Stopping Cross Sections of Some Hydrocarbon Gases for 40–200-keV Helium Ions*

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The stopping cross sections of air, CH_4, C_2H_2, C_2H_4, C_3H_8, $(CH_2)_3$, and C_3H_6 have been measured for incident helium ions in the energy range 40 to 200 keV. The data are in good agreement with previous experiment work, and have standard deviations between 2 and 6%. They obey an energy dependence $\epsilon=\beta E^{1/2}$ in accord with the calculation of Lindhard and Scharff. It has been shown that while there is probably an effect due to chemical binding on the helium-ion stopping cross sections of hydrocarbons, this effect is small.

I. INTRODUCTION

IN spite of the long-standing interest in the energy loss of helium ions and alpha particles, the stopping cross sections of very few materials have been measured. None of the scant data available on gases had been studied in the low-energy region by more than one investigator; thus, no check on the reliability of the results was possible.

In this experiment the stopping cross section of air has been measured to permit comparison with the experiment of Weyl[1] and the stopping cross section of ethylene has been remeasured with the present apparatus to permit comparisons with the measurements taken earlier at this laboratory. The stopping cross sections of methane, acetylene, propane, propylene, and cyclopropane were then measured to attempt to bridge the gap in helium-ion stopping-cross-section data and to permit studies of the Bragg rule with respect to low-energy helium ions. A differentially pumped gas cell containing a known length of gas at a known temperature and pressure was placed between analyzing and spectrometer magnets. A monoenergetic beam of helium ions was deflected 90° in the field of the magnetic spectrometer when the gas cell was evacuated. The energy change of the singly charged helium-ion component of the exit beam when a gas was admitted to the stopping cell was determined by measuring the decrease in the magnetic field necessary to restore the beam to the original 90° deflection angle. Previous work concerning energy-loss cross-section measurements for helium ions has been reviewed by Whaling.[2]

II. APPARATUS

The helium-ion beams used in this experiment were produced by the University of Nebraska Cockcroft-Walton accelerator,[3] using a radio-frequency ion source. The accelerating voltage was held to within 0.01% of the desired voltage.[4]

The stopping device consists of a differentially pumped gas cell, shown in Fig. 1. The stopping cell is made of a 3-in. o.d. brass tube 10.019±0.001 cm in length. The ends of this tube are sealed with two disks containing 0.025-in.-diam apertures. The sum of the separations between the stopping cell and the ends of the first differential pumping section is 1.985±0.002 cm. The stopping cell is differentially pumped by two 550-liter/sec diffusion pumps.

A 0.025-in.-diam defining aperture is located 50 cm from the end of the stopping cell between the stopping cell and the spectrometer magnet. This aperture narrowed the angle of acceptance to 5 min.

The pressure regulators on the tanks containing the gases to be tested were connected through shutoff valves to an isolation bubbler and a ballast chamber. A valve connected this chamber to a liquid-nitrogen cold trap and a mechanical pump which served to pump the manifold, lines, and regulators free of residual gases. The ballast chamber was connected through a metering valve to the stopping cell.

The temperature of the stopping cell was measured by a mercury thermometer in contact with the 5½-in. tube surrounding the gas cell.

The pressure of the gas in the stopping cell was measured by a Decker pressure meter[5] which was carefully calibrated by McLeod gauges. The output of the Decker pressure meter was connected to a recording potentiometer. A displacement in the pressure equilibrium of 0.1 μ resulted in a 2-mV output signal. In normal running conditions a change of 0.1 mV could be detected and the potentiometer rebalanced to elimi-

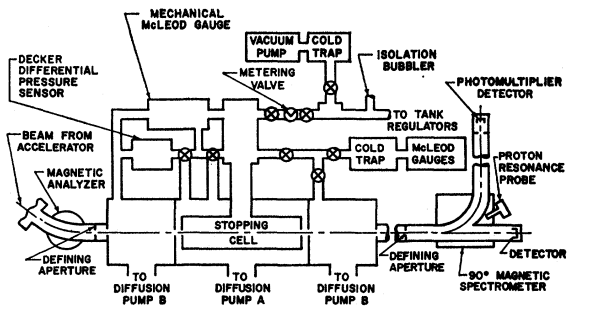

FIG. 1. Diagram of stopping cell and associated equipment.

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† Present address: University of Missouri at Rolla, Rolla, Missouri.
[1] P. K. Weyl, Phys. Rev. **91**, 742 (1953).
[2] W. Whaling, in *Handbuch der Physik*, edited by S. Flügge (Springer-Verlag, Berlin, 1958), Vol. 34, p. 193.
[3] C. J. Cook and W. A. Barrett, Rev. Sci. Instr. **24**, 638 (1953).
[4] L. H. Sohl, Master's thesis, University of Nebraska, 1960 (unpublished).
[5] Model 306-2F, 0 to +0.3 in. H_2O, The Decker Corporation, Bala-Cynwyd, Pennsylvania.

nate it. The drift in zero was negligible, being less than $\pm 0.1\ \mu$/h.

The pressure in the first differential pumping section was measured by a mechanical McLeod gauge.[6] It was found to be unnecessary to measure this pressure for all the readings, as the pressure in this region was a constant fraction of the stopping-cell pressure.

Two McLeod gauges were used as the ultimate pressure reference. One was constructed and calibrated in an earlier experiment,[7] and the other McLeod gauge[8] was calibrated by the manufacturer. The two gauges agreed within 0.6%, and the average reading was used.

The pressure of the outer differential pumping sections as measured by an ion gauge remained below 5×10^{-5} mm Hg.

The energy loss of the beam in passing through the gas sample was determined with the aid of a 90° magnetic spectrometer. The current in the magnet coils could be held constant to 0.01% for periods of 15 min or more. The magnetic field was measured with a proton resonance magnetometer based on the transitron circuit of Knoebel and Hahn.[9] The resonance frequency was measured with a General Radio type 620A frequency meter. The frequency meter could be calibrated against a crystal oscillator at no less than three places on each range of the scale. It was generally possible to determine the resonance frequency to better than 0.002 Mc/sec.

The beam of helium ions was detected by an electron multiplier (DuMont 6292) from which the glass tube had been removed. The beam of ions passed through a set of narrow slits and struck the first dynode of the electron multiplier. The amplified current was taken from the last dynode. A Kiethley Model 610 electrometer was used to read the current.

III. METHOD

With the stopping cell and differential pumping gap evacuated, the analyzing magnet was adjusted so that the beam of helium ions from the accelerator entered the stopping cell. After passing through the stopping cell the beam entered the magnetic spectrometer which was adjusted to deflect the beam into the 90° detector. Gas was then admitted to the stopping cell and the spectrometer magnet readjusted to deflect the singly charged helium-ion component of the beam leaving the stopping cell into the 90° detector.

Two methods were used to locate the maximum in the energy distribution of the beam (see Fig. 2). The first method involved adjusting the spectrometer magnet as carefully as possible to obtain the maximum beam current. The second method was a curve fitting procedure. Five readings of the beam current as a function of proton resonance frequency were taken. The

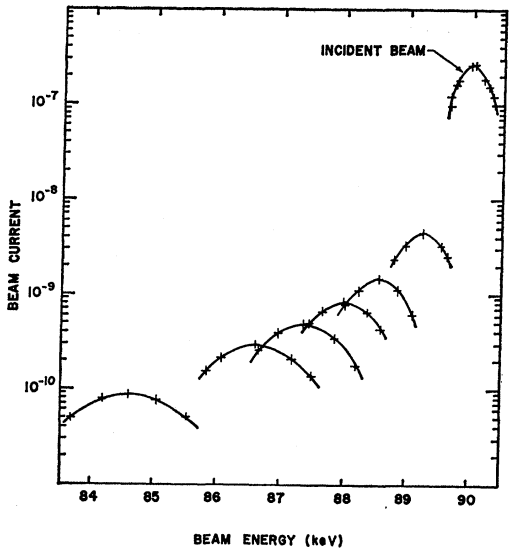

FIG. 2. Energy distributions of exit helium ion beams as a function of energy loss for an incident 90-keV helium ion beam.

points obtained were fitted by a computer to an adjusted Gaussian distribution giving an accurate indication of its center. This method permitted the use of the sloping sides of the energy distribution, where the change of beam current as a function of energy is large, to determine the location of the maximum beam current. The two methods gave excellent agreement.

The pressures of the gas in the stopping cell were generally in the pressure range 0.03 to 0.4 mm Hg. The gas pressure used was a function of the gas and the energy of the incident beam. Gas pressures which were too high resulted in excessive attenuation of the beam and an energy distribution of the exit beam too broad to permit accurate location of the maximum. At the extremely low pressures the energy losses were too small in relation to the energy distribution of the incident beam and resulted in large errors in the energy-loss measurements. Also, a variation in the measured stopping cross section at very low pressures was noted. The variation was indicated by cross-section measurements which increased with energy loss for very small energy losses but came to a constant value for larger energy losses. This effect was caused by an asymmetry in the emergent beam distribution for very small energy losses. It has been shown from probability theory[10] that even if the energy loss is the result of a very large number of individual contributions, a normal curve can be expected only if all of the energy-loss events are small compared to the half-width of the curve. If any of the energy-loss events are large compared to the half-width of the curve the most probable value of energy loss, which is the value measured in this experiment will not coincide with the mean value of the energy loss. This effect on the measured energy-loss

[6] J. T. Park, Rev. Sci. Instr. 35, 243 (1964).
[7] C. S. Cook, E. Jones, and T. Jorgensen, Phys. Rev. 91, 1417 (1953).
[8] Model GM100A, Consolidated Vacuum Company, Rochester, New York.
[9] H. Knoeble and E. Hahn, Rev. Sci. Instr. 22, 904 (1951).
[10] N. Bohr, Kgl. Danske Videnskab. Selskab, Mat.-Fys. Medd. 18, (1948).

cross sections was usually less than 10% and occurred in the measurements at low pressures with very low-energy losses where the uncertainty in a single measurement was already large.

The data obtained in the experiment were reduced by a computer which calculated both the stopping cross sections and the uncertainty in them for each experimental measurement.

IV. ERROR ANALYSIS

The energy of the proton beam from the accelerator has been found to be directly proportional to the reading on the calibrating potentiometer.[4] The maximum error in the stopping cross section due to the uncertainty in energy is 0.7%.[11]

The width of the incident beam at half-maximum depended slightly upon the focusing of the accelerator but was approximately $\frac{1}{2}$% of the beam energy. The peak of the incident beam could be located to better than 0.1% of the incident-beam energy by the method of simply adjusting the spectrometer magnet for maximum beam current. The curve-fitting method permitted more accurate location of the peak in the incident beam energy. The uncertainty of the location of the maximum using this method was computed for each case. The energy spread of the beam depended on the gas in the stopping cell, the energy loss, and the energy of the incident beam. The uncertainty in the energy of the exit beam was about 10% of the energy spread at half-maximum using the first method of locating the peak. If the curve fitting method was applied this uncertainty was considerably reduced and it was often only one-fourth of the uncertainty obtained by the first method. The energy loss sustained by the beam varied between $\frac{1}{2}$ and 5 keV and the uncertainty resulting from the energy loss varied from 1.5 to 15% and was necessarily calculated separately for each experimental measurement.

The purity of each gas and the uncertainty resulting from the impurities is given in Table I. All the gases, except air, are assumed to contain only the impurities specified by the manufacturer. Considerable effort was applied to prevent any contamination of the test gases.

TABLE I. Uncertainty in the stopping cross sections due to impurities.

Gas	Minimum purity	Uncertainty due to impurities (%)
Acetylene	99.5[a]	0.2
Cyclopropane	99.5[a]	0.2
Ethylene	99.5[a]	0.2
Methane	99.0[a]	0.5
Propane	99.5[a]	0.3
Propylene	99.0[a]	0.3

[a] Matheson Company, Incorporated.

[11] J. T. Park and E. J. Zimmerman, Phys. Rev. 131, 1611 (1963).

The connecting tubing, regulators, and valves were always leak tested and evacuated prior to the opening of the high pressure valve on the tank. The system was flushed with the test gas at least four times prior to the beginning of the stopping-cross-section measurements.

In the case of ethylene, most of the impurities have larger stopping cross sections than ethylene; hence, the result is to make the experimental cross section of ethylene about 0.2% too high. For this reason the reported values of the stopping cross section in Table III have been reduced by 0.2% from the measured values.

The uncertainty in the pressure as determined by the Decker meter was obtained from the average deviations of the readings when compared to the McLeod gauge readings in air, the uncertainty in corrections to the Decker meter calibration, and the uncertainty in the McLeod gauge readings. This uncertainty varies from 1.6 to 2.2%.

The uncertainty in the pressure of the gases for which the McLeod gauges were used directly is taken as 0.8%. In addition, the uncertainty in the measurement of the height of the mercury column of the McLeod gauges was taken to be 0.005 cm. The effects of mercury diffusion in the McLeod gauge[12] were taken into account.

The uncertainty due to the measurement of the length of the stopping cell and the first differential pumping gap is less than 0.01%. The lengths were corrected for thermal expansion. The temperature was readily determined to within 1°C by the mercury thermometer.

Table II gives a typical example of errors in a single measurement for 90-keV helium ions incident on cyclopropane with an energy loss of 3 keV. These uncertainties have been combined by standard root-mean-square error propagation methods. The statistical spread of the measurements was consistent with the calculated uncertainties. The uncertainties in a single measurement varied from 2 to 17%. Several such measurements were weighted and averaged to produce a single point.

TABLE II. Typical errors in single measurements of the stopping cross sections of cyclopropane for 90-keV helium ions.

Random errors	(%)
Uncertainty in energy loss	3.5
Uncertainty in pressure	2.0
Uncertainty in temperature	0.3
Uncertainty in relative frequency change	1.1
Root-mean-square value	4.3
Systematic errors	**(%)**
Uncertainty in energy	0.7
Uncertainty in McLeod gauge calibration	0.8
Uncertainty in length	0.01
Uncertainty due to impurities	0.2
Root-mean-square value	1.2

[12] H. Ishii and K. Nakayama, Vacuum Symposium Trans. 8, 519 (1961).

TABLE III. Helium-ion stopping cross sections (10^{-15} eV-cm^2/molecule).

Energy (keV)	Air/2 (%)	Acetylene (%)	Cyclopropane (%)	Ethylene (%)	Methane (%)	Propane (%)	Propylene (%)	ϵ (C)	ϵ (H)
40	16.2±5.0	35.7±5.0	59.8±5.0	42.2±4.0	31.1±5.0	69.5±5.0	62.7±5.0	12.9	4.1
60	19.7±4.0	48.7±4.0	80.0±3.0	57.0±3.0	40.3±3.5	97.0±4.0	82.8±3.0	18.0	5.2
90	23.2±3.0	60.2±3.5	103.9±2.5	70.0±2.0	47.3±3.0	115.5±3.0	107.2±2.5	24.2	5.5
140	29.1±2.5	75.9±3.0	131.2±2.5	87.5±2.0	57.8±2.5	142.5±2.0	133.6±2.5	31.3	6.4
200	36.0±2.5	89.5±2.5	153.0±2.5	102.8±1.7	70.2±2.5	168.9±2.0	153.7±2.5	35.7	8.0
α[a] (expt)	0.50	0.51	0.52	0.49	0.50	0.48	0.50		
α[a] (theo)[b]	0.50	0.50	0.50	0.50	0.50	0.50	0.50		
β[a] (expt)	2.51	1.52	1.13	1.29	1.00	1.22	1.22		
β[a] (theo)[b]	2.52	1.77	1.48	1.48	1.30	1.40	1.48		

[a] $\epsilon=\beta E^{\alpha}\times10^{-15}$ eV-cm^2/atom.
[b] Theoretical values of Lindhard—see text.

V. RESULTS AND CONCLUSIONS

The stopping cross sections and their uncertainties are given in Table III. The values in the tables are taken from smooth curves and the uncertainties shown are the standard deviations of the curves at that point.

Very few stopping cross section data are available for helium ions. Weyl[1] measured the energy loss in air in the energy range 150 to 400 keV so his data overlaps the present experiment from 150 to 200 keV. In Fig. 3, the reported values taken from Weyl's curve are denoted by circles and Weyl's measurements are shown as crosses. He obtains a curve roughly proportional to $E^{1/3}$ over the entire range and his reported values are taken from that curve. The present experiment is in very good agreement with the reported values but differs in the observed curvature.

Energy-loss cross sections may be obtained from the range measurements of Cook, Jones, and Jorgensen[7] on air. Differentiating the range measurements of Cook, Jones, and Jorgensen gave values which averaged 8% higher than the present experimental results. The value obtained for the energy-loss cross section by differentiating range measurements must necessarily be larger than that obtained in a differential experiment because the extremely narrow angle of acceptance in the differential energy-loss experiment largely eliminates the fraction of the beam undergoing energy loss due to nuclear scattering.

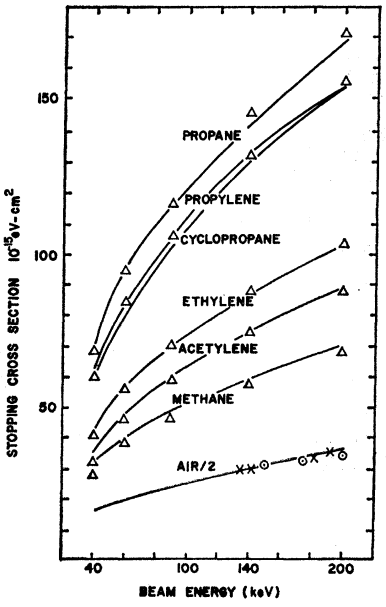

FIG. 3. Helium-ion stopping cross section for hydrocarbon gases. The solid curve is the best straight line through present experimental data. × Weyl (Ref. 1) data; ○ Weyl (Ref. 1) from smooth curves; △ Bragg-rule values.

The present experiment can also be compared to the previous measurements taken at this laboratory.[11] The measurements on ethylene and propane, where they overlap with the prior experiment, are essentially identical. This confirms the fact that the modifications in the apparatus have had no effect on the measured cross sections and that the comparisons of equipment and methods with other researchers using protons are valid.

Lindhard and Scharff[13] have calculated the loss of energy to electrons for ion velocities small compared to $v_0 z_1^{2/3}$, where z_1 is the nuclear charge of the incident ion and v_0 is the electron velocity in the first Bohr orbit. Their relationship is valid for helium ions with energy less than 250 keV. Rewriting their equation in terms of E, the energy of the incident ion, we obtain

$$\epsilon=\epsilon_0(8\pi e^2 a_0)[z_1 z_2/(z_1^{2/3}+z_2^{2/3})^{3/2}](E/E_0)^{1/2},$$

where z_2 is the nuclear charge of the stopping atom, a_0 is the Bohr radius, and ϵ_0 is of the order of 1–2 and E_0 is the kinetic energy of the incident ion when its velocity equals $v_0 z_1^{3/2}$.

In the helium-ion energy range 40–200 keV the energy loss of helium ions is predominately due to the electrons, as can be seen by examining Bohr's[10] expression for the nuclear contributions; hence, the above relation can be compared with the experiment in respect to the energy dependence. The experimental results showed some deviation from a direct power dependence on the energy at the lowest energies, and it must be noted that the power dependence on energy can be varied quite strongly without greatly increasing the uncertainty in the fitting of the data. The energy dependence of the helium-ion stopping cross sections is shown in Fig. 4. The theoretical[14] and experimental values obtained for the constants and exponents are given in Table III. The agreement with the calculation of Lindhard and Scharff is very good considering the complexity of the problem of calculating energy-loss cross sections in this

[13] J. Lindhard and M. Scharff, Phys. Rev. 124, 128 (1961).
[14] J. Lindhard (private communication).

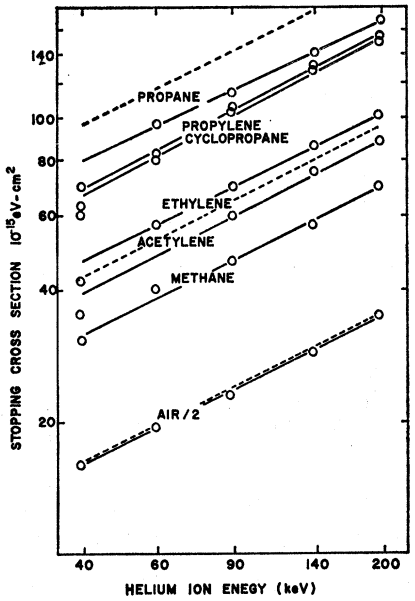

FIG. 4. Theoretical and experimental helium-ion stopping cross sections for hydrocarbon gases. o Experimental data. The solid curve is the best straight line through data. The dashed curve is Lindhard's calculation (Ref. 13, 14) for air/2, acetylene, and propane.

energy range for polyatomic molecules. The values of the exponents must be considered to have large uncertainties, but the experimental value of the exponents are all within 5% of the theoretical value of 0.5. The theoretical values for the constant average 18% higher than the experimental values.

Comparison of the data in this experiment with prior experiments using protons shows that in the energy range 40 to 200 keV the commonly used assumption that the stopping number is independent of the mass and charge of the incident ion is not valid. The ratio of the energy-loss cross sections of air for incident protons and incident helium ions at the same velocity is approximately 1.9. If the stopping number were independent of incident ion the ratio of the stopping cross sections should be as the ratios of the squares of the charges. $\epsilon_\alpha/\epsilon_p=(z_\alpha)^2/(z_p)^2$. The ratio is certainly not correct using the nuclear charges. Data for equilibrium fractions are available for 200-keV helium ions and 50-keV protons, incident on oxygen and nitrogen.[15] If the measured charge fractions are used to calculate the ratio of the stopping cross sections of helium ions and protons in air, a ratio of 1.15 is obtained. Thus, even where the equilibrium charge fractions are available, the use of a measured cross section for incident protons on a material to obtain the cross section for helium ions on the assumption that the stopping number was only a function of the velocity and the stopping material could produce an error of over 50% in this energy range.

The Bragg rule of additivity for a hydrocarbon molecule can be stated in the form $\epsilon(C_mH_n)=m\epsilon(C)+n\epsilon(H)$, where $\epsilon(C_mH_n)$ is the stopping cross section of the hydrocarbon molecule, and $\epsilon(C)$ and $\epsilon(H)$ are the stopping cross sections of atomic carbon and hydrogen, respectively. The failure of the Bragg rule for proton stopping cross sections has been illustrated.[2,11,16] If the present data are used to obtain the best values of $\epsilon(C)$ and $\epsilon(H)$ and the resulting values used to calculate the stopping cross sections of the hydrocarbon gases used, the agreement between the experimental curves and the calculated ones is quite good. Table III includes the calculated values of $\epsilon(C)$ and $\epsilon(H)$. The agreement between the experimental curves and the calculated stopping cross sections is shown in Fig. 3. The triangles represent the calculated values of the cross sections using the Bragg rule values of $\epsilon(C)$ and $\epsilon(H)$.

In addition to the large uncertainty in the calculated values of $\epsilon(C)$ and $\epsilon(H)$, the fact that the stopping cross section of propylene is slightly larger than that of cyclopropane is an indication of the effects of molecular structure on stopping cross sections. Cyclopropane, $(CH_2)_3$, and propylene, C_2H_6, have exactly the same number of carbon and hydrogen atoms but different molecular structure. The difference between the cross sections of the two gases is within the experimental error; however, in no case were the experimental measurements on cyclopropane as high as those on propylene at the same energy. Aniansson[17] measured the integral stopping power of hydrocarbons for 5.3-MeV α particles and noted that his value for stopping power of benzene was 2.2% larger than the value predicted from paraffin values on the assumption of additivity. This result combined with stopping power measurements made on hydrocarbons with protons[11] would indicate that the deviations from the Bragg rule due to molecular-bonding effects might occur at the maximum on the stopping cross section, i.e., at about 300 keV for He^+ and 70 keV for protons.

The conclusion of this analysis is that while there is evidence of an effect on the stopping cross sections of the hydrocarbon gases due to the molecular-binding configuration, it is a relatively smaller effect for incident helium ions than for protons in the energy range of 40 to 200 keV, and the Bragg rule values of $\epsilon(C)$ and $\epsilon(H)$ can be applied to obtain very useful information on the stopping cross section of a hydrocarbon gas.

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[15] S. K. Allison and M. Garcia-Munoz, in *Atomic and Molecular Processes*, edited by D. R. Bates (Academic Press Inc., New York, 1962).

[16] R. L. Platzman, in *Symposium on Radiobiology*, edited by J. J. Nickson (John Wiley & Sons, Inc., New York, 1952).

[17] G. Aniansson, Trans. Roy. Inst. Technol. Stockholm 174, (1961).