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RADIOISOTOPE DATING WITH AN ACCELERATOR: A BLIND MEASUREMENT

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ABSTRACT

The age of a sample of CO_2 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.

Work performed under the auspices of the U. S. Department of Energy.

RADIOISOTOPE DATING WITH AN ACCELERATOR: A BLIND MEASUREMENT

In a recent paper Muller 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 size of sample required (1). In the same paper he reported that the 88" cyclotron at Berkeley had been used in the first successful test of the technique: the tritium/deuterium 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 Helium-3, which are produced in the ion source with a charge-to-mass ratio very similar to that of tritium. Soon after the publication of that result, several other groups demonstrated (2,3) that the range-separation stage could be avoided in the case of carbon-14 if a tandem Van de Graaff accelerator were used in place of a cyclotron, since the main source of background, nitrogen-14, does not form negative ions. Accurate dates have now been obtained for samples with previously known ages up to about 40,000 years (4). The sensitivity appears to be limited by carbon-14 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. If that is accomplished, then it appears likely that the full anticipated potential (1) of accelerators for radioisotope dating will be achieved. For carbon-14 the age limit should be approximately 100,000 years for samples 100 mg in mass.

We report here on the first accelerator dating of a "blind" sample of carbon. The 88" 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 UCLA, who had measured the age using the standard radiocarbon "decay dating" technique invented by W. R. Libby. Our measured age of 6060 ± 800 years was obtained and announced (5) without prior knowledge of the age obtained by Berger's group.

The range separation technique relies on the observation that the range of carbon-14 is 1.3 times that of nitrogen-14 (1). The separation was accomplished 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 one micron thick. This window was supported on a tungsten grid with a clear aperture of approximately 65%. With the tungsten support, the window was able to hold a pressure difference of greater than one atmosphere.

The xenon gas cell was followed by 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 ten hours of running 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 carbon-14, carbon counts are registered in the detector telescope. The most plausible origin of the background counts is highly radioactive carbon within the cyclotron vacuum. Our level of background carbon-14 appears to be higher than the level of such background reported by the Rochester group (4).

The high carbon-14 background makes it necessary to make a subtraction in order to obtain an age. Figure 1 shows the results of a run 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 carbon-14. All three samples had been measured at R. Berger's radiocarbon laboratory. The data were fitted to a simple model with 5 constants: the age of the unknown; the background level; the overall normalization (i.e. the cyclotron efficiency); and two constants to describe the slow variation of the source output with time.

From the fit we obtained an age (6) for the unknown of 6060 ± 800 years. The errors are statistical only, and were obtained by varying the age parameter and refitting the data. The variation corresponding to a chi-square change of one gives the one-standard-deviation error estimate. 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 (7).

The age measured by Berger's group is 5230 ± 60 years. This result differs from ours by 830 years, about one standard deviation.

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5. Our results, including a promise to publish them, were mailed to R. Berger, W. F. Libby, and H. E. Gove on 13 December, the day before we learned the date obtained by Berger's group. This paper, except for the numbers appearing in the final paragraph, was written before we knew Berger's date. The need for taking such precautions is clearly expressed in F. G. Dunnington's description of his measurements of e/m for the electron in Phys. Rev. 52, 475 (1937).

6. We report the date in true years, rather than in "radiocarbon years" as is common in the radiocarbon literature. To convert our answer to radiocarbon years multiply by 5568/5730, the ratio of the "standard" half-life, to the best modern estimate published by H. Godwin, NATURE (London) 195, 984 (1962).

7. In reference (4) the Rochester group estimated the age of their Mt. Shasta sample to be 5700 ± 400 years. The age measured by the USGS using decay dating was 4590 ± 250 years, which differs from the Rochester answer by about 1100 years.

8. We are very grateful to R. Berger and W. F. Libby for their support, 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" cyclotron at the Lawrence Berkeley Laboratory. This work was supported by the Department of Energy.

FIGURE CAPTION

The $^{14}C/^{14}N$ ratio as a function of time. The ^{14}C was measured with an ionization-chamber, silicon-detector telescope. The ^{14}N was measured 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 sample known to contain no ^{14}C; and the unknown. The fit shown corresponds to an age of 6060 years.

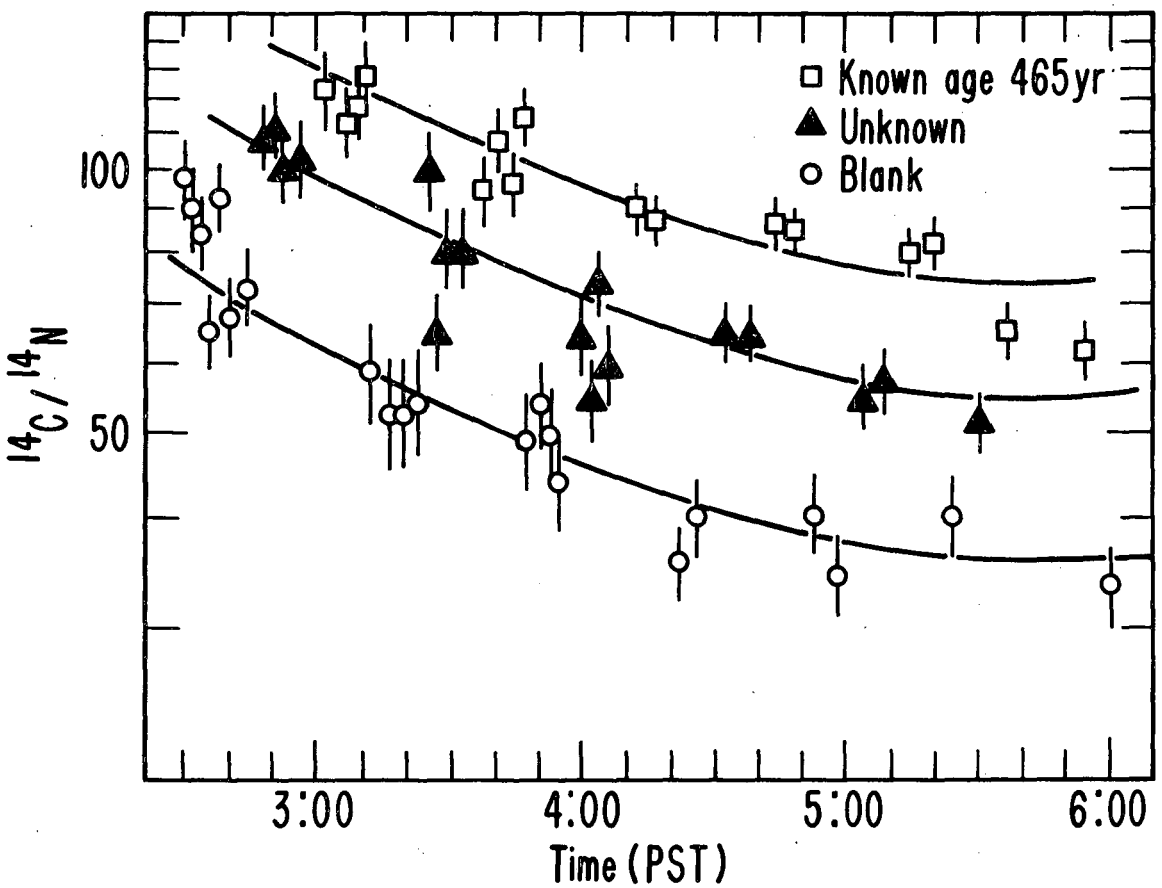

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Fig. 1

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