supplied these conventionally dated samples.
8. R. Middleton, *Nucl. Instrum. Methods* **122**, 35 (1974).
9. This standard deviation in the mean is calculated based on the use of the measured sample variance of the data and is commonly called the external error. This is a more meaningful error than the internal error when one is comparing the results from two independent measurements. See, for instance, D. W. D. Rogers, *Nucl. Instrum. Methods* **127**, 253 (1975).
10. E. J. Stephenson, paper presented at the fall meeting of the Division of Nuclear Physics, Rochester, N.Y., 27–29 October 1977.
11. We thank K. W. Allen, E. T. Hall, H. R. McK. Hyder, R. Middleton, and E. J. Stephenson for their advice and for valuable discussions; we thank T. S. Lund and the Tandem Operations Group at the University of Rochester Nuclear Structure Research Laboratory for their assistance in carrying out these experiments. We thank M. Rubin for supplying us with the charcoal samples. The Nuclear Structure Research Laboratory is supported by a grant from the National Science Foundation. The participants from the University of Toronto are supported in part under a grant from the National Research Council of Canada.

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Radioisotope Dating with an Accelerator: A Blind Measurement

Abstract. *The age of a sample of carbon dioxide has been determined by accelerating the carbon with a cyclotron and detecting the carbon-14 ions in the beam. Nitrogen-14 was eliminated as a background through the use of a range-separation technique. To avoid all possibility of experimenter bias, the measurement was conducted in a blind fashion.*

Muller recently proposed that the use of high-energy particle accelerators for the direct detection of radioactive atoms could greatly extend the age of samples that could be dated while simultaneously reducing the sample size required (*1*). He also reported in (*1*) that the 88-inch (224-cm) cyclotron at Berkeley had been used in the first successful test of the technique: the $^3H/^2H$ ratio had been measured to determine the age of a sample approximately one mean-life old. In that experiment a range-separation technique had been used to eliminate potential backgrounds, such as ^3He, that are produced in the ion source with a charge-to-mass ratio (e/m) very similar to that of ^3H. Soon after the publication of that result, several other groups demonstrated (*2*) that the range-separation stage could be avoided in the case of ^{14}C if a tandem Van de Graaff accelerator were used in place of a cyclotron, since the main source of background, ^{14}N, does not form negative ions. Dates have now been obtained for samples several milligrams in mass with previously known ages up to about 40,000 years (*3*). The sensitivity appears to be limited by ^{14}C associated with the accelerator. It may be possible to eliminate the background if the specific source can be found and removed, or if an accelerator with low radiation background is constructed specifically for radioisotope dating.

We report here on the first accelerator dating of a "blind" sample of carbon. The 88-inch cyclotron at Berkeley was used to accelerate $^{14}C^{3+}$ ions to an energy of 60 MeV, and the range-separation technique was used to separate and eliminate the background from $^{14}N^{3+}$. The sample was obtained in the form of CO_2 from R. Berger at the University of California at Los Angeles, who had measured the age using the standard radiocarbon "decay dating" technique invented by W. R. Libby. Our measured age of 5900 ± 800 years was obtained and announced (*4*) without prior knowledge of the age obtained by Berger's group.

The range-separation technique is based on the observation that the range of ^{14}C is 1.3 times that of ^{14}N (*1*). We accomplished the separation by passing the output beam of the cyclotron through a xenon gas cell about 10 cm long. Gas was used in order to assure a uniform material; xenon was chosen because its high Coulomb barrier minimizes nuclear interactions. The gas was separated from the vacuum of the cyclotron with a window of platinum and gold less than 1 μm thick. This window was supported on a tungsten grid with a clear aperture of approximately 65 percent. With the tungsten support, the window was able to hold a pressure difference of greater than 1 atm.

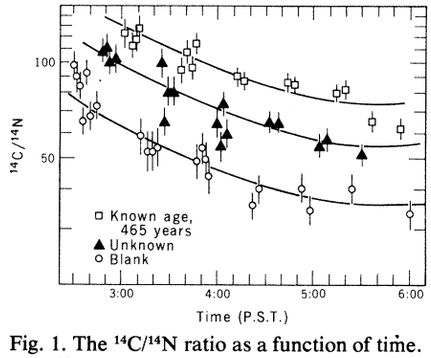

Fig. 1. The $^{14}C/^{14}N$ ratio as a function of time. The ^{14}C was measured with an ionization chamber–silicon detector telescope. We measured the ^{14}N by integrating the current from slits which collimate the beam soon after it emerges from the cyclotron and well before it enters the xenon gas cell. Three samples were alternated: one of known age 465 years, a blank known to contain no ^{14}C, and the unknown. The fit shown corresponds to an age of 5900 years. The time scale is in hours.

Adjacent to the xenon gas cell was an ionization chamber–silicon detector telescope to measure the ionization and total energy of the emerging particles. This telescope provided unambiguous identification of the nuclear charge of the particles emerging from the xenon cell.

The range-straggling curve for the nitrogen ions in the xenon gas cell was very steep. When the xenon pressure was maintained at 1.15 times the pressure corresponding to a 10-cm range of the nitrogen, all of the nitrogen ions stopped in the cell. In over 10 hours of operation with 1 to 10 nA of $^{14}N^{3+}$ incident on the xenon, not a single nitrogen ion emerged into the particle telescope to register a coincidence. Thus the discrimination against nitrogen was better than 10^{-14}.

Although the nitrogen background was effectively eliminated, another background has thus far limited the sensitivity of the measurements. Even when measuring a sample known to contain no ^{14}C, we found that carbon counts register in the detector telescope. The most plausible origin of the background counts is highly radioactive carbon within the cyclotron vacuum. Our level of background ^{14}C appears to be higher than that reported by the Rochester group (*3*).

Because of the high ^{14}C background, it is necessary to make a subtraction in order to obtain an age. Figure 1 shows the results of an experiment in which the cyclotron source gas was switched rapidly between three gas samples, one known to be 465 years old, the unknown, and a blank known to contain no ^{14}C. All three samples had been measured at Berger's laboratory. The data were fitted to a simple model with five constants: the age of the unknown, the background level, the overall normalization (that is, the cyclotron efficiency), and two constants to describe the slow variation of the source output with time.

From the fit we obtained an age (*5*) for the unknown of 5900 ± 800 years. The errors are statistical only and we obtained them by varying the age parameter and refitting the data. The variation corresponding to a chi-square change of one gives an estimated error of 1 standard deviation. Although our statistical error is twice that reported by the Rochester group, it is comparable to the accuracy they obtained for a sample of similar age (*6*).

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The age measured by Berger's group is 5080 ± 60 years. This result differs from ours by 820 years, about 1 standard deviation. The significance of the agreement is not in the accuracy of the cyclotron result, which does not compare with that attained by the standard decay-dating approach. For large samples that are not many half-lives old, the cyclotron technique may never match the accuracy obtainable by present radiocarbon laboratories. However, less than 150 mg of carbon was used in the measurement and most of this was used in the flushing of the manifold between sample changes; we anticipate that the amount of carbon used can be reduced by another order of magnitude. The ability to use small samples should greatly simplify the problems of selecting samples uncontaminated by recent carbon, and of dating objects too small or valuable to allow the extraction of the 1 to 10 g of carbon required in the past. If the background ^{14}C can be removed, either from the use of an external ion source or from the construction of a "clean" cyclotron, then it is possible that the full predicted sensitivity of the cyclotron technique will be achieved: 40,000 to 100,000 years for 10- to 100-mg carbon samples.

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4. Our results, including a promise to publish them, were mailed to R. Berger, W. R. Libby, and H. E. Gove on 13 December 1977, the day before we learned the date obtained by Berger's group. This report, except for the final paragraph, was written before we knew Berger's date. The need for taking such precuations is clearly expressed in F. G. Dunnington's description of his measurements of *e/m* for the electron [*Phys. Rev.* **52**, 475 (1937)].
5. We report the data in "radiocarbon years," using the standard lifetime for ^{14}C of 5570 years. The best modern estimate for the half-life is 5730 years [H. Godwin, *Nature (London)* **195**, 984 (1962)].
6. In (*3*) the group at the University of Rochester estimated the age of their Mount Shasta sample to be 5700 ± 400 years. The age measured by the U.S. Geological Survey, using decay dating, was 4590 ± 250 years, which differs from the Rochester date by about 1100 years.
7. We are grateful to R. Berger and W. R. Libby for their support and encouragement, and for supplying the blind sample. We have received important help in this work from L. W. Alvarez, H. Weiman, G. Wosniak, W. Erwin, L. Archambault, H. Dougherty, J. Yamada, and the staff of the 88-inch cyclotron at the Lawrence Berkeley Laboratory. This work was supported by the Department of Energy.

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Devonian Brachiopods from the Sillimanite Zone, Mount Moosilauke, New Hampshire

Abstract. *Devonian brachiopods, identifiable at the generic level, have been recovered from calc-silicate rocks more intensely metamorphosed and metasomatized than any other known fossil occurrence. The fossils are a key stratigraphic link between granulite facies rocks of central New Hampshire and fossiliferous rocks of western New Hampshire and Maine.*

Devonian brachiopods have been found in the sillimanite zone of the Littleton Formation, Mount Moosilauke, New Hampshire. The fauna is remarkably well preserved so that identification at the generic level is possible. The specimens are unique in that they have been metamorphosed at higher temperatures and have been subjected to more intensive metasomatism than any other known regionally metamorphosed fossils.

The fossils are significant because they paleontologically confirm Billings' (*1*) assignment of rocks of the Mount Moosilauke septum to the Devonian Littleton Formation. Furthermore, the brachiopods are a key stratigraphic link between the essentially unfossiliferous granulite facies metamorphic rocks of central New Hampshire and the fossiliferous Siluro-Devonian sections of western New Hampshire and Maine.

Generically identifiable brachiopods and a coral of late Early Silurian age have been reported from the sillimanite zone near Claremont, New Hampshire, 80 km south-southwest of the locality of the new discovery (*2*). The locality of the earlier recovery lies along the boundary between the staurolite-kyanite and sillimanite zones (*3*), presumably within the lower sillimanite zone. The fossils are preserved as coarsely crystalline calcite in a matrix consisting of quartz, diopside, grossular, and hornblende (*2*). The rocks of the older locality were metamorphosed at a lower temperature but higher pressure and have been less intensely metasomatized than the newly discovered fossils.

The fossils are located in the cascades of Beaver Brook, on the southwestern wall of Kinsman Notch, Mount Moosilauke 7-$^1/_2$′ quadrangle in New Hampshire. The bed of fossils (5 to 10 cm thick) is the innermost layer within a banded, tan or pink, calc-silicate unit (20 to 30 cm thick) that is interbedded with gray mica schist. The shells are aligned parallel to the bedding of the rock, are thoroughly disarticulated, little sheared, and unbroken. The rocks containing the fossils were correlated by Billings (*1*) with the Littleton Formation. The contact between the Littleton Formation and the Kinsman Quartz Monzonite is located 200 m east of the fossil locality.

The fauna recognized in the initial sample processed for fossils is as follows: *Acrospirifer* cf. *A. murchisoni; Leptocoelia* cf. *L. flabellites; Atrypa* cf. *A. "reticularis"; Leptostrophia* or *Protoleptostrophia* sp.; and a high-spired gastropod similar to *Loxonema*. The *Acrospirifer* is coarsely plicate and larger than similar-shaped spiriferids of Silurian age. *Leptocoelia sensu stricto* is known only from beds of Oriskany and Esopus age in the Northern Appalachians. *Atrypa* is not found in beds younger than early Late Devonian (Frasnian), nor earlier than late Lower Silurian (Upper Llandovery). Marine fossils of post-Early Middle Devonian (Onondaga, Eifelian) age are unknown in New England. Fossiliferous beds of Esopus age are rare in the Northern Appalachians, but beds of Oriskany age are widespread. Therefore, it is likely that the Beaver Brook fossils are of Oriskany age although an Esopus age cannot be ruled out.

The fossil shells consist of either calcite and quartz or wollastonite, calcite, and quartz. Impressions of the shells are composed of either grossular, diopside, and sphene, or grossular, diopside, sphene, and zoisite. The mica schists with which the fossils are interbedded contain the assemblage quartz–biotite–muscovite–andalusite–sillimanite (fibrolite)–staurolite (retrograde)–garnet–tourmaline–ilmenite (*4*). The Kinsman Quartz Monzonite is made of quartz–plagioclase (oligoclase–andesine)–potash feldspar–biotite–muscovite (*1*). Metamorphism of the fossils took place at a pressure of ≤3.8 kbar and a temperature of 625° ± 25°C; these conditions are estimated on the basis of the presence of the mineral assemblages listed above and their experimental calibration (*5*). Chemical metasomatism of the fossils included silicification of the shells during diagenesis or low-grade metamorphism followed by loss of CO_2 during high-grade metamorphism according to the reaction

$$CaCO_3 + SiO_2 = CaSiO_3 + CO_2$$

(calcite) (quartz) (wollastonite)

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