# Additivity studies of the stopping powers of eleven halogenated hydrocarbons for protons and $\alpha$ particles

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Recent accurate measurements of the stopping powers of eleven halogenated hydrocarbon compounds for protons and $\alpha$ particles in the respective energy intervals of 0.5–2.1 MeV and 1.6–3.0 MeV have been analyzed in the context of Bethe theory, modified by inclusion of Barkas-effect and Bloch terms. The values of various parameters of the formulation were ascertained through two- and three-parameter fits to the data. The results were generally consistent with anticipated deviations from the additivity rule.

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## I. INTRODUCTION

The stopping power of matter for various charged projectiles is a topic of vital importance in numerous areas of physics and related applied fields. It is often necessary to know the energy loss of a charged particle at a specified velocity traversing a known thickness of target material. If one is to avoid direct measurements on every such occasion, a comprehensive theoretical description of the process for any arbitrary projectile-target combination must be available. Considerable progress has been made toward this goal, but a consistently difficult aspect of the campaign has been the case of a target manifesting aggregation effects. That is, the basic theories pertaining to a broad range of projectile velocities, the Bohr and Bethe theories, originally applied strictly to pure monatomic targets in the gaseous state [1]. A target consisting of something as simple as an elemental target in the condensed state, or as a homonuclear diatomic molecule in the gaseous state, manifests bonding effects. The case of many atoms interacting simultaneously with a projectile and each other was first studied by Fermi, as the "density effect" [2]. The connection between the Bethe and Fermi theories was explained by Fano [1,3], who subsequently described how the Bethe theory can be adapted to molecules and condensed matter [1]. Both chemical bonding effects and physical state effects are usually grouped together as aggregation effects. Whenever aggregation effects pertain to a target, the target will herein be referred to as a composite material. Compounds, alloys, and mixtures clearly fall into this general category.

A first approximation in managing aggregation effects, originally advanced by Bragg [4], is to assume the linear additivity of stopping effects of the constituents. This assumption, known as "Bragg's rule" or the "additivity rule," serves as a point of departure in the study of aggregation effects. Thus stopping powers, stopping cross sections, and even parameters of modified Bethe-Bloch theory, are characterized by departures from the simple additivity assumption. The author has initiated several such studies in the past [5–15]. Progress in this area has been reviewed fairly recently [16–18]. The present study is a manifestation of continuing interest in the additivity problem, in this instance, in the stopping powers of several halogenated hydrocarbon gas targets for protons and $\alpha$ particles at energies in the interval of applicability of modified Bethe-Bloch theory [19]. These data seemed particularly interesting to analyze since several disagreements with earlier experimental results [20,21] were reported [19].

## II. METHOD OF ANALYSIS

The stopping power of a given elemental target for light projectiles at energies above 0.5 MeV/u can be described by modified Bethe-Bloch theory. The method will be presented briefly in two sections, the first containing the modified Bethe-Bloch formalism, and the second containing an additivity-based extension to composite target materials. Similar expositions have appeared recently [13–15].

### A. Modified Bethe-Bloch theory

An elemental target of atomic number $Z$ and atomic weight $A$ manifests a stopping power, for a projectile of atomic number $z$ and velocity $v=\beta c$, of

$$S=\frac{0.30708z^2Z}{\beta^2 A}L \tag{1}$$

in units of keV cm$^2$/mg, where $L$ represents the (dimensionless) stopping number per target electron. The stopping number consists of three terms:

$$L=L_0+\xi L_1+L_2\ , \tag{2}$$

where $L_0$ represents the basic stopping number:

$$L_0=\ln\frac{2mc^2\beta^2}{1-\beta^2}-\beta^2-\ln I-\frac{C}{Z}-\frac{\delta}{2}\ . \tag{3}$$

Here $I$ denotes the target mean excitation energy, $mc^2$ is the rest mass energy of the electron, $C$ is the sum of target shell corrections, and $\delta$ is the density effect correction needed for highly relativistic projectiles [22]. The mean

 

excitation energy can be calculated only for targets of low atomic number, as a matter of practicality, so that this quantity is often extracted from a fit to accurate stopping power measurements. Shell corrections can be obtained in the manner established by Bichsel [23–25], wherein the $K$- and $L$-shell corrections of Walske [26,27] are utilized with appropriate scaling factors applied to the $L$-shell correction to provide the $M$- and $N$-shell corrections:

$$C=C_K(\beta^2)+V_L C_L(H_L\beta^2)+V_M C_L(H_M\beta^2) + V_N C_L(H_N\beta^2) \ . \tag{4}$$

$C_K$ and $C_L$ refer to the aforementioned $K$- and $L$-shell corrections, respectively, and $V_i$ and $H_i$ ($i=L$, $M$, and $N$) refer to the scaling factors.

In Eq. (2) the $L_1$ and $L_2$ terms represent the higher-order $z$ terms. $L_1$ is the Barkas-effect correction, obtained from one of three extant formalisms [28–32]. Of the two treatments which appeared initially, the first [28–30] provides a method valid for low projectile velocities, whereas the second [31] describes a method valid for both low and very high projectile velocities. All three methods have been compared recently [33]. The method which clearly provides the best fit to measurements over the broadest energy interval [33] is the first, and it is this formalism that has been incorporated into the stopping number employed for analyses. In this particular formalism,

$$L_1=F(b/x^{1/2})/Z^{1/2}x^{3/2} \ , \tag{5}$$

where $F$ represents a function graphed in Ref. [28], $x=(18787)\beta^2/Z$, and $b$ is the single free (composite) parameter of the theory. $\xi$ appears as an amplitude of the Barkas-effect correction to reflect a controversy over inclusion of close-collision contributions to the Barkas effect. A brief history of parameters $b$ and $\xi$ indicates that $b$ was originally set at $1.8\pm0.2$ on the basis of fits to accurate stopping power measurements [28–30], but when the Bloch term [34] was reintroduced into the Bethe formula a strength factor of about 2 was suggested [35] for the Barkas-effect correction term [31] then used as a means of accounting for close-collision contributions. Soon thereafter, two of the three architects of the low-velocity Barkas-effect formalism [28–30] proposed that $\xi$ be held at unity, but that $b=1.4\pm0.1$, on the basis of fits to very accurate stopping power measurements with the Bloch term present [36]. A number of studies seeking to resolve this matter have been reported by the author [13–15,37–40], and the topic was reviewed less than a decade ago [41]. Although a theoretical study [42] purportedly resolved the issue, attempts were made to obtain best fits for each approach to close-collision contributions [35,36] in the current investigation.

The Bloch term [34] of Eq. (2) has the form

$$L_2=\Psi(1)-\mathrm{Re}[\Psi(1+iy)] \ , \tag{6}$$

where $\Psi$ is the digamma function [43] and $y=z\alpha/\beta$ with $\alpha$ representing the fine-structure constant.

Among the numerous parameters appearing in modified Bethe-Bloch theory, the target mean excitation energy $I$ and Barkas-effect parameters $b$ and $\xi$ are all independent of projectile energy, as indeed are the scaling parameters associated with the energy-dependent shell corrections. Whereas the Bloch term depends on projectile charge and energy, there are no free parameters therein. Moreover, the mean excitation energy is generally assumed to be independent of projectile identity.

A final possible modification to the Bethe-Bloch formula may be inserted in an attempt to describe the gain and loss of electrons by the projectile when it has slowed to a velocity comparable to those of target atomic electrons. This modification appears in some form of projectile effective charge [13,39,40]. One type of effective charge formalism often employed contains two parameters that are presumably independent of both projectile and target identity, as well as of projectile energy. However, systematics of the actual behavior of these parameters as functions of $z$ and $Z$ have been studied in some detail [40]. Fortunately, the projectile velocity interval selected for the present study was sufficiently high to avoid inclusion of any form of effective charge [40], and sufficiently low to avoid inclusion of the density effect correction [22].

### B. Additivity effects

Most real targets require some accommodation to the aforementioned aggregation effects, in which case the additivity rule is invoked. The Bethe-Bloch formula contains several target parameters that must be evaluated by appropriate averaging in the case of a composite-material target. Procedures for calculating average parameter values have been described previously [8,9,29,44]. Although the Bloch term [34] has no target-dependent parameters, the Barkas-effect correction term [28–30] requires averaging, as noted earlier [8,9,29]. Similarly, $L_0$ contains the target-dependent parameters, mean excitation energy and shell corrections, that must be assigned average values [8,9,44]. An obstacle in executing the averaging procedures is the general dearth of knowledge of the correct parameter values for target constituents. Mean excitation energies can generally be calculated from first principles only for low-$Z$ targets, for reasons of practicality. Moreover, aggregation effects have considerable influence on the value of this parameter characteristic of a given target material [8,9,45]. In the same vein, shell-correction parameters have been accurately established for only a few (elemental) target materials [23–25]. The parameter generally selected for a test of the additivity rule is the mean excitation energy, whose calculated average (Bragg) value $I_B$ can be obtained [1,44] as

$$\ln I_B=\frac{\Sigma_j n_j Z_j \ln I_j}{\Sigma_j n_j Z_j} \ , \tag{7}$$

where $n_j$, $Z_j$, and $I_j$, respectively, denote the atomic concentration, atomic number, and mean excitation energy of the $j$th component of the composite material.

Deviations from the additivity rule, with $I$ values exceeding the respective Bragg values, are expected to be

greatest for strong chemical bonds in low-$Z$ compounds, where a large fraction of the total number of electrons participate in the bonding. Moreover, for a given compound, the chemical binding effect is greater at lower projectile velocities, because the tightly bound inner shell electrons contribute less to the stopping than do the outer valence electrons [21].

The approach to additivity described above is to approximate the stopping power of a composite target as a linear combination of the stopping powers of the atomic constituents, which are weighted in proportion to their abundance [5–15,25,29,44]. Another approach, utilized by Neuwirth and Both [46] and Oddershede and Sabin [47], has been to apply the additivity rule to molecular fragments such as bonds or functional groups, with the result of improving the accuracy of the additivity predictions [25].

## III. PROCEDURE

The target gases investigated in the subject experiment [19] were 11 in number, each with average atomic number ($\overline{Z}$), average atomic weight ($\overline{A}$), and logarithmic average mean excitation energy ($I_B$) as shown in Table I, accompanied by the Walske-Bichsel shell-correction scaling parameters. Selected constituent mean excitation energies from which the additivity values were obtained appear in Table II. It should be noted that a rather different value for the mean excitation energy of Br (363 eV), and the same for Cl (174 eV), have reportedly been extracted from stopping power measurements with low energy protons [48].

The procedure utilized in the present study was similar to that of several recent studies [13–15]. A variation on the description of Ref. [15] follows.

Stopping power measurements can be analyzed with the modified Bethe-Bloch theory to extract various parameters of the formalism. Computer codes with a capability to search one-, two-, or three-parameter space have been described in detail earlier [37–40]. The quality of the fit is established by the root-mean-square relative deviation of calculated from measured stopping powers—a quantity assigned the symbol $\sigma$. Thus

TABLE I. Average atomic number ($\overline{Z}$), average atomic weight ($\overline{A}$), logarithmic average mean excitation energy ($I_B$), and shell correction scaling parameters assigned to the 11 halogenated hydrocarbon target materials.

| Target | $\overline{Z}$ | $\overline{A}$ | $I_B$ (eV) | $V_L$ | $H_L$ | $V_M$ | $H_M$ |
|---|---|---|---|---|---|---|---|
| $CF_3H$ | 6.80 | 14.003 | 100.0 | 0.60 | 1.00 | | |
| $CF_4$ | 8.40 | 17.601 | 107.1 | 0.80 | 1.00 | | |
| $CF_3Cl$ | 10.00 | 20.892 | 124.7 | 1.00 | 1.00 | | |
| $CF_3Br$ | 13.60 | 29.782 | 193.2 | 1.00 | 1.00 | 0.45 | 12.0 |
| $CF_2Cl_2$ | 11.60 | 24.183 | 139.3 | 1.00 | 1.00 | 0.20 | 12.0 |
| $C_2H_3Cl$ | 5.33 | 10.416 | 100.7 | 0.42 | 1.00 | | |
| $C_2H_3F_3$ | 5.25 | 10.505 | 87.8 | 0.41 | 1.00 | | |
| $C_2F_4Cl_2$ | 10.25 | 21.365 | 127.0 | 1.00 | 1.00 | | |
| $C_2H_2ClF_3$ | 7.25 | 14.811 | 110.2 | 0.66 | 1.00 | | |
| $C_2H_3ClF_2$ | 6.25 | 12.562 | 105.6 | 0.53 | 1.00 | | |
| $C_4F_8$ | 8.00 | 16.669 | 101.6 | 0.75 | 1.00 | | |

TABLE II. Constituent mean excitation energies for the 11 halogenated hydrocarbon target materials.

| Element | $Z$ | $I$ (eV) | References |
|---|---|---|---|
| H | 1 | 19.26 | [45] |
| C | 6 | 70.0 | [24] |
| F | 9 | 115 | [24] |
| Cl | 17 | 174 | [24] |
| Br | 35 | 343 | [24] |

$$\sigma=\left[\frac{1}{N}\sum_{i=1}^{N}\left[\frac{S_c-S_m}{\Delta S_m}\right]_i^2\right]^{1/2} \tag{8}$$

for measurements of stopping powers at $N$ energies, with $Sc_i$ representing the calculated stopping power at the $i$th energy, and $Sm_i$ and $\Delta Sm_i$ representing the measured stopping power and the statistical uncertainty in that quantity at the $i$th energy, respectively.

The number and accuracy of stopping power measurements in a particular experiment often support the extraction of two parameters, but rarely three. In the present study, the data [19] generally proved amenable to both two- and three-parameter fits. Two-parameter fits were conducted for $I$ (the mean excitation energy) and one of the two parameters associated with the Barkas effect term, i.e., either $b$ (the composite free parameter of the correction formalism [28–30]) or $\xi$ (the amplitude of the correction term). When $b$ was a search parameter, the value of $\xi$ was fixed at either 1, corresponding to the Ritchie-Brandt suggestion [36], or at 2, corresponding to the Lindhard suggestion [35] for another form of the correction term [31]. Similarly, when $\xi$ was a search parameter, $b$ was fixed at 1.36, a value consistent with the Ritchie-Brandt suggestion [36], and one which has proved generally satisfactory in a number of prior analyses [24,38,40,49–51], or at 1.8, corresponding to the Lindhard suggestion [35] for the other form of the correction term [31]. Both of these options for fixed $b$ and $\xi$ values were included in case superior fits might arise to select one of the two sides to the controversy concerning close-collision contributions to the Barkas-effect term [35,36]. Three-parameter fits were conducted for the mean excitation energy and both of the Barkas-effect term parameters ($I$, $b$, and $\xi$).

## IV. RESULTS

Each of the two types of analysis will be described in some detail, first the two- and then the three-parameter searches. A vexing problem encountered in several of the analyses was the existence of local minima, discovered during the searches. This impediment to progress occurred more often in the case of three- than two-parameter searches. The tactic employed to locate the (apparent) true minimum was to vary the starting intervals until all derived values lay within their respective starting intervals. The resulting value of $\sigma$ in such cases lay lowest of all. Hence this result was accepted as the true minimum in multiparameter space.

TABLE III. Results of searches for $I$ (mean excitation energy) and $b$ (parameter of the Barkas-effect term) with $\xi$ (amplitude of the Barkas-effect term) fixed at 1.0 and 2.0, including the minimum value of $\sigma$ (root-mean-square relative deviation of calculated from measured stopping powers) and the deviation of $I$ from an additivity prediction [$\Delta I/I_B=(I-I_B)/I_B$], for proton-projectile data.

| | | $\xi=1.0$ | | | | $\xi=2.0$ | | | |
|---|---|---|---|---|---|---|---|---|---|
| Target | $I_B$ (eV) | $I$ (eV) | $b$ | $\sigma$ | $\Delta I/I_B$ (%) | $I$ (eV) | $b$ | $\sigma$ | $\Delta I/I_B$ (%) |
| $CF_3H$ | 100.0 | 104.1 | 1.51 | 1.38 | 4.1 | 106.7 | 2.05 | 1.32 | 6.7 |
| $CF_4$ | 107.1 | 113.4 | 1.32 | 0.64 | 5.9 | 116.6 | 1.86 | 0.67 | 8.9 |
| $CF_3Cl$ | 124.7 | 126.5 | 0.86 | 0.85 | 1.4 | 132.2 | 1.30 | 0.94 | 6.0 |
| $CF_3Br$ | 193.2 | 206.1 | 1.34 | 0.73 | 6.7 | 213.6 | 1.73 | 0.70 | 10.6 |
| $CF_2Cl_2$ | 139.3 | 150.4 | 1.25 | 0.80 | 8.0 | 156.4 | 1.67 | 0.74 | 12.3 |
| $C_2H_3Cl$ | 100.7 | | | | | | | | |
| $C_2H_3F_3$ | 87.8 | 94.1 | 1.08 | 1.03 | 7.2 | 97.0 | 1.62 | 1.02 | 10.5 |
| $C_2F_4Cl_2$ | 127.0 | 124.4 | 1.01 | 1.18 | −2.0 | 131.8 | 1.41 | 1.11 | 3.8 |
| $C_2H_2ClF_3$ | 110.2 | 119.4 | 1.48 | 1.49 | 8.3 | 122.2 | 2.01 | 1.49 | 10.9 |
| $C_2H_3ClF_2$ | 105.6 | 113.2 | 1.39 | 1.14 | 7.2 | 114.8 | 1.86 | 1.09 | 8.7 |
| $C_4F_8$ | 101.6 | 105.0 | 3.00 | 0.77 | 3.3 | 106.3 | 3.45 | 0.77 | 4.6 |

### A. Two-parameter searches

#### *1. Proton measurements*

Proton stopping power data were analyzed for the energy interval 0.5–2.1 MeV, searching for $I$ and $b$ with $\xi$ fixed at 1.0 and 2.0, and for $I$ and $\xi$ with $b$ fixed at 1.36 and 1.80, with results as shown in Tables III and IV, respectively.

The reported measurements for the target $C_2H_3Cl$ proved incapable of fit by the Bethe-Bloch theory, yielding clearly absurd parameter values. This turn of events will be discussed extensively below.

The ten other target materials provided excellent fits for $\xi$ fixed, with average values of $\sigma$ of 1.0 for each fixed value of $\xi$, and eminently reasonable parameter values save possibly in the cases of $CF_3Cl$ (low-$b$ value of 0.86 for $\xi$ fixed at 1.0) and $C_4F_8$. In the latter instance the extracted values of $b$ (3.00 and 3.45) were remarkably high. Similarly, when $b$ was fixed $C_4F_8$ yielded values for $\xi$ of 0.16 and 0.28, which were remarkably low. All of these results for $C_4F_8$ were consistent in indicating that the measurements evinced a penchant for a small Barkas-effect correction. However, when $b$ was fixed at 1.80, two other target materials yielded rather high values of $\xi$: $CF_3Cl$ (4.08) and $C_2F_4Cl_2$ (3.61). Nonetheless, the average value of $\sigma$ remained at 1.0 for each fixed value of $b$.

Deviations from $I_B$ of the extracted value of $I$ ranged from −2.0% to +8.3% with an average of +5.0% for $\xi=1.0$, from +3.8% to +12.3% with an average of +8.3% for $\xi=2.0$, from +2.0% to +9.0% with an average of +6.0% for $b=1.36$, and from +2.3% to +13.8% with an average of +9.5% for $b=1.80$. Trends in deviations, such as decreases with increasing $\bar{Z}$, were not in evidence, nor were any obvious systematic variations based on bonds present in the compounds. The existence of such trends, whose effects are small, could well have been masked by fluctuations reflecting the considerable scatter, and possibly errors, in the measurements.

TABLE IV. Results of searches for $I$ (mean excitation energy) and $\xi$ (amplitude of the Barkas-effect term) with $b$ (parameter of the Barkas-effect term) fixed at 1.36 and 1.80, including the minimum value of $\sigma$ (root-mean-square relative deviation of calculated from measured stopping powers) and the deviation of $I$ from an additivity prediction [$\Delta I/I_B=(I-I_B)/I_B$], for proton projectile data.

| | | $b=1.36$ | | | | $b=1.80$ | | | |
|---|---|---|---|---|---|---|---|---|---|
| Target | $I_B$ (eV) | $I$ (eV) | $\xi$ | $\sigma$ | $\Delta I/I_B$ (%) | $I$ (eV) | $\xi$ | $\sigma$ | $\Delta I/I_B$ (%) |
| $CF_3H$ | 100.0 | 103.9 | 0.85 | 1.40 | 3.9 | 106.1 | 1.54 | 1.35 | 6.1 |
| $CF_4$ | 107.1 | 113.6 | 1.06 | 0.64 | 6.1 | 116.0 | 1.84 | 0.67 | 8.3 |
| $CF_3Cl$ | 124.7 | 133.0 | 2.17 | 0.96 | 6.6 | 140.9 | 4.08 | 1.08 | 13.0 |
| $CF_3Br$ | 193.2 | 207.7 | 1.08 | 0.72 | 7.5 | 216.4 | 2.34 | 0.69 | 12.0 |
| $CF_2Cl_2$ | 139.3 | 151.9 | 1.21 | 0.78 | 9.0 | 158.5 | 2.44 | 0.74 | 13.8 |
| $C_2H_3Cl$ | 100.7 | | | | | | | | |
| $C_2H_3F_3$ | 87.8 | 95.4 | 1.43 | 1.02 | 8.6 | 97.9 | 2.46 | 1.02 | 11.5 |
| $C_2F_4Cl_2$ | 127.0 | 129.9 | 1.80 | 1.12 | 2.3 | 139.0 | 3.61 | 1.05 | 9.4 |
| $C_2H_2ClF_3$ | 110.2 | 118.8 | 0.85 | 1.49 | 7.8 | 120.9 | 1.52 | 1.49 | 9.7 |
| $C_2H_3ClF_2$ | 105.6 | 111.8 | 1.00 | 1.15 | 5.9 | 114.8 | 1.88 | 1.10 | 8.7 |
| $C_4F_8$ | 101.6 | 103.6 | 0.16 | 0.76 | 2.0 | 103.9 | 0.28 | 0.76 | 2.3 |

### 2. α particle measurements

α particle stopping power measurements were analyzed for the energy interval 1.6–3.0 MeV, searching for pairs of parameters as in the proton data analysis, with results as shown in Tables V and VI.

The reported measurements for the target $C_2H_3Cl$ proved quite accommodating for fits by the Bethe-Bloch theory, in sharp contrast to the failure of the proton measurements. When this situation developed, the author contacted G. Reiter to inform him that the ratio of stopping powers for α particles and protons at the same velocity was considerably awry, whereas the α particle measurements provided acceptable fits by theory. The author surmised that a major error might have occurred during data reduction for the proton measurements. G. Reiter replied [52] that both $C_2H_3Cl$ and $C_2H_2ClF_3$ targets had furnished large experimental errors, as noted in Ref. [19]. It is true that the poorest fits were consistently manifested by these two compounds, as clearly shown in Tables III-VI. However, the discrepancy in fits of proton and α particle data for $C_2H_3Cl$ is not satisfactorily explained by the known experimental difficulties with the compound [19,52].

The 11 target gases yielded average values for $\sigma$ of 1.0 for all four combinations of two-parameter fits, even though the troublesome targets $C_2H_3Cl$ and $C_2H_2ClF_3$ were included. However, extracted $b$ values for $CF_3H$, $C_2H_3F_3$, and $C_4F_8$ with $\xi$ fixed were notably high, and extracted $\xi$ values with $b$ fixed were notably low. All of these results were indicative of a need for a small Barkas-effect correction. (This trend was discernible only for $C_4F_8$ in the case of proton projectiles.)

Deviations from the additivity rule, represented by the relative difference $(I-I_B)/I_B$, ranged from $-6.9\%$ to $+33.0\%$ with an average of $+5.1\%$ for $\xi=1.0$, from $-3.0\%$ to $+46.5\%$ with an average of $+12.5\%$ for $\xi=2.0$, from $-10.6\%$ to $+35.2\%$ with an average of $+4.4\%$ for $b=1.36$, and from $-8.8\%$ to $+46.8\%$ with an average of $+9.7\%$ for $b=1.80$. The largest deviations were associated with $C_2H_2ClF_3$ in each case, but several target gases showed deviations that are quite large in magnitude. Again, fluctuations induced in extracted parameter values by scatter and possible errors in the measurements undoubtedly could have precluded the discovery of discernible bond-dependent trends in these values.

## B. Three-parameter searches

### 1. Proton measurements

Few surprises attended the outcome of three-parameter fits of the proton measurements, the results of which are displayed in Table VII. Incongruous values of $b$ occurred for $CF_3H$, $CF_3Cl$, and $C_2H_3ClF_2$, whereas incongruous values of $\xi$ occurred for $CF_3H$, $CF_3Cl$, $C_2F_4Cl_2$, $C_2H_3ClF_2$, and $C_4F_8$. The average value of $\sigma$ lay just below 1.0. Additivity deviations, measured by $(I-I_B)/I_B$, ranged from $-2.2\%$ to $+19.5\%$ with an average of $+8.9\%$. The maximum deviation occurred for the $C_2H_3ClF_2$ target. Given the additional degree of freedom from a three-parameter search, the data would yield parameter values which manifested adjustment to the scatter, and possibly errors, in the measurements even more so than in the two-parameter case. Thus one might hold reduced expectations of discerning bond-dependent trends in extracted parameter values.

### 2. α particle measurements

Three-parameter fits of the α particle measurements yielded the parameter values shown in Table VIII. Extracted values of $b$ remained eminently plausible save for $CF_3H$ and $C_4F_8$, whereas extracted values of $\xi$ appeared to be remarkably low for $CF_4$, $CF_3Br$, $C_2H_3F_3$, and $C_4F_8$. Again the average value of $\sigma$ was slightly less than 1.0.

Deviations from additivity of the extracted mean excitation energy, $(I-I_B)/I_B$, fluctuated rather wildly, ranging from $-9.9\%$ ($C_2H_3F_3$) to $+36.5\%$ ($C_2H_2ClF_3$). The average deviation was only $+4.4\%$, however. The previous comment concerning bond-dependent systematics in

TABLE V. Results of searches for $I$ (mean excitation energy) and $b$ (parameter of the Barkas-effect term) with $\xi$ (amplitude of the Barkas-effect term) fixed at 1.0 and 2.0, including the minimum value of $\sigma$ (root-mean-square relative deviation of calculated from measured stopping powers) and the deviation of $I$ from an additivity prediction $[\Delta I/I_B=(I-I_B)/I_B]$, for α particle-projectile data.

| | | $\xi=1.0$ | | | | $\xi=2.0$ | | | |
|---|---|---|---|---|---|---|---|---|---|
| Target | $I_B$ (eV) | $I$ (eV) | $b$ | $\sigma$ | $\Delta I/I_B$ (%) | $I$ (eV) | $b$ | $\sigma$ | $\Delta I/I_B$ (%) |
| $CF_3H$ | 100.0 | 95.5 | 2.48 | 0.74 | −4.5 | 98.9 | 2.97 | 0.73 | −1.1 |
| $CF_4$ | 107.1 | 122.4 | 1.59 | 1.20 | 14.3 | 134.1 | 2.00 | 1.22 | 25.2 |
| $CF_3Cl$ | 124.7 | 124.2 | 1.20 | 0.81 | −0.4 | 138.2 | 1.60 | 0.77 | 10.8 |
| $CF_3Br$ | 193.2 | 194.3 | 1.59 | 0.60 | 0.6 | 205.1 | 1.99 | 0.74 | 6.2 |
| $CF_2Cl_2$ | 139.3 | 160.8 | 1.40 | 1.00 | 15.4 | 163.7 | 1.92 | 1.10 | 17.5 |
| $C_2H_3Cl$ | 100.7 | 99.2 | 1.79 | 1.44 | −1.5 | 105.6 | 2.21 | 1.45 | 4.9 |
| $C_2H_3F_3$ | 87.8 | 81.7 | 2.35 | 0.99 | −6.9 | 85.2 | 2.84 | 1.00 | −3.0 |
| $C_2F_4Cl_2$ | 127.0 | 129.2 | 1.20 | 0.54 | 1.7 | 143.2 | 1.60 | 0.67 | 12.8 |
| $C_2H_2ClF_3$ | 110.2 | 146.6 | 1.40 | 2.20 | 33.0 | 161.4 | 1.80 | 2.18 | 46.5 |
| $C_2H_3ClF_2$ | 105.6 | 110.6 | 1.80 | 0.79 | 4.7 | 119.1 | 2.19 | 0.80 | 12.8 |
| $C_4F_8$ | 101.6 | 101.2 | 2.47 | 0.59 | −0.4 | 106.6 | 2.85 | 0.58 | 4.9 |

TABLE VI. Results of searches for $I$ (mean excitation energy) and $\xi$ (amplitude of the Barkas-effect term) with $b$ (parameter of the Barkas-effect term) fixed at 1.36 and 1.80, including the minimum value of $\sigma$ (root-mean-square relative deviation of calculated from measured stopping powers) and the deviation of $I$ from an additivity prediction $[\Delta I/I_B=(I-I_B)/I_B]$, for $\alpha$ particle-projectile data.

| | | $b=1.36$ | | | | $b=1.80$ | | | |
|---|---|---|---|---|---|---|---|---|---|
| Target | $I_B$ (eV) | $I$ (eV) | $\xi$ | $\sigma$ | $\Delta I/I_B$ (%) | $I$ (eV) | $\xi$ | $\sigma$ | $\Delta I/I_B$ (%) |
| $CF_3H$ | 100.0 | 89.4 | 0.15 | 0.74 | −10.6 | 91.2 | 0.32 | 0.74 | −8.8 |
| $CF_4$ | 107.1 | 115.2 | 0.62 | 1.22 | 7.6 | 127.2 | 1.41 | 1.21 | 18.8 |
| $CF_3Cl$ | 124.7 | 132.7 | 1.39 | 0.85 | 6.4 | 138.0 | 2.52 | 0.89 | 10.7 |
| $CF_3Br$ | 193.2 | 200.9 | 0.80 | 0.57 | 4.0 | 200.1 | 1.46 | 0.67 | 3.6 |
| $CF_2Cl_2$ | 139.3 | 157.6 | 0.90 | 0.97 | 13.1 | 166.3 | 1.80 | 0.98 | 19.4 |
| $C_2H_3Cl$ | 100.7 | 93.1 | 0.42 | 1.44 | −7.5 | 100.0 | 1.05 | 1.43 | −0.7 |
| $C_2H_3F_3$ | 87.8 | 79.1 | 0.28 | 1.00 | −9.9 | 79.2 | 0.45 | 0.99 | −9.8 |
| $C_2F_4Cl_2$ | 127.0 | 138.2 | 1.40 | 0.58 | 8.8 | 160.4 | 3.01 | 0.65 | 26.3 |
| $C_2H_2ClF_3$ | 110.2 | 149.3 | 1.00 | 2.19 | 35.2 | 161.8 | 2.01 | 2.18 | 46.8 |
| $C_2H_3ClF_2$ | 105.6 | 109.6 | 0.58 | 0.84 | 3.8 | 110.6 | 1.00 | 0.79 | 4.7 |
| $C_4F_8$ | 101.6 | 98.5 | 0.25 | 0.64 | −3.0 | 96.9 | 0.34 | 0.59 | −4.6 |

parameter values extracted from the proton measurements applies here as well.

In general the outcome of three-parameter fits was encouragingly plausible values of all extracted parameters, thus providing enhanced credibility for the measurements.

### C. General observations

Several noteworthy positive attributes of the subject measurements have been described thus far. Another salient feature is the absence of a predominance of higher $I$ values extracted from proton data compared to those values taken from $\alpha$ particle data for the respective compounds. The balance actually lies in the opposite trend for the two-parameter fits, contrary to the findings of several recent studies [13,15,37,53], the most recent of which [53] pertained to the gas targets He and Ne, whose atomic numbers nearly span those of the compounds currently studied. The former trend [13,15,37,53] is present by a 6 to 4 margin in the case of the three-parameter fits, however.

A further objective of the present investigation was to learn if the subject measurements might evince a preference for one of the two approaches [35,36] to inclusion of close-collision contributions to the Barkas-effect correction, this preference being expressed by a better quality of fit. No such indication was present, as found in previous analyses [13–15,37,38,40,53]. In this connection, however, a difference between the two sets of fits was evident in the average deviations from additivity reflected in the mean excitation energies derived, in that the average value for the Lindhard suggestion [35] set was 1.5–2.5 times the corresponding value for the Ritchie-Brandt suggestion [36] set. It should be emphasized that if a given set of measurements did evince markedly better fits for one or the other suggestion [35,36], one might surmise the superiority of that approach in dealing with close-collision contributions to the Barkas-effect term. Evidence gathered so far indicates that if this question is to be resolved, the subject measurements will possess accuracy greater than that now considered state of the art.

In order to select starting intervals for the three-

TABLE VII. Results of searches for $I$ (mean excitation energy), $b$ (parameter of the Barkas-effect term), and $\xi$ (amplitude of the Barkas-effect term), including the minimum value of $\sigma$ (root-mean-square relative deviation of calculated from measured stopping powers) and the deviation of $I$ from an additivity prediction $[\Delta I/I_B=(I-I_B)/I_B]$, for proton-projectile data.

| Target | $I_B$ (eV) | $I$ (eV) | $b$ | $\xi$ | $\sigma$ | $\Delta I/I_B$ (%) |
|---|---|---|---|---|---|---|
| $CF_3H$ | 100.0 | 114.1 | 2.72 | 5.23 | 1.24 | 14.1 |
| $CF_4$ | 107.1 | 113.4 | 1.26 | 0.95 | 0.64 | 5.9 |
| $CF_3Cl$ | 124.7 | 121.9 | 0.46 | 0.49 | 0.82 | −2.2 |
| $CF_3Br$ | 193.2 | 212.9 | 1.71 | 1.94 | 0.70 | 10.2 |
| $CF_2Cl_2$ | 139.3 | 156.6 | 1.71 | 2.11 | 0.74 | 12.4 |
| $C_2H_3Cl$ | 100.7 | | | | | |
| $C_2H_3F_3$ | 87.8 | 96.4 | 1.58 | 1.89 | 1.02 | 9.8 |
| $C_2F_4Cl_2$ | 127.0 | 138.1 | 1.71 | 3.24 | 1.06 | 8.7 |
| $C_2H_2ClF_3$ | 110.2 | 119.4 | 1.46 | 0.98 | 1.49 | 8.3 |
| $C_2H_3ClF_2$ | 105.6 | 126.2 | 2.85 | 7.54 | 1.00 | 19.5 |
| $C_4F_8$ | 101.6 | 103.9 | 1.98 | 0.32 | 0.76 | 2.3 |

TABLE VIII. Results of searches for $I$ (mean excitation energy), $b$ (parameter of the Barkas-effect term), and $\xi$ (amplitude of the Barkas-effect term), including the minimum value of $\sigma$ (root-mean-square relative deviation of calculated from measured stopping powers) and the deviation of $I$ from an additivity prediction $[\Delta I/I_B=(I-I_B)/I_B]$, for $\alpha$ particle-projectile data.

| Target | $I_B$ (eV) | $I$ (eV) | $b$ | $\xi$ | $\sigma$ | $\Delta I/I_B$ (%) |
|---|---|---|---|---|---|---|
| $CF_3H$ | 100.0 | 95.3 | 2.49 | 1.00 | 0.74 | −4.7 |
| $CF_4$ | 107.1 | 115.4 | 1.25 | 0.55 | 1.20 | 7.7 |
| $CF_3Cl$ | 124.7 | 125.6 | 1.31 | 1.18 | 0.77 | 0.7 |
| $CF_3Br$ | 193.2 | 189.2 | 1.25 | 0.55 | 0.50 | −2.1 |
| $CF_2Cl_2$ | 139.3 | 159.8 | 1.49 | 1.11 | 0.95 | 14.7 |
| $C_2H_3Cl$ | 100.7 | 100.8 | 1.87 | 1.17 | 1.43 | 0.1 |
| $C_2H_3F_3$ | 87.8 | 79.1 | 1.89 | 0.48 | 0.99 | −9.9 |
| $C_2F_4Cl_2$ | 127.0 | 129.9 | 1.15 | 0.95 | 0.58 | 2.3 |
| $C_2H_2ClF_3$ | 110.2 | 150.4 | 1.49 | 1.19 | 2.19 | 36.5 |
| $C_2H_3ClF_2$ | 105.6 | 111.4 | 1.79 | 1.03 | 0.80 | 5.5 |
| $C_4F_8$ | 101.6 | 99.0 | 2.21 | 0.65 | 0.59 | −2.6 |

parameter searches, it was necessary to choose either the Ritchie-Brandt suggestion [36] set of fits ($b$ fixed at 1.36 and $\xi$ fixed at 1) or the Lindhard suggestion [35] set ($b$ fixed at 1.80 and $\xi$ fixed at 2) from the two-parameter fits. Reference values of the parameters were (quite arbitrarily) taken from the former set, so that the three-parameter search results might be expected to conform more closely with the former set, as indeed was the case.

Only one of the compounds, $CF_3Br$, contained Br, whose prescribed [24] mean excitation energy was 343 eV. If this value of mean excitation energy for Br were raised to the recently obtained value [48] of 363 eV, the already moderate deviations from additivity of extracted $I$ values would be lowered further—not necessarily a desired outcome for heuristic reasons. However, it is clear from results in Tables III and IV that in the case of proton measurements the compounds $CF_3Br$ and $CF_2Cl_2$ provided some of the highest deviations from additivity, so that reduction of the deviation for these two (highest-$\bar{Z}$) compounds could well be appropriate. These compounds rated attention for another reason in the original report of the measurements, in that the curves of their stopping cross sections for both types of projectile crossed each other at comparable projectile velocities [19].

Finally, one can readily advance a heuristic argument to the effect that all negative deviations of mean excitation energy from the Bragg value, especially those large in magnitude, are suspect. Since the constituent mean excitation energies are probably quite reliable, this observation merely suggests that minor experimental errors may be present to influence the results of detailed analysis by modified Bethe-Bloch theory. Nonetheless, the general results of the current study surely establish the subject data as the benchmark measurements for such targets, despite considerable scatter in the data.

## V. SUMMARY AND CONCLUSIONS

Recent measurements of the stopping powers of 11 halogenated hydrocarbon gaseous targets for protons and alpha particles [19] have been analyzed in terms of modified Bethe-Bloch theory to extract parameters of the formulation. These measurements proved amenable to both two- and three-parameter fits, generally yielding plausible values of the extracted parameters while providing excellent fits. The additivity assumption was tested, using the mean excitation energy as the test parameter. Results indicated no definitive trends, presumably because of the accuracy level of, and/or minor experimental errors in, the published data. For example, in the two-parameter fits the four compounds with lowest $\bar{Z}$ showed a correlation with low deviations of $I$ values from $I_B$, contrary to expectation. The absence of any systematic trends in the three-parameter fit results was somewhat expected, since the accuracy level of the measurements would suggest some difficulty in obtaining a well-defined trio of parameter values. Nonetheless, these measurements represent a welcome supplement to, and extension of, related data from the Baylor University group [54].

The author places the highest credibility in results from two-parameter fits of proton data, first with $b$ fixed at 1.36, and second with $\xi$ fixed at 1.0. Recommended values of the parameters are $\xi=1.0$, $b=1.36$, and $I=1.055I_B$ for all compounds studied. These values can be refined when measurements of greater accuracy over broader projectile-energy intervals become available. Moreover, uncertainties reflecting the density and statistical accuracy of the measurements can be assigned to the extracted parameter values, in accordance with a technique devised and described previously [38,40]. Since the method of calculation is quite time consuming, only typical values of uncertainties are generally computed. The results of applying this technique to the case of a selected target, $CF_2Cl_2$, were that $\Delta I=2.2$ eV, $\Delta b=0.06$ for $\xi$ fixed at 1.0 and $\Delta I=2.8$ eV, $\Delta\xi=0.13$ for $b$ fixed at 1.36. These values can serve as guidelines for corresponding uncertainties in parameter values for any of the two-parameter fits, since both proton and $\alpha$ particle data sets possessed essentially equal densities of, and uncertainties in, stopping power measurements [19].


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