

## PROGRESS IN RADIOCARBON DATING WITH THE CHALK RIVER MP TANDEM ACCELERATOR

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**ABSTRACT.** The evolution of a tandem accelerator  $^{14}\text{C}$  dating system at Chalk River is recounted. Background problems and sources of instability are discussed and solutions are described. Details of sample chemistry and source preparation are presented.

### INTRODUCTION

Accelerator-based radioisotope dating is an important development for situations where sample sizes are limited and for isotopes of very long half-life. The technique can also be used for studying isotopic ratios of stable isotopes of possible interest in geochemical or other disciplines. Chalk River work in this field is motivated by its application to ground water dating in connection with nuclear waste disposal research and by a general interest in the use of nuclear physics techniques for problems in other scientific disciplines. The accelerator improvements required for accurate dating measurements will also be of benefit to the rest of the experimental physics program. There are major problems with the direct counting technique, however, that arise from the necessity of operating complex accelerators as quantitative instruments, a mode for which they have not been designed and for which little experience exists. Our goal is a dedicated reliable system for routine use that will produce accurate results with minimum expenditure of human and accelerator time.

Our first experiments were described at the Rochester Conference (Andrews and others, 1978). In those and subsequent measurements we cleanly detected  $^{14}\text{C}$  against a background of other ions, dated some samples of known age or  $^{14}\text{C}$  content, and established an initial system background level corresponding to an age limit of 45 to 50,000 years. In the course of this work we identified several instability problems limiting the accuracy of our results and we have spent most of our time trying to eliminate them. We have also successfully tested an automatic data acquisition and control system.

Figure 1 is a schematic diagram of the Chalk River MP Tandem complex. Most of our work has been done with the Q3D spectrometer (location 1, fig 1). Its remoteness from the accelerator poses stability and transmission problems caused by the high momentum selectivity of the line which are, as yet, unsolved. We, therefore, moved to a position on the  $70^\circ$  line (location 2, fig 1) that is much closer to the tandem. Here, the previous problems were resolved but unexpectedly serious backgrounds were encountered that render this position unacceptable. Our next attempt will be on the  $56^\circ$  line in target room 1 (location 3). This should combine the stability associated with the  $70^\circ$  location with the spectral purity of the Q3D data.

In this paper we shall describe our experiments at the Q3D and on the 70° line as well as those proposed on the 56° line. We shall also describe our sample preparation methods.

### Q3D experiments

The Q3D and subsequent experiments were done with 40MeV  $^{14}\text{C}$  ions produced by gas or foil stripping to charge state 4 at a terminal voltage of 8 MV. The apparatus used in the Q3D work was described at Rochester (Andrews and others, 1978). It consisted of a gas proportional  $\Delta E$  counter and a silicon surface barrier E counter located in the spectrometer focal plane as well as a start detector in the spectrometer target chamber to measure the time-of-flight (TOF) over a 7m path between this detector and the E focal plane detector. Typical spectra are shown in figure 2. It is seen that the Z of the ions is clearly defined by  $\Delta E$  and the mass numbers by the TOF signal. Similar mass identification comes from the E signal except that a low energy tail extends from the  $^{13}\text{C}$  peak across the position of mass 14. The  $^{12,13}\text{C}$  background peaks arise from ions injected when the source inflection magnet is set for mass 14 that have undergone charge exchange collisions with residual gas in the accelerator tubes and emerge with magnetic rigidity equal to that of the  $^{14}\text{C}$  particles. Since these  $^{12,13,14}\text{C}$  ions have equal rigidities, both their energies and velocities scale inversely with their masses. Thus, eg, the velocities and

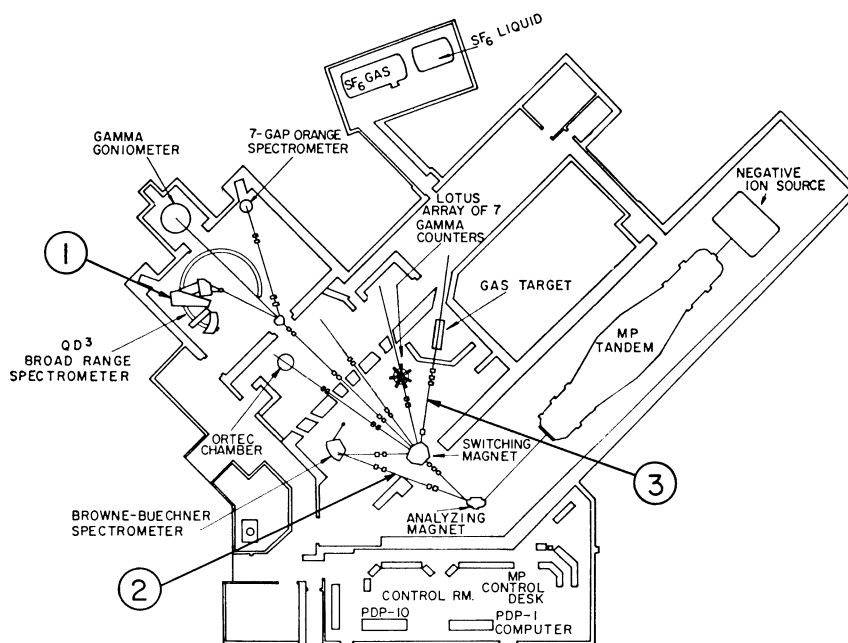


Fig 1. Layout of the Chalk River MP Tandem Complex with locations of  $^{14}\text{C}$  dating experiments indicated by number as follows: 1) Q3D spectrometer; 2) 70° line from the analyzing magnet; 3) 56° line from the switching magnet. Approximate distances to the three positions are 31, 9 and 15m, respectively.

energies of the  $^{14}\text{C}$  and  $^{13}\text{C}$  ions differ by about 7.7 percent. These background ions can be blocked by either an electrostatic bend (the electrostatic rigidity varies as  $E/q$ , energy/charge) or by a crossed electric and magnetic field velocity filter placed ahead of the particle detectors. For  $^{14}\text{C}$ , given the TOF information, these background ions pose no problem but for  $^{36}\text{Cl}$ , eg, the spurious  $^{35,37}\text{Cl}$  would be injected in much larger numbers so that they would have to be blocked to achieve a tolerable count rate in the detector system. We have chosen the velocity filter technique for our work at the  $70^\circ$  and  $56^\circ$  positions.

Since the Q3D is so far from the accelerator, efficient beam transport to it is a problem. Small changes in accelerator steering often result in large fluctuations in transport efficiency. Another problem at this location is that the energy acceptance at the Q3D is only 1 to 2KV on the accelerator terminal. During  $^{14}\text{C}$  counting there is no beam for energy stabilization and thus terminal stability (about 1 in 8000) must be maintained by the generating voltmeter (GVM), an electromechanical device that senses the electric field generated by the accelerator high voltage terminal. A con-

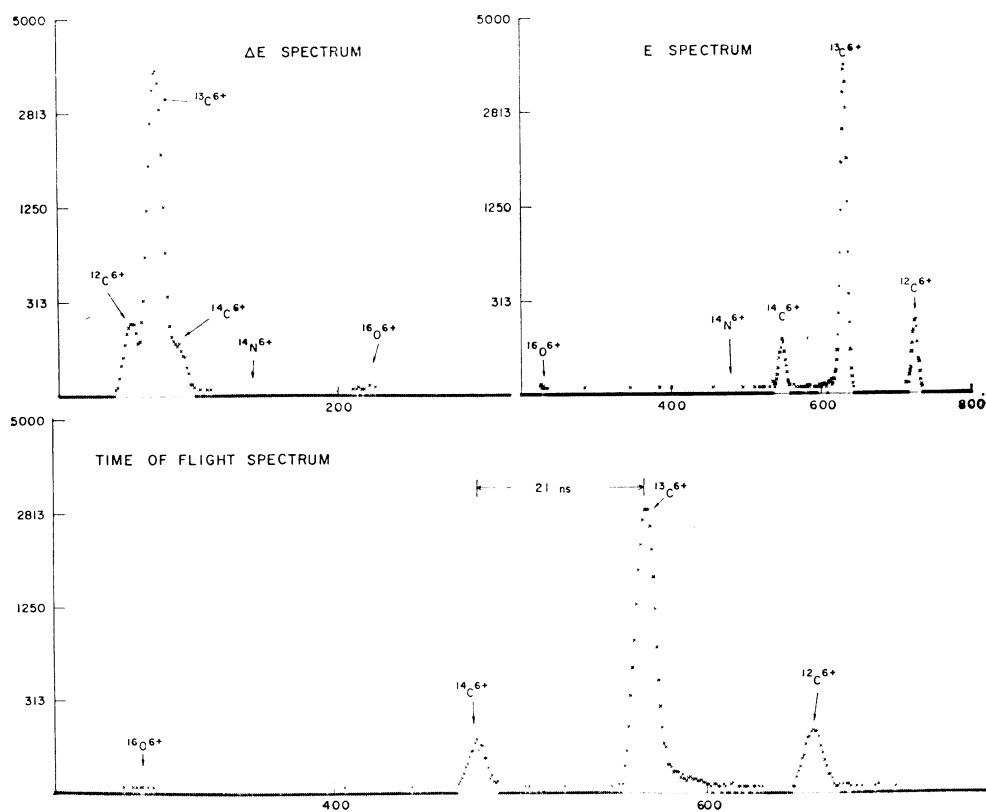


Fig 2. Typical  $\Delta E$ , residual energy,  $E$ , and time-of-flight spectra from a  $^{14}\text{C}$  experiment at the Q3D. Spectra are plotted on a square root ordinate scale.

centrated effort has thus far failed to reduce the GVM fluctuation to a level significantly below the energy acceptance of the spectrometer. This residual noise in the GVM is not yet understood; it may be an intrinsic property of the device, due perhaps to varying surface charges on the moving parts. Because of these difficulties it seemed prudent to make the experiment less sensitive to accelerator voltage and beam position fluctuations.

### 70° line experiment

The 70° line passing from the analyzing magnet into target room 1 is one of our few locations with beam optics suitable for a velocity filter. Guided by the very clean spectra seen at the Q3D, we set up a very simple system: a velocity filter at the exit of the analyzing magnet acting vertically and a silicon surface barrier E counter just behind the image cup. We later added a  $\Delta E$  gas proportional counter in front of the E detector. A more than adequate energy acceptance corresponding to 30 to 40KV on the accelerator terminal was achieved by opening the horizontal image slits just ahead of the E counter.

Most of the tests at this location were done with a reactor irradiated graphite sample that produced  $^{14}\text{C}$  count rates of 20-30 per second. This high rate facilitates tuning and in tests of stability and reproducibility it removes counting statistics as a significant source of error. The effectiveness of the velocity filter is illustrated in figure 3, which shows data from the irradiated sample with the velocity filter on and off. There is essentially perfect suppression of the spurious peaks of  $^{12}\text{C}$ ,  $^{13}\text{C}$ ,  $^{16}\text{O}$ , etc. The region around the  $^{14}\text{C}$  peak is shown in more detail in figure 4 for data taken with the irradiated graphite and with carbon prepared from NBS standard oxalic acid (assumed 105 percent of modern  $^{14}\text{C}$  concentration). These data were taken with a  $2\text{mg cm}^{-2}$  mylar foil in front of the E counter that provided  $\Delta E$  information for particle identification. The

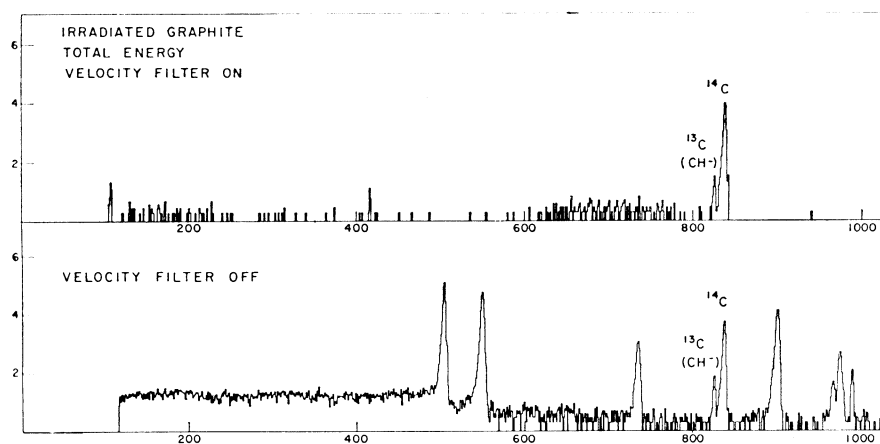


Fig 3. Spectra taken with the velocity on and off at the 70° position with a single silicon E counter.

higher energy peak is primarily  $^{14}\text{C}$ , the other is  $^{13}\text{C}$  generated by  $^{13}\text{C}$  injected as  $^{13}\text{CH}^-$  that travelled through the accelerator as a normal beam in the same charge states as the  $^{14}\text{C}$ . The final energy is slightly lower because a molecular ion is injected rather than a simple ion as for  $^{14}\text{C}$ . This macroscopic beam reaches the analyzing magnet with the wrong rigidity but a few ions suffer charge-changing collisions with the residual gas in the magnet box at just the right place to emerge in the direction of the detector. Similarly, under the  $^{14}\text{C}$  peak, there could be a contribution from  $^{13}\text{C}$  injected with mass 14 because of the relatively poor selectivity of the inflection magnet. This form of spectral contamination is particularly insidious because the ions, of approximately equal energy, have velocities varying inversely as the square root of the mass so that the  $^{13}\text{C}$  and  $^{14}\text{C}$  velocities differ by only 3.8 percent rather than 7.7 percent as for the  $^{13}\text{C}$  and  $^{14}\text{C}$  ions with the same rigidity in the previous case. The velocity filter is, therefore, much less efficient as shown by the strong presence of the contaminated peak in figure 3 with the velocity filter on. Furthermore, since the velocities of the  $^{13}\text{C}$  and  $^{14}\text{C}$  ions are so similar, the rates of energy loss differ by less than 5 percent (*cf* figure 5) which is comparable to the 5 to 7 percent resolution attainable with these  $\Delta E$  counters.

Judging from data presented at Rochester by the McMaster and Oxford groups (Nelson and others, 1978; Barratt and others, 1978), this type of background has been seen previously. However, the earlier data were taken with much narrower slit settings so that the peaks were not

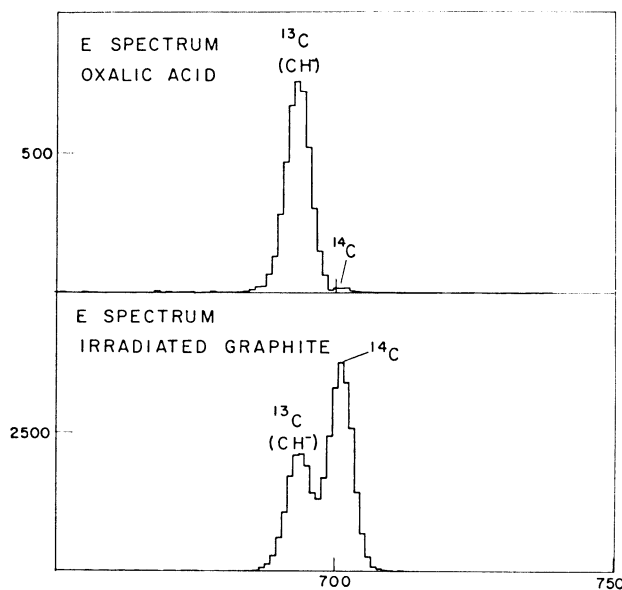


Fig 4. Spectra from irradiated graphite and NBS oxalic acid samples showing the vicinity of the E peak in more detail. Velocity filter is on and a  $2\text{mg cm}^{-2}$  mylar foil is in front of the E counter. The  $^{13}\text{C}$  background peak arises from charge exchange in the magnet box as described in the text.

as prominent. Fortunately, these spurious ions can easily be removed by a second magnetic or electrostatic analysis between the analyzing magnet and the detector system. It should be noted that electrostatic analysis ahead of the analyzing magnet is useless in this respect since it passes all ions which have the same  $E/q$  upon emergence from the accelerator. It is, therefore, important in small facilities to ensure that additional electric or magnetic analysis follows the analyzing magnet.

*Future experiments*

Our next attempt will be on the  $56^\circ$  line in target room 1 just after the switching magnet. The latter provides the second magnetic analysis required to remove the backgrounds found at the  $70^\circ$  position. A velocity filter will be placed just after the second magnet and our  $\Delta E$  gas proportional, silicon E counter system will be used. It is expected that the energy acceptance and transmission will be good and that the spectra will be clean. A major difference will be operation at constant terminal voltage rather than at constant magnetic rigidity. This should permit simultaneous optimization of  $^{12,14}\text{C}$  transmission through the accelerator and result in better stability in their ratio. Cycling of the beam transport system is feasible for this location close to the tandem; it was not thought to be so for the Q3D experiments.

After we have achieved an acceptable level of stability and reproducibility we shall return to measuring samples of known and then unknown

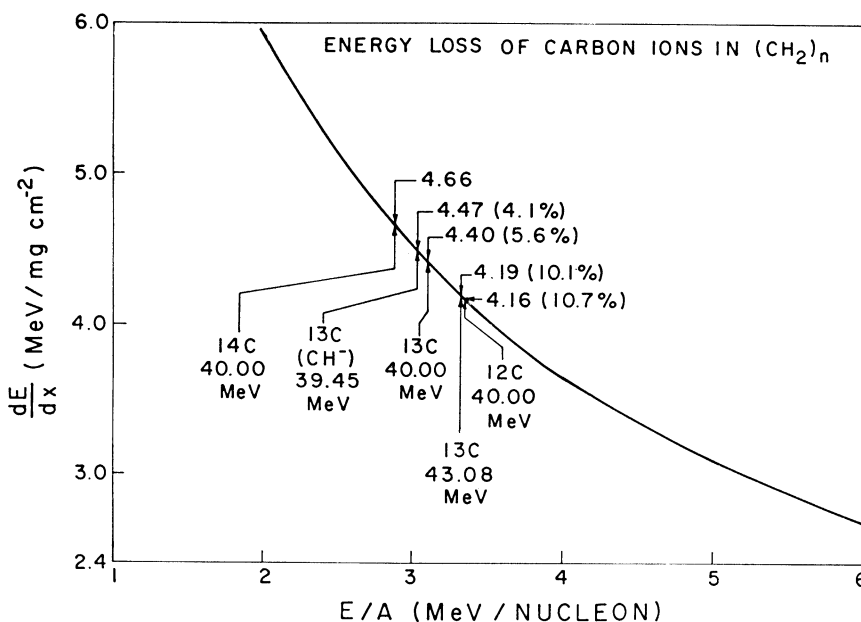


Fig 5. Energy loss of carbon ions in hydrocarbons vs specific energy.  $\Delta E$  values for various background groups are shown. The numbers in parentheses are the percent difference in  $\Delta E$  relative to 40MeV <sup>14</sup>C.

age. Exploratory experiments will also be carried out for other isotopes, particularly  $^{36}\text{Cl}$ , which is of interest in hydrology.

#### Sample preparation

With the exception of graphite, our samples were in the form of carbon black prepared by reduction of  $\text{CO}_2$  by hot magnesium metal. The small size of sample involved greatly facilitates what was a tedious, time-consuming procedure when used by Anderson, Arnold, and Libby (1951) for preparing 20g samples for the original screen wall solid carbon counter.

For a 25mg carbon preparation, a small evacuated stainless steel furnace (1.25cm ID, 10cm long) containing 110mg Mg turnings (10 percent excess) is heated to a dull red heat with a hand torch and the sample  $\text{CO}_2$  admitted. Reaction is complete within 3 to 4 minutes, with no residual gas remaining. The furnace is finally heated to a bright red heat to ensure conversion of any magnesium carbides to elemental carbon. After cooling, the solid residue is transferred to a 40ml centrifuge tube, washed free of Mg and MgO with hot HCl and water and air dried. Adsorption of radioactivity from the atmosphere or washwaters is of no concern since no radioactivity measurements are involved and adsorbed atmospheric  $\text{CO}_2$  will be desorbed during initial pump-down of the hot, high vacuum ion source.

Samples from 5 to 100mg were prepared by this procedure with carbon recoveries of 85 to 90 percent. Low yields were experienced when a greater excess of Mg was used, evidently due to difficulty in converting the carbides to carbon.  $^{13}\text{C}$  measurements indicate that isotopic fractionation at the yields normally realized is not significant. Total preparation time is about 2 hours.

For sample mounting the carbon is moistened with just enough water to make it sticky (about 10mg  $\text{H}_2\text{O}$  on 5mg carbon), pressed manually

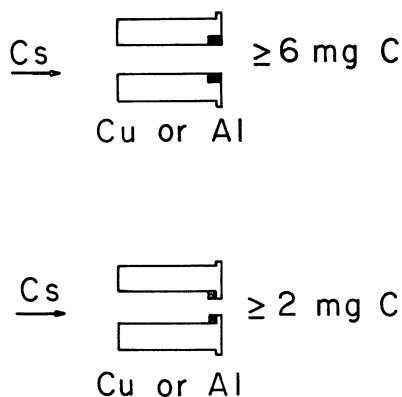


Fig 6. Two sputter source sample geometries. At the top, carbon is sputtered by reflected Cs ions as well as those passing through the central hole. Numbers are the smallest sample sizes used to the present in each geometry.

into Cu or Al sample holders for the sputter ion source and dried under a heat lamp. Two mounts, shown in figure 6, have been used. The one with the sample inside the Cs<sup>+</sup> beam tube provides better protection from sputtering of material from adjacent mounts. Samples down to 2mg may be mounted in this holder. Our source mounts will probably change in the course of development work to improve our source output.

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