

Long-Lived Radioactive Carbon: C¹⁴

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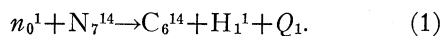
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A long-lived activity has been obtained by deuteron bombardment of graphite. This activity has been shown by chemical methods to be isotopic with carbon. The same activity has been obtained by slow neutron irradiation of nitrogenous compounds (NH₄NO₃, NH₄NO₂, etc.). The radiations consist of low energy negative electrons with a range of 19±2 mg/cm² aluminum (145±15 kev upper energy limit). No soft or hard gamma-rays could be detected. The nuclear reactions must be C¹³ (*d, p*) C¹⁴ and N¹⁴ (*n, p*) C¹⁴. A sample has been followed in a screen wall Geiger counter for 9 months and has shown no decay. From cross section and yield estimates the half-life of C¹⁴ is probably 10³-10⁵ years. The production, isolation, and detection of C¹⁴ for tracer work is described.

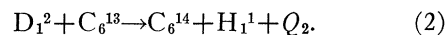
THERE are 5 known isotopes of carbon, stable C¹² (~99 percent abundance), stable C¹³ (~1 percent abundance), C¹⁰ (8.8-second half-life),¹ C¹¹ (21-minute half-life),² and C¹⁴ (half-life 10³-10⁵ years).³ The production and properties of C¹⁴ will be described in this paper. In view of the possible application of C¹⁴ as a tracer,⁴ a discussion of relative yields and specific activities obtainable is included together with a brief description of detection methods.

It is of interest to review briefly the history of C¹⁴. In 1934 Kurie, using a Wilson cloud chamber, studied the fast neutron disintegration of nitrogen and observed in addition to the known⁵ alpha-particle emission, another type of transmutation,⁶ less frequent, in which singly charged heavy particles were emitted. Kurie suggested these were protons due to



Later Bonner and Brubaker,⁷ irradiating nitrogen with slow neutrons, observed a few heavy tracks (~1.06-cm range) which they attributed

to alpha-particles. Burcham and Goldhaber⁸ demonstrated these tracks were protons and suggested they originated from reaction (1). Bonner and Brubaker⁹ agreed with Burcham and Goldhaber and moreover estimated $Q_1 = 0.58 \pm 0.03$ Mev. From the known mass difference between neutron and proton, C¹⁴ was estimated to be unstable with respect to N¹⁴ by ~0.17 Mev so that it could be expected that C¹⁴ would be a beta-emitter of relatively long half-life. McMillan¹⁰ found a long-lived soft radiation from metal scraped from inside the cyclotron vacuum chamber and suggested it might be due to C¹⁴ formed by the reaction



Unfortunately, the sample was accidentally lost before any chemistry was performed. However, the upper energy limit of the beta-radiation was measured by Libby and Lee¹¹ using a screen wall Geiger counter and found to be ~10 kev. (It seems quite likely this activity was due to H³ which was discovered by Alvarez and Cornog.¹²) Shortly after, the writers had occasion to prepare N¹³ (by deuteron bombardment of graphite) for use as a tracer in nitrogen fixation experiments. The same graphite target was used in all bombardments and after ~1000 μ amp. hr. of 8-Mev

¹ Barkas, Creutz, Delsasso, Fox and White, Phys. Rev. **57**, 562 (1940).

² Cockcroft, Gilbert and Walton, Proc. Roy. Soc. (London) **A148**, 225 (1935).

³ S. Ruben and M. D. Kamen, Phys. Rev. **57**, 549 (1940); **58**, 194 (1940).

⁴ C¹¹, despite its short half-life has already been used rather extensively as a tracer, cf. G. T. Seaborg, Chem. Rev. **27**, 199 (1940).

⁵ N. Feather, Proc. Roy. Soc. (London) **A136**, 709 (1932).

⁶ F. N. D. Kurie, Phys. Rev. **45**, 904 (1934); **46**, 330 (1934).

⁷ T. W. Bonner and W. M. Brubaker, Phys. Rev. **48**, 469 (1935).

⁸ W. E. Burcham and M. Goldhaber, Proc. Camb. Phil. Soc. **32**, 632 (1936).

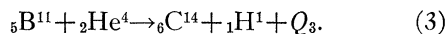
⁹ T. W. Bonner and W. M. Brubaker, Phys. Rev. **49**, 223 (1936); **49**, 778 (1936).

¹⁰ E. M. McMillan, Phys. Rev. **49**, 875A (1936).

¹¹ W. F. Libby and D. D. Lee, Phys. Rev. **55**, 245 (1939).

¹² L. W. Alvarez and R. Cornog, Phys. Rev. **56**, 613 (1939).

deuterons it was burned to CO_2 which was precipitated as CaCO_3 . A fraction of this material, when mounted on the inner surface of a cylinder and counted in a screen wall counter of the type developed by Libby,¹³ gave an effect of 16 ± 2 counts/min. It was possible to show this effect was not due to natural contamination of the chemical reagents used. It seemed desirable, however, to obtain stronger samples before drawing any positive conclusions. It was apparent that a prolonged bombardment was necessary and, due to the heavy schedule under which the cyclotron was operating, this was not possible until recently. In the meantime B_2O_3 was bombarded in a gas-tight target chamber with 32-Mev alpha-particles accelerated in the 60" cyclotron in a search for C^{14} produced by



It had been observed that when B_2O_3 is bombarded with deuterons the major fraction of the newly formed C^{11} is expelled from the target as CO .^{14,15} After $\sim 10 \mu$ amp. hr. of alpha-particle bombardment, the gas from the target chamber was introduced into an ionization chamber connected to an FP-54 electrometer tube and amplifier circuit.¹⁶ No evidence for any long-lived activity was found.

At this time Pollard¹⁷ and Bower and Burcham¹⁸ presented additional evidence for the production and instability of C^{14} . Pollard investigated the protons from reactions (2) and (3). Using 3-Mev deuterons, the longest proton range was 81.1 ± 2 cm yielding $Q_2 = 8.21$ Mev. Bombardment of boron with ThC alphas produced protons with a 97.6-cm range; Po alpha-particles gave a proton group of 42.1-cm range. These experiments yielded an average $Q_3 = 0.66$ Mev and using these values he proposed the mass of C^{14} to be 14.00780 ± 0.00020 and thus unstable

¹³ W. F. Libby, *Phys. Rev.* **46**, 196 (1934).

¹⁴ Yost, Shinohara and Ridenour, *J. Chem. Phys.* **3**, 133 (1935).

¹⁵ Ruben, Kamen and Hassid, *J. Am. Chem. Soc.* **61**, 661 (1939); **62**, 3443 (1940).

¹⁶ We are indebted to Dr. E. Segrè for the loan of the electrometer and for aid in many of the measurements made with it.

¹⁷ E. Pollard, *Phys. Rev.* **56**, 1168 (1939).

¹⁸ J. C. Bower and W. E. Burcham, *Proc. Roy. Soc. (London)* **A173**, 391 (1939).

with respect to N^{14} by ~ 300 kev. His attempts to detect an activity ascribable to C^{14} after $\sim 6 \mu$ amp. hr. bombardment of graphite with 3-Mev deuterons were unsuccessful. Bower and Burcham, using 0.8-Mev deuterons, found a 48-cm group of protons which they attributed to reaction (2) with $Q_2 = 6.1$ Mev. Recently Bennett, Bonner, Hudspeth and Watt¹⁹ have confirmed this observation, using targets enriched in C^{13} .

ISOLATION OF C^{14} AFTER PROLONGED DEUTERON BOMBARDMENT

By resorting to the method of internal targets²⁰ it was finally found possible to produce appreciable amounts of an activity which was proven chemically to be isotopic with carbon. A probe target, covered with a small quantity of Aquadag, was inserted into the vacuum chamber and subjected to a $40\text{-}\mu$ amp. beam of 7–8 Mev deuterons for ~ 120 hr. The probe was frequently removed and examined and the Aquadag renewed whenever necessary. The target prepared in this way proved unsatisfactory for prolonged bombardment and was finally removed and examined for activity. For a second bombardment, the graphite was electroplated with copper and hard soldered to the probe surface. The graphite was of a type prepared for use as an anode in a high power rectifier tube. This target proved more satisfactory and was able to withstand 500–600 μ amperes of ion current without appreciable deterioration. This probe was placed far into the vacuum chamber to intercept high currents ($\sim 500 \mu$ amperes) of 3–4-Mev deuterons until 13,000 μ amp. hr. had been accumulated.

Since high specific activities are necessary for many tracer experiments it is apparent that the bombardment of carbon enriched in C^{13} is desirable. Professor H. C. Urey and Dr. D. W. Stewart very kindly sent us 0.63 gram of ~ 23 percent C^{13} as amorphous carbon prepared by the reduction of enriched NaCN with magnesium at $\sim 400^\circ\text{C}$. A portion of this carbon was placed

¹⁹ Bennett, Bonner, Hudspeth and Watt, *Phys. Rev.* **58**, 478 (1940). Holloway and Moore (*Phys. Rev.* **57**, 1086A (1940)) were unable to detect the protons from $\text{C}^{13}(d, p)\text{C}^{14}$. They placed a lower limit of 1.2×10^{-28} cm² on the value for the cross section of this reaction at 1 Mev.

²⁰ R. R. Wilson and M. D. Kamen, *Phys. Rev.* **54**, 1031 (1938).

in a steel mold and subjected to high pressure²¹ (40 tons/in.²). The material failed to cohere and examination under a microscope revealed a granular structure quite different from graphite found to press well. Since no equipment for very high temperature work was available, the carbon was sent to the Edgewater Works of the National Carbon and Carbide Corporation, at the suggestion of Dr. H. C. McPherson. In these laboratories, after considerable effort, Dr. McPherson succeeded in preparing several satisfactory targets. The procedure employed by Dr. McPherson involved dilution of the amorphous carbon with a suitable amount of pitch binder. The mixture was pressed into a mold to make a slab 2 mm thick and 1 cm square. The slab was baked at 1200°C and then "graphitized" (heated in vacuum) at 2800°C to drive off all volatile matter. The resultant material contained 9.3 percent C¹³, a reduction by a factor of more than two from the starting material. (Such loss is unavoidable with small quantities of carbon). The slab was sprayed with molten copper (Schoop process) to provide a tenacious metallic backing for soldering the material to the water-cooled probe surface. It was necessary in some instances to repeat the spraying since the slabs showed a tendency to embrittle and disintegrate when reheated. The slabs which survived this treatment possessed about $\frac{1}{5}$ the heat conductivity of the best graphite obtainable and proved to be adequate as target material. One slab mounted with phos-copper solder on a probe withstood 10 hours' bombardment with 300 μ amperes of 3-4-Mev deuterons and showed no visible deterioration.

CHEMICAL IDENTIFICATION

The graphite was burned in a stream of oxygen and the resultant gases passed over hot cupric oxide and finally absorbed in Ca(OH)₂. The precipitated CaCO₃ was filtered and carefully washed. The filter paper containing the precipitate, when placed over a thin-walled aluminum Geiger counter (0.1 mm wall thickness), failed to show any activity. A portion ($\frac{1}{3}$) of this sample, spread out in a thin film inside a cylinder which was introduced into a screen wall counter, was

²¹ We are indebted to the Department of Civil Engineering for cooperation in these experiments.

active (~400 counts/min.). In the counting arrangement used, the cylinder containing the sample could be slid to and fro over the counting region, thus enabling alternate readings of sample activity and background to be taken. The procedure was repeated using ordinary CaCO₃ prepared in the same way to test for activity in the reagents. The CaCO₃ precipitate obtained in this way showed no activity.

The active CaCO₃ was treated with acid and the CO₂ collected. After drying, this was introduced into a Geiger counter and gave an effect of ~2000 counts/min. The 5-fold increase in activity was due to increased solid angle and elimination of self-absorption. The active gas was absorbed in Ca(OH)₂ and reconverted into CO₂ several times with no loss in specific activity. However a preliminary measurement indicated the range of the radiations in aluminum to be approximately 12 mg/cm²; which is close to the known value for S³⁵. The procedure used to this point did not exclude the possibility that part or all of the activity was due to S³⁵O₂. Carrier (inactive) SO₂ was mixed with the active gas and the mixture treated with KMnO₄ in sulfuric acid solution to oxidize the SO₂ to SO₄²⁻. There was no loss in gaseous activity. In another experiment the mixture of the gaseous activity and inactive SO₂ was absorbed in alkali and the SO₃²⁻ oxidized to SO₄²⁻ with I₃⁻. The CO₂ was liberated by acidifying the solution and absorbed in Ca(OH)₂. The CaCO₃ was active, there being no loss in specific activity.

PRODUCTION OF C¹⁴ BY NEUTRON TRANSMUTATION OF NITROGEN

It was realized from the very beginning that deuteron bombardment of carbon was not the best method for preparing C¹⁴ in high specific activity suitable for tracer work. The transmutation of N¹⁴ by slow neutrons (reaction 1) offered many attractive features: (1) Possibility of obtaining very high specific activity since the target material is not carbon but nitrogen. (2) Production of C¹⁴ is achieved as a by-product of cyclotron operation and thus does not interfere with other research. (3) Any strong neutron source will suffice. It is not necessary to go to high bombarding voltages (3-4 Mev as is the case for

deuteron bombardment). (4) The nitrogenous material, also containing some hydrogen, can profitably replace water or paraffin as a shielding material.²²

While the slow neutron experiments are by no means complete, the results obtained thus far conclusively indicate this to be the most practical method for C^{14} production.

Two five-gallon carboys filled with saturated solutions of ammonium nitrate were placed in the region opposite the deflector of the Berkeley medical cyclotron. The solutions were irradiated with neutrons during a period of ~ 6 months. Approximately $40,000 \mu$ amp. hr. of deuteron bombardment at 16 Mev of various targets (principally beryllium and phosphorus) took place in this time. A chemical analysis was performed in which carbon monoxide, carbon dioxide, methane, carbon, methanol, cyanide and formaldehyde were added as carriers and then separated and analyzed for radioactivity. After vigorous and prolonged shaking the gaseous compounds were pumped off. The gases were oxidized to carbon dioxide and absorbed in $Ca(OH)_2$. The $CaCO_3$ was very active, containing 10^5 counts/min. By the chemical methods described above the activity was shown to be isotopic with carbon.

Charcoal and other sediment was separated from the NH_4NO_3 solution by filtration. This proved to be completely inactive. Cyanide was precipitated as $AgCN$ which also was inactive. Methanol and formaldehyde were distilled from an aliquot (~ 10 percent) of the NH_4NO_3 and the distillate divided into 2 portions. From one portion the insoluble hydrazone of formaldehyde prepared by the addition of excess 2,4 dinitrophenyl hydrazine was found to be inactive. The methanol from the other portion was volatilized and passed over hot cupric oxide. The resulting CO_2 was not active. For future experiments a more complete and elaborate chemical analysis has been devised.

In addition to NH_4NO_3 solutions, several samples of solid NH_4NO_3 (~ 1 lb.) as well as concentrated NH_4NO_2 solutions have been irradiated with neutrons. C^{14} was found only in the

²² C. Lapointe and F. Rasetti, Phys. Rev. **58**, 554 (1940), have found the total capture cross section of nitrogen for thermal neutrons to be 1.2×10^{-24} cm². Hydrogen has a cross section of 0.2×10^{-24} cm².

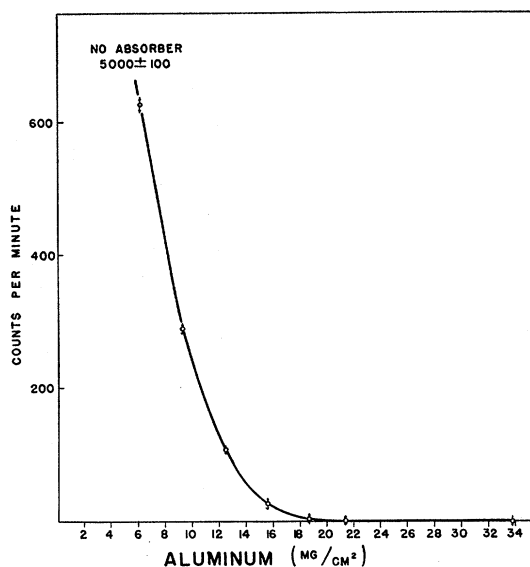


FIG. 1. Absorption of radiation from C^{14} in aluminum.

gaseous fraction. The solid NH_4NO_3 was irradiated in sealed soft glass containers. No volatile activity could be pumped off the solid even after addition of carrier gases. However, when the solid was dissolved in water the gaseous fraction contained 1000 counts/min. Evidently the gases are firmly trapped and are released only when the crystals are completely dissolved.

A large number of other nitrogen compounds are being irradiated not only to determine the best conditions for large scale²³ C^{14} production but also to investigate the chemistry of the recoiling carbon as a function of the chemical environment. Experiments are now in progress with urea, melamine, dicyandiamide, etc.²⁴

PROPERTIES OF THE LONG-LIVED RADIATION

The absorption of the radiation in Al was measured by wrapping the screen wall of the Geiger counter with thin Al foils. With the first CaC^*O_3 sample (prepared by deuterons on carbon) which had an activity of only 400 counts/min. (background was 120/min.), the activity coming through 12 mg/cm² Al was 0 ± 6

²³ It is a matter of common knowledge that NH_4NO_3 is thermodynamically unstable. However, it seems certain that its solution at room temperature offers no explosion hazard.

²⁴ We are indebted to the American Cyanamid Corporation for the gift of large quantities of these materials.

counts/min. However with a very thin layer (~ 0.1 mg/cm²) of CaC*O₃ (produced from NH₄NO₃) which had an activity of 5000 counts/min., 0.5 percent \pm 0.1 percent of this effect was still observable at 13 mg/cm². The first estimate of the range was too low and it seems a more accurate value is 19 ± 2 mg/cm². The absorption curve is shown in Fig. 1. Since the activity coming through 19 mg/cm² is < 0.1 percent of the initial effect it is safe to conclude that γ -rays of ~ 100 kev or more are absent. If the radiation consisted wholly of positrons the annihilation γ -rays should have given an effect of 100 counts/min. coming through the Al.²⁵ A comparison of the absorption in Cellophane and Al indicates the absence of soft γ -rays. 3.26 mg/cm² of Cellophane absorbed 69 ± 4 percent while the same amount of Al absorbed 71 ± 4 percent of the radiation.²⁶ Therefore it seems safe to conclude negative electrons are being emitted and the unstable isotope is C¹⁴. If the upper energy limit of the beta-ray spectrum is taken to be $145,000 \pm 15,000$ ev and the mass of N¹⁴ to be 14.007526,²⁷ the mass of C¹⁴ is calculated to be 14.00768. It is also possible, with the neutron-proton mass difference,²⁷ to determine $Q_1 = 0.64$ Mev. The estimate of Bonner and Brubaker⁹ of 0.58 ± 0.03 Mev is in agreement with this value.

The activity of a sample has been followed for 9 months and no decay observed. From this fact alone the half-life must be longer than 30 years. It is possible to make rough estimates of the half-life from the yields and cross sections. The C¹³(d, p)C¹⁴ experiments indicate 10^3 – 10^4 years while the N¹⁴(n, p)C¹⁴ data suggest 10^4 – 10^5 years. A lower limit of 1000 years is not unreasonable. Using the same technique employed in the H³ work,²⁸ ($n_0^1 + {}_3\text{Li}^6 \rightarrow {}_2\text{He}^4 + {}_1\text{H}^3$), Goldhaber concludes²⁹ the half-life to be longer than 300 years.

The upper energy limit of ~ 150 kev coupled with the long half-life ($\sim 10^3$ years) is quite unexpected. It would have been supposed that

the decay of C¹⁴ to N¹⁴ would be an allowed transition analogous to the He⁶–Li⁶ case. Dr. P. Morrison has calculated that on this basis, a maximum half-life value of the order of a day is to be expected. Since the actual half-life is quite certainly a factor of 10^5 – 10^6 greater it follows that C¹⁴–N¹⁴ decay involves a forbidden transition. In consequence,³⁰ it must be supposed that configurations with high angular momentum make appreciable contributions to the ground state of C¹⁴. A similar³¹ situation has been found in the case of Be¹⁰, a nucleus which might be expected to behave like He⁶, i.e., decay with a short half-life. Actually, despite the high upper energy limit of the emitted electrons (~ 580 kev), Be¹⁰ seems to have a very long half-life (about 10^3 – 10^5 years).

It is of interest to note that because of their long half-lives the production of macroscopic amounts of C¹⁴ and Be¹⁰ is at hand and in the near future it may be possible to determine their nuclear spins, etc. If the half-life of C¹⁴ is assumed to be 10^4 years, then 0.1 microgram is present in the sample prepared from the transmutation of nitrogen.

In conclusion, a few remarks may be made regarding the measurement of C¹⁴. For small samples (up to ~ 50 mg), counting as gaseous CO₂ seems to be the simplest and most sensitive method. Carbon dioxide, with a trace of foreign gas (H₂O, C₂H₅OH, O₂, etc.) has been found³² in this laboratory to be a quite satisfactory counter gas. However, since CO₂ requires relatively high counting voltages, the source of very steady high potential may be a limiting factor. For the measurement of large samples of low specific activity the use of thin carbonate (Li₂CO₃, etc.) layers inside a screen wall counter is the most efficient technique.

In many respects it is unfortunate that the half-life of C¹⁴ is so long, since, for the production of strong samples, long time exposures are required. On the other hand, it is possible to concentrate C¹⁴ by various isotope separation methods (chemical exchange, thermal diffusion, ultracentrifugation, etc.) and thus material of low

²⁵ This estimate is based on measurements with a positron emitter (Na²²) under the same conditions.

²⁶ A more detailed investigation of the spectrum by Dr. J. Halpern and Mr. R. C. Raymond, using a Wilson cloud chamber, is now in progress.

²⁷ Hahn, Flügge and Mattauch, Ber. **73**, 1 (1940).

²⁸ R. D. O'Neal and M. Goldhaber, Phys. Rev. **57**, 1086 (1940).

²⁹ Conference on Applied Nuclear Physics, Massachusetts Institute of Technology, 1940.

³⁰ We are indebted to Professor J. R. Oppenheimer and Dr. L. I. Schiff for valuable discussions on this point.

³¹ E. M. McMillan and S. Ruben, unpublished.

³² T. H. Norris, private communication.

specific activity (including the end products of tracer experiments), can be reconcentrated. A given sample of C^{14} (and C^{13}) may, therefore, be used repeatedly. When the use of C^{14} and C^{13} becomes widespread enough, it should be feasible for a commercial company to perform this task at a nominal cost.³³

³³ The Eastman Corporation, at the suggestion, and with the cooperation, of Professor H. C. Urey, has begun the concentration of N^{15} on a small scale. It is reasonable to believe that the interest and demand for carbon tracers will far exceed the supply of the radioactive isotopes and, therefore, the large scale concentration of C^{13} is of considerable importance.

Note added in proof.—The estimate for the C^{14} half-life was made by three different methods (1) the yield of C^{14} coupled with an extrapolation of Amaldi's curve (cf. Bethe's review) for the $C^{12}(d, p)C^{13}$ reaction and Pollard's value for the yield ratio $C^{13}(d, p)C^{14}/C^{12}(d, p)C^{13}$, (2) the $C^{13}(d, p)C^{14}$ cross section was estimated by comparison with the $B^{10}(d, n)C^{11}$ yield at bombarding energies of

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7–8 Mev, (3) the C^{14} yield from the slow neutron irradiation of NH_4NO_3 . Method (1) gave $2 \cdot 10^4$ years while (2) yielded $2 \cdot 10^8$ years. Schultz and Watson, *Phys. Rev.* **58**, 1047 (1940), have found Pollard's $C^{13}(d, p)C^{14}/C^{12}(d, p)C^{13}$ yield ratio was high by a factor of 5–10. This seems to bring the first two C^{14} half-life estimates into agreement at 10^8 years. The $N^{14}(n, p)C^{14}$ experiments suggest a still longer half-life for C^{14} . Hence it seems reasonable to regard 10^8 years as a lower limit.

Ionization and Dissociation of Diatomic Molecules by Electron Impact

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An extension of the mass spectrometric method of study of ionization and dissociation processes occurring in diatomic molecules under electron impact and the results of further investigation of the molecules H_2 , CO , NO , N_2 , and O_2 are discussed. The shapes of the peaks which appear in the graph of resolved ion current as a function of ion accelerating potential are analyzed and accounted for in terms of the wave-mechanical theory of the diatomic molecule and the characteristics of the mass spectrometer. It is indicated how the study of the peak shape coupled with appearance potential measurements has made possible conclusions concerning the nature of the dissociation process and the structure of the molecule. Differences between previous mass spectrometric data and those from other sources have been clarified either by obtaining better data with the mass spectrometer or by formulation of a satisfactory interpretation of the difference based on conclusions of an analysis of the ion peak shape. Values of dissociation energies, ionization potentials and interpretations of the processes that occur are given.

INTRODUCTION

COLLISIONS of electrons of low velocity with diatomic molecules may produce charged fragments of the molecule. The data on the minimum electron energy necessary to produce these fragments, the identification of them and of the processes involved may permit one to determine the heat of dissociation of the molecule. The relative abundance of the ion products

and the probability of a process as a function of the electron energy are also of interest.

In the usual mass spectrometer the ions of a given m/e value are segregated and the dependence of the ion current on the energy of the bombarding electrons and on the gas pressure in the collision chamber is determined.^{1–4} In the

* Now at the Bell Telephone Laboratories, New York.

¹ For a review of all work previous to 1931 see H. D. Smyth, *Rev. Mod. Phys.* **3**, 347 (1931). Reviews have also been given by H. Kallmann and B. Rosen, *Physik. Zeits.* **32**, 521 (1931); W. de Groot and F. M. Penning, *Handbuch*