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He stripping for AMS of ^{236}U and other actinides using a 3 MV tandem accelerator

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ABSTRACT

Interest in the long-lived radioisotope ^{236}U ($t_{1/2}$ = 23.4 million years) has significantly increased recently, due to the emergence of environmental and earth science applications. Presently, only a few (AMS) accelerator mass spectrometry instruments are suited for this measurement. One major limitation is the relatively low total detection efficiency (on the order of 10^{-4}), which is partly caused by a low stripping yield of the positive ion charge state selected after the tandem accelerator. It has been shown that high yield can be achieved using helium as stripper gas for uranium ion energies below 0.35 MeV. Here we investigate the potential of He stripping of U at the 3-MV tandem accelerator VERA. Phenomenological charge state distributions for U and Th are presented for terminal voltages from 1.0 to 1.7 MV. These terminal voltages provide better background rejection than possible below 1 MeV, and are suited to the widely used 1–3 MV workhorses of many accelerator mass spectrometry laboratories. The methods can be applied to other actinides also.

1. Introduction

Decay counting, ICP-MS (inductively coupled plasma mass spectrometry) and TIMS (thermal ionization mass spectrometry) methods have been used for most investigations on environmental actinides [1], but accelerator mass spectrometry (AMS) has long been used when exceptional sensitivity is required [2,3]. Recently, new applications of these isotopes at the lowest abundance levels have brought the technique into focus.

^{236}U can be produced by (thermal) neutron capture from ^{235}U and by (n,3n) reactions of high-energy neutrons on ^{238}U. ^{236}U/U from natural production by cosmic rays is expected to be quite low [4], and has escaped detection so far, but the ratios derived from radiogenic production in high grade ores are within the capabilities of suitable AMS instruments [5,6]. The most recent environmental applications evolved around the discovery of ^{236}U from global fall-out [7,8], with a strong focus on oceanography [9–11], but also first soil science studies [12,13] and first measurements in ice cores have been published recently [14].

For ^{236}U, AMS measurements of ores and environmental samples have demonstrated the capability to determine isotopic abundance levels down to ^{236}U/U = 10^{-12}, which are far beyond the capabilities of other methods. Still, it is yet unclear where the real abundance limit of AMS is, since no material with an isotopic ratio below 10^{-12} has been separated so far. Depending on sample size and isotopic ratio, the final detection limit can be imposed by chemistry background, instrumental background caused by incomplete suppression of the naturally abundant uranium isotopes, and atom counting efficiency. The factors contributing to efficiency are: (1) the chemical yield, for which typically more than 50% are reported; (2) the negative ionization yield in the ion source, 0.3% as estimated by Fifield et al. [15]; (3) the yield of the charge state selected after the tandem accelerator, e.g. 5% for $^{236}U^{5+}$ at 3.0 MV terminal voltage [16]; (4) the ion optical transmission through the instrument, which however may approach 100% in dedicated AMS machines; and (5) the detector efficiency between 15% and 100%, depending on the type of particle detector used. The AMS group at ETH Zurich [17] has shown that a significant improvement in efficiency is achieved by using the 3+ charge state and helium as stripper gas when using a terminal voltage of approximately 0.3 MV.

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We have implemented a helium gas stripping system for our AMS instrument VERA based on a 3 MV tandem accelerator. Since most background processes in actinide AMS arise from interactions of unwanted ions with residual gas, with cross sections generally decreasing with higher energy, an advantage of higher energy is expected. In our case the limitation to particle energy is the analyzing magnet with a mass energy product of 176 MeV amu. While this allows transporting the 5+ charge state at the design terminal voltage of 3 MV, we are limited to 2.5 MV for the 4+, and 1.7 MV for the 3+ charge state. Our terminal voltage for the 3+ charge state of actinides is 3–5 times higher than the voltage range studied by Vockenhuber et al. [17] for actinide transmission.

This study aims at demonstrating the potential of helium gas stripping of uranium to the 3+ charge state at these energies with regard to enhancing the AMS technique for ^{236}U. To gain insight into the physics behind the special behavior of helium stripping, data on thorium is also presented. Although abundance sensitivity is not as crucial for the other actinides Pu, Am, Np, *etc*, because no naturally abundant isotopes of these elements exist, their measurement will also gain from higher stripping yields. We look at the key background problems and their possible resolution, and give the resulting limits of the method.

2. Experimental

The vacuum system of VERA is based on cryo-pumps. While these provide excellent pumping speed and unproblematic handling, they are unsuited to pump helium, which impeded continuous operation with helium as stripper gas. Consequently, the pumps before and after the tandem accelerator were replaced by turbo-molecular pumps (Pfeiffer HiPace 1200). These pumps provide a compression ratio above 10^8 for air, which makes sure that measurements of other AMS isotopes, usually with argon as stripper gas, are not compromised. For helium, a compression ratio of 2×10^5 is specified. The dry forepump (Adixen ACP 28), which achieves an operating pressure below 10^{-2} mbar for helium stripping, prevents contamination with pump oil also in case of accidents.

VERA is equipped with a stripper gas handling system that allows changing the gas without opening the tandem accelerator. This is made possible by a PFA line with valves both at the terminal stripper gas bottle and on the ground side. While high voltage is present at the terminal, this line must be filled with an insulation gas (N_2 at 10 bars). After slight modifications of the original design (NEC SDH-9 pelletron manual, National Electrostatics Corporation, Middelton, WI, USA) we are now able to switch between different stripper gases within half an hour.

Except for some initial experiments, so called shorting rods were used for terminal voltages below 2.0 MV. These rods shorten out the rear end of the acceleration tubes before and after the terminal, thus maintaining the field gradients at the entrance of the tubes at the design values for good ion optics. We consider the ratio between injection energy and terminal voltage to be the relevant measure for choosing the shortened fraction. VERA is designed for 70 keV injection energy, and 3.0 MV terminal voltage. For actinides, we reduce the injection energy to 56 keV, which is the limit imposed by VERA's injection magnet. This suggests shortening about 60% of the accelerator tube sections for 1.0 MV and about 40% for 1.4 MV, respectively. Nevertheless, during the experiments presented here, 4/9 of the tubes were shortened as a compromise for both voltages. For the 3+ charge state at 1.65 MV (for transporting mass 238) 3/9 of the acceleration tubes were shortened.

The original appliance for shorting rods consisted of steel and nylon rods of 1/9 length of the accelerator tubes which are screwed together and pushed through O-ring sealed feedthroughs into the accelerator structure. Although this equipment is to our knowledge routinely used at many other accelerators, we found it too cumbersome and prone to failure for routine AMS operation. Additionally, once the optimum conditions for actinide measurements were established, we decided to sacrifice the flexibility of shorting segments. For the terminal voltage of 1.65 MV with 3/9 of the acceleration tubes shortened, we machined un-segmented shorting rods. The nylon rods originally employed were avoided by using a steel rod instead, which can be unscrewed and retracted after plugging the actual shorting rod into a slot at the terminal. Deploying or retracting of the shorting rods is now possible in about 15 min.

The maximum achievable transmission of ^{238}U and ^{232}Th to the Faraday cup after the analyzing magnet was determined for the charge states 5+ (at a terminal voltage of 3.0 MV), 4+ (2.45 MV and 2.0 MV), and 3+ (1.7 MV, 1.4 MV, and 1.0 MV). This was done for several different pressures of the stripper gas. Because of ion optical and scatter losses, and losses due to charge exchange on residual gas outside of the stripper, these charge state transmissions are phenomenological and are not fully related to the underlying cross sections. They are however of practical importance for use in AMS measurements.

The lower charge states omitted in the list given above cannot be bent by the analyzing magnet, and could therefore not be investigated by this straightforward approach. In a second set of experiments we employed the Wien filter right after the accelerator to resolve the charge states and assess the complete charge state distributions for our machine, for terminal voltages of 1.0, 1.4 and 2.0 MV. Fig. 2 was obtained by scanning the voltage U applied at the Wien Filter. Very narrow ($\Delta x = 0.2$ mm aperture) slits were used in front of the Faraday cup after the Wien filter (see Fig. 1) to sufficiently resolve the individual peaks with charge states q at voltages U_q; however, the peaks still overlap. Therefore, Gaussian fits $i_q G(U|U_q, \sigma_q)$ with parameters i_q, U_q and σ_q were applied to these peaks to unfold the distribution $i(U)$ into the different charge states. i_q is the maximum current measured for charge state q. The peak width σ_q (in V) corresponds to a spatial beam width s_q of several mm at the slit position, therefore the impact of the finite size Δx of the slit aperture can be neglected. The correspondence between peak width σ_q and beam width s_q is, however, different for different charge states q, which has to be considered when calculating the particle current I_q (in particles per second) of a certain charge state q:

$$I_q = \frac{1}{q\Delta x}\int i_q G(U(x)|U_q,\sigma_q)dx = \frac{1}{q\Delta x}\int i_q G(U|U_q,\sigma_q)\frac{dx}{dU}d$$

where $\frac{dx}{dU}$ is the dependency of the beam position at the slits on the voltage applied to the Wien filter, which is proportional to $\frac{q}{E}$ (with E the particle energy) for constant magnetic field and mass m. For the tandem accelerator E is approximately proportional to $q+1$, thus:

$$\frac{dx}{dU} \approx \alpha\frac{q}{q+1}$$

The proportionality constant α can in principle be calculated from the parameters of the Wien filter, but can be neglected if only the relative proportions of the charge states are investigated.

In the third set of experiments the full separation capability of our high-energy beam line was used. Background and detection efficiency of the AMS system for the 3+ and the 5+ charge state were investigated. The main separation is provided by the analyzing magnet with 1.27 m radius and the ESA with 2 m radius. The Wien filter and the switching magnet do not have sufficient resolution to help separating $\Delta m/m \sim 236$, but provide additional

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