

# $^{236}\text{U}$ AMS measurement at CIRCE

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**Abstract** In order to measure the isotopic ratio of actinides, the upgrade of the accelerator mass spectrometry system at the Center for Isotopic Research on Cultural and Environmental heritage at the Second University of Naples, Italy, was performed. The beam emittance of  $^{238}\text{U}$  and the isotopic abundance sensitivity of  $^{236}\text{U}$  were measured on the present beam line. Utilizing a 16-strip silicon detector, the sensitivity of  $^{236}\text{U}/^{238}\text{U} \approx 1 \times 10^{-11}$  was obtained.

**Key words** uranium, AMS, beam emittance, sensitivity

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## 1 Introduction

$^{236}\text{U}$  plays an important role in nuclear safeguards and in determining the extent of environmental contamination in modern theaters of war due to the use of depleted uranium weaponry, as well as the application to track nuclear releases. The key feature of the accelerator mass spectrometry (AMS) method is the significant suppression of scattering tails due to the high ion energies and the complete elimination of molecular interference. AMS is proving to be an exquisitely sensitive tool for studying long-lived actinides. More and more AMS laboratories are adding an actinide capability to their repertoire [1–6].

This paper describes the upgrade of the AMS system at the Center for Isotopic Research on Cultural and Environmental heritage (CIRCE) at Caserta, Italy. The measurement procedure and a sensitivity of  $^{236}\text{U}$  detection by using a 16-strip silicon detector are described.

## 2 $^{236}\text{U}$ AMS measurement at CIRCE

### 2.1 Upgrade of CIRCE AMS system

The CIRCE AMS facility is based on a 3 MV pelletron tandem accelerator, built by National Elec-

trostatics Corporation. The initial AMS system was presented at AMS-10 [7]. The schematic layout of the new setup is shown in Fig. 1. For detailed information of the main elements, see Refs. [7, 8]. This section focuses on the upgrade of the CIRCE AMS system, aiming to achieve isotopic abundance sensitivity for  $^{236}\text{U}$  better than  $10^{-11}$  and better background suppression. The first step is to add a switching magnet with several dedicated beam lines for other applications and another purification stage.

The decision on how to arrange the switching magnet ( $B=1.3$  T,  $\rho=1.760$  m and  $ME/q^2=252.5$  MeV amu/ $e^2$ ) was based on ion optical calculation. The COSY infinity code was implemented to simulate the individual ion trajectories [9].  $^{236}\text{U}$  and  $^{238}\text{U}$  beams have been analyzed from the image point of the high energy analysing magnet with a relative energy difference of  $\Delta E/E=0.001$ , which corresponds to the energy resolution of the electrostatic analyzer (ESA) [10]. According to the calculated result, the switching magnet was located 0.5 m behind the ESA [10]. In Fig. 1, the upgraded setup at CIRCE is shown, where the dipole switching magnet has been inserted.

### 2.2 Acceleration and transmission

Samples of a few milligrams  $\text{U}_3\text{O}_8$  powder mixed

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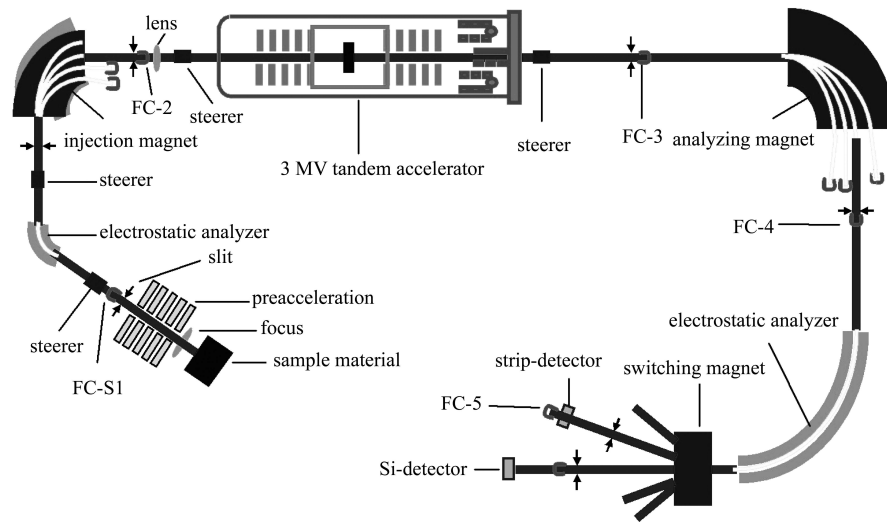


Fig. 1. Schematic layout of the CIRCE AMS system with the switching magnet.

with iron powder of equal mass, as a binder, are pressed into a 1 mm diameter aluminum holder. Negative molecular ions  $^{238}\text{U}^{16}\text{O}^-$  obtained from operating a high intensity Cs-sputter source are extracted from the sample. According to the maximum rigidity of the injection magnet, the negative ion beam is pre-accelerated to 50 keV, and selected by an electrostatic analyzer and a  $90^\circ$  injection magnet before the accelerator. The injection magnet object and image slits in vertical and horizontal directions were all close to  $\pm 1$  mm, in order to increase the mass resolution ( $M/\Delta M \approx 500$ ). The current of  $^{238}\text{U}^{16}\text{O}^-$  in a Faraday Cup FC-2 is in the range of 50–300 nA. The terminal voltage was carefully chosen and compared with the other labs [2–5]. The advantages of a high terminal voltage are lower background and higher tandem transmission. On the other hand, higher energy requires a higher field in the analyzing and switching magnets. In the CIRCE AMS system,  $^{236}\text{U}^{5+}$  is transmitted at a terminal voltage of 2.900 MV.

In order to optimize the beam transmission through the various apertures, we tune the ion-optical components with  $^{238}\text{U}^{16}\text{O}^-$  at a terminal voltage of 2.875 MV. The focusing and steering elements are optimized by maximizing the current in the Faraday cup (FC-4), after the analyzing magnet. This is automatically done by our own program. Here, the slits are set to  $\pm 2$  mm. According to the currents in the Faraday cup FC-2 and FC-4, the maximum stripping yield of 5+ charge state is about 3.1%, at a pressure of about 0.93 Pa. The relationship between the stripper gas pressure and the stripping yield of 5+ charge state is shown in Fig. 2.

The analyzing magnet, the electrostatic analyzer,

the switching magnet and all of the focusing and steering elements are optimized by maximizing the current in the Faraday cup FC-5 (see Fig. 2). The transmission efficiency between the Faraday cup FC-4 and the Faraday cup FC-5 at  $20^\circ$  is about 80%.

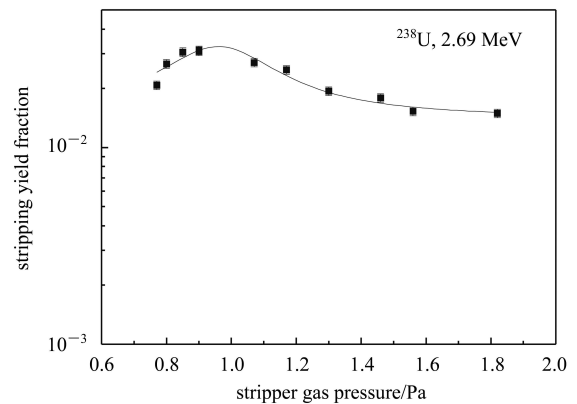


Fig. 2. Measured stripping yield fraction versus stripper gas pressure for  $^{238}\text{U}$  ions at 2.69 MeV energy for charge state 5+ and for argon as the stripper gas. The maximum of the stripping yield corresponds to 0.93 Pa.

### 2.3 Beam emittance measurement

In order to locate the optima the position of the TOF detector and decide the necessary active area of the stop detector, a simple emittance measurement unit (EMU) is used to implement the beam emittance of  $^{238}\text{U}$ . The EMU consists of a front slit and a rear wire. The front slit is made of stainless steel, and the size of the aperture is 38 mm $\times$ 0.5 mm. The diameter of the copper wire is 0.1 mm. The front slit is 152 cm behind the exit of the switching magnet. The distance between the slit and the wire is 117 cm. The

results indicate that the boundary emittance of  $^{238}\text{U}$  in the  $X$  and  $Y$  planes is  $(9.5\pm 1.0)$  and  $(2.7\pm 0.3)$  mm-mrad, respectively. The beam profile has an approximately Gaussian shape. The phase diagrams of the boundary emittance of  $^{238}\text{U}$  ions are shown in Fig. 3. Due to the larger angular scattering

of the heavier ions in the stripping gas, the measured beam boundary emittance for heavy ion beams (e.g., for  $^{238}\text{U}^{5+}$  with the energy of 17.1 MeV the emittance is  $(9.5\pm 1.0)$  mm-mrad), is larger than the light ions (e.g., for  $^{13}\text{C}^{3+}$  with the energy of 10.2 MeV the emittance is  $(6.8\pm 0.7)$  mm-mrad).

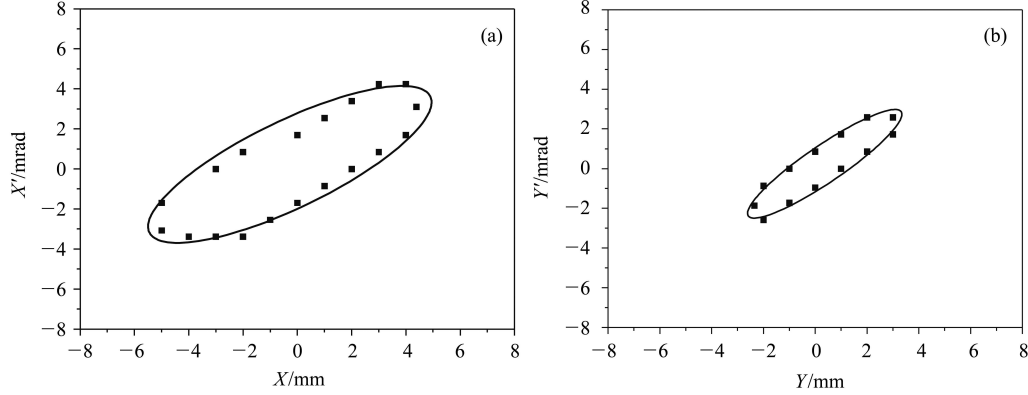


Fig. 3. Phase diagram of the boundary emittance of  $^{238}\text{U}$  ions (a)  $X$ -plane; (b)  $Y$ -plane.

According to the emittance of  $^{238}\text{U}$ , we can deduce that the beam waist of  $^{238}\text{U}$  is located approximately 80 cm behind the switching magnet, which is in agreement with the calculation results. The diameter of the beam profile of the detector emplacement, which is 2 m behind the waist, is about 20 mm. So a large active area detector is necessary for high detection efficiency.

## 2.4 AMS measurement

The detection system consists of a Faraday cup (FC-4, not FC-5, since the FC-5 can not be remotely controlled) that measures the current of the abundant isotope (e.g.,  $^{238}\text{U}$ ) and a 16-strip silicon detector that identifies and counts the rare isotope (e.g.,  $^{236}\text{U}$ ). The 16-strip silicon detector with an active area of 58 mm $\times$ 58 mm, is positioned 1.8 m downstream from the 4 mm diameter collimator positions (80 cm from the switching magnet). The distance between the centres of two adjacent strips is 3.625 mm.

Using fast switching of the potential of the insulated vacuum box, the 90° injection magnet provides fast-switched bouncing of the injected ion mass between  $^{238}\text{U}^{16}\text{O}^-$  and  $^{236}\text{U}^{16}\text{O}^-$ . Typical injection times for  $^{236}\text{U}$  are 120 s, and for 10 s for  $^{238}\text{U}$ . Since the rigidity between  $^{238}\text{U}^{5+}$  with the energy of 17.07 MeV (i.e., terminal voltage 2.875 MV) and  $^{236}\text{U}^{5+}$  with the energy of 17.21 MeV (terminal voltage 2.9 MV) is the same, we don't need to change the field of the analyzing magnet. The potential of the insulated vacuum box of the injection magnet, the

terminal voltage and the voltage of the ESA are controlled by an automated program. Faraday cup FC-4 is inserted to measure the current when the  $^{238}\text{U}$  ions are injected. The total measuring time for  $^{236}\text{U}$  is 600 s.

Two kinds of samples were measured a few times in the present system from February 2009 to July 2009. A 'standard' with a nominal ratio  $^{236}\text{U}/^{238}\text{U}\approx 1\times 10^{-8}$ , obtained by adding a spike of  $^{236}\text{U}$  to the sample KkU VERA-in-house-U standard [11,12], and the KkU itself with  $^{236}\text{U}/^{238}\text{U}=(6.98\pm 0.32)\times 10^{-11}$ , were measured. The ion distribution in the strip detector of 'standard' and KkU are shown in Fig. 4. The value of the  $Y$  axis is the ratio between the counts per strip and the total ones of 16 strips.  $^{236}\text{U}$  counts are distributed from strip-8 to strip-12, according to the 'standard' sample. Almost 100% of the  $^{236}\text{U}$  counts are located in the region of interest, from strip-9 to strip-11. The unnormalized values of isotopic abundance of 'standard' and KkU sample are  $^{236}\text{U}/^{238}\text{U}=(9.51\pm 0.36)\times 10^{-9}$  and  $^{236}\text{U}/^{238}\text{U}=(8.23\pm 0.60)\times 10^{-11}$ , respectively. The results were obtained several times, indicating the good reproducibility of the system (shown in Fig. 5).

Moreover, in order to get the detection limit of our system, we measured different samples obtained starting from the 'standard' sample with ratio  $^{236}\text{U}/^{238}\text{U}\approx 1\times 10^{-8}$  with serial dilutions. A weighted purified uranyl nitrate ( $\text{UO}_2(\text{NO}_3)_2\cdot 6\text{H}_2\text{O}$ ) sample with the atomic ratio of  $^{236}\text{U}/^{238}\text{U}\approx 1\times 10^{-8}$  was dissolved in concentrated  $\text{HNO}_3$  for 4 h in a beaker. The

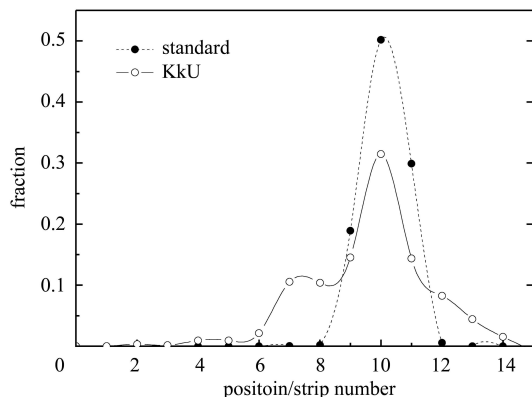


Fig. 4. Normalized fractions (the ratio of counts per strip to the total counts in 16 strips) versus horizontal position of the 16-strip silicon detector.

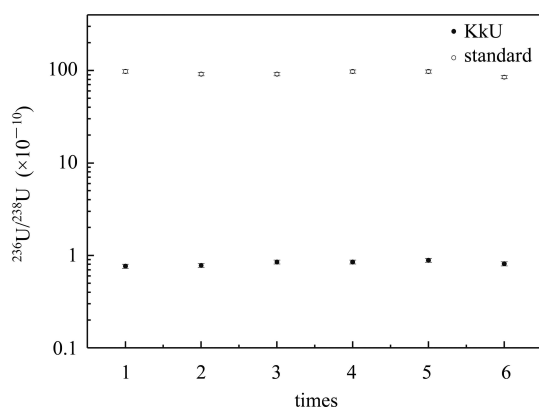


Fig. 5. Results of 'standard' and KkU sample for different time measurements.

distilled water was added until the concentration of  $^{238}\text{U}$  was  $40\ \mu\text{g}/\text{ml}$ . 5 ml of uranium solution was diluted to 50 ml of solution. This initial dilution was 1:10. Then 5 ml of this dilution was added to 45 ml of  $\text{H}_2\text{O}$ , further diluting the sample. This same process was continued. So the serial uranium solution with concentrations of 40, 4, 0.4 and  $0.04\ \mu\text{g}/\text{ml}$ , was prepared. An amount of  $\text{FeCl}_3$  solution (equivalent to 2 mg Fe) was added to 50 ml in serial uranium solutions in a centrifuge tube. The concentrated

$\text{NH}_4\text{OH}$  was added until uranium and iron hydroxide co-precipitated. The precipitation was centrifuged and separated. The separated precipitation by combustion at  $700\ ^\circ\text{C}$ , and the chemical form of the AMS sample  $\text{U}_3\text{O}_8+\text{Fe}_2\text{O}_3$  was obtained. Three Uranium samples with 2, 20, and 200 micrograms, were measured and compared with the sample with 2 mg. The results show that the samples with the  $^{238}\text{U}$  amount up to  $20\ \mu\text{g}$  yield an isotopic value in good agreement with the sample with 2 mg. Unfortunately, we can't get a good result from the sample with the amount of  $2\ \mu\text{g}$  due to the 'zero' current in FC-4.

### 3 Results and discussions

A high detection efficiency of actinide measurement was set up in CIRCE, Italy. The transmission efficiency between the low energy injection magnet and the detector is about 2.5%. The detection efficiency is close to 100% due to the 16-strip silicon detector with a large active area. The performance of the actinide detection system is as good as expected. The unnormalized value,  $^{236}\text{U}/^{238}\text{U} = (8.23 \pm 0.60) \times 10^{-11}$ , was obtained by measuring the 'KkU' sample. According to the results of 'KkU' (see Fig. 4), we can deduce the sensitivity of our detection system, which is  $\sim 1 \times 10^{-11}$ .

An amount of Uranium in the sample as low as  $20\ \mu\text{g}$  is measured, corresponding to  $200\ \text{fg}\ ^{236}\text{U}$ , and the results are in agreement with the sample of a few milligrams. The result indicates that the present detection limit in CIRCE is better than  $5 \times 10^8$  atoms.

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