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# Applications of a compact ionization chamber in AMS at energies below 1 MeV/amu

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#### Abstract

The increasing demand for measuring long-lived radionuclides with small AMS machines at energies below 1 MeV per nucleon raises the need for compact detectors which still have a decent energy resolution and allow for a clear identification of the incident particles. Based on a design by the AMS group at the ETH Zurich a compact gas ionization chamber was built and installed at the 3 MV tandem AMS facility VERA (Vienna Environmental Research Accelerator). The main challenge in AMS is the detection of rare isotope species in the presence of strong isotopic and isobaric interferences. The task of the ionization chamber is the suppression of the unwanted isobar by separating the ions via their different stopping powers. Measurements of <sup>36</sup>Cl at VERA showed an achieved suppression of the unwanted stable isobar <sup>36</sup>S of  $3 \times 10^{-4}$  and measurements of <sup>10</sup>Be showed an achieved suppression of <sup>10</sup>B of at least  $3 \times 10^{-6}$ . Additional suppression of the isobaric ions can be achieved by a degrader foil technique applied to <sup>10</sup>Be measurements by G.M. Raisbeck. In combination with the new ionization chamber the achieved suppression of <sup>10</sup>B is at least  $10^{-10}$ . Measurements of blank samples at VERA show that the background for AMS with <sup>10</sup>Be is below  $2 \times 10^{-15}$ . © 2008 Elsevier B.V. All rights reserved.

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#### 1. Introduction

Accelerator Mass Spectrometry (AMS) is a powerful technique for the detection of long-lived radioisotopes at typical abundances in the range of  $10^{-12}$ – $10^{-15}$ . The precise determination of such low abundances has found various applications as tracers or chronometers in geology, archaeology and biomedicine. Tuniz et al. give a detailed overview of the technique as well as a discussion of its numerous applications [1].

One of the main tasks in AMS measurements is the separation of the rare radioisotope from stable isobaric background. There are various ways to achieve this separation. In some cases this task can be efficiently fulfilled inside the ion source using negative ions. Examples for these cases are <sup>14</sup>C, <sup>26</sup>Al and <sup>129</sup>I where the stable isobars <sup>14</sup>N, <sup>26</sup>Mg and <sup>129</sup>Xe do not form negative ions. In other cases negative molecular ions can be used. One example is the measurement of <sup>41</sup>Ca where the interference of <sup>41</sup>K is strongly suppressed by using <sup>41</sup>CaH<sub>3</sub><sup>-</sup> ions through the instability of <sup>41</sup>KH<sub>3</sub><sup>-</sup>. Another possibility is the separation after acceleration using different methods like energy loss in matter, using the mean charge state in a gas-filled magnet or full stripping in a subsequent stripper stage.

The scope of this work is to show the application of a compact ionization chamber to achieve the separation of different isobars in AMS.

#### 2. Setup of the detector

At the laboratory for ion beam physics at the ETH Zurich a compact ionization chamber was developed for use

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with their sub-MeV tandem AMS machines [2]. The aim of such detectors is to separate the stable isobar from the radioisotope via their different energy loss in a gas. Based on this design a similar detector was built at the VERA laboratory for use at the VERA 3 MV tandem AMS facility.

Owing to its compact design the detector fits into a standard DN100CF cross piece. It can be inserted and retracted without breaking the vacuum. The active detector volume has a length of about 6 cm. A scheme of the inner part is shown in Fig. 1. The anode is split into two consecutive regions with equal lengths, which allows a simultaneous measurement of the energy loss in two different regions. The preamplifier electronics is built with CR-110 charge sensitive preamplifier modules from Cremat Inc. (Watertown, MA, USA). These compact hybrid modules have a small footprint, which allows mounting the preamplifiers for the two signals inside the active detector volume close to the anodes. This reduces the electronic noise due to short cable lengths. A major breakthrough for the use of ionization chambers at energies below 1 MeV/amu was the development of thin silicon nitride foils [3]. These foils are pinhole free and commercially available from Silson Ltd. (Northampton, UK) with thicknesses down to 30 nm. They are remarkably homogeneous, which produces almost no low energy tails. In our setup windows of square shape with a side length of 5 mm are used. They come with a native silicon frame of  $10 \text{ mm} \times 10 \text{ mm}$  that can be glued onto the entrance of the ionization chamber. The 50 nm foil used in our setup withstands easily a gas pressure above 100 mbar inside the detector. The Frisch grid [4] consists of gold coated tungsten wires (20 µm thickness) glued or soldered onto a stainless steel frame. The applied voltage between anode and cathode is 200 V. The Frisch grid was set to 60% of the full voltage using a resistive voltage divider. The distances between the cathode, the Frisch grid and the anodes are 25 mm and 5 mm, respectively. As counting gas isobutane is used. A separate gas regulation system is used to maintain a constant pressure inside the detector. The pressure was chosen such that the best possible separation for the measured isobars was achieved.

The output of the preamplifiers is sent to a dual channel spectroscopic amplifier (Ortec model 855) for amplification and pulse shaping. The resulting signals are sent, via two ADCs gated by the signal from the first anode, to the VERA data acquisition.



Fig. 1. Schematic of the active detector volume.

#### 3. Results

For the first performance tests of the detector a  $^{13}$ C particle beam with an energy of 12 MeV was produced. The detector was filled with 95 mbar of isobutane to fully stop the particles inside the active volume. The energy resolution (FWHM) of the measured signal was found to be 100 keV (0.8%). This remarkably high resolution outperforms a silicon detector by far. Also measurements with heavier ions were performed. For a 12 MeV Cu beam an energy resolution of 220 keV (1.8%) and for a 16.4 MeV Au beam a resolution of 400 keV (2.4%) were measured.

### 3.1. <sup>41</sup>Ca

In the case of <sup>41</sup>Ca the stable isobar is <sup>41</sup>K. As mentioned earlier this can be suppressed by using CaH<sub>3</sub><sup>-</sup> ions. But as the hydride is highly hygroscopic special care has to be taken during preparation and handling of the sample material. Another approach is using CaF<sub>3</sub><sup>-</sup> ions, which are easy to handle but show a much smaller suppression of KF<sub>3</sub><sup>-</sup>. The CaF<sub>3</sub><sup>-</sup> ions are extracted from CaF<sub>2</sub> sample material. After the acceleration stage ions of charge state 4<sup>+</sup> with an energy of 16 MeV were selected. The spectra of a typical sample material and a blank are shown in Fig. 2. The measurement time was 422 s. The measured isotope ratio of <sup>41</sup>Ca/Ca of the unknown sample was  $5.1 \times 10^{-12}$ . The blank sample was measured to have  $2.4 \times 10^{-13}$ .

## 3.2. <sup>36</sup>Cl

In <sup>36</sup>Cl measurements the stable isobar is <sup>36</sup>S. A suppression to a certain degree of sulfur can be achieved by chemical treatment of the sample material and putting it into a special sample holder containing virtually no sulfur, but a relatively high background of <sup>36</sup>S remains. To achieve a decent separation a high particle energy of 28 MeV using the  $7^+$  charge state of the ions was chosen. Instead of fully stopping the particles inside the active region of the detector a low pressure of 35 mbar of isobutane was used. In this way the counts of <sup>36</sup>Cl ions show up in a region of the spectra which is empty of events of scattered <sup>36</sup>S particles. The spectra of a typical sample material and a blank are shown in Fig. 3. The bin for the <sup>36</sup>Cl events was chosen in such a way that only the right half of the peak area was used. This reduces the detection efficiency by half but in this way a suppression of the unwanted isobar  ${}^{36}$ S of  $3 \times 10^{-4}$  could be achieved. The measured isotope ratio of  ${}^{36}$ Cl/Cl was  $4.6 \times 10^{-12}$ . The blank was measured to have  $5.5 \times 10^{-15}$ .

3.3. <sup>10</sup>Be

AMS measurements of <sup>10</sup>Be are influenced by a strong interference of the stable isobar <sup>10</sup>B. Previous measurements at VERA were performed using BeO as sample material and extracting BeO<sup>-</sup> beams from the sputter ion source. The interfering isobar is stopped in front of a silicon detector using an absorber. A different approach using a degrader foil after the high energy mass separation was

developed by G.M. Raisbeck [5]. We adopted this method for <sup>10</sup>Be measurements at VERA. After mass separation the



Fig. 2. Separation of 16 MeV <sup>41</sup>Ca and <sup>41</sup>K measured with the ionization chamber. The horizontal and the vertical axis show the energy loss and the residual energy, respectively. The left figure show a sample material with an unknown amount of <sup>41</sup>Ca whereas the right figure shows the measurement of a blank sample. The position of <sup>41</sup>Ca and <sup>41</sup>K counts are marked with circles. The measured isotope ratio of <sup>41</sup>Ca/Ca of the sample is  $5.1 \times 10^{-12}$ . The blank was measured to have  $2.4 \times 10^{-13}$ . The measurement time was 422 s and the average <sup>40</sup>Ca<sup>4+</sup> current was 0.1 µA in both cases.



Fig. 3. Separation of 28 MeV  ${}^{36}$ Cl and  ${}^{36}$ S measured with the ionization chamber. The ions were not stopped inside the active volume. The horizontal axis shows a transformation of the first anode signal whereas the vertical axis shows the energy loss in the second anode region. The left figure shows a sample material with an unknown amount of  ${}^{36}$ Cl whereas the right figure shows the measurement of a blank sample. The position of  ${}^{36}$ Cl and  ${}^{36}$ S counts are marked with circles. The measured isotope ratio of  ${}^{36}$ Cl/Cl was  $4.6 \times 10^{-12}$ . The blank was measured to have  $5.5 \times 10^{-15}$ . The measurement time was 100 s and the average  ${}^{35}$ Cl<sup>7+</sup> current of the blank sample was  $0.5 \,\mu$ A.



Fig. 4. Separation of <sup>10</sup>Be and <sup>10</sup>B measured with post-stripping and the ionization chamber. The left figure shows a typical spectrum from <sup>10</sup>Be standard material with an isotope ratio of <sup>10</sup>Be/<sup>9</sup>Be of  $9.5 \times 10^{-11}$ . The right figure show a sum of several runs performed on BeO blank material. No <sup>10</sup>Be events have been counted in this sample. The average <sup>9</sup>Be<sup>2+</sup> current of the blank sample was 2.5  $\mu$ A.

ion beam passes through a 260  $\mu$ g/cm<sup>2</sup> thick silicon nitride foil. The  ${}^{10}$ B ions lose more energy in the foil than the  ${}^{10}$ Be ions, and thus can be partially separated by a 20° switching magnet. The <sup>10</sup>Be and the remaining <sup>10</sup>B are injected directly into the detector. The energy of the ions before the degrader foil was 7.2 MeV with a charge state of  $2^+$ . After the foil, ions of charge state  $4^+$  were selected with the magnet and subsequently sent to the detector. The gas pressure (95 mbar) was chosen to achieve the largest separation between the two different isobars. Fig. 4 shows a spectrum of a <sup>10</sup>Be standard material with a <sup>10</sup>Be/<sup>9</sup>Be isotope ratio of  $9.5 \times 10^{-11}$  and a spectrum of a BeO blank material. The different counts in the spectra are marked. The measurement time for the blank material was 4400 s with an average  ${}^{9}\text{Be}^{2+}$  current of 2.5  $\mu$ A. During this time no event of <sup>10</sup>Be has been counted. Due to the long measurement time, background of <sup>9</sup>Be coming from <sup>9</sup>BeOH<sup>-</sup> can be seen. The <sup>9</sup>BeH survives the stripping process in the terminal and is subsequently destroyed in the stripper foil. The resulting <sup>9</sup>Be ions experience in isobutane nearly the same energy loss like the <sup>10</sup>Be but enter the detector with a slightly lower energy. Measurements of different blank samples show a background for <sup>10</sup>Be measurements at VERA of below  $2 \times 10^{-15}$ .

#### 4. Conclusion

The development of thin silicon nitride foils allowed the use of small gas ionization chambers at AMS facilities with energies at or below 1 MeV/amu. The decent energy resolution as well as particle identification via their energy loss allows measurements of radioisotopes like <sup>41</sup>Ca and <sup>36</sup>Cl previously difficult to measure at 3 MV machines. In combination with the post-stripping method, lower <sup>10</sup>Be isotope ratios can be achieved at 3 MV AMS facilities compared to former measurements. This allows using such facilities in geological applications, which require measurements of low abundances.

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