BREAKTHROUGH OF THE MINI-CYCLOTRON MASS SPECTROMETER FOR ^{14}C ANALYSIS

MAOBAI CHEN, DEMING LI, SENLIN XU, GUOSHENG CHEN, LIGONG SHEN, XIANGSHUN LU, WEIYU ZHANG, YUEXIANG ZHANG, ZONGKUN ZHONG and YINGJI ZHANG

Shanghai Institute of Nuclear Research, Academia Sinica, P.O. Box 800204, Shanghai 201800 and T. D. Lee Physics Laboratory, Fudan University, Shanghai 200433 China

ABSTRACT. We present results of current measurements on the super-sensitive mini-cyclotron as an accelerator mass spectrometer, successfully developed at Shanghai Institute of Nuclear Research (SINR). We describe new ideas and unique techniques adopted for increasing the transmission efficiency in the injection, acceleration and extraction region, for eliminating various backgrounds and for improving the precision of ^{14}C analysis, which have led to the breakthrough of our Shanghai Mini-Cyclotron Accelerator Mass Spectrometry project. We also discuss further development of the prototype facility.

INTRODUCTION

A cyclotron was first used as an accelerator mass spectrometer (AMS) by L. W. Alvarez in 1939 in an experiment that demonstrated the stability of ^{3}He (Alvarez 1981). In 1977, R. A. Muller published an article in which he suggested that a cyclotron could be used as a mass spectrometer to detect ^{14}C (Muller 1977). Following the initiative of the Berkeley group (Welch *et al.* 1987), we began the Shanghai Mini-Cyclotron Accelerator Mass Spectrometry (SMCAMS) project in late 1985. The first phase of the project has now been completed. Using resonance analysis, the cyclotron AMS can sensitively discriminate among the background particles even with the mass difference resulting only from mass defect. Thus, there is less tail phenomenon on a mass spectrum. A cyclotron should be the most suitable for a super-sensitive mass spectrometer, but it is not, for the following reasons:

1. Because stable negative ions, such as N^-, do not exist for isobars of many radioactive nuclei to be analyzed, the required resolution for AMS is greatly decreased if negative ions of a sample particle are analyzed. Cyclotrons do not usually can accelerate negative heavy ions, whereas tandems do.
2. In dating applications, one must measure the ratio of the radioactive isotope to its corresponding stable isotope, such as $^{14}C/^{13}C$, by alternately accelerating both negative ions. It is easier for a tandem AMS to change one electric parameter sequentially, or even to measure all three isotopes simultaneously (Purser 1992). It is more difficult for a large cyclotron to change many electric parameters as well as magnetic field sequentially.
3. The beam intensity in all small cyclotrons developed in the 1950s (Clark 1984) and in the 1980s (Bertsche *et al.* 1990) was too low to count rare isotopes.

Thus, tandem AMS dating has overtaken the cyclotron AMS technique during the past decade.

As a super-sensitive mass spectrometer, the mini-cyclotron has the following advantages common to both tandem and cyclotron AMS:

1. It is capable of accelerating negative ions, which can be extracted directly for measurement without the need for stripping. Positive ions can also be analyzed.
2. It retains the function of resonance analysis of the cyclotron AMS. Alternate acceleration can be undertaken without changing the magnetic field, because the relativistic effect is negligible at very low energy (50 keV).
3. Most important, it can be set up at almost any laboratory for ^{14}C analysis, because of its low operating costs, small size, low energy, low magnetic field and low power consumption (12 kW).

Proceedings of the 15th International ^{14}C Conference, edited by G. T. Cook, D. D. Harkness, B. F. Miller and E. M. Scott. RADIOCARBON, Vol. 37, No. 2, 1995, P. 675–681

KEY TO THE BREAKTHROUGH

A mini-cyclotron AMS for ^{14}C analysis should not only efficiently deliver the heavy negative $^{14}C^-$ ions, but it should also eliminate all backgrounds, especially $^{13}CH^-$ ions, the ion closest to $^{14}C^-$. The mass resolution needed for discriminating $^{14}C^-$ from $^{13}CH^-$ is *ca.* 1800. According to the resolution formula of a cyclotron

$$R = M/\Delta M = 360^\circ \, nh/\Delta Q = 2\, nh \tag{1}$$

where $\Delta Q = 180^\circ$ is the defined maximum amount needed for shifting the radio frequency (RF) phase of $^{13}CH^-$ ions to the deceleration phase, n is the maximum acceleration turn number experienced by the $^{13}CH^-$ ions with $\Delta Q = 180^\circ$, and h is the harmonic number, *i.e.*, the ratio of RF to $^{14}C^-$ ion revolution frequency. High resolution can be achieved by increasing the product, nh. However, the increase of either n or h would require strict mechanical constraints and great electrical stability, as well as precision of the isochronous magnetic field on a mini-cyclotron itself. Hence, this would jeopardize the particle acceptance of the mini-cyclotron.

A large turn number, n, would 1) hamper the operation of negative heavy $^{14}C^-$ ions with very low energy; 2) enlarge the diameter of the mini-cyclotron magnet; or 3) tighten the turn spacing so as to damage the injection and extraction of particles. We were forced to meet the high-resolution requirements by increasing the harmonic number, h. Although a high harmonic number would greatly enhance some effects, such as the energy spread of the beam, RF phase divergence and convergence of particles and the coupling effect between longitudinal and transversal motion, it would result in deteriorated beam quality and density. Thus, it is unreasonable for cyclotrons to operate with high harmonics, primarily because the sinusoidal wave (sine wave) accelerating voltage is used in all existing cyclotrons.

Attempts were made in the 1950s to build small cyclotrons as mass spectrometers (Clark 1984). However, all failed to achieve abundance sensitivity of higher than 10^{-9} due to low beam intensity (10^{-15} A). This is the case for the modern small Cyclotrino (10^{-9} A) at Berkeley (Bertsche *et al.* 1990).

To obtain ^{14}C counts from the SMCAMS, we focused on increasing the $^{12}C^-$ beam intensity by improving the transmission efficiency in the acceleration region as well as the injection and extraction efficiency. To overcome the effect of the high-harmonic operation on the transmission efficiency in the acceleration region of a mini-cyclotron, we used a triangular-wave accelerating voltage instead of the usual sine-wave accelerating voltage (Chen, Li and Gao 1989). Calculations indicated (Lu 1992) that the transmission efficiency using a triangular-wave voltage would increase by *ca.* 50 times compared with sine-wave voltage. The results of beam tuning were also satisfactory.

To fully use the powerful function of the triangular-wave voltage and to minimize the energy spread of the beam, the RF phase of particles should be located in the linear area of the real triangular wave voltage when crossing the accelerating gap. This is because non-linearity exists on both ends of the real shape of the triangular wave voltage. Nevertheless, the force of the electric focusing field (phase focusing) approaches zero near the center of the linear area of the triangular wave voltage, which makes the ion motion unstable and renders insignificant the adoption of the triangular-wave voltage to increase beam intensity. To keep ion motion stable, we must rely on magnetic focusing by using a non-uniform magnet. We believe a uniform magnet is inappropriate for AMS with a cyclotron, where the analyzed particles circle inside the magnet for many turns rather than for <1 turn, as is the case in conventional mass spectrometry. Thus, we designed a non-uniform magnet with high flutter.

The nickel-coated yoke constitutes the vacuum chamber of the mini-cyclotron, the first time such a design has been used. The isochronous magnetic field is adjusted by correcting the shape of the sectors without using usual trim coils.

After we successfully built a ramp generator of V (peak to peak) = 1 kV and shaped the isochronous magnetic field, the beam tuning indicated a transmission efficiency in the acceleration region of >20% under harmonic h=16 operation. Such a transmission efficiency is affected by many factors, which include linearity of the real triangular-wave voltage, isochronous deviation of the shaped magnetic field, vacuum and aperture of the accelerating (dee) electrodes.

For mini-cyclotron AMS, one should choose external injection for: 1) promptly changing samples; 2) greatly reducing memory effect; 3) adopting a high-yield Cs sputter negative ion source; and 4) applying an emitting voltage (20 kV) that is much higher than the accelerating voltage (500 V). It is also important that the single-particle detector—a dynode and microchannel plate (Friedman *et al.* 1988; Zhang *et al.* 1991)—which can count the particle number but cannot distinguish the particle, should be located outside the mini-cyclotron to avoid the strong X-ray interference inevitably induced by the beam hitting inside the vacuum chamber of the mini-cyclotron. Thus, we should pay more attention to improving the injection and extraction efficiency.

As for axial injection in a cyclotron, it is usually accomplished by passing particles through a *mirror* or a *spiral* inflector. We designed a pair of spherical electrostatic injection deflectors—vertical injection deflector and horizontal injection deflector (Fig. 1)—and obtained a total injection efficiency of *ca.* 60%.

Because the energy gain per turn in SMCAMS is small (Chen, Gao and Li 1989), the next goal for improving injection and extraction efficiency is to obtain a turn spacing wide enough to clear the

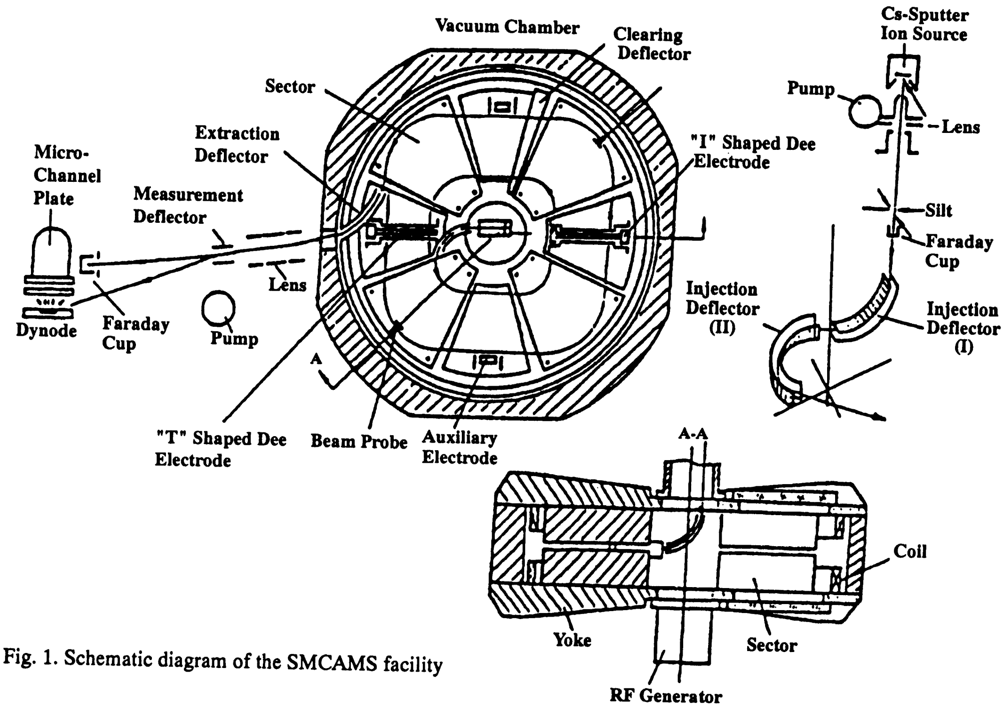

Fig. 1. Schematic diagram of the SMCAMS facility

horizontal injection deflector in the first turn and to enter the horizontal extraction deflector at the last turn. The magnetic perturbation method often used in large cyclotrons to obtain the desired turn spacing is not applicable to the mini-cyclotron. In our mini-cyclotron with a non-uniform magnet of high flutter, we use accelerating (dee) electrodes with zero angle for convenience of assembly. These electrodes constitute differential electrodes with a sandwich structure, equivalent to two adjacent dee-dummy dee structures in series (Chen, Xu and Li 1990). The feature of such an electrode arrangement is that the effective dee voltage experienced by particles crossing the electrode gap increases as the harmonic number, h, increases, and can be adjusted by changing the geometric parameters, D (gap width), T (dee width) and H (dee aperture) of the structure rather than by merely adjusting the dee voltage. To meet the high-energy gain demanded for injection and extraction, and to keep the lowest energy gain within working radii, we adopted a pair of auxiliary electrodes and asymmetric width varied electrodes with wedge shape. This is based on the above-mentioned feature of the differential electrode and the recently explored phase divergence and convergence phenomenon under high harmonic operation (Chen, Xu and Li 1990) to obtain the necessary turn spacing of 6 mm. This new type of electrode is simple and practical.

We have begun to take steps toward eliminating interference of backgrounds, particularly ^{13}CH and X-rays by:

- Analyzing negative ions of ^{14}C to remove the isobar ^{14}N$^-$
- Defining ΔQ = 180° or R = 2nh to determine the necessary turn numbers to ensure eliminating ^{13}CH$^-$
- Establishing a prohibitive zone of 6 cm wide. Theoretically, ^{13}CH$^-$ with ΔQ = 180° will start to decelerate inward before it approaches this zone, whereas ^{14}C$^-$ particles will continue to accelerate outward for *ca.* 30 additional turns to reach the extraction region. If there are any unwanted particles in this zone, their energy and direction of motion would probably be unsuitable for the extraction condition of the deflector.
- Using the extraction deflector as a final electrostatic analyzer
- Placing the single particle detector outside the mini-cyclotron, and protecting it from strong X-rays by lead shielding
- Adding two additional deflectors during alternate acceleration: a) the *measuring deflector*, to which a pulsed rectangular voltage is applied while detecting ^{14}C$^-$ to deflect the ions onto the dynode of the single particle detector. This voltage will be suppressed while measuring ^{12}C$^-$ or ^{13}C$^-$, as they will strike an adjacent Faraday cup; b) the *clearing deflector*, to which a pulsed voltage is applied at the moment when the alternate ^{12}C$^-$ or ^{13}C$^-$ acceleration is turned into ^{14}C$^-$ acceleration.

In our mini-cyclotron, all particles emitted from the ion source are simultaneously injected into the vacuum chamber without using an analyzing magnet prior to the cyclotron, because most of the unwanted particles, except ^{13}CH$^-$ ions, can easily be eliminated by powerful resonance analysis. However, the lack of the pre-analyzing magnet makes measuring ^{12}C$^-$ and ^{13}C$^-$ difficult.

In dating, one must measure sequentially the ratio of the detected radioactive isotope being analyzed to its corresponding stable isotope, such as ^{14}C/^{12}C and ^{13}C/^{12}C. Thus, contrary to conventional cyclotrons, where only one type of ion can be accelerated, the ^{12}C$^-$, ^{13}C$^-$ and ^{14}C$^-$ ions must be accelerated alternately in a mini-cyclotron AMS. It is a complex task for a mini-cyclotron, even though it reaches a low final energy and requires no adjustment to the magnetic field. Because the mass difference between ^{14}C and ^{12}C is *ca.* 15% and the magnetic field is fixed, most electric parameters must be sequentially changed for *ca.* 15%, according to the orbit similarity theory.

The transmission efficiency of different particles ($^{12}C^-$, $^{13}C^-$ and $^{14}C^-$) from the exit of the ion source down to the detector must always be calibrated against a standard or known sample, which we can also use to monitor and correct for any change of the transmission efficiency. A blank sample is also used to monitor the background level of the detector. A feature critical to precision of analysis is the flat-top transmission characteristic of the mini-cyclotron AMS in the injection, acceleration and extraction region. Even if there is an inevitable minor fluctuation of critical parameters, the transmission efficiency must not change or induce fractionation.

RESULTS

Figure 2A shows the $^{14}C^-$ and $^{13}CH^-$ frequency response (counts *vs.* frequency) for a carbon sample made from sugar. It shows that the ^{14}C counting is *ca.* 100 cpm under *ca.* 15 μA of ^{12}C from ion source, and the ratio of peak to valley on the curve reaches 3. Figure 2B is the same curve for a blank sample. No counts occur at the frequency position corresponding to the peak of $^{14}C^-$ counts, which implies that the adjacent $^{13}CH^-$ ions have been thoroughly suppressed. Further, background counts for a blank sample show a detection limit of 10^{-15}. Figure 3A, B shows the frequency response for $^{12}CH_2^-$, $^{13}CH^-$ and for $^{12}CH^-$, $^{13}C^-$. It is apparent from Figure 3 that the resolution of the SMCAMS has approached *ca.* 3000, which is much higher than needed (*ca.* 1800) for distinguishing ^{14}C from ^{13}CH. These measurements indicate that the SMCAMS facility is now able to perform AMS.

CONCLUSION

The first phase of the SMCAMS project established our facility and obtained ^{14}C counts to demonstrate our innovation. For practical applications, we must build a microcomputer-controlled system for alternately accelerating $^{12}C^-$, $^{13}C^-$ and $^{14}C^-$, and equip the vertical Cs ion source with a fiber-optic-controlled multi-sample device for sequentially changing samples. We must also improve the performance of the prototype, especially the injection and extraction efficiencies, and explore methods of alternate acceleration and ^{14}C analysis on a mini-cyclotron.

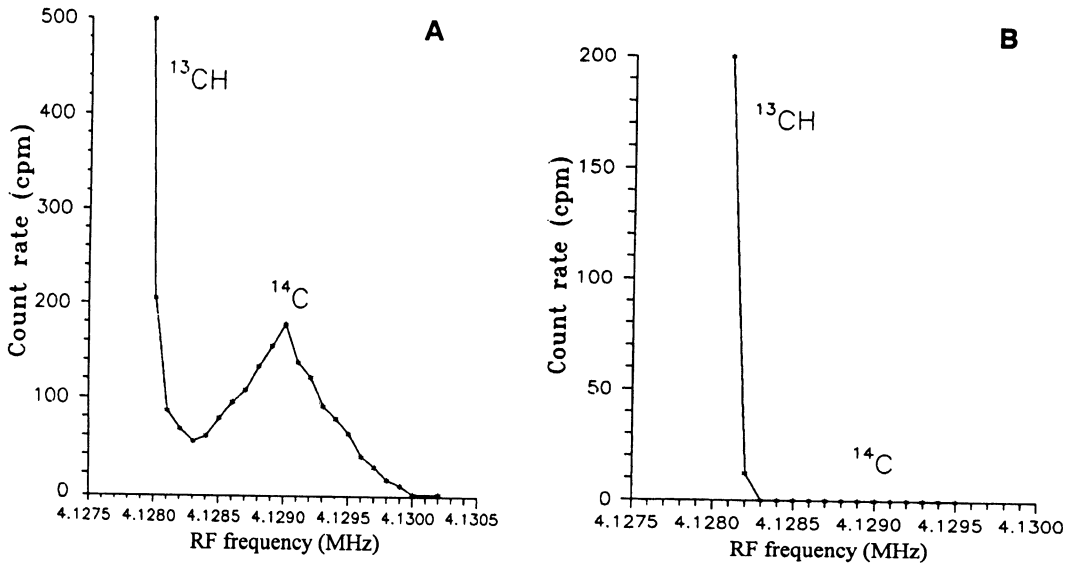

Fig. 2. A. ^{14}C and ^{13}CH frequency response curve for a carbon sample made from sugar. B. ^{14}C and ^{13}CH frequency response curve for a blank sample.

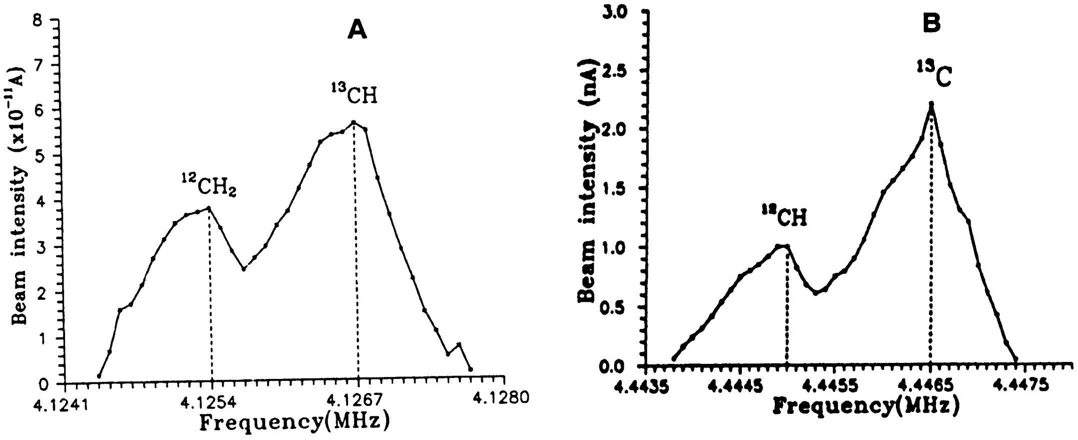

Fig. 3. Frequency response curves for blank samples: A. $^{12}CH_2$ and ^{13}CH; B. ^{12}CH and ^{13}C

The computer system has now been completed and the alternate acceleration is being tested. We have just finished designing the multi-sample device for our vertical ion source and expect to test this device in late 1995. We have also refined our beam tuning and remeasured the whole ion spectrum of $^{12}C^-$, $^{13}C^-$, $^{12}CH^-$, $^{14}C^-$, $^{13}CH^-$ and $^{12}CH_2{}^-$ (Fig. 3). The ratio of peak to valley on the ^{14}C and ^{13}CH frequency response curve has now increased to 4, as shown in Figure 4. This illustrates the improvement of the resolution; the flatter shape of the peak is better for ^{14}C-counting precision. We have learned the causes for beam loss in the central region of the mini-cyclotron, and are confident of improving transmission efficiency by refining the central region and increment of the dee aperture as well as the vacuum. We will reinforce the X-ray shielding to further improve counting operation.

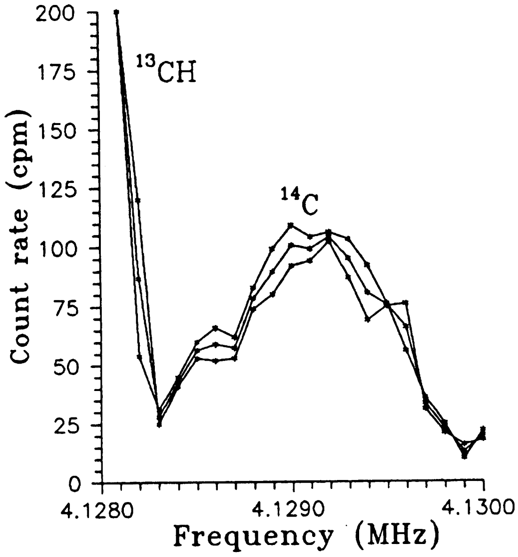

Fig. 4. Current-measured ^{14}C and ^{13}CH frequency response curve for a carbon sample made from sugar

ACKNOWLEDGMENTS

This research was supported by the National Natural Science Foundation of China and Academia Sinica. We are also indebted to Prof. Fujia Yang for his strong support and encouragement.

REFERENCES

Alvarez, L. W. 1981 The early days of accelerator mass spectrometry. *In* Kutschera, W., ed., *Proceedings of the Symposium on Accelerator Mass Spectrometry.* ANL/PHY 81-1, Springfield, National Technical Information Service: 1–15.

Bertsche, K. J., Karadi, G. A., Muller, R. A. and Paulson, G. C. 1990 Detection of radiocarbon in the cyclotrino. *In* Yiou, F. and Raisbeck, G. M., eds., Proceedings of the 5th International Conference on Accelerator Mass Spectrometry. *Nuclear Instruments and Methods in Physics Research* B52: 398–402.

Chen, M. B., Gao, W. Z. and Li, D. M. 1989 Design criteria on a mini cyclotron as a mass spectrometer for dating. *Nuclear Instrument and Methods in Physics Research* A278: 402–408.

Chen, M. B., Li, D. M. and Gao, W. Z. 1989 Adopting a non-sinusoidal wave dee voltage in minicyclotron as mass spectrometers for dating. *Nuclear Instruments and Methods in Physics Research* A278: 409–416.

Chen, M. B., Xu, S.L. and Li, D. M. 1990 Determination of the shape of the dee electrodes in the SINR minicyclotron used as AMS for ^{14}C dating. *Nuclear Instrument and Methods in Physics Research* A297: 47–59.

Clark, D. 1984 Cyclotrons as mass spectrometers. *In* Marti, F., ed., *Proceedings of the 10th International Conference on Cyclotrons.* New York, The Institute of Electrical and Electronics Engineers: 534–539.

Friedman, P. G., Bertsche, K. J., Michel, M. C., Morris, D. E., Muller, R. A. and Tans, P. P. 1988 Low background-rate detector for 40 keV ions using a conversion dynode and a microchannel-plate electron multiplier to reject low-energy ions, electrons, and photons. *Review of Scientific Instruments* 59(1): 98–111.

Lu, X. S. (ms.) 1992 Study of Effects of Non-Ideal Factors on the Minicyclotron as AMS. Master's dissertation. Shanghai Institute of Nuclear Research: 105 p.

Muller, R. A. 1977 Radioisotope dating with a cyclotron. *Science* 196: 489–494.

Purser, K. H. 1992 A high throughput ^{14}C accelerator mass spectrometer. *In* Long, A. and Kra, R. S., eds., Proceedings of the 14th International Radiocarbon Conference. *Radiocarbon* 34(3): 458–467.

Welch, J. J., Bertsche, K. J., Friedman, P. G., Morris. D. E., Muller, R. A. and Tans, P. P. 1987 A low energy cyclotron for radiocarbon dating. *Nuclear Instruments and Methods in Physics Research* B18: 202–214.

Zhang, Y. G., Wei, Y. G.,Wang, Q. Y. and Qian,W. G. 1991 A position-sensitive microchannel-plate detector for 27 keV ions. *Nuclear Instruments and Methods in Physics Research* A302: 76–82.