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THE UTRECHT ACCELERATOR FACILITY FOR PRECISION DATING WITH RADIONUCLIDES

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The Utrecht facility for accelerator mass spectrometry is described. The set-up with an EN tandem accelerator is designed for measurements of a broad range of long-lived radionuclides and of stable trace elements. In particular, dating measurements with ^{10}Be and ^{14}C can be performed with high precision by rapid switching between the measurements of the isotopes. Optimal beam transmission through the accelerator is achieved with a new injection system in conjunction with the installation of axially symmetric acceleration tubes with spirally inclined electrodes. The first $^{14}C/^{12}C$ measurement with this set-up is reported.

1. Introduction

During the last three years the Utrecht tandem facility has been adapted for accelerator mass spectrometry [1]. The set-up is suitable for measurements of a broad range of long-lived radionuclides and stable trace elements. The first measurements have been performed recently.

For ^{10}Be and other long-lived radionuclides an accuracy of 5–10% in the isotope ratios is in most cases sufficient, but for ^{14}C a higher precision is required. Such a high precision provides an optimal extension towards milligram samples and short measuring times of the high quality ^{14}C counting data (with accuracies as high as 0.3%).

With accelerators such a high precision can only be achieved with a dedicated set-up which utilizes the electrostatic character of the tandem accelerator. Such a set-up has been described in 1977 by Purser [2] and has first been realized in Zürich [3]. It allows a (quasi) simultaneous measurement of the radionuclide ^{14}C and the stable ^{12}C and ^{13}C isotopes, instead of measuring these isotopes in separate runs at different accelerator settings. Such a dedicated set-up also requires optimal beam control. This can be achieved with optimal beam transmission through the accelerator by minimizing the beam losses.

2. The facility

Fig. 1 shows a schematic lay-out of the Utrecht facility for accelerator mass spectrometry with the EN tandem accelerator. The standard 20° injector, which is located between the 90° injector and the accelerator, is not shown in this figure.

In the 90° injector [4] the negative ions, produced from the sample in the inverted sputter source [5], are pre-accelerated before they are injected into the accelerator. Optimal beam transmission through the accelerator is achieved with this injection system. Apart from the 90% transparency of the gridded lens in the accelerator and the known stripping efficiency no other beam losses occur. In this respect the pre-acceleration, the gridded entrance lens in the accelerator and the axially symmetric acceleration tubes with spiral inclined field electrodes are essential (see sect. 2.2).

The characteristic feature of the set-up, which makes it dedicated for precise dating measurement, is the electrostatic character of the beam transport devices, including the accelerator itself. The 13° electrostatic analyser is placed close to the accelerator in the beam line used for nuclear physics experiments. For these experiments the left electrode in the beam direction is turned out. The beam for the mass spectrometry measurements is bent towards the set-up consisting of a 90° magnet and a large detection chamber which contains the ionization counter and the Faraday cups.

Through the mass independence of the electrostatic beam transport devices, the set-up can be used for acceleration of different ion masses (isotopes) at one accelerator setting. Rapidly alternating beam pulses of ^{12}C, ^{13}C and ^{14}C have the same optimal beam transmission. The last transport device, the analysing magnet, bends the ^{14}C beam in the ionization chamber and the ^{12}C and ^{13}C beams in the Faraday cups. One mass dependent process, however, is still present: the velocity dependent stripping process. But this can be corrected for in the calibration procedure.

The rapid switching, with a frequency of 10 Hz, between the measurements of the different isotopes, is fast with respect to slow terminal voltage variations. It

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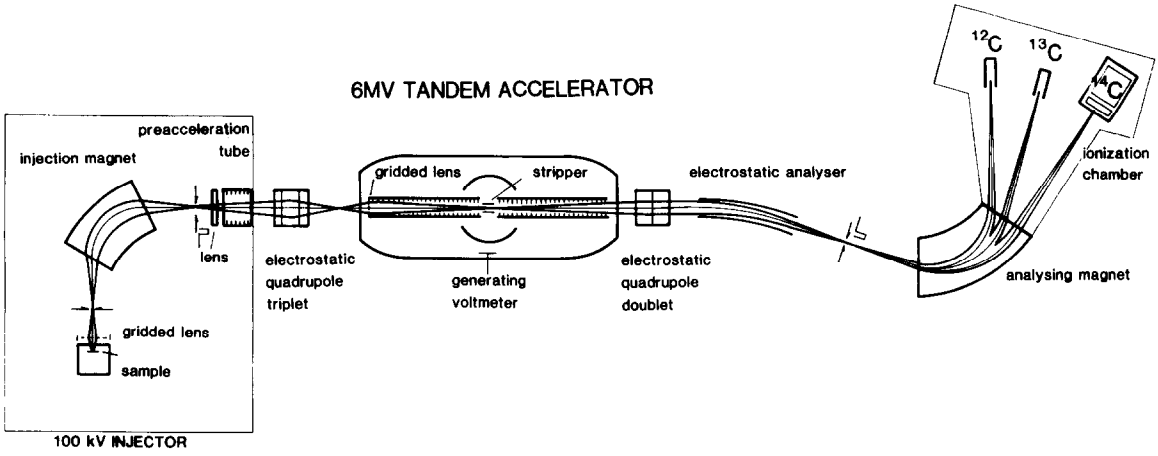

Fig. 1. The Utrecht facility for accelerator mass spectrometry.

thus simulates the simultaneous measurement of the isotopes. In this way the isotope ratio becomes independent on variations in beam current and of the poor reproducibility of the accelerator settings. Moreover, high beam currents ($\gtrsim 10\ \mu$A) can be used in the short beam pulses without affecting the accelerator condition.

2.1. Technical details

Technical details about the set-up concerning the 90° injector, the accelerator and the detection arrangement are summarized in table 1. The injection system with devices at a high-voltage level (see sect. 2.2) is operated via fiber-optics control [4,6] which serves 32 stations with 16-bit datawords in a serial transfer.

2.2. Beam transport calculations

For the design of the set-up beam transport calculations were performed with the program OPTRYK [7]. In these calculations a (too) large emittance of 14π mm · mrad · $(\mathrm{MeV})^{1/2}$ was assumed to ensure good spatial acceptance.

Fig. 2 shows the beam optics up to the stripper, which is the determining factor in the accelerator acceptance. Ions emerging from the source are focussed in the

Table 1
Technical details of the set-up

A. Injector	
Ion source	Inverted sputter source
Injection magnet	Double focussing, second order corrected, bending angle 90°, radius 40 cm, gap 6.0 cm; $m/\Delta m = 300$, mass–energy product 6 amu · MeV
Pre-acceleration	100 kV; acceleration tube: length 45 cm, diameter 9.0 cm
Insulation transformers	3 phase, 20 kV.A, 100 kV epoxy insulation; 1 phase, 4 kV.A, 30 kV epoxy insulation
Cooling	Freon-113 for magnet coils, kerosene for source and pumps
Remote control	Fiber-optics communication
Vacuum	2×10^{-7} Torr; Diffstaks 700 and 300 l/s
B. Tandem accelerator	
Acceleration tubes	Four spiral inclined field acceleration tubes; first tube in LE-part is provided with a gridded lens
Terminal voltage	Max. 6 MV (for mass spectrometry)
Generating voltmeter stabilization	$V/\Delta V \leqslant 2500$
Stripper	Gas or C-foil ($2\ \mu g/cm^2$)
Vacuum	5×10^{-7} Torr; two combinations of 700 l/s Diffstak and Vacion pumps
C. HE set-up	
Electrostatic analyser	Bending angle 13°, length 200 cm, gap 6.0 cm, turnable electrodes
Analysing magnet	Single-focussing, bending angle 90°, radius 86 cm, gap 2.5 cm; resolution $p/\Delta p = 1100$; mass–energy product 52 amu · MeV
NMR stabilization	$B/\Delta B \approx 2\times10^6$
Vacuum	2×10^{-7} Torr; two 700 l/s and two 300 l/s Diffstak pumps

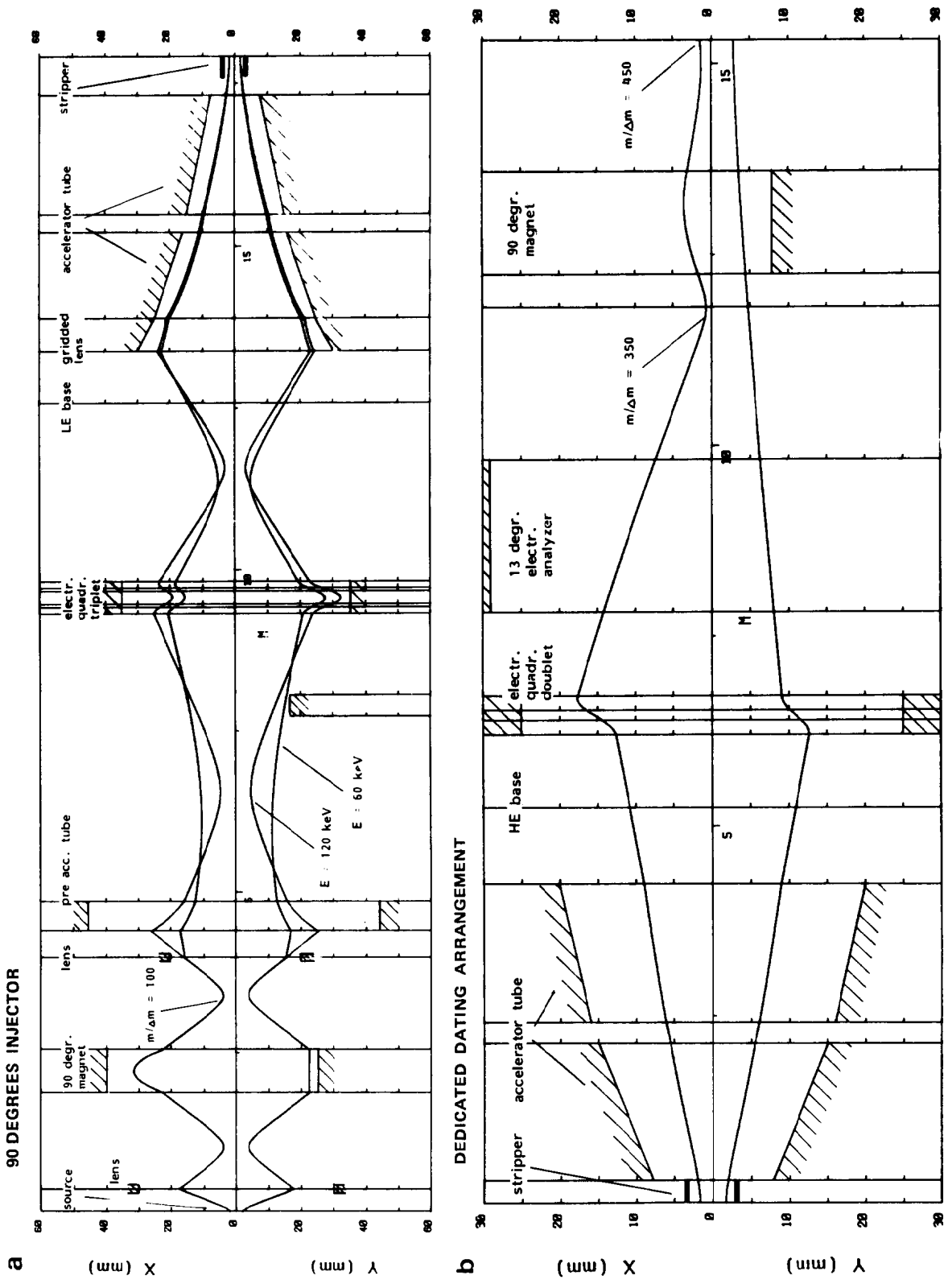

Fig. 2. Beam transport calculations for (a) the LE-part and (b) the HE-part of the accelerator. Shaded areas show spatial limitations.

object slits of the injection magnet by a gridded lens, which has small aberrations [8]. This lens reduces the divergence of the beam such that a magnet gap of 60 mm suffices. The double-focussing 90° magnet with a high mass resolution ($m/\Delta m \approx 300$) is used to reduce competing background ions. The injector has been built on an insulated platform so that ions can be accelerated in a 100 kV pre-acceleration tube. Usually the ions are pre-accelerated to 65 keV before they are transported with the electrostatic quadrupole triplet lens towards the accelerator.

Five electrostatic *xy*-steerers located before the object and behind the image slits of the injector magnet, in front of the 20° injector, the triplet and the accelerator, are used to correct for misalignments. In practice the steerers have to correct only for small deviations.

The gridded entrance lens in the first accelerator tube is essential for a good beam transport. It replaces the very strong and terminal voltage dependent lens effect at the entrance of the tube by a weak ($f \approx 1$ m) and adjustable lens, such that the beam can be focussed in a 3 mm diameter spot in the stripper (gas or foil). Not only the gridded lens is important, but also the axially symmetric character of the spiral inclined field acceleration tubes. In case acceleration tubes with slotted electrodes are used, the lens effect due to the slotted electrodes is asymmetric and comparable in strength with that of the gridded lens, so that simultaneous focussing in the horizontal and vertical plane cannot be reached.

The beam optics for the beam from the stripper towards the detection arrangement is shown in fig. 2b. The electrostatic quadrupole lens focusses the beam through the electrostatic analyser which bends the beam horizontally over 13°. Since the 90° analysing magnet ($R = 86$ cm) is single-focussing, the beam is focussed horizontally in the object slits and vertically in the detector located in the image plane. In order to achieve a sufficient spatial separation of about 10 cm between the Faraday cups and the detector in the image plane of the magnet, the object distance has been taken $R/2$ so that the image is at about $2R$ without losing much resolution.

2.3. Injection and measurement of ^{12}C and ^{13}C beam pulses

The ^{12}C and ^{13}C beam pulses are produced by hv-pulsing of the vacuum chamber of the 90° injection magnet. Special devices have been developed to produce the pulsed beam sequence [9] and to measure the corresponding charges.

The time structure of the beam pulses is determined by that of the hv-pulsing of the vacuum chamber. The hv pulses accelerate the $^{12}C^-$ and $^{13}C^-$ ions to a magnetic rigidity equal to that of the $^{14}C^-$ ions for which the magnet is set. For the 15 keV ions emerging from the ion source this implies hv pulses of 2.50 and 1.15 kV for ^{12}C and ^{13}C, respectively.

In beam transport calculations (similar to those in fig. 2a) for the LE-end, the lens effects have been checked which occur at the insulation gaps. In the case that 4 kV pulses are applied at these gaps located within 30 cm of the object and image slits of the injection magnet, these lens effects are negligibly small ($f \approx 10$ m).

Well-defined time sequences are derived from the main clock in the beam sequencer, a 2 MHz oscillator. These time signals are used to operate solid-state switches via fiber-optics communication. In this way up to three 1.4 kV power supplies can be stacked. One of these stacked power supplies is adjustable so that any voltage up to 4.2 kV can be selected. With proper timing and safety checks, the power supplies are switched sequentially. The total rise time is about 20 μs, determined by the fiber-optics communication. The flatness of the hv pulse is better than 0.1%.

The pulses of both ^{12}C and ^{13}C are separated by half a cycle time. These are independently adjustable in time (0.1–0.9 ms) and voltage (0–4.2 kV), whereas the cycle time can be choosen between 0.1 and 0.9 s. A "Pulse Valid" signal is present when the hv pulse is within 0.1% of the maximum value. This signal, properly delayed to compensate for the beam flight time, starts the corresponding current integrator (sensitivity 3 pC to 100 nC), which produces a pulse train with a frequency (maximum 1 MHz) proportional to the accumulated charge. The intrinsic electronic accuracy of these integrators is better than 0.1%.

For each beam pulse of ^{12}C and ^{13}C, a new frequency is produced. These frequencies are counted in scalers during the ^{14}C acquisition period. The isotope ratios are independent on the computer dead time. This has been achieved by disabling the ^{12}C and ^{13}C scalers by computer-busy signals.

3. First results

The installation of this set-up was completed two weeks before this conference. The results obtained in this short period are described here. The beam pulsing system and the current integrators work according to design. The data acquisition system, based on scalers and 11-bit ADCs interfaced with CAMAC to a PDP-11/34 computer, is used for on-line data reduction.

The measurements were performed with graphitized [10] medicinal carbon, which produced in the inverted sputter source a reasonably stable beam current of 10 μA $^{12}C^-$. The ions were accelerated by a 5 MV terminal voltage, and the 4^+ charge state was selected. The ^{14}C yield obtained with the ionization chamber is excel-

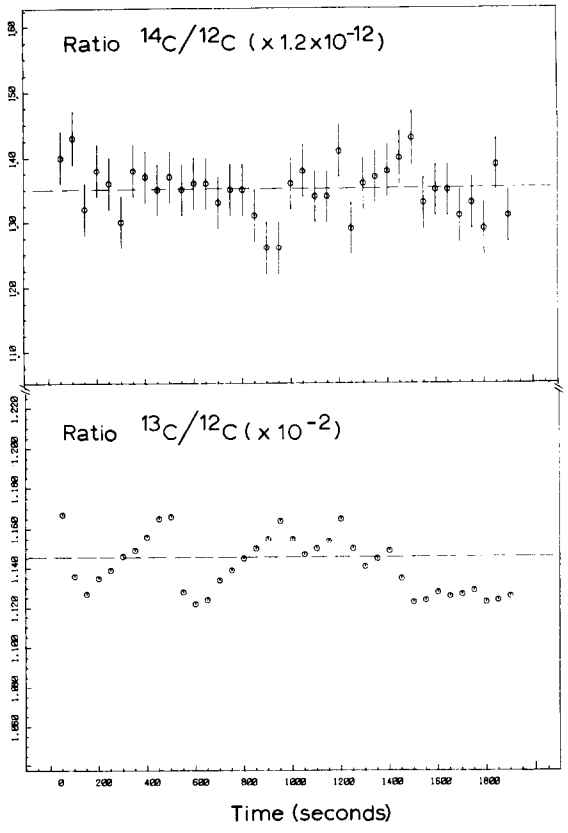

Fig. 3. First results with a recent carbon sample obtained with pulsed beams. Each data point represents a measuring time of 50 s. The $^{14}C/^{12}C$ ratio determined is $(1.350\pm0.007)\times1.2\times10^{-12}$ and the $^{13}C/^{12}c$ ratio is $(1.146\pm0.003)\times10^{-2}$. The variations in the $^{13}C/^{12}C$ ratio reflect long time correlations.

lently identified. The first isotope ratios measured with pulsed beams are shown in fig. 3. Each data point represents a measurement of 50 s; the total measuring time was 1800 s. The mean value obtained for the $^{14}C/^{12}c$ ratio is $(1.350 \pm 0.007)\times 1.2\times 10^{-12}$, quite reasonable for recent material. The quoted error (standard deviation) is quite small. The reproducibility, however, is poor in comparison with this high-precision value. Deviations of several percents, found in repeated measurements, are related to initial problems in the measurement of the current, as can be seen from the simultaneously recorded $^{13}C/^{12}c$ ratio. The value obtained, $(1.146 \pm 0.003)\times 10^{-2}$, deviates from the natural abundance ratio. Moreover, the $^{13}C/^{12}C$ ratio shows long time correlations due to interference with the 50 Hz of the mains.

In order to avoid interferences of this type, the beam sequencer has now been modified to produce beam sequences which shift continuously in time relative to the line frequency. After several seconds possible correlations are averaged out in this way.

The promising first results obtained with the new set-up indicate that reproducible high-precision ^{10}Be and ^{14}C data can be produced with this facility. A research program for archeology, geology and oceanography will be carried out. Simultaneously measurements are prepared of other radionuclides like ^{26}Al, ^{36}Cl, and ^{129}I and trace elements.

References

[1] A.E. Litherland, Ann. Rev. Nucl. Part. Sci. 30 (1980) 437; W. Kutschera, Radiocarbon 25 (1983) 677.
[2] K.H. Purser and P.R. Hanley, Proc. 1st Conf. on Radiocarbon dating with accelerators, ed., H.E. Gove, University of Rochester (1977).
[3] W. Wölfli et al., Radiocarbon 25 (1983) 115.
[4] K. van der Borg A.P. de Haas, A. 14 Hoogenboom, B.A. Strasters, A. Vermeer and N.A van Zwol., Nucl. Instr. and Meth. 220 (1984) 115.
[5] B.A. Strasters, A. Vermeer and N.A. van Zwol, Nucl. Instr. and Meth. 220 (1984) 109; J.L. Yntema and P.J. Billquist, Nucl. Instr. and Meth. 199 (1982) 637.
[6] A.P. de Haas et al., to be published in Nucl. Instr. and Meth.
[7] T.J.L. Greenway, computer code OPTRYK, Internal Report, Oxford University.
[8] M.Y. Bernard, J. Phys. Radium 8 (1953) 451.
[9] H. de Vries and K. van der Borg, to be published in Nucl. Instr. and Meth.
[10] R.D. Shuiling, Inst. of Earth Sciences, University of Utrecht, private communication.