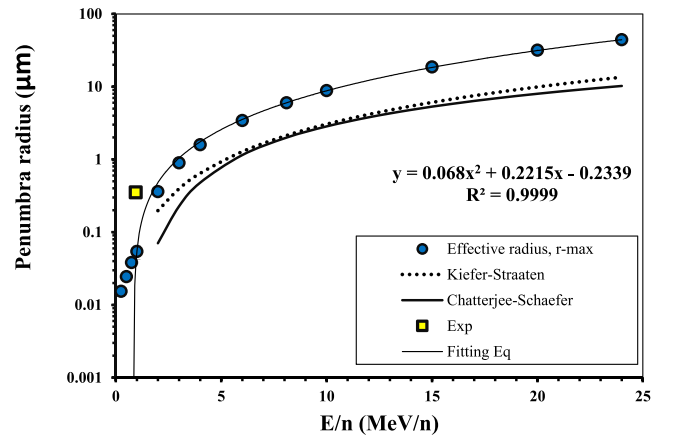


Fig. 10. The predicted effective radius, penumbra radius, in water as a function of the ion's energy is presented for 85 ions under investigation. The suggested penumbra formula is also compared to Kiefer-Straaten and Chatterjee-Shaefer formula. The Experimental point is the penumbra radius for oxygen in silicon at energy 0.94 MeV/n and is shown for guidance due to the lack in experimental data.

ion path may not be an appropriate assumption and needs to be revised and angular percentage of δ -electrons must be considered. At large distances, the dose is mainly due to the slowing down of energetic electrons and the model is not able to recognize such slowing rate. The reason for this is not clear but may be the δ -ray range, number of collisions as well as the rate of electron energy loss are not easy to be exactly determined.

The deduced linear energy transfer *LET* of the studied 85 ions by the current approach are excellent in the Bethe-Bloch region of Bragg peak where the obtained *LET* of the ions are almost equal *SRIM* and Benton-Henke data, see Figs. 8 and 9. But in the Thin Down region, the calculated *LET* is less than the expected (*SRIM*) ones by 30% as stated before. In Thin Down region where the ion energy becomes very small, the ion's characteristic (effective charge, range, straggling, *LET* etc.) as well as the ion's behavior are dramatically changing in such a way that it is difficult to be predicted. It is known that in Thin Down region, nuclear process have higher cross-sections at lower ion energies (Rutherford scattering), that low-energy electrons are more massively produced. In addition, current paradigm does not take into account electron excitation energy, the elastic nuclear collision between the incident ion with surrounding medium and the energy of the produced free radicals [35]. It is worth to remember that the scope of the present work is δ -ray interaction with the surrounding target atoms and hence, these aspects are not taken into account.

Adjusting the integration limits enable to deduced Penumbra radius of the studied ions, Fig. 10. In this figure, the predicted effective radius, penumbra radius, in water as a function of the ion's energy is presented for 85 ions under investigation. The suggested penumbra formula is also compared to Kiefer-Straaten and Chatterjee-Shaefer formula. The Experimental point is the penumbra radius for oxygen in silicon at energy 0.94 MeV/n and is shown for guidance due to the lack in experimental data. Fortunately, the experimental data for the lateral distribution of charge due to oxygen in silicon [15] is closer to the suggested formula in this work. Therefore, one may claim that the present approach predicts the penumbra radius more accurately than the other models. Experimental data for lateral distribution of charge for the different ions of different energy are needed for further examination of the present formula. As expected, as the ions energy increases, the liberated δ -electron energies increases (Eqs. (3) and (4)) leading to an increased penumbra radius. Penumbra radii obtained by the present work and radii deduced from Eq. (13) and Eq. (14) are in the same order of magnitude (μm). However, the present radii are 3 times larger than penumbra radii deduced by other formula. The present calculations are limited to the scope of delta-ray action; therefore, penumbra radius is defined in this study as a static concept. The physical phase of ionizing radiation energy deposition which include thermalization of delta-rays is limited in time to about 10^{-13} s. At this point the track is still not in chemical equilibrium, and reactive species are heterogeneously distributed. However, in water, reactive oxygen species (ROS) created due to incident ions diffuse in the bulk water and therefore, the concept of radiation track in water medium is a dynamical concept as it is modeled in RITRACKS [55] for example. In polymer, effective track (penumbra) radius due to incident ion may be described as the radial extension of reactive species formed rather than the maximum distance at which δ -rays escape out of ion's trajectory. Free radicals created can diffuse and enhance the size of the radial zone affected by quasi-instantaneous effect of delta-rays [42]. Therefore, penumbra radius may have dynamic dimensions rather than static ones.

7. Conclusion

Radial dose distributions of different ions of different energies in water have been improved by applying the effective δ -ray range, r_{max} . Effective δ -ray range was deduced by adjusting the radial dose integration limits. Detail steps were given and the final radial dose integration was solved numerically using the Mid-Point Method. Radial dose for p, d, alpha, C, O, I, Ne and Fe at energies 0.25, 0.50, 0.75, 1, 2, 2.4, 2.569, 3, 4, 6, 8.1, 10 and 24 MeV/n were determined. The model was able to reproduce successfully the radial dose for such ions compared to experimental as well as the Geant-4 DNA. The variation in radial dose multiplied by r^2 ($D(r) \times r^2$) as a function of the radial distance, r was estimated and the variation between the different calculations are clearly demonstrated. All ions are following the inverse

square relation $\frac{1}{r^2}$ where all ions showing straight line in agreement with experimental as well as Monte Carlo estimation.

The model is able to predict the ion's *LET* exactly compared to *SRIM* and Benton-Henke data in the *Bethe-Bloch* region of Bragg's peak where the electronic energy transfer is dominant. However, in the *Thin down* region it found some difficulty to determine it exactly due to the fact that the current paradigm is based on δ -ray action and at this low energy region it cannot describe properly other actions than those of delta rays (nuclear scattering, electron excitation, low energy electrons interactions and energy of free radicals).

Penumbra radii in water for the 85 ions understudy were determined. New formula for penumbra radius is suggested in the present work and tested experimentally and compared to other formula. However, the newly suggested formula needs further examination by new experimental data.

The present model is offering a simple alternative to time consuming Monte Carlo simulations and can be conveniently used in hadron therapy dosimetry. It can be used for estimating the radial dose of the ion; it's *LET* as well as lateral distribution of charge around the ion's path (its penumbra radius).

Despite the success obtained by adding the effective δ -ray range, the present approach still suffering some limitation especially at the larger radii where the $\frac{1}{r^2}$ behavior is not exactly fulfilled. Radial dose distribution is of particular significance for improving the understanding of radiobiology experiments with heavy ions, radiation protection issues and radiation risks associated for heavy ion exposure. Therefore, further refinement still required where angular distribution of δ -ray must be considered and incorporated into calculations.

Latent track structure in PADC and other solid state nuclear track detectors will be examined using the current approach. The assumption that ions of the same *LET* can have completely different radial dose and different penumbra radii must be tested in the future using current approach.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgments

The authors gratefully acknowledge and appreciate the unknown referee who had helped to improve the quality of the work by their valuable comments and Nermeen Ahmed Fadl for her effort in collecting and calculating the data.

Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.nimb.2019.10.031>.

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