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DIRECT DETECTION OF ^{14}C AT THE HARWELL TANDEM

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ABSTRACT. Direct detection of ^{14}C using the Harwell 6MV Tandem accelerator has been achieved using a gaseous ion source. The implications of our results for the use of such sources in this role and for the machine, in general, are discussed.

INTRODUCTION

A great deal of interest has recently been aroused by the possibility of using various heavy ion accelerators for trace element analysis. ^{14}C detection is initially of particular interest because of its important role in the dating of artifacts (Gove, 1978).

The use of the Harwell 6MV Tandem Van de Graaff accelerator as a trace element analysis facility is currently being evaluated. This progress report will concern itself with our experiences in ^{14}C dating. The ion sources used on the tandem are all of the gaseous variety. Although the most favored source for the trace element analysis facility is the Middleton type of negative-ion sputter source, it was decided to proceed with the project using the existing gas source pending the arrival of a sputter source. Furthermore, it was felt that a gas source, although inferior to a sputter source in brightness and memory effect, provided sufficient advantage to justify further investigation. Of particular interest is that possible feed gases such as carbon dioxide and methane are easily derived from archaeologic samples by well understood and easily controlled processes (Burke and Menschein, 1955), whereas the problem of the preparation of samples for sputter sources has yet to be satisfactorily solved.

To date, ^{14}C accelerated from this ion source has been detected with a suitable beam line detection system consisting of a Wien filter and an E-ΔE ionization chamber as shown in figure 1.

Ion sources

The Harwell Dawton-type carbon ion source (Dawton, 1963) is shown in figure 2. It consists of a mercury pool cathode arc discharge source with a gas charge exchange canal. It is not especially useful for ^{14}C dating in its present state because it produces beams of only $\frac{1}{2}$ μA of ^{12}C. Nevertheless, it is sufficient to determine whether further development of this type of gas source is worthwhile.

Carbon dioxide was used in the source initially but the resultant carbon beams were unreliable; methane was found subsequently to give more consistent results. The gas charge exchange canal was fed with specially prepared 'dead' methane containing no ^{14}C. The carbon beam has a magnetic rigidity consistent with the process $CH_4^+ \rightarrow C^-$ at the charge exchange canal (Dawton, 1973, pers commun). In addition, large beams of oxygen were produced by the source at inflector magnet settings near those for C^-. While gaining experience in the use of the ion source

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for carbon beams the nature of the beam was diagnosed by using a scatter foil above the analyzer magnet to look at the equilibrium charge state distribution of the ions from the foils at the center terminal as previously described by Freeman and Hooton (1973). Because the equilibrium charge state distribution of ^{12}C can be easily distinguished from that of ^{16}O, the correct settings for the ion source and inflector magnet could be confirmed.

An important property of any ion source used for dating is that there should be negligible memory of the previously dated sample during a dating run. The measured memory effect of the ion source described, above, is of the order of 10 percent. This is the result of a direct ^{14}C measurement, first with a sample of CH_4 enriched in ^{14}C to 300 times modern level, and then, a sample of modern CH_4. By contrast, a bench test of a duoplasmatron source having the same volume showed a much larger memory effect when run on carbon dioxide for many hours and then on ^{20}Ne. The residual ^{12}C beam was often larger than when using CO_2. The carbon beam lasted for many weeks and finally necessitated a complete overhaul and cleaning of the source head, whereupon it was

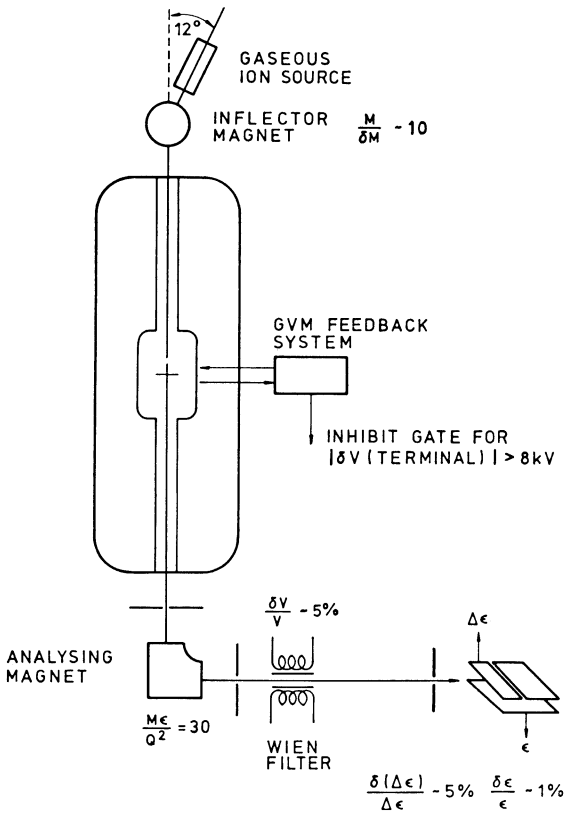

Fig 1. Schematic diagram of the Harwell Tandem showing relevant features.

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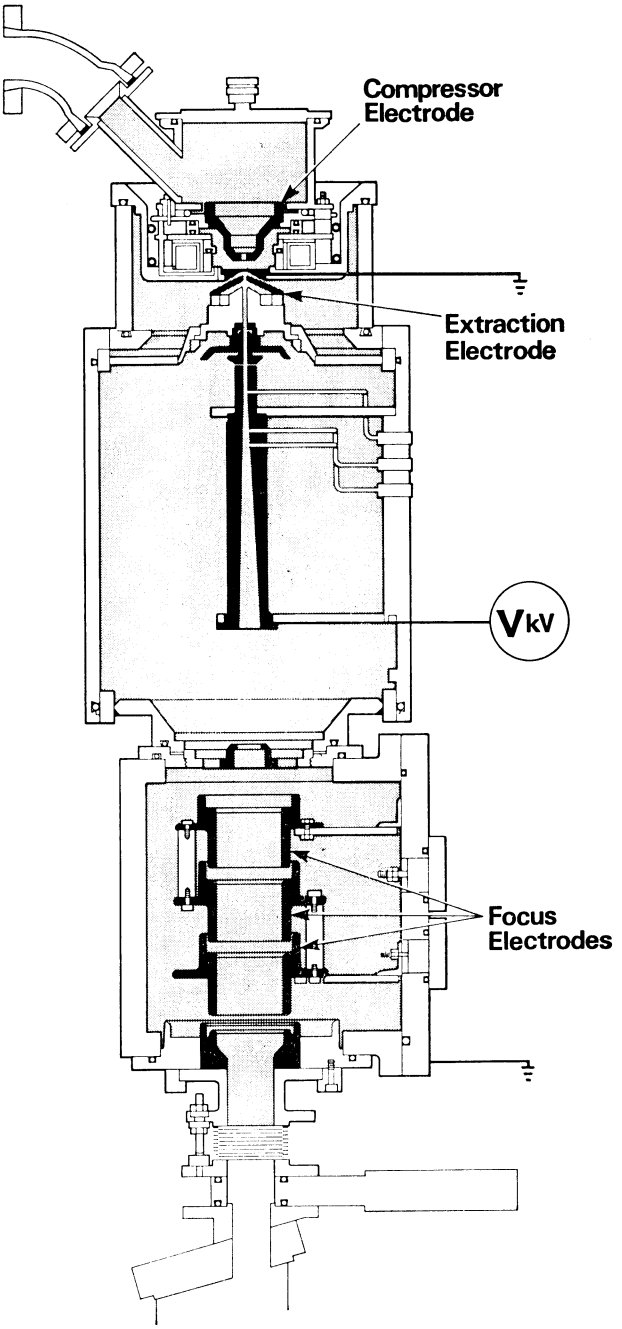

Fig 2. Mercury pool cathode arc discharge source used for carbon beams, showing complete electrode structure with the gaseous charge exchange canal directly beneath. The canal voltage V was chosen to be 25kV.

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found that not only was the inside wall of the source covered with sooty deposits but also the extract electrode contained small chips of solid carbon. A similar test with a PIG source for a cyclotron showed a much smaller memory effect of about 1 part in 2000 of the original ^{12}C beam (Jones, 1978, pers commun). This latter source has a small volume of about 5ml and a simple two-electrode structure. Hence, apart from possible effects of source temperature and feed gas, compactness and simplicity of gaseous ion sources are necessary properties if they are to be suitable for further development in this role.

Center terminal stabilization

The standard slit stabilization technique cannot be applied to maintain center terminal stability when low intensity trace element beams are inflected into the machine. A feedback system based on comparison of a Generating Voltmeter output with a reference voltage was used instead. Furthermore, it was noticed in the early stages of this project that a substantial amount of background in the ionization chamber arose from large terminal instabilities and sparks which introduced large spurious beams into the system for short intervals. This type of event was eliminated by generating a veto level whenever the terminal voltage altered by more than ± 8kV. This level then inhibited the data collection electronics whenever a terminal instability occurred.

Beam line apparatus

The ion source inflector magnet has a mass resolution $M/\Delta M$ of 10 and the important beam selection process is achieved with the analyzer magnet at the exit of the machine. Most of the beams observed when the analyzer is set for ^{14}C are ion source-related and arise by various processes. Some of these are the product of charge exchange processes in the machine tubes as discussed by Purser and others (1977). Not all of the beams have the same velocity as ^{14}C and may be eliminated by using a crossed field type of velocity filter. The ^{14}N beam, however, has the same velocity as ^{14}C and discrimination between them may be achieved with an E-ΔE ionization chamber.

The crossed field filter consists of a 15cm long parallel plate capacitor between the poles of an electromagnet. Beams not fulfilling the transmission condition are deviated from the beam axis and resolution is achieved by a defining slit set at a suitable distance from the crossed field region. In practice, it was found that about 20kV across the plates and 250 gauss magnetic fields were sufficient for about 5 percent velocity resolution.

The gas counter is set up in the beam path and consists of a split anode, two grid design described by Erskine, Braid, and Stolzfus (1976) using isobutane as the filler gas. This device obtains a ΔE signal from the anode and a total energy signal from the cathode. It has a 1.75μm thick Makrofol window foil supported on a 90 percent transmission hexagonal grid of Cu/Be. The window grid is biased to −50V to ensure more complete charge collection, thus improving ΔE pulse height by about 10 percent. The typical operating pressure of the counter was 80 to 90torr, the energy resolution 1 percent, nuclear charge resolution 1/20.

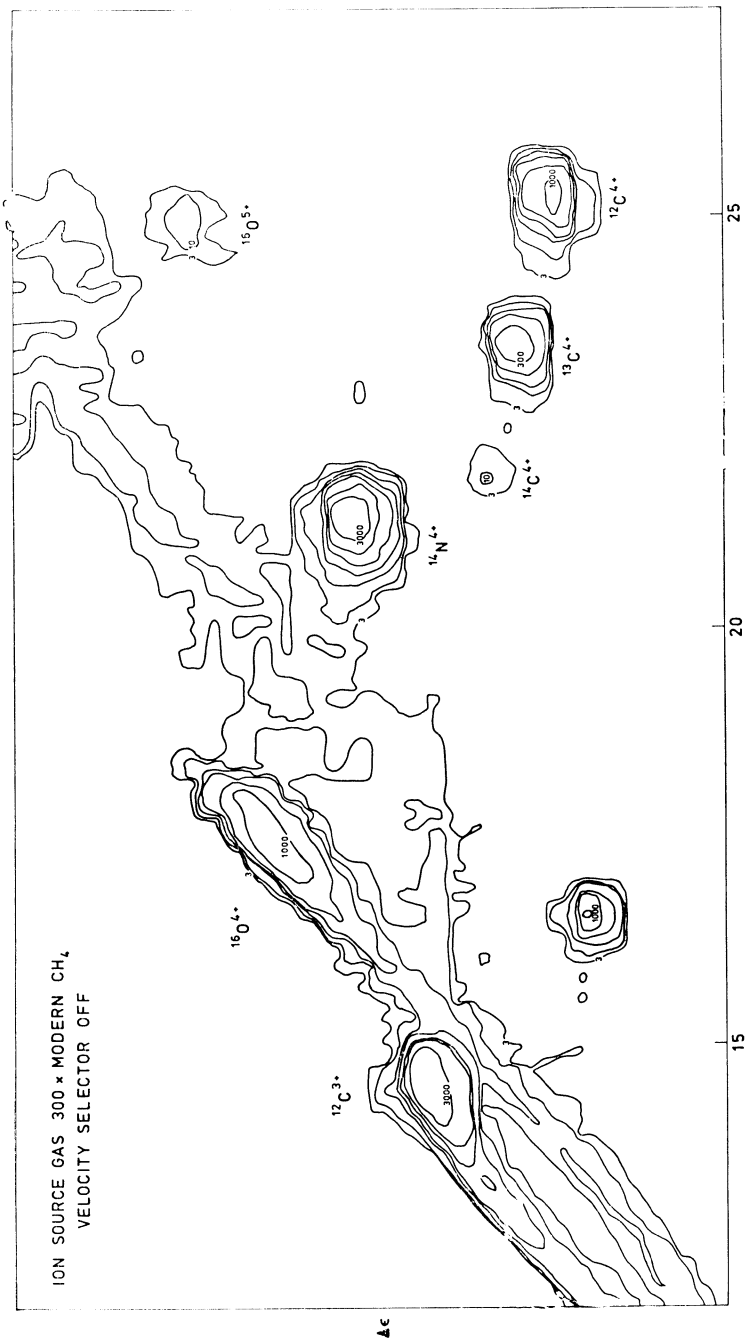

Fig 3. ϵ-$\Delta\epsilon$ spectrum with machine set to $^{14}C^{4+}$ at 22.5MeV. The velocity filter was not switched on and the beam contaminants feature strongly.

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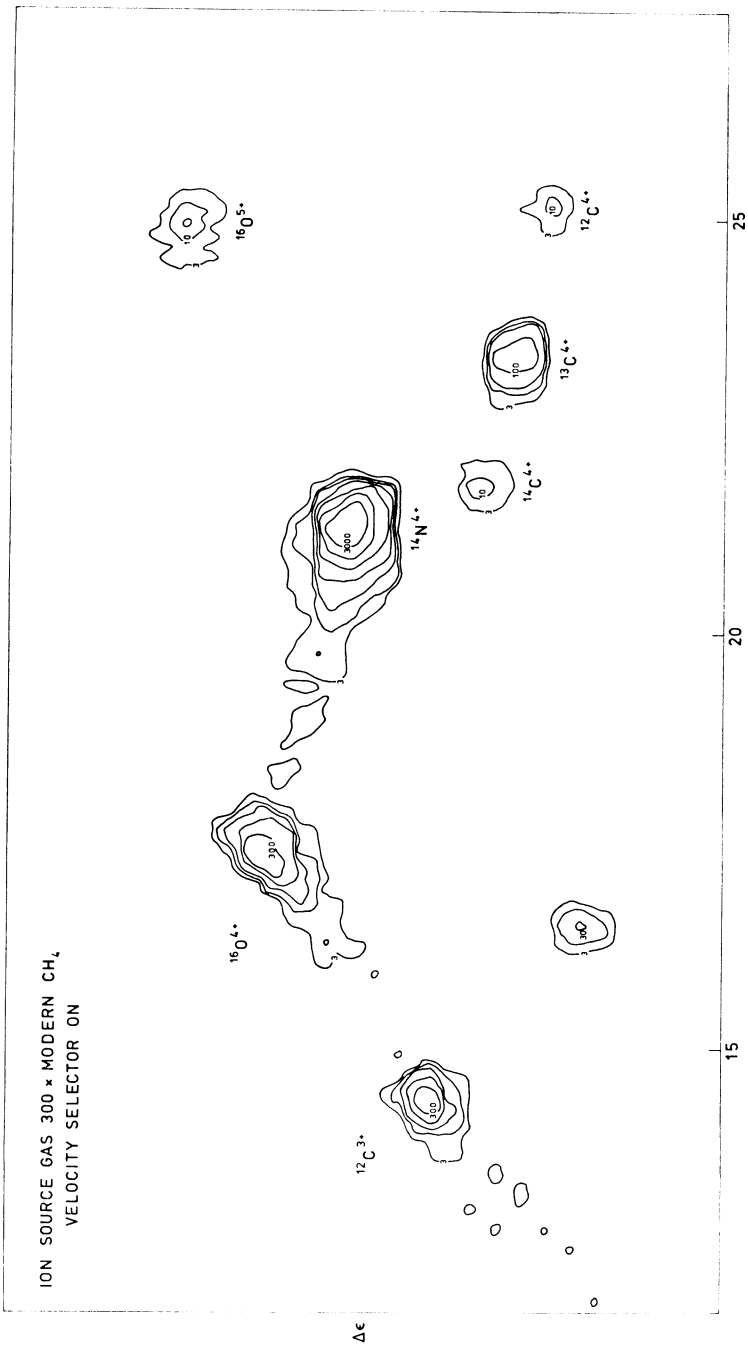

Fig 4. ϵ-$\Delta\epsilon$ spectrum with the same machine setting but with velocity selection. The spectrum is noticeably cleaner.

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RESULTS

The E and ΔE signals were collected in a two-dimensional multichannel analyzer gated with the terminal voltage excursion level. The source gas used was methane enriched 300 times modern to ensure reasonably high counting rates. Figures 3 and 4 show spectra taken with the ion source inflector and machine analyzer set for ^{14}C. The effect of velocity selection is quite marked. In the vicinity of the ^{14}C peak are the ^{13}C and ^{12}C peaks selected out of the energy continua in the machine tubes by the analyzer magnet. At higher energy there is a yield of ^{14}N, unattenuated by the velocity filter. This ^{14}N yield appears to be generated by the process $^{14}NH^- \rightarrow {}^{14}N^{4+}$ at the center terminal stripper since the ^{14}N beam maximizes at a terminal voltage higher by 1/75 of its setting for ^{14}C transmission. The analyzer therefore appears to be selecting the tail of $^{14}N^{4+}$ generated by the above process. Similar observations have already been made by Nelson, Korteling, and Stott (1978).

CONCLUSION

^{14}C has been counted directly on the Harwell Tandem using a gaseous ion source. Experience with gas sources has demonstrated that memory effect is a major disadvantage that makes them unsuitable for dating.

In the course of our experiments we have found that the ^{12}C transmission of the Tandem is 15 percent; lower than expected (Goldie and Trump, 1974). Poor source emittance and matching optics combined with a relatively poor vacuum is believed to cause this. Steps to overcome these problems are planned.

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DISCUSSION

Damon: If you said that the memory effect is 0.1 percent in a PIG source, this is not a severe limitation for some purposes. What does provide the limitation for using a ^{14}C gas source in a tandem accelerator?

Shea: There is perhaps a slight confusion about our ion source experiments. The PIG source has not a negative ion source. Its memory is certainly not a severe limitation but a lot of development would be necessary to convert to negative ion operation. The primary limitations associated with the existing tandem ion source are the 10 percent memory effect and the poor carbon currents generated.

Klein: In your experiments with the PIG source, did you measure the carbon currents from the source when gases containing no carbon (such as O_2 or Ne) were being used? Such currents might result from carbon absorbed in, or contained by the electrodes.

Shea: The PIG electrodes were actually of tantalum but the point is well taken. We did not investigate the source of the residual carbon beam, but assumed it was a memory.

Purser: Could you point out why the $^{14}N^{4+}$ peak is so much more intense than the $^{14}C^{4+}$ peak, even though the ^{14}C is enriched by a factor of 300 above recent? Do you think N^- ions are produced from the PIG source?

Shea: The $^{14}N^{4+}$ peak is consistent with the acceleration of NH^- to the center terminal and with stripping to N^{4+}. Elevating the terminal voltage by 1/75 very much increases the nitrogen yield as may be expected on this hypothesis. We did not investigate ^{14}N generation from the PIG source. Our main concern was to estimate the memory effect. The PIG was a positive ion source and so N^- production could not be measured.