MEASUREMENT OF ¹⁰Be AND ²⁶Al WITH A TANDETRON ACCELERATOR MASS SPECTROMETER FACILITY

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ABSTRACT. A description is given of the use of a Tandetron accelerator mass spectrometer system for measuring the isotopes ¹⁰Be and ²⁶Al.

INTRODUCTION

At the third symposium on accelerator mass spectrometry at Zurich, we discussed a procedure for measuring ¹⁰Be with a Tandetron accelerator mass spectrometer. During the past year we have used this procedure to measure cosmogenic ¹⁰Be in over 430 natural samples, principally polar ice, marine and lacrustine sediments, but also cosmic spherules, rainwater, coral, fossil organic deposits and ocean particulate matter. We briefly review here the present status of our measurement technique, including background levels, efficiency and reproducibility. During the past few months we have also developed a procedure for measuring the isotope ²⁶Al with the Tandetron operating at a nominal terminal voltage of 1.75 MV. The details of this procedure, which has an efficiency and background levels comparable to higher energy machines, are presented below.

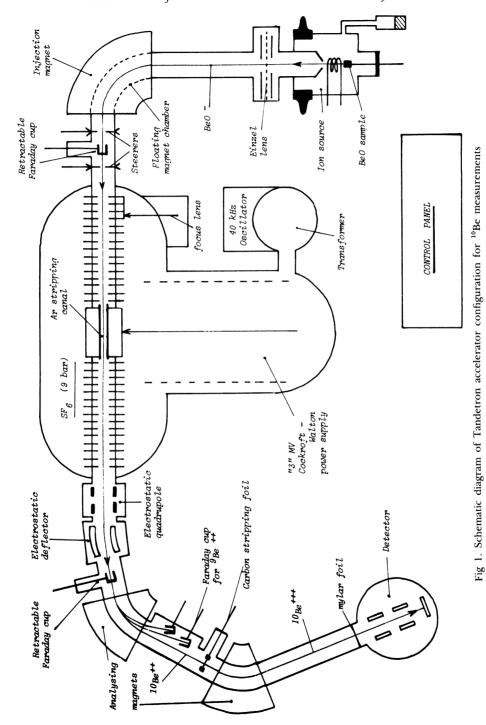
 $^{10}\mathrm{B}\epsilon$

A schematic diagram of the Tandetron operation for ¹⁰Be analysis is shown in Figure 1. A detailed description of the procedure used is given in Raisbeck *et al* (1984). Although the technique used during the past year has changed little since our earlier description, the much larger experience obtained since that time allows us a significantly improved appreciation of our capabilities. These are summarized below.

Reproducibility

For most of our measurements we continue to include a 7% instrumental uncertainty in the reported results. This is approximately the observed long term (months) variability observed in the measured 10 Be/ 9 Be ratio of our 10 Be standard. However, since we usually measure the standard 2 or 3 times during the day, and normalize our results to them, it is likely that the actual instrumental error is usually less than this. For example, in a recent series of sediment measurements, we measured 18 different samples two or more times, on different days. The average variability of these duplicates was 2.7%. In the same series, there was one section of fairly homogeneous sediment in which we processed 8 different samples. The standard deviation of these 8 measurements was 3.1%. Thus, for a homogeneous series of samples, which are not limited by statistics, and when the machine is stable, it is probable that the precision is of the order of 3% or so.

Although the above results indicate that we have the potential, under favorable conditions, to make measurements to $\sim 3\%$, there are also periods in which we observe greater variability in our standard, even over a



period of several hours. As mentioned in Raisbeck *et al* (1984), we believe the most important cause of this variation are small ($\sim 5 \times 10^{-4}$) changes in the fields of the high energy magnets. We have found the 10 Be to be much more sensitive than 14 C to changes in these fields, probably because of spreading in the beam due to breakup of the BeO molecule and diffusion in the carbon foil used to eliminate 10 B. The magnet gaps are too non-uniform to accept a NMR probe and our Hall probes are not temperature-stabilized. Since the temperature in the building housing the accelerator often varies over several degrees during the day, the drift in the Hall probes presently makes them unusable for controlling the magnets.

Background

Raisbeck *et al* (1984) estimated our background as 10 Be/ 9 Be ~3 × $^{10^{-14}}$. Over the past year we have measured this background a number of times, with values falling in the range $1 \pm 0.3 \times 10^{-14}$. Since this is much lower than any of the samples we have measured to date, we have not made any attempt to improve on it. It should be noted, however, that this background is due to real 10 Be, and not stray counts or 10 B induced 7 Be. Thus, it appears that our beryllium (BeO and Be(NO₃)₂, Merck) has a 10 Be level similar to that found by Middleton *et al* (1984) for several other sources of commercial beryllium.

Efficiency

Our overall efficiency (10 Be detected per BeO $^-$ injected into Tandetron) has remained relatively constant at 5–6% throughout the past year. However, increased experience with the source operation has improved the BeO $^-$ output available. For a normal sample (prepared with 250 μ g of 9 Be carrier) we now typically obtain $1-2\mu$ A of BeO $^-$, although we have had as much as 4μ A. On occasion, when samples are not sufficiently pure chemically, we still must limit the BeO $^-$ current because of an intolerable (> 40,000 cps) 10 B count rate.

Data acquisition

Cycling between 9 Be and 10 Be, and data recording, are still being done manually. However, the Δ E-E spectra from the multichannel analyzer are now transfered to diskette by way of a micro-PDP11/23 computer. We plan to use this computer to automatize the data acquisition in the near future.

26 A

We have discussed (Raisbeck *et al* 1979; Raisbeck & Yiou, 1984) the motivation for measuring ²⁶Al by AMS, both in terrestrial and extraterrestrial matter. For example, measurements in marine sediments, in conjunction with ¹⁰Be, might provide a dating method insensitive to production variations (Lal, 1962), while measurements in cosmic spherules, again in conjunction with ¹⁰Be, can give information on the origin and irradiation conditions of these spherules (Raisbeck *et al*, 1983).

In considering the procedure to develop for measuring ²⁶Al with the Tandetron, we were influenced by the following factors: 1) the present voltage capabilities of the machine (~2 MV); 2) the desire to maximize the effi-

ciency through the machine (and thus to use an abundant charge state); 3) the advantages of using a prime number for the charge state, 4) the desire to retain the capability of making $\Delta E - E$ identification in the detector; 5) the desire to minimize the changes necessary between ¹⁰Be and ²⁶Al measurements. Taking into consideration the above factors, we decided to try to develop a technique using: 1) Al⁻ injection from Al₂O₃ samples from a General Ionex Model 860 source; 2) analysis of Al⁺³ at the output of the machine, and 3) retention of the mylar window and $\Delta E - E$ configuration of the detector. These choices, therefore, differ significantly from those adopted by Aardsma (1984) using a similar Tandetron. Using the Al⁺³ charge state, and the present current limits of the high-energy magnets, we

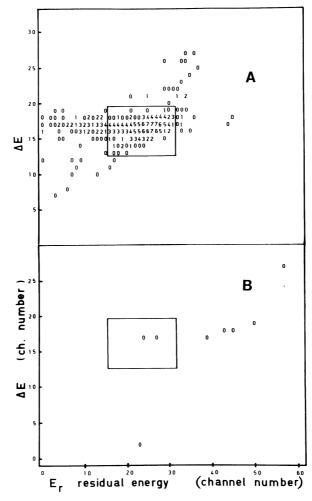


Fig 2. (A) Spectra of ΔE vs E_r (residual energy) for Al_2 O_3 standard having $^26Al/^{27}Al=4.87\times 10^{-10}$. The numbers in the spectra (n) indicate the number of events in a given channel (N) through the relationship $2^{n-1} < N \le 2^n$. The number of events in ^{26}Al window is 1009. (B) Spectra of blank Al_2O_3 giving $^{26}Al/^{27}Al \sim 7\times 10^{-14}$

were forced to limit our terminal voltage to a nominal value of 1.75 MV (we have not yet calibrated our GVM with a nuclear reaction). We calculated, however, that this would still permit $\Delta E - E$ identification in the detector.

Figure 2 shows a spectrum from an Al_2O_3 standard having $^{26}Al/^{27}Al = 4.87 \times 10^{-10}$. This Al_2O_3 was made by diluting a calibrated ^{26}Al solution, obtained from LMRI, Saclay, and transformed to the oxide. The ionization detector was operated at a pressure of 30 torr of 90% Ar–10% methane mixture, and 250 volts cathode voltage. The overall transmission, from Al^- injected into the machine to $^{26}Al^{+3}$ counted in the detector is ~10%. This is comparable to that obtained in a higher energy tandem (Middleton *et al*, 1983). Figure 2B shows an example of a spectrum from a blank Al_2O_3 . The background, which appears to consist of real ^{26}Al events, corresponds to an $^{26}Al/^{27}Al$ ratio of $\sim 7 \times 10^{-14}$. This is about an order of magnitude higher than observed at the University of Pennsylvania (Middleton *et al*, 1983). This blank run was made immediately after the standard at 4.87×10^{-10} . It is, therefore, quite possible that the background represents a memory effect, at a level of $\sim 10^{-4}$, quite consistent with that observed by Middleton *et al* (1983).

One of the main difficulties in measuring 26 Al by AMS in a tandem accelerator is the relatively poor emission for Al $^-$ ions. To date, the best current we have been able to obtain is ~ 100 nA. This is insufficient to permit measurements of 26 Al in terrestrial sediments, where the expected ratio is 26 Al/ 27 Al $\sim 10^{-14}$, without prior enrichment (Raisbeck & Yiou, 1981). However, the technique described above is capable of determining 26 Al in various extraterrestrial material, and we are presently using it to determine 26 Al in cosmic spherules.

CONCLUSION

A Tandetron accelerator mass spectrometry system operating at a terminal voltage of \sim 2 MV appears capable of measuring the isotopes ¹⁰Be and ²⁶Al with performances comparable to higher energy tandems.

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REFERENCES

Aardsma, G E, 1985, Accelerator mass spectrometry of ²⁶Al at 1.2 MV using charge state +2: Nuclear Instruments & Methods, vA238, p 170–172.

Lal, D, 1962, Cosmic ray produced radionuclides in the sea: Jour Oceanog Soc Japan, v 18, p 600–614.

Middleton, R, Klein, J, Brown, L and Tera, F, 1984, ¹⁰Be in commercial beryllium: Nuclear Instruments & Methods, v B5, p 511–513.

Middleton, R, Klein, J, Raisbeck, G M and Yiou, F, 1983, Accelerator mass spectrometry with ²⁶Al: Nuclear Instruments & Methods, v 218, p 430–438.

Raisbeck, G M and Yiou, F, 1981, Electromagnetic isotope separation as a method of improving the detection limits of accelerator spectrometry: Nuclear Instruments & Methods, v 186, p 483–486.

—————1984, Production of cosmogenic nuclei and their applications: Nuclear Instruments & Methods, v B5, p 91–99.

Raisbeck, G M, Yiou, F, Bourles, D, Lestringuez, J and Deboffle, D, 1984, Measurement of

¹⁰Be with a Tandetron accelerator operating at 2 MV: Nuclear Instruments & Methods,

New with a Tandetron accelerator operating at 2 MV: Nuclear Instruments & Methods, v B5, p 175–178.

Raisbeck, G M, Yiou, F, Klein, J, Middleton, R, Yamakoshi, K and Brownlee, D E, 1983, ²⁶Al and ¹⁰Be in deep sea stony spherules; evidence for small parent bodies: Lunar Planetary Sci, v 14, p 622–623.

Raisbeck, G M, Yiou, F and Stephan, C, 1979, ²⁶Al measurement with a cyclotron: Jour Physique-Lettres, v 40, p L241–L244.