Semiempirical Calculation of the Differential Energy Expended per Ion Pair for Fission Fragments in H_2†

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High and low limits for the variation of w_{ff}, the differential energy expended per ion pair, are calculated for the average-energy Cf^{252} fission fragment in H_2. The electronic and nuclear stopping cross sections are obtained from the "unified" range theory for heavy ions constructed by Lindhard and co-workers and from recent experimental energy-distance data. Published experimental data of primary ionization cross sections for protons in hydrogen over the appropriate recoil energies yield the high limit. The low limit is obtained when protons are considered to expend energy in the creation of an ion pair at the same rate for their whole trajectory as they do at very high energies. The variation of w_{ff} with fission-fragment energy can be calculated if total ionization cross sections of the stopping-medium knocked atoms and energy-distance curves of the incident particles are known.

I. INTRODUCTION

An interesting problem connected with the primary effects produced by the passage of heavy charged particles through matter is the dependence on the incident particle energy of the energy expended in the formation of an ion pair. Reviews on the general subject have been published by Binks[1] and by Valentine and Curran.[2] Since an interest in over-all effects is generally the case, a quantity W defined as the average energy expended per ion pair (E/I) has frequently been used. Fission fragments, however, are known to lose their energy mainly by two processes which are predominant in different energy ranges: inelastic collisions with electrons at high energies and elastic collisions with the screened nuclei at low energies. This change in the energy loss process suggests that the primary species formed in the interaction will vary in quantity, if not in nature, along the path.[3] For fission fragments, then, it seems of greater interest to study the dependence of w, the differential energy expended per ion pair (dE/dI), on the incident particle energy, rather than of W, which is an average. A simple expression relates the two;

$$1/w=(1/W)(1-d\ln W/d\ln E). \tag{1}$$

If I, the number of ions produced, and E are proportional throughout the whole trajectory, then w equals W. This is not the case, however, for heavy charged particles.

Studies of W_r, the average energy expended per ion pair for recoil particles, have been made in the low energy range, 100 to 170 keV. Stone and Cochran,[4] Madsen,[5] and Jesse and Sadauskis[6] have measured W_r for various α emitters in gases. Although their results differ, it is certain that the ratio W_r/W_α is not unity in this energy range. Schmitt and Leachman[7] measured the ionization produced by fission fragments of various energies in N_2, Ne, Ar, and a mixture of Ar and CO_2. They found that in the energy interval between 25 and 100 MeV the number of ions produced is a linear function of the particle energy within the accuracy of the experiment. Although w_{ff} is a constant in that energy range, it is observed that W_{ff} for the full energy fragments is not equal to W_α in those gases.

The energy dependence of w for different systems has been investigated. Jesse[8] obtained experimental data for the variation of w_α and W_α in N_2 and C_2H_4. Dalgarno and Griffing[9] calculated the variation of w_e and W_e with electron energy in atomic hydrogen. Erskine[10] made a similar calculation for α particles in helium. At low energies, he found that since the efficiency in the production of ions decreases, both w_α and W_α increase. This decrease in efficiency should be more marked for fission fragments since elastic collisions with atoms at the end of the fragment path are more important than for lower mass projectiles, but apparently no detailed calculations have been made heretofore. Since general interest in the use of fission fragments, particularly in radiation chemistry, has risen in the past years,[11] it seemed opportune to attempt a similar study to Erskine's.[10] A calculation was therefore made of the low and high limits of w_{ff} as a function of energy for the average energy Cf^{252} fission fragment (atomic mass=126, atomic number=49, and initial energy=91.4 MeV) in H_2.

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II. THE ANALYTICAL EXPRESSION FOR w

The differential energy expended per ion pair w can be expressed as the ratio of the stopping power and the specific ionization of the particle in the medium;

$$w=(-dE/dx)/(dI/dx). \tag{2}$$

To obtain w as a function of the energy, it is necessary to know the variation of dE/dx and dI/dx with E.

The specific ionization of the particle can be described[12] as

$$\frac{dI}{dx}=N\left(S^e+\int_0^{E_m'} S(E,E')I'(E')dE'\right), \tag{3}$$

where S^e is the cross section for the production of ion pairs including ionization produced by ejected electrons in the collision of a particle with a target molecule. The integral represents the integrated cross section for ionization produced by the recoil gas molecules or atoms. The cross section $S(E,E')$ represents the probability of the production of a recoil atom of energy E' by a particle of energy E. $I'(E')$ is the number of ions produced by the recoil atom of energy E' and E_m' is the maximum energy transferred to a gas atom by a particle of energy E in an elastic collision;

$$E_m'=[4MM'/(M+M')^2]E. \tag{4}$$

The stopping power of the incident particle in the gas is similarly described as

$$-dE/dx=N(b^e+b^n), \tag{5}$$

where b^e is the stopping cross section per gas molecule for the loss of energy to electrons in an inelastic collision and b^n for the loss of energy to recoil atoms in an elastic collision.

Defining the kernel

$$k(E,E')=S(E,E')E' \Big/ \int_0^{E_m'} S(E,E')E'dE', \tag{6}$$

and since by definition the integral in the denominator of the right-hand side of Eq. (6) is b^n, then the following expression can be obtained:

$$1-\frac{w^e}{w}=\frac{b^e-w^eS^e}{b^e+b^n}+\frac{1}{1+b^e/b^n}\int_0^{E_m'} k(E,E')1-\frac{w^e}{W'}dE', \tag{7}$$

where W' equals the ratio E'/I' and w^e, a constant, represents the value of w at very high velocities. This constant is assumed to be independent of the nature of the incident particle.

Knipp and Ling's[12] calculation of the fission fragment ionization defect assumes that the first term is zero during the stopping process. This is strictly correct if the rate of energy loss by the incident particle to electrons (b^e) and the rate of primary ionization including that produced by secondary electrons are strictly proportional through the constant w^e. This approximation can be justified at very low velocities since energy loss to recoil atoms predominates; thus $b^n \gg b^e$ and the first term of Eq. (7) tends to zero. At high velocities, energy loss to electrons is predominant, thus $b^e \gg b^n$ and the same term tends to zero. In the latter energy range, the experimental evidence obtained by Schmitt and Leachman[7] confirms this approximation. At some intermediate energies, this term might possibly have negative or positive values but it will be assumed to equal zero throughout the whole trajectory of the incident particle.[12] Then Eq. (7) reduces to

$$1-\frac{w^e}{w}=\frac{1}{1+b^e/b^n}\int_0^{E_m'} k(E,E')1-\frac{w^e}{W'}dE'. \tag{8}$$

If the value of W' is equal to w^e throughout the whole trajectory of the knocked atoms, then one observes that w is equal to w^e throughout the whole trajectory of the incident particle.

III. W_p' FOR PROTONS IN HYDROGEN GAS

The variation of W_p', the average energy expended per primary ion pair for protons in H_2, is calculated from measured ionization cross sections s_i', since

$$dI_p'/dx=s_i'N_{H_2}, \tag{9}$$

where N_{H_2} is the atomic density of the hydrogen gas. Curran and Donahue[13] obtained s_i' in the range of proton energies between 4.0 and 36.0 keV, Foguel' *et al.*[14] between 12.3 and 56.7 keV, Afrosinov *et al.*[15] between 60 and 180 keV, and Hooper *et al.*[16] between 200 and 1100 keV. A theoretical formula for the primary ionization cross section of protons in hydrogen atoms was developed by Bates and Griffing.[17] Hooper *et al.* made a correction to this theoretical expression in order to apply it to the case of molecular hydrogen and found excellent agreement with their experimental data. For proton energies higher than 1.1 MeV, the corrected formula of Bates and Griffing is used to obtain s_i'.

The energy-range curve[18] for protons in H_2 was used to integrate dI_p'/dx and to obtain I_p', the number of primary ion pairs produced in the stopping of a proton of energy E'. Finally, W_p' was calculated as a function of E' (see Fig. 1).

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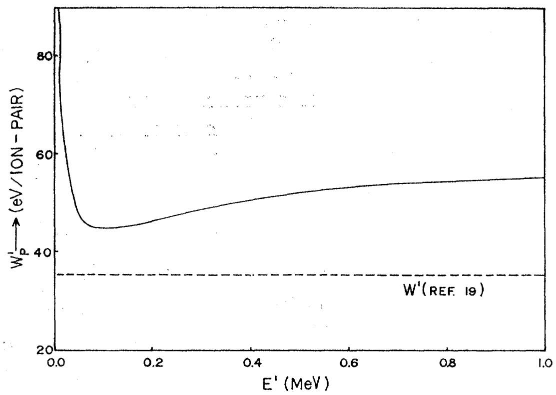

FIG. 1. W_p' is the energy loss per primary ion pair for protons in H_2 as a function of proton energy E'.

The values of W_p' calculated from s_i' are much higher than the experimental values of W', the average energy expended per ion pair for protons in H_2. Bakker and Segré[19] found W' for 340-MeV protons in hydrogen gas equal 35.3 eV/ion pair. Gerthsen's[20] experimental values of W' for low-energy protons (27 to 45 keV) in H_2 are similar to those in air, i.e., approximately 36 eV/ion pair. The difference between W_p' and W' arises from the ionization produced by the secondary electrons knocked by the protons. This ionization is not considered in s_i', or W_p', while W' includes the total ionization, both primary and secondary, produced in the passage of the proton through the gas. The use of W_p' in Eq. (8) will yield a high limit for w_{ff}, as will be seen later.

IV. THE ENERGY LOSS CROSS SECTIONS: b^e AND b^n

Theoretical expressions for the electronic stopping power $(-dE/dx)_e$ of heavy charged particles have been found to underestimate measured energy losses.[21] A semiempirical use of one of these theoretical expressions

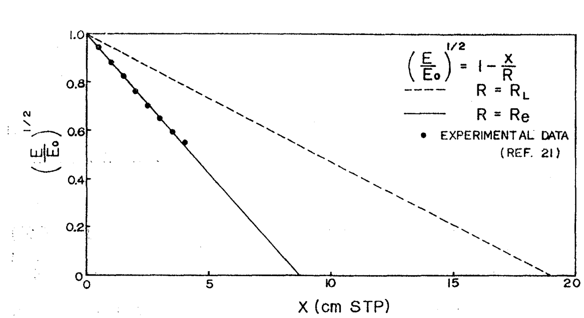

FIG. 2. Energy loss as a function of thickness of H_2 gas traversed, for the average energy Cf^{252} fission fragment. R_L is the electronic range of the fission fragment as predicted by the Lindhard theory. R_e is the extrapolated electronic range.

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together with experimental data in the 30–100-MeV energy range may be employed to obtain values for the cross sections.[22]

Lindhard *et al.*[23] have suggested that the electronic stopping power is proportional to the velocity of the fission fragment in the velocity range between zero and v_1, where v_1 is equal to $v_0Z^{2/3}$. A similar deduction was made from Bohr's classical expression in the case of heavy stopping materials.[24–26] Consequently,

$$(-dE/dx)_e = kE^{1/2}. \quad (10)$$

Integrating and defining an electronic range as $R_e = 2E_0^{1/2}/k$, the energy distance relation is given by

$$E/E_0 = (1 - x/R_e)^2. \quad (11)$$

This equation is applicable only in the range of energies where nuclear stopping is negligible. In Fig. 2, a plot of the experimental values of $(E/E_0)^{1/2}$ as a function of x,

TABLE I. Energy loss cross sections and high limit of w_{ff} as a function of the energy of the average energy Cf^{252} fission fragment.

E (MeV)	b^e 10^{-19} (MeV cm²)	b^n 10^{-19} (MeV cm²)	w^e/w_{ff} [a]
0.5	0.280	0.415	0.460
1.0	0.355	0.403	0.650
2.0	0.570	0.270	0.769
3.0	0.700	0.240	0.833
5.0	0.904	0.180	0.909
7.5	1.108	0.139	0.939
10.0	1.275	0.113	0.955
17.5	1.666	0.0775	0.975
25.0	2.000	0.0600	0.981
50.0	2.830	0.0355	0.991
75.0	3.460	0.0258	0.993
90.0	3.815	0.0225	0.996

[a] w^e equals 35.3 eV/ion pair.

in cm of H_2 at STP, yields an extrapolated value of R_e equal to 8.75 cm (STP). The initial energy of the average energy Cf^{252} fission fragment is taken to be equal to the arithmetic average of the initial energies of the light and heavy Cf^{252} fission fragments measured by the time-of-flight experiment of Fraser and Milton[27] and corrected for prompt neutron emission. In the same figure, the straight line corresponding to the Lindhard prediction[23] is shown.

Similarly, the nuclear stopping power $(-dE/dx)_n$ is obtained from a universal curve calculated by Lindhard

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et al.[23] They report good agreement with experimental-range data for low-energy heavy particles in light-mass stopping media.

A numerical integration is made of the total stopping power, that is, the sum of $(-dE/dx)_e$ and $(-dE/dx)_n$. The electronic power expression used is that of Eq. (10), where k is calculated from the extrapolated electronic range obtained from the experimental data, i.e., 8.75 cm. In Fig. 3, the experimental data points and the calculated energy-distance curve are shown. Although the experimental points correspond to total energy losses and were used to obtain the extrapolated R_e and k of Eq. (7), the fitting is good because nuclear stopping (b^n) is negligible compared to electronic stopping (b^e) in the range above 30 MeV. The total range obtained from the numerical integration of the sum of $(-dE/dx)_e$ and $(-dE/dx)_n$ is found to equal 7.6 cm (STP) which appears reasonable by comparison with other fission fragment ranges although no explicit data on the range of Cf^{252} fragments in hydrogen apparently exist.

Since the electronic and nuclear stopping powers are proportional to their respective energy stopping cross sections, values of b^e and b^n are obtained as a function of energy from the numerical integration described above. These are tabulated in Table I and plotted in Fig. 4.

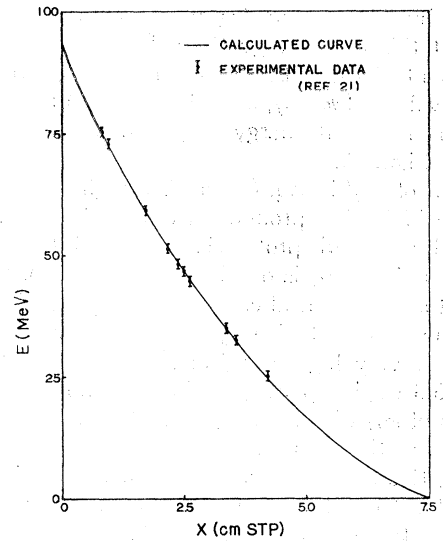

FIG. 3. Numerical integration of total stopping power. The parameters used are: for the incident fission fragment, $M=126$, $Z=49$, $E_0=91.4$ MeV, and for the stopping medium, hydrogen, $M'=1$, $Z'=1$.

V. CROSS SECTION FOR THE PRODUCTION OF RECOIL ATOMS, $S(E,E')$

The integral of Eq. (8) contains $S(E,E')$, the cross section for the production of recoil atoms of energy E' by an incident particle of energy E. This cross section is extracted from an elastic-scattering analysis by Lindhard *et al.*[23] that is based on a Thomas-Fermi potential. Their expression for the nuclear stopping power in terms of a dimensionless energy ϵ and a dimensionless range ρ is

$$\left(\frac{d\epsilon}{d\rho}\right)_m=\int_0^\epsilon \frac{f(t^{1/2})}{\epsilon}d(t^{1/2})\,, \qquad (12)$$

where

$$\epsilon=\frac{EaM'}{ZZ'e^2(M+M')}\,,\quad \rho=\frac{4\pi a^2RNM'M}{(M+M')^2}\,. \qquad (13)$$

The parameter t is equal to $\epsilon^2(E'/E_m')$ and a is equal to $0.8853a_0(Z^{2/3}+Z'^{2/3})^{-1/2}$. In these expressions, e is the electron unit charge, R is the range of a particle of energy E, a_0 is the hydrogen Bohr radius, M and Z are the atomic mass and atomic number of the incident particle, while M' and Z' are those of the stopping medium. The function $f(t^{1/2})$ is given in their Fig. 1, where it is observed that at high values of $t^{1/2}$, the differential scattering cross section joins smoothly that given by the Rutherford expression for elastic scattering, i.e., $f(t^{1/2})$ equals $1/(2t^{1/2})$. By algebraic manipulation the required cross section is found to be

$$S(E,E')=\frac{\pi a^2 f(t^{1/2})}{2t^{3/2}}\frac{\epsilon^2}{E_m'}\,. \qquad (14)$$

The transition energy E_t' occurs approximately at $t^{1/2}=7$, that is, $E_t'=49E_m'/\epsilon^2$. Using Eq. (14) and knowing the form of the function $f(t^{1/2})$ in the different ranges of E', the values of $S(E,E')$ are calculated as a function of E' for a given E in the incident particle energy range between 0.5 and 90.0 MeV.

VI. RESULTS AND DISCUSSION

The value of w^e used throughout the calculation of w^e/w_{ff} is approximated to be equal to the value of W'

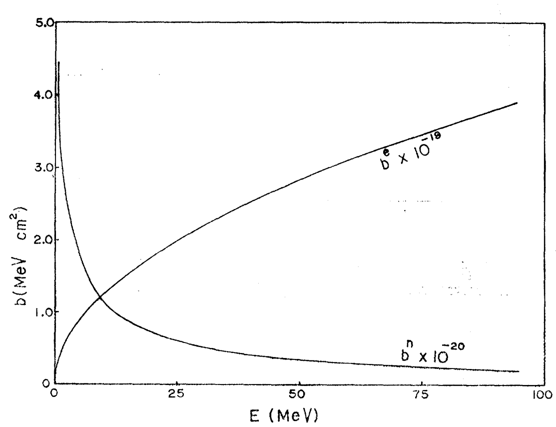

FIG. 4. Electronic and nuclear stopping cross sections for the average energy Cf^{252} fission fragment in H_2 as a function of the fragment energy.

obtained by Bakker and Segrè[19] for 340-MeV protons in H_2. At this high energy the value of w^e should be independent of the nature of the incident particle. In Table I, values of w^e/w_{ff} are tabulated as a function of the fission fragment energy. A plot of these values is shown in Figure 5.

The use of W_p' in Eq. (8) yields a high limit for w_{ff} since the ionization produced by secondary electrons knocked by energetic protons is not taken into account. The low limit of w_{ff} is obtained when it is considered that protons will expend energy to produce an ion pair during the whole of their trajectory at the same rate as they do at very high energies; e.g., at 340 MeV, W' equals 35.3 eV/ion pair. In order to obtain the exact variation of w_{ff} with the fragment energy, the value of W' as a function of proton energy is needed. Unfortunately, this is not known. Gerthsen's[20] experimental values of W' can be compared to those of Jesse and Sadauskis[6] for α particles. They observed that above 1 MeV there was no variation of W_α in H_2 but did observe a variation in air. Since α particles and protons are very similar, it seems doubtful that at the low energies used by Gerthsen, 20 times less than the ones of Jesse and Sadauskis, the value of W' should still be constant.

The constancy of w_{ff} in the high-energy range is an expected result since energy losses by elastic collisions with the screened nuclei of the gas atoms are negligible. Consequently, almost all of the energy is lost in inelastic collisions with the electrons. In this energy range, as was assumed in the derivation of Eq. (8), the rate of energy loss to electrons is closely proportional to the rate of primary ionization including that produced by ejected electrons. This is consistent with Schmitt and Leachman's observations[7] in other gases.

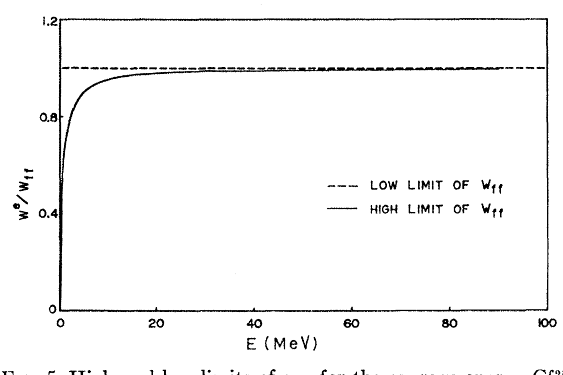

FIG. 5. High and low limits of w_{ff} for the average energy Cf^{252} fission fragment in H_2 as a function of the fragment energy.

Jesse[8] finds that for α particles in H_2 and the noble gases W_α/W_e is a constant in the α-energy range between 1 and 9 MeV. W_e is the constant value observed for electrons in the energy range of 3 to 50 keV. This implies that W_α is a constant very nearly equal to w_α, that is, I_α is proportional to E_α. In terms of Fig. 5, a plot of w^e/w_α would yield a horizontal line at unity with a very sharp break towards the origin at very low energies (less than 1 MeV). Unfortunately, the theory developed by Lindhard and co-workers does not apply to those cases where low-mass particles are stopped by low-mass media,[23] and this does not allow a similar calculation for the system measured by Jesse.

The observed ionization defect[28] in the determination of fission fragment energies with ionization chambers is a result of the increase in w_{ff}. If the ionization defect is defined as $D=(E-w^eI)$, then from the calculated values of w^e/w_{ff}, D can be obtained since

$$D=\int_0^{E_0}(1-w^e/w_{ff})dE. \qquad (15)$$

If $(1-w^e/w_{ff})$ is plotted as a function of E and the area under the curve measured, then D is found to equal 2.75 MeV in the case of the high limit of w_{ff} and zero in the low limit. The ionization defect for fission fragments in H_2 has not been measured, but Schmitt and Leachman[7] obtained experimental D's for N_2, Ne, and Ar between 4.3 and 6.5 MeV.

In summary, it is possible to calculate the ionization efficiency of heavy charged particles such as fission fragments, if the total ionization cross section of the stopping-medium knocked atoms and energy-distance curves of the incident particles are known. All the available evidence indicates that the energy dependence of these efficiencies is different from that of low-mass charged particles, e.g., α particles.

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