# AMS measurements in nuclear physics and astrophysics at VERA

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***Abstract:*** Accelerator mass spectrometry (AMS) represents a powerful technique with excellent sensitivity for the detection of long-lived radionuclides through ultra-low isotope ratio measurements. Research activities using AMS cover a wide variety of domains, e.g. $^{14}$C-dating, geological, biomedical applications, environ-mental and climate studies, and also nuclear physics related measurements.
The Vienna Environmental Research Accelerator (VERA) represents a state-of-the-art AMS facility active in a wide range of such applications. In particular, a substantial part is devoted to nuclear physics and astrophysics applications. AMS being independent on half-lives offers a powerful technique for such investigations. The measurement of neutron and proton capture cross-sections has become one main research topic at VERA. Various samples have been irradiated for that purpose. After the activation the amount of longer-lived radionuclides was quantified using the technique of AMS. In a fusion environment particularly long-lived activation products may lead to significant long-term waste disposals. For such nuclides production cross-sections and induced activities are key parameters for safety and design analyses. The quantification of actinides via AMS is not affected by isobaric molecular interferences. Different kinds of sources producing actinides are reflected in different signatures of e.g. Pu isotopes, like isotopic ratios and concentration levels. This information allows to identify human activity and to reconstruct its neutron history in environmental samples. Also, AMS is applied for searching for feeble traces of long-lived radionuclides as signatures of recent close-by supernova explosions.
An overview on recent research activities at VERA with respect to nuclear physics and nuclear astrophysics is presented. In addition, the actual detection limits for long-lived radionuclides are given.

## Technique of Accelerator Mass Spectrometry

Accelerator mass spectrometry (AMS) is a mass-spectrometric technique based on a tandem-accelerator, commonly used for quantifying long-lived radionuclides within a wide range of applications [1,2]. AMS has been utilized for the measurement of minute concentrations of isotopes over the whole mass range and is characterized by a low measurement background and high detection efficiency. In contrast to other mass spectrometric techniques, like e.g. ICP-MS, AMS is not affected by isobaric molecular interferences.
In most cases, negatively charged ions are produced in a Cs-sputter source. To this end, solid sputter targets have to be produced from the sample to be analyzed by AMS (new developments use also gaseous samples for $^{14}$C applications). The typical sample masses are of the order of mg per sputter sample. The sample material itself is used up during the measurement. Negative ions are pre-accelerated, energy- and mass-selected by passing an electrostatic deflector and an injection magnet, respectively. The ions are injected into a tandem accelerator. At the terminal a gas or foil stripper is utilized to strip-off electrons. The negatively charged ions then leave the accelerator positively charged after being accelerated a second time. With the stripping process, molecules are destroyed and only atomic particles with different charge states are produced. A second analyzing magnet is used to select a specific positive charge state. VERA is based on a 3-MV tandem accelerator, which produces particle energies between 10 and 25 MeV.
Currents are measured for stable isotopes with off-line Faraday cups (see Fig. 1) positioned at both sides, the low-energy side after the mass-selective injection magnet (e.g. $^{209}Bi^-$ or $^{12}C^-$ and $^{13}C^-$), and after the analyzing magnet ($^{209}Bi^{5+}$ or $^{12}C^{3+}$ and $^{13}C^{3+}$). The radionuclides are directly counted with a particle detector (count-rate of $^{210}Bi^{5+}$ or $^{14}C^{3+}$). AMS provides isotope

ratios, the count-rate ratio of radionuclide and stable isotope (e.g. $^{210}Bi/^{209}Bi$, $^{14}C/^{12}C$ or $^{14}C/^{13}C$) with current and count-rate measurements performed sequentially.

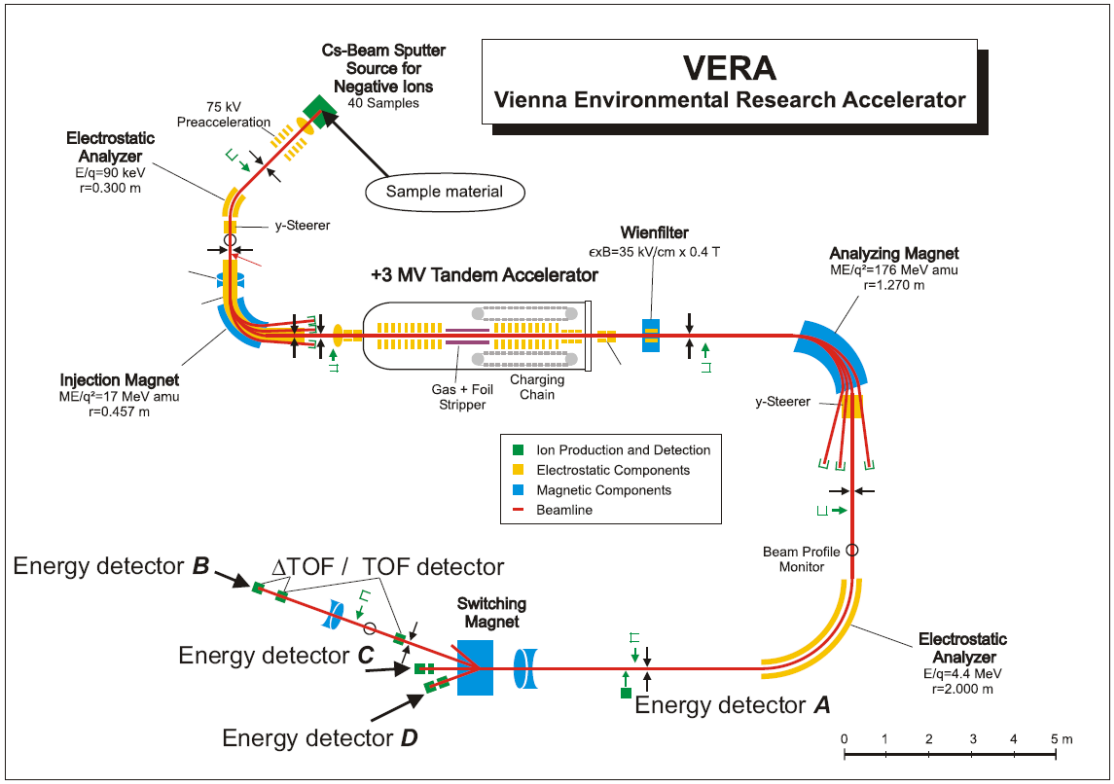


***Figure 1.*** Schematic layout of the VERA facility. Currents of stable ions are measured in sequential mode with Faraday cups positioned after the injection magnet (low-energy section, negative ions) and after the analyzing magnet (high-energy section, positive ions). Various detectors are available for counting rare isotopes (detectors A to D): The heavy-ion beamline (which is also used for $^{236}U$ and $^{210}Bi$-detection) consists of time-of-flight (TOF) detectors and an ionization chamber (E).

In contrast to other mass-spectrometric techniques, the acceleration and the stripping process leads to the destruction of molecular isobars. With the use of the second mass spectrometer at the high-energy side, the beam is free of molecular interference (the molecule $^{209}BiH$ is destroyed and break-up products are deflected by the second magnet when measuring the radionuclide $^{210}Bi$).
Isotopic interference is still possible due to charge-exchange reactions in the residual gas (e.g. “leaking” $^{209}Bi$ in case of $^{210}Bi$ detection, i.e. $^{209}Bi$ is injected as $^{209}BiH^-$ into the tandem, charge exchange at the high-energy side allows the break-up product $^{209}Bi$ to enter the particle detector although the setup is tuned for mass 210). Such interferences are strongly reduced by additional filters. At VERA a Wien-filter, a double-focusing electrostatic analyzer and additional magnetic deflectors are available for this purpose. However, a finite number of isotopic background particle still enter the detector. Time-of-flight (TOF) technique is applied to discriminate isotopic interferences: different masses with the same energy (constricted by the preceding filters) are distinguished by their different flight time.
Atomic isobars (e.g. $^{210}Po$ is the isobar to $^{210}Bi$) cannot be removed from the beam by selective filtering. However, further reduction of isobaric and isotopic interferences for lower masses can be achieved by a dedicated particle detection system. The different energy-loss of isobars is utilized for that purpose: The now widely-used silicon nitride membranes (SiN) offer an unprecedented homogeneity. New developments in compact ionization chambers with significantly improved energy resolution, allow a substantial reduction of isobaric interferences.

The typical isotope ratios in AMS are between $10^{-11}$ and $10^{-16}$, depending on the radionuclide under investigation. This ultra-sensitivity allows quantifying natural concentrations of long-lived radionuclides in our environment (the natural concentration of $^{14}C$, $^{14}C/^{12}C = 1.2\times10^{-12}$). It is also this dynamical range, which makes AMS a versatile tool for many applications. The detection limit in most cases is given by the machine or sample-background signal registered with the particle detector and varies strongly with the specific radionuclide measured. Together with samples of unknown isotope ratios, reference materials with well-known ratios are measured in periodic intervals as well. They serve as standards to check long-term drifts of the beam transmission through the beamline and provide the absolute scaling factor for the final ratios. Another important parameter is the overall-efficiency, i.e. the fraction of particles detected relative to those available in the sputter sample. It depends on the negative ion formation probability and the transmission of the particles through the AMS facility up to the particle detector and varies strongly for different isotopes, and ranges between $10^{-4}$ and up to 10% (see Table 1).

***Table 1.*** *Basic features of some radionuclides measured at VERA*

| Radio-nuclide | Half-life (yr)[1] | Overall efficiency[2] | Detection limit[3] | Precision | Remark |
|---|---|---|---|---|---|
| $^{10}Be$ | $(1.36 \pm 0.05)*10^6$ | $> 1\times10^{-3}$ | $<10^{-15}$ | < 2 % | isobar $^{10}B$ |
| $^{14}C$ | $(5730 \pm 40)$ | $1\times10^{-1}$ | $< 3\times10^{-16}$ | < 0.5 % | no stable isobar* |
| $^{26}Al$ | $(0.71 \pm 0.02)*10^6$ | $3\times10^{-4}$ | $< 6\times10^{-16}$ | < 1.0 % | no stable isobar* |
| $^{36}Cl$ | $(3.0 \pm 0.02)*10^5$ | n.m. | $< 10^{-13}$ | 2-5 % | isobar $^{36}S$ |
| $^{41}Ca$ | $(1.03 \pm 0.05)*10^5$ | n.m. | $< 10^{-13}/ 10^{-15}$ | 2-5 % | isobar $^{41}K$ $CaF_2$/ $CaH_2$ |
| $^{55}Fe$ | $(2.73 \pm 0.03)$ | n.m. | $< 1\times10^{-15}$ | < 2 % | no stable isobar* |
| $^{129}I$ | $(15.7 \pm 0.4)*10^6$ | $1\times10^{-2}$ | $2\times10^{-14}$ | 2 % | no stable isobar* |
| $^{182}Hf$ | $(8.90 \pm 0.09)*10^6$ | $1\times10^{-4}$ | $1x10^{-11}$ | 10 % | isobar $^{182}W$ |
| $^{210m}Bi$ | $(3.04 \pm 0.06)*10^6$ | n.m. | $< 5\times10^{-12}$ | 10 %[4] | no isobar |
| $^{236}U$ | $(23.42 \pm 0.03)*10^6$ | $5\times10^{-4}$ | $< 2\times10^{-12}$ | 3 % | no isobar |
| $^{244}Pu$ | $(80.0 \pm 0.9)*10^6$ | $5\times10^{-4}$ | --[5] | 3 % | no isobar |

* no stable negatively charged isobar is extracted from the source (e.g. $^{14}N^-$ is not stable).
[1] see e.g. Ref. [13]
[2] atoms detected per atoms in sample; 'n.m.' means 'not measured'.
[3] expressed as isotope ratio
[4] detection of $^{210m}Bi$ is under development.
[5] $^{244}Pu$ background is less than 1 count per hour measuring time.

AMS measurements are performed for radionuclides with half-lives between a few years and up to hundred million years. It reflects that this technique offers a powerful tool through ultra-low isotope-ratio measurements irrespective of half-lives and decay schemes of reaction products. In combination with the very low masses needed, AMS measurements of long-lived radionuclides represents a technique with a much higher sensitivity compared to decay-counting techniques. This can easily be seen from the following calculations: 1 mg of carbon contains $5\times10^{19}$ $^{12}C$ atoms. At natural levels, the isotope ratio of $^{14}C/^{12}C = 1.2*10^{-12}$ converts into a total of 60 million $^{14}C$ atoms in 1 mg carbon. An overall efficiency of 5 percent (see Tab. 1), corresponds to a total of 3 million $^{14}C$ counts in principle to be registered. At natural levels a $^{14}C$ count rate of 100 per second is measured with the particle detector in AMS. In order to get a statistical uncertainty of 1% only 100 seconds of counting time are needed. If decay counting is used, and assuming a detection efficiency of 100%, a count rate of $3\times10^{-4}$ (i.e. an activity of 0.3 mBq) is measured. To get the same statistical uncertainty, a counting time of

more than 1 year is needed! To summarize, AMS is orders of magnitude more efficient for long-lived radionuclides compared to decay counting techniques. This is mainly a consequence of the long half-life and the small sample masses needed.
For some radionuclides Table 1 lists basic features of interest in AMS for the specific facility VERA: Their half-lives span a range from 2.7 years ($^{55}$Fe) to 80 million years ($^{244}$Pu). Overall efficiency means the fraction of particles in a sample, which can be counted with the particle detector. The next column in Fig. 1 depicts the detection limit, either determined by isobaric interferences or by the instrumental background. Also listed is the precision of the AMS measurement if not limited by counting statistics.
The dedicated AMS facility VERA provides the ability for quantifying nuclides over the whole mass range [3]. Measured radioisotopes include $^{10}$Be, $^{14}$C, $^{26}$Al, $^{36}$Cl, $^{41}$Ca, $^{55}$Fe, $^{129}$I, $^{182}$Hf, $^{210m}$Bi, $^{236}$U and $^{244}$Pu. AMS facilities based on larger tandems allow to quantify a few additional isotopes in the medium mass range where suppression of isobaric interferences asks for higher particle energies.

## Comparison of AMS with other techniques for cross-section measurements

In particular, well-established data on production-rates and cross sections of long-lived radionuclides are often highly desired. Especially long-lived radionuclides have often been inaccessible to decay counting techniques, e.g. because of low activity or an unfavorable decay scheme. Cross-section measurements can be classified into two complementary techniques: direct and indirect methods. The direct method makes use of the detection of the prompt and characteristic radiation associated with the production of a specific nuclide, or selectively detects the reaction product itself by means of the recoil separator technique. This method is characterized as an on-line technique. A second and independent method makes use of the activation technique, with sample irradiation and subsequent measurement of the reaction product. After the irradiation the number of produced radioactive nuclei can be quantified either by decay-counting or by mass spectrometric methods. This method is mostly restricted to radioactive products; however, it represents a very sensitive technique due to potential long irradiation periods.
AMS represents an independent and complementary method to the above mentioned direct measurements. A comparison of AMS results allows studying systematic contributions to the total uncertainty associated e.g. with direct methods, which otherwise are hard to quantify. In contrast to other mass spectrometric techniques, like e.g. ICP-MS, AMS is characterized by a low measurement background and, most important, it is not affected by isobaric molecular interferences.

## Applications of AMS in nuclear physics and nuclear astrophysics

Improved and highly accurate nuclear data are urgently required for the design of advanced reactor concepts (Gen IV, ADS) or for the design of nuclear fusion devices, like ITER. The determination of cross sections via the combination of the activation technique and AMS represents an important indirect method. Similarly, there is a clear need for more data to support our understanding of nucleosynthesis processes in astrophysical scenarios. One important contribution to the study of nuclear processes occurring in stars can be provided by measurements of nuclear reactions at accelerator based facilities in combination with AMS.
AMS is also used to quantify absolute concentrations of long-lived radionuclides in a bulk material: Concentrations of rare radionuclides in our environment have the potential to provide unique information. The measurement of such radionuclides allows tracing anthropogenic activities or environmental processes. Similarly, spurious amounts of extra-terrestrial input into terrestrial archives can be investigated by AMS. In the following, with a few examples related to nuclear physics and nuclear astrophysics, a sketchy overview of AMS applications in this field is given.

### $^{235}$U(n,$\gamma$)$^{236}$U – a prime example for actinide measurements

Existing data for the capture channel of $^{235}$U have been measured by time-of-flight techniques via detection of the prompt capture $\gamma$-rays. A major difficulty in these experiments is the safe discrimination against the strong $\gamma$-background from the competing fission channel, therefore those data might suffer from systematic uncertainties. Using AMS, any interference from the fission channel is completely excluded. Activations can be performed with very small samples of natural uranium. This method for measuring the neutron-capture cross section of $^{235}$U has

the advantage that the involved systematic uncertainties are in no way correlated with the uncertainties inherent to the TOF technique. In particular, measurement methods for the detection of the long-lived radionuclides were established at VERA, where suppression of isobars is not required (no stable isobar for $^{236}$U detection) [4]. To suppress interference from neighboring masses (isotopic interference by $^{235}$U and $^{238}$U), the resolution of VERA was recently increased, both by improving the ion optics of existing elements and by installing a new electrostatic analyzer after the analyzing magnet. Interfering ions which pass all beam filters are identified with a high-resolution time-of-flight (TOF) system. In collaboration with various neutron-producing facilities, AMS measurements are performed for neutron energies between thermal and 500 keV.

**$^{209}$Bi(n,γ)$^{210m}$Bi – of interest for ADS and nuclear astrophysics**

The reaction $^{209}$Bi(n,γ) leading to the short-lived $^{210g}$Bi ($t_{1/2}$ = 5 days) and the long-lived isomer $^{210m}$Bi ($t_{1/2}$ = 3.0 Myr) is studied at VERA for thermal energies and in the keV energy range. In the design of future high-power spallation sources a eutectic Pb-Bi target is envisaged, also Pb-Bi may be used as a coolant for accelerator driven systems. The neutron capture reactions on Pb and Bi were considered of key relevance for the design of such systems. In addition, the neutron capture reaction $^{209}$Bi(n,γ) terminates the s-process in nucleosynthesis, because no stable or sufficiently long-lived nuclide can further be produced via slow neutron capture processes (see e.g. [5,6]). Although no stable isobar exists for $^{210}$Bi, the decay product of $^{210g}$Bi, $^{210}$Po ($t_{1/2}$ = 138.4 days), with a much higher negative ionization yield, interferes with $^{210m}$Bi in AMS measurements. The low cross-section value requires suppressing efficiently interference from the neighbouring mass $^{209}$Bi (like for $^{236}$U measurements, see above). Measurements have been started first to investigate the thermal cross section [7].

**Neutron-capture reactions in the keV energy range for s-process studies**

The measurement of neutron and proton capture cross-sections relevant to nuclear astrophysics has become one main research topic at VERA. This work continuous and adds to previous measurements e.g. performed at Rehovot, Argonne and Munich [8-10]. Various samples have been irradiated for that purpose at Forschungszentrum Karlsruhe [11]. After the activation the amount of longer-lived radionuclides is quantified using the technique of AMS. Examples are $^{9}$Be(n,γ)$^{10}$Be, $^{13}$C(n,γ)$^{14}$C, $^{14}$N(n,p)$^{14}$C, $^{40}$Ca(n,γ)$^{41}$Ca and $^{54}$Fe(n,γ)$^{55}$Fe in the neutron energy range from thermal to keV [12,13]. These reactions are of interest in Big-bang nucleosynthesis and in the slow-neutron capture process (s-process) in stars.

**Long-lived radionuclides as activation products in a fusion environment**

In a fusion environment particularly long-lived activation products may lead to significant long-term waste disposals and radiation damage. Many of these production cross sections are not well-known making it difficult to calculate concentration limits [14]. With the high neutron flux in a fusion reactor also impurities in structure materials may lead to significant or dominating activations. For such nuclides production cross-sections and induced activities are key parameters for safety and design analyses. At VERA a program to measure cross sections for various long-lived radionuclides in the neutron energy range relevant for nuclear fusion is ongoing. Within that program, in collaboration with various neutron-producing facilities (e.g. IRMM Geel, Forschungszentrum Dresden-Rossendorf, Forschungszentrum Karlsruhe), cross sections are measured for reactions like $^{14}$N(n,p)$^{14}$C, $^{27}$Al(n,2n)$^{26}$Al, $^{54}$Fe(n,np+d)$^{53}$Mn, $^{56}$Fe(n,2n)$^{55}$Fe, $^{60}$Ni(n,2n)$^{59}$Ni, etc. for neutron energies from 13 to 20 MeV [15,16].

**AMS measurements in environmental samples for nuclear forensic studies**

The concentrations of rare radionuclides in our environment have the potential to provide unique information. The measurement of such radionuclides allows tracing anthropogenic activities or environmental processes. Man-made radionuclides will enter the environment via different processes, e.g. from nuclear weapons tests, as accidental local fallout products, from nuclear-fuel reprocessing plants, or from industrial or medical applications. For the trans-uranium elements like plutonium, by far their largest signals stem from artificial sources. Different kinds of origin are reflected in different signatures like isotopic ratios and concentration levels [17,18]. Its detection represents therefore a proper means for proving human activity. Besides $^{238}$Pu, all other Pu isotopes, i.e. from $^{239}$Pu to $^{244}$Pu, do not suffer from stable isobaric interferences.

**Search for long-lived radionuclides as signatures of a recent close-by supernova explosion**

Some ten years ago, it was pointed out that some radionuclides produced in a supernova explosion might have been incorporated into terrestrial archives [19]. We continue to explore the AMS detection of very feeble natural traces of such long-lived radionuclides, like $^{244}$Pu ($t_{1/2}$ = 81 Ma) and $^{247}$Cm (15.6 Ma). These isotopes may be present on Earth in suitable archives (e.g. deep-sea sediments [20] and deep-sea manganese crusts [21]) as the remnants of supernovae, which happened "close" in space and time (<100 light-years, < 100 million years ago). Such a finding would be of great interest in nuclear astrophysics complementing the recent detection of possibly supernova-produced $^{60}$Fe [22]. The principle of the measurement is similar as for environmental samples. The expected extremely small concentrations makes AMS the favorite method.

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