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THE IONIZATION POTENTIAL OF DNA AND LIQUID WATER

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ABSTRACT

There are multiple applications in medical physics and space radiation health, such as hadron therapy for cancer treatment ofthe irradiation of biological systems by energetic ion beams. Therefore, for a better control and understanding of the effects of radiation damage in living tissues, it is necessary to advance an accurate description of the energy loss from the ion beam to the target. In the present work PPA of the dielectric formalism has been used to calculate the probability for an energetic proton to produce electronic excitations in DNA as well as the average energy of the target electronic excitations as a function of the projectile energy. Our results show that the protons with energy between 0.05 MeV to 2.5 MeV are very efficient in producing secondary electrons in DNA, which are able to produce strand breaks and could be very effective for the biological damage of malignant cells.

The ionization potential (the so-called I-value) is the most important material quantity in the calculation of the electronic stopping power of energetic ions (>1 MeV u−1) through Bethe's formula and represents its main source of uncertainty at high energies, I_{DNA} found to be ≅ 80.01 eV and $I_{H2O} \cong 78.17eV$. Our present results are compared with previous work and show good agreement.

Keywords: Energy-loss function, Dielectric formalism, DNA target, Liquid water, Energy loss, Proton beam, Avarge energy, Ionization potential.

1. INTRODUCTION

The interaction of heavy ions with matter and the transference of their energy to matter are key for understanding the biological effects of radiation on living tissue. Ionizing radiation defines those radiations that primarily interact with matter by charged particles[1].

Using energetic ion beams for radiation therapy has become a promising technique because high doses can be deposited locally at tumor sites, reducing the damage to the surrounding critical organs.Hadron therapy exploits the enhanced energy deposition taking place at the end of the range of energetic ion beams (the so-called Bragg peak) [2].

Most calculations of the energy loss of ions in solids are based on the Bethe theory [3], where the most important factor is the mean excitation energy of the target. At high projectile energies (in the Bethe region), the stopping powers predicted by the Bethe theory are within a few percent of the experimental data [4].

An effort to study the interaction of energetic ion beams with liquid water at intermediate energies has been carried out recently, since water represents over 80% of the content of the cells

of soft tissues [5, 6]. A little work has been done concerning the electron energy loss of energetic ions in DNA targets.

Research on the effects of radiation on DNA, the most important biological material, is very active, because determining the relationship between the energy deposited by fast particles in the target and the damage they cause is important to radiation biophysics [7, 8]. DNA damage can be produced by direct ionization and excitation of DNA electrons [9] or by indirect chemical reactions of water radiolysis products with DNA [10]. Even electrons with subionizing energies can cause lethal lesions in DNA [11, 12].

Therefore, a detailed study of the energy loss of ions in biological targets (such as DNA or liquid water) is desirable to improve our understanding and modeling capabilities of the action of radiation in ion-beam cancer therapy [8, 13, 14]

In this paper we calculate the ionization potential in DNA and the probability per unit path length P (T, E) and the mean energy $< E(T) >$ of the electronic excitations produced in DNA for a wide range of projectile energy (0.05 Mev/nucleon to 2.5 Mev/nucleon) by using program of Fortran 60.

2. THEORETICALBACKGROUND

2.1.Description of the Target ELF

The key parameter to obtain reliable results for the energy losses is the energy-loss function (ELF) of the material, Im[-1/ϵ(k,ω)], since it contains all the information about the electron excitation spectrum of the target. Thus it is essential to use a good description of the target ELF for the whole k-ω plane (that is, the Bethe surface) [2].

The well-known Lindhard dielectric function is applicable to only a limited number of so-called nearly-free-electron materials, like aluminum. For other targets, a commonly used method for obtaining the ELF is to employ the experimental energy-loss function at k = 0 (optical limit) and extend it to k > 0 by introducing suitable dispersion relationship schemes [15].

Experimental information about the ELF at k = 0 can be obtained for a number of materials, including liquid water and DNA, from the measurements of optical magnitudes[16].However, experimental information about the ELF at k≠ 0 is limited. For this reason, it is necessary to model the evolution of the optical ELF with finite k in order to calculate magnitudes such as P and $\langle E \rangle$

The Plasmon Pole Approximation (PPA) is applicable for a projectile of high velocity compared to the Fermi velocity $\vec{v} >> \vec{v}_F$, to give a good approximation to the Lindhard dielectric function. It accounts for the collective electron-gas behavior at small $\vec{k}$ and the free-particle behavior at large $\vec{k}$, are included [17].

$$\in(k,\omega) = 1 + \frac{\omega_p^2}{\omega_g^2 + \beta^2\kappa^2 + \frac{\kappa^4}{4} - \omega(\omega + i\gamma)} \qquad (1)$$

The constant $\beta = (3/5)^{1/2}\vec{v}_F$ is the propagation of density disturbances in an electron gas, $\vec{v}_F$ being the Fermi velocity of the electrons in the medium [18, 19].

At high velocities $v > v_F$one can use the Plasmon Pole Approximation ***(PPA)*** ***[20]*** for the dielectric function in the limit of no damping process(i.e., $\gamma \to 0$) [21].

$$\mathrm{Im}\left[\frac{-1}{\epsilon(k,\omega)}\right] = \frac{\pi\omega_p^2}{2A}\delta(\omega - A) \qquad (2)$$

With $A^2 = \Omega_p^2 + \beta^2 k^2 + k^4/4$

At low velocities $v < v_F$, the dielectric function is described as [21].

$$\mathrm{Im}\left[\frac{-1}{\epsilon(k,\omega)}\right] \simeq \begin{cases} \frac{2k\omega}{(k^2+k_D^2)^2} & \text{for } k \le 2k_F \\ 0 \quad \text{for } k > 2k_F \,, \end{cases} \tag{3}$$

with a screening constant,

$$k_D^2 = \frac{4K_F}{\pi}$$

2.2.The Probability and Average Energy of Electronic Excitation

When a swift projectile with mass M1, atomic number Z1, kinetic energy T and charge q moves inside a solid, it induces electronic excitations in the material, losing energy in the process. In the energy range (from 0.05 Mev / nucleon to 2.5 Mev/ nucleon). These electronic excitations can correspond to excitations or ionizations of individual electrons or even excitations of collective modes in the target electron gas.

The dielectric formalism [22] provides a way of studying the response of the electronic system of the target to the perturbation represented by the projectile. Within this framework the probability per unit path length P_q(T, E) that a projectile with charge state q and energy T produces in the target an excitation of energy E =ħω irrespective of its momentum, ħk, is given by:

$$P_q(T,E) = \frac{M_1 e^2}{\pi \hbar^2 T} \int_{k_{\min}}^{\infty} \frac{\mathrm{d}k}{k} \rho_q^2(k) \mathrm{Im}\left[\frac{-1}{\varepsilon(k,\omega)}\right], \tag{4}$$

where $k_{min} = \omega/\sqrt{2T}/M1$, e is the absolute value of the electron charge and ρ_q (k) is the Fourier transform of the projectile charge density. Hence, the mean energy lost by the projectile per unit path length (the so called stopping power or stopping force) can be calculated integrating over all possible energy transfer E

$$\left.\frac{\langle \Delta T \rangle}{\Delta x}\right|_q = \int_0^{\infty} \mathrm{d}E E P_q(T,E). \tag{5}$$

The mean energy of the electronic excitations <E_q (T)> induced by the projectile can be written as

$$\langle E_q(T) \rangle = \frac{\int_0^{\infty} \mathrm{d}E E P_q(T,E)}{\int_0^{\infty} \mathrm{d}E P_q(T,E)}. \tag{6}$$

The charge state q of the projectile inside the target can vary through capture and loss processes and depends on its energy T. However, when charge equilibrium is reached, the probability Φ_q (T) of finding the projectile in a charged state remains constant for each incident energy T. Here we obtain the values of Φ_q(T) for hydrogen projectiles in DNA using the parameterization provided by the CasP code [23], which uses Bragg's additivity rule for compound targets. We average over all possible charge states (q = 0 and 1 for H) in order to obtain the energy distribution, P (T, E) and the mean energy, <E (T)> of the electronic excitations produced in the target as

$$P(T,E) = \sum_{q=0}^{1} \emptyset_q(T) P_q(T,E) \tag{7}$$

$$\langle E_q(T) \rangle = \frac{\int_0^{\infty} dE E \sum_{q=0}^{1} \emptyset_q(T) P_q(T,E)}{\int_0^{\infty} dE \sum_{q=0}^{1} \emptyset_q(T) P_q(T,E)} \tag{8}$$

Where the calculation of probability and mean energy requires the description of the projectile charge density through ρ q (k), and of the target excitation spectrum by means of its energy-loss function (ELF), Im[-1/ϵ(k,ω)].

2.3. The Ionization Potential

The mean excitation energy (the so-called *I*-value) is the most important material quantity in the calculation of the electronic stopping power of energetic ions (>1 MeV u $^{-1}$) through Bethe's formula and represents its main source of uncertainty at high energies [24].Basically, the average ionization potential is defined [25] as the average value of the excitation energies over all atomic states ($E\ I$) weighted by their transfer probability to continuum ($f\ i$).

The I-value can be generally determined in three ways: (i) from an analysis of stopping power (and range) measurements, (ii) from optical absorption data and (iii) by ab initio calculations. The last method is not yet applicable to condensed targets (liquids and solids) [6].

In this paper we will discuss the calculation of I (eV) from the analysis of stopping power measurements.

2.3.1. The Born Approximation

The electronic (or collision) stopping power (S_{col}) of a material for a charged projectile represents its mean energy loss per unit path length due to inelastic Coulomb collisions with target electrons and is formally obtained from the differential-in-energy-transfer inelastic cross section as follows [26]:

$$S_{col} = \int_0^{E\,max} E\,\frac{d\sigma}{dE}\,dE \qquad \textbf{(9)}$$

where σ is the macroscopic inelastic cross section (or inverse inelastic mean free path) and E is the energy transfer from the projectile to the target electrons leading to ionization and/or (discrete) excitation events.

For protons the upper limit of integration is $E_{max} \approx 4T$ where $T = mv^2/2$ or $T = (m/M)\tau$ with m the electron rest mass (mc^2= 511 keV), and v, τ and M are the proton velocity, kinetic energy and rest mass (Mc^2= 938 MeV), respectively.Assuming sufficiently fast (but still non-relativistic) projectiles of fixed charge leads to the following expression for the electronic stopping power [27],

$$S_{Born} = \frac{z^2}{\pi a_0\, T}\int_0^{Emax} E\ dE \int_{q_{min}}^{q_{max}} \frac{1}{q}\ Im\left[\frac{-1}{\epsilon(E,q)}\right] dq \qquad (10)$$

The limits of integration over *the* q in equation (10) are $q_{max/min} = \sqrt{2M\,(\sqrt{\tau}} \pm \sqrt{\tau - E})$. It follows from equation (10) that the imaginary part of the inverse dielectric function, Im [$-1/\epsilon(E, q)$], the so called target energy-loss function (ELF). It should be highlighted that in order to calculate the electronic stopping power according to equation (10), the ELF must be known over the whole energy–momentum plane; this is the so-called Bethe surface of the material [28]. There are, a numerical evaluation of the Bethe surface of a liquid or amorphous solid is not feasible, and even for simple molecules [29].

2.3.2. The Bethe Approximation

Bethe showed that equation (10) reduces to the following asymptotic form in powers of $1/T$ [30]:

$$S = AT^{-1}\ ln(T) + BT^{-1} + CT^{-2} + \ldots \qquad (11)$$

By using the f sum rule and a distinction between low- and high-q collisions [31] :

$$f \text{ sum rule: } \frac{2}{\pi E_p^2}\int_0^{\infty} E\ Im\ [-1/\in\ (E,0)]dE = 1\ , \qquad (11a)$$

$$\text{conductivity sum rule : } \frac{2}{\pi E_p^2}\int_0^{\infty} E\ Im\ [\in\ (E,0)]dE = 1, \qquad (11b)$$

$$\text{perfect screening sum rule: } \frac{2}{\pi}\int_0^{\infty} \frac{1}{E}\ Im\ [-1/\in (E,0)dE + Re[1/\in (0,0)] = 1. \qquad (11c)$$

Since the Born approximation is essentially a high-T approximation, equation (11) automatically provides the most important contributions to the stopping power. Specifically, the terms of order T^{-1} will be dominant at high energies, whereas the term of order T^{-2} provides the principal correction at lower energies. The coefficients A, B and C depend on target properties related to the ELF. Importantly, the coefficients A and B of the first-order terms (T^{-1}) are obtained solely from the optical limit of the ELF [6]:

$$A=\frac{E_p^2}{2a_0} \quad (12)$$

$$B= A \ln\left(\frac{4}{I}\right) \quad (13)$$

where *I* is the ionization potential which, for condensed systems, is defined by :

$$\ln(I) = \frac{\int_0^\infty E \ln(E) Im[-1/\in(E,0)dE}{\int_0^\infty E\, Im[-1/\in(E,0)dE} \quad (14)$$

Eqs. (11- 14) leads to uncorrected Bethe stopping power formula,

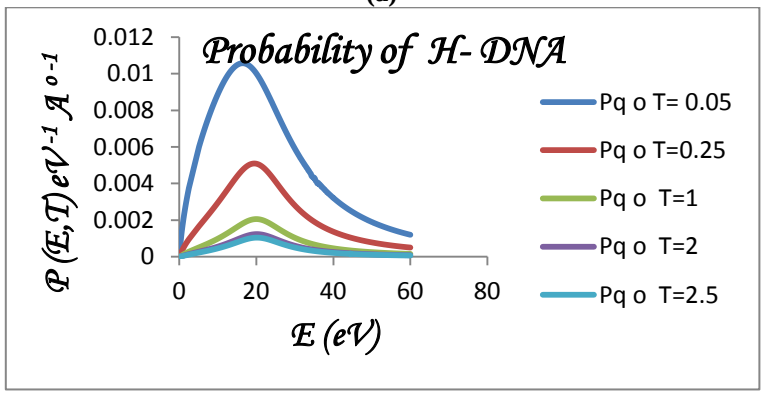
$$S_{Bethe}=\frac{z^2 E_p^2}{2a_0 T}\ln\left(\frac{4T}{I}\right) \quad (15)$$

3. RESULTS AND DISCUSSION

In the present work the calculation of the probability per unit path length, P(T,E) based on Eqs. (4,7), for the proton beam (having incident energies (T = 0.05MeV - 2.5 MeV) of producing an electronic excitation of energy E in DNA and H2O is shown in Figs. (1) & (2) By using PPA has given in Eq. (1). The probability P decreases as the proton energy increases, indicating that a larger number of electronic excitations are produced when lower incident energies T are used. The value of P predicted by PPA agree at large T, and this is very important because this range of T corresponds to the Bragg peak, where most of the projectile energy is deposited. Good agreement achieved with previous work given in [32, 33].

Fig-1. Probability per unit path P that H projectile induces electronic excitation of energy E in DNA for T=(0.05- 2.5) Mev/a.u when modeling the ELF for (a) Λ_o (b) Λ .

(a)



(b)

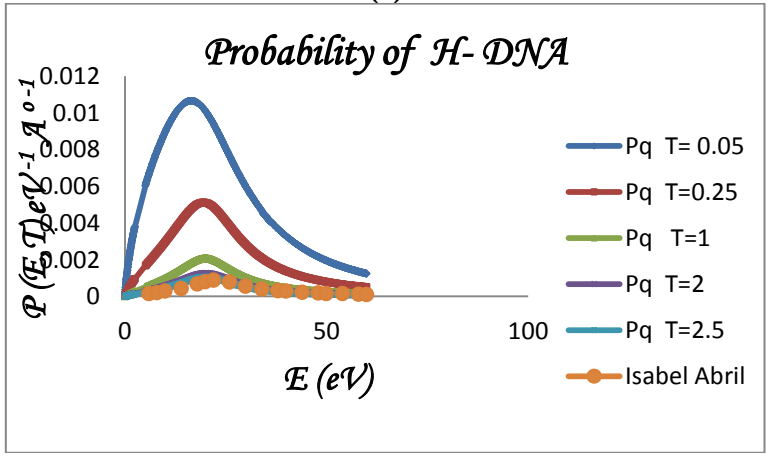
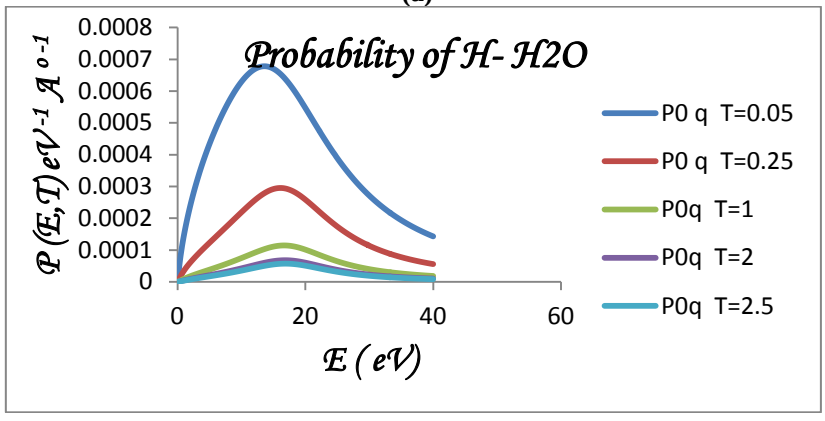
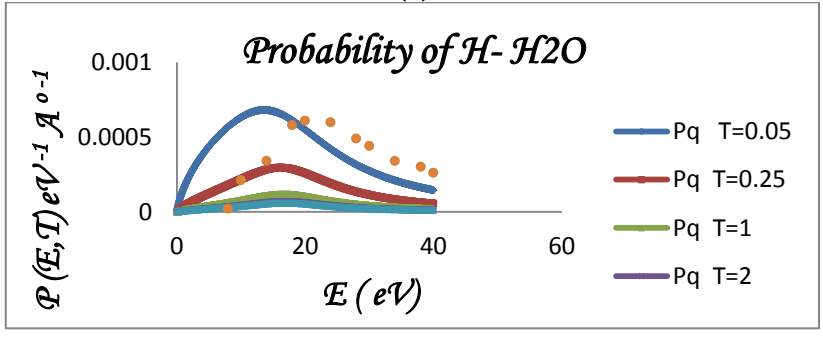


Fig-2. Probability per unit path P that H projectile induces electronic excitation of energy E in Liquid water for T=(0.05- 2.5) Mev/a.u when modeling the ELF for (a) Λ_o (b) Λ .

(a)



(b)



The mean energy $\langle E(T)\rangle$, Eq. (8), of the electronic excitations produced by a proton in DNA and Liquid water is depicted in Figs. (3) & (4). It is seen that $\langle E(T)\rangle$ increases with the proton energy T, being around 9-10 eV at T = 0.001 MeV and around 80 eV for T = 10 MeV. This value agree with previous work [16, 34] taking in the considration screening length Λ_0.

Fig-3. Mean energy $\langle E_q(T)\rangle$of the excitations induced by a hydrogen projectile in DNA as a function of the projectile energy T for screening length Λ_o and Λ .

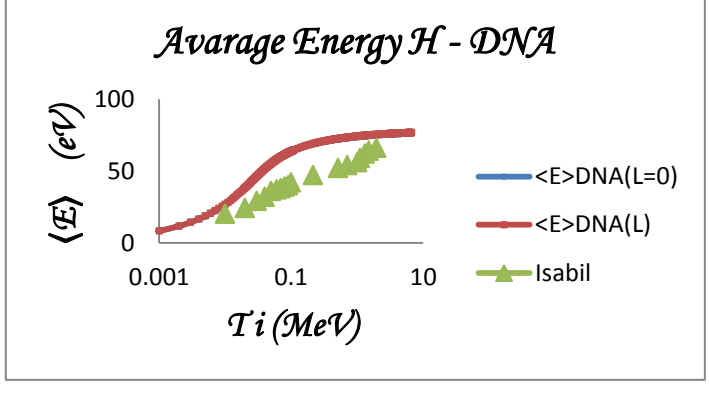
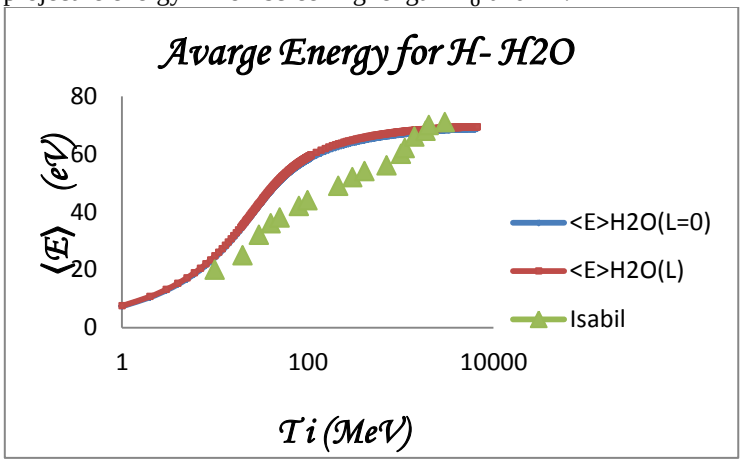


Fig-4. Mean energy $\langle E_q(T)\rangle$of the excitations induced by a hydrogen projectile in H2O as a function of the projectile energy T for screening length Λ_o and Λ .



Accurate values of the ionization potential I(eV) for biological materials such as DNA are desirable because a difference in the I value of only a few percent might cause sizable changes in the range and stopping maximum of therapeutic ion beams [35, 36], which could be biologically relevant for the accuracy of the energy deposited in nanometer volumes [37].

The PPA allows an accurate description of the dielectric properties of DNA including information regarding the condensed state and the ionization potential allows us to calculate how electrons in different shells contribute to the mean excitation energy I, which is obtained from Eq. (14) when the transfer energy $\hbar\omega \longrightarrow \infty$. Figs.(5) & (6) show the value of I in (eV) of DNA and Liquid water as a function of the transferred energy E in eV. When $\hbar\omega \longrightarrow \infty$, the ionization energy I = 80.01 eV using the program AuFa- H- DNA-1.For which is written in Fortran -90 [38].The value of I we obtained is somewhat lower than that of 81.5 eV calculated previously by Abril, et al. [2] and higher than that of 77.9 eV calculated by LaVerne and Pimblott [39] and clearly differs from that of ,85 eV obtained when applying Bragg's rule in ICRU Report 49 [40] as well as from that of 86.64 eV calculated by Tan, et al. [41], because each author used the same experimental ELF to calculate I values and that could be due to the different extrapolation of the ELF at intermediate energy transfers used by each group , where there are no experimental data. So it is interesting also to compare with an ionization potential I of liquid water where a value of $I_{\text{liq.water}} \cong 78.17$ eV was recently obtain by using a similar procedure that used before.

Fig-5. The ionization potential (*I*) of the stopping power theory is calculated as a function of the maximum energy transfer in DNA from equation (14).

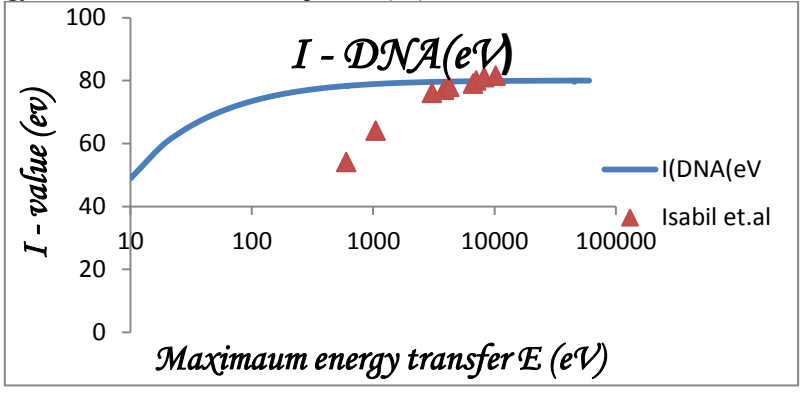
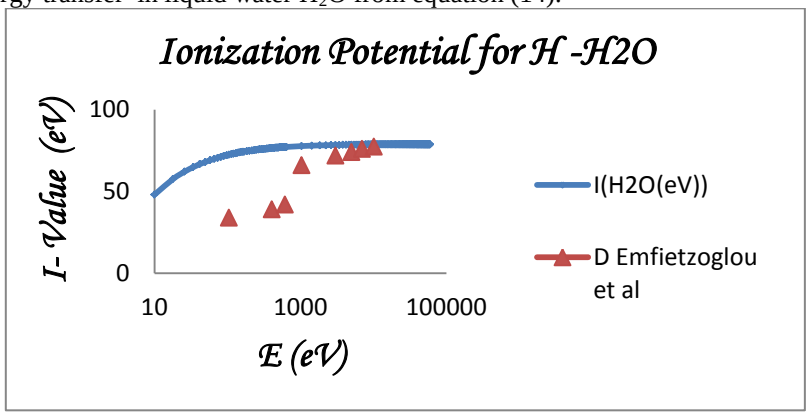


Fig-6. The ionization potential (I) of the stopping power theory is calculated as a function of the maximum energy transfer in liquid water H_2O from equation (14).



4. CONCLUSIONS

In the present work the PPA of the dielectric formalism has been applied to calculate the probability, the average energy of electronic excitation and ionization potential of the hydrogen ion beam in DNA target by using a program AuFa- H- DNA-1.For.

The spectral distribution of the electronic excitations induced by proton in DNA is calculated and found that , regardless of proton energy, the probability distribution P , of electronic excitations has a maximum around 20 eV. Besides, P(E,T), which is related to the number of electronic excitations of a given energy E, decreases with the incident proton energy T. On the other hand the mean energy $\langle E(T)\rangle$ of the electronic excitations increases monotonically with the proton energy T being around 9-10 eV at T = 0.001 MeV and around 80 eV for T = 10 MeV.

The ionization potential which is calculated in present work I = 80.01 eV gives a good agreement with I. Abril et. al results I= 81.5 [2], and conclude that the protons with energy between 0.05 MeV to 2.5 MeV are very efficient in producing secondary electrons, which are able to produce strand breaks, and it was a significant to compare I_{DNA} with $I_{\ liquid\ water}$, we found that $I_{\ liquid\ water}$= 78.17 eV . In the other hand $I_{DNA}$is $\sim 2.2\%$ larger than $I_{\ liquid\ water}$ by using the same procedure of calculation.

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