

AMS of ^{93}Zr : Passive absorber versus gas-filled magnet

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ABSTRACT

Two different isobar separation techniques were tested for the detection of the long-lived fission product ^{93}Zr ($T_{1/2} = 1.64 \cdot 10^6$ a) using Accelerator Mass Spectrometry (AMS), i.e. a passive absorber and a gas-filled magnet, respectively. Both techniques were used in combination with a Time-of-Flight path for the identification of the stable neighboring isotopes ^{92}Zr and ^{94}Zr . The passive absorber was represented by a stack of silicon nitride foils for high flexibility regarding the thickness for optimal isobar separation. Ion beams with a large variety of energies, between 80 and 180 MeV, were provided for this experiment by the tandem accelerator at the Maier-Leibnitz Laboratory in Garching, Germany. With these beams, the stopping powers of ^{93}Zr and ^{93}Nb as a function of energy were determined experimentally and compared to the results obtained with the simulation program SRIM. Considerable discrepancies regarding the energy dependence of the two stopping power curves relative to each other were found. The lowest detection limit for ^{93}Zr achieved with the passive absorber setup was $^{93}\text{Zr}/\text{Zr} = 1 \cdot 10^{-10}$. In comparison, by optimizing the gas-filled magnet set-up, ^{93}Nb was suppressed by around six orders of magnitude and a detection limit of $^{93}\text{Zr}/\text{Zr} = 5 \cdot 10^{-11}$ was obtained. To our knowledge, these results represent the lowest detection limit achieved for ^{93}Zr until now.

1. Introduction

The development of accelerator mass spectrometry (AMS) has provided an ultra-sensitive tool for the detection of long-lived radionuclides with applications in many fields of science [1–3]. Considering the long half-life of the zirconium isotope ^{93}Zr of 1.64 ± 0.06 Ma [4], its detection by AMS seems favorable compared to decay counting. Activity measurement of low concentrations of ^{93}Zr is also difficult because it decays with a probability of $(73 \pm 6)\%$ via the emission of a low energy beta (maximum energy: 60 or 91 keV [5,4]). ^{93}Zr plays an important role in different fields of research, as for example in the s-process nucleosynthesis taking place in the asymptotic giant branch stars [6]. The reaction flow depends on the cross-section of $^{92}\text{Zr}(n, \gamma)^{93}\text{Zr}$, which has to be known with a high precision for astrophysical models in order to calculate the resulting neutron density. However, experimental data on this cross-section are still limited [7]. Additionally, ^{93}Zr is a relevant topic in nuclear waste management as it is produced with a high yield in nuclear fission [8], as well as by

neutron capture on ^{92}Zr since most power reactor designs use Zircaloy as fuel rod cladding [9]. Thus, studies of its migration behaviour in the environment in case of accidental releases from a final repository are needed. Recently, the utilization of ^{93}Zr as reactor neutron dosimeter for advanced nuclear reactor designs in the energy range between 1 keV and 1 MeV, has been proposed [10]. Furthermore, a possible new application of ^{93}Zr is the dating of geological samples for a time-scale shorter than the common uranium-lead method [11]. All these applications require a highly sensitive detection method in order to measure minute amounts of ^{93}Zr .

Whereas many other long-lived radionuclides like ^{14}C , ^{10}Be , ^{26}Al etc. are routinely analyzed by AMS, there are only preliminary studies to develop the method for ^{93}Zr [12–14]. This is mainly due to interfering background of the stable isobar ^{93}Nb and the two stable isotopes ^{92}Zr and ^{94}Zr , which makes the clear identification of ^{93}Zr extremely challenging. In this work, we present our studies to develop the AMS method for ^{93}Zr which reaches sufficiently high sensitivities to allow measurements of samples with widespread applications. For this

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purpose, we compared the detection of ^{93}Zr using a passive absorber with our gas-filled magnet system.

2. Sample material

For the detection of ^{93}Zr , ZrO_2 enriched in ^{92}Zr (94%) and with only a small concentration of ^{94}Zr (1%), was irradiated for different time lengths at the OSIRIS research reactor situated at the CEA Saclay research center. The exact irradiation history of the samples can be found in Ref. [10]. As an ICP/AES analysis of the enriched material before the irradiation found a rather high concentration of ^{93}Nb of 167 ± 17 ppm, Nb was chemically separated by anion exchange chromatography [15] reducing the Nb concentration to a few ppm [16]. This material served as ^{93}Zr reference sample in the following experiments.

In order to determine the sensitivity in our experiments, two different blank materials were available: ZrO_2 powder with natural isotopic composition to which no dedicated Nb separation chemistry was applied and the same enriched ZrO_2 as used for the irradiation which was chemically prepared correspondingly to the reference sample to allow a direct comparison.

The passive absorber was represented by a stack of SiN foils (10×10 mm window, 1038 ± 5 nm thickness each, with a density of 2.7 ± 0.1 g/cm 3 [17]) purchased from Silson Ltd., Northampton, England.

3. Methods

3.1. AMS with ^{93}Zr at the Maier-Leibnitz-Laboratory

The AMS setup at the Maier-Leibnitz-Laboratory (MLL) in Garching, Germany, which is schematically shown in Fig. 1, consists of a single-cathode Cs-sputter ion source, a 90° injector magnet, an 18° electrostatic deflection, a 14 MV tandem accelerator, a 90° analyzing magnet, and several Wien filters for isotopic background suppression. After the switching magnet there are two different setups dedicated to the ion

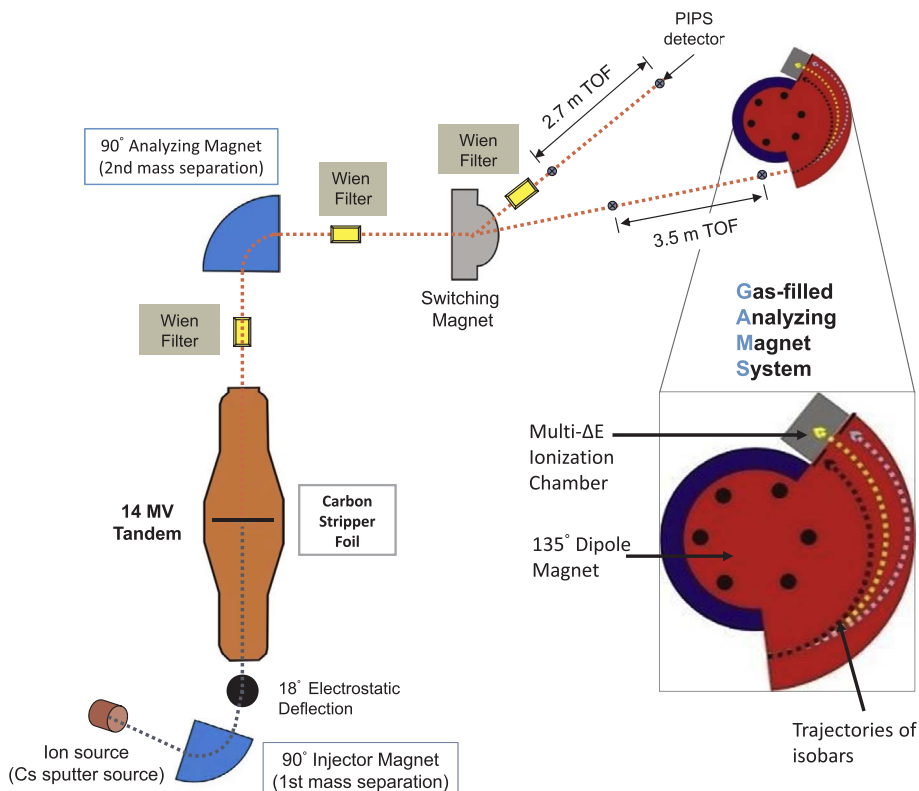


Fig. 1. Schematic drawing of the AMS setup at the MLL in Garching (not to scale). Blue and red dashed lines indicate negatively and positively charged ions, respectively. In the gas-filled magnet, the ions form an average charge state indicated with a yellow dashed line, compared to particles with higher and lower Z (black and grey dashed lines, respectively). Refer to the main text for a more detailed explanation of the gas-filled magnet. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

identification for AMS. The first beam line is equipped with a Time-of-Flight system developed for the detection of actinides and the search for super-heavy elements [18,19], for which no stable isobaric background occurs. The second beamline uses a gas-filled magnet system for the separation of the radioisotope from their stable isobars [20,21]. An additional Time-of-Flight path is installed in front of the gas-filled magnet.

Zr was extracted as ZrO^- from a ZrO_2 matrix mixed with Cu powder at a volume ratio around 1:1, which showed sufficiently high and, most important, stable currents. In that way, a few 100 nA of $^{90}\text{ZrO}^-$ were measured in the Faraday cup after the first 90° magnet. In contrast, hydride and carbide molecules of ^{90}Zr showed currents below 50 nA in this Faraday cup. From a zirconium fluoride matrix currents of ZrF_4^- were obtained which were in the same order of magnitude as ZrO^- extracted from ZrO_2 . However, ZrF_4^- yields were not stable and quickly dropped (within 2 h of sputtering) below 5 nA, probably due to surface effects taking place at the ionizer. In order to find the maximum sensitivity, the beam energies provided by the accelerator have been varied between 80 MeV and 180 MeV. After selecting the respective p/q ratio with the high-resolution 90° analyzing magnet ($\Delta m/m = 1/2500$), the beam was directed to one of the AMS beam lines. If required, the beam intensity of the stable isotopes could be attenuated by inserting steel plates with different densities of small holes which define the attenuation factor (about 1150 for 2 attenuators and 33 for the third one).

3.2. Time-of-Flight (ToF) with a passive absorber

The start signal of the ToF was generated by a micro-channel plate (MCP) detector when the ions passed through a thin carbon foil producing secondary electrons. A PIPS (Passivated Implanted Planar Silicon) detector from Canberra coated with a 25–35 nm thick aluminum layer for better timing performance and an active area of 450 mm 2 , was used both as stop detector and for the energy measurement. With this detection system, a ToF resolution of 0.6 ns FWHM was achieved. The Wien filter installed in the ToF beam line could not be

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