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THE OXFORD RADIOCARBON ACCELERATOR FACILITY

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The Oxford ^{14}C accelerator has operated with beam for some 400 hours. This report describes the progress made towards achieving dates from milligram samples with the required accuracy of better than 2%. In summary, it shows how ^{14}C is relatively easily detected, but that the overall beam optical system is, at present, rather sensitive to effects which prevent reliable maintenance of the necessary isotope ratio stability. These effects can probably be eliminated by careful attention to details of the design rather than by major modifications.

GENERAL DESCRIPTION OF BEAM TRANSPORT SYSTEM. Figure 1 shows a general plan of the beam transport system. ^{12}C and ^{13}C are both measured after acceleration of pulses of beam (pulse length 0.1msec to 10msec for ^{12}C, 1msec to 100msec for ^{13}C, mark space ratio of 2%), and collection in Faraday cups in the focal plane of the analyzer magnet, which is set to bend the $^{13}C^{3+}$ beam 90°. ^{14}C is measured by counting each ion as it is identified from two energy loss (dE/dX) measurements and a total energy measurement from a gas counter fitted with a half-micron polypropylene window. As ^{14}C is only 0.1 to 1% of the ion beam leaving the analyzer magnet at 90°, excess ions, which depress the count rate of the detector, are removed with a Wien (velocity) filter. The velocity resolution (half-width) of the filter is variable, to a maximum of 2%. The mass resolution of the analyzer magnet is 0.05%.

The accelerator is a '3MV' Tandetron, capable of operating up to 2.4MV. The maximum voltage for reliable operation appears to be ca 2.1MV at present, but this should improve. Ideally, the terminal voltage would be chosen to optimize production of C^{3+}, and possibly to ensure that isotopic fractionation in the stripping process is at its most reproducible. The terminal voltage stability is controlled solely by a GVM feedback loop, and at 2MV, is within ± 300V, with drifts of up to 1000V over several hours. Negative ion beams are stripped by carbon foils, or, more usually, by O_2 gas leaked into the stripper canal. Optimum stripping is not realized, since all leaked gas is pumped through the accelerating tubes. However, the overall beam transmission is quite satisfactory.

Selected masses are injected with a 90°, 450mm radius double focusing magnet with a mass resolution (FWHM) of 1%. $^{12}C^-$, $^{13}C^-$ ions are injected by electrostatically accelerating the beams through the magnet, using gap lenses at the magnet

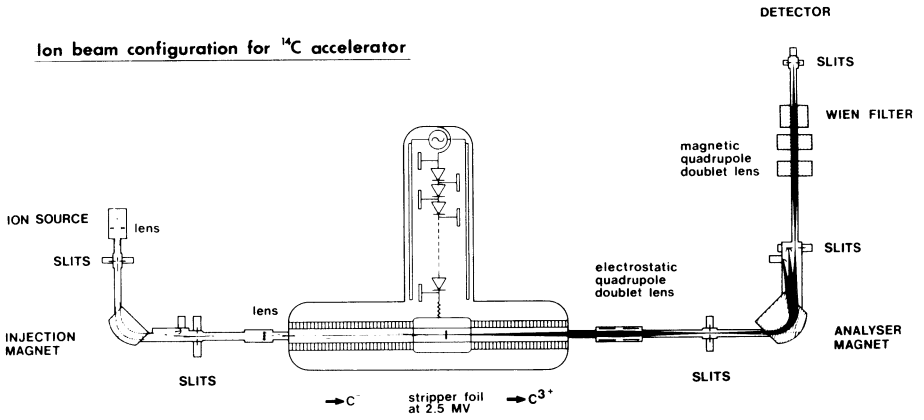


Fig 1. Beam transport system for ^{14}C accelerator (to scale)

waists. The electrostatic field is pulsed according to the schedule described above. All beam steering elements in the complete system are energy-dependent, rather than momentum-dependent. C^- beams are produced from graphite, using one or other of two Cs sputter sources, which are described below.

ION SOURCES AND SAMPLE TARGETS. Both ion sources were described previously (Hedges, Wand, and White, 1980; White, in press). The reflected source is more complex in operation and performance. It can accept 20 samples of graphite deposited on Ta wires. The 5mm length of graphite is scanned repetitively past the Cs^+ sputtering beam. Figure 2 shows the essentials of the arrangement.

In the alternative source (IS3; see figure 2) a single sample is accommodated. The operation of the source is more straightforward, but is limited by unsatisfactory fabrication procedures for the annular spherical ionizer. The source works best when the sample of graphite is crushed from the original Ta wire on which it was deposited and pressed into a hole of 1mm diameter in the center of an expendable Al holder. We have also operated (for a short time) the reflected beam source on graphite samples similarly crushed and pressed into holes of 1.2mm diameter. The resulting currents are 1/2 to 2/3 the current produced from wires; the sample is probably consumed more efficiently. In principle, it should be possible to design the IS3 source to take multiple samples. We believe that this source offers the best long-term arrangement for ^{14}C dating, but will continue to work both in parallel. Both sources have similar characteristics in performance, which also extends to finer details of isotopic ratio stability, and background ^{14}C

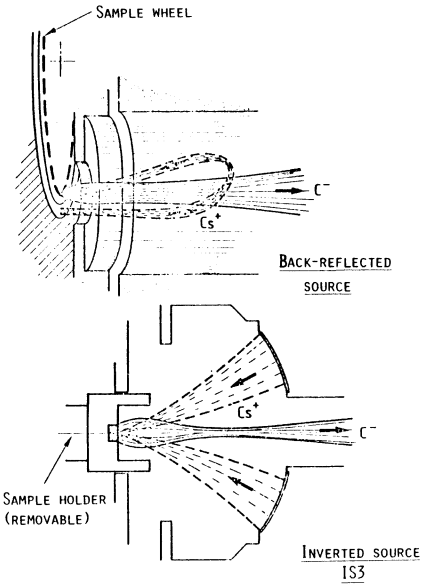


Fig 2. Schematics of beam arrangements in the back reflected and inverted sputter sources, showing sample mounting

contamination. Typical beam currents are in the 1 to 10μA region, using samples of ca 2mg of deposited graphite. When given full attention, both sources are capable of producing up to 5 times this beam current however.

BEAM TRANSPORT. The current injected as mass 14 (mainly $^{13}CH^-$) is in the region of 1nA or less. Up to 5μA ($^{12}C^-$) at least, there is no change in overall transmission with current, although there is a significant increase in radiation at 2MV. The beam waist sizes, measured at slits, and the optical field strengths for optimum transmission, agree well with the original beam optics calculations.

The particle transmission through the accelerator at 2MV is ca 60 to 65%. This could be improved to perhaps 70 to 75% with a terminal gas stripper pump. Foil stripping at this voltage is significantly less efficient, due to greater scattering. In all cases, transmission should improve at higher voltage. The yield of C^{3+} is ca 30 to 35%, on a particle basis, from gas stripping. It is unlikely that equilibrium stripping can be obtained in our system at present, and the expected value is 42%. Transmission around the analyzer magnet and, for the 90° (ie, ^{14}C) beam, to the detector, is 95 ± 5%. For the smallest aperture (1.5mm diameter) before the detector, this is reduced to 80 to 85%, probably as a result of terminal voltage fluctuations (see below).

The stability of the terminal voltage can be monitored by measuring the position of the $^{12}C^{3+}$ beam inside the 97° Faraday cup. This can be measured to within 0.1mm, corresponding to a

value of ± 25V on the terminal. There is an additional ripple at 50Hz and 300Hz with a maximum amplitude of 400V on the terminal, and it is necessary to synchronize the phase of the short $^{12}C^-$ beam pulse with the phase of the mains supply in order to eliminate a major source of noise in the measurement. Reduction of this ripple on the terminal can only be achieved with a fast-response feedback loop using an error signal derived from the Tandetron power supply. However, the ripple, while somewhat reducing transmission under conditions of highest resolution, should not lead to long-term instabilities. Monitoring of the beam position shows that the terminal is subject to random drifts typically in the range of 200V over 10 minutes, although periods of much higher stability can occur. The maximum excursion is probably ca 400V, although long-term drifts, probably due to thermal changes in the GVM and its circuitry, can lead to drifts of up to 1KV. These changes are important in the context of the stability of the $^{14}C/^{12}C$ ratio measurements.

DETECTION OF ^{14}C. With the beam transport system properly aligned, >95% of the particles reaching the detector, even with a large final aperture of 4mm, are ^{14}C ions, using a sample of oxalic acid standard (see fig 3). With the Wien filter turned off, the dominant ion is $^{13}C^{3+}$, with the same magnetic rigidity as the detected ^{14}C, and therefore, less energy. Some oxygen is also observed. There can be a small background spectrum from particles originating in the accelerating tubes during conditioning behavior, ie, irrespective of beam. Nitrogen is only observed when the injector magnet is tuned to inject mass 15 (NH^-), when the flux is maximized for a rigidity corresponding to the breakup of NH. There is no difference in observed background between foil stripping and gas stripping. (O_2 is used in preference to Ar simply because it is pumped better by our ion pumps.)

DETECTION OF ^{14}C FROM 'DEAD' SAMPLES. Although we have not done much work on the reduction of ^{14}C backgrounds, preliminary results give a ^{14}C signal equivalent to ca 60,000 years BP. This signal is undoubtedly due to ^{14}C nuclei originating from the ion source, and very probably from the region of sputtering itself. If the Cs^+ beam is interrupted before striking the target, no ^{14}C atoms are detected (age >80,000 years). Bare Ta and Pt wires have given ages between these two values. Since some C^- is detected (generally <1nA and falling with time), the ^{14}C ratio corresponds to fairly 'old' carbon. Interestingly, the oil from the turbo-pump has an equivalent age of 11,000 years. Much more work is needed to locate the sources of contamination, but it is gratifying that there is no significant cross-contamination between samples.

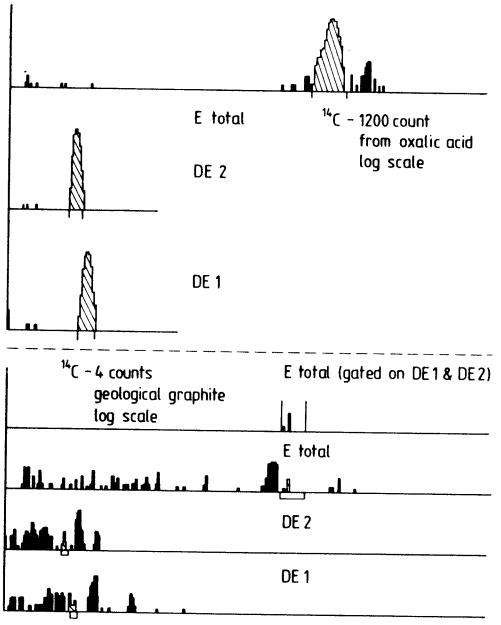


Fig 3. Energy spectra from the ^{14}C detector

MEASUREMENT OF THE ISOTOPE RATIO $^{14}/12$. This ratio is measured by comparison with that from a standard, and fractionation in the system which is linear with mass, ie, dependent on dynamic effects rather than, eg, beam alignment, is corrected for by reference to the change in $^{13}/12$ ratio. Thus, absolute measurements are not required, but the system must have a high degree of stability for accurate (<2%) measurements. Stability was measured under conditions similar to those for actual dating, but is not yet adequate for dating.

Stability of electronic measurements. By feeding stable currents into the gated integrators (which measure the pulsed beams entering the precision Faraday cups), and using a radioactive source before the detector, the dating experiment can be mimicked. The overall stability between runs is better than 1% provided the temperature of the gated integrators themselves is kept within 5°F.

Stability during a run with a single sample. Both ratios must be considered, and can be measured separately. As a rule, the $^{13}/12$ ratio is reproducible for a period of up to ca 3 hours, to within 1%. Occasionally, drifts in the value of 1 to 2% over 30 minutes are observed, and sometimes step changes of

2 to 3% take place. Such variations, which we suspect reflect varying source conditions, occur in ca 20% of runs of ca 30 minutes.

The stability of the $^{14}/12$ ratio is more difficult to measure, because of the time required to accumulate adequate statistics. We used Fermi graphite enriched to ca 4 times modern and observed quite severe drifts of the $^{14}/12$ ratio with time, nearly always falling away from the initial (set-up) value. We ascribe these to terminal voltage drifts which show a similar time-dependent behavior, and have the appropriate magnitude. We should eliminate this problem by stablizing the voltage on a signal from the pulsed ^{12}C beam position at the analyzer magnet. Table 1 lists other possible effects from the pulsed ^{14}C transmission, although there is no reason to expect other beam transport components to drift by the required amount. It is, of course, vital that the transmission of beam from the analyzer to detector can be kept constant to better than 1%, since these changes cannot be compensated.

^{14}C transmission will also probably be affected by the same causes of variation in the $^{13}/12$ ratio (apart from the linear, dynamical effects), which will have to be eliminated by study of the $^{13}/12$ ratio, since measurements to this accuracy on the $^{13}/12$ ratio take much more time.

Stability for runs with a single sample which is removed and replaced. Movement of the sample causes major changes in the $^{13}/12$ ratio, typically of ± 5%, sometimes of up to 15%. Very often the $^{14}/12$ ratio is correlated, but is not proportional to the mass difference. To some extent the same $^{13}/12$ ratio can be produced by exact repositioning of the sample, but movement of the order of 0.3mm, along beam axis, or 0.5mm, transversely, is sufficient to change the ratio significantly. We have evidence that these changes arise from only slight displacements in the three isotopic beams (see below).

Stability for runs between two samples of identical composition. The stability of these conditions is dominated by the problems mentioned above. One interesting and important feature that has emerged is that the injection conditions that maximize the transmission can be reproducibly different for different samples. In particular, a field shift of 0.5G in 2000 may be necessary to maximize the same isotopic beam from different samples on the target wheel. This corresponds to a shift of source position (transverse to the beam) of 0.25mm. Since the sputtering Cs^+ beam has a focused diameter of ca 1mm, with the target of similar size, such apparent shifts are not difficult to understand.

Stability conditions for dating. Accurate dating requires adequate stability under condition 4, above (to better than 1%) on account of dynamical fractionation effects, eg, occurring

TABLE 1. Change in HEMS system to produce 2%(5%) transmission change

Instrument	Beam	Aperture(mm)	Change in value
Terminal V	^{12}C,^{13}C pulsed		1.8 (2)kV @ 2MV
	^{14}C detector	1.5	120 (180)V
		2.3	450V
		4	550V
Analyzer magnet	^{12}C,^{13}C		18G = 0.18%
	^{14}C	1.5	0.3G = 3 x 10^{-5}
		4	1.5G = 1.5 x 10^{-4}
Wein filter	^{14}C only	1.5	0.2 (0.4)%
		4	1 (1.5)%
Magnetic quadrupole doublet lens	^{14}C only	1.5	0.5 (0.8)%
		4	1 (1.5)%
Electrostatic quadrupole doublet lens	all isotopes		5 (6)%
Stripper	(Attenuation)		~8% pressure change in gas
	C^{3+} fraction		25 (60)kV
Injector magnet	all isotopes		0.4 (0.6)G = 2 x 10^{-4}
Pulser voltage	^{13}C,^{12}C only		0.5(0.8)% = 1 x 10^{-4} for DE/E total
Injector lens			1.4 (2.3)%
Ion source lens			0.9 (2)%
Injector steerers (4)			17, 6, 17, 4%

in chemical preparation, and possibly in the mass spectrometer, and a demonstration that the measurement of $^{14}/12$ is indeed linear with the $^{14}/12$ ratio. Since the ^{14}C background can be shown to be sufficiently small, ie, below 1% of the sample requiring accurate dating, and the detector counts all particles, we do not expect difficulties in establishing linearity. However, the uncontrolled variations in $^{13}/12$ and $^{14}/12$ ratios prevent a demonstration of this at present.

THE EFFECT OF BEAM ALIGNMENT ON ISOTOPIC DISCRIMINATION. Table 2 shows the measured change in various functions of the system which bring about isotopic discrimination. The injection system is particularly sensitive, and angular or displacement changes of the order of that found in changing from one sample to the next are sufficient to bring about discrimination of the

TABLE 2. Change in HEMS system to produce 2%(5%) change in $^{13}/12$

Instrument	% change	Degree of fractionation change / transmission change
Ion source lens	1.1 (3) %	0.8
Injector lens	1 (2.5) %	1.4
Electrostatic quadrupole doublet	14%	0.3
Injector magnet	0.5,0.65 G (1G) ie 2.2×10^{-4}	0.8
Pulser voltages	0.5 (0.8) %	1.0
Injector steerers		
horizontal 1	10%	1.4
vertical 1	10%	0.6
horizontal 2	20%	0.8
vertical 2	6%	0.7
Analyzer magnet	15G = 1.5×10^{-4}	

magnitude observed. (It is most unlikely that run to run reproducibility is affected by lack of stability in the electronic control of the beam transport fields.)

Reference to the curve showing the observed change in $^{13}/12$ as a function only of the injector magnetic field (fig 4A), suggests that if the two transmission curves for $^{12}C^-$ and $^{13}C^-$ are not exactly superimposable, ie, if the isotopic beams are not exactly co-linear, a change in the field for optimum transmission ~0.5G, will produce isotopic discrimination of the magnitude seen. This is shown in Figure 4B, where a shift of 0.5G would give a 2% change in isotope ratio for a displacement equivalent to 0.25G in the two stable isotope beams. Two effects therefore multiply each other: slight changes in beam source position and direction from sample to sample, and lack of co-linearity in the injected isotope beams.

OUTLOOK. We have not collected enough evidence, nor made sufficient changes to the beam transport system to make predictions of performance with confidence. However, before the requisite accuracy can be achieved or any dynamical correction to the isotopic discrimination becomes appropriate, the following must be accomplished: 1) stabilization of the terminal voltage to within ± 150V at all times; 2) corrections to the injection system, either magnetically or with steerers synchronized with the pulsed isotopic beam, to reduce the isotope ratio sensitivity to misalignment; 3) understanding and reduction of the change of apparent source position for change of sample.

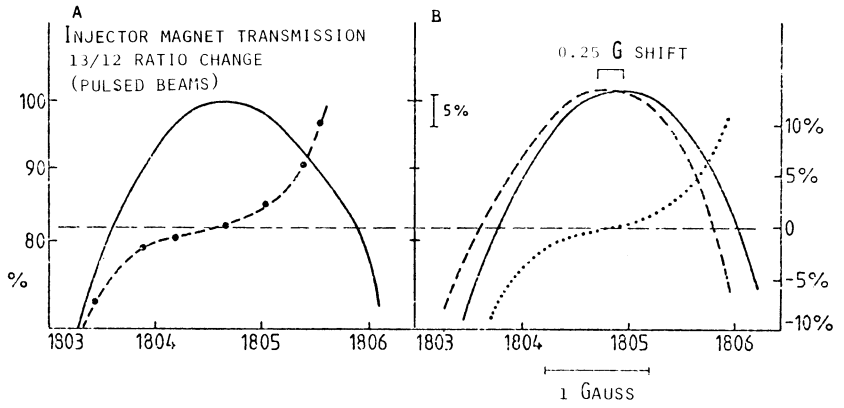


Fig 4A. Transmission of ^{13}C ion beam through the accelerator as a function of injector magnetic field. The dashed curve is the measured change in $^{13}C/^{12}C$ ratio as a result of changing the injector field.

Fig 4B. The continuous curve is a repeat of the transmission curve in 4A. The cashed curve is a similar curve, shifted by 0.25G, such that, if it represented the transmission of ^{12}C ion beam, the resulting isotopic discrimination would be given by the dotted curve.

REFERENCES

Hedges, REM, Wand, JO, and White, NR, 1980, The production of C^- beams for radiocarbon dating with accelerators: Nuclear Instruments and Methods, v 173, p 409-421.

White, NR, in press, The inverted spherical ionizer sputter ion source (IS3): Nuclear Instruments and Methods, in press.