ARE COMPACT AMS FACILITIES A COMPETITIVE ALTERNATIVE TO LARGER TANDEM ACCELERATORS?

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ABSTRACT. In the last decade, small and compact accelerator mass spectrometry (AMS) systems became available operating at terminal voltages of 1 MV and below. This new category of instruments has become competitive for radiocarbon detection to larger tandem accelerators and many of these instruments are successfully used for ^{14}C dating or biomedical applications. The AMS group at ETH Zurich has demonstrated that small instruments can be built, which allow measurements also of other radionuclides such as ^{10}Be, ^{26}Al, ^{129}I, and the actinides. ^{41}Ca measurements can be performed with sufficient sensitivity for biomedical applications. A summary of recent developments made at the 500kV Pelletron in Zurich is given and its performance is compared with that of a commercial compact instrument from the company High Voltage Engineering Europe (HVEE) in Amersfoort, the Netherlands, operating at 1MV at CNA in Seville, Spain, as well as with that of larger AMS facilities. It turns out that the ion optics, stripper design, and the detection system are critical for the performance.

INTRODUCTION

It is our dream to have smaller, simpler, and reliable accelerator mass spectrometry (AMS) facilities at low cost. At the same time, high performance (low background, high yield, and high precision) and the flexibility to measure many different isotopes is desirable. The construction of compact AMS facilities started about 13 yr ago with several proposals for dedicated instruments for radiocarbon measurements (Hughey et al. 1997a,b; Mous et al. 1997; Suter et al. 1997) based on the concept that interfering molecules in charge state 1+ and 2+ can be destroyed by collisions with the stripper gas (Litherland 1984; Lee et al. 1984). These developments were driven especially by the needs of biomedical applications requiring a large sample throughput at low cost (Vogel et al. 1990; Purser 1994). The instrument then built by the ETH/PSI group in Zurich, Switzerland, in collaboration with National Electrostatics Corporation (NEC) in Wisconsin, USA, demonstrated that competitive ^{14}C dating is possible with accelerators operating at a terminal voltage as low as 500 kV, selecting ions in charge state 1+ for the final analysis (Suter et al. 1999; Synal et al. 2000a,b). The first experiments with this system clearly showed that interfering molecules of mass 14 such as $^{13}CH^+$ and $^{12}CH_2^+$ can be reduced by about 10 orders of magnitude in collisions with the target atoms in the terminal stripper gas (Jacob et al. 2000). This was an essential step forward to AMS at low energy. Meanwhile, we could demonstrate that molecules can be sufficiently destroyed at even lower energies (~200 keV). Based on these results, a smaller instrument (2.5 × 3 m^2) has been developed providing high-stability, high-reproducibility ^{14}C measurements with low background and a transmission through the accelerator of more than 40% (Synal et al. 2004, 2007; Wacker et al. 2010). The results obtained with this instrument demonstrated that compact AMS systems are competitive with many larger instruments for ^{14}C dating. Commercial instruments are now on the market based on our developments. NEC offers a compact AMS system based on a 0.5MV Pelletron accelerator (Goslar et al. 2004; Roberts et al. 2004) and a single stage AMS facility equipped with an air-insulated high-voltage platform (Klody et al. 2005; Skog 2007). HVEE has developed a compact AMS system based on a 1MV Tandetron accelerator to analyze ^{14}C in charge state 2+. This flexible instrument has been designed for measurements of light and heavy radioisotopes (Klein et al. 2006, 2007; Chamizo et al. 2008).

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Speculations whether a low-energy ^{14}C AMS system based on a tandem accelerator could be also used for the detection of other radionuclides were already made before the first compact instruments were available (Suter 1998). The ETH compact AMS facility TANDY operating at voltages up to 600 kV was primarily built to demonstrate the potential for ^{14}C dating, but it was designed to provide a universal platform to explore the potential of small AMS instruments for the analysis of other radioisotopes. The analyzing magnets at the low-energy (LE) and high-energy (HE) side provide magnetic fields high enough to bend also heavier isotopes of interest (Figure 1). Soon after the demonstration of successful ^{14}C measurements, tests with other radioisotopes provided promising results. Based on further systematic studies, the instrument has been significantly improved. Meanwhile, the feasibility for measurements has been demonstrated for ^{10}Be, ^{14}C, ^{26}Al, ^{41}Ca, ^{129}I, and actinides (Fifield et al. 2004; Stocker et al. 2004, 2005; Suter 2004; Wacker et al. 2005; Grajcar et al. 2007; Suter et al. 2007; Müller et al. 2008).

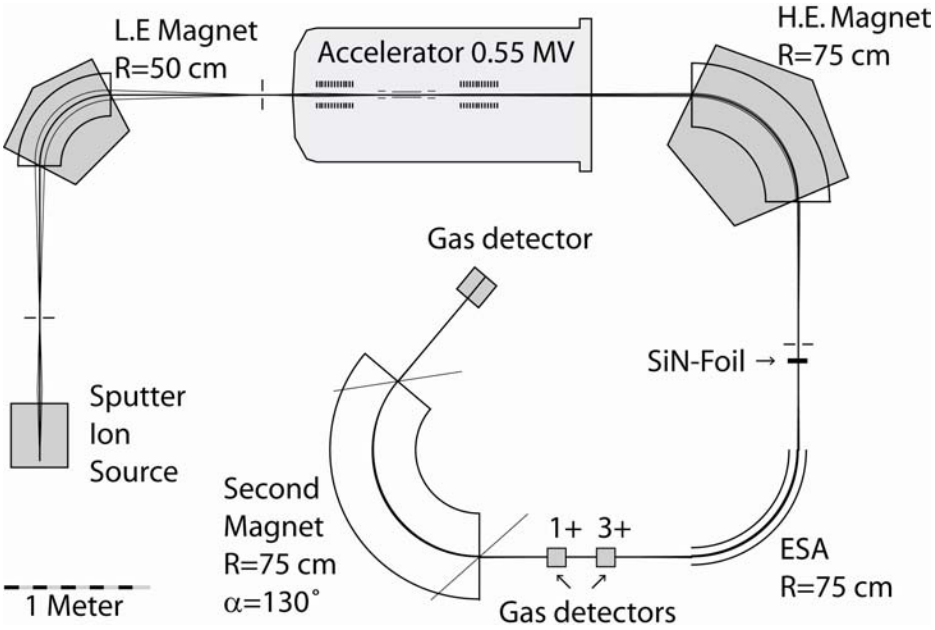

Figure 1 Layout of the compact AMS facility TANDY at ETH with the recently installed second HE magnet

In this paper, the basic challenges for designing a flexible low-energy AMS system for light and heavy ions are discussed. A brief description of the ETH compact facility TANDY is given. Its present performance is discussed and compared to that of the 1MV facility from HVEE (Klein et al. 2006, 2007; Chamizo et al. 2008) and to that typically obtained at larger AMS instruments. Finally, conclusions and ideas for improvements are presented.

INSTRUMENTATION

The basic arrangement of the new compact instruments is similar to larger AMS facilities where a ratio of the radioisotope counting rate (detector) to the stable (or abundant long-lived) isotope ion current (Faraday cup) is measured. The advantage of the new generation of compact AMS facilities is that investment and operating costs are significantly lower than at larger facilities. In addition, these instruments occupy less space and can therefore be placed inside regular-sized buildings with regular-sized laboratories (Figure 2).

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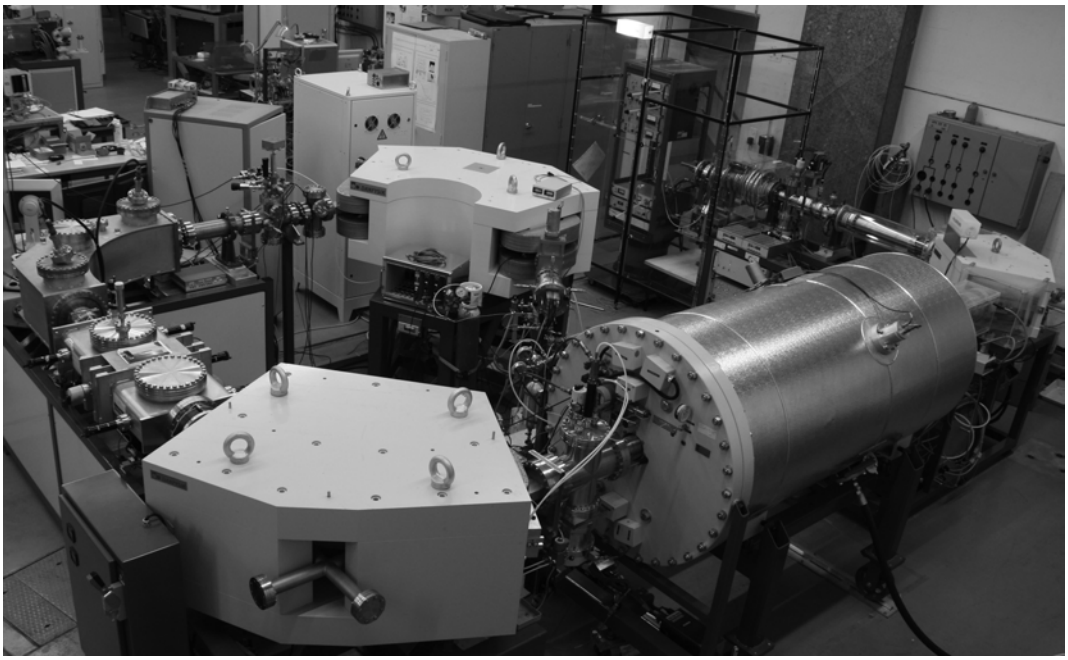

Figure 2 Compact AMS facility TANDY at ETH showing the recently installed second HE magnet

The original design of the compact AMS instrument TANDY at ETH was made before all the information on stripping yields, scattering losses, and destruction of molecular were available. Also, the contributions of the various background sources were not known at that time. So the first design idea was to build a compact and simple instrument for ^{14}C dating with the option to explore the potential of low-energy AMS over the whole mass range (Suter et al. 2000; Synal et al. 2000b). To keep the instrument simple, no lenses were installed except in the ion source. All further focusing is provided by the accelerator tubes and by the electric and magnetic deflection units. For proper focusing of the beam into the stripper canal, the source potential has to be about 10% of the terminal voltage. The facility is equipped with an ion source from NEC (40-sample multicathode MC-SNICS). The performance of this source was sufficient to explore the potential of AMS at low energies, but the currents were not high enough to perform competitive measurements for radionuclides such as ^{10}Be and ^{26}Al. Therefore, the source was upgraded in the last year to provide higher beam currents based on the experience of our extensive ion sources developments and those from other AMS facilities. First tests yielded significantly higher beam currents. The 90° magnet at the LE side (r = 50 cm) was available from other research programs at PSI. It has a mass-energy product of 5.9 amu MeV/e^2, which allows bending all light ions at full source energy (60 keV) and heavy ions such as iodine and plutonium at energies of 45 and 23 keV, respectively. The accelerator is of the Pelletron type from NEC with a nominal maximal terminal voltage of 500 kV. The high voltage is stabilized with a generating voltage meter (GVM), which controls the charging system of the Pelletron. It can be operated up to 600 kV and stable measurements are possible up to about 550 kV. The accelerator is a prototype and not identical to the design of compact ^{14}C dating facilities offered by NEC. Our prototype has 1 charging chain system on the high-energy side, whereas the commercial compact ^{14}C dating facilities have it at the low-energy end. Also, the ion optics and the pumping systems are different from instruments manufactured by NEC. The stripper is equipped with 2 turbomolecular pumps in the terminal (details in Jacob et al. 2000). It is designed to transmit particles with angles of up to

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about 14 mrad. The HE 90° magnet with a mass energy product of 33 amu MeV/e^2 can analyze light ions in charge state 1+ at full energy and 3+ ions up to ^{129}I. Actinides in the 3+ charge state can only be analyzed at a reduced terminal voltage of ~300 kV (Fifield et al. 2004). For 1+ ions, the HE magnet together with the subsequent electrostatic analyzer (ESA) forms an energy focusing system to compensate the beam energy spread caused by the energy-loss straggling in the stripper gas and for fluctuations in the terminal voltage. The whole system has been configured for negative atomic ions stripped in charge state 1+. Due to the focusing properties of the high-energy acceleration tube, the focal planes for ions in charge state 3+ are ~21 cm closer to the magnet and ~29 cm closer to the ESA, respectively (Stocker et al. 2005). These distances turn out to be practically identical for Be^{1+} ions injected as BeO^- molecules. In order to detect the 3+ ions and also ^{10}Be from BeO molecules properly in the corresponding focal plane, special removable detectors were developed (Stocker et al. 2004), which can be placed in the appropriate focal plane 1+ and 3+ (Figure 1). The detector entrance window consists of a thin silicon nitride foil with a thickness of 30 to 100 nm, depending on the size of the window and the detector gas pressure (Döbeli et al. 2004; Stocker et al. 2005). Originally, ORTEC preamplifiers were used for our gas detectors, resulting in electronic noise of ~32 keV FWHM (calibrated using the pulse height of protons). The noise was significantly reduced (FWHM: 15–16 keV) by mounting CREMAT preamplifiers directly onto the anode plates (Stocker et al. 2005). A compact detector design has at least 2 advantages: a short drift distance for the electrons also reduces their diffusive movement leading to a better localization of the electrons between the anodes and therefore providing a better ΔE-E resolution; a good spatial localization of the electrons leads to a short collection time, which in turn implies a fast signal rise time enabling the detector to handle high count rates with low pile-up rates (Schulze-König et al. 2010). Systematic studies showed that electronic noise was still an essential and limiting factor for light ions. A further reduction of the electronic noise could be achieved with newly available preamplifiers from AMPTEK (FWHM: 8.6 keV) (Suter et al. 2007; Müller et al. 2008). This required a new design with the preamplifier mounted outside of the detector while keeping the dimensions of the detector electrodes about the same. The whole detector is now mounted on an end flange of the beam line.

The vacuum conditions in the acceleration tubes before and after the stripper canal are not ideal because a small fraction of the stripper gas leaks out of the stripper. In these regions, the probability for charge changing and scattering processes is strongly enhanced, which in turn leads to tails in the energy and the angular distributions. Particles from neighboring masses can thus pass a spectrometer consisting of 1 magnet and 1 electrostatic analyzer. In order to eliminate this background, larger AMS systems are often equipped with a second magnet on the high-energy side. Originally, it was hoped that these background effects could be kept small enough with an appropriate stripper design or/and by using time-of-flight detectors. After several years of experience, however, we came to the conclusion that an additional magnet is the best solution to improve the performance of the system in this respect. Especially for ^{10}Be measurements, an additional magnet provides not only the elimination of isotopic background, but also allows to define an independent energy window for the ^{10}B-^{10}Be separation, when the degrader foil method is applied (Müller et al. 2008). In order to keep the overall dimension of the facility small, a 130° deflection magnet with a bending radius of 75 cm was chosen providing short focal distances (Figures 1 and 2) (Suter et al. 2010). With this arrangement, all masses analyzed with the first magnet can also pass the second magnet. Tests after installation in spring 2009 indicate that the expected improvement in performance can be achieved.

RESULTS

Performance is primarily characterized by the overall efficiency, background, and precision. It has been demonstrated that ^{14}C can be measured with precision in the per mil range (Goslar et al. 2004;

Synal and Wacker 2010; Wacker et al. 2010). This aspect has been a topic in many other papers and is therefore not discussed here. For the measurement of other radionuclides, an important question to answer is whether a sufficient ion yield can be obtained with compact facilities. When discussing the number of atoms of the sample reaching the detector, the ion source yield is an essential parameter. However, small and large AMS systems use in general similar ion sources and this aspect is thus not discussed here any further. This leaves the terminal stripping yield and the losses on apertures and in the stripper canal as the relevant factors, although some additional losses might occur in the particle identification system. Beam losses depend on the phase space extracted from the ion source and how it matches the ion optical design and beam-limiting apertures. Good experimental data on stripping yields in the energy range below 1 MeV are sparse in the literature and no reliable model is available to estimate these yields at low energies. Therefore, transmission measurements as function of stripper density have been performed for the most relevant AMS radioisotopes using our compact AMS facility (Jacob 2001; Stocker et al. 2004; Stocker 2006). The pressure dependency of the transmission yields information on small angle scattering losses in the stripper. For light ions, single scattering is the dominant process for losses and models (Lindhard et al. 1968) for scattering on screened potentials give good estimates on these losses (Suter 2004). For heavier ions, multiple scattering is significant and appropriate models (Sigmund and Winterbon 1974) provide good estimates. Also, simulations with the SRIM program (http://www.srim.org/) give good results on small angle scattering losses.

For heavier elements, the magnetic field of the HE magnet(s) is not necessarily strong enough to bend the charge state with the highest yield. Typically for ions heavier than Al, a higher charge state has to be selected. For the proper selection of the charge state, also the dependency of background (of molecules or m/q ambiguities) on charge states has to be considered. In addition, the energy needed for proper particle identification in the gas ionization detector determines which charge state is to be selected. This is particularly critical for small AMS systems with low ion energies. Table 1 shows the charge states chosen based on the above considerations, associated yields, beam losses, and transmission values for the radionuclides measured with our compact AMS system.

Table 1 Stripping yields and scattering losses for various radioisotopes at the compact AMS facility in Zurich. The numbers given are for the most abundant stable isotope under conditions used for measurements.

Element	Terminal voltage (MV)	Charge state	Stripping yield	Scattering losses	Transmission
Be	0.53	1	63%	9%	57%
C	0.47	1	50%	15%	43%
Al	0.45	1	42%	45%	23%
Ca	0.5	3	5%	27%	4%
I	0.5	3	10%	18%	8%
Th	0.3	3	24%	37%	15%

Be^{1+} ions provide an almost constant high stripping yield of about 60–65% over a large energy range (170–500 keV), corresponding to terminal voltages ranging of 0.4–1.2 MV when BeO^- is injected (Figure 3). For carbon, charge state 1+ also gives a high yield of more the 50% in the range 250–550 keV (Jacob et al. 2000). For a 1MV accelerator, charge state 2+ is most appropriate for carbon. The highest yield for Al in the 500-kV region can be obtained by analyzing 1+ ions, but the interference of various isobaric molecules requires a relatively high stripper density (~2 $\mu g/cm^2$), leading to beam losses of 40 to 50%. Charge state 2+ cannot be used because of the intense $^{13}C_2$ interference.

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Therefore, the selection of 3+ ions has to be considered. ^{41}Ca measurements for biomedical application are made with $^{41}CaF_3^-$ beams. The injection of this heavy molecule leads to relative low energies of calcium ions in the stripper with relatively low stripping yields for 2+ and 3+ ions (10–15% and 3–5%, respectively) (Stocker et al. 2004). This is, however, sufficient to measure biomedical samples with $^{41}Ca/^{40}Ca$ isotopic ratios in the range of 10^{-9} to 10^{-10}. For ^{129}I and actinides, 3+ ions are the most appropriate to select and, compared to larger instruments, relatively high stripping yields (>10%) can be obtained. In this respect, compact AMS facilities are very attractive. On the other hand, beam losses due to small angle scattering can be important and have to be considered in the design of the stripper canal and ion transport. These losses rise up to 50%. Proper selection of the stripping gas may help to reduce these losses. Argon gas has been used for stripping at most laboratories and also at the ETH compact AMS facility. At low energies, beam losses due to small angle scattering could be significantly reduced by using lighter gases (e.g. N_2, O_2). In some cases, these gases give also higher stripping yields (Wittkower and Betz 1973). Also, cross-sections for molecular dissociation have to be regarded in this optimization process.

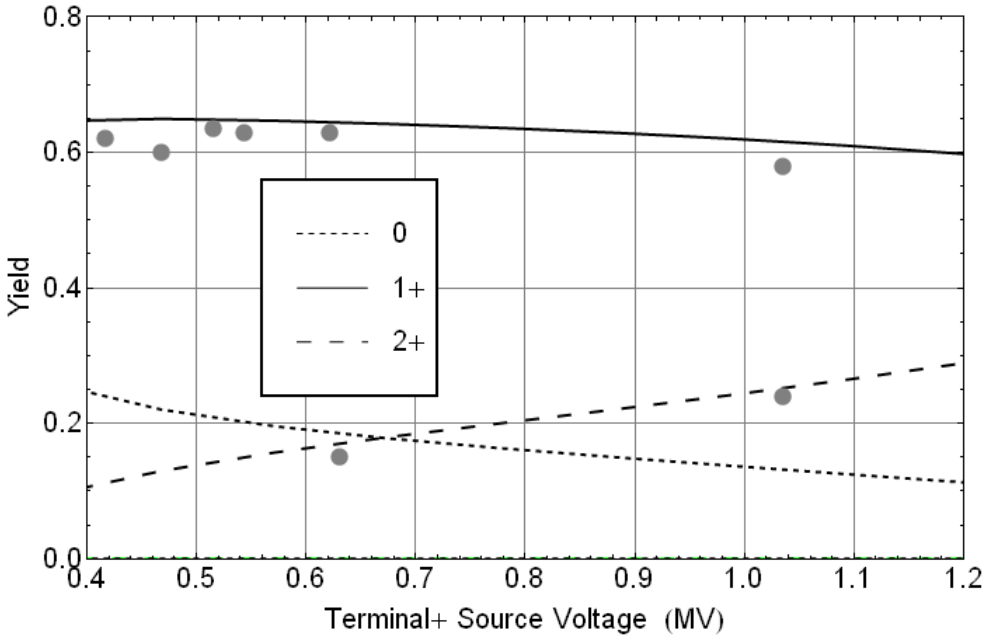

Figure 3 Stripping yields (lines) for ^{9}Be based on the extrapolation of charge changing cross-sections measured at higher energies (Niklaus et al. 1994). The data points are based on transmission measurement at ETH (Grajcar et al. 2004) and at CNA in Seville. The data show that high stripping yields of more than 60% can be obtained over a large energy range.

Background levels for the various radioisotopes are listed in Table 2. Background problems are more severe at lower energies for the following reasons: Cross-sections for scattering and for the relevant charge exchange processes are significantly larger (Niklaus et al. 1994); hence, these processes are responsible for the increased probability of neighboring masses passing the mass spectrometers and reaching the final detector. Also, particle identification with gas ionization chambers using the ΔE-E detection technique is much more difficult at low energies because the signals and their separation are smaller. Molecules in low charge states (1+ or 2+) might survive the stripping process, if the stripper density is not high enough. On the other hand, higher stripper densities lead

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to larger beam losses and increased background from scattering. The stripper geometry has to be designed so that good vacuum conditions are maintained in the acceleration tubes, but at same time beam losses should be small (Jacob et al. 2000). Maximizing yields and minimizing background at the same time is not always possible, so compromises have to be made. For many radioisotopes, certain background components (isobars, molecules, M/q ambiguities) can be identified with an appropriate detector system. Detectors with sufficient energy resolution at these low energies were not available when this project started, but meanwhile the detector resolution has been improved by successively optimizing the performance of gas ionization chambers.

Table 2 Performance data for the ETH compact facility and 1MV instrument at CNA. The transmission values are those for the rare isotope under measurement conditions. All background data are shown as isotopic ratio of radioisotope to stable isotope and are based on experiments before the second HE magnet was installed, expect for ^{10}Be. Due to the absence of a stable isotope, no background ratios are listed for the actinides. Significant improvements are expected for ^{26}Al and the actinides with this magnet. For comparison, data for larger facilities are given in the form of ranges as the performance varies widely from facility to facility.

	ETH Zurich NEC compact 0.5MV		Seville HVEE compact 1MV		Larger >2.5MV	
Element	Transmission %	Background 10^{-14}	Transmission %	Background 10^{-14}	Transmission %	Background 10^{-14}
^{10}Be[a]	10	<0.1	8	3	10–38	0.05–1
^{14}C	43	0.3	43	0.3	40–62	0.06–0.3
^{26}Al	23	1	25	5	15–43	0.05–1
^{41}Ca[b]	5	300			3–5	0.1–5
^{129}I	8	6	10	30	4–13	0.5–10
Actinides	12–15		12–16		1–6	

[a]Injected as BeO^-. Transmission given for ^{10}Be (including that through the degrader foil).
[b]Injected as CaF_3^-.

One of the most difficult and challenging problems with compact AMS instruments is the measurement of ^{10}Be, because the intense background of the isobar ^{10}B has to be suppressed by 8–10 orders of magnitude. At higher energies, an absorber method can be applied. ^{10}B can be eliminated completely because it is stopped in the absorber while ^{10}Be is reaching the detector (Klein et al. 1982). At low energies of ~1 MeV, the range straggling leads to a significant overlap in the stopping distance, so that this method is not applicable for AMS facilities operated with terminal voltages below 1 MV (Figure 4). An alterative has been shown by Raisbeck et al. (1984, 1987), who used a degrader foil technique that worked quite well at beam energies at 5.2 MeV. This method was also extensively studied at ETH. Significant progress was made by using very homogeneous Si_3N_4 foils, allowing a reduction of the ^{10}B rate by 4 orders of magnitude (Grajcar et al. 2004, 2007; Müller et al. 2008). Disadvantages of this method are the additional count rate losses due to energy straggling and angular straggling as well as the losses due to the distribution of the beam into various charge states, so this degrader foil method reduces in total the ^{10}Be rate by about a factor of 4. In addition, the detector development was essential to reach the required additional suppression power of about 6 orders of magnitude. At higher energies, as can already be obtained with the 1MV facility from HVEE, ^{10}Be identification in the presence of intense ^{10}B interference becomes less serious, because the energy loss differences of ^{10}Be and ^{10}B in the detector are larger. The residual limiting factor for ^{10}Be measurements at low energies using the degrader foil technique was background at the ^{10}Be/Be

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level of 10^{-13}, which was caused by ^{9}Be (Müller et al. 2008). With an additional mass filter, this background can be removed.

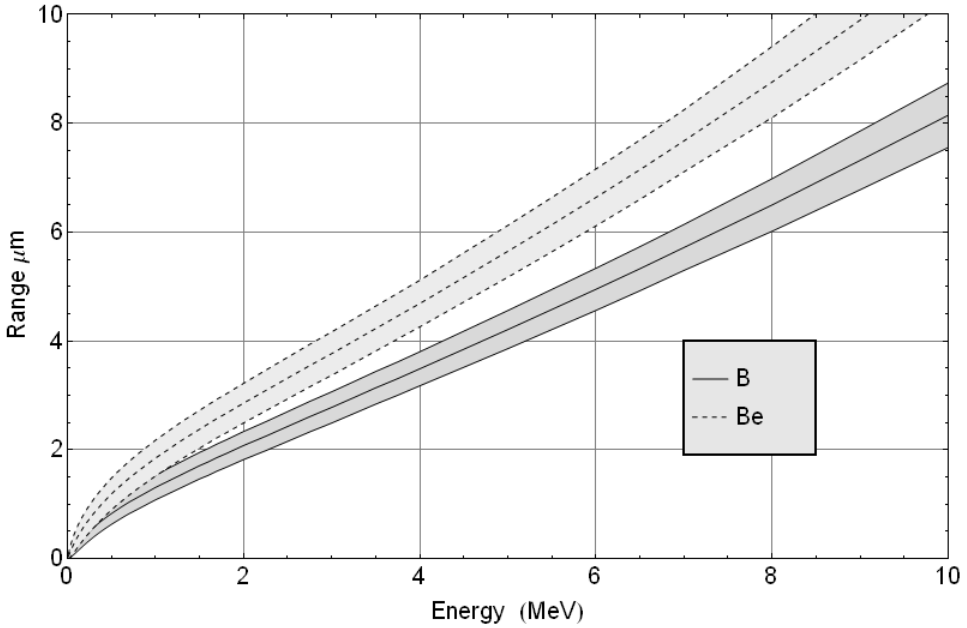

Figure 4 Range straggling (2-σ interval) for ^{10}B and ^{10}Be in Si_3N_4, based on the SRIM program. A clear overlap of the distributions is visible at energies below 1 MeV.

Another way to solve the isobar problem without using a degrader was proposed by the Isotrace Laboratory at the University of Toronto, Canada. By extracting BeF^- beams, the ^{10}B counting rate can be reduced by many orders of magnitude, because BeF^- ions do not form stable molecules (Zhao et al. 2004). With this technique, ^{10}Be can be clearly separated from ^{10}B in a gas ionization chamber alone. Several tests have been performed at ETH to estimate the potential of this method. It was found that only relatively small negative ion currents could be extracted from BeF_2 and $BaBeF_4$ targets, making routine measurements of low isotopic concentrations tedious (Grajcar et al. 2007; Müller et al. 2008).

PERFORMANCE

Here, we discuss the performance of the compact AMS facility at ETH and compare it with that of the 1MV AMS facility at CNA in Seville, based on published data (Table 2). Performance parameters of larger instruments are also listed, but as ranges only because the performance varies strongly from instrument to instrument.

^{10}Be: We could demonstrate that ^{10}Be/Be ratios can be measured with compact AMS instruments operating at terminal voltages of 520–570 kV using a degrader foil method with silicon nitride membranes 60–100 nm thick. An overall transmission of ~10% is obtained and the background is less than 1×10^{-15}. The key to this achievement is a high-resolution gas ionization detector and a second magnet in the high-energy mass spectrometer. Good sample preparation chemistry is a prerequisite to keep the ^{10}B counting rate in the detector at acceptable levels. The 1MV AMS facility at CNA is presently capable of measuring ^{10}Be with a background level of $2–4 \times 10^{-14}$ and an overall transmis-

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sion of about 10%. Their energy spectra clearly indicate scattering events with lower energies, which are most probably caused by scattering on the deflector plates.

^{14}C: Many compact AMS facilities are in operation either based on the tandem principle or as a so-called single stage system. These systems have demonstrated that they are powerful tools for ^{14}C dating. Performance parameters have been reported in many publications and are therefore not discussed here. One special note on the usage of charge state 2+ at 1MV facilities should be made. Several earlier attempts with Si solid state detectors showed that the interference from two $^{7}Li^{1+}$ ions arriving simultaneously at the detector is a limiting factor that cannot be resolved with those detectors. With gas ionization detectors, however, this interference can be identified due to the pulse height defect, which is different for C and Li (Klein et al. 2007). The use of 2+ ions, however, has the advantage that the measurements can be performed at significant lower stripper density because the destruction of molecules is more efficient in this case.

^{26}Al: ^{26}Al measurements are performed in charge state 1+. A high stripper density is needed to break up the abundant molecules of mass 26 ($^{12}C^{14}N$, $^{13}C_2$, $^{10}B^{16}O$, ^{25}MgH). Molecules surviving the stripping process can be identified in the detector except for MgH (Stocker et al. 2005). Early measurements had background levels of about 10^{-14}. If this background would be due to ^{27}Al, it should now be removed by the second magnet. The original NEC MC-SNICS ion source provided relatively low currents, making the system unattractive for routine measurements. With the upgraded system (ion source, second magnet), we expect a performance sufficient for routine applications.

^{41}Ca: This isotope has the isobaric interference ^{41}K. First experiments were made with CaH_3^- beams, which allow a strong reduction of ^{41}K background, but sample preparation and handling is too complex for routine applications in biomedical research. CaF_3^- also provides a large reduction in the ^{41}K intensity, so that biomedical applications are possible even when no full ^{41}K-^{41}Ca separation is possible in the final detector (Schulze-König et al. 2010). A peak shift indicates when a high K contamination is present in the sample. With careful sample preparation as well as clean ion source conditions, the ^{41}Ca/Ca background ratio is in the range of 10^{-11} to 10^{-12}. A transmission of about 5% is possible for ions in the 3+ charge state.

^{129}I: Initially, ^{129}I was measured in charge state 4+. The M/q ambiguity $^{97}Mo^{3+}$ produced some background and the stripping yield was relatively small (4%). The yield could be increased to about 8% (Alfimov et al. 2010) by selecting the 3+ charge state. Under appropriate operating conditions as well as applying adequate sample chemistry, the ^{129}I/I background ratio is 6×10^{-14}. M/q ambiguities ($^{27}Al^{16}O^+$ and $^{86}Br^{2+}$) can be separated if their counting rates are not too high. The molecular interference ($^{27}Al^{16}O^+$) can be suppressed by higher stripper pressure, whereas the scattered ^{127}I into the detector can be removed by the additional mass filter. This performance is sufficient for most applications. All iodine measurements are now performed with our compact AMS facility.

Actinides: Measurements of the actinides require sequential counting of several isotopes in the detector due to the absence of an abundant stable or long-lived isotope (except for U). Actinides cannot be measured at the maximal terminal voltage because of the limitation of the HE magnet. Surprisingly, a relatively high transmission of about 12–15% can be obtained at a terminal voltage of 300 kV (Fifield et al. 2004; Wacker et al. 2005). This is much higher than all large AMS facilities measuring actinides report. The compact AMS instrument is now used routinely for applications. ^{231}Pa is now also routinely measured with this instrument (Christl et al. 2007). Tests for ^{236}U were performed and a detection limit of about 10^{-9} was due to the background of ^{235}U. This background can by eliminated to a large extent with the new magnet. Similar performance is quoted for Pu isotopes for the 1MV facility at CNA.

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One would expect that a 1MV AMS system should have a better performance than a 0.5MV system, because scattering losses should be smaller and stripping yields should in general be higher. Yet, comparison with the instrument in Seville shows about the same transmission values. The background there is actually higher for several radioisotopes (^{10}Be, ^{26}Al, ^{129}I). This indicates that there is still potential for improvement. Comparing performance parameters with those of larger facilities, the compact systems can compete with many dedicated AMS facilities with terminal voltages of 2 MV or higher. These larger facilities are needed for measurements of radioisotopes with interferences of stable isobars. One exception is ^{10}Be with its large relative difference in nuclear charge compared to ^{10}B, making isobar separation in a gas ionization detector possible.

CONCLUSIONS

Within 10 yr of development at ETH, a new generation of universal compact AMS systems has emerged. With this new category of small AMS instruments, cosmogenic radionuclides such as ^{10}Be, ^{14}C, and ^{26}Al can be measured with background levels quite similar to those of many larger instruments and with sufficient efficiency for many applications. The best large instruments have significantly higher transmissions for ^{10}Be and also ^{26}Al. Compact facilities can therefore not fully replace larger instruments. ^{36}Cl, another important cosmogenic radionuclide, cannot be measured at natural concentrations with this type of instrument. But for heavy ions such as I as well as the actinides, surprisingly good performance has been obtained. Especially for actinides, transmissions are higher than those at any of the large AMS facilities.

Very essential for success was the improvement of gas ionization detectors. Electronic noise could be reduced by a factor of nearly 4. With silicon nitride foils, the energy-loss straggling in the entrance window has been reduced significantly. A compact design and good electronic shielding improved the resolution further. Isobar suppression for ^{10}Be measurements can be as high as 6 orders of magnitude. Molecules can be identified in some cases and M/q ambiguities can be clearly separated.

With a second magnet in the HE analyzing beam line, the background induced by neighboring masses has been strongly reduced. This magnet increases the complexity of the system and its cost, but a design has been found that does not increase the overall size of the instrument. The potential of this magnet has not yet been fully explored, but initial tests indicated that the background from neighboring isotopes is now significantly reduced and in most cases not a limiting factor any more.

We have demonstrated that the low-energy AMS concept is a very attractive alternative to larger instruments for many applications due to the lower cost and smaller space requirement. Especially for ^{14}C laboratories that want to expand their application program to other isotopes, these compact instruments are best suited. The companies manufacturing AMS facilities have realized the potential of this category of instruments. The prototype of a universal AMS facility from HVEE operated at CNA in Seville has shown promising results for many isotopes, but a comparison with the ETH instrument clearly demonstrates that there are possibilities for improvements in that instrument. NEC is manufacturing a compact AMS system based on 500kV Pelletron, which is designed for ^{14}C analysis, but with some modification it should also allow ^{10}Be measurements with the degrader foil method as well as the detection of other radioisotopes.

Further developments of compact facilities seem to be feasible. There is a potential for improving the detector performance. Energy resolution might be further improved. Based on our experiments, the best operating conditions concerning the necessary stripper densities are now roughly known. The scattering losses under these conditions have also been studied. Based on this information, it

seems that the stripper geometry could be improved in order to reduce scattering losses. Also, a modification of the LE mass spectrometer should be considered. A magnet with a larger gap and higher magnetic field could enhance beam transmission, especially for the actinides.

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REFERENCES

Alfimov V, Synal H-A. 2010. ^{129}I AMS at 0.5MV tandem accelerator. *Nuclear Instruments and Methods in Physics Research B* 268(7–8):769–72.

Chamizo E, López-Gutierrez JM, Ruiz-Gómez A, Santos FJ, García-León M, Maden C, Alfimov V. 2008. Status of the compact 1 MV AMS facility at the Centro Nacional de Aceleradores (Spain). *Nuclear Instruments and Methods in Physics Research B* 266(10): 2217–20.

Christl M, Wacker L, Lippold J, Synal H-A, Suter M. 2007. Protactinium-231: a new radionuclide for AMS. *Nuclear Instruments and Methods in Physics Research B* 262(2):379–84.

Döbeli M, Kottler C, Stocker M, Weinmann S, Synal H-A, Grajcar M, Suter M. 2004. Gas ionization chambers with silicon nitride windows for the detection and identification of low energy ions. *Nuclear Instruments and Methods in Physics Research B* 219–220:415–9.

Fifield LK, Synal H-A, Suter M. 2004. Accelerator mass spectrometry of plutonium at 300 kV. *Nuclear Instruments and Methods in Physics Research B* 223–224: 802–6.

Goslar T, Czernik J, Goslar E. 2004. Low-energy ^{14}C AMS in Poznań Radiocarbon Laboratory, Poland. *Nuclear Instruments and Methods in Physics Research B* 223–224:5–11.

Grajcar M, Döbeli M, Kubik PW, Maden C, Suter M, Synal H-A. 2004. ^{10}Be measurements with terminal voltages below 1 MV. *Nuclear Instruments and Methods in Physics Research B* 223–224:190–4.

Grajcar M, Döbeli M, Kubik PW, Synal H-A, Wacker L, Suter M. 2007. New concepts of ^{10}Be AMS at low energies. *Nuclear Instruments and Methods in Physics Research B* 259(1):173–7.

Hughey BJ, Klinkowstein RE, Shefer RE, Skipper PL, Tannenbaum SR, Wishnok JS. 1997a. Design of a compact 1 MV AMS system for biomedical research. *Nuclear Instruments and Methods in Physics Research B* 123(1–4):153–8.

Hughey BJ, Shefer RE, Klinkowstein RE, Zhao X-L, Kieser WE, Litherland AE. 1997b. Experimental investigation of the destruction of 1 MeV $^{12}CH_2$ molecules in single and double stripping foils. *Nuclear Instruments and Methods in Physics Research B* 123(1–4):186–92.

Jacob S. 2001. Beschleunigermassenspektrometrie (AMS) von ^{14}C bei tiefen Energien [PhD dissertation]. Dissertation Nr. 14071; ETH Zürich.

Jacob SAW, Suter M, Synal H-A. 2000. Ion beam interaction with stripper gas—key for AMS at sub MeV. *Nuclear Instruments and Methods in Physics Research B* 172(1–4):235–41.

Klein J, Middleton R, Tang H. 1982. Modifications of an FN tandem for quantitative ^{10}Be measurement. *Nuclear Instruments and Methods in Physics Research B* 193(3):601–16.

Klein MG, Mous DJW, Gottdang A. 2006. A compact 1 MV multi-element AMS system. *Nuclear Instruments and Methods in Physics Research B* 249(1–2):764–7.

Klein MG, van Staveren HJ, Mous DJW, Gottdang A. 2007. Performance of the compact HVE 1 MV multi-element AMS system. *Nuclear Instruments and Methods in Physics Research B* 259(1):184–7.

Klody GM, Schroeder JB, Norton GA, Loger RL, Kitchen RL, Sundquist ML. 2005. New results for single stage low energy carbon AMS. *Nuclear Instruments and Methods in Physics Research B* 240(1–2): 463–7.

Lee HW, Galindo-Uribarri A, Chang KH, Kilius LR, Litherland AE. 1984. The $^{12}CH_2^{2+}$ molecule and radiocarbon dating by accelerator mass spectrometry. *Nuclear Instruments and Methods in Physics Research B* 5(2):208–10.

Lindhard J, Nielsen V, Scharff M. 1968. Approximation method in classical scattering by screened coulomb fields. *Det Kongelige Danske Videnskabernes Selskab Matematisk-fysiske Meddelelser* 36(10):1–32.

Litherland AE. 1984. Accelerator mass spectrometry. *Nuclear Instruments and Methods in Physics Research B* 5(2):100–8.

Mous DJW, Purser KH, Fokker W, van den Broek R, Koopmans RB. 1997. A compact ^{14}C isotope ratio mass spectrometer for biomedical applications. *Nuclear Instruments and Methods in Physics Research B* 123(1–4):159–62.

Müller AM, Christl M, Döbeli M, Kubik PW, Suter M, Synal H-A. 2008. ^{10}Be AMS measurements at low energies (E < 1 MeV). *Nuclear Instruments and Methods in Physics Research B* 266(10):2207–12.

Niklaus TR, Bonani G, Guo Z, Suter M, Synal H-A.

1994. Optimising tandem accelerator stripping efficiency by simulation of charge changing processes. *Nuclear Instruments and Methods in Physics Research B* 92(1–4):115–21.

Purser KH. 1994. A future AMS/chromatography instrument for biochemical and environmental measurements. *Nuclear Instruments and Methods in Physics Research B* 92(1–4):201–6.

Raisbeck GM, Yiou F, Bourles D, Lestringuez J, Deboffle D. 1984. Measurement of ^{10}Be with a Tandetron accelerator operating at 2 MV. *Nuclear Instruments and Methods in Physics Research B* 5(2):175–8.

Raisbeck GM, Yiou F, Bourles D, Lestringuez J, Deboffle D. 1987. Measurements of ^{10}Be and ^{26}Al with a Tandetron AMS facility. *Nuclear Instruments and Methods in Physics Research B* 29(1–2):22–6.

Roberts ML, Culp RA, Dvoracek DK, Hodgins GWL, Neary MP, Noakes JE. 2004. The ^{14}C AMS system at The University of Georgia. *Nuclear Instruments and Methods in Physics Research B* 223–224:1–4.

Schulze-König T, Maden C, Denk E, Freeman SPHT, Stocker M, Suter M, Synal H-A, Walczyk T. 2010. Comparison of ^{41}Ca analysis on 0.5 MV and 5 MV AMS systems. *Nuclear Instruments and Methods in Physics Research B* 268(7–8):752–5.

Sigmund P, Winterbon KB. 1974. Small angle multiple scattering in the screened Coulomb region. *Nuclear Instruments and Methods* 119:541–57.

Skog G. 2007. The single stage AMS machine at Lund University: status report. *Nuclear Instruments and Methods in Physics Research B* 259(1):1–6.

Stocker M. 2006. AMS bei tiefen Energien [PhD dissertation]. Dissertation nr 16787; ETH Zürich.

Stocker M, Bertschinger R, Döbeli M, Grajcar M, Jacob S, Scheer J, Suter M, Synal H-A. 2004. Status of the PSI/ETH compact AMS facility. *Nuclear Instruments and Methods in Physics Research B* 223–224:104–8.

Stocker M, Döbeli M, Grajcar M, Suter M, Synal H-A, Wacker L. 2005. A universal and competitive compact AMS facility. *Nuclear Instruments and Methods in Physics Research B* 240(1–2):483–9.

Suter M. 1998. A new generation of small facilities for accelerator mass spectrometry. *Nuclear Instruments and Methods in Physics Research B* 139(1–4):150–7.

Suter M. 2004. 25 years of AMS—a review of recent developments. *Nuclear Instruments and Methods in Physics Research B* 223–224:139–48.

Suter M, Jacob S, Synal H-A. 1997. AMS of ^{14}C at low energies. *Nuclear Instruments and Methods in Physics Research B* 123(1–4):148–52.

Suter M, Huber R, Jacob SAW, Synal H-A, Schroeder JB. 1999. A new small accelerator for radiocarbon dating. In: Applications of Accelerators in Research and Industry. Proceedings of the Fifteenth International Conference. *AIP Conference Proceedings* 475:665–7.

Suter M, Jacob SWA, Synal H-A. 2000. Tandem AMS at sub-MeV energies—status and prospects. *Nuclear Instruments and Methods in Physics Research B* 172(1–4):144–51.

Suter M, Döbeli M, Grajcar M, Müller A, Stocker M, Sun GY, Synal H-A, Wacker L. 2007. Advances in particle identification in AMS at low energies. *Nuclear Instruments and Methods in Physics Research B* 259(1):165–72.

Suter M, Chamizo E, Müller AM, Synal H-A. 2010. The relevance of ion optics for the development of small AMS facilities. *Nuclear Instruments and Methods in Physics Research B* 268(7–8):722–5.

Synal H-A, Wacker L. 2010. AMS measurement techniques after 30 years: possibilities and limitations of low energy systems. *Nuclear Instruments and Methods in Physics Research B* 268(7–8):701–7.

Synal H-A, Jacob S, Suter M. 2000b. New concepts for radiocarbon detection systems. *Nuclear Instruments and Methods in Physics Research B* 161–163:29–36.

Synal H-A, Jacob S, Suter M. 2000b. The PSI/ETH small radiocarbon dating system. *Nuclear Instruments and Methods in Physics Research B* 172(1–4):1–7.

Synal H-A, Döbeli M, Jacob S, Stocker M, Suter M. 2004. Radiocarbon AMS towards its low-energy limits. *Nuclear Instruments and Methods in Physics Research B* 223–224:339–45.

Synal H-A, Stocker M, Suter M. 2007. MICADAS: a new compact radiocarbon AMS system. *Nuclear Instruments and Methods in Physics Research B* 259(1):7–13.

Vogel JS, Turteltaub KW, Felton JS, Gledhill BL, Nelson DE, Southon JR, Proctor ID, Davis JC. 1990. Application of AMS to the biomedical sciences. *Nuclear Instruments and Methods in Physics Research B* 52(3–4):524–30.

Wacker L, Chamizo E, Fifield LK, Stocker M, Suter A, Synal H-A. 2005. Measurement of actinides on a compact AMS system working at 300 kV. *Nuclear Instruments and Methods in Physics Research B* 240(1–2):452–7.

Wacker L, Bonani G, Friedrich M, I Hajdas, Kromer B, Němec M, Ruff M, Suter M, Synal H-A, Vockenhuber C. 2010. MICADAS: routine and high-precision radiocarbon dating. *Radiocarbon* 52(2–3):252–62.

Wittkower AB, Betz HD. 1973. Equilibrium-charge-state distributions of energetic ions ($Z > 2$) in gaseous and solid media. *Atomic Data and Nuclear Data Tables* 5(2):113–66.

Zhao X-L, Litherland AE, Doupé JP, Kieser WE. 2004. The potential for AMS analysis of ^{10}Be using BeF^-. *Nuclear Instruments and Methods in Physics Research B* 223–224:199–204.