160 keV ^{26}Al-AMS with a single-stage accelerator mass spectrometer

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ABSTRACT

Proof-of-principle ^{26}Al-AMS analysis is achieved with a single-stage accelerator mass spectrometer (SSAMS) utilising very low ion energy. The SSAMS operates by discriminating against atomic isobar interference in a negative ion source and suppressing molecules with thick gas stripper. Resulting 1+ ions counting is with a surface barrier detector. The NEC designed SSAMS for ^{14}C analysis is a popular model accelerator mass spectrometer and the developed further capability might be a significant addition to established ^{26}Al-AMS capacity. Measurements at these energies should also be sufficient for alternative ^{26}Al positive-ion mass spectrometry (PIMS).

1. Introduction

^{26}Al is a cosmogenic and anthropogenic radionuclide with diverse applications in geological, environmental and biological sciences [1–6]. Highly sensitive accelerator mass spectrometry (AMS) is able to detect ^{26}Al as low as 1 atom in 10^{16} total sample Al atoms, but the technique sufferers from the low electron affinity of aluminium. The resulting difficulty in producing large Al$^-$ currents from the caesium sputter ion sources used makes these measurements slow and expensive with limited accuracy.

The MV tandem accelerator mass spectrometers typically employed in ^{26}Al measurements will also be capable of ^{10}Be detection. This measurement of in situ produced terrestrial cosmogenic radionuclides is normally preferred because of the correspondingly larger ions beams that can be made from common sample material. Nevertheless Al-AMS is needed in additional to Be-AMS to understand complicated exposure history, and for the other applications. Accordingly, developing aluminium AMS on small radiocarbon spectrometers could promote Al-AMS and it applications by expanding the installed base of capable instruments, with implications for analysis cost reductions.

Carbon and aluminium AMS rely on negative-ion atomic isobar suppression. The ^{14}N and ^{26}Mg interferences to ^{14}C and ^{26}Al do not form stable anions and are suppressed in the negative ion source. The remaining molecular interference can in principle be removed with relatively little energy in a compact system. Our single-stage accelerator mass spectrometer (SSAMS) destroys molecular

interferences to ^{14}C with ion energy of <300 keV, and such low energy ^{26}Al-AMS has now been demonstrated too.

2. Experiments

Fig. 1 is of the SUERC SSAMS featuring a caesium sputter ion source which accommodates a carousel of up to 134 prepared samples including reference standards, etc. The source is biased to produce negative ions of energy up to 45 keV at ground potential. The resulting ion beam is momentum analysed by a 90° double focusing bending magnet rated to a mass energy product of 1 (a.m.u. MeV) and injected into the accelerator. The magnet vacuum manifold is electrically isolated to allow a switchable bias, known as the magnet bias sequencer (MBS), to be applied. The MBS locally increases ion energy such that lower-mass isotopes can match the rigidity of higher-mass atoms and can selectively be injected into the accelerator in turn. An off-axis multi-channel Faraday cup is aligned on the inside radius of the magnet to measure the ^{13}C and ^{12}C during the ^{14}C and ^{13}C MBS injection cycles respectively. The following accelerator is a single-stage, air insulated, 250 kV deck that houses all of the high ion energy diagnostics at potential. Once there the negatively charged ions enter a 1 m long stripper canal with a 1 cm inside diameter that is at a relatively high pressure, or gas thickness, compared to those in high-energy accelerators, using nitrogen with a input flow rate of 0.85 sccm. The thick stripper gas destroys molecules through multiple low energy collisions but only significantly produces a 1+ charge-state ion beam [7]. Post stripper ion beam analysis is with a second 90° double focusing magnet rated to a mass energy
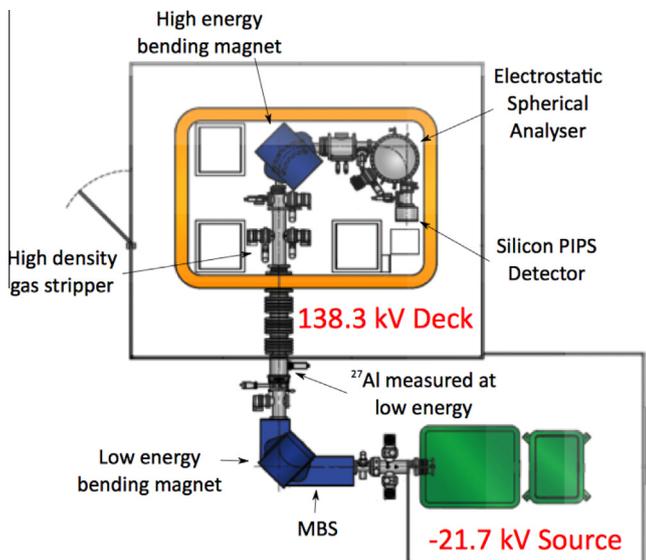
Fig. 1. Schematic of the SSAMS used for ^{26}Al-AMS (courtesy of NEC).

product of 5, to separate out the fractions of a molecule from the beam of interest. Two further off-axis Faraday cups are aligned to measure the ^{12}C and ^{13}C ions on the inside radius of the magnet. The rare isotope is transmitted on-axis to a 90° pre-detector electrostatic spherical ion energy analyser (ESA) and then, through a bounced deflector (SPAD) to the detector. The SPAD stops scattered particles damaging the detector during the stable isotope MBS injection periods. The detector is a silicon Canberra PIPS detector.

To measure aluminium the SSAMS was modified from its standard carbon set-up. ^{14}C is the maximum mass the dipole magnets are designed to transmit at full energy with the current power supplies and so the energy of the ion beam was approximately halved on both the low-energy and high-energy side of the spectrometer in order to transmit aluminium ions, which are almost twice as massive as carbon atoms. Samples were biased to −6.5 kV within the ion source and the source biased to 15.2 kV from ground for a total accelerator injection energy of 21.7 kV. The ion extraction cone in front of the sputtered sample surface was biased to 19.8 kV for efficient ion collection, but this did not add to the total beam energy in the configuration employed. The source typically produced ∼1 μA of ^{27}Al$^-$. Similarly, the deck potential was required to be reduced to 138.3 kV, which gave the aluminium ions a total energy of 160 keV. This was the maximum energy of ^{27}Al that the high-energy magnet could transmit to an off-axis cup. The beam focusing of the acceleration tube was poor at this reduced potential resulting in decreased ion optic transmission. In addition, the stable isotope of aluminium, ^{27}Al, is heavier than the rare isotope and therefore would ordinarily be measured by an off-axis Faraday cup on the outside of the magnet radius, which the SSAMS does not have either at ground or on the high-voltage deck. Therefore the first two time-states of the MBS were used to inject ^{26}Al and ^{27}Al into the accelerator in repeated sequence and a third state pulled the ^{27}Al into the low-energy off-axis cup on the inside of the injection-magnet radius. The MBS was used to generate the ratio of high-energy ^{26}Al to low-energy ^{27}Al as the measure of high-energy to low-energy stable isotope ratio was stable. Several Al_2O_3 samples mixed with silver in 1 part Al_2O_3 to 2 parts Ag were used to tune and test the new configuration. Three Nishiizumi standard material (Al01-4-1, Al01-4-2 and Al01-5-3) [8] and commercial blank Al_2O_3 from Fisher and Assure were normalised to Purdue Z92-0222 primary standard (nominally ^{26}Al/^{27}Al = 4.11 × 10^{-11}).

3. Results

Repeated sample measurements were consistent, which limited sample measurement precision to Poisson counting statistics. The two secondary standard materials, Al01-4-2 and Al01-5-3, were each measured as multiple samples and their distributions are shown in Fig. 2a and b respectively. These show a normal or Gaussian distribution due to statistical variation, with no systematic error or addition scatter beyond the average measured error. Table 1 is a table of the average results of several samples for the three types of standard material used normalised to the primary standard. All of the results are within one standard deviation of the nominal values.

The under-gradient acceleration tube limited the focusing of the ion optics and the reduced velocity ion interaction with the thick stripper gas resulted in a combined accelerator and stripper ion transmission of only ∼1%. The low aluminium current, although high by AMS standards, and the low transmission resulted in low counting statistics and relatively large error for the low ratio standard, Al01-5-3. The solid-state detector employed has a higher
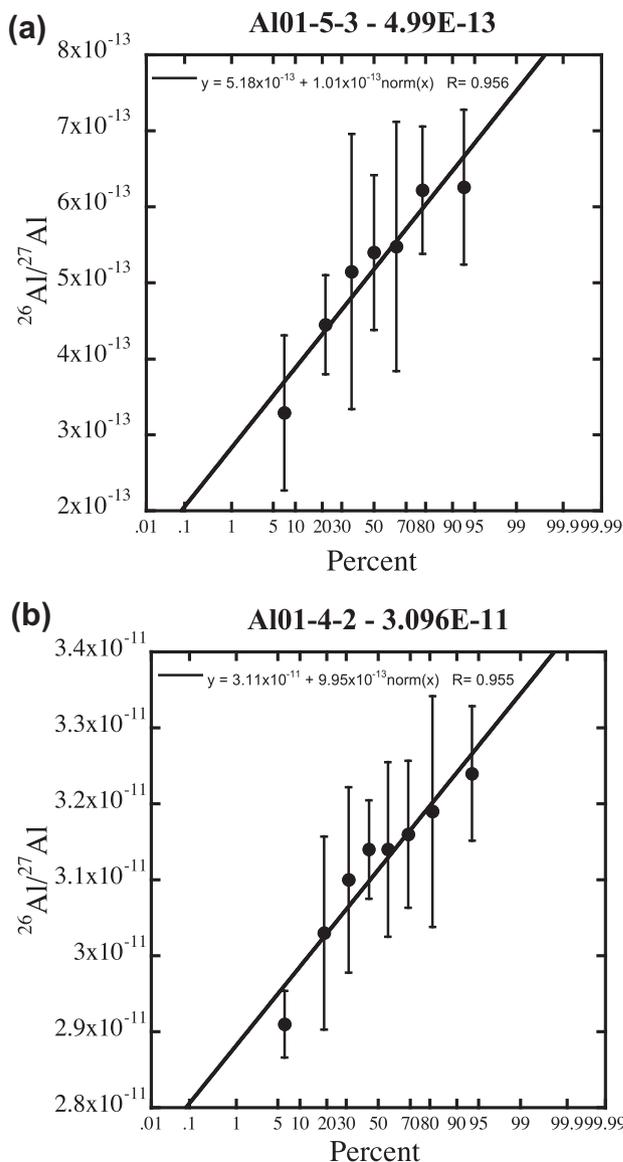

Fig. 2. Probability distribution plots of standard material (a) Al01-5-3 and (b) Al01-4-1 measured at 160 keV on the SSAMS.

Table 1
Table of the measured Al standard material.

Nishiizumi standard	Nominal $^{26}Al/^{27}Al$ ratio	Number of samples	Measured $^{26}Al/^{27}Al$ ratio	Error (%)
Al01-4-1	7.44×10^{-11}	3	7.48×10^{-11}	1.5
Al01-4-2	3.096×10^{-11}	8	3.11×10^{-11}	3.2
Al01-5-3	4.99×10^{-13}	7	5.18×10^{-13}	23

electronic noise level than conventional gas ionisation detectors and dark count measured in the detector (when the detector is blocked by a Faraday cup and no real signal can register counts) is 1.8 ± 0.3 (std) counts per minute. The PIPS detector registers the dark counts regardless of the deck potential and any alternative detectors would need to be well shielded against the noisy environment on the deck. A 1 μA primary standard produces 155 counts per minute and therefore the dark count correction is only 1%. The count rate for a low ratio standard, such as the Al01-5-3, is almost two orders of magnitude lower than the Purdue and therefore a dark count correction of around 50% is required. This is significant for the background and low ratio standard and must be corrected for and further experiments would require a low noise detector. Background samples produced only one or two counts after dark count correction and set an upper limit to the background measurement of mid/high 10^{-14}. The experiment background is ultimately limited by the low ion transmission that in principle could be addressed by improving the ion optics with stronger dipole magnets for full acceleration of the aluminium beam, etc. The results in Fig. 2 and Table 1 are dark count corrected.

Conventional high-energy high-charge state Al-AMS can be subject to interference from other mass elements with the same magnetic rigidity, such as ^{13}C, ^{9}Be, ^{17}O, ^{35}Cl and ^{37}Cl [9]. The use of the single positively charged $^{26}Al^{1+}$ makes it impossible for any of those interferences to be transmitted to the detector. The primary source of interference for a compact system utilising a charge state <3+ is molecules surviving the interaction with the stripper. The stripper thickness can be increased to ensure sufficient interaction to destroy all molecules. This will however increase the scattering and associated losses. The balance between molecular destruction and transmission is yet to be found and would be better done on a system optimised to transmit aluminium. Moreover, Al-AMS will be immune to another SSAMS issue. Compact low-energy spectrometers are sensitive to interference from their stable isotope [10,11] injected as a hydrogenated molecule. In carbon AMS, for example, the mass 14 ^{13}CH molecule is injected into the accelerator. A small percentage of the ^{13}C from the molecule is scattered in the stripper canal or in the residual stripper gas in the high-energy magnet and transmitted to the detector. However the hydrogenated stable aluminium isotope, ^{27}AlH, is too heavy to be injected into the spectrometer because the stable isotope is already heavier than the rare isotope, therefore this interference is not possible with aluminium.

4. Discussion

In order to fully utilise the compact spectrometer for aluminium, it will be necessary to up-grade some of the components. A high-energy off-axis Faraday cup on the outside of the magnet's radius will be required to measure the high-energy $^{26}Al/^{27}Al$ ratio. In addition, stronger dipole magnets on the high and low energy side of the spectrometer will be required to improve the ion optics and ion beam stripper interaction and ultimately yield higher transmission. If the maximum potential of the SSAMS deck is utilised along with the source potential, the aluminium could gain

an energy of 350 keV. Wittkower and Betz [12] shows that Al^{1+} yield increases with decreasing energy and at 400 keV in nitrogen a yield of 47% should be expected. Measurements on the ETH Tandy have achieved a transmission of 25% of the 1+ charge state at 460 keV in argon [13]. The losses to below the expected yield are likely due to scattering. However, transmission typically increases when the stripper gas is changed to He, with losses due to scattering almost halving for some species [14] (although data for aluminium is not present). Transmission data for all the parameters above combined does not yet exist but a conservative estimate of the transmission of aluminium on the SSAMS at full energy should be >20%. Even 10% transmission combined with the 1 μA $^{26}Al^-$ output achieved with our ion source would provide a competitive measurement time compared to other systems.

In addition to measuring ^{26}Al at low energies from a conventional negative-ion sputter source, it should also be possible to measure aluminium from a plasma source that produces a positive ion beam and remove the ^{26}Mg interference by converting the positive ion beam to a negatively charged ion beam away from the ion source. The SUERC SSAMS has been used to demonstrate this principle with radiocarbon detection from a positive ion source at low energy [15]. The SUERC SSAMS has recently been up-graded with a low-energy, 45°, rotatable ESA, which allows two ion sources to be attached to the injector [11] to pursue this alternative scheme. The additional ion source on the SSAMS is a Pantechnik electron cyclotron resonance (ECR) Nanogan, which can create positive ion beams of multiple charge-states. In the case of radiocarbon measurement molecular interference was partially removed by selecting the $^{14}C^{2+}$ from the ion source and the remainder destroyed in a charge exchange cell. The nitrogen interference was removed by extracting the negative carbon ion from the charge exchange cell. This process should also work for the suppression of ^{26}Al interferences and take advantage of the high efficiency ECR source capable of producing much higher beam currents than a conventional caesium sputter ion source. Fast and efficient measurements with increased precision will expand the scope of ^{26}Al application and potentially provided an improved alternative to ^{10}Be analysis.

5. Conclusion

The SUERC SSAMS has been used to demonstrate ^{26}Al AMS at 160 keV. Standards measurements have shown reliable ratios. Backgrounds are limited by the low transmission ion optics but show no significant signal above detector noise. Up-grades to the ion optics should result in improved transmission and provide a compact, low cost alternative to large conventional tandem accelerators for aluminium AMS.

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