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The Radial Distribution of Dose around the Path of a Heavy Ion in Liquid Water

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Abstract

Monte Carlo calculations of the radial distribution of dose in liquid water, incorporating energy deposition due to primary excitations and ionizations, have been performed for protons of energy 1, 10, 20, 50, and 100 MeV. By combining these results with earlier semi-empirical formulae used in track structure theory calculations, a corrected analytic formulation has been developed which on radial integration closely reproduces the value of stopping power for protons in the energy range 0.1–1000 MeV. After including a β -dependent ‘effective charge’ formula, this corrected formulation is tested against all published measurements of radial distribution of dose from energetic ions in gaseous media. Though some inconsistencies at the closest and the farthest reaches of the radial distribution of dose remain, the overall agreement is very satisfactory, indicating that the ‘effective charge’ Z^* , and Z^{*2}/β^2 scaling are phenomenologically valid concepts for describing the radial dose from heavy ions of energies above $\sim 0.5\text{MeV}/\text{amu}$.

1. Introduction

The delta-ray theory of track structure (Butts and Katz, 1967; Katz and Kobetich, 1969; Katz *et al.*, 1972; Katz, 1978) makes no attempt at following the detailed pathways from the initial array of excitations and ionizations around the path of a heavy ion penetrating the detector medium, to the finally observed physical or biological endpoint. Instead, the approximation is made that the detector may be calibrated by exposing it to gamma-rays, whereby the targets in the detector are bathed in a uniform field of secondary electrons following the gamma irradiation. The response of a detector to a given dose of gamma-rays is interpreted as the fraction of available targets which have been affected by radiation, or as the probability that a target will be activated at this gamma-ray dose. If the average radial distribution of dose from delta-rays about the ions’s path is known, the gamma-ray calibration serves to approximate the radial distribution of effect, *i.e.* the radial probability of target activation. Except as inherently present in the calibrating dose-response function, fluctuations for gamma-rays and delta-rays are neglected. The expectation value of the single-particle action cross-section can then be specified as the integral of this radial probability over all radii, and the fraction of targets in the detector activated to the observed endpoint, *i.e.* the response of a detector after a measured fluence of particles, calculated.

In this scheme, the radial distribution of delta-ray dose around the path of a heavy ion represents a transfer function, relating the (non-linear) response of a detector after uniform doses of secondary electrons generated by gamma-rays, to the response of this detector after a non-uniformly distributed dose of delta-rays accompanying the passage of charged heavy particles through the detector medium.

How good an approximation is the calibration of a detector with gamma-rays? Since the response of a detector to electrons can be expected to depend on their energies, one should calibrate the detector with electrons whose energy spectrum approximates the energy spectrum of the delta-rays. In practice, however, the electron energy spectrum varies from shell to shell with radial distance from the ion’s path, making it impossible to perform a suitably matched exact calibration. Ultimately, neglecting the difference between the radially varying electron energy spectrum from delta-rays and the energy spectrum of the secondary electrons following gamma-ray irradiation can be justified by the considerable success of this first-order model in describing the response of physical detectors, such as the appearance of tracks in nuclear emulsions (Katz and Kobetich, 1969), the light output of scintillators (Katz and Kobetich, 1968), or thermoluminescent dosimeters (Waligórski and Katz, 1980), and the response of biological systems: inactivation of enzymes and viruses (Butts and Katz, 1967), survival of

mammalian cells (Roth *et al.*, 1976; Katz *et al.*, 1985), or generation of neoplastic transformations in such cells (Waligórski and Katz, 1986), after high-LET irradiations.

At the time the model was conceived (Butts and Katz, 1967), no measurements of the radial distribution of dose were available. The original formula was constructed using a number of simplifying assumptions, among which were the use of a linear energy-range relationship for electrons in aluminum, normal electron ejection, and the Rutherford formula for the distribution of delta-rays.

Since then, several measurements of this distribution have been made, in air (Varma *et al.*, 1976), hydrogen (Baum *et al.*, 1968), and tissue-equivalent gas (Baum *et al.*, 1974; Wingate and Baum, 1976; Menzel and Booz, 1976; Varma *et al.*, 1977, 1980; Varma and Baum, 1980), and corresponding calculations performed using the Monte Carlo technique (Berger *et al.*, 1972; Paretzke *et al.*, 1974; Hamm *et al.*, 1976; Turner *et al.*, 1980, 1980a; Todo *et al.*, 1982; Zaider *et al.*, 1983).

Recently, using more accurate power-law energy-range relationships for electrons and a selected value for the ionization potential of electrons in the detector, a more accurate formula describing the radial distribution of delta-ray dose has been developed (Zhang *et al.*, 1985).

Over the last few years a Monte Carlo code has been developed to calculate proton and alpha-particle tracks, with the inclusion of all primary excitation and ionization events accompanying the passage of these ions through liquid water (Hamm *et al.*, 1985).

The aim of the present work is to develop a semi-empirical analytic formula for the radial distribution of delta-ray dose which adequately reproduces the results of these Monte Carlo calculations for protons, extending continuously over a wide range of proton energies and which has been so adjusted that on radial integration it approximates the value of proton stopping power. By incorporating the previously used (Butts and Katz, 1967; Zhang *et al.*, 1985) energy-dependent effective charge formula, $Z^* = Z^*(\beta)$ (where β is the relative velocity of the ion), and the Z^{*2}/β^2 factor, we are able to calculate the radial distribution of dose for any ion species of any energy within the applicable energy range (0.1–1000 MeV/amu). Our formula is then compared to the available measurements of radial distribution of dose (Varma *et al.*, 1976; Baum *et al.*, 1974; Wingate and Baum, 1976; Menzel and Booz, 1976; Varma *et al.*, 1977, 1980; Varma and Baum, 1980), to other semi-empirical calculations (Fain *et al.*, 1974, 1974a; Chatterjee *et al.*, 1973) and to other Monte Carlo calculations (Paretzke *et al.*, 1974; Zaider *et al.*, 1983).

Within our approximative scheme, we assume that the corrected formula includes all primary excitation and ionization events close to the ion's path. Use of this formula may be justified if better agreement is achieved

between track theory calculations and experimentally measured high-LET detector response. We show that this is indeed so for the inactivation of dry enzymes and viruses (Waligórski *et al.*, 1986) and for the ferrous sulphate (Fricke) (Katz *et al.*, 1986) and alanine (Waligórski *et al.*, 1986) dosimeters. We may then address the question of the contribution of primary effects to the response of physical and biological detectors.

2. The Monte Carlo Calculation

The Monte Carlo computer code OREC (Hamm *et al.*, 1985; Hamm *et al.*, 1985, and references therein) was used to calculate the passage of a heavy charged particle or electron and all secondary electrons through the initial physical stage of interactions (10^{-15} s) in liquid water. A complex dielectric response function, $\epsilon(w, q)$ was developed for liquid water, where $\hbar w$ and $\hbar q$ are the energy and momentum transferred by the charged particle (proton) to the medium (\hbar is Planck's constant/ 2π). Collective effects in the condensed phase have thus been included *a priori*. The macroscopic cross-sections, or inverse mean free paths, for any kind of inelastic interaction are then obtained directly from $-Im(1/\epsilon)$, the negative of the imaginary part of $1/\epsilon$. In contrast to vapor, energy can be shared collectively by large numbers of electrons in liquid water. A suitable algorithm for treating initially delocalized excitations in the liquid has been developed and applied, under the assumption that the probability of a given mode of de-excitation is proportional to $-Im(1/\epsilon)$ and depends on the distance from the particle track. The electronic transitions were divided into six specific excitation and five ionization events. The numeric information assembled in the proton energy range 1–100 MeV leads to electronic stopping powers for protons which differ less than 10% from tabulated values.

To obtain the cross-sections needed for the transport of secondary electrons in water, the angular distributions of electrons produced in inelastic collisions were computed by a simple algorithm, giving an isotropic distribution for small values of energy transfer and a 'free electron' distribution at large energy transfers. To account for the appreciable changes of the directions of electron travel at low energies, elastic scattering of electrons of those energies was computed on the basis of phase-shift analysis and joined to the Thomas-Fermi model at higher energies. Also, in contrast to heavy ions, electron exchange was taken into account explicitly.

Tracks of protons of energy 1, 10, 20, 50, and 100 MeV were calculated using the OREC code. The energy deposited was added in several concentric cylindrical shells around the proton's path. Thus, histograms representing the radial distribution of energy deposited were obtained. These are shown in Figure 1.

3. The Corrected Formula for the Radial Distribution of Dose

Zhang, Dunn, and Katz (1985) have developed the following formula describing the radial distribution of dose around the path of a heavy ion (see Appendix 1).

$$D_1(t) = \frac{Ne^4 Z^{*2}}{amc^2 \beta^2 t} \left[\frac{\left(1 - \frac{t + \theta}{T + \theta}\right)^{1/2}}{t + \theta} \right] \quad (1)$$

where $D_1(t)$ is the dose deposited in a coaxial cylindrical shell of thickness dt at a distance t from the path of an ion of effective charge Z^* moving with a relative velocity $\beta = v/c$ (c is the speed of light) through the detector medium containing N electrons per cm^3 , m is the mass of the electron. The Rutherford cross-section for delta-ray production from atoms having ionization potential $I = 10$ eV, normal ejection and power law range (r)-energy (w) relationship for electrons, are assumed. The range-energy relationship is based on a two-component fit to the available experimental data (Kobetich and Katz, 1968; Iskef *et al.*, 1983, see Appendix 1) concerning ranges of electrons in aluminum:

$$r = kw^\alpha \quad (2)$$

where

$$k = 6 \times 10^{-6} \text{ g cm}^{-2} \text{ keV}^{-\alpha} \quad (3)$$

$$\text{For } w < 1 \text{ keV, } \alpha = 1.079,$$

$$\text{and for } w > 1 \text{ keV, } \alpha = 1.667 \quad (4)$$

θ is the 'range' of an electron of energy $w = I$

$$\theta = k (0.010 \text{ keV})^{1.079} = 4.17 \times 10^{-8} \text{ g cm}^{-2}. \quad (5)$$

The kinematically limited maximum delta-ray energy is

$$W = 2mc^2 \beta^2 / (1 - \beta^2). \quad (6)$$

This translates to the maximum range of delta-rays:

$$T = kW^\alpha \quad (7)$$

where the choice of α (see equation (4)) depends on the velocity β of the ion. We calculate

$$\text{for } \beta < 0.03, \alpha = 1.079, \text{ and}$$

$$\text{for } \beta > 0.03, \alpha = 1.667. \quad (8)$$

For water

$$\frac{2\pi Ne^4}{mc^2} = 1.369 \times 10^{-7} \frac{\text{erg}}{\text{cm}} = 8.5 \text{ keV mm}^{-1}. \quad (9)$$

Like in the earlier work of Butts and Katz (1967) and of Zhang *et al.* (1985), the effective charge of an ion of Z elementary charges, moving with a relative velocity β is calculated from the expression of Barkas (1963)

$$Z^* = Z[1 - \exp(-125 \beta Z^{-2/3})]. \quad (10)$$

The radial distribution of delta-ray dose in water, calculated for protons of energies 1, 10, 20, 50, and 100 MeV using expression (1) are compared in Figure 1 with results of the Monte Carlo calculation. Results of radial integration of equation (1), expressed as the ratio of the proton stopping power (LET_∞) at the corresponding proton velocity, β , for β ranging from ~ 0.01 to 0.9 (corresponding to proton energies in the range 0.05–1,000 MeV), are shown in Figure 2. The algorithm used for calculating LET_∞ , based on the tables of Janni (1982) is given in Appendix 2.

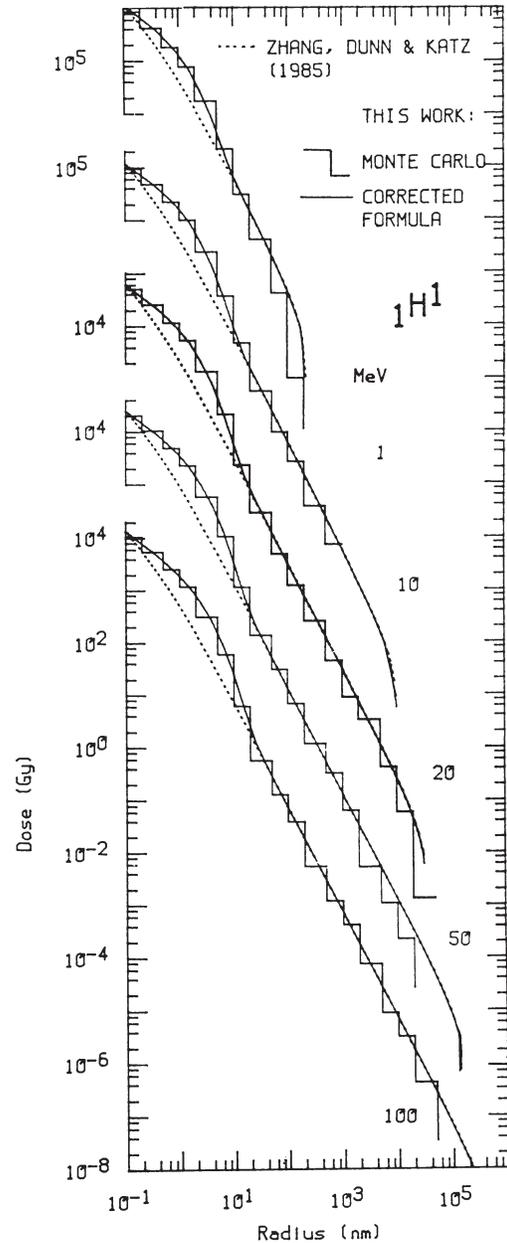


Figure 1. Radial distribution of energy deposited around the path of protons of energies 1, 10, 20, 50, and 100 MeV. Monte Carlo calculations (in liquid water) are presented as histograms. Full lines: equation (11), dotted lines: equation (1).

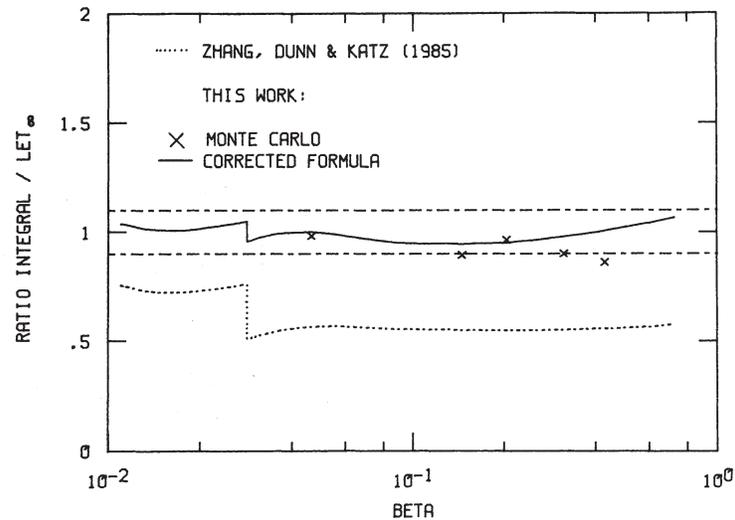


Figure 2. Ratios of the radially integrated distributions of energy of Figure 1 (\times —Monte Carlo calculations for 1, 10, 20, 50, and 100 MeV, full line: equation (11), dotted line: equation (1)), to proton stopping power, LET_{∞} , plotted vs. relative speed of proton, β . Broken lines represent 10% error limits.

To account for the “missing” radial dose due to primary events in the region of $t = 1$ –10 nm (see Figure 1), we seek a correction to equation (1) in the form

$$D_2(t) = D_1(t) (1 + K(t)) \quad (11)$$

where we find it convenient to seek an expression of the type $K(t) = at \exp(-at)$.

After making some semi-empirical adjustments, we arrive at the following expression:

(a) for $t > B = 0.1$ nm

$$K(t) = A \left(\frac{t-B}{C} \right) \exp - \left(\frac{t-B}{C} \right) \quad (12.1)$$

where

$$B = 0.1 \text{ nm}$$

$$C = 1.5 \text{ nm} + 5 \text{ nm} \times \beta$$

and

$$A = 8 \times \beta^{1/3}, \text{ for } \beta < 0.03$$

or

$$A = 19 \times \beta^{1/3}, \text{ for } \beta > 0.03.$$

(b) for $t < B = 0.1$ nm

$$K(t) = 0. \quad (12.2)$$

The corrected expression for $D_2(t)$ (equations (11) and (12)) features a “hump” at radial distances $t = 1$ –10 nm and reduces to the expression of Zhang *et al.* (1985) (equation (1)) at greater t .

4. Results

The results of Monte Carlo calculations of radial distribution of dose in liquid water around the path of protons of energies 1, 10, 20, 50, and 100 MeV are presented

as nested histograms in Figure 1, together with the corresponding radial distributions of dose calculated using equation (1) and equation (11). These distributions, after radial integration, are compared with proton stopping power LET_{∞} , calculated using the algorithm described in Appendix 2. In Figure 2 the results of integrating the Monte Carlo histograms and equations (1) and (11), are presented in the form of ratios of LET_{∞} , as a function of β , the relative speed of the proton. The corrected formula reproduces the proton stopping power to within 10% over the range of β from 0.015 to 0.9, while the original formula (equation (1)) yields about half of this value. The “step” at $\beta = 0.03$ results from changing the electron range power law constant from 1.079 to 1.667 (see equation (4), equation (8) and equation (12)).

We compare the radial dose distributions calculated using both formulae with the measured distributions, for light particles (Figure 3), and for relativistic and slow heavy particles (Figure 4 (A & B) and Figure 4 (C & D) respectively). Comparison with other Monte Carlo calculations of Paretzke (1974), of Zaider *et al.* (1983), and with semi-empirical calculations of Fain *et al.* (1974, 1974a) are presented in Figure 3 (A), Figure 4 (A), and Figure 4 (B, C, & D) respectively.

The complete set of published experimental data on the radial distribution of dose measured in air, tissue-equivalent gas, hydrogen, and water vapor, consists of 15 different sets of measurements. For reasons of economy in publication, only nine of these have been illustrated, in Figures 3 and 4.

5. Discussion

The published measurements of the radial distribution of dose span a fairly wide range of ion species

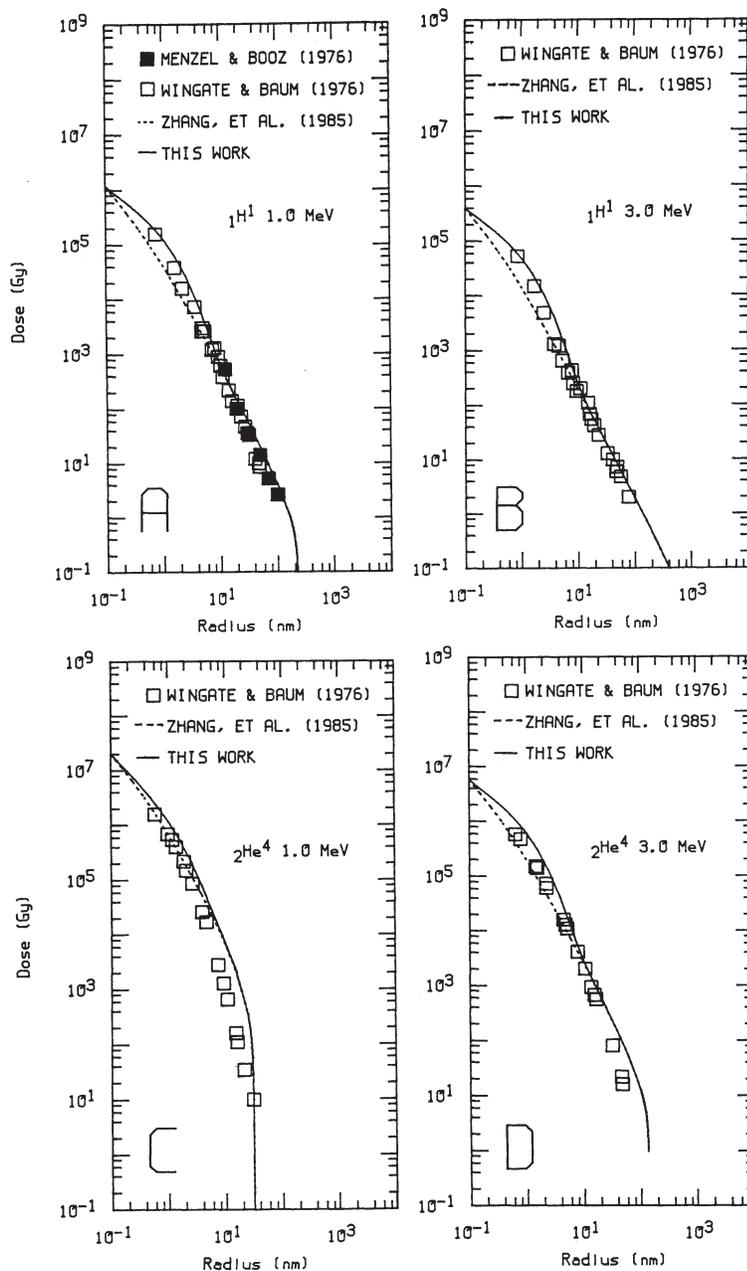


Figure 3. Comparison of measured radial distributions of dose for **A)** 1 MeV protons (Baum *et al.*, 1974; Wingate and Baum, 1976); **B)** 3 MeV protons (Wingate and Baum, 1976); **C)** 1 MeV α -particles (Wingate and Baum, 1976); **D)** 3 MeV α -particles (Wingate and Baum, 1976), with equation (11) = full lines, and equation (1) = dotted lines.

and energies. Measurements of this distribution have been made for protons of energies 1 MeV ((Wingate and Baum, 1976; Menzel and Booz, 1976), Figure 3(A)), 2 MeV (Wingate and Baum, 1976; Menzel and Booz, 1976), and 3 MeV ((Wingate and Baum, 1976), Figure 3(B)), deuterons of 1 MeV and 2 MeV (Menzel and Booz, 1976) and helions of energies 1 MeV (Figure 3(C)), 2 MeV and 3 MeV (Figure 3(D)) (Wingate and Baum, 1976). While our formulae reproduce the measurements made for helions somewhat less accurately (see Figures 3(C) and

3(D)), the agreement between our calculations and the remaining measurements, including those not shown, is quite adequate.

The same is true for measurements of energetic helions of 930 MeV ((Varma *et al.*, 1976), Figure 4(A)), and 377 MeV/amu ⁴⁰Ne ions ((Varma and Baum, 1980a), Figure 4(B)) as well as for slow heavy ions: 42 MeV ⁷⁹Br₃₅ ((Varma *et al.*, 1980), Figure 4(C)), 61.9 MeV ¹²⁷I₅₃ ((Baum *et al.*, 1974), Figure 4(D)), 41.1 MeV ¹⁶O (Varma *et al.*, 1977), 38.4 MeV ¹⁶O and 33.25 ¹²⁷I ions (Baum *et al.*, 1974).

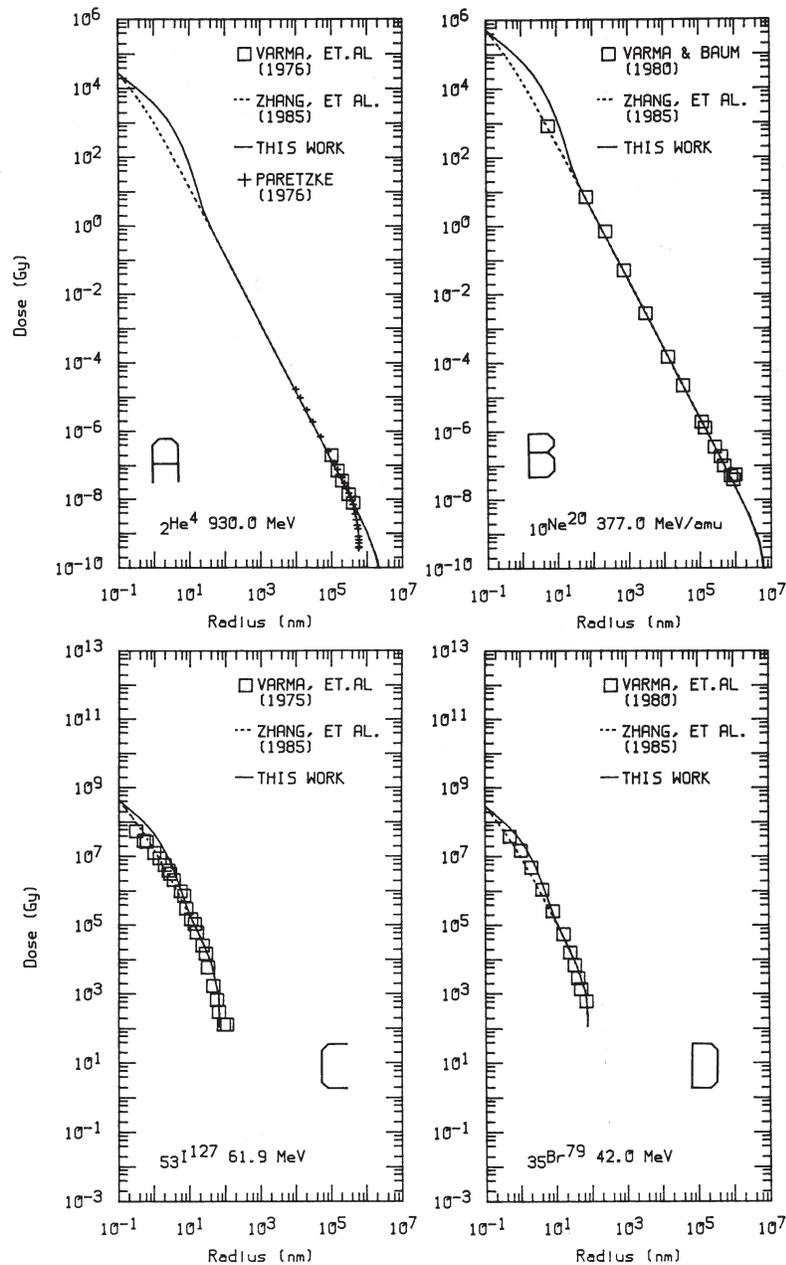


Figure 4. Comparison of measured radial distributions of dose for A) 930 MeV α -particles (Varma *et al.*, 1976), B) 377 MeV/amu α -particles (Varma and Baum, 1980), C) 61.9 MeV iodine 127 (Baum *et al.*, 1974), D) 42 MeV bromine 79 ions (Varma *et al.*, 1980) with equation (11) = full lines, and equation (1) = dotted lines. Crosses in panel A indicate the Monte Carlo calculation of Paretzke (Varma *et al.*, 1976).

We also find good agreement between results of our calculations and those of Fain *et al.* (1974, 1974a) for 2 MeV/amu ^{12}C , 8.1 MeV/amu ^{40}Ne and 90 MeV/amu ^{56}Fe ions, as shown in Figures 5(B), 5(C), and 5(D), respectively. In the region below ~ 30 nm, results of these calculations lie between our "corrected" and "uncorrected" distributions.

Our corrected formula (equation (11)) appears to reproduce very well the result of the Monte Carlo calcu-

lation of Zaider *et al.* (1983) (Figure 5(A)), except for the region below ~ 1.5 nm. This indicates that the differences between the PROTON and DELTA codes (Zaider *et al.*, 1983) which apply to water vapor, and the OREC code for liquid water on which our corrected formula is based, need to be clarified in the region where primary effects are important.

The overall results are somewhat surprising though gratifying. We use a rather simplistic calculation of the

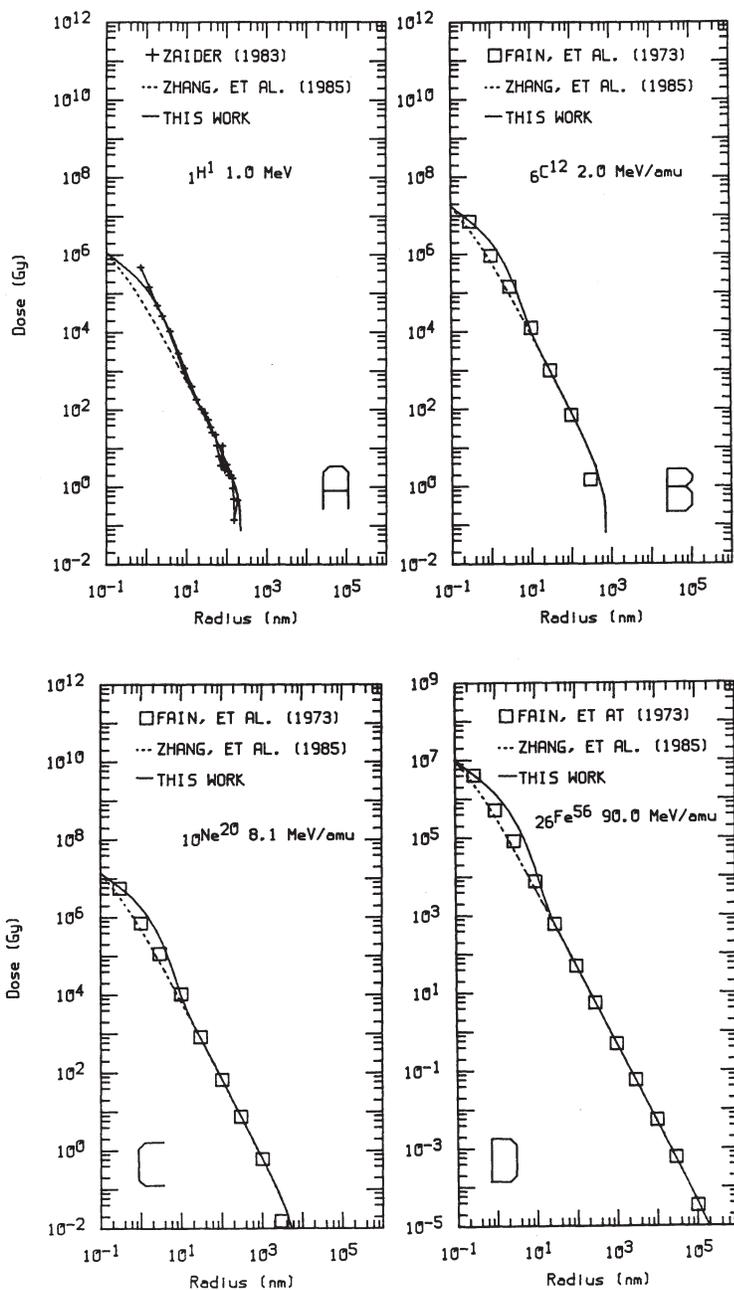


Figure 5. Comparison of calculated radial distributions of dose for **A)** 1 MeV protons (Zaider *et al.*, 1983); **B)** 2 MeV/amu carbon 12; **C)** 8.1 MeV/amu neon 20; and **D)** 90 MeV/amu iron 56 ions with equation (11) = full lines, and equation (1) = dotted lines. Data in panels B, C, and D are from Fain *et al.* (1974, 1974a).

radial distribution of dose in which all components are greatly oversimplified. We compare our calculations to measurements made in a variety of gases. Our principal electron data (equations (3) and (4)) are taken from measurements of electron ranges in aluminum. The electron density constant (equation (9)) is for liquid water. We assume normal ejection of delta-rays and use an arbitrary value of ionization potential (equation (5)). We exploit an “effective charge” formula (equation (10)) origi-

nally fitted to range and stopping power experiments in nuclear emulsion.

It seems that the radial distribution of dose is relatively insensitive to these details, except for (a) ion energies below 0.05 MeV/amu, and (b) both very small and very large radial distances.

Judging from the appearance of Figures 3, 4, and 5, in the region of radial distances below 30 nm, our “uncorrected” formula (equation (1)) appears to fit experimen-

tal data better than the corrected one. However, only the latter reproduces the proton stopping power. It appears that in the region where primary effects are important, energy-corrected values of W , the energy to form an ion pair, rather than a single value of W for all radial distances, should be used experimentally. This point has already been raised by Fain (Fain *et al.*, 1974, 1974a) and by Paretzke (Paretzke *et al.*, 1974). The available data do not permit us to suggest any form of radial correction of the W value.

In Figure 6 we display experimental data as plots of radial dose multiplied by the square of the radial distance and by β^2/Z^{*2} , where the "effective charge," Z^* , is calculated using equation (10). Data are displayed in groups of similar β . Three such groups, of energies 1, 0.5, and 0.25 MeV/amu were selected. Here, differences between the measured data points and our distributions can be visualized more readily. The 1 MeV/amu group consists of particles of charge 1 with practically no "effective charge" correction. The distribution of data points provides us with a measure of the overall consistency of the experimental technique. Judging from the roughly similar spread of data points around our calculation for the 0.5 MeV/amu group, we conclude that the "effective charge" formula appears to describe the effects of the complicated process of charge exchange to present experimental precision. Whether the same conclusion can be drawn for the 0.25 MeV/amu data points, is

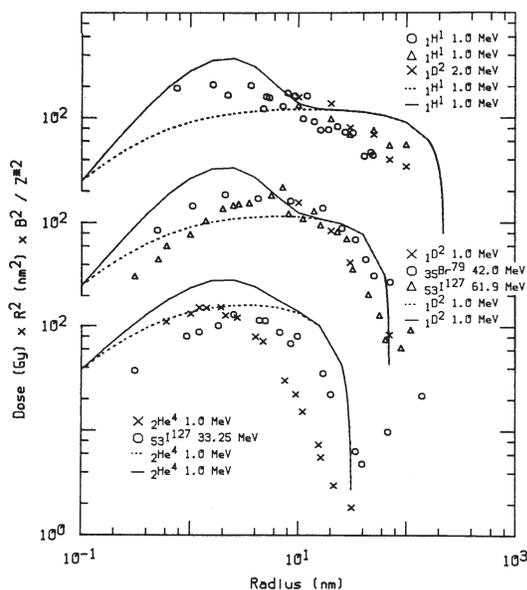


Figure 6. Measured radial distributions of dose, multiplied by the square of the radial distance and by β^2/Z^{*2} , for ions of 1 MeV/amu (uppermost group), 0.5 MeV/amu (central group), and 0.25 MeV/amu (lowest group). Full lines represent equation (11), dotted lines equation (1). Key to sources of data: 1 MeV H; circles = Wingate and Baum (1976); triangles = Menzel and Booz (1976); 2 MeV D = Menzel and Booz (1976); 1 MeV D = Menzel and Booz (1976); 42 MeV Br = Varma *et al.* (1980); 61.9 MeV I = Baum *et al.* (1974); 1 MeV He = Wingate and Baum (1976); 33.25 MeV I = Baum *et al.* (1974).

debatable, at least until further measurements are made. Some anomalous data points for the 61.9 MeV and the 33.25 MeV $^{127}_{53}\text{I}$ ions are attributed by the authors of these measurements (Baum *et al.*, 1974) to scattered gas atoms and Auger electrons ejected from the incident ion.

The existence of a "track core" in the radial distribution of dose has been postulated by some authors (Mozumder and Magee, 1966; Magee and Chatterjee, 1980; Chatterjee and Magee, 1980), as connected with the Bohr adiabatic radius (*e.g.* Brandt and Ritchie, 1974). Our Monte Carlo calculations performed for liquid water do not confirm the existence of such a core at any region close to the ion's path.

6. Conclusions

Our corrected formula for the radial distribution of dose, despite its simplicity, offers a surprisingly good description of the available experimental data. We are able to adequately reproduce the ion's stopping power over a wide and continuous range of ion charges and speeds. The "effective charge" formula appears to represent a valid phenomenological description of the speed-dependent charge exchange process, for ions up to iodine at energies exceeding 0.5 MeV/amu, for purposes of calculating the radial distribution of dose. Clearly, more experimental data and more accurate Monte Carlo calculations would help us to further clarify the validity of our formula at regions close to the ion's path and near the outer reaches of the distribution of radial dose.

The original "uncorrected" formula has been shown to reproduce quite accurately the measured cross-sections for enzyme and virus inactivation (Zhang *et al.*, 1985), even though the radial integration of this formula yields only about half of the ion's stopping power. This gives rise to an interesting speculation that the response of some detectors may be basically related to the effects of the dose deposited by secondary processes, while other detectors may be "sensitive" also to energy depositions due to primary excitations close to the ion's path.

Our dose calculations and our consequent calculations of the detector response are based on averaged quantities. The success of these calculations suggests that knowledge of the detailed spectrum of energy depositions in nanometer or micrometer subvolumes and of its dependence on the separation of these volumes, may be superfluous when interpreting experimental data which are accurate at best to about 15%.

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Appendix 1. Development of the Radial Distribution of Dose Formula

The expression (1) for the radial dose distribution

$$D_1(t) = \frac{Ne^4 Z^{*2}}{amc^2 \beta^2 t} \left(\frac{1 - \frac{t + \theta}{T + \theta}}{t + \theta} \right)^{1/\alpha} \quad (\text{A.1})$$

is developed by the comparison of two calculations. In one of these the electrons of the medium are assumed to be initially bound with ionization potential I (which we have arbitrarily taken to be 10 eV), and the electron range-energy relation is linear. In the other the electrons are assumed to be free and the range-energy relation is given by a power law. The two-component power law fit to the range vs. energy data for electrons in aluminum is shown in Figure A.1. In both cases all secondary electrons (delta-rays) are assumed to be ejected normally and to travel in straight line paths. Primary energy deposition is neglected.

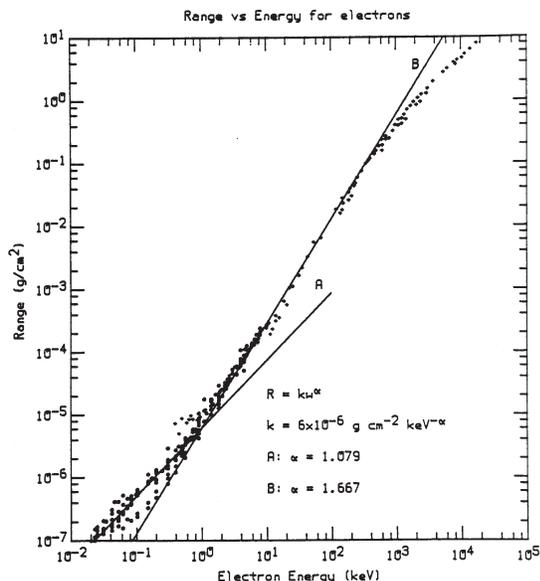


Figure A.1. Range vs. energy for electrons in aluminum, as fitted by two power law segments. Data from Kobetich and Katz (1968) and Iskef *et al.* (1983).

We start with the Rutherford formula for delta-ray production from a medium containing N electrons per unit volume, bound with ionization potential I

$$dn = \frac{2\pi Ne^4 Z^{*2}}{mc^2 \beta^2} \left(\frac{dw}{(w+I)^2} \right) \quad (\text{A.2})$$

where dn is the number of delta-rays per unit pathlength of energy between w and $w + dw$, Z^* is the "effective charge number" of the ion, e is the electron charge and β is the ion's speed relative to the speed of light.

Case 1

We write the range(r)-energy(w) relation for electrons as

$$r = kw \quad (\text{A.3})$$

so that

$$\frac{dw}{dr} = \frac{1}{k} \quad (\text{A.4})$$

If w_i is the energy of an electron of initial energy w , range r after penetrating thickness t of material

$$w_i = k(r - t) \quad (\text{A.5})$$

and

$$\frac{dw_i}{dt} = -k. \quad (\text{A.6})$$

Thus, we may write an expression for the dose $D_0(t)$ in a cylindrical shell of radius t , thickness dt , whose axis is the ion's path as

$$D_0(t) = \frac{1}{2\pi t dt} \int_{w_i}^W \left(-\frac{dw_i}{dt} \right) dt \frac{dn}{dw} dw \quad (\text{A.7})$$

where

$$W = 2mc^2 \beta^2 / (1 - \beta^2) \quad (\text{A.8})$$

is the (kinematically limited) maximum delta-ray energy. Straightforward integration leads to

$$D_0(t) = \frac{Ne^4 Z^{*2}}{mc^2 \beta^2 t} \left(\frac{1}{t + \theta} - \frac{1}{T + \theta} \right) \quad (\text{A.9})$$

where we have written

$$kw_i = t; \quad kI = \theta; \quad kW = T. \quad (\text{A.10})$$

As compared to the earlier work of Butts and Katz (1967), where $I = 0$, the effect of the ionization potential has been to replace t and T in the final bracket by $t + \theta$ and $T + \theta$, respectively.

Case 2

When the range-energy relation is

$$r = kw^\alpha \quad (\text{A.11})$$

we note that

$$\frac{dr}{r} = \alpha \frac{dw}{w} \quad (\text{A.12})$$

and

$$kw_i^\alpha = r - t \quad (\text{A.13})$$

$$w_i = \left(\frac{r - t}{k} \right)^{1/\alpha} = w \left(1 - \frac{t}{r} \right)^{1/\alpha} \quad (\text{A.14})$$

whence

$$-\frac{dw_i}{dt} = \frac{w}{\alpha r} \left(1 - \frac{t}{r} \right)^{1/\alpha - 1} \quad (\text{A.15})$$

Table A.1. Power expansion coefficients for LET(E) and range, $R(E)$ for protons in water. Units of LET: MeV g⁻¹ cm². Units of range: g cm⁻¹. Units of proton energy E : MeV/amu

	$1.E - 3 < E < 6.E - 2$	$6.E - 2 < E < 1.E + 1$	$1.E + 1 < E < 1.E + 3$	$1.E + 3 < E$
k	1	2	3	4
a_0	$-0.374474 E + 1^*$	$+0.553740 E + 1$	$+0.549002 E + 1$	$+0.163141 E + 2$
a_1	$-0.115638 E + 2$	$-0.712500 E 0$	$-0.670708 E 0$	$-0.541066 E + 1$
a_2	$-0.471453 E + 1$	$+0.886293 E - 3$	$-0.210655 E - 1$	$+0.618546 E 0$
a_3	$-0.911414 E 0$	$+0.190340 E - 2$	$-0.501344 E - 2$	$-0.232769 E - 1$
a_4	$-0.869548 E - 1$	$-0.108231 E - 1$	$+0.113600 E - 2$	0.0
a_5	$-0.328797 E - 2$	$+0.141793 E - 2$	0.0	0.0
a_6	0.0	$+0.649495 E - 3$	0.0	0.0
b_0	$-0.664955 E + 1$	$-0.603737 E + 1$	$-0.608610 E + 1$	$-0.141562 E + 2$
b_1	$+0.157200 E + 1$	$+0.163896 E + 1$	$+0.171916 E + 1$	$+0.498610 E + 1$
b_2	$+0.338162 E 0$	$+0.770354 E - 1$	$-0.493530 E - 2$	$-0.390014 E 0$
b_3	$+0.536396 E - 1$	$-0.350977 E - 1$	$+0.860489 E - 2$	$+0.124950 E - 1$
b_4	$+0.283165 E - 2$	$+0.304331 E - 2$	$-0.114098 E - 2$	0.0
b_5	0.0	$+0.192069 E - 2$	0.0	0.0
b_6	0.0	0.0	0.0	0.0

* Powers of 10 are given as numbers following E (e.g. $10^{-2} = 1.0 E - 2$; $10^2 = 1.0 E + 2$).

Taking $I = 0$ in equation (A.2) we find

$$D(t) = \frac{Ne^4 Z^{*2}}{amc^2 \beta^2 t} \int_{w_i}^w \frac{1}{r} \left(1 - \frac{t}{r}\right)^{1/\alpha - 1} \frac{dw}{w} \quad (\text{A.16})$$

Substituting from equation (A.12)

$$D(t) = \frac{Ne^4 Z^{*2}}{\alpha^2 mc^2 \beta^2 t} \int_t^T \left(1 - \frac{t}{r}\right)^{1/\alpha - 1} \frac{dr}{r^2} \quad (\text{A.17})$$

which is integrated by the substitution $y = 1/r$ to yield

$$D(t) = \frac{Ne^4 Z^{*2}}{amc^2 \beta^2 t} \left(\frac{\left(1 - \frac{t}{T}\right)^{1/\alpha}}{t} \right). \quad (\text{A.18})$$

From case 1 we note that the transition from the free to the bound electron case is made by replacing t and T within the final bracket by $t + \theta$ and $T + \theta$, respectively. We make the same substitution in equation (A.18) to yield equation (A.1).

Thus, while equation (A.1) is not derived directly, by integration, for the case of bound electrons, it is written so as to yield the correct functional form in the limiting cases; where $\alpha = 1$ and where $I = 0$. The use of the ionization potential makes it possible to find the total energy deposited by delta-rays by radial integration of the dose without an even more arbitrary specification of the lower limit of integration.

Appendix 2. Algorithm for Calculating Stopping Power and Range of Heavy Ions in Water

Least squares polynomials were fitted to the values of stopping power LET_p and range R_p of protons in water, as given in the tables of Janni (1982)

proton LET (MeV cm² g⁻²).

$$\ln \text{LET}_p(E_k) = a_{k0} + \sum_{j=1}^6 a_{kj} (\ln E_k)^j \quad (\text{A.19})$$

proton range (g cm⁻¹)

$$\ln R_p(E_k) = b_{k0} + \sum_{j=1}^6 b_{kj} (\ln E_k)^j \quad (\text{A.20})$$

where the values of energy ranges E_k and power expansion coefficients are given in Table A.1.

To calculate the values of LET(E) and range $R(E)$ for an ion of atomic mass amu and energy E (MeV/amu), "rest" charge Z , and relative speed β , select the appropriate energy range E_k ($E_{k-1} < E < E_{k+1}$), and use the expressions (Barkas and Berger, 1964):

$$\text{LET}(E) = (Z^*/Z_p^*)^2 \text{LET}_p(E) \quad (\text{A.21})$$

$$R(E) = (\text{amu}/Z)^2 (R_p + C) \quad (\text{A.22})$$

where

$$\text{if } \beta < Z/69, \text{ then } C = 2.284 \times 10^{-3} \beta Z^{5/3} \quad (\text{A.23})$$

else

$$C = 3.341 \times 10^{-5} Z^{8/3} \quad (\text{A.24})$$

Z^* and Z_p^* are calculated using the "effective charge" formula (equation (10)) for the ion "rest" charge Z and proton "rest" charge $Z_p = 1$, respectively.

Tables of stopping power and range of ions in water have been generated (Katz, 1985), and a program using this algorithm implemented on the Hewlett-Packard HP-41C pocket calculator (Waligórski, 1985). Both are available from the authors on request.