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Heavy ion AMS with a “small” accelerator

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Abstract

Compared to heavy ion AMS at large tandem accelerators (TV $\geqslant$ 8 MV) and for cases where stable isobar interference is absent, a small facility like VERA (TV = 3 MV) is shown to be able to offset the disadvantage of lower ion energy. After testing the system with ^{210}Pb/^{208}Pb isotope ratio measurements, we prove this by a measurement of ^{236}U at natural levels in a material separated before 1918 and therefore free from anthropogenic contamination. An isotopic ratio of $^{236}U/^{238}U = (6.1 \pm 0.4) \times 10^{-11}$ was found for this material.

Keywords: AMS; Uranium; U-236; Joachimsthal

1. Introduction

Among the various methods of mass spectrometry, accelerator mass spectrometry (AMS) is the most sensitive method to measure isotope ratios in the range from 10^{-10} to 10^{-16}. The common belief was that for AMS of heavy elements, the use of large high-energy accelerators was essential. This, however, was questioned at the IsoTrace Laboratory in Toronto, where ^{236}U was first detected at natural levels at a terminal voltage of only 1.6 MV [1]. In [2] this group gives a detailed description of a suggested broad-range precision AMS spectrometer especially adapted to the needs of actinides. The Vienna Environmental Research Accelerator, VERA [3], set up 1995, complies with this design in several important points.

VERA is designed to allow the transport of ions of all elements, from the lightest to the heaviest. However, VERA is based on a 3-MV Pelletron tandem accelerator and can only reach particle energies up to about 20 MeV. With existing detection techniques, isobar separation and identification is not feasible at these low energies (except for the lightest AMS isotope, ^{10}Be). The VERA heavy ion program tries to establish measurement methods which work for the long-lived radionuclides where suppression of isobars is not required. Among these are ^{210}Pb, ^{236}U and all heavier ions where no stable isobars exist.

To suppress neighboring masses, the resolution of VERA was 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

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are identified with a high-resolution time-of-flight (TOF) system, using a 0.6 μg/cm^2 diamond-like carbon (DLC) foil in the start detector, which substantially reduces beam straggling.

We have measured ^{210}Pb to evaluate the heavy ion detection system because well-known spike and dilution material were readily available. Due to the short half-life, applications for competitive AMS measurements may exist only in niches. ^{210}Pb ($T_{1/2} = 22.3$ a) is routinely measured by decay counting [4].

In nature, ^{236}U ($T_{1/2} = 23.4$ Ma) is produced from ^{235}U by the capture of thermal neutrons. The isotope ratio ^{236}U/^{235}U can therefore be used as a monitor of the thermal neutron flux, integrated over geological time scales. Within the last 100 million years, ^{236}U is a much more sensitive monitor than the depletion of ^{235}U due to neutron-induced fission.

At depths below the influence of cosmogenic particles, remaining neutron sources are from spontaneous and induced fission of uranium, and from (α, n) reactions on light nuclei. For natural samples, ratios of ^{236}U/^{238}U between 7×10^{-10} [1] and $\sim 10^{-14}$ are expected. This natural abundance of ^{236}U is likely to be surpassed by anthropogenic contributions in many places, since the ^{236}U/^{238}U ratio in spent nuclear fuel is 0.1–0.5%. Applications of ^{236}U as an environmental tracer were already performed at detection levels above 10^{-8} [5]. AMS measurements of ^{236}U/^{238}U ratios of ores from various uranium deposits are presented in [6].

2. Measurement technique

We refrained from using lead oxide [7] as sputter material, since ^{210}Pb^{16}O$^-$ will unavoidably be accompanied by a beam of ^{208}Pb^{18}O$^-$. Similar arguments rule out PbS, which was the other target material successfully tested in [7]. Since fluorine has only one stable isotope (^{19}F) and since all fluorides show high electron affinities, we decided to investigate PbF_x^- ions. Lead fluoride (PbF_2) can easily be precipitated from an aqueous solution with hydrofluoric acid (HF). Typically we obtain 70 nA of $^{208}PbF_3^-$ from pure PbF_2 targets, put into our 40 position MCSNICS sputter source.

Pure U_3O_8 targets yielded on average 50 nA of $^{238}UO^-$. U_3O_8 was produced by combusting uranylnitrate ($UO_2(NO_3)_2 \cdot 6H_2O$) at 700 °C in air. It is important that the combustion is complete, because remnants of nitrogen form $^{238}U^{14}N^-$, an isobar to $^{236}U^{16}O^-$. Up to now, we have not tried UF_x as target material.

VERA's injector consists of a 45° electrostatic analyzer and a 90° bending magnet. An additional quadrupole doublet in front of the magnet allows for the correction of saturation effects at the magnet pole edges, which are unavoidable considering the large mass range from 12 to 270 amu used. With a new object slit at the optimal ion optical position we obtain a mass resolution of $M/\mathrm{FWHM}(M) \sim 900$ sufficient to resolve actinides [8]. At a 3-MV terminal voltage, the lowest charge state we can bend with our 90° analyzing magnet is 5+. Both for U^{5+} and Pb^{5+} ions we achieved a yield between 1% and 3% using argon gas for stripping at the terminal. This is significantly lower than the equilibrium charge-state yield of ~8% expected by the semi-empirical formula in [9], probably due to angular scattering in our stripper.

Previous measurements for ^{210}Pb [8] were performed with the original small Wien filter after the analyzing magnet intended for light ions only [3]. Recently, we replaced the Wien filter by an electrostatic analyzer with a bending angle of 90° and a radius of 2 m. A gap of 4.5 cm and a maximum plate voltage of ±100 kV is able to bend ions with energy/charge ratios of 4.4 MV. This analyzer almost completely suppresses the background for ^{210}Pb (Fig. 1(a)). All horizontal slits in the analyzer are set to ±2 mm, which results in a nominal resolution $M/\mathrm{FWHM}(M) = 635$ for the analyzing magnet and $E/\mathrm{FWHM}(E) = 1000$ for the electrostatic analyzer. The width of the beam was less than 1 mm (FWHM) at all slits and the beam divergence was 2.0 mrad (FWHM), measured for a $^{197}Au^{5+}$ beam at 18 MeV.

Presently, only TOF detectors can resolve the interfering neighboring masses which happen to pass the ion optical filters. The common type of time pick-off detectors deflects ion-induced electrons from a thin carbon foil onto a microchannel plate. We obtained two such detectors built at the

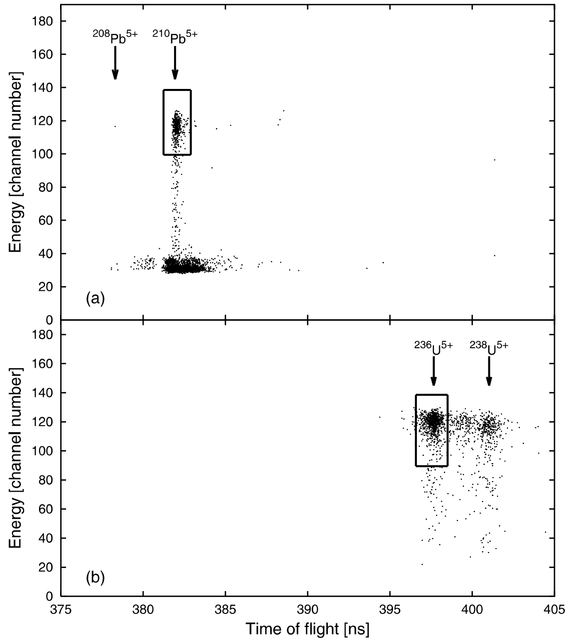

Fig. 1. Time-of-flight versus energy spectra for isotope ratio measurements of (a) $^{210}\mathrm{Pb}/^{208}\mathrm{Pb} = 4.2 \times 10^{-10}$ and (b) $^{236}\mathrm{U}/^{238}\mathrm{U} = 6.1 \times 10^{-11}$. Single events are shown as points. The low-energy events in (a) are due to background ions with mass/charge = 42. 438 counts of ^{210}Pb were collected in 600 s. In (b) 890 counts of ^{236}U were collected in 8400 s. The rectangular windows were set with materials of higher isotope ratios. The low-energy tails are due to incomplete charge collection in the ionization chamber.

LMU Munich [10]. In the start detector we used a DLC foil of 0.6 μg/cm^2 thickness produced by glow-discharge sputtering of graphite in a low-density krypton plasma [11]. The foil was floated onto a stainless steel frame with a fine mesh of nominally 90% transmission and a circular aperture of 12 mm. Investigations on the energy loss in the foil verified the thickness. The low angular scattering allowed a flight path of 1.52 m with reasonably high transmission (see below). A similar foil with 2.0 μg/cm^2 and a diameter of 18 mm was used in the stop detector. After optimizing the electronics, we obtained a time resolution of 340 ps for $^{208}\mathrm{Pb}^{5+}$ at 18 MeV (the separation of neighboring masses of equal magnetic rigidity is 1.8 ns). During the ^{236}U measurement the failure of a fast amplifier reduced the time resolution to about 1 ns. However, this was still sufficient for the measurement. 15 cm after the stop detector, the ion energy is measured by a small gas ionization chamber with 5% energy resolution which is used only to sort out ambiguities in mass/charge ratios. Typical spectra are shown in Fig. 1.

The measurement procedures for ^{210}Pb and for ^{236}U are similar, and shall be described only for ^{236}U. The injector setup for $^{236}\mathrm{U}^{16}\mathrm{O}^-$ was scaled from a $^{238}\mathrm{U}^{16}\mathrm{O}^-$ pilot beam. The analyzer was scaled from a setup found by maximizing the detector count rate for an attenuated $^{238}\mathrm{U}^{5+}$ beam. The optimizations were performed fully automated with our AUTOMAX algorithm [12]. In both the injector and in the analyzer it is impossible to measure ^{238}U-related currents in the respective offset cups when the components are tuned for ^{236}U, because the separation of the beams is too small. Therefore our (hardware implemented) fast beam switching system is not sufficient to switch between ^{236}U and ^{238}U. However, it allows us to switch fast between measurements of $^{238}\mathrm{U}^{16}\mathrm{O}^-$ in an offset cup in the injector and either $^{238}\mathrm{U}^{5+}$ or $^{236}\mathrm{U}^{5+}$ in the analyzer. Software control alternates between these two modes every five minutes. Thus, output variations from the ion source can be traced at a rate of several measurements per second, whereas the variations in the 5+ yield are slow enough for accurate interpolation.

For ^{210}Pb we determined that out of $(1.6 \pm 0.8) \times 10^5$ sputtered ^{210}Pb atoms one atom was actually detected. The largest loss is due to the low ionization yield of $(2 \pm 1) \times 10^{-3}$ for PbF_3^-.

3. Dilution series

To check our techniques, dilution series for both ^{210}Pb and ^{236}U (see Fig. 2) were carried out. AMS usually requires a standard material of known isotopic ratio, which does not yet exist for ^{236}U. Therefore we tried to obtain absolute data by precisely calibrating the detector efficiency. This was done by measuring the ratio of $^{234}\mathrm{U}^{5+}/^{238}\mathrm{U}^{5+}$ with attenuated beams so that only $^{238}\mathrm{U}^{5+}$ is measured as current, while $^{234}\mathrm{U}^{5+}$ is measured as a count rate in the detector. With the resulting detector efficiency of (33 ± 1)% we obtained for the

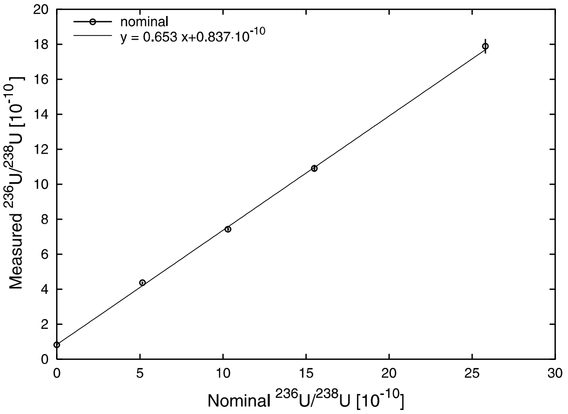

Fig. 2. Nominal versus measured $^{236}U/^{238}U$ ratios for the uranium dilution series.

unspiked uranium reagent used for dilution an efficiency-corrected $^{236}U/^{238}U$ of $(8.4 \pm 0.4) \times 10^{-11}$. This result is compatible with uranium from a uranium mineral deposit, although the origin of the chemical reagent is not specified by the manufacturer. This material was used as a preliminary standard in the following measurements.

The dilution for ^{210}Pb started from a lead solution with $^{210}Pb/^{208}Pb = 2.0 \times 10^{-8}$, whereas pure ^{236}U was available in dried nitrate form (99.97 at.%). In both cases, the dependency between the nominal concentrations calculated from the dilution factors and the measured values, normalized to the preliminary standard, is strictly linear. The linearity shows that for relative measurements the precision is better than 5%. However, the ratio between the measured and the nominal values is only 0.65 ± 0.02. The reason for this deviation has yet to be resolved, and it emphasizes the need for a reliable standard material.

4. A first result

A first sample of uranium of known origin was readily available. We own a fairly large amount (180 kg) of uranylnitrate $(UO_2(NO_3)_2 \cdot 6H_2O)$, which was prepared in the "K.u.K. Uranfabrik Joachimsthal" before 1918. It was separated from uranium ore mined in Joachimsthal (now Czech Republic) and has been stored in sealed glass bottles in our basement since then. It can be assumed to be free of any contamination by anthropogenic ^{236}U. Fig. 1(b) represents the complete statistics collected on this material. Our result for the "K.u.K. Joachimsthal" uranylnitrate is $^{236}U/^{238}U = (6.1 \pm 0.4) \times 10^{-11}$. We have decided to use this material as our reference material in future measurements and would be pleased to share it with other laboratories interested in measuring natural ^{236}U.

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References

[1] X.L. Zhao, M.J. Nadeau, L.R. Kilius, A.E. Litherland, Nucl. Instr. and Meth. B 92 (1994) 249.

[2] K.H. Purser, L.R. Kilius, A.E. Litherland, X.L. Zhao, Nucl. Instr. and Meth. B 113 (1996) 445.

[3] W. Kutschera, P. Collon, H. Friedmann, R. Golser, P. Hille, A. Priller, W. Rom, P. Steier, S. Tagesen, A. Wallner, E. Wild, G. Winkler, Nucl. Instr. and Meth. B 123 (1997) 47.

[4] G. Wallner, K. Irlweck, Radiochim. Acta 78 (1997) 173; X.-L. Zhao, L.R. Kilius, A.E. Litherland, T. Beasley, Nucl. Instr. and Meth. B 126 (1997) 297.

[5] M.A.C. Hotchkis, D. Child, D. Fink, G.E. Jacobsen, P.J. Lee, N. Mino, A.M. Smith, C. Tuniz, Nucl. Instr. and Meth. B 172 (2000) 659.

[6] D. Berkovits, H. Feldstein, S. Ghelberg, A. Hershkowitz, E. Navon, M. Paul, Nucl. Instr. and Meth. B 172 (2000) 372.

[7] R. Middleton, A Negative Ion Cookbook, University of Pennsylvania, 1989 (unpublished).

[8] R. Golser, G. Federmann, W. Kutschera, A. Priller, P. Steier, C. Vockenhuber, in: J.L. Duggan, I.L. Morgan (Eds.), Applications of Accelerators in Research and Industry – Sixteenth International Conference, American

Institute of Physics Conference Proceedings 576, 2001, p. 627.

[9] R.O. Sayer, Rev. Phys. Appl. 12 (1977) 1543.

[10] D. Müller, T. Feastermann, A. Gillitzer, G. Korschinek, E. Nolte, R. Scheuer, Nucl. Instr. and Meth. B 52 (1990) 366.

[11] V.K. Liechtenstein, T.M. Ivkova, E.D. Olshanski, I. Feigenbaum, R. DiNardo, R. Hellborg, R.A. Weller, H.L. Wirth, Nucl. Instr. and Meth. A 397 (1999) 140.

[12] P. Steier, S. Puchegger, R. Golser, W. Kutschera, A. Priller, W. Rom, A. Wallner, E. Wild, Nucl. Instr. and Meth. B 161–163 (2000) 250.