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# First application of calorimetric low-temperature detectors in accelerator mass spectrometry

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## Abstract

For the first time, calorimetric low-temperature detectors were applied in accelerator mass spectrometry, a well-known method for determination of very small isotope ratios with high sensitivity. The aim of the experiment was to determine with high accuracy the isotope ratio of $^{236}U/^{238}U$ for several samples of natural uranium, $^{236}U$ being known as a sensitive monitor for neutron flux. Measurements were performed at the VERA tandem accelerator at Vienna, Austria. The detectors consist of sapphire absorbers and superconducting transition edge thermometers operated at $T \approx 1.5\,K$. The relative energy resolution obtained for 17.39 MeV $^{238}U$ is $\Delta E/E = 4–9 \times 10^{-3}$, depending on the experimental conditions. This performance enabled to substantially reduce background from neighbouring isotopes and to increase the detection efficiency. Due to the high sensitivity achieved, a value of $^{236}U/^{238}U = 6.5 \times 10^{-12}$ could be obtained, representing the smallest $^{236}U/^{238}U$ ratio measured until now.

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*Keywords:* Calorimetric low-temperature detectors; Accelerator mass spectrometry; Heavy ion physics

## 1. Introduction

Accelerator mass spectrometry (AMS) is a well-known method for the determination of very small isotope ratios with high sensitivity [1]. In comparison to conventional mass spectrometry, the use of accelerated ion beams provides substantial advantage in the quality of isotope separation and background suppression, and therefore, allows the determination of isotope ratios down to a level of $10^{-10}$–$10^{-16}$ with high precision. $^{236}U$ represents one of the heaviest nuclides of interest for AMS. Being produced in nature by capture of thermal neutrons in the reaction $^{235}U(n,\gamma)^{236}U$ and having a half-life of 23.4 million years, the relative abundance of $^{236}U$ provides an excellent neutron flux monitor integrated over geological time scales [2,3]. Thus, besides other applications, $^{236}U$ can be used to prove the existence of an enhanced neutron flux due to natural "reactor-like" conditions [3]. In

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natural uranium, the isotope ratio is expected to be of the order of $^{236}U/^{238}U = 10^{-10}$–$10^{-14}$, dependent on its history and surroundings.

Under the present conditions at the VERA AMS facility, background in AMS measurements for very heavy ions is mainly due to neighbouring isotopes which have, due to various charge exchange processes, the same magnetic rigidity $ME/q^2$ ($M$ being the mass and $q$ the charge state of the ion) and therefore pass through the high-energy magnetic analyzer and, after additional charge exchange, also through the electrostatic analyzer (see Ref. [3] for detailed discussion). Background situation expected for the case of 17.54 MeV $^{236}U^{5+}$ is displayed in Fig. 1. Since standard energy detectors (e.g. ionization chambers, etc.) do not provide sufficient resolution to resolve these background peaks, in the previous measurements [2,3] time-of-flight (TOF) spectrometers combined with the ionization chambers were used. The level of $^{236}U/^{238}U$ ratio sensitivity reached was limited to $6 \times 10^{-11}$, mainly due to the limited detection efficiency of only 20%. Calorimetric low-temperature detectors (CLTDs) have already been demonstrated [4] to provide an excellent relative energy resolution of $5$–$6 \times 10^{-3}$, a linear energy response and high-detection efficiency for relatively low ion energies $E = 0.1$–$1$ MeV/amu, typical for AMS. Such detectors are, therefore, well suited to replace the standard TOF/energy detection scheme and to resolve the isotope of interest from neighbouring isotopes by their high-energy resolution power alone (see Fig. 1). This leads to a substantially improved detection efficiency, especially important for the detection of very rare isotopes.

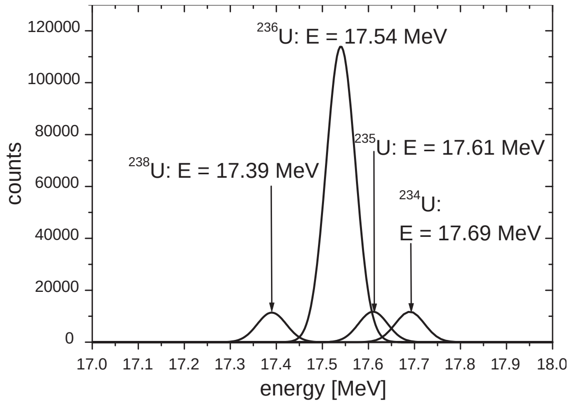

Fig. 1. Simulation of background separation (Gaussian line shapes). The ratio of $^{236}U$: $^{234,235,238}U$ is assumed to be 10:1, the energy resolution to be $\Delta E/E = 4.6 \times 10^{-3}$.

The aim of the present investigations was therefore to apply CLTDs in an AMS experiment in the determination of the isotope ratio $^{236}U/^{238}U$ for various samples of natural uranium in order to establish a precise material standard for the isotope ratio and to improve the level of sensitivity to $\leqslant 10^{-11}$.

## 2. Detector design and experimental setup

The detectors used for the present investigations consist of a thin film superconducting aluminium strip thermometer operated at $T \sim 1.5$ K, which is evaporated onto a sapphire substrate serving as absorber with a thickness of 330 μm and an area of approximately $3 \times 2.5\,mm^2$. A detailed discussion of layout and preparation of the detectors can be found in Ref. [5]. The detectors are mounted in a pumped $^4He$ window cryostat operated at temperatures between 1.2 and 1.6 K. To avoid energy straggling and efficiency losses of the low-energy heavy ions, entrance windows were replaced by a system of four slits with dimensions of $3.5 \times 15\,mm^2$. The setup was mounted at the end of the beamline of the VERA 3MV tandem accelerator. This dedicated AMS machine provides $U^{5+}$ beams with $E \approx 17.5$ MeV. Typical count rates ranged from $10\,s^{-1}$ down to $10^{-2}\,s^{-1}$ for the sample with lowest $^{236}U$ abundance. For the determination of the isotope ratio the radioisotope $^{236}U$ was detected in the CLTD, while for the "stable" $^{238}U$ the beam current was measured in a Faraday cup. For detailed description of AMS measurement procedure see Refs. [2,3]. In contrast to the setup described in these references, no TOF detector was included in the present measurements.

## 3. Results

The detector performance was tested using a beam of $^{238}U$, reduced in intensity, with an incident energy of $E = 17.39\,\mathrm{MeV}$. The corresponding spectrum is displayed in Fig. 2. Compared to previous measurements [4], the energy resolution was improved to $\Delta E = 80\,\mathrm{keV}$, corresponding to a relative resolution of $\Delta E/E = 4.6 \times 10^{-3}$. With this performance, the background contribution of neighbouring isotopes leaking through the magnetic and electrostatic filters of VERA (see Fig. 1) may be well separated.

For the first AMS measurements presented below, the detector performance under running conditions was unfortunately worse, but already with a resolution of $\Delta E/E = 9.1 \times 10^{-3}$, essential parts of the background could be separated. Several samples of natural uranium were investigated (see Table 1). The samples named "Joachimsthal" were prepared from uranium ore material which was stored before 1918 and thus, does not contain any contamination due to nuclear bomb explosions etc. The results (still preliminary!) are displayed in Table 1 and in Fig. 3. Only minor contributions of $^{238}$U and $^{234}$U were observed whereas separation of $^{235}$U was not possible. The difference between samples Joachimsthal 1 and 2 may result from the fact that they were not collected at the same location inside the mine. Within errors, the result of Joachimsthal 1 is in agreement with previous measurements [2,3]. The systematic errors quoted are mainly due to the uncertainities in the transmission of the $^{236}$U ions from the VERA facility to the detector. As compared to the earlier measurements [2,3], a reduction of the systematic errors due to increase in transmission from $(0.20 \pm 0.06)$ to $(0.65 \pm 0.10)$ was achieved, mainly due to the improved detection efficiency.

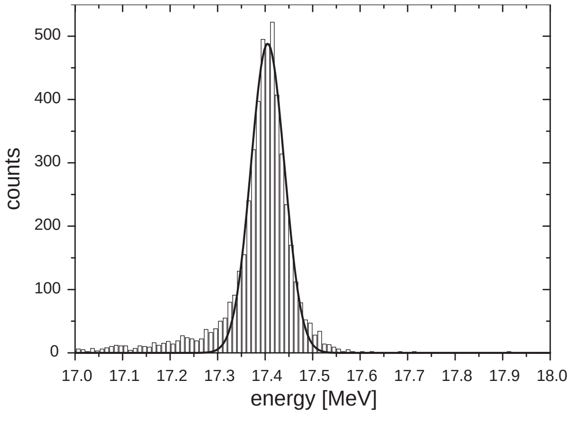

Fig. 2. Energy spectrum for $^{238}$U at $E = 17.39\,\mathrm{MeV}$.

Table 1
Results of isotope ratio measurements with AMS

| Sample | $^{236}$U/$^{238}$U ($10^{-11}$) |
|---|---|
| Joachimsthal 1 | $4.46 \pm 0.09_{\mathrm{stat}} \pm 0.70_{\mathrm{syst}}$ |
| Joachimsthal 2 | $2.72 \pm 0.09_{\mathrm{stat}} \pm 0.43_{\mathrm{syst}}$ |
| Joachimsthal 1 [2,3] | $6.1 \pm 0.4_{\mathrm{stat}} \pm 1.8_{\mathrm{syst}}$ |
| Bad Gastein | $0.65 \pm 0.19_{\mathrm{stat}} \pm 0.10_{\mathrm{syst}}$ |

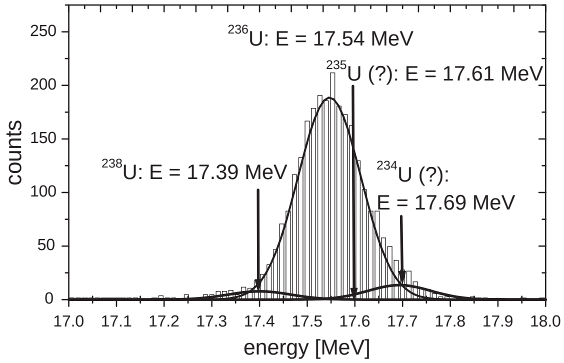

Fig. 3. Spectrum of AMS measurement for sample Joachimsthal 1.

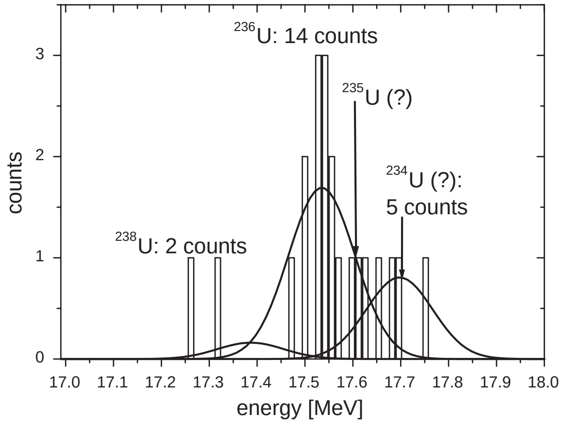

Fig. 4. Spectrum of AMS measurement for sample from Bad Gastein spring water.

With the increase in sensitivity obtained, it was possible for the first time to investigate one sample of uranium extracted from 5 l of spring water from Bad Gastein, Austria, for which an isotope ratio of $^{236}\mathrm{U}/^{238}\mathrm{U} \leqslant 10^{-12}$ was expected. Due to the low uranium concentration in the spring water, the amount of sample material was limited and only one measurement of 30 min could be performed. Fig. 4 shows the energy spectrum obtained. The result for the isotope ratio is $6.5 \pm 2.2 \times 10^{-12}$ (see Table 1). This represents the smallest isotope ratio ever measured for $^{236}\mathrm{U}/^{238}\mathrm{U}$. Future measurements with optimised energy resolution and increased detection efficiency will allow to measure even lower $^{236}\mathrm{U}/^{238}\mathrm{U}$ ratios.

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