Radioisotope Dating with Accelerators

Complete analysis of minute samples shows promise of extending power of the method

BY DIETRICK E. THOMSEN

Radioisotope dating is one of the contributions of nuclear physics to archeology. It allows for calculation of the time since a given object was taken out of equilibrium with the atmosphere — that is, the time since the object was buried or submerged. Alone, or in correlation with biological (dendrochronological) or artistic evidence, the radioisotope technique can be used to determine the date at which a fossil was alive or a human artifact was made.

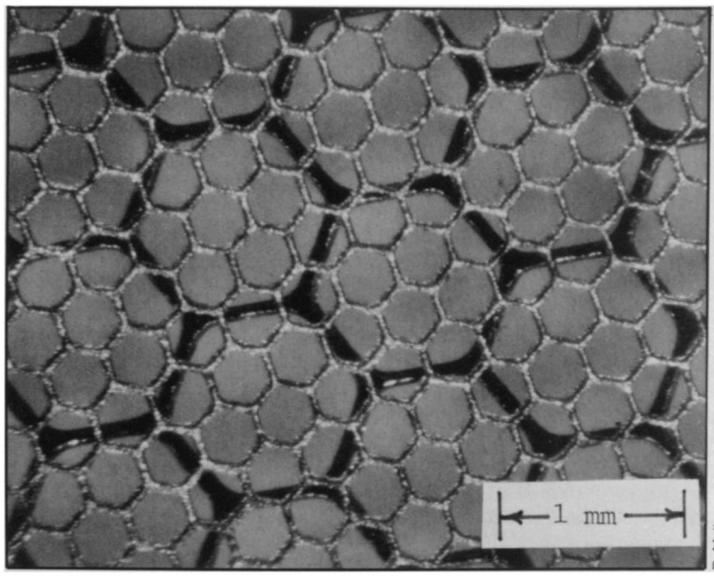
The isotope usually used for the technique is radioactive carbon 14, which is made in the atmosphere by cosmic rays and constantly ingested by living organisms. The cosmic rays make carbon 14 out of nitrogen 14 at a rate of approximately 13.5 atoms per minute for each gram of carbon on the earth's surface. Thus, each gram of carbon in equilibrium with the atmosphere will exhibit 13.5 decays of carbon 14 per minute, more or less. If the object containing the carbon has been buried for a long time, the decay rate will be less, because a proportion of the carbon 14 will have been depleted. Knowing that the half-life of carbon 14 is about 5,730 years, one can calculate the amount of time the sample has been buried from the current decay rate.

The technique is straightforward, and, in the hands of an expert, can produce accurate results. But it can take a long time and require what, for archeological remains, is often a sizable sample. For ages between 5,000 and 10,000 years something on the order of 10,000 carbon 14 disintegrations must be observed. This will take between 1 and 10 grams of carbon and 1.5 to 15 hours of counting. That is a lot of time and what can be a large slice of a rare and valuable item.

A method that takes less time and mass is under development using particle accelerators at the University of California's Lawrence Berkeley Laboratory and the University of Rochester (SN: 6/25/77, p. 405). It consists essentially of counting all the carbon 14 atoms now present in an archeological sample rather than the rate at which decay is taking place. This type of analysis opens the additional possibilities of using isotopes other than carbon 14 to establish dates with scales ranging from dozens to hundreds of years (for tritium) to tens of millions of years (for beryllium 10).

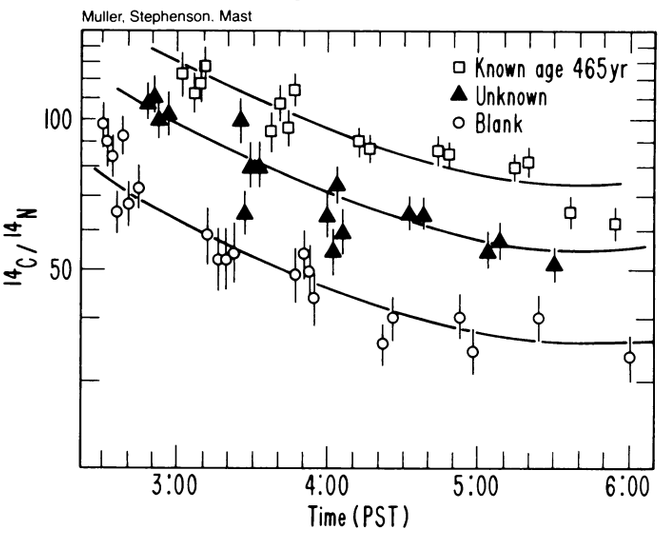
The usual way of making such a trace-element analysis is to vaporize and ionize the sample and put it through a mass spectrometer. The mass spectrometer separates ions according to the ratio of their electric charge to their mass. This method of separation has a certain built-in ambiguity, because ions of different elements may have the same charge-to-mass ratio. The ambiguity is tolerable in many applications, but it prevents using an ordinary mass spectrometer for the kind of trace element analysis involved in radioactive dating. The counts of the wanted ions tend to be so low that a small background count of intruders with the same charge-to-mass ratio can make the determination impossible. For instance, carbon 14 ions with a charge of 6 cannot be distinguished from nitrogen 14 ions with the same charge.

The way out of the problem is to use a particle accelerator as a mass spectrometer. The accelerator gives the particles much more energy than an ordinary mass spectrometer does, and this permits discrimination between ions of nearly the same charge-to-mass ratio. Generally, different ions will have different ionization rates and therefore penetrate barriers to different distances even though their



Entry to the xenon cell that separates nitrogen 14 from carbon 14 in the Berkeley dating experiments is through this pair of superimposed tungsten screens that support a gold-plated perforated platinum window.

Stephenson, Muller adjust xenon cell.



Experimental runs done in the early morning compared carbon 14 to nitrogen 14 ratios in three samples. Figures for the blank sample gave the internal cyclotron background and permitted computation of the age of the unknown by comparison with the known.

charge-to-mass ratio is the same.

At LBL the 88-inch sector-focused cyclotron is being used in a series of experiments to test the feasibility of the method. Richard A. Muller, one of the chief LBL experimenters in the program, described the progress up to last spring in an article in the April 29 SCIENCE At the University of Rochester, a tandem Van de Graaff machine is the instrument chosen by a group led by Harry Gove and including C. L. Bennett, M. R. Clover, and W. E. Sondheim of Rochester, A. E. Litherland and R. P. Beukens of the University of Toronto, and K. H. Purser and R. B. Liebert of General Ionex Corp.

To distinguish carbon 14 from the intruding nitrogen 14, the LBL group uses a cell of xenon gas. The xenon will stop the nitrogen 14, because it loses energy faster by ionizing the xenon, but will pass the carbon 14. Xenon was chosen because it has a very low probability of nuclear interactions with the two ions, but the choice of a gas rather than a solid required the design of a complicated window between the gas cell and the vacuum of the cyclotron. A foil of platinum a third of a micron thick will pass the ions, but it will not support the pressure of one atmosphere of xenon. To make the platinum strong enough, it was reinforced with two screens of tungsten, at the mesh screen on top of a coarse mesh one. Pinholes in the platinum were covered with gold foil. This arrangement succeeded in passing the ions and holding the xenon pressure.

For carbon dating, the tandem Van de Graaff used at Rochester has the advantage of not requiring such a discriminator. The reason is that a tandem Van de Graaff accelerates negative ions, atoms with an extra electron bound to them. Most elements will bind an extra electron. The exceptions are the heavier noble gases — and nitrogen 14. Therefore, the Van de Graaff will accelerate carbon 14 but not nitrogen 14, and a nitrogen 14 stopper is not necessary. This advantage leads Gove to predict that when an accelerator dedicated to carbon 14 dating is built, it will be a Van de Graaff. However, if other radioisotopes are used for dating, the contamination problems for the two kinds of accelerator are similar.

The idea of using cyclotron as a mass spectrometer goes back to 1939 when Luis W. Alvarez and R. Cornog used it in their discovery of the rare stable isotope helium 3. Muller says the technique was not used from that time to the present series of experiments.

The first use of the cyclotron as mass spectrometer in the present work was not for radioisotope dating, but in a search for quarks with unit charge that was carried out by Muller, Alvarez, William R. Holley and Edward J. Stephenson (also reported in the April 29 SCIENCE). Quarks are, according to theory, the subunits out of which particles like protons, neutrons and their many relatives are built. The most widely accepted form of quark theory (originated by Murray Gell-Mann and George Zweig) contained three quarks, each of which had either one-third or two-thirds of the electric charge of the electron, which is the basic unit of charge for subatomic particles. However, at about the same time, M. Han and Y. Nambu developed a version of the theory that employed nine quarks, each with unit charge.

The Han-Nambu version at first gained fewer adherents than the Gell-Mann-Zweig version because of the principle called Occam's Razor: Given rival explanations of the same phenomenon, choose the simplest one. But in recent years previously unknown properties of subatomic particles have been discovered. To account for them, the number of quarks in the Gell-Mann-Zweig version has had to be raised to nine, so now the Han-Nambu version seems a more feasible alternative.

If unit charge quarks really exist, and some of them are floating around unattached to other quarks, they should appear as an "anomalous" kind of hydrogen. That is, a quark with the same electric charge as a proton but much more mass should play the role of an unusually heavy hydrogen nucleus in chemistry and atomic physics. So an experiment was mounted with the 88-inch cyclotron to search for such ultraheavy hydrogen. None was found, and the experimenters so reported before going on to radioactive dating experiments.

From analysis of hydrogen samples in the search for unit-charge quarks, the experimenters went on to dating by the radioactive hydrogen isotope, tritium. Tritium has a short half-life (17.8 years), but it still can be useful for dating water samples in hydrology, oceanology and meteorology. For example, it may be desirable to know how long it will take to refill an underground reservoir of water that is being tapped. Tritium analysis can give an idea, by telling how long ago the water present at the time of tapping went out of contact with the atmosphere, where tritium is made. One of the first tests used a sample of water known to be 24 years old. The cyclotron analysis produced an age of 33 years. In future, the experimenters hope to refine the accuracy of tritium dating. A large number of samples of known age to practice on can be obtained by opening bottles of vintage wine.

Since the publication last spring, the experimenters have been working on carbon 14 dating, and have successfully obtained dates for some known samples. Muller describes archeologists as excited by the idea, because the accelerator method tends to require a few to a few hundred milligrams of material compared to grams for the older method. This means that dating will be possible for objects that contain too little material for the older method and with much less damage for larger objects. A work of art, an ancient document or an ancient fabric would be subject to much less injury in the dating process.

Archeologists, especially Willard Libby, who was one of the developers of the original carbon 14 dating technique, and Rainer Berger, have been supplying samples with known dates for the physicists to use in proving the reliability of the method. In the past, the LBL group has known the age of the sample they were experimenting with. Recently they succeeded in dating a blind sample, a procedure Muller insists is the final way to test the method. In a paper submitted to SCIENCE, Muller, Stephenson and Terry S. Mast report that they have dated some carbon monoxide submitted by Berger, without knowing the date Berger's group had determined by the usual carbon 14 counting method, and come out with a result within one standard deviation of the age determined by the Berger group.

The experiment was done by switching the cyclotron beam back and forth among the unknown, a sample of known age and a sample known to contain no carbon 14. The last, the blank sample, was necessary because there is a source of carbon 14 somewhere in the cyclotron. Muller, Stephenson and Mast determined the date of the unknown and sent it out to Berger and various other interested scientists. They wrote the paper except for the last paragraph, in which the dates are compared, and then asked Berger what his date was. The cyclotron got 6,060 ± 800 years. The Berger group's date was 5,230 ± 60 years.

The Rochester group has not done a blind sample, but they have determined dates of several known samples, including one of age comparable to that of the LBL group's blind sample, and with a smaller statistical error. The results so far indicate that use of accelerators will extend the power of carbon 14 dating. Carbon 14 dates now rarely go beyond 20,000 years. "We expect to have no trouble dating back to 100,000 years," says Muller.

Everyone interested in accelerator dating methods expects them to increase the number of radioisotopes used for dating. Gove says there are many that scientists would like to use. In addition to carbon 14, his group has made some tries with aluminum 26, but they have found difficulty with contamination by magnesium 26. One of the big possibilities is beryllium 10, and it is in the prospective use of this species that Muller points to an advantage for the cyclotron. Beryllium 10 is made in the atmosphere. It mixes gradually with the air and then with ocean water, finally settling out on the sea bottom. It will be necessary to determine the manner and rate of this deposition in order to establish a dating scale for use with beryllium 10. But if that can be done, the long half-life of beryllium 10, 1.5 million years, will permit the extension of radioisotope dating to geological samples up to 35 million years old or older. □