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Accelerator mass spectrometry of the heaviest long-lived radionuclides with a 3-MV tandem accelerator

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Abstract. A 3-MV Pelletron tandem accelerator is the heart of the Vienna Environmental Research Accelerator (VERA). The original design of the beam transport components allows the transport of ions of all elements, from the lightest to the heaviest. For light ions the suppression of neighboring masses was sufficient to measure isotopic ratios of $^{14}C/^{12}C$ and $^{26}Al/^{27}Al$ as low as $10^{-15}$and $^{10}Be/^{9}Be$ down to 10^{-13}. To suppress neighboring masses for the heaviest radionuclides in the energy range of 10 – 20 MeV, the resolution of VERA was increased both by improving the ion optics of existing elements at the injection side and by installing a new high-resolution electrostatic separator at the high-energy side. Interfering ions which pass all beam filters are identified with a Bragg-type ionization detector and a high-resolution time-of-flight system. Two ultra-thin DLC (Diamond-Like Carbon) foils are used in the start and stop detector, which substantially reduces losses due to beam straggling. This improved setup enables us to measure even the heaviest long-lived radionuclides, where stable isobaric interferences are absent (e. g. ^{236}U and ^{244}Pu), down to environmental levels. Moreover, the advantage of a "small" and well manageable machine like VERA lies in its higher stability and reliability which allows to measure these heavy radionuclides more accurately, and also a large number of samples.

Keywords. AMS; U-236, Pu-244

1. INTRODUCTION

Among the various applications of accelerators, Accelerator Mass Spectrometry (AMS) reaches into many different research areas and is therefore one of the most interdisciplinary ones [1]. The combination of mass spectrometers and "small"

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tandem accelerators (0.5 – 15 MV) allows to measure minute isotope ratios (10^{-10} – 10^{-16}) or small amounts (10^5 to 10^8 atoms) of long-lived radionuclides in samples from various spheres of our environment. Since the radioactive atoms are detected directly rather than by their infrequent radioactive decay, the measuring times and sample sizes are reduced by many orders of magnitude compared to conventional decay counting.

The AMS facility at the University of Vienna, the Vienna Environmental Research Accelerator (VERA), is designed to measure many long-lived radionuclides across the whole nuclear chart [2]. The lighter radionuclides of interest, such as ^{10}Be ($t_{1/2} = 1.5 \times 10^6$ a) [3], ^{14}C (5.73×10^3 a) [4] and ^{26}Al (7.2×10^5 a) [5], have been routinely measured for several years with a large number of samples. A first step towards the heavier ions was done by a ^{129}I (1.6×10^7 a) measurement with a time-of-flight system [6]. However, limitations from the ion optics of the injection system and the low resolution of the Wien-filter of the original setup of VERA made measurements of even heavier ions impossible. The necessary modifications of the VERA facility for measurements of even the heaviest radionuclides and first results of the two interesting radionuclides ^{236}U (2.34×10^7 a) and ^{244}Pu (8.1×10^7 a) will be presented.

2. THE VERA AMS FACILITY

VERA is a dedicated AMS facility based on a +3 MV-tandem accelerator built by National Electrostatics Corporation (NEC) in Wisconsin, USA. Different to many nuclear physics applications, the pre-treated sample material of a few mg is put into the ion source and analyzed by two mass spectrometers (called injector and analyzer) linked with a tandem accelerator.

A schematic layout of the VERA facility is shown in fig. 1. The negative ions from the "Multi-Cathode Source of Negative Ions by Cesium Sputtering" (MC-SNICS) are mass selected with a 45° electrostatic analyzer (ESA) and a 90° injection magnet at energies in the 70 keV range and injected into the accelerator. The terminal voltage is measured by a generating voltmeter and can be kept constant at 3 MV ± 300 V. At the terminal the ions loose electrons in a differentially pumped channel ("stripper") filled with a few microbar of argon, where molecular ions are destroyed. The ions with positive charge states are accelerated a second time by the same potential. This results in an energy of several MeV. The analyzing mass spectrometer (similar to the injection side) efficiently removes molecular break-up products. The double focusing 90° analyzing magnet can bend the heaviest elements with charge state 5+ or higher at 3 MV terminal voltage. In the original setup a velocity filter (Wien-filter) followed the analyzing magnet, but due to the low resolution, this filter has been replaced by a high-resolution electrostatic analyzer (see section 3). Finally the selected ions are counted in an appropriate particle detector.

Our relative low energies (10 – 20 MeV) do not allow a separation of interfering stable isobars (except for ^{10}B in the detection of ^{10}Be). For a separation of isobars an energy of at least 1 MeV per nucleon is needed. The main advantage of a small, fully computer controlled machine is the higher (long-term) stability of its components, and that it can be operated by a single person. To achieve the ultimate sensitivity and stability, all components are designed to avoid beam losses. High selectivity is obtained by several elements with lower resolution and high transmission. Furthermore, the possibility to measure up to 40 samples in one

computer controlled batch allows not only to measure a large number of samples, but also to switch fast (within a few minutes) between unknown samples and standard materials. This reduces errors from systematic uncertainties and long-term variations.

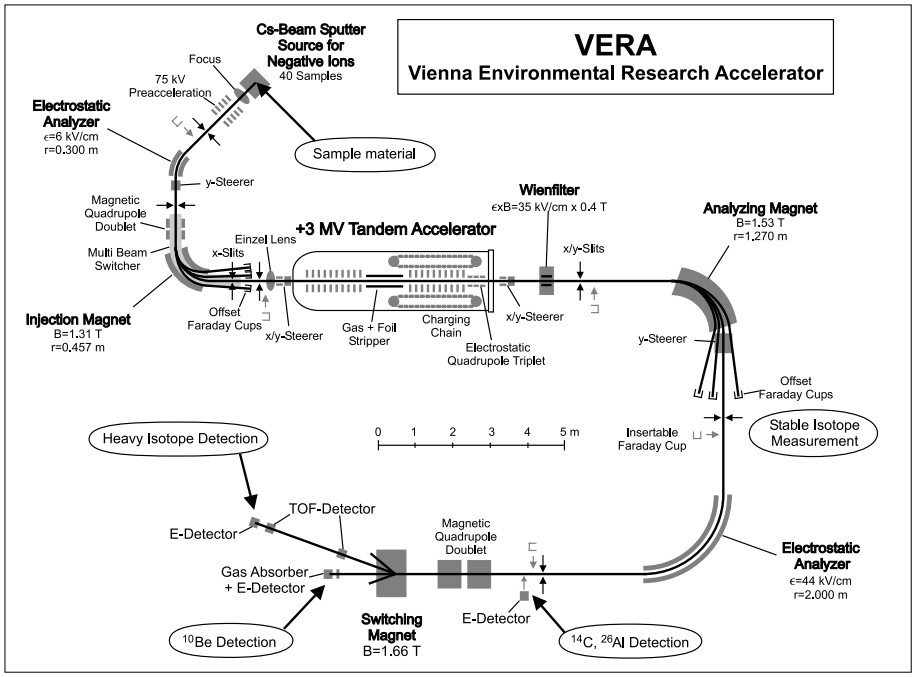

Figure 1. Schematic layout of the VERA facility after the upgrade for heavy ions.

For heavy ions further developments were necessary since the experimental difficulties increase with the mass of the ions. The relative mass difference between the radionuclides of interest and its neighboring isotopes generally becomes smaller for higher masses, so both the injector and the analyzer must provide sufficient resolution. The resolution can be enhanced by reducing the apertures of the slits to the limit of the beam size.

3. UPGRADE FOR HEAVY ION MEASUREMENTS

An important modification was the installation of additional, moveable slits after the injection magnet (see fig. 2). If the opening of the slits are reduced to ~ 1 mm, the mass resolution of our injector can increased be to $M/\mathrm{FWHM}(M) \sim 900$, whereas the transmission is still above 80%. The slits are insulated from the mounting flange and manual controls. This is required, because they are mounted inside the multi Faraday cup chamber, which can be biased up to 13 kV. With the high injection mass resolution, the use of PbF_3^- to avoid simultaneous injection of other lead isotopes and a time-of-flight (TOF) system for additional mass discrimination, we were able to demonstrate the measurement of ^{210}Pb (22.3 a) down to isotope ratios of $^{210}\mathrm{Pb}/^{208}\mathrm{Pb} = 3 \times 10^{-11}$ [7]. These experiences revealed, however, the need of an additional high-resolution filter element at the analyzing side.

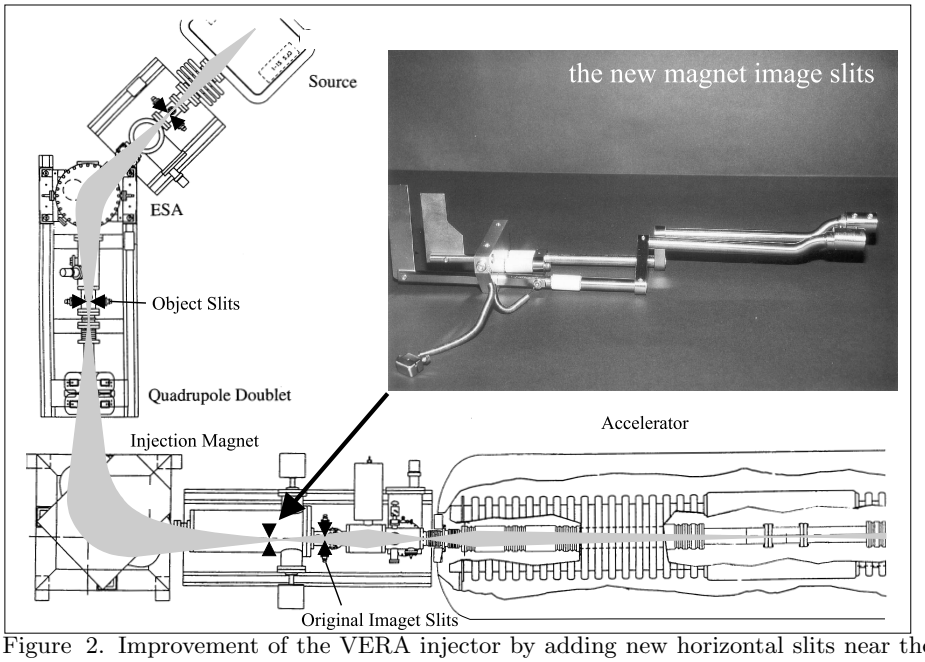

Figure 2. Improvement of the VERA injector by adding new horizontal slits near the real stigmatic beam waist after the injection magnet. The position is 40 cm closer than the original slit assembly, which is outside the multi Faraday cup chamber.

In 2001 we replaced the relatively weak velocity filter (Wien-filter) of the original setup by a high-resolution electrostatic analyzer (ESA) with a bending angel of 90° and a radius of 2 m. The ESA was built by Danfysik in Denmark. The spherical shape of the aluminum electrodes (maximum voltage of ± 100 kV at a distance of 45 mm) provide both horizontal and vertical focussing. The ESA was positioned in such a way, that the image point of the analyzing magnet formed the object point of the ESA. The vacuum chamber is sealed with Viton and reaches a vacuum below 10^{-7} Torr. The good vacuum is essential to reduce charge changing and scattering of the ions inside the ESA. The Wien-filter has been moved to a location between the exit of the tandem accelerator and the analyzing magnet (see fig. 1) for future use as an additional analyzing component. The nominal energy resolution of the ESA, $E/\mathrm{FWHM}(E) = 1000$, together with the high momentum resolution of the analyzing magnet, $p/\mathrm{FWHM}(p) = 635$, provides sufficient separation between the isotopes of even the heaviest elements (if the opening of all horizontal slits at the analyzing side is set to 4 mm). The remaining background stems from charge changing and scattering processes of intense beams that accompany the ions of interest. It can be reduced by additional filters and distinguished by the heavy ion detector.

Since different detectors are used for different ions, a switching magnet (with a preceding magnetic quadrupole doublet) was mounted at the end of the ESA exit beam line (see fig. 1). The heavy ion detector is mounted at the + 20° exit port. For heavy ions the switching magnet fulfills an important function as an additional stage of background suppression (see section 4). The heavy ion detector provides both TOF and energy signals. Only the TOF detector has sufficient resolution to distinguish neighboring isotopes.

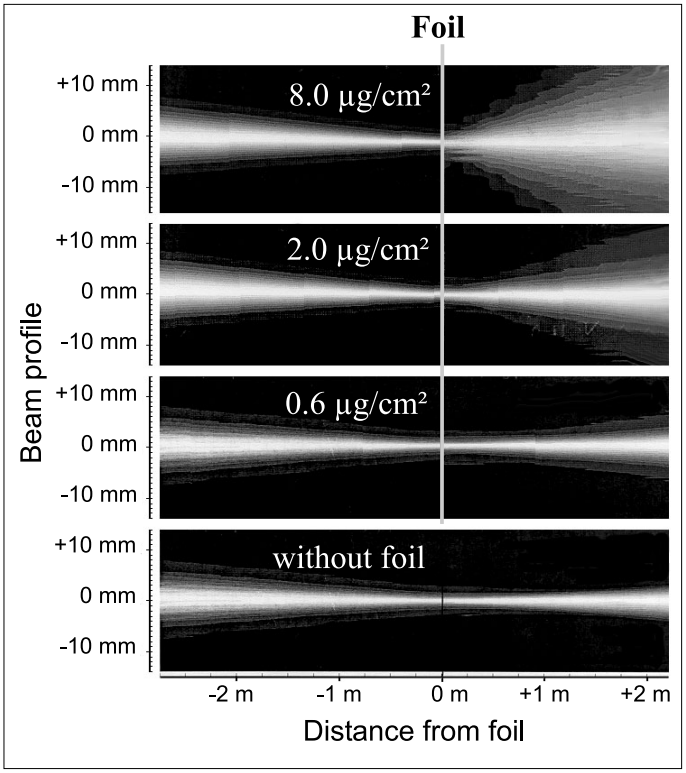

Figure 3. Scattering properties of diamond-like carbon (DLC) foils for 18 MeV ^{197}Au derived from phase space measurements. For the phase space measurement a moveable slit and a NEC-type beam profile monitor have been combined to measure the emittance of the ion beam. As the phase space contains the complete information of the beam, this allows to extrapolate the profile along the optical axis. The thicker foils (2.0 and 8.0 μg/cm^2) have a significant effect on the beam quality, whereas with the 0.6 μg/cm^2 thick foil the beam divergence remains almost unchanged.

The TOF signal is derived from the time-of-flight between two similar timing detectors (start and stop detector) separated by a flight path of 1.5 m. The ions induce secondary electrons at an ultra-thin carbon foil. The electrons are deflected by an electrostatic mirror, collected and multiplied by a micro channel plate. The time resolution FWHM(t) of the whole time-of-flight setup is 300 – 400 ps, which corresponds to a time resolution of FWHM(t)/t = 10^{-3} for e. g. ^{238}U ions at 18 MeV.

The diamond-like carbon foils (DLC foils) used at the start and stop detectors were produced at the Kurchatov Institute in Moscow by glow-discharge sputtering of graphite inside a low-density krypton plasma [8]. These foils are the thinnest carbon foils available anywhere, and are mounted on a flat Cu mesh (1.2 μm wire thickness) of 90% transmission. For the start detector we used a foil with a thickness of 0.6 μg/cm^2 and an aperture of 16 mm. These foils induce minimal energy loss and almost no angular straggling to the passing ions, as shown by phase space measurements (see fig. 3) [9]. This is a main advantage to minimize the losses of ions due to scattering. In addition, we can increase the flight path to get a better relative time resolution. For the stop detector we use a slightly thicker foil (2 – 4 μg/cm^2) with a larger aperture of 18 mm. Because of the short distance to the

final energy detector, angular scattering is not so critical.

For the energy measurement we use a Bragg-type ionization chamber with an energy resolution of FWHM$(E)/E \sim 3$ – 5%, which is better than for a surface barrier detector ($\sim$ 10%). For our purpose the Bragg-type detector is sufficient since it is used to distinguish ions with different charge states. These can still pass the filters, if they have the same M/q and E/q as the wanted ion. At high count rate they can blind the detector. So, for heavy ion measurements it is necessary to work with a charge state q, which shares no common divider with M (for a detailed discussion see [10]). With this system, a detector efficiency of 20 – 30% from the image point of the analyzing magnet to the final detector is achieved.

4. DETECTION OF ^{236}U

^{236}U with a half-life of 23.4 million years is one of the heaviest radioisotopes of interest for AMS. ^{236}U is produced in nature from ^{235}U by neutron capture. The isotopic ratio ^{236}U/^{238}U depends strongly on the thermal neutron flux in the material. This makes the radioisotope very suitable as a neutron monitor. The isotope ratios of ^{236}U/^{238}U in natural samples are expected to be in the range of 10^{-10} – 10^{-14}. The sensitivity is not limited by counting statistics, but by background from other nuclides. For first measurements we used samples from the uranium mine "K. k. Uranfabrik Joachimsthal", from which we still have a large amount (stored in sealed bottles since 1910) in the basement of the Institute for Isotope Research and Nuclear Physics in Vienna. The isotopic ratio is determined by switching between two setups: The $^{238}U^{5+}$ current is measured after the analyzing magnet in a Faraday cup, whereas the $^{236}U^{5+}$ ions are counted in the heavy ion detector. The injected negative ions are $^{238}U^{16}O^{-}$ (mass 254) and $^{236}U^{16}O^{-}$ (mass 252) respectively, which are analyzed as $^{238}U^{5+}$ and $^{236}U^{5+}$ (figure 4).

The main difficulty of the AMS measurement of ^{236}U is the intense neighboring beam of ^{238}U. Although most of the ^{238}U are suppressed at the injector side and by the analyzing magnet a small fraction of this intense beam can interfere the ^{236}U measurement even if the expected separation in the ion-optical filters is large. The main reason for this "leakage" of interfering ions are charge changing processes due to residual gas within the elements. Angular scattering on the residual gas, on electrodes, slits or vacuum chamber walls can also allow background to pass a filter. However, the scattering cross section is in the order of 10^{-20} cm^2 whereas the cross section of charge changing is 10^{-16} – 10^{-15} cm^2 [11]. The leakage can be reduced by simply adding more filter elements, which provides several orders of magnitude of suppression for each additional element.

In the original beam line configuration only the analyzing magnet provided suppression of neighboring masses for heavy ions. Since the separation between $^{236}U^{5+}$ and interfering $^{238}U^{5+}$ after the Wien-filter was only 0.8 mm (the detector aperture was 4 mm at this time), a real ^{236}U measurement was not feasible. In the TOF-versus-energy spectrum (fig. 5a) we see that the tail of a background peak related to ^{238}U extends into the area where the ^{236}U is expected. The counts in this area correspond to a background in ^{236}U/^{238}U of 1×10^{-8}.

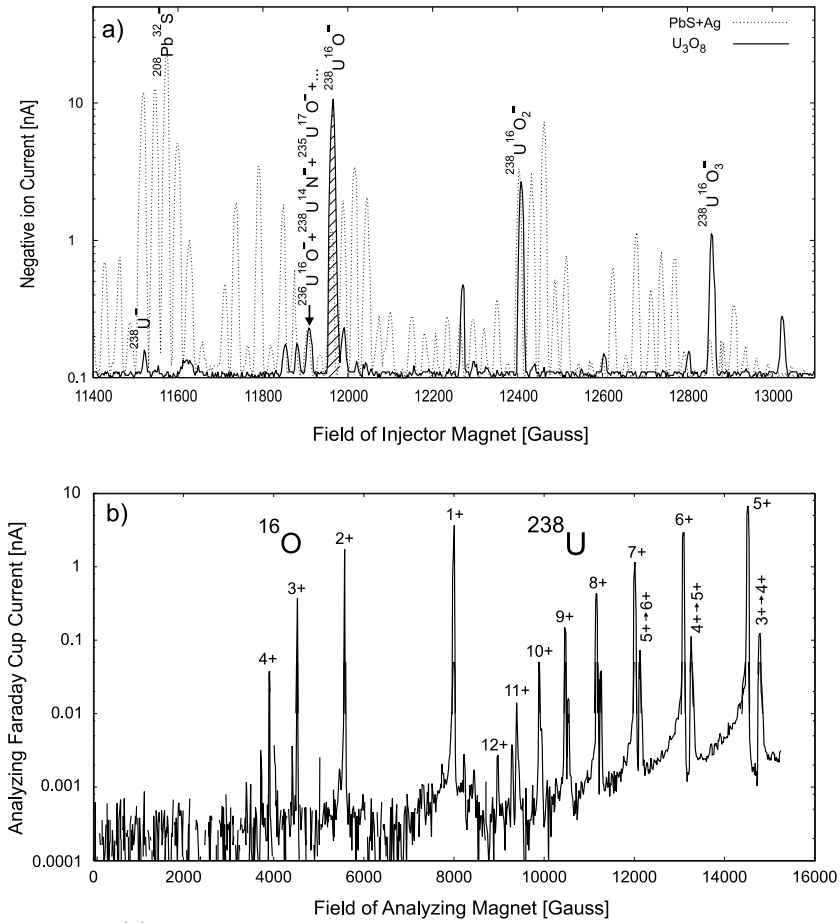

Figure 4. (a) Injector magnet scan of negative ions from a U_3O_8 and from a PbS+Ag sample. The rich negative-ion mass spectrum from PbS+Ag is used to calibrate the mass scale. The $^{238}U^{16}O^-$ peak (mass 254) yields the highest current. For the ^{236}U measurement we inject mass 252 ($^{236}U^{16}O^-$). The strongest contribution to the visible peak is probably due to $^{238}U^{14}N^-$ (the ^{14}N probably stems from the uranylnitrate $UO_2(NO_3)_2 \cdot 6\ H_2O$ used as raw material).
(b) Analyzing magnet scan of the $^{238}U^{16}O^-$ peak (mass 254). At lower magnetic fields there are the different charge states of oxygen (^{16}O) which have gained different energy in the accelerator. For ^{238}U we see the charge states down to 5+ which is the lowest charge state we can bend with the magnet. Between these peaks, we can see a continuum caused by charge changing processes within the accelerator tubes. Every peak has a tail towards lower energies of ions which have changed from a lower charge state to a higher one, e.g. from 4+ to 5+. They have less energy than the regular 5+ ions, and the energy deficit depends on the position where the charge change took place. The probability is highest close to the gas stripper, where the vacuum is worst. The tail ends in a peak at the magnetic rigidity of ^{238}U ions with the energy of regular 4+ ions, but with charge state 5+. This peak is caused by charge changing from 4+ to 5+ between the accelerator exit and the analyzing magnet entrance, which comprises a relatively long section (~ 5 m) of the beam line.

The high resolution of the new ESA (replacing the Wien-filter) reduced this background efficiently. The separation at the image slits of the ESA is $\Delta x = 2r\Delta E/E$.

In our case $^{238}U^{5+}$ with the same magnetic rigidity as $^{236}U^{5+}$ is separated by 34 mm, whereas the opening of the slits is ± 2 mm. In the TOF-versus-energy spectrum (fig. 5b) a clear peak corresponding to $^{236}U^{5+}$ is visible. The measured isotopic ratio $^{236}U/^{238}U$ of $(6.1 \pm 0.1) \times 10^{-11}$ agrees with the expected value for a sample from uranium ore [12]. Additionally there is a peak where we expect $^{238}U^{5+}$ with the same magnetic rigidity (momentum over charge state) p/q as $^{236}U^{5+}$ ($\Delta t/t = \Delta M/M$). In our understanding, these ^{238}U ions must undergo two charge changes to obtain the correct energy to pass the analyzing magnet. The first change from 4+ to 5+ has to take place inside the accelerator column shortly after the terminal (a frequent process due to the comparable bad vacuum there), the second charge change from 5+ back to 4+ happens inside the ESA and causes a change of the bending radius. From a certain position near the exit of the ESA the $^{238}U^{5+}$ ions will pass through the image slits.

Since the ion now have the wrong charge state (4+ instead of 5+) they can be suppressed even by a component of low selectivity. This has been investigated with the switching magnet added as an additional filter element. Since the path inside the switching magnet is short the separation between neighboring uranium isotopes with the same energy is only 2 mm. On the other hand, the residual background before the switching magnet is dominated by ^{238}U ions in charge state 4+ which can be completely suppressed as can be seen in the spectrum of fig. 5c. The ^{238}U peak corresponding to the same magnetic rigidity has disappeared. Pleasingly, the background which we interpreted as $^{238}U^{5+}$ with the same energy (E/q) as $^{236}U^{5+}$ ($\Delta t/t = \frac{1}{2}\Delta M/M$) is also suppressed. For a detailed discussion see [13].

Fig. 5c suggests that we can measure isotopic ratios much lower than 10^{-11}. We plan to demonstrate this with uranium separated from ambient rocks containing much less ^{236}U. To reach an isotopic ratio of 10^{-14} with a realistic $^{238}U^{5+}$ current of 10 nA we will collect one count every 8000 seconds.

5. DETECTION OF ^{242}PU AND ^{244}PU

The long-term stability and reliability of VERA is especially important to detect small amounts of atoms (10^5 – 10^8) in a sample. Often only a few counts of a radionuclide can be collected over a measuring time of hours. The measurements of natural ^{244}Pu with a half-life of 81 million years is such a chase. One application of ^{244}Pu is the search for a recent (within the last several 100 million years) supernova, which may have produced enough ^{244}Pu to leave a detectable signal in slowly accumulating reservoirs such as deep sea sediments.

First measurements of targets containing trace amounts of plutonium show promising results. We measured the $^{242}Pu/^{244}Pu$ ratio in a ^{244}Pu standard prepared at Argonne National Laboratory. The isotopic ratio of $^{242}Pu/^{244}Pu$ determined from α-activity is 0.0112, and this value could be reproduced by our AMS measurement in a sample diluted to contain only $\sim 2 \times 10^7$ atoms of ^{242}Pu. The ^{242}Pu and ^{244}Pu spectra are shown in fig. 6. A total of 191 ^{242}Pu atoms were detected (fig. 6a). Since only part of the sample was sputtered, we obtain a lower limit of the overall efficiency (counts/atoms in sample) of $\sim 3 \times 10^{-5}$, which agrees well with the expected efficiency, and is considerably higher than the efficiency of $\sim 3 \times 10^{-6}$ reported recently for plutonium AMS measurements at a large tandem accelerator [14].

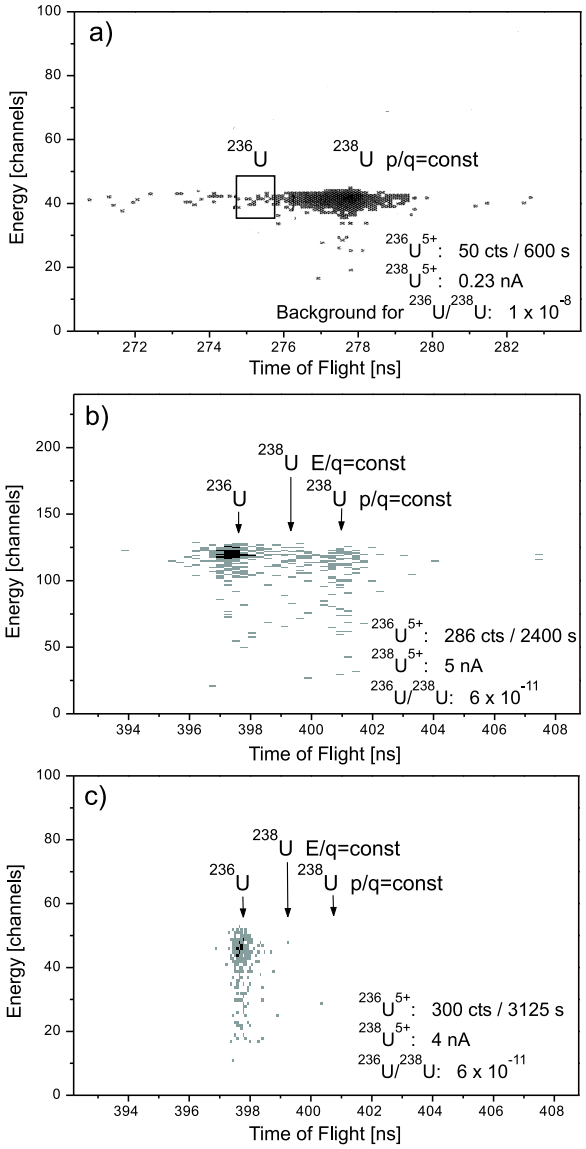

Figure 5. Comparison of energy (E) versus time-of-flight (TOF) spectra measured at different stages of the VERA upgrade for the detection of ^{236}U (injected as $^{236}U^{16}O^{-}$ and analyzed as $^{236}U^{5+}$).
(a) Original setup with analyzing magnet and Wien-filter.
(b) After replacing the Wien-filter with the ESA. Compared to spectra (a) and (c), the TOF resolution in this measurement was worse due to a defective amplifier.
(c) Compared to (b), an additional analysis of the beam is provided by the 20° bend in the switching magnet.

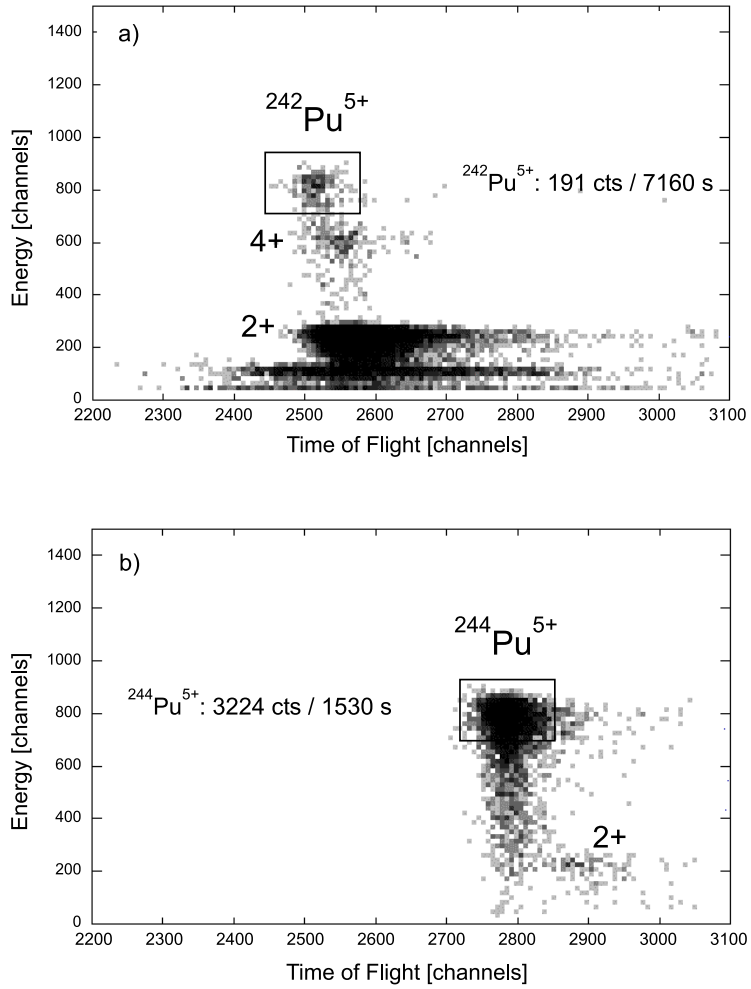

Figure 6. Energy versus TOF spectra measured for a ^{244}Pu calibration sample. The ^{242}Pu/^{244}Pu isotope ratio determined from α-activity was 0.0112. An aliquot containing only $\sim 2 \times 10^7$ ^{242}Pu atoms was measured with AMS, resulting in a ^{242}Pu/^{244}Pu isotope ratio of 0.0127 ± 0.0008.
(a) ^{244}Pu spectrum, injected as $^{242}Pu^{16}O^-$ and analyzed as $242Pu^{5+}$.
(b) ^{244}Pu spectrum, injected as $^{244}Pu^{16}O^-$ and analyzed as $244Pu^{5+}$.

After the upgrade, VERA fulfills the requirements for a universal AMS facility for cases where no stable isobars exist. The good stability of the comparably small machine and the possibility to measure up to 40 samples in one computer controlled batch makes our AMS system suitable for applications where a large number of samples must be measured with high sensitivity.

6. ACKNOWLEDGEMENT

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