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MEAN EXCITATION ENERGIES CALCULATED FROM
STOPPING-POWER AND RANGE DATA

(Thesis)

Patricia Dalton
J.E. Turner

Submitted as a thesis by Patricia Dalton to the Graduate School of Vanderbilt University in partial fulfillment of the requirements for the degree of Master of Science.

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FROM STOPPING-POWER AND RANGE DATAPatricia Dalton
and
J. E. Turner

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ABSTRACT

Mean excitation energies (I-values) have been calculated from the data of nine key experiments that measured absolute stopping power, relative stopping power, or range. The experiments reviewed are those of Bakker and Segrè (1951); Thompson (1952); Sachs and Richardson (1953); Brolley and Ribe (1955); Burkig and MacKenzie (1957); Zrellov and Stoletov (1959); Nielsen (1961); Barkas and von Friesen (1961); and Nakano, MacKenzie, and Bichsel (1963). The older data have often been analyzed with insufficient allowance for the contribution of shell corrections, and new shell corrections (C/Z) for the entire atom, as described by Fano,* have been included in this analysis. I-values obtained from relative stopping-power measurements have been normalized to $I = 163$ eV for aluminum and $I = 314$ eV for copper.

The results of the analysis indicate that 1) the absolute stopping-power data of Sachs and Richardson does not fit the general trend described by the other data;

* U. Fano, "Penetration of Protons, Alpha Particles, and Mesons." Annual Reviews of Nuclear Science, E. Segrè, Ed., Vol. 13, p. 1. Annual Reviews, Inc., Palo Alto (1963).

2) a standard reference on stopping power, National Bureau of Standards Handbook 79, Stopping Powers for Use with Cavity Chambers (1961), which does not include the more recent data, does not include adequate shell corrections; 3) agreement is good with a recent report of the National Academy of Sciences - National Research Council, Studies in the Penetration of Charged Particles in Matter (1964), which includes suggested I-values based on empirical estimates of the shell corrections; and 4) a good estimate of I (eV) can be found from the equations

$$I = (10.2 \pm 2.07) + (11.8 \pm 0.55)Z, \quad \text{for } Z \leq 13$$

$$I = (56.1 \pm 8.76) + (8.61 \pm 0.37)Z, \quad \text{for } Z > 13$$

I. INTRODUCTION

A. The Stopping Power Formula

A heavy charged particle slows down in matter in a series of inelastic electronic collisions that produce ionization and excitation of the atoms in the absorbing medium. Bohr^{1,2} described the collisions by an impact parameter, the incident ion's distance of closest approach to the target particle. In a quantum-mechanical analysis Bethe^{3,4} characterized the collisions in terms of the momentum transferred. Assuming that the ion is moving much faster than atomic electrons and using the first Born approximation, Bethe calculated the differential cross section, $d\sigma_n$, for the inelastic scattering of an ion from an initial to a final momentum eigenstate with simultaneous excitation of an atom from its ground state to an excited state, n . He found σ_n , the total cross section for excitation to the state n , by integrating $d\sigma_n$ over all scattering angles. The product of σ_n and the excitation energy, E_n , summed over all possible final states of the atom, is the average energy lost per collision, $\sum_n E_n \sigma_n$. When multiplied by the number of atoms per unit volume, N ,

this quantity gives dE/ds , the average linear rate of energy lost by the ion in a homogeneous medium, that is, the absorber's stopping power.

$$-\frac{dE}{ds} = N \sum_n E_n \sigma_n \quad (1)$$

Bethe's explicit evaluation of the quantity on the right in equation (1) is

$$-\frac{dE}{ds} = \frac{4\pi z^2 e^4}{mv^2} NZ \left\{ \ln \frac{2mv^2}{I(1-\beta^2)} - \beta^2 \right\} \quad (2)$$

Here ze is the charge on the incident heavy ion which has a velocity v . Equations (1) and (2) imply that the medium traversed is an element and that all the atoms are alike; in a compound or mixture NZ represents the number of electrons per unit volume. In equation (2), m is the electron rest mass, β is the ratio of the particle velocity to the velocity of light, and I is the mean excitation energy of the absorber. The quantity in brackets is often called the absorber's stopping number, B .

The quantity I in the stopping power formula is defined by equation (3).

$$\ln I \equiv \sum_n f_n \ln E_n \quad (3)$$

where the optical dipole oscillator strength, f_n , gives a measure of the probability of excitation of an atom from its ground state to the energy level E_n . I represents a weighted average of the energy transferred to individual

atoms in collisions.

The Bethe theory requires that $v \gg v_e$, that is, the velocity of the incident particle must be greater than the velocity of the orbital electrons in the stopping material. However, inner electrons in intermediate- and high-Z elements have speeds near that of light, and the condition $v \gg v_e$ cannot always be satisfied. The requirement also may not be met when low-energy particles slow down in low-Z materials. For this reason a shell-correction term, C/Z , a function of the incident particle energy and the atomic number of the absorber, is included in the stopping power formula. Equation (2) is then written

$$-\frac{dE}{ds} = \frac{4\pi z^2 e^4}{mv^2} NZ \left\{ \ln \frac{2mv^2}{I(1-\beta^2)} - \beta^2 - \frac{C}{Z} \right\} \quad (4)$$

(The notation here is explained above and is the same as that used in the review article by Fano⁵).

B. Discussion of I-values

Knowledge of stopping powers is important both theoretically and practically. For example, the absorbed dose from charged particles (energy deposited per unit absorber mass) is proportional to the stopping power of the absorber. The amount of energy deposited in an ionization chamber or shielding material also depends upon stopping power. Data on the variation of stopping power

and range with particle energy are often used to determine energies and masses in nuclear cross-section measurements. A heavy charged ion's stopping power is related to the absorbing medium by the parameters NZ and I .

To calculate stopping power, the mean excitation energy of the absorber must be known. On the basis of the Thomas-Fermi statistical model of the atom, Bloch⁶ showed that $I = kZ$, where k is about 15 eV for low- Z absorbers and about 10 eV for high- Z elements. In principle I could be obtained from equation (3). However, most elements have excitation energies (E_n) in the range 10 to 1000 eV, and in this range the oscillator strengths (f_n) are poorly known. I -values have been calculated^{3,4,7-11} only for the elements through beryllium.

For most elements information about I is provided by experimental data on stopping power or range. As can be seen from equation (4), these measurements yield determinations of the quantity $\ln I + C/Z$. In analyzing the data to obtain I , one must say how much each of the terms, $\ln I$ and C/Z , contributes to the stopping power. Similarly, an uncertainty in C/Z leads to uncertainty in the value of I . As a result, a variety of I -values for many elements has been published over the past 15 years, even when stopping power measurements were consistent.

An analysis of the data from key experiments would be useful for two reasons.

Often stopping powers are found by comparison to the stopping power of aluminum, and hence the value of I found for the measured material depends upon the I -value chosen for aluminum. Since about 1960 a value of 163 eV has become well established for the mean excitation energy of aluminum, and it would be convenient to have a table of I -values corrected to $I = 163$ eV for aluminum.

The magnitude of the shell-correction term has most often been underestimated, and occasionally it has been ignored altogether. Discrepancies between the theory and the data indicate that shell corrections are not negligible. It would also be useful to have a set of I -values based on the most recent determinations of the shell corrections.

The most extensively used corrections have been those of Walske^{12,13} who used hydrogenic wave functions to calculate the separate contributions of K- and L-shell electrons to the stopping power. Khandelwal and Merzbacher¹⁴ extended this method to M-shell electrons. However, these corrections do not treat all electrons in heavy elements. The use of hydrogenic wave functions for M-shell electrons may be questionable since, due to screening of the nucleus by inner shell electrons, the M-shell electrons do not experience a $1/r$ potential.

A new method for obtaining C/Z corrections for the

entire atom has been outlined by Fano.⁵ This method extends the evaluation of the differential cross section beyond Bethe's dipole approximation and also includes the effect of collisions that transfer more energy than $2mv^2$, the upper limit assumed by Bethe. It is shown in reference 15, where the formula (5) for C/Z is derived, that these two contributions are equal. The expression

$$\frac{C}{Z} \sim 2 \left[\frac{m \langle v^2 \rangle_0}{2mv^2} + \frac{m^2 \langle v^4 \rangle_0 + \frac{10\pi}{3} \frac{\hbar^2 e^2}{m} \langle \rho(0) \rangle_0}{(2mv^2)^2} \right] \quad (5)$$

approximates the total shell correction for an atom to the order $1/v^4$ when electron correlations are neglected. The quantity $\langle v^2 \rangle_0$ is the mean squared velocity of an atomic electron in the ground state of the atom, averaged over all electrons in the atom. The number $\hbar$ is $h/2\pi$, where h is Planck's constant. The mean electron density at the center of the atom is given by $\langle \rho(0) \rangle_0$. This expression for the shell correction is independent of the atomic model used in the calculation and depends only on the ground-state properties of the atom. It is also shown in reference 15 that to the order $1/v^2$ the electron correlation contributes less than 3% to the shell correction. These shell corrections have been included in the analysis reported in this paper.

C. Other Corrections to the Penetration of Charged Particles

Use of the dipole oscillator strength in equation (3) implies that the positions of electrons are not correlated over distances greater than $\hbar/g$, where g is the average momentum transferred in a collision.⁵ While this assumption is valid for low-density gases, often it is not true for condensed media. Fermi¹⁶ corrected the stopping power formula to include this "density effect" (δ) on the slowing down of a heavy charged particle. When the density effect is included, the stopping number, B , is written

$$B = \ln \frac{2mv^2}{I(1-\beta^2)} - \beta^2 - \frac{C}{Z} - \frac{1}{2}\delta \quad (6)$$

Straggling and multiple scattering also characterize the penetration of heavy charged particles. Because the number and kinds of collisions in which a charged particle slows down in matter are described by statistical distributions, there is a spread in both the path lengths of particles and their energies at a given depth when the particles are stopping under the same experimental conditions. This is straggling. Corrections for multiple scattering compensate for the irregular path of a charged particle, for the depth of penetration of the particle in a stopping medium will be shorter than the particle's total path length. At particle velocities comparable to

those of outer electrons in the absorber atoms, charge exchange occurs and the stopping power theory is no longer valid.

II. TYPES OF EXPERIMENTS AND DATA

A. Description of Data

Several different kinds of experiments yield information about mean excitation energies. The experiments can be classified according to the parameter measured: stopping power relative to a reference absorber, absolute stopping power, or range.

1. Relative Stopping-Power Measurements

In relative stopping-power measurements, the thickness of the test absorber is found that will produce the same energy loss as will a given thickness of the reference absorber for incident particles of equal energy. The ratio of these thicknesses is the relative stopping power.

Equation (4) for the stopping power can be rewritten

$$\left[\frac{dE}{ds} \right]_R = 0.3071 \frac{Z^2 Z_R}{\beta^2 A_R} \left\{ \ln \frac{1.022 \times 10^6 \beta^2}{1 - \beta^2} - \beta^2 - \ln I_R - (C/Z)_R \right\} \quad (7)$$

The subscripts "R" refer to the standard absorber, and the density correction has been ignored. If the stopping power of the test absorber is given by a similar equation with the subscripts "R" replaced by "X", then the relative

stopping power, S , is given by

$$S = \frac{(dE/ds)_x}{(dE/ds)_R} = \frac{Z_x A_R}{Z_R A_x} \left\{ \frac{f(\beta^2) - \ln I_x - (C/Z)_x}{f(\beta^2) - \ln I_R - (C/Z)_R} \right\} \quad (8)$$

where

$$f(\beta^2) = \frac{1.022 \times 10^6 \beta^2}{1 - \beta^2} - \beta^2.$$

The measured quantity is S , and the mean excitation energy of the test absorber, I_x , can be found from equation (8).

The shell corrections, C/Z , must be known for both the reference and test absorbers, and of course, I_R must be known for the reference absorber.

2. Measurements of Absolute Stopping Power

A measurement of absolute stopping power, which is a direct determination of the energy loss of charged particles in an absorber, presents greater experimental difficulties than the measurement of relative stopping powers. The I -value of the absorbing material can be calculated from the absolute value of the stopping power directly by equations (4) or (7).

3. Range Measurements

The range of a charged particle is related to stopping power and I by equation (9).

$$R = \int_0^E \frac{I}{(-dE/ds)} dE \quad (9)$$

This equation gives the "c.s.d.a. range".⁵

Because the stopping power formula does not hold at very low particle velocities, equation (9) cannot be used directly to calculate I-values. The range equation must be rewritten

$$R = R_0(E_0) + \int_{E_0}^E \frac{I}{(-dE/ds)} dE \quad (10)$$

where $R_0(E_0)$ is the measured range of particles of energy E_0 .

To determine I from a range measurement, one estimates the probable I-value of the absorber and numerically integrates the reciprocal of the stopping power formula over the energy interval E_0 to E . A measured value of $R_0(E_0)$ is added to the result of the numerical integration to obtain the total range. The process of estimating I-values and numerically integrating the reciprocal stopping power formula is repeated until the value of R obtained by equation (10) matches the experimentally measured range.

B. Description of Analysis

The review that follows includes five experiments that measured relative stopping powers, two that measured absolute stopping powers, and two that measured range.

Most measurements of relative stopping power are made with respect to aluminum or copper. Because the measure-

ments are relative, the I-value found for a test absorber depends upon the I-value chosen for the reference absorber. The experiments in this review have been normalized to $I = 163$ eV for aluminum and $I = 314$ eV for copper.¹⁷

The new shell corrections, as described by Fano and Turner^{5,15} and tabulated by Marusak and Turner,¹⁸ have been included in the analysis of each experiment. Corrections for density, multiple scattering, and straggling are usually small and were often included in the published data. Unlike the shell corrections, these corrections are well established, and they are included in the work reported here without any additional attempt to verify them.

Before correcting I-values, the experimenters' results were verified according to the methods published in their reports to insure that the recalculated I-values do not differ from the published ones simply because of an arithmetic error. Thus the new I-values reported here reflect 1) a normalization of relative stopping-power measurements to $I = 163$ eV for aluminum or $I = 314$ eV for copper, and 2) the effect of the new shell corrections.

In their report the National Academy of Sciences - National Research Council Subcommittee on Penetration of Charged Particles¹⁹ has presented a comprehensive review of stopping-power theory and data. For each experiment reviewed in this thesis, the I-values have been compared

to those indicated for the data by the calculated tables
in the NAS-NRC report.²⁰

III. ANALYSIS OF NINE EXPERIMENTS

A. Bakker and Segrè

In 1950 Bakker and Segrè²¹ measured the mass stopping power (MeV/g/cm^2) of 340-MeV protons in 11 elements relative to copper. They found the range of these protons was 93.7 g/cm^2 of copper then replaced about 30 g/cm^2 of the copper with a material of unknown stopping power and redetermined range. The range of the unknown-plus-copper absorber was compared to that of the pure copper absorber to find the mass stopping power of the unknown relative to copper.

Bakker and Segrè reported mass stopping powers, relative to copper and converted to aluminum, and also stopping powers per electron relative to aluminum. The I-values they calculated from their data were based on $I = 150$ eV for aluminum. They made a small correction for nuclear scattering but omitted shell corrections in their calculation of I.

This analysis has treated Bakker and Segrè's values of stopping powers per electron relative to aluminum as the "raw data." Bakker and Segrè's results were corrected by using 163 eV instead of 150 eV for the I-value of aluminum,

TABLE 1. Calculation of I-values from Relative Stopping-Power Data of Bakker and Segrè for 340-MeV Protons

Element	Stopping power per electron, relative to aluminum*	I-value reported by Bakker and Segrè (eV)	I-value indicated by NAS-NRC report ²⁰ (eV)	New shell correction, C/Z ¹⁸	New value of I (eV)	New value of I/Z
H ₂ in CH ₂	1.280	15.6	17.0	0	17.5	17.5
3-Li	1.184	34.0	36.9	0	37.4	12.5
4-Be	1.113	60.4	65.9	0	66.6	16.7
6-C	1.084	76.4	82.6	0	83.5	13.9
13-Al	(1.000)	(150)	(163)	0.003	(163)	(12.5)
26-Fe	0.941	243	269	0.010	266	10.2
29-Cu	0.924	279	301	0.013	297	10.2
47-Ag	0.873	428	457	0.028	444	9.4
50-Sn	0.859	479	507	0.030	491	9.8
74-W	0.814	697	721	0.056	687	9.3
82-Pb	0.804	758	785	0.066	742	9.0
92-U	0.786	881	900	0.078	847	9.2

* Column 4, Table I, of Reference 21

an increase of 8%. If shell corrections are ignored, this change increases I for other elements by an average of 7.3%, ranging from 6% for uranium to 9.8% for H_2 in CH_2 .

The inclusion of shell corrections increases the I -values of H_2 , Li, Be, and C by less than 1%. For elements with atomic number greater than 13, I -values are reduced by about 1% for Fe and Cu and by nearly 8% for U.

Table 1 is a summary of the reanalyzed data.

B. Burkig and MacKenzie

Burkig and MacKenzie²² in 1957 reported the stopping powers of 23 metals relative to aluminum. An aluminum foil and a test foil were exposed to a collimated beam of 19.8-MeV protons, and the position and angle of each foil were adjusted so that the protons emerging from the foils had the same velocity. Additional aluminum behind the targets brought the protons to rest. The ionization chamber used for detection was thus operated in the region of the Bragg peak where the system was at maximum sensitivity. Their comparison measurements yielded stopping powers per electron relative to aluminum.

Using $I = 166.4$ eV for aluminum and incorporating Walske's shell corrections, Burkig and MacKenzie calculated I -values for Be, Fe, Cu, Ag, W, Au, and Pb. They found the ratio I/Z to be about 12.6.

TABLE 2. Calculation of I-values from Relative Stopping-Power Data of Burkig and MacKenzie for 19.8-MeV Protons

Element	Stopping power per electron relative to aluminum*	Value of I reported by Burkig and MacKenzie	Value of I indicated by NAS-NRC report ²⁰	New shell correction, C/Z^{18}	New value of I	New value of I/Z
		(eV)	(eV)		(eV)	
4-Be	1.166	64.0	67.5	0.003	67.8	17.0
13-Al	(1.000)	(166.4)	(163)	0.0404	(163)	(12.5)
20-Ca	0.970	---	185	0.0704	187	9.3
22-Ti	0.936	---	221	0.0788	223	10.1
23-V	0.918	---	243	0.0834	245	10.7
26-Fe	0.886	328.8	286	0.0938	289.5	11.1
28-Ni	0.872	---	307	0.0988	311	11.1
29-Cu	0.867	366.0	315	0.1028	318.6	11.0
30-Zn	0.866	---	316	0.1058	319	10.6
41-Nb	0.816	---	405	0.1378	407	9.9
42-Mo	0.809	---	418	0.1404	422	10.1
45-Rh	0.800	---	437	0.1488	440	9.8
46-Pd	0.793	---	451	0.1518	456	9.9
47-Ag	0.791	587.0	455	0.1548	459.6	9.8
48-Cd	0.789	---	459	0.1568	464	9.7
49-In	0.782	---	475	0.1598	481	9.8
50-Sn	0.782	---	475	0.1618	480	9.6
73-Ta	0.716	---	643	0.2308	644	8.8
74-W	0.706	920.5	667	0.2342	678.0	9.2
77-Ir	0.703	---	676	0.2448	682	8.9
78-Pt	0.695	---	698	0.2478	711	9.1
79-Au	0.692	997	707	0.2518	719.6	9.1
82-Pb	0.677	1070	752	0.2618	774	9.4
90-Th	0.691	---	710	0.2872	698	7.7

* Column 6, Table I, of Reference 22

Although Burkig and MacKenzie did use shell corrections, they were smaller than the C/Z corrections described by Fano and Turner.^{5,15} At proton energies as low as 20 MeV, shell corrections contribute significantly to the stopping power of high-Z elements and are important even for low-Z elements. I-values for W, Au, and Pb are nearly 30% lower than Burkig and MacKenzie's when the new shell corrections are used in calculating I. Renormalization of I for aluminum accounts for about 2% of the decrease in I-values. The new I-values are listed in Table 2.

C. Nakano, MacKenzie, and Bichsel

Nakano, MacKenzie, and Bichsel²³ used a method similar to Burkig and MacKenzie's²² and measured the stopping power of 28.7-MeV protons. The protons lost about 2 MeV in thin foils of Be, Ti, V, Co, Ni, Ag, Ta, W, Ir, Au, and a comparison foil of aluminum.

Nakano, MacKenzie, and Bichsel reported mass stopping powers and stopping powers per electron relative to aluminum but did not report any estimates of I for the elements they studied. I-values have been calculated here from their data, with and without shell corrections. For Ti, V, Co, Ni, and Cu, the value of I is 4-6% lower when the new shell corrections are included; I for Ag is about 10% lower; and the I-values of Ta, W, Ir, and Au are 16-18% lower than the

TABLE 3. Calculation of I-values from Relative Stopping-Power Data of Nakano, MacKenzie, and Bichsel for 28.7-MeV Protons

Element	Stopping power per electron, relative to aluminum*	Value of I indicated by NAS-NRC report ²⁰ (eV)	New shell correction, C/Z ¹⁸	New value of I (eV)	New value of I/Z
4-Be	1.1840	57.4	0.002	56.9	14.2
13-Al	(1.0000)	(163)	0.0302	(163)	(12.5)
22-Ti	0.9409	227	0.0613	224	10.2
23-V	0.9185	258	0.0646	254	11.0
27-Co	0.8942	294	0.0766	290	10.7
28-Ni	0.8780	321	0.0796	318	11.4
29-Cu	0.8692	336	0.0816	334	11.5
47-Ag	0.7923	500	0.1276	501	10.7
73-Ta	0.7143	718	0.1979	739	10.1
74-W	0.7023	794	0.2009	790	10.7
77-Ir	0.7035	752	0.2102	777	10.1
79-Au	0.7014	759	0.2172	782	9.9

* Column 4, Table I of Reference 23

uncorrected I-values. The corrected I-values agree to within 3% with those indicated for the data by the NAS-NRC report.²⁰ A summary of the calculations appears in Table 3.

D. Brolley and Ribe

Brolley and Ribe's²⁴ experiment in 1955 was one of the few to determine stopping powers of heavy charged particles in gases. They measured the stopping power of 8.86-MeV deuterons in H₂, He, N₂, O₂, Ne, Ar, Kr, Xe, CH₄, and CO₂ relative to air, and they also found absolute stopping powers for 4.43-MeV protons in H₂, air, and Kr. Scattered protons, or deuterons of the same velocity, were directed down the length of a chamber filled with the stopping gas. In the deuteron experiment they observed the relative amounts of air and the test gas required to give the same energy loss to deuterons having an incident energy of about 10 MeV. When they measured absolute stopping powers, the energy lost by protons in the chamber was observed; the energy lost was slightly more than 1 MeV.

Brolley and Ribe report their raw data and the stopping cross sections they obtained from these data. With the use of their values of dE/ds , determined from both the relative and absolute measurements, and including the new shell corrections, I-values have been calculated as shown in Table 4. The experiment of Brolley and Ribe is valuable

TABLE 4. Calculation of I-values from Data* of Brolley and Ribe

Element	Experimental stopping power†	dE/ds	Value of I indicated by NAS-NRC report ²⁰	New shell correction, C/Z ¹⁸	New value of I	New value of I/Z
	(eV-cm ² x 10 ⁻¹⁵)	(MeV/g/cm ²)	(eV)		(eV)	
1-H	0.335	200.24	22.5	0	20.0	20.0
2-He	0.582	87.605	52.1	0.0042	44.3	22.1
6-C	1.58	79.254	82.3	0.03856	72.6	12.1
7-N	1.76	75.706	99.4	0.04864	90.2	12.9
Air	1.82	74.797	---	0.04978	94.3	12.9
8-O	1.98	74.560	106	0.0508	98.3	12.3
10-Ne	2.29	68.359	141	0.07084	132	13.2
18-Ar	3.72	56.104	201	0.12385	189	10.5
36-Kr	6.10	43.984	368	0.202	350	9.7
54-Xe	8.06	36.984	520	0.2442	480	8.9

* Absolute stopping-power measurements were made with 4.43-MeV protons; stopping-power measurements relative to air were made with 8.86-MeV deuterons.

† Column 2, Table III of Reference 24

because it deals with gaseous elements, for which few other data exist, and also because the measurements were made with low-energy particles so that shell corrections are an important factor in the stopping power. Shell corrections reduce carbon's I-value by nearly 4% and xenon's by 22% below the uncorrected values.

The values of I reported here are consistently lower than those given by the NAS-NRC²⁰ for these stopping-power data, from 5% for Kr to 16% for He; however, they do not vary greatly from the trend of I-values established by the other experiments that have been reviewed here.

E. Thompson

Thompson²⁵ in 1952 found stopping powers for high-energy protons in H, C, N, and O relative to copper. He also verified that the stopping power, S_{cpd} , of a compound A_aB_b is given by $aS_A + bS_B$, and from his measurements of the stopping powers of chlorine compounds, he inferred the stopping power of chlorine.

Thompson's method was similar in principle to that of Bakker and Segrè.²¹ He obtained the range of a standard copper absorber and of an absorber in which the test material replaced part of the copper. The targets were chosen to reduce the energy of the proton beams from 340 MeV to 200 MeV. The ionization loss of the proton beam in

TABLE 5. Calculation of I-values from Thompson's Relative Stopping-Power Data for 270-MeV Protons

Element or compound	Stopping power relative to copper*	Thompson's value of I	Value of I indicated by NAS-NRC report ²⁰	New shell correction, C/Z ¹⁸	New value of I [†]	New value of I/Z
		(eV)	(eV)		(eV)	
1-H ₂	0.0472	18.2	22.0	0	21.9	21.9
6-C	0.2455	70.2	81.6	0	82.2	13.7
7-N ₂	0.2837	76.3	88.5	0.001	89.1	12.7
8-O ₂	0.3188	88.3	102.2	0	102.8	12.9
H-saturated	0.04797	15.5	18.5	0	18.6	18.6
H-unsaturated	0.04879	13.0	15.6	0	15.7	15.7
C-saturated	0.24627	69.3	79.4	0	80.0	13.3
C-unsaturated	0.24674	67.2	78.1	0	78.7	13.1
C-highly chlorinated	0.2509	57.9	67.6	0	68.0	11.3
N-amines, nitrates, etc.	0.2785	89.4	103.6	0.001	104.1	14.9
N-in ring	0.2870	68.8	80.1	0.001	80.6	11.5
=O ⁻	0.3187	88.5	102.4	0	103.1	12.9
O	0.3226	79.8	92.4	0	93.1	11.6
17-Cl-all	0.6335	153.7	175.7	0.004	176.5	10.4

* Tables I and II of Reference 25

† Based on I = 314 eV for copper

the test substances was compared to that of the protons in the standard copper absorber.

Thompson reported stopping powers and values of I for H, C, N, O, and Cl as elements and in compounds. He based his calculations of I on Bakker and Segrè's I -value for copper, 279 eV. Thompson did not include shell corrections in his computations.

Correcting Thompson's data with $I = 314$ eV for copper results in an increase of 18% in I for H, a 15% increase in I for C, N, and O, and a 14% increase in I for Cl.

Since Thompson's data are for 270-MeV protons, shell corrections are not of great importance for low- Z elements. Their inclusion in the calculations raises I for H by 2%, I for C, N, and O by 1.5%, and I for Cl by 1% over the uncorrected I -values.

The new I -values, summarized in Table 5, agree to within less than 1% with the I -values indicated by the NAS-NRC report²⁰ for Thompson's stopping-power data.

F. Sachs and Richardson

In 1951 Sachs and Richardson²⁶ measured absolute stopping powers. A scattered beam of 18-MeV protons from a cyclotron described a circular path, passed through a metal foil, and was detected by an ionization chamber. The radial displacement in the distribution of protons was a measure of

the energy lost by the protons in the foils.

They published their energy-loss data for aluminum and their calculated I-values for Al, Ni, Cu, Rh, Ag, Cd, Sn, Ta, Au, and nylon. In 1953²⁷ they corrected a numerical error in their results and reported $I = 168$ eV for aluminum. They mentioned corrections for nonparticipating K electrons, but except for aluminum, they did not take these corrections into account when calculating I-values.

When the new shell corrections are included in the calculation of I from Sachs and Richardson's energy-loss data for aluminum, the corrected value of I for Al is 151 eV. This is 10% lower than their value, but the decrease is not entirely explained by the inclusion of shell corrections in calculating I. Even if shell corrections are ignored, their data for aluminum indicates a value no greater than $I = 160$ eV.

An analysis of Sachs and Richardson's data for metals other than aluminum is difficult because they published only their I-values and not their primary data. To correct their I-values, their measurements of dE/ds were inferred by a back-calculation based on their published I-values and the constants they reported using in the stopping-power formula. This was easily done because they did not include shell corrections in their calculation of I, except for aluminum. The new shell corrections were then included in the stopping-power formula and a new value of I obtained,

TABLE 6. Calculation of I-values from Absolute Stopping-Power Data of Sachs and Richardson for 17.8-MeV Protons

Element	Stopping power, dE/ds*	Value of I reported by Sachs and Richardson	Value of I indicated by NAS-NRC report ²⁰	New shell correction, C/Z ¹⁸	New value of I	New value of I/Z
	(MeV/g/cm ²)	(eV)	(eV)		(eV)	
13-Al	21.933	168	153	0.0444	151	11.6
28-Ni	18.051	399	358	0.1058	360	12.9
29-Cu	16.947	435	386	0.1088	391	13.5
45-Rh	14.025	799	641	0.1568	684	15.2
47-Ag	13.987	796	639	0.1628	678	14.4
48-Cd	13.727	792	637	0.1648	673	14.0
50-Sn	13.281	853	675	0.1698	721	14.4
73-Ta	11.722	1148	840	0.2402	904	12.4
79-Au	11.032	1383	(greater than 1000)	0.2608	1067	13.5

* For aluminum, dE/ds is reported in Table I of Reference 26; for other metals, dE/ds has been inferred from the I-values reported in Table III of Reference 27.

as shown in Table 6.

The corrected I-values obtained from Sachs and Richardson's data are much lower than those they listed; however, comparison of these corrected I-values with the results of other experiments indicates that Sachs and Richardson's data are not consistent with most other data. Some sources²⁸ have suggested that the discrepancies are due to multiple scattering that took place in their foils and for which Sachs and Richardson apparently did not correct their data. Whatever the reason for the disparity, these data do not fit the trend described by other experiments. For this reason, although the data are shown here for comparison, they have not been included in the average I-values listed in Table 11 in the concluding section of this thesis.

G. Nielsen

Nielsen²⁹ in 1961 measured the absolute stopping powers of protons and deuterons between 1 and 5 MeV in Be, Al, Ni, Cu, Ag, and Au. Particles from an accelerator passed through a metal foil and their distribution was recorded on a photographic emulsion. The spread in the distribution of these particles, as compared to the distribution of particles that did not pass through a foil, was a measure of the energy loss and straggling of the

TABLE 7. Calculation of I for Aluminum from Nielsen's Absolute Stopping-Power Data for Protons of 1 to 5 MeV Energy

Proton energy	Stopping power, dE/ds*	Nielsen's uncorrected† value of I	New shell correction, C/Z ¹⁸	New value of I	New value of I/Z
	(MeV/g/cm ²)	(eV)		(eV)	
1.59]	127.7]	135	0.1074	165.9	12.8
1.59]	126.7]		0.1074	169.8	13.1
1.99]	110.9]		0.108	165.5	12.7
2.03]	107.9]	184	0.108	171.5	13.2
2.06]	107.4]		0.108	165.6	12.7
2.50]	95.96]		0.107	156.5	12.0
2.48]	94.97]	180	0.107	162.1	12.5
2.52]	92.99]		0.1069	166.6	12.8
3.01]	82.56]		0.1039	165.8	12.7
3.03]	81.57]	180	0.1038	173.1	13.3
3.06]	82.16]		0.1036	162.1	12.5
3.56]	73.62]		0.1006	162.6	12.5
3.58]	73.13]	175	0.1005	164.2	12.6
3.61]	72.53]		0.1003	162.5	12.5
4.17]	66.57]		0.0966	154.9	11.9
4.17]	65.58]	175	0.0966	164.2	12.6
4.19]	66.17]		0.0965	154.3	11.9
4.21]	65.78]		0.0963	158.0	12.1
4.21]	64.59]	---	0.0963	168.7	13.0
4.47]	63.20]		0.0942	152.7	11.7
4.54]	61.71]		0.0937	163.0	12.5
				Avg.: 163	Avg.: 12.5

* From Fig. 4 of Reference 29

† Table A of Reference 29

TABLE 8. Summary of Calculation of I-values from Nielsen's Absolute Stopping-Power Data* for Protons of 1 to 5 MeV Energy

Element	Nielsen's average value of I [†]	Average per cent reduction in I due to C/Z correction	Value of I indicated by NAS-NRC report ²⁰	New value of I	New value of I/Z
	(eV)	(%)	(eV)	(eV)	
4-Be	56	3.6	65.6	55.6	13.9
13-Al	179	10.7	178	163	12.5
28-Ni	371	18.5	338	308	11.0
29-Cu	376	18.4	342	315	10.9
47-Ag	577	25.2	514	461	9.8
79-Au	942	21.2	893	777	9.8

* From Fig. 4 of Reference 29

† Table A of Reference 29

particles in the stopping material. For each metal several measurements of dE/ds were made for particles in the energy range 1 to 5 MeV.

In the calculation of I for aluminum Nielsen incorporated Walske's C_k corrections, which reduce the value of I by about 5%. The C_k corrections are about 1/5 the shell corrections used here, which lower I for aluminum by 11%. Nielsen did not apply shell corrections in her calculations for metals other than aluminum. The new shell corrections lower I -values for Ni and Cu by 18% and for Ag and Au by more than 21%.

The calculations and a comparison with I -values indicated by the NAS-NRC²⁰ report for Nielsen's data are summarized in Tables 7 and 8.

H. Zrelov and Stoletov

Zrelov and Stoletov³⁰ in 1959 found that the range of 658-MeV protons in copper is 257.6 g/cm². Including Walske's C_k corrections, a density correction, and an adjustment for multiple scattering, they concluded that the mean excitation energy of copper is 305 ± 10 eV.

They also measured the stopping power of 635-MeV protons in H, Be, C, Fe, Cd, and W relative to copper, and they calculated I -values for these elements based on $I = 305$ eV for copper.

From the range measurement for copper, it was found here that with no shell corrections, $I = 310\text{-}311$ eV. According to Zrelov and Stoletov, their estimated shell corrections lowered I by about 3 eV and the density correction reduced I by approximately 2 eV, and thus agreement in methods of calculating I from the range measurement is indicated. When the new shell corrections were used in the calculation, the I -value of copper was found to be 307 eV. The NAS-NRC²⁰ report indicates that if $R = 257.6$ g/cm² for copper, then I is 309 eV.

I -values for the other elements were calculated from Zrelov and Stoletov's relative stopping-power data with the new shell corrections and $I = 307$ eV for copper. Since the proton energy was very high, the shell corrections do not greatly affect the stopping power. When the new shell corrections are included, I for Cd is reduced by 1.3% and for W by 3.5%.

Table 9 is a summary of the results of the calculations.

I. Barkas and von Friesen

In 1961 Barkas and von Friesen³¹ measured the total range of high-energy protons in Al, Cu, Pb, U, and photographic emulsion. From their range measurement in aluminum they concluded that the initial proton energy was 752.2 MeV,

TABLE 9. Calculation of I-values from Zrelov and Stoletov's Range Measurement for 658-MeV Protons in Copper and Stopping-Power Measurements for 635-MeV Protons Relative to Copper

Element	Relative stopping power per electron* for 635-MeV protons	Value of I reported by Zrelov and Stoletov (eV)	Value of I indicated by NAS-NRC report ²⁰ (eV)	New shell correction, C/Z ¹⁸	New value of I (eV)	New value of I/Z
1-H	3.016	15	15	0	13.6	13.6
4-Be	1.167	61	62.0	0	61.7	15.4
6-C	1.268	85	86.6	0	86.2	14.4
26-Fe	1.034	273	277	0.005	276	10.6
29-Cu	(1.000) (R=257.6 g/cm ² for 658-MeV protons)	(305)	309	0.007	(307)	(10.6)
48-Cd	0.887	468	469	0.020	461	9.6
74-W	0.794	680	684	0.041	662	8.9

* From Table II of Reference 30

Parentheses denote reference absorber for relative stopping-power measurements

assuming $I = 163$ eV for aluminum. They used their measurements of total range to determine stopping powers for these protons in Al, Pb, U, and emulsion relative to copper. Relative stopping powers were found for protons in the energy-loss intervals 750-600 MeV, 600-450 MeV, 450-300 MeV, and 750-0 MeV. I -values were calculated for Cu, Pb, U, and emulsion; Walske's shell corrections were included.

From Barkas and von Friesen's total range measurements, I -values were calculated with and without the new shell corrections. When the shell corrections were not included, the values of I for Cu, Pb, and U agreed to within less than 1% with the uncorrected I -values listed by Barkas and von Friesen. When the new shell corrections were made, the I -value for copper was only slightly lower, the I -value for lead decreased by 6%, and the I -value of uranium was 7% lower. The NAS-NRC report²⁰ indicates I -values for Pb and U about 3% lower than these corrected values.

Table 10 shows the results of the calculations.

J. Other Experiments

The experiments reviewed here are representative of the large number of stopping-power and range measurements that yield information about mean excitation energies. Data have been analyzed here for 37 elements with atomic

TABLE 10. Calculation of I-values from Data of Barkas and von Friesen on Total Range of 752.2-MeV Protons

Element	Total range* (g/cm ²)	Corrected value of I reported by Barkas and von Friesen† (eV)	Value of I indicated by NAS-NRC report ²⁰ (eV)	New value of I, including C/Z corrections ¹⁸ (eV)	New value of I/Z
29-Cu	314.91	323	322	321	11.1
82-Pb	415.62	826	809	786	9.6
92-U	432.50	917	892	864	9.4

* Table II of Reference 31

† Table IV of Reference 31; Walske's shell corrections were used

numbers from 1 to 92; the experiments selected for review were performed with protons and deuterons in the energy ranges 1-30 and 250-750 MeV. An exhaustive review has not been attempted. Additional data are not expected to substantially alter the results reported in the concluding section of this thesis; however, four more experiments are noted here for completeness of reference.

In 1955 Rybakov³² published range data for protons of 1-7 MeV in Fe, Cu, Mo, Cd, Sn, Ta, and Pb. Bichsel, Mozley, and Aron³³ in 1957 measured the range of 6- to 18-MeV protons in Be, Al, Cu, Ag, and Au and found $I = 166.45$ eV for aluminum. Bichsel and Uehling³⁴ in 1960 obtained range data for 1- to 20-MeV protons in Al, Cu, Ag, and Au and calculated an I -value of 163 eV for aluminum. Walske's shell corrections were included in the analyses of the latter two of these experiments. Recently Andersen, Garfinkel, Hanke, and Sørensen³⁵ reported measurement of the stopping power of aluminum for 5- to 12-MeV protons and deuterons; their results agreed to within 2% with the stopping-power tables of Barkas and Berger.²⁰

IV. CONCLUSION

Figure 1 shows the mean excitation energy, I , plotted against atomic number, Z . Each measurement of I considered here is shown, and all I -values include the new shell corrections described by Fano and Turner.^{5,15} I -values obtained from relative stopping-power data have been normalized to $I = 163$ eV for aluminum and $I = 314$ eV for copper. The circled points represent the data of Sachs and Richardson,^{26,27} and it can be seen from Figure 1 that I -values calculated from these data do not fit the trend described by the results of other experiments.

Table 11 lists average values of I and I/Z based on the data in Figure 1. Some of the average values of I were obtained from only one measurement of stopping power or range, while others represent an average of 4 or 5 measurements. For example, seven of the nine experiments analyzed included measurements that led to I -values for copper, but only one experiment included cobalt.

The accuracy of most stopping-power and range measurements is about 1%. However, since the natural logarithm of I appears in the stopping-power formula, a 1% error in the

FIGURE 1. Individual Corrected Values of I

TABLE 11. New Average Values of I

Element	Average* value of I (eV)	Average* value of I/Z	References
1-H	18.2	18.2	21,24,25,30
2-He	44.3	22.2	24
3-Li	37.4	12.5	21
4-Be	61.7	15.4	21,22,23,29,30
6-C	81.2	13.5	21,24,25,30
7-N	89.6	12.8	24,25
8-O	101	12.6	24,25
10-Ne	132	13.2	24
13-Al	163	12.5	29
17-Cl	176	10.4	25
18-Ar	189	10.5	24
20-Ca	187	9.35	22
22-Ti	224	10.2	22,23
23-V	250	10.8	22,23
26-Fe	277	10.7	21,22,30
27-Co	290	10.7	23
28-Ni	312	11.2	22,23,29
29-Cu	316	10.9	21,22,23,29,30,31
30-Zn	319	10.6	22
36-Kr	350	9.72	24
41-Nb	407	9.93	22
42-Mo	422	10.0	22
45-Rh	440	9.78	22
46-Pd	456	9.91	22
47-Ag	466	9.93	21,22,23,29
48-Cd	462	9.64	22,30
49-In	481	9.82	22
50-Sn	486	9.71	21,22
54-Xe	480	8.89	24
73-Ta	692	9.47	22,23
74-W	704	9.52	21,22,23,30
77-Ir	730	9.47	22,23
78-Pt	711	9.12	22
79-Au	760	9.62	22,23,29
82-Pb	767	9.36	21,22,31
90-Th	698	7.76	22
92-U	856	9.30	21,31

* Data of Sachs and Richardson^{26,27} are not included.

stopping power will result in an uncertainty in I of about 5%. In relative stopping-power measurements the uncertainty in I is proportional to $\frac{\Delta S}{S} \ln \frac{2mv^2}{I_R}$, where I_R is the mean excitation energy of the reference absorber. Thus at low particle velocities the same uncertainty in S will result in a smaller error in I than at high velocities. A standard error, represented by the error bars in Figures 2 and 3, has been computed for some of the average values of I . This standard error is about 2% for the elements considered, which is consistent with the accuracy of the experimental measurements.

Average values of I plotted against Z appear in Figure 2. Straight lines of the form $I = a + bZ$, as predicted by Bloch's theory,⁶ were fitted to the average I -values by the least-squares method, weighted according to the number of measurements that determined an average value of I . Two lines, one for $Z \leq 13$ and one for $Z > 13$, fit the data better than a single line for all Z . The equations of these lines are shown below, (11) and (12), and although Bloch's theory may not be valid for low- Z elements, the straight lines fit the data well.

$$I = (10.2 \pm 2.07) + (11.8 \pm 0.55)Z, \quad Z \leq 13 \quad (11)$$

$$I = (56.1 \pm 8.76) + (8.61 \pm 0.37)Z, \quad Z > 13 \quad (12)$$

FIGURE 2. New Average Values of I

Ninety-five per cent confidence intervals are shown for a and b. For $Z \leq 3$ equation (11) gives the experimental value of I listed in Table 11 to within $\pm 22\%$; for $4 \leq Z \leq 13$ the I -value obtained from equation (11) will be within about $\pm 3\%$ of the value listed in Table 11. Equation (12) gives $I \pm 4\%$ of the I -values in Table 11, except for Cl, Ar, Ca, and U, where the value of I from equation (12) will be $\pm 17\%$ of the value listed in the table.

Figure 3 shows I/Z plotted against Z . The solid curve is joined smoothly to the straight lines fitted to the data and shown in Figure 2. In 1961 the National Committee on Radiation Protection and Measurements published National Bureau of Standards Handbook 79,²⁸ and the dotted line in Figure 3 is derived from the I -values suggested in Table 2 of the Handbook. This curve does not include the data of Zrelov and Stoletov,³⁰ Barkas and von Friesen,³¹ Nakano, MacKenzie, and Bichsel,²³ or Nielsen,²⁹ nor does it take full account of the shell corrections.

The use of the new shell corrections^{5,15,18} has led to I -values that are consistent with those suggested in the report of the NAS-NRC Subcommittee on the Penetration of Charged Particles.¹⁹ The broken line in Figure 3 is the curve suggested in that report.¹⁷ The variation between the solid and broken lines represents a variation of about 1% in $\ln I$, which is of the order of accuracy in the exper-

FIGURE 3. New Average Values of I/Z

iments. The new shell corrections used here are based on theory, and the results agree well with the tables of Barkas and Berger²⁰ in the NAS-NRC report, which are based on an empirical formula for the shell corrections.

One's judgment of the usefulness of additional measurements depends on whether he is interested in stopping power or in mean excitation energies. The stopping-power data now available are probably sufficiently accurate for most applications. However, since the values of I depend upon measurements of stopping power and range, and since a 1% error in the experimental measurement is reflected as an error of about 5% in I , additional accurate experimental work would be of benefit in leading to better values of I .

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