Stopping Power of Some Metallic Elements for 19.8-Mev Protons*

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The proton beam of the UCLA cyclotron, adjusted to 19.8 Mev, was used to measure the stopping power per electron relative to aluminum for 23 elements from Be to Th. An approximate straight-line relationship was found as a function of $\log Z$ with deviations noted from Ta to Th, and from Ca to Fe. The latter deviation seems to be associated with the number of electrons in the $3d$ shell.

INTRODUCTION

IN recent papers[1] deviations have been reported from a smooth variation of stopping power as a function of Z. The anomalies appear to be correlated in some way with the electronic shell structure of the elements. The experiment described herein achieves greater accuracy than the previous work in the vicinity of 20 Mev, so that deviations can be more precisely determined.

The source of protons was the external beam of the cyclotron with an energy spread of approximately 150 kev. Metallic elements which could be obtained in the form of thin foils were used as samples: Be, Ca, Ti, V, Fe, Ni, Cu, Zn, Nb, Mo, Rh, Pd, Ag, Cd, In, Sn, Ta, W, Ir, Pt, Au, Pb, and Th. About 1 sq in. of each of these elements, a few mils thick, was compared with a stack of Al foils of nearly equivalent stopping power. Small variations in effective thickness of either the Al stack or the sample material were achieved by rotating the foil to change the angle of incidence of the proton beam, and adjustment was made so that the energy of the protons emerging from either the sample or the Al stack was the same, as detected by a differential ionization chamber. Whenever possible, direct comparisons were made between elements as a check on the comparisons with Al.

EXPERIMENTAL

To compare two materials it was necessary to move them successively into the path of the beam and adjust the angle of one of them so that the emerging protons had the same energy in each case. The cyclotron beam is subject to rapid variations in energy, both periodic and random, of the order of 0.1%. The apparatus was designed to permit accurate comparisons of foils in spite of these cyclotron variations.

Foil Shuttle

The two foils to be compared were placed in frames, one above the other, on the top of a shaft eccentrically connected to a motor-driven wheel. When in motion this caused the foil holder to move up and down at a rate of 4 times a second with a length of travel such that the beam passed through points from the top of the upper foil to the bottom of the lower one. As the beam was collimated to $\frac{1}{8}$-inch diameter, a $\frac{1}{8}$-inch swath was scanned during the motion. Provision was made for moving the foil holder from left to right in $\frac{1}{8}$-inch steps to permit scanning the entire area. One of the foil frames could be rotated $\pm 10°$ about a horizontal axis by advancing or retracting a micrometer screw. This permitted varying the apparent thickness by 1.5%. Stacks of Al foil were selected whose stopping power did not differ by more than this from the various test foils.

Interposed between the test foils and the ionization chamber was a range-adjusting Al absorber which reduced the energy of the protons to a few Mev so that they would reach the sharply varying portion of the Bragg curve within the chamber and give maximum sensitivity to energy changes. A thickness of absorber appropriate to each sample was selected from a series of 1-mil steps and fine adjustments were made by rotation.

Ionization Chamber

The ionization chamber was a parallel plate type with two plates 3 inches long and 1 inch wide held 1 inch apart on Lucite supports. One of the plates was divided into two sections $1\frac{1}{2}$ inches long, insulated from each other, while the other plate was held at -900 volts. The assembly was placed inside a brass chamber which could be evacuated and filled with a mixture of argon and 5% CO_2. Protons entered the brass chamber through a copper window and a $\frac{1}{8}$-inch collimating hole, traversing the two sections in series. Since protons emerge from the cyclotron in bursts, pulses of ionization current were collected in the chamber. The relative size of the electron pulses from the two halves of the divided plate depended upon the position of the combined Bragg and straggling curve within the chamber. They could be made equal by varying the thickness of the range-adjusting absorber. These pulses were applied to the grids of a difference amplifier and the output was fed directly to the vertical plates of an oscilloscope.

A dc voltage, taken from a potentiometer which rotated with the motion of the foil holder, was fed to

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[1] C. P. Sonett and K. R. MacKenzie, Phys. Rev. **100**, 734 (1955); Bader, Pixley, Mozer, and Whaling, Phys. Rev. **103**, 32 (1956); Green, Cooper, and Harris, Phys. Rev. **98**, 466 (1955).

the horizontal plates of the oscilloscope. This caused the electron beam to sweep back and forth across the screen as the foils moved up and down. The pattern therefore displayed, side by side, pulses from protons which passed through points from the top of the upper foil to the bottom of the lower one. This made it possible to observe differences of 0.1% in the stopping thickness of the two foils as well as to exhibit nonuniformities in the individual foils which could be partially overcome by visual averaging. Some foils were found to have as much as 2% taper. Accurate comparisons of the order of 0.1% could not, of course, be made in these cases.

Measurement

To make a comparison, the range-adjusting absorber was rotated to give minimum differential pulse height when the beam traversed the thicker foil. The thinner foil was then rotated to give the same pulse height. Measurements were made of the required angle of rotation on each side of the vertical and an average value used. Small periodic fluctuations in the energy of the beam could be seen to put an envelope on the pulse heights but did not invalidate comparisons. Large random fluctuations due to sparking were easily detected and measurements were made during periods of steady operation.

Foil Preparation

The samples were prepared by clamping pieces of the material between aligned steel blocks, which had been ground and polished as a pair. The excess foil was then shaved off with a razor blade. This technique was found to give smooth edges and the same area for every foil cut to within 0.1%. Each was measured with a Bausch and Lomb comparator to determine the width and length to 0.0002 inch, or the area to 0.04%. The foils were cleaned and weighed both before and after the comparisons to 0.05 mg, corresponding to 0.03% for the lightest material and 0.01% for the heaviest.

TABLE I. Results of aluminum comparisons.

	Individual foil values for stopping power per electron relative to aluminum				Average value	Relative mass stopping power
Be	1.166	1.166			1.166±0.005	1.073±0.005
Ca	0.982	0.983			0.970±0.020	1.008±0.020
Ti	0.938	0.933	0.932	0.941	0.936±0.006	0.888±0.006
V	0.918	0.918	0.918	0.918	0.918±0.002	0.860±0.002
Fe	0.886	0.886	0.884		0.886±0.002	0.856±0.002
Ni	0.871	0.872	0.872		0.872±0.002	0.863±0.002
Cu	0.867	0.867	0.868		0.867±0.002	0.821±0.002
Zn	0.866	0.867	0.866		0.866±0.002	0.819±0.002
Nb	0.816	0.817			0.816±0.002	0.746±0.002
Mo	0.807	0.811			0.809±0.005	0.735±0.005
Rh	0.805	0.795			0.800±0.007	0.726±0.007
Pd	0.791	0.790	0.795	0.794	0.793±0.003	0.710±0.003
Ag	0.789	0.792	0.793		0.791±0.003	0.715±0.003
Cd	0.789				0.789±0.002	0.700±0.002
In	0.782+	0.781			0.782±0.002	0.693±0.002
Sn	0.782	0.782			0.782±0.002	0.680±0.002
Ta	0.719	0.718	0.710		0.716±0.005	0.597±0.005
W	0.706	0.707	0.706		0.706±0.002	0.590±0.002
Ir	0.702	0.704	0.703		0.703±0.002	0.581±0.002
Pt	0.696	0.696	0.693		0.695±0.002	0.576±0.002
Au	0.691	0.694			0.692±0.003	0.576±0.003
Pb	0.679	0.675			0.677±0.003	0.556±0.003
Th	0.691	0.692			0.691±0.002	0.556±0.002

TABLE II. Results of cross checks.

Element No. 1	Element No. 2	Direct experimental ratio of stopping power	Ratio of element 1 to element 2 calculated from Al comparison
Zn	V	0.938±0.002	0.943±0.003
Cu	Fe	0.975±0.004	0.979±0.003
	(4 separate pairs of foils)	0.976±0.003	
		0.972±0.005	
		0.987±0.007	
Cu	Ni	0.991±0.002	0.994±0.003
		0.992±0.002	
		0.994±0.002	
	(7 separate pairs of foils)	0.994±0.002	
		0.995±0.002	
		0.993±0.002	
		0.992±0.002	
Pd	Mo	0.983±0.005	0.980±0.005
Pd	Cd	1.011±0.005	1.005±0.003
Sn	Cd	0.994±0.002	0.991±0.003
Pd	Rh	0.996±0.005	0.991±0.008
Rh	Nb	0.989±0.005	0.980±0.008
Au	Ta	0.980±0.005	0.972±0.005
W	Pt	1.017±0.002	1.017±0.003
W	Ir	1.013±0.008	1.004±0.003
Th	Ni	0.792±0.002	0.792±0.003

ERRORS

The uncertainty in measurement of the surface density of each foil was ±0.05%. The uncertainty in the surface density of the stacks of Al was ±0.05%. The uncertainty in setting the angle θ of the rotated foil led to an uncertainty in $\cos\theta$ of 0.15% for the uniform foils. Thus the total probable error is ±0.19%, except for the most nonuniform foils, where the angular setting was much more in question and the probable error is estimated as 0.5%. These values are based on repeated measurements of individual foils. The deviations in results from various runs fall within the probable error.

Spectroscopic analysis showed the materials to be pure to within 0.25% and with impurities of similar Z, so that the error introduced was less than 0.1%.

In the case of Ca, which oxidized rapidly, the amount of oxygen present at the middle of the run was estimated, on the basis of the weight gained during the measurement (about 2%) and a chemical determination of the amount of Ca in a fresh sample of the foil (about 95%). The large error assigned to calcium is based on foil nonuniformities and also upon uncertainties in the amount of oxidation and absorption of water of crystallization by CaO during the run.

RESULTS

As different amounts of energy were lost in the various foils, all relative stopping power values are corrected to 19.8 Mev. Table I shows the results of comparisons

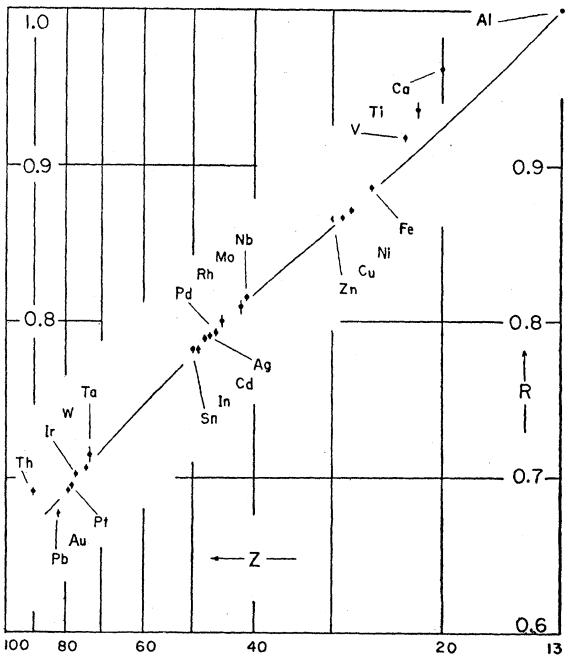

FIG. 1. Stopping power per electron relative to aluminum, R, as a function of log$Z$. The Be point is not shown on this graph but appears in Fig. 2. A smooth curve has been drawn through those points which appear to show a regular behavior of I/Z.

of different samples of each material with Al. Average values of relative stopping power per electron and relative mass stopping power are given.

Table II shows the results of the cross checks in which direct comparisons were made between elements. Seven sets of selected foils from a plentiful stock were used in the Cu−Ni cross check and 4 sets in the Cu−Fe check. In general the foils were not quite as uniform as the selected samples used for the aluminum comparison. These runs served to indicate the accuracy of the method as well as to reduce the error for these points.

INTERPRETATION AND CONCLUSIONS

In Fig. 1, relative stopping power per electron is plotted as a function of log$Z$. Values of the mean ionization potential for a few selected elements have been calculated from the present data by David O. Caldwell of the Massachusetts Institute of Technology. The results are shown in Table III. Corrections for nonparticipating electrons were obtained from the papers of Walske.[2] In the heavier elements Caldwell made at least rough estimates of the corrections even up to the O shell. His calculations were based on an I value for aluminum of 166.4 ev from the 18-Mev total range data of Bichsel and Mozley.[3] With the exception of Be the general trend can be fitted by I/Z approximately equal to 12.6. An estimate based on the direct energy loss measurement of Sachs and Richardson,[4] with a corrected[5] I value of 163.1 ev for aluminum, implies a slightly lower value of about 12.4 for most of the elements.

Some sort of estimate of I/Z can be made from the present relative measurements alone. If the values for Be and Pb are ignored (the value for Pb must be either anomalous or in error—see Fig. 1), the rest of the elements in Table III, which might be called typical, can be fitted within 1% by an I/Z value of 12.0. However, very little weight can be put in this estimate, since the shell corrections in the heavy elements are hardly accurate enough to justify this procedure, and the assumption must be made that a constant value of I/Z exists, which appears to be very unlikely in view of the fluctuations reported in this paper. The extremes range from 11 for Ti to 13 for Pb. The low value of I/Z for Ca is not stressed as being significant, since the deviation is actually only a little greater than the large estimated uncertainty. The high value of 16 for Be ($I=64$ ev) agrees within experimental error with the value of 64 ev predicted by Bohr[6] on the basis of the polarization effect. It appears that I/Z for most elements is significantly higher than 11.5, which is the value that enters into the calculation of the current range-energy curves.[7]

The points in Fig. 1 are shown again in Fig. 2 with ordinates and abscissa multiplied by functions of particle velocity and other factors. This method of plotting, due to Lindhard and Scharff,[8] allows comparison of stopping-power data taken at different energies and with different particles. Some of the measurements, by other investigators, in the vicinity of 18 Mev were on an absolute basis, but the majority were made relative to aluminum and have been normalized to aluminum with an I value of 163 ev. A line through the present data, normalized on this basis, fits the absolute data of Brolley and Ribe[9] at 8.86-Mev deuteron energy and 4.43-Mev proton energy. Almost all previous measurements in this intermediate energy range can be fitted within experimental error to this same line, which has

TABLE III. Ionization potentials after application of shell corrections. All relative data have been normalized to an aluminum value of 166.4 ev.

	Be	Al	Fe	Cu	Ag	W	Au	Pb
I (ev)	64.0	166.4	328.8	366.0	587.0	920.5	997	1070
I/Z	16.0	12.8	12.6	12.6	12.5	12.4	12.6	13.0

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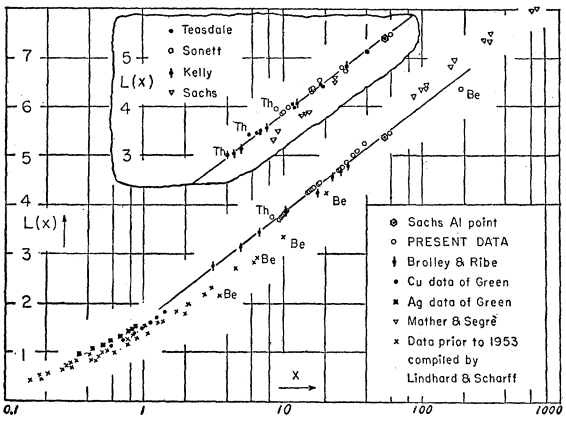

FIG. 2. The stopping power per electron $L(x)=(1/NZ)(dE/dx)\times(mv^2/4\pi z^2e^4)$ is plotted against $x=(v^2h^2)/(e^4Z)$. All relative data in the 12- to 32-Mev range have been normalized to $I=163$ ev for aluminum. In order to display individually all points in this energy range, some of the previous data are shown displaced vertically.

been displaced vertically in Fig. 2 to allow separate points to be seen. Shown are the relative data of Kelly[10] (37-Mev α's), Teasdale[11] (12-Mev protons, obtained in this laboratory), Sonnet[1] (20-Mev protons), and the absolute data of Sachs[4] (18-Mev protons). Only the values for six heavy elements, Rh through Au, obtained by Sachs, show an obvious deviation. These deviations, if real, would indicate that I/Z should be about 14 for heavy elements.[5] Richardson[12] has pointed out that a tedious and hardly worthwhile correction should be applied to these points, arising from angular multiple scattering for the heavier elements in the nonuniform magnetic field of the cyclotron which was used to determine the energy loss. The correction is not considered to be significant for aluminum, and hence the aluminum I value of 163 ev was used in normalizing the data shown in Fig. 2.

Disagreement is also noted for some elements, shown on the original Lindhard and Scharff plot, which overlap the 4.43-Mev data. These are chiefly measurements of Madsen[13] at energies of 2 Mev and below. However, recent data of Green, Cooper, and Harris[1] and Chilton, Cooper, and Harris,[14] at energies up to 1 Mev, come much closer to an extrapolation of the line. Their points also make it appear that no single line can represent all data at low energy. The high-energy data of Mather and Segrè[15] from 340-Mev proton range data, as pointed out by Caldwell,[5] also differs from the low-energy measurements.

The variation of stopping power per electron with atomic number agrees in general with former measurements, but the accuracy is such that individual element variations are observed that seem to be well outside experimental error. Efforts were made to correlate these variations with physical properties such as density, conductivity, etc., without success, leaving the electronic shell structure as the most likely causative factor. The most striking anomalies occur for Ca ($3d^04s^2$), Ti ($3d^24s^2$), and V ($3d^34s^2$), which have been hitherto unmeasured in this energy range. Calcium is unique in the periodic table, being the first element with a completely empty d shell followed by a full s shell.

Similar but smaller effects might be expected as the $4d$ shell is filled. Unfortunately, of the three foils that could have shown such an effect, Nb ($4d^45s$), Mo ($4d^55s$), and Rh ($4d^85s$), the latter two were not very uniform, so no changes in slope could be determined. In the heavy elements, Ta through Pb, the slope of stopping power *vs* log$Z$ appears to increase, with the exception of Th which is anomalously high. All previous workers have found a high value for Th (see Fig. 2). Th ($6d^27s^2$) is followed by Pa ($5f^26d7s^2$), showing that the outer electron configuration in this region of the periodic table is not normal and could be responsible for the high value of stopping power.

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