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RESULTS OF TESTS AND MEASUREMENTS FROM THE NSF REGIONAL ACCELERATOR FACILITY FOR RADIOISOTOPE DATING

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ABSTRACT. Tests of performance of the tandem accelerator mass spectrometer at the NSF Regional Facility at the University of Arizona are discussed. Results of measurements on some tree rings and on some archaeologic samples are presented.

SYSTEM PROPERTIES

The accelerator and associated instrumentation were delivered to our laboratory by General Ionex Corporation, Newburyport, Massachusetts, on September 15, 1981. The installation was completed by November, and ^{14}C was first detected on February 2, 1982. In this paper, we present some results of tests and measurements made with the accelerator. The ion source is a cesium sputter type in which the cesium beam is incident upon a flat target at an angle of ca 35° to the normal to the target face. With graphite targets we have been able to produce currents of ^{12}C, analyzed through the injection magnet, of nearly 20 micro-amperes. The accelerator has been operated at a voltage of 2.2MV. For the ^{14}C measurements, a terminal voltage of 1.8MV was used. The system is operated using an argon gas stripper. We do not, as yet, have a vacuum pump in the terminal. Figure 1 shows, for a terminal voltage of 1.8MV, the transmission of the system,

$$T = \frac{\text{particle current of } {}^{12}C^{q} \text{ at high-energy end}}{{}^{12}C^{-} \text{ at low-energy end}} ,$$

as a function of gas pressure as measured by a Penning ionization gauge located at the high-energy end of the accelerating tube. For $^{12}C^{+++}$ ions, the maximum transmission occurs when this pressure is 5×10^{-6} Torr, which is about a factor of ten higher than the pressure with no stripper gas. Figure 2 shows the transmission as a function of terminal voltage, when the stripper is set at its optimum value for the 3^{+}

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charge state. The transmission for $^{12}C^{+++}$ is 20%. An extrapolation of the 3^+ curve indicates that at a terminal voltage of about 2.4MV, this transmission would reach a maximum of ca 25%. In figure 3, the transmission for 3^+ ions is plotted as a function of ion velocity at the terminal. The velocities of ^{12}C, ^{13}C, and ^{14}C are indicated for a terminal voltage of 1.8MV, and, as can be seen, ^{14}C ions are produced 20% less efficiently than are ^{12}C ions.

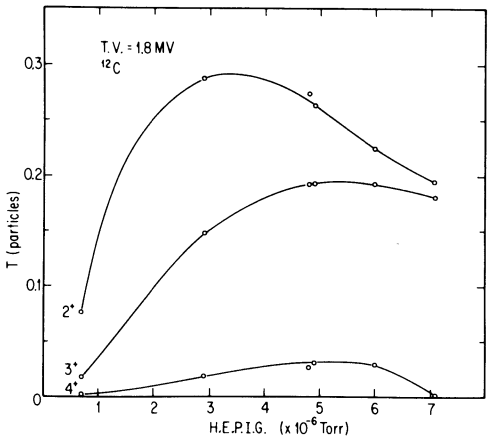

Fig 1. Particle transmission, T, through the accelerator of various charge states as a function of stripper-gas pressure, measured at the high-energy end of the accelerator tube

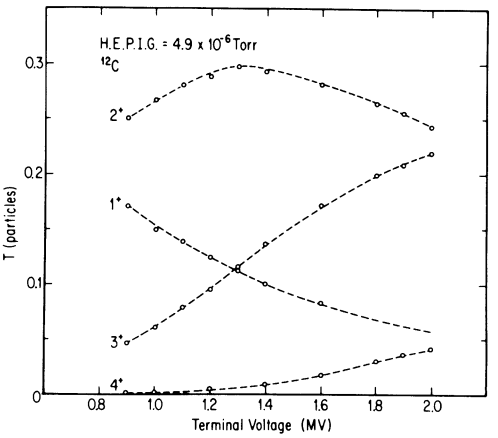

Fig 2. Particle transmission, T, vs terminal voltage

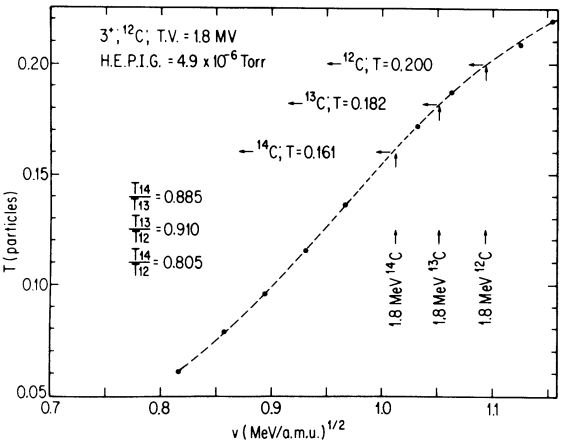

Fig 3. Particle transmission of $^{12}C^{+++}$ ions vs velocity of ^{12}C ions. Velocities of ^{14}C, ^{13}C, and ^{12}C ions with kinetic energies of 1.8MeV are indicated.

Ions of ^{14}C are detected in a silicon surface-barrier detector. Nitrogen ions are eliminated in one of two ways. The silicon detector can be operated in conjunction with a gas-ionization detector from which $\Delta E/\Delta x$ pulses are obtained for use in discriminating against nitrogen pulses. At times we use an alternative method for rejecting pulses from nitrogen. A thick absorber, 250 $\mu g/cm^2$, placed in front of the silicon detector is used to reduce the energy of the nitrogen ions by enough so that they can be rejected by energy discrimination in the silicon detector.

From the currents obtained from the ion source and from the accelerator transmission shown in figures 2 and 3, we calculate that for a current of 2.5 μamp $^{12}C^-$ (a realistic value from archaeologic samples) a ^{14}C count rate of 3 c/sec should be obtained for a modern sample. We do, in fact, observe such count rates.

TESTS WITH GRAPHITE

For most of the early tests, targets of spectroscopic graphite were used. The graphite was pumped free of nitrogen and irradiated in a nuclear reactor to produce a ^{14}C content of 1.57 $\pm$ 0.05 times the content of modern (1950) wood. We used these samples to measure the reproducibility of $^{14}C/^{13}C$ ratios in a series of targets. The methods used are best des-

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cribed by referring to the schematic diagram in figure 4.

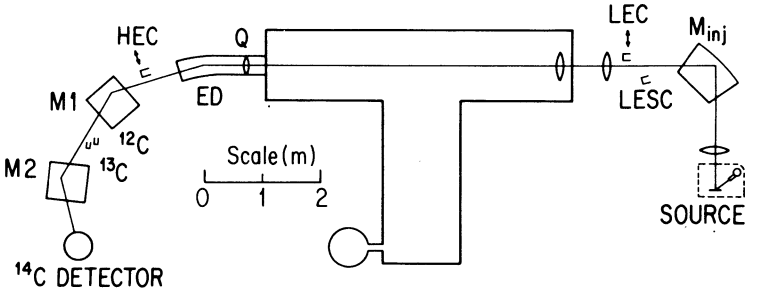

Fig 4. Schematic diagram of system. Abbreviations in the figure are: M_{inj}, injection magnet; LEC, low-energy Faraday cup (this cup can be moved in and out of the beam); LESC, low-energy side cup; Q, electrostatic quadrupole focus; ED, electrostatic deflector; M1 and M2, high-energy magnets; ^{13}C and ^{12}C, high-energy Faraday cups

The procedures used were as follows: 1) with a suitable target, enriched by a factor 2 to 10 in ^{14}C, set ion source controls, lenses, and other electrostatic and magnetic elements to optimize ^{14}C rate in detector; 2) move standard sample into appropriate position in ion source; 3) measure ^{14}C rate in detector. Counting time is from 2 to 6 minutes; 4) manually adjust injection magnet to pass M = 13. With a current integrator, measure ^{13}C current in high-energy side cup. Counting time is 10 to 30 seconds; 5) move unknown sample into position in the ion source; 6) repeat steps 3) and 4); 7) calculate the ratio

$$R = \frac{(^{14}C/^{13}C)_u}{(^{14}C/^{13}C)_s} .$$

Results of $^{14}C/^{13}C$ ratios made with a pair of graphite targets are shown in figure 5. Each point represents a cycle with the injection magnet, and gaps occur where the targets were interchanged. The measurement time for ^{14}C was 200 seconds and for ^{13}C, 10 seconds; targets were cycled about every 10 minutes. Two points should be noted. First, for target

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#6, results were very stable. The standard deviation of the mean value of all the points is 0.5%, and is essentially equal to the precision from counting statistics. Second, the ratio for the upper target exhibits a trend with time. The problem is probably with the target itself. Quantitative results obtained with graphite targets appear to be very sensitive to properties of the target. We have observed that as the cesium beam produces cratering on the surface of the graphite target, $^{14}C/^{13}C$ ratios have changed by as much as a factor of two. In fact, the ratio begins to change before any obvious surface defects appear. From the results for target #6, we conclude that, in this set of data, systematic errors introduced by system fluctuations, by cycling of the injection magnet, and by moving the target in and out of position, were less than statistical errors, which were 0.5%.

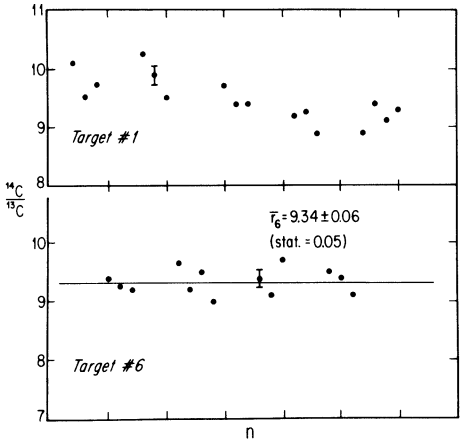

Fig 5. A plot of measurements of the ratio $^{14}C/^{13}C$ from irradiated graphite targets. Each point represents a cycle of the injection magnet and each gap in the points indicates a change on the target. The flags indicate the size of statistical error on individual points.

TESTS WITH Al_4C_3

We have tried many ways in which to prepare accelerator targets from cellulose and/or CO_2. Charcoal is easy to prepare from wood or cellulose and alternatively, CO_2 can be reduced to carbon by reduction with hot magnesium. Carbon in either of these forms is either mixed with a binder or heated to form a carbide. The product is packed into a sample holder. We have tried mixtures such as Ag-C, Pb-C Cu-C and KBr-C, and compounds such as Fe_3C, Mn_3C and Al_4C_3 and have had

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mixed and inconsistent results. At the present time, Al_4C_3 appears to be most satisfactory of the compounds. We consistently produce targets that yield from one to three micro-amps of $^{12}C^-$, resulting in 2 to 4 counts/sec from 1890 wood samples. The Al_4C_3 targets are easy to make and appear to stand up well under bombardment. Although cratering does occur in Al_4C_3 targets after a few hours of use, isotopic effects similar to those from graphite targets have not been observed in the 3% measurements made with these targets. Figure 6 illustrates that results of $^{14}C/^{13}C$ in two "real" targets can be made to 3%, and we expect that simply counting longer will improve that precision.

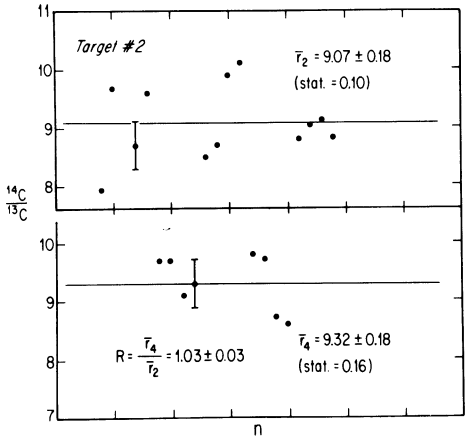

Fig 6. Plots similar to those in figure 5, but for Al_4C_3 targets.

BACKGROUND

Measurements of background are made by optimizing the parameters of the instrument with a target containing ^{14}C, and then measuring ^{14}C rates produced from a carbon target that contains no ^{14}C. Targets made from old (age > 58,000 yr) graphite, from bottled CO_2, and from limestone have been used. Results seem to depend on a variety of parameters which are not simple to define quantitatively. In figure 7, the ^{14}C rate from various targets is plotted against the total output of carbon ions from the ion source, as measured by the $^{12}C^-$ current in the low-energy cup. For these data, the gas-ionization chamber was used in conjunction with the silicon detector to ensure that the residual counts have the $\Delta E/\Delta x$ values of carbon. The figure includes data obtained with graphite and with Al_4C_3 targets made from old CO_2 and from limestone.

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The value of the ^{14}C rate extrapolated to $i(^{12}C) = 0$, 1 or 2 counts per 1000 seconds, is approximately the rate observed with a pure aluminum target. Current of $^{12}C^-$ in the low-energy cup observed from pure aluminum is generally a few nano-amperes. Most Al_4C_3 targets of interest produce currents of < 4 micro-amperes, so that, for the present, a background is used as indicated by the horizontal lines in the curve; i e , we assume a ^{14}C background rate = 0.02 ± 0.01 counts/sec. We expect that, eventually, this rate will be decreased. In terms of sample age, if a sample were 41,000 years old, its signal would be about equal to the background.

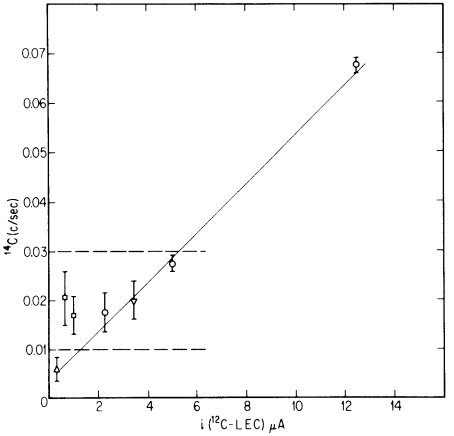

Fig 7. Background count rates plotted vs total carbon current from the ion source. Circles were obtained with graphite targets, and the others, with Al_4C_3 targets.

ARCHAEOLOGIC MEASUREMENTS

Measurements were made with Al_4C_3 targets prepared from tree rings provided by C W Ferguson, from corn obtained from Bat Cave by Austin Long, and from several CO_2 samples prepared by R E Taylor, from bones of a skeleton known as "Sunnyvale girl". The results of these measurements are listed in Table 1. Column 3 lists measured values of the ratio

$$R = \frac{(^{14}C/^{13}C)_{sample}}{(^{14}C/^{13}C)_{standard}} \quad .$$

The measured ratio of $^{14}C/^{13}C$ in 5050 BC tree rings and in AD 1890 tree rings is shown as R = 0.50 ± 0.03. The expected value for this ratio (R = 0.48) was obtained from the calibration curves of Klein et al (1982). The age of the Bat-cave

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corn as shown agrees well with a recent measurement by Long (pers. commun; 1982) on charcoal taken from that cave.

TABLE 1.Results of measurements

Sample	Standard	Ratio (measured)	Radiocarbon Age ^{14}C years B P ($T_{\frac{1}{2}}$ = 5568 yr)
Tree ring, 5050 BC	Tree ring, AD 1890 (ratio from calibration (1))	0.50 ± 0.03 0.48	
Bat-cave corn	Tree ring, AD 1890	0.84 ± 0.10	1460 ± 800
Sunnyvale, 1437A	Tree ring, 5050 BC	1.36 ± 0.10	3600 ± 600
Sunnyvale, 1437B	Tree ring, 5050 BC	1.16 ± 0.05	4850 ± 400
Sunnyvale, 1437D	Tree ring, 5050 BC	1.19 ± 0.05	4650 ± 400

Results of measurements on CO_2 from three different fractions of bone from the "Sunnyvale girl" are presented. According to Taylor (pers. commun; and (1983)), sample 1437A is from total acid-soluble organics, sample 1437B is from total base-soluble organics, and sample 1437D is from total insoluble organics, after gelatin conversion. Figure 8 shows results of $^{14}C/^{13}C$ measurements in which targets prepared from 5040-5050 BC tree rings and from sample 1437D (CO_2 from total insoluble organics) were alternated in the ion source. It is clear that the $^{14}C/^{13}C$ ratios of the two targets are comparable.

Measurements were made in collaboration with L A Currie and G A Klouda of the National Bureau of Standards on several atmospheric and standard samples. These measurements were made with our original target chamber and will be repeated. One result of interest is that a $^{14}C/^{13}C$ ratio was measured with a statistical precision of 10% with an Al_4C_3 target fabricated from oxalic acid which contained slightly < 100 μg of carbon.

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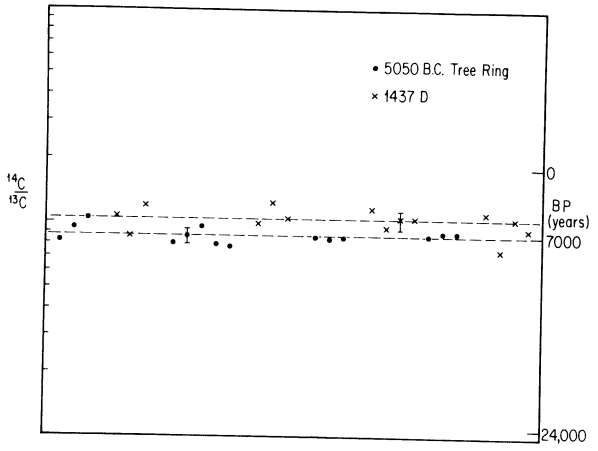

Fig 8. Successive measurements of $^{14}C/^{13}C$ ratios in Al_4C_3 targets made from 5050 BC tree rings (•) and from CO_2 from Sunnyvale bones (x). The dotted lines are averages of the two sets of points. The flags indicate the size of the statistical error for individual measurements. The right hand ordinate shows the time scale.

SUMMARY

We have presented results of initial tests of the General Ionex Corporation tandem accelerator facility which has been installed at the University of Arizona as part of a National Science Foundation Regional Facility for Radioisotope Dating. Some important results are: 1) at a terminal voltage of 1.8MV, the transmission of $^{12}C^-$ through the accelerator to $^{12}C^{+++}$ is 20%, sufficient to perform precise measurements of $^{14}C/^{13}C$ ratios; 2) to a precision of 0.5%, $^{14}C/^{13}C$ ratio measurements can be made in which the errors are predominately statistical. These measurements were made with irradiated graphite targets, and include all of the magnet cycling and target changing that are necessary to make relative age measurements; 3) experience with a large number of mixtures and compounds indicate that Al_4C_3 is a reasonable material from which to fabricate accelerator targets. Results were obtained from one such target which was fabricated from 100 μg of carbon; 4) with Al_4C_3 targets fabricated with CO_2 obtained from geophysical and archaeological samples, measurements of the ratio:

$$R = \frac{(^{14}C/^{13}C)_{sample}}{(^{14}C/^{13}C)_{standard}}$$

have been made with standard deviations of 3%. Again, these standard deviations were limited primarily by statistical uncertainties. It is our expectation that further experiments with more total counts collected will lower these uncertainties; 5) background measurements indicate that, at present, the real signal from a 40,000-year-old sample is equal to the background from a dead carbon sample. The "machine" background, obtained by looking for a signal from a blank aluminum target, is ca 60,000 years; 6) preliminary results of measurements on tree rings and on some archaeologic samples are presented.

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