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Energy-Loss and Stopping-Power Measurements between 2 and 10 MeV/amu for ^{12}C, ^{14}N, and ^{16}O in Silicon

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The energy deposited in a 94.6-μm-thick totally depleted silicon detector by beams of ^{12}C, ^{14}N, and ^{16}O was measured at incident energies up to 10 MeV/amu. Theoretical stopping powers for these ions are derived from best theoretical values for protons and a universal distribution of ion charges as a function of energy. The effect of a recently derived z^3 correction is included. This correction is found to improve the fit to the experimental values.

I. INTRODUCTION

In the preceding paper[1] (hereafter referred to as I), it was shown that the z^3 correction to theoretical values for the stopping power, derived by Ashley *et al.*,[2] is required to fit measured values of the energy loss by ^3He and ^4He ions in silicon. In the present paper that analysis is extended to the energy loss by heavier ions, namely, ^{12}C, ^{14}N, and ^{16}O, in silicon. Northcliffe[3] states that the distinction between the stopping powers for protons and α particles on one hand and those for heavier ions on the other is arbitrary but real; arbitrary since the same mechanisms apply for both, real since the light ions remain completely ionized over the energy range of interest while the heavier ones do not. As detailed by Northcliffe, the calculation of ion charge as a function of energy is an extremely difficult task; however, he provides experimentally determined universal curves for the charge state of heavy ions in aluminum in the energy range of interest here. The difference between the universal curves for different materials should be small; particularly for two materials as similar in electronic structure as aluminum and silicon.

Recently, Betz published an extensive review[4] of the experimental and theoretical situation concerning charge states of heavy ions ($16 \leq z \leq 92$) which penetrate through gaseous and solid targets. He notes that the only feasible means of determining charge distributions, as opposed to average charge, is by interpolating from experimental data. Average charge data obtained by Martin[5] for ^{16}O in Si at 5–36 MeV agree very well with the values obtained from Northcliffe's universal curves for Al.

The misestimate of energy loss due to multiple scattering effects and asymmetry in the energy-loss distribution which were of minor significance in the He data are even less important here, since the effects decrease with increasing ion mass.[6,7] The contribution of nuclear stopping[8] was calculated and found to be insignificant, i.e., less than 0.01% of the total stopping power in this energy range.

The results presented here are not biased by channeling effects in the silicon detectors used. Measurements on 5–36-MeV ^{16}O and ^{12}C ions by Martin[5] show the full width of the channeling directions for these ions in Si to be about 1°. The measurements reported here used particles incident at up to ± 1.5 deg from the normal to the plane of the silicon wafer. If any channeling were occurring it would have produced an asymmetrically broadened, possibly double-peaked energy-loss distribution. Such distributions were not observed, so channeling cannot be an important factor affecting the data. In addition, the ^4He-^3He results of I, taken with the same experimental setup and detectors, also show that channeling is not significant for the directions of particle travel through the silicon surface barrier detectors used in this experiment.

II. EXPERIMENTAL RESULTS

The experimental apparatus and technique were described in I and will not be repeated here. The errors in the ^{12}C and ^{16}O data are similar to those found there, namely up to a 1% systematic error from the measurement of detector thickness[9] and 1–2% random errors in the single parameter analyzer values for incident energy and energy loss. The results and the limits for the random error are shown in Fig. 1.

In the course of analyzing the ^{14}N data, it became apparent that the measurements of incident and residual energies were meaningless due to a failure in the 750-μm detector while the measurements of the energy loss in the 94.6-μm detector remained valid. The nominal beam energy, defined by the absorber foils and the analyzer magnet settings, had not been expected to be particularly accurate. However, analysis of the ^{12}C and ^{16}O measurements

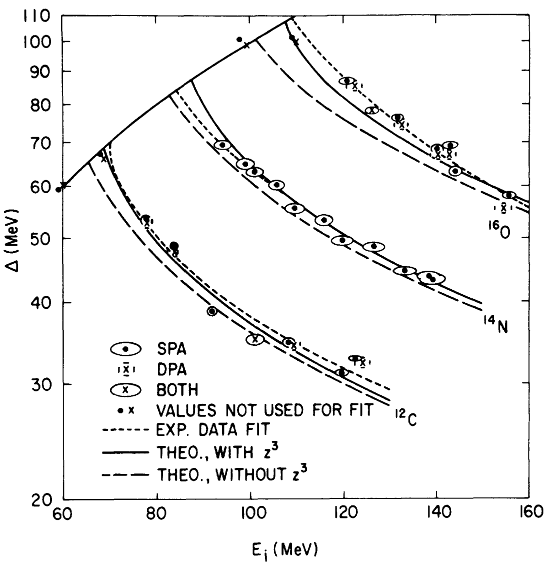

FIG. 1. Plot of energy-loss data for ^{12}C, ^{14}N, and ^{16}O incident on 94.6-μm-thick silicon detector. SPA refers to the single-parameter analyzer. DPA refers to the dual-parameter analyzer. Character size indicates error limits except for points not used in obtaining the experimental fit.

disclosed that the incident energy was less than the nominal energy by $(5.5 \pm 3.0)\%$. Therefore, the ^{14}N data were plotted as the energy loss versus the nominal energy less 5.5%. These data may contain both systematic and random errors in the incident energy. However, the resulting plot, especially its shape which would not be affected by systematic errors in the input energy correlates well with the theory affording additional weight to the evidence of the ^{12}C and ^{16}O data.

III. STOPPING POWER

A. Experimental

In a manner similar to that used in I, stopping powers S (MeV cm^2/g) were determined by use of the expression

$$S(\mathcal{E}) = -\frac{dE}{dx} = A(\mathcal{E}+r)^{1-n} , \qquad (1)$$

where x is the path length (g/cm^2), $\mathcal{E} = E$ (MeV)/M (amu) is the energy per unit mass of the ion, and A, r, and n are constants determined by fitting the energy-loss function derived from (1) to the data. Best-fit values are given in Table I for ^{12}C, ^{14}N, and ^{16}O. As shown in Fig. 1 the fits obtained are less satisfactory than those for the He data in I. This is partially because Eq. (1) is a poorer approximation to the stopping power for heavier ions, particularly at low energies. While this introduces some additional error in the comparison between experimental and theoretical results, we believe it to be insignificant compared to the uncertainty due to the scatter in the data. Values of stopping power calculated from (1) with the parameters of Table I are given in Table II.

B. Theoretical

A corrected form for the stopping power of an ion of atomic number z and energy per unit mass $\mathcal{E}$ (MeV/amu) in a material of atomic number Z has been suggested by Ashley *et al.* and used as Eq. (5) of I. Since only a fraction ϕ_i $(z, \mathcal{E}, Z)$ of the ions at any point in the material is in any particular charge state z_i, we assume that the corrected stopping power for heavy ions can be obtained, in general, by summing over all nonzero ϕ_i:

$$S_c(z, \mathcal{E}, Z) = \sum_i \phi_i(z, \mathcal{E}, Z) S_c(z_i, \mathcal{E}, Z) = \sum_i \phi_i(z, \mathcal{E}, Z) S_b(z_i, \mathcal{E}, Z) \times [1 + z_i u(\mathcal{E}, Z)] . \qquad (2)$$

Here $S_b(z_i, \mathcal{E}, Z)$ is the "best" previous theoretical value for ions in charge state z_i, i.e., the z^2 theoretical result using the Born approximation including all corrections. The z^3 factor $u(\mathcal{E}, Z)$ that results from the work of Ashley *et al.* has been defined and tabulated for silicon in I. For the He work it was possible to use the theoretical stopping power results of Bichsel and Tschalär[10] for ^{3}He and ^{4}He in Si directly. The present work uses the fact that in the z^2 approximation the stopping power of an ion of energy per unit mass $\mathcal{E}$ in charge state z_i is related to that of a proton, $S_p(\mathcal{E}, Z)$, by

$$S_b(z_i, \mathcal{E}, Z) = z_i^2 S_p(\mathcal{E}, Z) \qquad (3)$$

in the region $\mathcal{E} > 0.5$ MeV/amu, where the proton charge state is unity. Thus, the extensive theoretical results of Janni[11] for protons incident on a wide variety of elements and compounds may be used in the determination of stopping power for heavy ions. Here for consistency with the He work in I, the results of Bichsel and Tschalär for protons in Si have been used.

Since the required charge state measurements have not been made for Si, the values of ϕ_i were taken from the universal curves of Northcliffe (Fig. 3, Ref. 3) which are given in terms of the

TABLE I. Experimental stopping-power parameters for use in Eq. (1).

Incident ion	A $\frac{\text{MeV}^n\,\text{cm}^2}{\text{g amu}^{n-1}}$	n	r $\frac{\text{MeV}}{\text{amu}}$
^{12}C	8225.9	1.787	0.544
^{14}N	34790.0	2.157	3.680
^{16}O	270420.0	2.735	6.501

velocity parameter

$$\xi = 137\beta/z \approx 6.35\mathcal{E}^{1/2}/z\ , \tag{4}$$

and hence are independent of Z.

The z^3 corrected stopping power is then given by

$$S_c(z, \mathcal{E}, Z) = S_p(\mathcal{E}, Z)\sum_i \phi_i(\xi)\, z_i^2[1 + z_i u(\mathcal{E}, Z)]\ , \tag{5}$$

which can be written in a form analogous to that for an ion in the single charge state z:

$$S_c(z, \mathcal{E}, Z) = S_b(z, \mathcal{E}, Z)[1 + z\, U(z, \mathcal{E}, Z)]\ . \tag{6}$$

Here $S_b(z, \mathcal{E}, Z)$ is the "best" previous theoretical value

$$S_b(z, \mathcal{E}, Z) = z^2 \gamma_{\mathrm{rms}}^2\, S_p(\mathcal{E}, Z) \tag{7}$$

and U is the z^3 factor

$$U(z, \mathcal{E}, Z) = u(\mathcal{E}, Z)\, \gamma_{\mathrm{rmc}}^3/\gamma_{\mathrm{rms}}^2\ , \tag{8}$$

where γ_{rms} and γ_{rmc} are the fractional root-mean-square and -cube charges

$$\gamma_{\mathrm{rms}}^2 = \sum_i \phi_i(\xi)\left(\frac{z_i}{z}\right)^2 \tag{9}$$

and

$$\gamma_{\mathrm{rmc}}^3 = \sum_i \phi_i(\xi)\left(\frac{z_i}{z}\right)^3\ . \tag{10}$$

Northcliffe's curves for $\phi_i(\xi)$ depend upon his particle spectrograph results for their validity (see Ref. 3, footnote 9) and constitute a body of data independent of the stopping-power measurements which he made at the same time. These results were obtained principally with heavy ions in Al and Formvar. Additionally, Northcliffe[3] has found γ_{rms}^2 from (9) for B, C, N, O, F, and Ne ions in Al both from his universal curves and from other available charge state measurements, which were made mostly for solid materials other than Al. From an equation equivalent to (7) he computed the stopping powers and the comparison appears satisfactory (see Ref. 3, Fig. 6 and 7, $\mathcal{E} = 0.5$ to 2 MeV/amu). Thus, while it would be desirable to use values of ϕ_i measured for Si, the universal curves provide a reasonably good alternative.

Values of S_b and S_c calculated from (7) and (6) for 1–10-MeV/amu ^{16}O, ^{14}N, and ^{12}C are shown in Table II. Although Eq. (1) will fit the S_b and S_c values to $\pm 2\%$ from 2 to 10 MeV/amu, the deviation becomes somewhat larger at 1 MeV/amu. The theoretical energy losses were obtained from the stopping powers by a numerical integration method for direct comparison with the data without the intermediate fit to Eq. (1). These results are plotted in Fig. 1 together with the fits to the experimental data.

IV. DISCUSSION

A. Effective Charge and the z^3 Correction

As stated above, the charge state of the ions incident on the stopping material was determined by use of Northcliffe's universal curves, $\phi_i(\xi)$ in (9) and (10). It has been noted that the true degree of ionization in the material may differ from the mean due to effects such as distortion of the charge density of the atoms of the material[12] or penetration of the charge cloud of the ion by electrons of the material.[13] A further possible complication is that the observed mean charge, which must be measured external to the surface, may differ from its value inside the solid due to stripping of the electrons from the departing ions.[14] It appears, however, that this effect is only important for lower energies and heavier ions than those of interest here. It has also been pointed out[14,15] that restrictions in the derivation leading to the theoretical result (6) may not be satisfied, hence precluding the

TABLE II. Stopping-power data for ^{12}C, ^{14}N, and ^{16}O, where S_b is the best previous theoretical stopping power, S_c incorporates the z^3 correction, "Expt. fit" is the stopping power derived from the fit to the experimental data (all in units of MeV cm^2/g) and "% Diff." is the difference between Expt. fit and S_c relative to Expt. fit.

$\mathcal{E}$ [a]	^{12}C				^{14}N				^{16}O			
MeV/amu	$S_b(\mathcal{E})$ [b]	$S_c(\mathcal{E})$ [c]	Expt. fit	% Diff.	$S_b(\mathcal{E})$	$S_c(\mathcal{E})$	Expt. fit	% Diff.	$S_b(\mathcal{E})$	$S_c(\mathcal{E})$	Expt. fit	% Diff.
1	4350	5043	5848	13.8	5756	6805	5834	−16.6	7592	9171	8196	−11.9
2	3532	3850	3948	2.5	4492	4950	4663	−6.2	5588	6223	6596	5.7
3	2834	2999	3042	1.4	3721	3971	3865	−2.7	4711	5067	5439	6.8
4	2370	2468	2501	1.3	3164	3315	3289	−0.8	4011	4228	4572	7.5
5	2044	2110	2139	1.4	2745	2848	2855	0.2	3506	3654	3904	6.4
6	1796	1841	1877	1.9	2424	2494	2516	0.9	3137	3240	3378	4.1
7	1610	1643	1678	2.1	2170	2223	2246	1.0	2807	2885	2956	2.4
8	1455	1480	1522	2.8	1972	2012	2025	0.6	2556	2614	2611	−0.1
9	1334	1354	1395	2.9	1808	1839	1842	0.2	2347	2394	2326	−2.9
10	1231	1247	1290	3.3	1672	1698	1687	0.7	2171	2209	2087	−5.9

[a] ^{12}C, $M = 11.996\,71$ amu; ^{14}N, $M = 13.999\,23$ amu; ^{16}O, $M = 15.990\,52$ amu.
[b] Obtained from Eq. (7) by use of the theoretical stopping powers contained in Ref. 8, $I = 173.5$ eV.
[c] Theoretical calculation including z^3 effect of Ref. 2, i.e., S_b as corrected in Eq. (6).

use of $\gamma_{\rm rms}^2$. The usual approach is, therefore, to replace $\gamma_{\rm rms}^2$ in Eq. (7) by an effective fractional charge

$$\gamma_{\rm eff}^2 = z^{-2}[S_b(z, \mathcal{E}, Z)/S_p(\mathcal{E}, Z)]_{\rm expt} \ . \qquad (11)$$

where the values in the bracket are now the experimental results. Defined in this way $\gamma_{\rm eff}$ is simply the parameter necessary to produce the observed ion stopping power. Nevertheless, in the region in which the z^3 correction is significant there is little justification for use of (11), since (7) cannot give the correct theoretical result. Rather, in order for the concept of effective charge to retain physical significance, it is necessary to include its effect on $\gamma_{\rm rmc}$ in the z^3 corrected stopping power, Eq. (6).

B. Stopping Power

Only one experimental point was obtained for which the energy of the ion leaving the 94.6-μm detector was significantly less than 2 MeV/amu. That point, which occurs in the ^{16}O data, was not used in fitting the energy-loss function since it was strongly affected by range straggling. One is not surprised then by the disparities between S_c and "Expt. fit" at 1 MeV/amu in Table II. The experimental data and the associated "Expt. fit" values obtained from Eq. (1) give stopping powers in the 2–10-MeV/amu region with an uncertainty that we estimate as $\pm 2\%$. Equation (1) should not be extrapolated outside this region with the values of A, r, and n given in Table I.

In the 2–10-MeV/amu region the rms deviation of S_c from the experimental fit is 2.3% for ^{12}C and 5.2% for ^{16}O. For S_b this deviation is 5.8% for ^{12}C and 9.2% for ^{16}O. The S_c deviation for ^{14}N is about the same as that for ^{12}C, but the possible systematic error in the E_i values used in Fig. 1 for this ion prevents attachment of any particular significance to this result. The deviation of the ^{12}C data from the theoretical curve barely exceeds the estimated experimental uncertainty. The disagreement between the ^{16}O data and theory, as embodied in Eq. (5) suggests that some remaining correction to theory, particularly the charge state function, may be necessary as z increases, but the disagreement is insufficient to suggest the exact form for such a correction. A reduction in the value of I would improve the agreement, but a decrease of 5 eV produces only a 1% increase in $S_c(\mathcal{E})$.

As discussed in I, a new z^3 correction was recently calculated by Jackson and McCarthy.[16] While this new correction is patterned after that of Ashley *et al*[2] it differs in details. A preliminary comparison of the new calculation of S_c with our experimental data provides rms deviations of 2.8% for ^{12}C and 5.8% for ^{16}O if their preferred value of $\sqrt{2}$ is used for the Lindhard-Scharff parameter. These deviations are 2.4% for ^{12}C and 5.4% for ^{16}O if a value of $\sqrt{3}$ is used for that parameter. The fit to our ^{14}N data is slightly better than with the Ashley theory.

The percentage differences shown in Table II are due to the experimental error and, to a lesser extent, to the failure to obtain an exact fit of the data by use of Eq. (1). In general, the experimental results show a definite requirement for the z^3 correction to the theoretical approach previously accepted and are, for the most part, in reasonable agreement with the corrected theory.

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