Analysis and application of heavy isotopes in the environment

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A R T I C L E   I N F O

A B S T R A C T

A growing number of AMS laboratories are pursuing applications of actinides. We discuss the basic requirements of the AMS technique of heavy (i.e., above ~150 amu) isotopes, present the setup at the Vienna Environmental Research Accelerator (VERA) which is especially well suited for the isotope ^{236}U, and give a comparison with other AMS facilities. Special emphasis will be put on elaborating the effective detection limits for environmental samples with respect to other mass spectrometric methods.

At VERA, we have carried out measurements for radiation protection and environmental monitoring (^{236}U, ^{239,240,241,242,244}Pu), astrophysics (^{182}Hf, ^{236}U, ^{244}Pu, ^{247}Cm), nuclear physics, and a search for long-lived super-heavy elements ($Z > 100$). We are pursuing the environmental distribution of ^{236}U, as a basis for geological applications of natural ^{236}U.

1. Introduction

Accelerator mass spectrometry (AMS) is characterized by its outstanding abundance sensitivity and by the complete suppression of molecular ion species. Depending on the nuclide, the detection limit for heavy, long-lived radioisotopes can be several orders of magnitude below that of the conventional mass spectrometric methods thermal ionization mass spectrometry (TIMS) and inductively coupled plasma mass spectrometry (ICP-MS) and the new method of resonance ionization mass spectrometry (RIMS). Compared to decay counting, mass spectrometry allows one to measure minute isotopic ratios of long-lived nuclides by detecting the atoms directly rather their infrequent decay. Based on realistic assumptions, for actinides AMS can provide a higher detection efficiency then α-spectrometry if the half life is longer than ca. 10^3 years. The high sensitivity extends the domain of applicability to almost every compartment of the environment, where many such radionuclides occur at detectable levels, both of anthropogenic and natural origin.

2. The AMS technique of actinides

VERA, hosted by the Faculty of Physics, Universität Wien, Vienna, Austria, is an example of an AMS facility based on a tandem accelerator and used for AMS [1]. A prerequisite for AMS of actinides is the capability of the bending magnets to transport the heavy ions. The most populated charge state of ^{238}U after gas stripping grows from 1+ to 5+ for terminal voltages between 0.5 and 14 MV terminal voltage, while the energy of this charge state grows from 1 to 84 MeV. To our knowledge, no AMS facility based on a tandem accelerator is presently equipped with a magnet able to bend the most populated charge state of actinide ions at the maximum terminal voltage. At the 3-MV tandem at VERA, this would require 1.77 m radius for the 3+ charge state, whereas only 1.27 m are installed. All laboratories take resort to higher, less populated charge states, with lower magnetic rigidity. In several cases, also the terminal voltage has to be reduced. The resulting lower energy also reduces sensitivity (see below).

At VERA, we use oxygen as stripper gas. As observed first by Betz [2], oxygen yields higher average charge states for very heavy ions. This effect seems to be amplified for the yield of the less populated higher charge states used by AMS. In routine measurements, we observed yields of up to 5% for ^{238}U^{5+}, while argon yields only 3.5%. Higher charge states would be expected for foil stripping, but it suffers from the exceptionally large straggling of the heavy, slow

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ions in the comparably thick stripper foils. While the gas stripper is operated on the order of 0.1 μg/cm² at VERA, the thinnest available carbon foils (0.6 μg/cm², [3]) are still much thicker. This causes losses when we try to transport this beam to the rare ion detector, resulting in the same overall efficiency as with gas stripping.

Since the relative mass difference between adjacent isotopes is smaller for higher masses, higher resolution is required. For actinides, the minimum resolution must be well above 250. This condition is usually satisfied at the high-energy side of most facilities. At VERA, we work with all slits closed to 4 mm aperture except for the image slit after the analyzing magnet, which is set to 6 mm. Using the simple formulas of Ref. [4] this corresponds to a mass resolution of $M/\text{FWHM}(M) = 420$ for the magnet and an energy resolution of $E/\text{FWHM}(E) = 670$ for the ESA. The beam width (FWHM) at the waists is typically 1 mm in the injector and 2 mm in the analyzer.

At VERA, the injector magnet is set to a mass resolution of 450 for actinide measurements. Molecular isobars appear to a certain extent at almost every mass in the injector of an AMS facility. Due to the very small mass difference these ions cannot be removed efficiently by additional ion optical separators, and limit the possible background suppression in the injector.

AMS facilities of all sizes (tandems of 0.5–14 MV terminal voltage, e.g. [5–9], and even at a linear accelerator at 221 MeV [10]) have performed measurements on actinides already, and several new groups have presented work at the AMS-11 conference. Chemical suppression of the closest naturally abundant background elements, uranium or thorium, can alleviate the need for the ultimate abundance sensitivity. This is certainly not true for the trace isotope ^{236}U which cannot be chemically separated from the naturally abundant uranium isotopes. Our discussion of the AMS abundance sensitivity will thus focus especially on ^{236}U.

Ionization chambers are usually used as final energy detectors which allow the separation of background ions with lower charge state (see below). Only time-of-flight (TOF) detectors presently provide sufficient resolution to also separate interference from neighboring masses.

3. Abundance sensitivity of ^{236}U at VERA

Charge change along the acceleration tubes in the tandem accelerator is known to produce an energy continuum between the regular ions which change charge only in the terminal stripper (at the level of 10^{-3} at VERA [1]). Additional charge exchange events on residual gas take place throughout the beam line. This is the dominating process leading to leakage of "wrong" ions through the high-resolving filters of an AMS analyzer. Fig. 1 illustrates this process: we assume the two main filters, the magnet and the ESA, have each a suppression factor of 10^4 for "wrong" ions. This results in a total suppression of 10^8 if the ions do not match the settings for neither the magnet (i.e., they have the same momentum/charge) or the ESA (same energy/charge). Thus, ion species with a "wrong" mass which match either the setting of the magnet or the ESA will be enriched by a factor 10^4 relative to the remaining background after the filters, which results in background peaks.

Since traditionally magnets are the highest resolving components in AMS facilities, the particles with the same magnetic rigidity form the most pronounced feature. A detailed investigation for VERA is shown in Ref. [1]. It turns out that charge exchange at a critical position within the ESA is the relevant process (additional to a charge exchange within the accelerator tubes). Since the ions are in a "wrong" charge state after the ESA, they can be suppressed by a relatively low-resolution filter ($M/\text{FWHM}(M) \ll 250$) like our switching magnet. The background of ions with the same electric rigidity is less clearly visible in the spectra presented in Ref. [1],
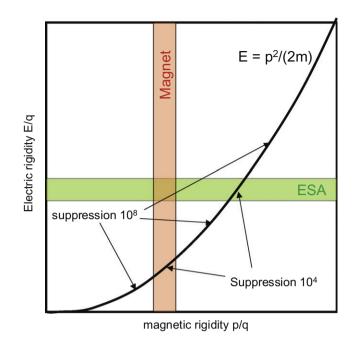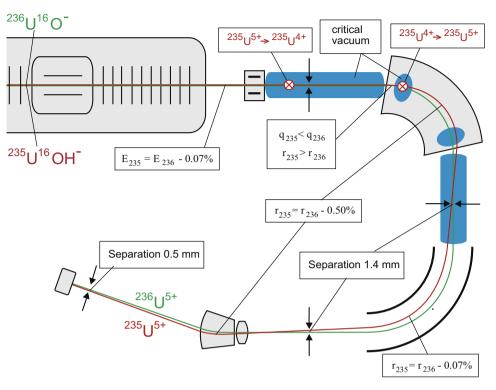

Fig. 1. High-resolution components carve peaks out of the continuous background.

but are the dominating background after the switcher magnet. We have now clarified its origin in detail (Fig. 2). $^{235}U^{5+}$ which is injected as $^{235}U^{16}OH^-$ or $^{235}U^{17}O^-$ does not obtain exactly the same energy as $^{236}U^{5+}$, since it gets a smaller share of the energy when the molecular negative ion breaks up at the terminal. The relative energy difference of -7×10^{-4} is too small to be resolved in the ESA. Also these ions need two charge exchange processes to leak through the spectrometers, one in the beam path between VERA's Wien filter and the analyzing magnet to 4+, and one at a certain position close to the entrance of the analyzing magnet back to 5+, which brings the ions back to the center of the beam line. Alternatively, a first charge change can take place shortly before the exit of the analyzing magnet, and the second in the beam tube between magnet and ESA. Our TOF detector with 2.8 m flight path and 0.7 ns resolution (FWHM) for ^{238}U at 18 MeV provides sufficient resolution to resolve the relevant ions. Fig. 3 shows a TOF-spectrum of a low-^{236}U material (6×10^{-12}) from a measurement which strongly suffered from such background (which might be caused by moisture in the samples or ion source). We have further tested this explanation by closing the vacuum pumps of the respective sections, which resulted in an about 10-fold pressure increase. The count rate in the background peak increased by a factor 10 for the pump before the magnet, and by a factor of 2 for the pump positioned after the magnet. This observation emphasizes the importance of good beamline vacuum in the analyzer.
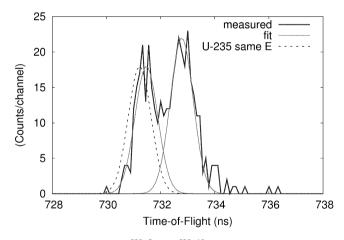
Higher energy is in principle advantageous. The acceleration reduces the beam emittance, which results in higher resolution of the spectrometer. Additionally, most reactions on residual gas, which are responsible for background, have cross sections proportional to E^{-a}, with $a > 0$ [2]. Since, in general, one of such reaction is required for a "wrong" particle to pass a high-resolution filter, the benefit of higher energy should multiply for every additional component.

4. Comparison with other facilities

A recent review paper [11] presents various AMS facilities active in the field of actinides. We can thus restrict ourselves to discussing the sensitivity limits obtained at the various laborato-

Fig. 2. Background of $^{235}U^{5+}$ with almost the same energy. By two charge changes on residual gas in the shaded regions marked as "critical vacuum" the $^{235}U^{5+}$ ions from $^{235}U^{16}OH^-$ can enter the final particle detector.

Fig. 3. TOF-spectrum showing $^{236}U^{5+}$ from $^{236}U^{16}O^-$ (right peak) and background of $^{235}U^{5+}$ from $^{235}U^{16}OH^-$ (left peak). The fit to the data reveals that the $^{235}U^{5+}$ ions have not exactly the same energy as the $^{236}U^{5+}$ ions (the expected peak for the same energy is shown as dashed curve), since they inherit a larger fraction of the energy of the respective molecular negative ion (see text).

Table 1
Sensitivity limits for ^{236}U.

Method	Facility, laboratory, or project		Lowest $^{236}U/U$ reported on real samples	Sensitivity limit quoted
α–spectroscopy		[12]		2×10^{-5}
AMS	ANTARES, Menai	[13]	7×10^{-9}–1.5×10^{-8}	1×10^{-8}
	CAMS, Livermore	[14]	4×10^{-9}	1×10^{-9}
	VERA, Vienna	[15]	5×10^{-12}	5×10^{-13}
	ANU, Canberra	[16]	1×10^{-12}	1×10^{-13}
TIMS	IRMM, Geel	[17]	2×10^{-10}	
ICP-MS	LLNL, Livermore	[14]	1.5×10^{-8}–2×10^{-7}	1.5×10^{-8}
	VIRIS, Vienna	[18]	4×10^{-8}	1×10^{-8}
	REIMEP-18	[19]	3×10^{-8}	
RIMS	Larissa, Mainz	[20]		$>4 \times 10^{-8}$
	IRMM, Geel	[17]	2×10^{-10}	

ries. Table 1 presents abundance sensitivities for $^{236}U/U$ found in the literature. The limits given are probably mainly rough estimates, and thus we do not bother with their precise definition in the following.

Two AMS laboratories are presently targeting at natural isotopic abundances of $^{236}U/U$ below 10^{-10}: the tandem facility at ANU in Canberra, Australia, and VERA. This goal emphasizes the need for the ultimate abundance sensitivity. Additionally, it requires a relatively large amount of uranium (~mg) in the sputter samples, to achieve sensible counting statistics on ^{236}U. Both laboratories have a high-resolution injector magnet, and high-resolution magnetic and electric components in the analyzer, followed by a TOF and en-

ergy detector. Despite VERA uses two additional low-resolution filters for which no equivalent at the ANU setup exists, the abundance sensitivity limit at ANU seems to be even lower, which might be attributed to the higher terminal voltage used.

The other two AMS facilities (CAMS at LLNL in Livermore, California, USA, and ANTARES at ANSTO in Menai, Australia) shown in Table 1 are focusing on significantly higher isotopic ratios in anthropogenically influenced material, which alleviates the demand for abundance sensitivity; however, significantly smaller sample sizes down to a few ng are investigated. The analyzers combine high-resolution magnetic and electric separation followed by an ionization chamber, without a TOF detector. Such small samples require to measure also the abundant isotopes ^{238}U, ^{235}U, and ^{234}U with a particle counting detector. Generally, the requirements come closer to those for the measurement of other anthropogenic, environmental actinides like plutonium and neptunium without stable isotopes.

Table 2
Detection efficiency and limits for Pu.

		Atoms/count	Detection limit (number of atoms in sample)
AMS	(Many laboratories) [22]	10^4	$X \times 10^4$ of ^{241,242,244}Pu 2×10^5 ^{239}Pu, limited by procedural blanks
ICP-MS	[21] [23]	10^3	10^7 quoted for 10% uncertainty
TIMS	[24]	20	$\sim 3 \times 10^4$ quoted "for precise ratio measurement"
RIMS	[25]	2.5×10^4	

5. Comparison with other methods

Every detection method has specific problems. Due to the short half-life of ^{241}Pu (14.4a), β-counting gives the best sensitivity limit for this isotope. α-Decay counting is still the most widespread method to measure environmental $^{239/240}$Pu concentrations, because of the much smaller instrumental effort. A recent review [21] presents the status of plutonium measurements with ICP-MS.

A main limitation for plutonium, where no interference from a stable isotope exists, is the detection efficiency, i.e., the number of counts in the final detector per atom in the environmental sample. Limitations of the various methods are presented in Table 2. Both ICP-MS and TIMS have a higher detection efficiency than AMS. However, for both methods the effective sensitivity for ^{239}Pu is limited by residual ^{238}UH or tailing from ^{238}U in the sample. The sensitivity of <5 fg (1.3×10^7 atoms) reported in Ref. [21] is outperformed by AMS, where 0.1 fg (2.5×10^5 atoms) was achieved [22], limited by the process blank count rate. For ^{244}Pu, where laboratory contamination is unlikely, the sensitivity of AMS is limited by the detection efficiency to $\sim 10^4$ atoms.

TIMS has the highest detection efficiency. Ref. [24] reports that 1 out of 20 atoms can be detected, and that for 3×10^4 ^{240}Pu atoms a precise ^{240}Pu/^{239}Pu measurement is possible. To reduce background, the spectrometer is located in a clean room, and every sample is screened with ICP-MS to prevent contamination of the instrument.

The main advantages of ICP-MS quoted are simplicity of sample preparation, high sample throughput, widespread availability in laboratories worldwide, and relatively straightforward operation. The disadvantages are connected to the injection of the sample as an aqueous aerosol, which fosters hydride formation, and to the high temperature of the plasma, which leads to a large emittance of the beam. In modern setups, the aerosol is dried in the injection tube, and the ion beam may be cooled in a gas filled RF quadrupole.

Decay counting cannot distinguish between ^{239}Pu and ^{240}Pu because of the very similar α-energy. Similarly, α-lines from ^{235}U limit ^{236}U/^{238}U to ratios above 2×10^{-5} [12]. The long-lived isotopes ^{237}Np, ^{242}Pu, ^{244}Pu, and ^{236}U are accessible by decay counting only in rare cases. Mass spectrometric methods including AMS cannot detect ^{238}Pu, because of the interfering natural isobar ^{238}U (this limitation does not apply to RIMS). Since most AMS facilities use the 5+ charge state to measure Pu, ^{240}Pu^{5+} is often limited by pile-up from mass/charge ambiguities. At VERA, we have observed problems with ^{192}Pt^{4+}, ^{144}Nd^{3+}, and ^{48}Ti$^+$ so far, depending on which elements were introduced into the ion source in previous beam times. For many samples, only switching to the 7+ charge state, which however reduces the stripping yield to about 1%, makes a ^{240}Pu/^{239}Pu measurement possible. We hope that our new injector [26], which will be used only for the main AMS isotopes, will avoid this problem.

6. Applications of heavy, long-lived radionuclides at VERA

Refs. [21,11] review environmental applications of actinides, for mass spectrometry in general and especially for AMS, respectively.

At VERA, we have established a procedure to determine the complete 'isotopic vector' with the respective best-suited method for each isotope (α-spectrometry for ^{238}Pu/$^{239+240}$Pu; AMS for ^{240}Pu/^{239}Pu, ^{242}Pu/^{239}Pu, and ^{244}Pu/^{239}Pu; and liquid scintillation counting (LSC) for ^{241}Pu/Pu), and verified the procedure with reference materials [27]. Though this approach yields the best possible results, all isotopes but ^{238}Pu are accessible with AMS alone, at reduced sensitivity for ^{241}Pu compared to LSC, but at much lower preparation effort. Fig. 4 shows as an example a plot of soil samples collected from Naßfeld, Salzburg country, Austria, the region in Western Europe most strongly affected by fallout from the Chernobyl accident (typical Pu isotope ratios for various sources are given in Ref. [23]). Since this is a region of high precipitation, also the concentration of global fallout nuclides is relatively high. The AMS results clearly suggest an origin from global fallout. The
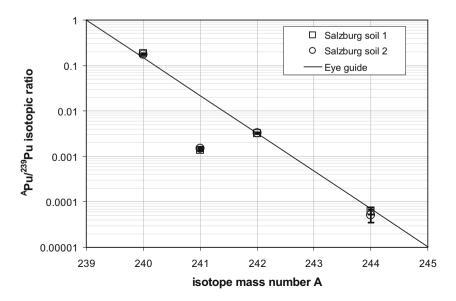

Fig. 4. Pu in soil samples from Naßfeld, Salzburg, Austria. The data is in agreement with global fallout (see text).

ratio of ^{240}Pu/^{239}Pu is 0.18. ^{244}Pu would not be present in used reactor fuel, but is observed at the level expected from the exponential mass trend of ^{239}Pu, ^{240}Pu, and ^{242}Pu. The deviation of ^{241}Pu from the trend corresponds to a decay age of roughly 50 years.

In the Salzburg soil samples, also ^{236}U was found at isotopic ratios up to 1.5×10^{-6} [15]. ^{236}U was also present in surface waters from nearby rivulets [28]. Due to the high fallout of ^{137}Cs and ^{90}Sr from Chernobyl in this region, an origin from that source cannot be excluded. To complement these measurements by samples less affected by Chernobyl, and to assess whether ^{236}U may be used as a tracer for reactor fuel releases, we have carried out exploratory measurements in river sediments near the Garigliano Nuclear Power Plant, South Italy, which is presently in the decommissioning phase [29]. In the cooling water drain channel of the plant, and in river sediment 1 m downstream of the mouth of the drain channel, the ^{236}U/^{238}U isotopic ratio is slightly, but significantly higher compared to samples from several km upstream of the power plant. Several km downstream the same values were found as upstream. This finding demonstrates the potential of ^{236}U as a fingerprint of releases from nuclear facilities.

However, to our surprise, the sensitivity of AMS allowed to unambiguously detect ^{236}U also upstream of the power plant, at isotopic ratios as high as ^{236}U/U = 2.5×10^{-8}. ^{236}U can be produced in thermonuclear explosions via ^{238}U(n, 3n), and has been detected in global fallout by Ketterer et al. [30]. In a joint effort with a group from the Research Institute for Radiation Biology and Medicine, Hiroshima University, Japan, and the Low Level Radioactivity Laboratory, K-INET, Kanazawa University, Nomi, Ishikawa, Japan [31], we were able to obtain a relatively precise first value of ^{236}U/^{239}Pu 0.23 ± 0.02 from a site in Japan representative for global fallout.

The high selectivity achieved at VERA also allows us to look for "exotic" heavy nuclei, where only very few, if any, real events can be expected. We have put significant effort in detecting ^{182}Hf [32] and ^{244}Pu [33] from interstellar matter, which could have entered deep sea sediments similarly to ^{60}Fe [34]; and we are pursuing a search for long-lived, super-heavy elements in nature [35].

7. Summary and conclusions

The sensitivity of AMS for actinides is not limited by the accelerator terminal voltage. Also for the most demanding case, ^{236}U, the 3-MV tandem accelerator VERA can achieve an abundance sensitivity which is sufficient even for the smallest isotopic ratios expected in environmental samples. However, careful investigations of background processes are required to eliminate all possible background. No other mass spectrometric method can presently achieve the abundance sensitivity of AMS for ^{236}U well below 10^{-11}.

The environmental and geological applications of long-lived anthropogenic isotopes are generally similar to those of short-lived nuclides measured by decay spectrometry, i.e. as environmental tracers or time markers. However, they provide complementary information because of different chemical behavior and source terms. Especially ^{236}U, for which the advantages of AMS over other mass spectrometric methods are most pronounced, is omnipresent in the environment. Further investigations on the distribution and environmental pathways of global-fallout ^{236}U are likely to open up applications in oceanography, hydrology and sedimentology. The much less abundant natural nuclides may allow to extend the time range to geological scales.

The Pu isotopes and other actinides, which are (almost) completely of anthropogenic origin, can also be measured by other mass spectrometric methods, and partly with higher instrumental detection efficiency (several %). However, in practical measurements, AMS has achieved higher sensitivity, due to the higher robustness against uranium-related background from the sample matrix.

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