

Stopping of Low-Energy H⁺ and He⁺ Ions in Plastics*†

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The stopping cross sections of polystyrene, polyethylene, and Pliolite S-5A for 40- to 340-keV H⁺ and He⁺ ions have been measured. A nearly monoenergetic analyzed beam of ions of known energy was deflected 90° in the field of a magnetic spectrometer. A thin film of the stopping material was placed in the beam path at the spectrometer entrance. The energy loss of the beam was determined by measuring the decrease in magnetic field necessary to restore the beam to the 90° deflection angle. The film thickness was obtained by weighing a known area of film. The probable errors in the measurements are 2.3–2.8% for H⁺ and 3–4% for He⁺. Proton stopping cross-section measurements on gold with this equipment agree with the results obtained by Bader, Pixley, Mozer, and Whaling. Above 75 keV the He⁺ cross sections vary linearly with the ion velocity.

I. INTRODUCTION

THE need for accurate low-energy proton and alpha stopping cross sections in hydrocarbon solids has been pointed out by Zimmerman.¹ Proton stopping cross sections have been measured by Reynolds *et al.*,² in the energy range of 30 to 600 keV in hydrocarbon gases. These data indicate that the Bragg rule of additivity does not apply to hydrocarbon gas molecules for proton energies below 150 keV. Furthermore the difference between the stopping powers of H₂O vapor² and D₂O ice³ suggests that there may be a dependence on the physical state. Since the dependence of the stopping cross section on physical state and molecular structure is not known, the cross sections of solid hydrocarbons cannot be calculated reliably from existing data. Because it is possible to obtain very thin self-supporting films of certain plastics, an experimental determination of the proton stopping cross sections for the following three solid hydrocarbons has been made: polyethylene (CH₂)_n, polystyrene (C₃H₃)_n, and Pliolite S-5A (C₁₂H₁₄)_n. A simultaneous determination of the alpha-particle stopping cross sections was carried out for the same materials.

II. METHOD

Plastic films for this experiment were cast on clean glass microslides by the method of Revell and Agar.⁴ The solvent for polystyrene and Pliolite S-5A was benzene and warm toluene was used for polyethylene. Films uniform in thickness over an area of more than 5 cm² and ranging from 5 to 50 μg/cm² were produced.

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¹ E. J. Zimmerman, *Phys. Rev.* **99**, 1199 (1955).

² Reynolds, Dunbar, Wenzel, and Whaling, *Phys. Rev.* **92**, 742 (1953).

³ W. A. Wenzel and W. Whaling, *Phys. Rev.* **87**, 499 (1952).

⁴ R. Revell and A. Agar, *Brit. J. Appl. Phys.* **6**, 23 (1955).

The uniformity was easily determined by observing the variation of interference color across the film when it was floating on water. Before the film was removed from the slide a suitable region was selected and a predetermined area was cut out. The film was then floated off on water from which it was mounted on an aluminum frame.

To determine the weight of the films a Rodder model E quartz fiber microbalance⁵ was used. The balance combines an equal arm beam and a torsion fiber to obtain a ratio of load capacity to sensitivity of 4×10⁶. This feature was desirable in order to determine to 1% or better the weight of 100 μg films mounted on 30 mg frames. The balance was calibrated with weights of 0.5, 1, and 2 mg certified by the National Bureau of Standards to 0.5 μg. Reproducible weight measurements were consistently obtained within ±0.15 μg or 0.2%, whichever was larger. In order to manipulate and weigh the films, they were mounted on frames of 2-mil aluminum foil each with 3 windows of 0.5 cm² open area. Films of 5.32 cm² were mounted on these frames and were self-supporting across the windows. The films were transferred from the water surface to the frame by pulling the immersed frame vertically out of the water with the film attached to the top edge. The frames were weighed before and after mounting the film to obtain the film weight.

Since the films were weighed in air it was necessary to investigate whether the water absorption by the plastics would seriously affect the weight measurements. A simple quartz-fiber microbalance which was capable of determining weight changes to 0.5 μg was constructed and placed in a vacuum system. With this balance it was observed that the weight of the film generally increased almost immediately when the mechanical fore pump was turned on. It was also observed that further weight increase occurred if the film was left in the vacuum system for a period of one day or longer at a pressure of 10⁻⁵ mm of Hg. A similar weight increase occurred on films which had been in the energy-

⁵ Obtained from Microtech Services Company, Los Altos, California.

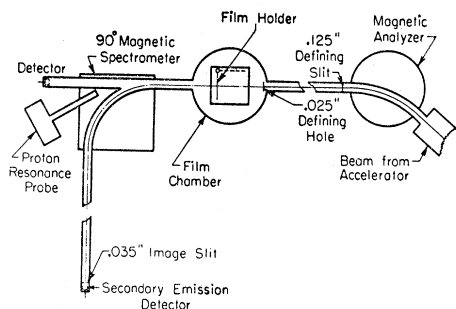


FIG. 1. Schematic diagram of apparatus.

loss vacuum chamber. The weight increase was independent of the film thickness, indicating that it was a surface phenomenon. Moreover, the initial increase during fore-pump evacuation could be eliminated by inserting a trap kept at -78°C between the pump and the chamber. It is well known that unbaked metal vacuum systems pumped on by untrapped oil pumps have a residual atmosphere of organic and water vapor.⁶ Hence it is believed that the weight increase was due to deposition of organic vapors of the vacuum system on the film and frame surfaces. By using the trapped system it was possible to establish that no weight change greater than 1% occurred on $40\text{-}\mu\text{g}/\text{cm}^2$ films due to removal of the atmosphere. For polystyrene and polyethylene this is in agreement with the work of Day⁷⁻⁹ who has studied water absorption of thin films of these materials with a vacuum torsion balance. The weight increase of the films due to organic vapor deposition could not be eliminated but a discussion of how it was minimized follows later in this section.

Beams of protons and helium ions from 30 to 350 keV were produced by the Nebraska Cockcroft-Walton accelerator. To prevent damage to the films, low-intensity beams were required. These were obtained by operating the radio-frequency ion source at low power with helium gas. Residual hydrogen in the accelerator was always adequate to supply a proton beam. The beam intensity was adjusted to a desired value for each energy by adjusting the rf input power while the accelerator was in operation. The accelerating voltage was monitored and regulated to 0.01% by means of a high-voltage resistance divider and electronic feedback system.¹⁰ The feedback system compares a small fraction of the accelerating voltage with a preset voltage on a type *K* potentiometer and it is thereby ultimately compared with a standard cell. The accelerating voltage is thus determined by the ratio of the resistances in the high-voltage resistance divider.

⁶ J. Blears, *J. Sci. Instr. Suppl.* No. 1, 36 (1951).

⁷ A. G. Day, Report British Elect. Research Association, L/T 292, 1953 (unpublished).

⁸ A. G. Day, Paper presented at the Llandudno Conference on Film Structure and Adhesion, June 7-11, 1955 (unpublished).

⁹ A. G. Day (private communication).

¹⁰ C. J. Cook and W. A. Barrett, *Rev. Sci. Instr.* **24**, 638 (1953).

Before entering the energy-loss chamber, the beam was analyzed by deflecting it 40° in a magnetic field.

The energy loss ΔE of an ion beam passing through a thin film was measured with the aid of a 90° magnetic spectrometer. Figure 1 shows the general arrangement of the apparatus and the location of the beam apertures. The resolution obtained with the illustrated system was well suited to the energy spreads encountered in both the incident and exit (from the film) beams. The magnetic field was determined with a proton resonance magnetic field meter. The transitron circuit of Hahn and Knoebel¹¹ was used as a basis in the construction of this meter. The resonance frequency was measured with a General Radio 620-A frequency meter in the 10 to 45 Mc/sec range and a Signal Corps BC-221 meter in the 5 to 10 Mc/sec range. Both of these instruments were capable of an absolute accuracy of 0.01%. A secondary emission detector in which the electrons released when the proton beam impinged on a brass emitter were collected was used to detect the beam. The current was amplified with a conventional high-

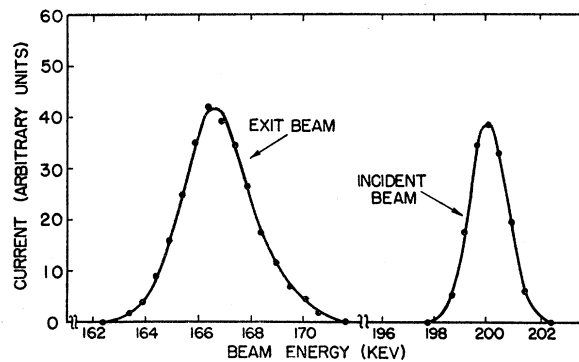


FIG. 2. Energy distributions of an incident and exit 200-keV proton beam from a $42.9\text{-}\mu\text{g}/\text{cm}^2$ polystyrene film.

impedance dc amplifier and detected with a sensitive galvanometer. The system was capable of detecting a beam of 10^{-14} ampere. An ordinary Faraday cup detector collecting the beam from the analyzing magnet was used to determine the proper alignment and focus of the beam.

The spectrometer was used to obtain the energy loss ΔE in terms of the initial energy E_1 determined from the accelerating potential. The spectrometer field was measured with the film in and out of the beam and ΔE was calculated by assuming $E = kB^2$, where B is the spectrometer magnetic field and k is a constant. The assumption that k is a constant was tested by plotting the accelerating voltage setting against B^2 over the complete energy range for both protons and singly charged helium ions. The plot was found to be linear within $\frac{1}{2}\%$ for protons and within 1% for helium ions. One reason that k might not be constant is that the

¹¹ H. Knoebel and E. Hahn, *Rev. Sci. Instr.* **22**, 904 (1951).

fringing field outside the pole gap might not be strictly proportional to the field in the gap. This will happen when the magnet becomes saturated. The maximum field, required for the 350-keV He⁺ beam, was 8600 gauss which is well below saturation for mild steel of which the magnet was constructed. It thus seems reasonable to assume that k is constant over the entire energy range for protons which require a maximum field strength of only 4300 gauss. The second reason is that the beam energy may not be strictly proportional to the accelerating voltage setting. That it is proportional, however, follows from the fact that k is constant for protons and that the plot of B^2 vs E is linear over the entire energy range for protons. Hence k must be constant within 1% to at least 8600 gauss because of the linearity of the B^2 vs E curve for He⁺ ions. Below 100 keV, where the high-voltage resistance divider is not appreciably disturbed by corona and leakage, the beam energy is within 1% of the high-voltage setting. Since the beam energy is a linear function of the voltage setting over the whole energy range the energy is taken

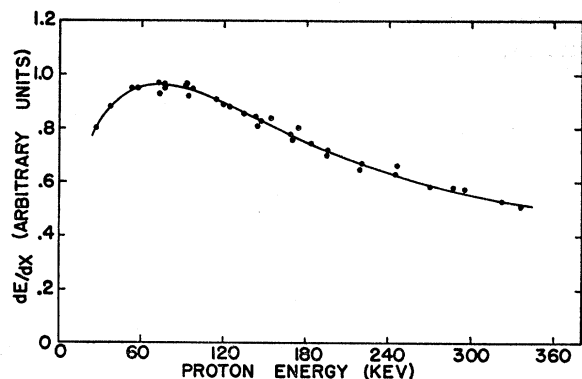


FIG. 3. "Shape curve" for protons on polystyrene.

as being within 1% of that voltage setting for all energies. Thus the energy loss ΔE was calculated from the equation $\Delta E = E_1(\Delta B/B_1)(2 - \Delta B/B_1)$, where $\Delta B = B_1 - B_2$ and B_1 and B_2 correspond to E_1 and E_2 , the incident and exit energies, respectively.

The energy distributions of an incident and exit 200-keV proton beam are shown in Fig. 2. The energy loss ΔE is defined as the difference between the energies of the peaks of these distributions. In measurements only the peaks were determined by observing the detector response in the region of the peak as a function of the spectrometer field. The stopping cross section is then given as $\Delta E/\Delta x$ at $E = (E_1 + E_2)/2$, where Δx is the film thickness.

The film was mounted on a film holder in the film chamber which could be rotated into and out of the path of the beam. External controls also allowed the film to be moved horizontally and vertically with respect to the beam so that different portions of the film could be exposed to the beam. Incident beam

TABLE I. Estimated uncertainties in absolute proton cross sections.

1. Balance calibration	$\pm 0.2\%$
2. Balance reproducibility	$\pm 0.2\%$
3. Film contamination	$\pm 1.0\%$
4. Film area	$\pm 0.4\%$
5. $\Delta B/B_1$	$\pm 0.6\%$
6. Energy	$\pm 2.0\%$
7. Impurity	$\pm 0.5\%$

currents of 10^{-9} to 10^{-11} ampere were used without noticeable damage to the films in the time necessary to make a measurement. This was ascertained in two different ways. Films which were known to be uniform in thickness because of their uniform interference color sustained the same ΔE on all portions of the film. Moreover since the energy loss could be continuously observed, any systematic change in ΔE was quickly detected as a change in the position of the peak. These observations further demonstrated that there was no appreciable contamination of the film surface due to the polymerization of adsorbed organic vapors by the beam.

In order to minimize the effects of surface contaminants already discussed which caused the weight of the films to increase while they were in the vacuum system, the following method was used. In the low-energy region, where relatively thin films ranging from 6 to 10 $\mu\text{g}/\text{cm}^2$ were necessary, only relative cross sections were obtained. Films of 15 to 20 $\mu\text{g}/\text{cm}^2$ were used for relative measurements at higher energies. The data from several films were fitted together at common energy points and a smooth curve was drawn through the points. Figure 3 shows a typical "shape" curve. Although the hydrocarbon contamination on the thinnest films might be as much as 10% of the film thickness, none of the curve shapes of hydrocarbon cross sections differ by more than 10% so that at most a 1% error is made in the shape. Absolute cross-section measurements for protons were made at 200 keV with films of 40 to 50 $\mu\text{g}/\text{cm}^2$. The weight increase for such films was usually less than 1%. In order to obtain an average ΔE for the films used in absolute measurements the energy loss was determined at twelve different points on the film. The mean of the absolute cross sections of two films was used together with the relative values of the "shape" curve to obtain absolute cross sections at all other energies.

Because of the larger energy losses sustained by He⁺ ions it was difficult to make measurements on films

TABLE II. Final uncertainties in stopping cross sections.

	Protons	Alphas	
1. Polystyrene	$\pm 2.3\%$	$\pm 3.0\%$	
2. Polyethylene	$\pm 2.3\%$	$\pm 4.0\%$	< 100 keV
		$\pm 3.0\%$	≥ 100 keV
3. Pliolite S-5A	$\pm 2.8\%$	$\pm 3.5\%$	

TABLE III. Proton stopping power (kev-cm²/microgram) of plastics.

Energy (kev)	Polystyrene	Polyethylene	Pliolite S-5A
25	0.806
30	0.864
32	...	0.984	...
40	0.931	1.040	...
50	0.973	1.083	...
54	0.984
60	0.994	1.108	1.006
70	1.002	1.119	1.024
80	0.998	1.119	1.028
90	0.992	1.105	1.023
100	0.978	1.083	1.010
120	0.926	1.034	0.968
140	0.879	0.990	0.915
160	0.832	0.946	0.866
180	0.780	0.900	0.823
200	0.738	0.856	0.786
225	0.689	0.804	0.743
250	0.644	0.755	0.704
275	0.604	0.711	0.669
300	0.578	0.676	0.635
325	0.551	0.648	0.604
335	0.540	0.640	0.592

thicker than 20 $\mu\text{g}/\text{cm}^2$ with these ions. Hence the absolute stopping cross section for He⁺ ions was obtained from the proton curve by measuring the ratio of the ΔE 's of He⁺ ions and protons in several films. This ratio together with the proton cross section and the He⁺ "shape" curve determines the stopping cross sections at all energies for helium ions.

III. ACCURACY

Table I gives the estimated errors contributing to the final uncertainty in the measured absolute proton cross sections. The main source of uncertainty in the weight determination was the contamination due to water vapor and hydrocarbon vapor. Each of these effects contributed 1.0% or less, but their effects on

TABLE IV. Alpha stopping power (kev-cm²/microgram) of plastics.

Energy (kev)	Polystyrene	Polyethylene	Pliolite S-5A
35	...	0.98	...
40	0.96	1.07	1.05
50	1.10	1.22	1.17
60	1.21	1.33	1.26
70	1.30	1.43	1.34
80	1.39	1.52	1.41
90	1.46	1.60	1.48
100	1.53	1.67	1.54
120	1.63	1.81	1.66
140	1.74	1.92	1.77
160	1.83	2.03	1.87
180	1.92	2.12	1.96
200	2.00	2.22	2.05
225	2.11	2.33	2.15
250	2.20	2.43	2.25
275	2.29	2.52	2.34
300	2.36	2.61	2.43
320	2.43	2.68	2.50
340	2.49

ΔE tend to cancel, so it is believed that 1.0% is a reasonable estimate for this uncertainty. The estimate of the $\Delta B/B_1$ uncertainty was obtained from the spread of 12 measurements at different spots on a film of uniform thickness (no interference color variations over the film). The uncertainty in the energy is 1.0% but it contributes twice to the error in the absolute cross section. The first contribution enters in the equation for ΔE . The second contribution results from transferring the error in the independent variable (E) to the dependent variable (stopping cross section). Thus the absolute stopping cross-section data can be properly combined with the relative data and all of the cross sections presented as a function of definite energies. This contribution is pessimistically taken as 1%. Since the two energy contributions are not independent they must be added before being combined with the other uncertainties. The 0.5% contribution of impurities is an estimate of the effect of the impurities in the raw film material and those introduced during film production. The final uncertainty in the proton absolute cross section is then $\pm 2.4\%$ which is reduced to $\pm 1.7\%$ because measurements on two films of each material were averaged. In the case of Pliolite S-5A, however,

TABLE V. Proton stopping power of gold.

Energy (kev)	dE/dx (kev-cm ² /microgram)
184	0.121 \pm 0.003
235	0.119 \pm 0.003
286	0.111 \pm 0.003
326	0.107 \pm 0.003

one of the films was considerably nonuniform so that an error of $\pm 2.4\%$ was assigned. The uncertainties in the relative values of the cross sections at different energies was estimated from the scatter of the points about the best curve drawn through them to be about 1.5%. The final uncertainties are given in Table II.

The uncertainty in the alpha-particle data must include the errors for the proton stopping cross sections as well as the error in the ratio measurements and shape determinations. The ratio measurements contribute 1.3% and the shape determinations contribute 1.5% except for polyethylene below 100 kev where the shape uncertainty is 3.0%. The final uncertainties in the alpha stopping cross sections are also shown in Table II.

IV. RESULTS

Tables III and IV give the results of these measurements. These tabulated values were obtained from the smooth "shape curve" adjusted to the mean value of the measured absolute cross section.

Stopping cross sections of gold were obtained in order to compare the results with those of other workers in the field. Gold films were evaporated on glass slides, floated off on water, mounted and weighed in a manner

similar to that of the plastic films. The results of the measurements are given in Table V. These results are in excellent agreement with those obtained at the California Institute of Technology.¹² Earlier measurements on gold carried out in Chicago and in Denmark¹³ are about 20% lower than the Caltech measurements. However, another recent determination by Green, Cooper, and Harris of Ohio State¹⁴ agrees to 1.5% with the Caltech data in the interval of 400 to 600 kev where they overlap. Furthermore, because of this discrepancy the Caltech group has checked the internal consistency of their measurement by determining the ratio of the stopping power of gold to copper by two independent methods.

The stopping power of gold for alpha particles at 299 kev was found to be 0.316 ± 0.008 kev-cm²/μg. The ratio of the alpha stopping cross section to the proton stopping cross section at 300 kev in gold is 2.87 ± 0.09 . This ratio agrees with the less precise value of 3.02 ± 0.45 obtained by Wilcox.¹³

Figure 4 shows the helium ion cross sections plotted as a function of the ion velocity. Except possibly at energies below 75 kev, these graphs are straight lines. Although the slopes of the lines all differ slightly they are equal within error. The straight lines extrapolate to positive values of dE/dx at zero energy and these intercepts are also equal within error. The velocities are too high for the considerations of Fermi and Teller,¹⁵ giving such a linear dependence, to be valid.

We do not here give an analysis of the proton cross

¹² Bader, Pixley, Mozer, and Whaling, *Phys. Rev.* **103**, 32 (1956).

¹³ S. K. Allison and S. D. Warshaw, *Revs. Modern Phys.* **25**, 779 (1953).

¹⁴ Green, Cooper, and Harris, *Phys. Rev.* **98**, 466 (1955).

¹⁵ E. Fermi and E. Teller, *Phys. Rev.* **72**, 399 (1947).

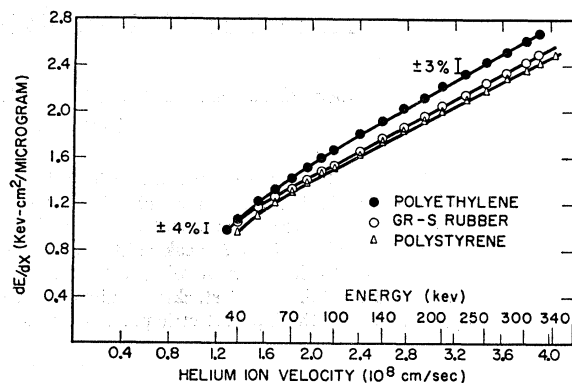


Fig. 4. Helium ion cross sections as a function of ion velocity. (Note.—“GR-S rubber” should read “Pliolite S-5A.”)

sections in terms of atomic cross sections (using some additivity rule to construct the molecular cross section) because we hope to present a complete analysis of these data, together with the corresponding data of Reynolds *et al.*,² in a forthcoming paper, in which we propose an additivity rule for hydrocarbons which is a modification of the Bragg rule. In terms of that analysis our data are completely consistent with those of Reynolds *et al.*

V. ACKNOWLEDGMENTS

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