

## ACCELERATOR MASS SPECTROMETRY OF $^{14}\text{C}$ AT THE AUSTRALIAN NATIONAL UNIVERSITY

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**ABSTRACT.** A  $^{14}\text{C}$  measurement capability has been developed on the 14UD accelerator at the Australian National University. At present, this system operates on a medium-precision, low-throughput basis with slow cycling between isotopes. We describe unusual features of the system, and review preliminary experience with this mode of operation, in sample preparation, and with a recently installed injection system.

### INTRODUCTION

The accelerator mass spectrometry (AMS) program based on the 14UD Pelletron accelerator at the Australian National University has been committed historically to the measurement of  $^{36}\text{Cl}$  in groundwater samples (Fifield *et al.* 1990, 1987), and  $^{36}\text{Cl}$  remains the main thrust of the program. More recently, a limited  $^{14}\text{C}$  AMS capability has been implemented to satisfy the need within the University for a small-sample capability to complement the existing decay-counting facilities of the Radiocarbon Laboratory. The system operates on a medium-precision, low-throughput basis with slow cycling between isotopes, and has some unusual features. Most notably, there is no electrostatic analyzer in the system, which makes it possible to transport both the  $^{14}\text{C}$  ions and the stable beams to the detector station.

During the first three months of 1991, we installed a new injection system on the accelerator. In parallel with this installation of the new injection system, we also studied graphitization procedures with a view to simplifying the procedure in order to permit a greater degree of batch processing.

### THE ANU $^{14}\text{C}$ SYSTEM

The ANU 14UD Pelletron accelerator is a shared facility. Basic research in nuclear physics accounts for 80% of the beam time, with AMS taking the remaining 20%. A consequence of the shared nature of the facility is that both financial and political constraints have been placed on developments specifically for AMS. For example, the single ion source is shared with the Nuclear Physics users, and the accelerator's analyzing magnet is not a multiport device and thus, cannot support a fast-cycling system. One consequence of these limitations is that any AMS must, at least at present, be performed in a slow-cycling mode.

### Features of the $^{14}\text{C}$ System

Negative carbon ions are produced in a General Ionex 832 ion source operated in reflected geometry. Typical graphite sample sizes are ~1 mg. The slow-cycling procedure consists typically of 30-sec measurements on each of the stable beams, followed by a 5-min counting interval for  $^{14}\text{C}$ . Unusual features of the system are:

1. There is no electrostatic analyzing element after acceleration. Nevertheless, the counting rates from charge-exchanged  $^{12}\text{C}$  and  $^{13}\text{C}$  ions are  $<100\text{ s}^{-1}$  due to the excellent vacuum of a few times  $10^{-8}$  Torr in the acceleration tubes of the 14UD accelerator. Such rates pose no difficulties to the ionization chamber, which detects the  $^{14}\text{C}$  ions.
2. The absence of an electrostatic analysis element makes it possible to transmit all three carbon beams to the detector station, where the stable beams are measured in a removable Faraday cup immediately in front of the detector. In order to accomplish this without changing all the magnetic elements of the post-acceleration beam transport, both the terminal voltage of the accelerator and the field of the injection magnet are changed while switching from one beam to another. Terminal voltages of 7.00, 6.46 and 6.00 MV are used for  $^{12}\text{C}$ ,  $^{13}\text{C}$  and  $^{14}\text{C}$ , respectively, ensuring that the magnetic rigidities of all three beams are identical after acceleration. Such a system avoids any concerns about variations in transmission of the  $^{14}\text{C}$  ions from the final Faraday cup to the detector. It is also worth noting that, if the fractionation introduced by the terminal stripper foil could be measured independently, it would be possible to measure the  $^{14}\text{C}/^{12}\text{C}$  ratio without reference to a standard ( $^{36}\text{Cl}$  measurements are already performed in just this way). At present, all measurements are made relative to the ANU sucrose standard, but the raw  $^{14}\text{C}/^{12}\text{C}$  ratios determined directly from the  $^{14}\text{C}$  count rate and the  $^{12}\text{C}$  beam current have proven to be useful indicators of system performance.
3. A fast beam chopper is used to attenuate the intense  $^{12}\text{C}$  beam by a factor of 80 before injection into the accelerator, thereby avoiding the heavy loading of the accelerator that would result from injecting the full beam of  $\sim 10\ \mu\text{A}$ . This chopper is switched off during the  $^{13}\text{C}$  and  $^{14}\text{C}$  phases of the measurement sequence.

### The Upgraded Injection System

Figure 1 shows the layout of the new injection system. The major improvement over the former system is the replacement of the old single-focusing injection magnet by a double-focusing magnet. This brings in its train much greater control over the beam optics *via* object and image slit assemblies and beam scanners immediately upstream of each set of slits. The image slits fulfill the additional function of intercepting the unwanted beam deflected by the beam chopper in the direction orthogonal to the bending plane of the magnet.

Focusing and steering have also been rearranged in the new system. A waist is produced at the object slits by a novel design of lens (Weisser 1991), which decelerates the beam before it enters the pre-acceleration tube, thereby increasing the strength of the lens effect at the entrance to the tube. A steering capability is also provided by splitting the electrode of this lens into four quadrants. Focusing and steering after the image slits to prepare the beam for injection into the accelerator is accomplished by an electrostatic quadrupole triplet, with six independent power supplies to permit steering and focusing.

### PRELIMINARY RESULTS

During 1989, before the installation of the upgraded injection system, 15 days of accelerator time were devoted to establishing a  $^{14}\text{C}$  capability and to performing measurements on a few samples of archaeological and geological interest. This experience showed the following:

1. We could obtain beam currents up to  $12\ \mu\text{A}$  from 1–3 mg graphite samples produced by either the iron/zinc or iron/hydrogen process (see the next section, below).

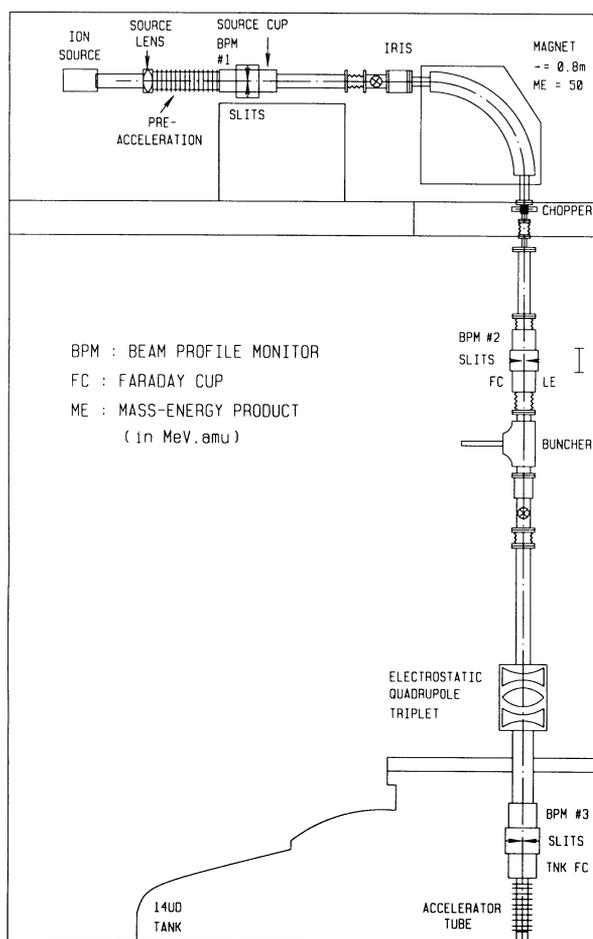


Fig. 1. The new injection system for the 14UD accelerator at the Australian National University

2. We could make measurements with a precision of  $\sim 4\%$ . The major factor limiting precision was the reproducibility of repeated measurements rather than counting statistics. We believe that this rather poor reproducibility arose from temporal variations in the transmission through the injection system and accelerator. Because  $\sim 75\%$  of the beam was lost between ion-source and the high-energy end of the accelerator, small changes in beam direction could lead to significant changes in transmission. Beam optics calculations showed that the single-focusing injection magnet seriously compromised the remaining beam transport, leading to poor transmission. Hence, the new injection system described above was installed. We made no  $^{14}\text{C}$  measurements during 1990.

Following the completion of the new injection system, a total of only two days of beam time were available to study  $^{14}\text{C}$ . We measured a series of standards to become familiar with the new system and to pinpoint any shortcomings. We observed a significant improvement in transmission over the old system in this first run. Transmission is still only  $\sim 50\%$ , however, and we suspect a misalignment of one of the elements. An advantage of the new system is that, with the beam scanners and remotely-controlled slits, we now have powerful tools to diagnose the problem, and we are actively pursuing further improvements in transmission.

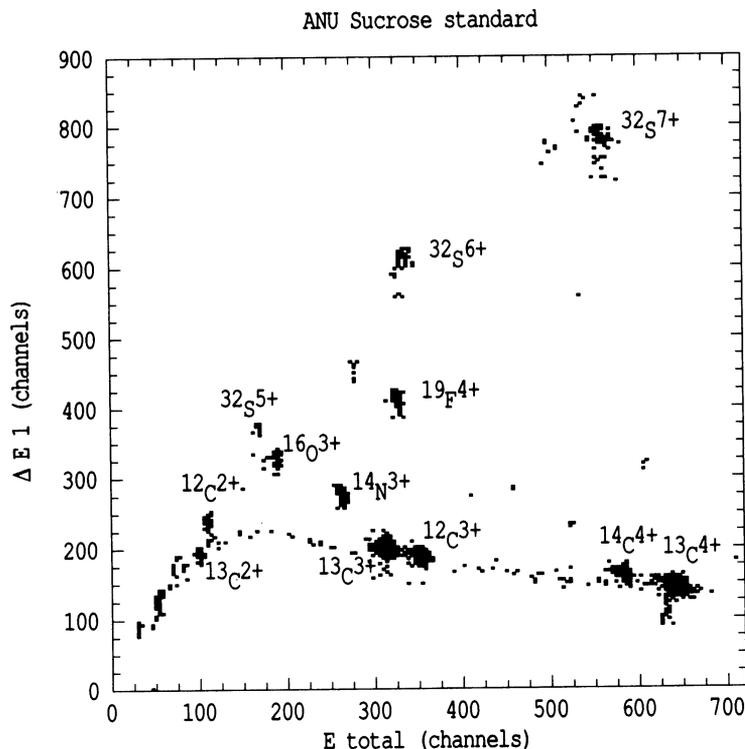


Fig. 2. Two-dimensional plot of energy loss vs. total energy from the ionization chamber, used to count the  $^{14}\text{C}$  ions. The energy loss signal is taken from near the front of the counter.

Despite the non-ideal conditions, typical  $^{14}\text{C}$  count rates from an ANU sucrose standard (151 pMC) were  $10\text{ s}^{-1}$  at  $^{12}\text{C}$  negative ion currents of  $6\ \mu\text{A}$ . Figure 2 shows a two-dimensional representation of data from the ionization chamber. The sample was a sucrose standard, and the total count rate of  $\sim 50\text{ s}^{-1}$  was due predominantly to the 3+ and 4+ carbon ions (remembering that there is no electrostatic analyzer in our system). The very much weaker groups of  $^{32}\text{S}$ ,  $^{19}\text{F}$ ,  $^{16}\text{O}$  and  $^{14}\text{N}$  are presumably due to  $\text{SF}_6$  and air in the residual gas in the accelerator tubes.

Preliminary indications from repeated measurements on the standards are that the reproducibility is already close to the 1% level, and we envisage that once the “teething” troubles are resolved, the system will be able to handle  $\sim 100$   $^{14}\text{C}$  samples per year with a precision between 1 and 2%.

#### DEVELOPMENTS IN SAMPLE PREPARATION

Two methods for the conversion of  $\text{CO}_2$  to graphite have been widely used. The first method (Slota *et al.* 1987) uses zinc to reduce  $\text{CO}_2$  to  $\text{CO}$ , followed by a catalytic reduction of  $\text{CO}$  to graphite on iron, whereas the second (Vogel *et al.* 1984) uses hydrogen gas in place of zinc. Of these two methods, only the former lends itself to a sealed tube “batch process” approach, because water does not have to be removed as the reaction proceeds. Consequently, we have taken some preliminary steps toward establishing the feasibility of such a batch process for the zinc/iron method.

As a first step, we placed the zinc and iron in separate 15-cm-long silica tubes attached to a small-volume manifold with its own pressure transducer. Following the standard procedure of first

purifying the iron and zinc by heating to 400°C in hydrogen, and then reducing the CO<sub>2</sub> with the zinc and iron at temperatures of 450°C and 600°C, respectively, we obtained very good graphite targets. However, both the time for starting and completing the reaction were long (typically 4–6 h and 12–18 h, respectively) and variable, presumably because of the distance between the iron and zinc.

The next step was to place both the zinc and the iron in a single horizontal tube, with the iron at the closed end and the zinc held in place half-way along the tube by silica wool. Care was taken to ensure a gap above the zinc. Small tube furnaces heated the zinc and iron independently. This arrangement proved to be markedly superior to the two-tube system. Reaction and starting times were much reduced to 4 h and 1 h, respectively. Figure 3 shows a typical pressure vs. time curve for the single-tube reaction.

In light of this experience, we are building a system that permits the simultaneous processing of several samples using a pair of block furnaces (one each for the zinc and iron). Although we are incorporating a pressure transducer for each sample into the design at this stage, we hope that, with experience, we should be able to go to a sealed-tube system, which will do away with the need for the pressure measurement.

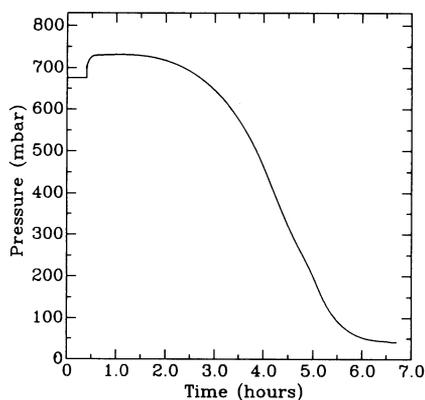


Fig. 3. Pressure as a function of time for a single-tube catalytic conversion of CO<sub>2</sub> to graphite using the zinc/iron method

## CONCLUSION

The establishment of a <sup>14</sup>C capability on the ANU 14UD Pelletron accelerator arose from the increasing need within the university for a small-sample capability to complement the decay-counting facilities of the Radiocarbon Laboratory. Such a capability is presently under development, and we anticipate that it will be fully operational by the end of 1991. On a shared accelerator such as this one, it would be unrealistic to expect to achieve the 0.5% precision reached by dedicated AMS laboratories, but the anticipated precision of 1–2% is adequate for many problems in both archaeology and the environmental sciences.

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