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^{10}Be ANALYSES WITH A COMPACT AMS FACILITY—ARE BeF_2 SAMPLES THE SOLUTION?

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ABSTRACT. The injection of $^{10}BeF^-$ instead of $^{10}BeO^-$ into a compact accelerator mass spectrometry system with a terminal voltage of 0.58 MV was investigated, because BF^- molecules are unstable and isobaric interference of ^{10}B with ^{10}Be can thus be significantly reduced. We describe the method we developed to prepare BeF_2 samples. ^{10}Be was measured in a segmented gas ionization detector. Separation of ^{10}Be from ^{10}B could be achieved both for ions in the 1+ charge state with an energy of 0.8 MeV and in the 2+ charge state with an energy of 1.4 MeV. The 2+ ions are better separated, whereas the 1+ charge state has a higher transmission. $^{10}Be/^{9}Be$ ratios ($\sim 10^{-12}$) in a suite of rock samples were successfully determined for exposure dating in either charge state and compared with measurements made on the 6MV tandem.

INTRODUCTION

The main difficulty in ^{10}Be accelerator mass spectrometry (AMS) is the high intensity of the isobaric ^{10}B beam (Table 1) when samples are prepared as BeO. With a compact AMS facility operating at a terminal voltage of ≤1 MV, the energy of the ions is too low to efficiently remove ^{10}B using the foil stack method (Grajcar et al., forthcoming) or a gas absorber before the final detector. However, if $^{10}BeF^-$ is injected instead of $^{10}BeO^-$, the ^{10}B intensity is significantly reduced, allowing particle identification in a gas ionization detector (Zhao et al. 2002). This reduction is due to the lifetime of the electronic ground state of BF^- being much less (<1 fs) than the roughly 10 μs needed for the negative ions to reach the terminal stripper. Though exited electronic states of BF^- may have longer lifetimes, the total boron rate is still reduced significantly (Deuw et al. 2002).

The aim of this work was to find a method to prepare samples which would produce high BeF^- currents in a Cs sputter ion source, and to investigate the potential of a small AMS facility for ^{10}Be measurements using BeF_2.

METHODS

Sample Preparation

Aqueous ^{10}Be standard and blank solutions containing 0.1–0.5 mg of beryllium as nitrate or chloride were dried and 100 μL of a 1% silver solution ($AgNO_3$) was added to increase the bulk for easier sample handling. A solution of 2 mL of 40% HF was added and evaporated at 100 °C until dry to produce fluorides; this step was repeated twice. The samples were then baked at 420 °C for 90 min to reduce the silver to its metallic form. The dry BeF_2 with silver was then stored under vacuum in a desiccator because BeF_2 is very hygroscopic. The samples were mixed with 4 mg of niobium powder and then pressed into Al sample holders.

Niobium was added to improve electrical and thermal conductivity. These samples produced readily high BeF^- currents and had low ^{10}B counting rates in the detector, compared with other metals, such as silver, iridium, palladium, copper, and iron, which were also tested.

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In addition to blanks and standards, we prepared rock samples for measurement on both our small 0.6MV and the 6MV AMS facility for a comparison. The chemical isolation of the Be from quartz after spiking with ^{9}Be carrier was performed according to Ochs and Ivy-Ochs (1997). Those samples were then split into 2 fractions. One fraction was used to make BeO targets and the other for BeF_2 samples, as described above for the standards and blanks.

Measurement

The BeF_2 samples were measured using the compact PSI/ETH Tandy AMS system (Synal et al. 2000; Stocker et al., forthcoming; Grajcar et al., forthcoming; Döbeli et al. 2002). The BeF^- molecules, which were injected into the accelerator operating at the maximal terminal voltage of 0.58 MV, were dissociated in argon stripper gas. Both Be^+ and Be^{2+} ions were analyzed. ^{10}Be was measured in a gas ionization counter filled with isobutane. A 50-nm silicon nitride foil was used as a detector window (Döbeli et al. 2002), which allows the ion identification even at very low energies (Grajcar et al., forthcoming). An energy loss (ΔE) and a residual energy (E_{Res}) signal were derived from 2 anodes.

RESULTS

Targets made from commercial BeF_2 and the BeF_2 targets made according to the sample preparation procedure above, both mixed with Nb powder, gave BeF^- beam currents of up to 0.2 μA. The ^{10}B-counting rate was typically at the level of 10^{-9} to 10^{-10} relative to the ^{9}Be rate (Table 1), which is consistent with Zhao et al. (2002). This corresponded to counting rates of less than 1 kHz in the detector for typical beam currents, low enough to not degrade the energy resolution of the detector.

Table 1 Comparison of the 0.6 and the 6MV AMS facilities. In the present configuration of the compact facility, the ^{10}Be beam is not well focused into the detector, resulting in a much lower ^{10}Be transmission compared to ^{9}Be.

	BeF_2 with 0.6 MV		BeO with 6 MV
	1+	2+	3+
Extracted ions	BeF^-		BeO^-
LE current (nA)	100–200		2000–4000
^{10}B/^{9}Be (HE side)	10^{-10}–10^{-9}		10^{-4}
^{10}B (Hz)	100		10^{8}
Charge state	1+	2+	3+
Ion energy (MeV)	0.8	1.4	19
^{9}Be transmission	50%	12%	17%
^{10}Be transmission	25%	4%	15%

The isobars ^{10}Be and ^{10}B have different stopping powers, and so can be separated by their ΔE–E signals in the ionization chamber. Figure 1 shows a 2-dimensional spectrum for ions in charge state 2^+ (E = 1.4 MeV). A standard material with a ^{10}Be/^{9}Be ratio of about 10^{-10} was used. The ^{10}B peak of similar intensity is well separated. A third peak of similar height was found between the ^{10}B and the ^{10}Be peak when the stripper gas was optimized for the highest transmission of ^{10}Be. This peak can be attributed to $(^{9}BeH)^{2+}$ molecules surviving all the way into the detector. This demonstrates the potential of this gas ionization detector for the identification of molecular ions, with AMS at low energies, where molecules in charge state 1+ or 2+ can survive the stripping process. By increasing the gas pressure in the stripper to about 1 μg/cm^2, the peak in the 2+ charge state spectrum disap-

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pears, e.g., most of molecules have been destroyed. At this higher pressure, the ^{9}Be transmission is reduced by about 10% due to scattering processes in the stripper gas. The same gas pressure was sufficient to destroy molecules in the 1+ charge state. We did not find a significantly higher rate of 1+ molecules, though one would expect them to be formed more easily. All further measurements were made at the higher stripper pressure.

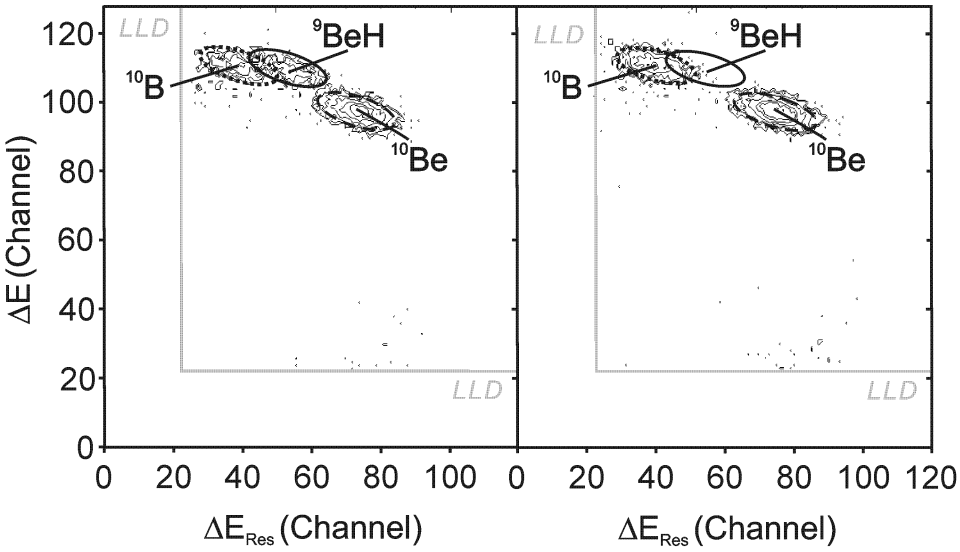

Figure 1 Left: spectrum of a ^{10}Be standard in charge state 2+ and for a stripper gas pressure of 0.3 µg/cm^2 (optimum for maximal transmission of ^{9}Be+). A $(^{9}BeH)^{2+}$ peak shows between $^{10}B^{2+}$ and $^{10}Be^{2+}$. The $(^{9}BeH)^{2+}$ can be eliminated if the gas pressure is raised to 1 µg/cm^2 (shown on the right).

In the upper part of Figure 2, the 2-dimensional spectrum of the same standard material is compared with one of a geological sample. In this case, the detector was optimized for the best Be-B separation. The peaks are well separated, with peak widths (FWHM) of about 50 keV. A suppression of about 5 orders of magnitude can be obtained with a peak integration of 90%. A similar peak separation has been shown in previous experiments with BeO beams (Grajcar, forthcoming). Measurements with blanks samples gave values of 4×10^{-14}, i.e., higher than the result from the BeO measurements using the large tandem accelerator (Table 2) and higher than what one would expect from a tail of the boron peak. This hints at some other background source. Further experiments are needed to identify the cause for this background.

In the lower part of Figure 2, the spectra of the same samples are shown when analyzing ^{10}Be in charge state 1+ (E = 0.8 MeV). The peak widths are about the same, but the peak separation is about a factor of 2 smaller. This allows still some ^{10}Be-^{10}B separation, but a reasonable background suppression can only be obtained with narrow and asymmetric software gates for ^{10}Be as shown in Figure 2. Then, only about 60% of the real ^{10}Be were integrated and the blank values were still at a level of 10^{-13}. The ^{10}B contribution to the background is now negligible. Setting the gates tighter does not improve the background.

The ^{9}Be transmission given in Table 1 has been determined from current measurements on the low-energy side and after the high-energy magnet. The high transmission of about 50% for charge state 1+ reflects the high stripping yield obtained at the terminal voltage of 580 kV (corresponding to

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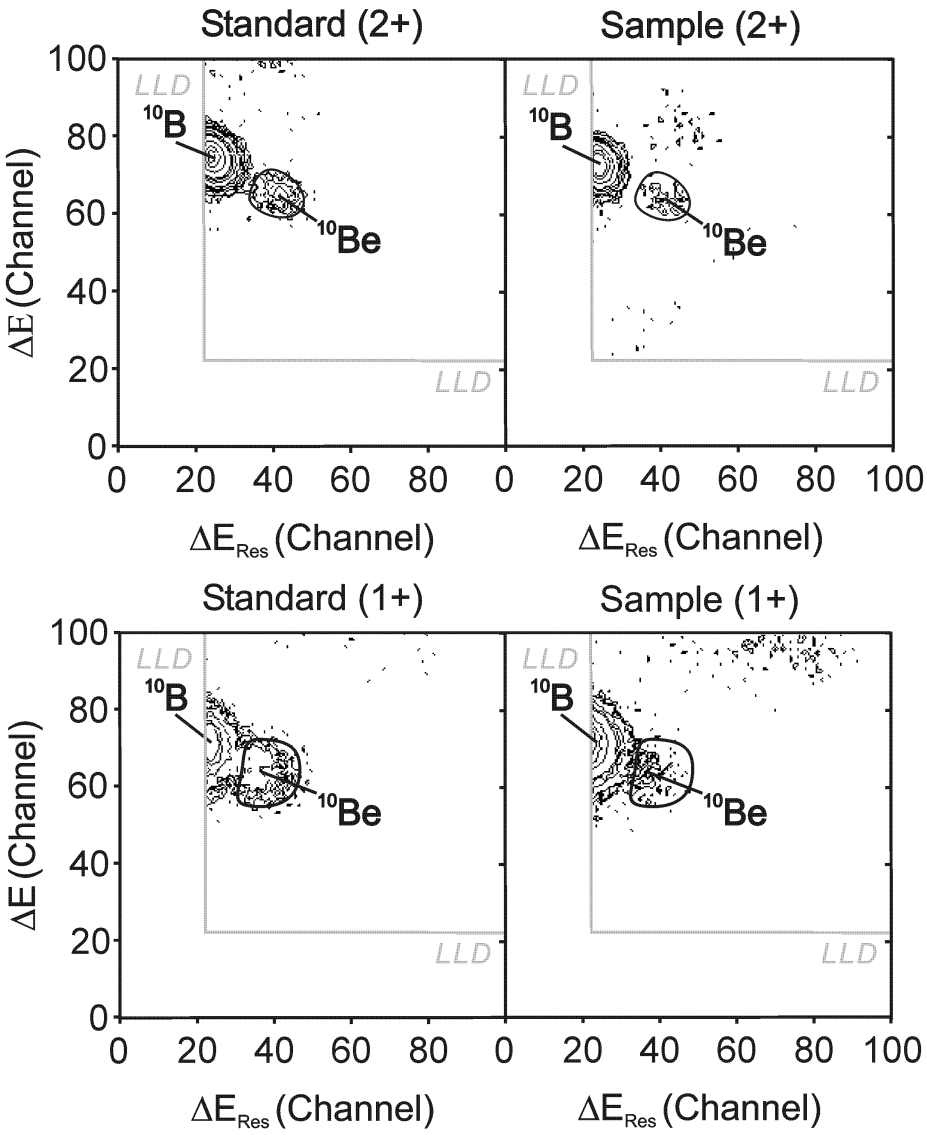

Figure 2 Two-dimensional spectra of ΔE versus ΔE_{Res} for a standard ($^{10}Be/^{9}Be = 10^{-10}$) and a geological sample. The 1.4-MeV $^{10}Be^{2+}$ is clearly separated from $^{10}B^{2+}$. The 0.8-MeV $^{10}Be^{+}$ is only partially separated from $^{10}B^{+}$. Events within the black contour lines were accepted as ^{10}Be events while suppressing ^{10}B by a factor of more than 10^4. The grey lines show the low-level discriminator settings to cut off electronic noise.

205 keV for ^{9}Be when injecting BeF^{-}) and the large acceptance of the high-energy Faraday cup. For charge state 2+, the stripping yield is about a factor of 4 smaller. About a factor of 2 is lost from there to the detector for 1+ and about a factor of 3 for 2+ ions.

The normalized ^{10}Be/Be ratios of the rock samples are consistent with those obtained with the 6MV AMS facility, for which a description can be found in Synal et al. (1997).

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Table 2 Results for geological samples measured on the 6MV Tandem and on the 0.6MV Tandy accelerator. Each rock sample was spiked with 0.5 mg ^{9}Be and was split after Be separation to produce a BeO and a BeF_2 target. The BeF_2 sample was measured both in the 1+ and the 2+ charge state.

	Tandy (1+), BeF_2		Tandy (2+), BeF_2		Tandem (3+), BeO	
	^{10}Be/^{9}Be		^{10}Be/^{9}Be		^{10}Be/^{9}Be	
Sample	10^{-12}	Error	10^{-12}	Error	10^{-12}	Error
S 1	5.1	12%	5.1	11%	5.06	3%
S 2	1.8	21%	not measured	not measured	1.56	4%
S 3	8	17%	7.5	26%	9.75	2%
S 4	0.5	31%	0.8	41%	0.55	4%
Blank	0.09	14%	0.04	70%	0.01	24%

DISCUSSION

BeF_2 targets of good quality can be prepared with reasonable effort using the described procedure. The hygroscopic nature of the fluorides requires the storage of the samples in a dry environment. This is inconvenient; on the other hand, BeO is more toxic. The beam currents from BeF_2 samples are still clearly lower than those obtained from BeO; however, the ^{10}B rate is significantly reduced. Remaining ^{10}B entering the detector is likely to be injected as $(^{10}B^{18}O^{1}H)^{-}$, $(^{10}B_2{}^{9}Be)^{-}$, or $(^{10}BF)^{-}$ in a meta-stable exited electronic state.

The high stripping yield of 1+ ions makes this charge state very attractive, leading to an overall transmission higher than at most existing AMS facilities. The optics system for charge state 1+ is relatively easy to improve by positioning the detector in the focal plane of the high-energy mass spectrometer and by increasing the size of the detector window, which is currently only 5 mm × 5 mm. For the charge state 2+, the situation is more complex. The high-energy accelerator tube acts as a stronger lens, leading to a convergent beam at the exit of the accelerator. To overcome this problem, a more complicated ion optics system for proper focusing would be needed.

The blank ratios are still about 1 order of magnitude higher than can be achieved at larger facilities, but a further background reduction seems to be feasible. The energy resolution of the detector can be improved, as systematic studies clearly showed that the electronic noise is presently the limiting factor for light ions. Signals from a pulser fed directly into the preamplifier showed only a 10% reduction in resolution, indicating that the noise of the preamplifier represents the main contribution. Energy straggling in the entrance window and the gas are significantly smaller. Based on preamplifier specifications, one can expect a further improvement with an appropriate design of the detector and the electronics and by the appropriate choice of the counter gas. A reduction of the peak width of 20–40% does not seem unrealistic. This should provide a reasonable ^{10}Be-^{10}B separation for the 1+ charge state.

CONCLUSION

We have demonstrated that BeF beams are a means of permitting ^{10}Be AMS at low energies. For the first time, environmental samples have been measured with a compact AMS operated at 580 kV. The results are promising, but at the moment, the method is not competitive with that used at larger facilities. There is potential for improvement and it appears feasible that a facility can be developed meeting the required specifications for applications in Earth Sciences.

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