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The AMS measurement of ^{236}U at CIRCE

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Abstract Accelerator mass spectrometry (AMS) is an ultrasensitive technique for measuring long-lived actinides, e.g., ^{236}U, ^{237}Np and Pu isotopes. In order to improve the detection limit for actinides abundance, and to increase the detection efficiency in actinides AMS measurement, a 16-strip silicon detector was used to identify actinides at the Center for Isotopic Research on Cultural and Environmental heritage in Caserta, Italy. The sensitivity of ^{236}U/^{238}U was 1×10^{-11} by special resolution and 5.0×10^{-12} by time resolution. The pulse height defect of ^{236}U in an ion-implanted silicon detector in the low-energy range with 17.26 MeV is presented.

Keywords Uranium · Accelerator mass spectrometry · Sensitivity · Pulse height defect

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1 Introduction

The long-lived anthropogenic radio nuclides of ^{236}U ($T_{1/2} = 2.348 \times 10^7$ a), ^{237}Np ($T_{1/2} = 2.1 \times 10^6$ a), ^{239}Pu ($T_{1/2} = 2.4 \times 10^4$ a) and ^{240}Pu ($T_{1/2} = 6.6 \times 10^3$ a) have been released into the environment by nuclear weapons testing, nuclear accidents, fuel reprocessing and decommissioning of nuclear power plants. These radio nuclides play important roles in environmental monitoring, nuclear safeguards and nuclear forensic studies. The main sources of ^{236}U are from the thermal neutron reaction of ^{235}U and the alpha decay of ^{240}Pu.

The nature abundances of ^{238}U, ^{235}U and ^{234}U are approximately 99.272, 0.72 and 0.054%, respectively. Due to the difference of ^{236}U concentration between natural uranium and the spent uranium by 7 to 11 orders of magnitude, ^{236}U/^{238}U isotopic ratio is a powerful indicator of uranium resource. Usually, the activity of ^{236}U in environmental samples is much lower than the exemption limit, and analyzing ^{236}U does not require special radiochemical laboratories. However, determination of ^{236}U is not an easy task, because low quantities of ^{236}U are usually mixed with the high abundance of ^{238}U and ^{235}U, considered as potential sources of strong interferences. Since the energy difference between the alpha particles emitted by ^{236}U and ^{235}U is less than 0.10 MeV, detection of ^{236}U alpha particles emission is interfered with ^{235}U alpha particles and other isotope emissions, caused by energy tailing. The ^{236}U/^{238}U isotopic ratio is limited in 2.6×10^{-5} in alpha spectrometry. With the improvements in sample preparation methods and detection techniques, thermal ionization mass spectrometry (TIMS), inductively coupled plasma mass spectrometry (ICP–MS) and accelerator mass spectrometry (AMS) are becoming the most popular and

powerful tools for the determination of uranium isotopic measurements.

ICP–MS and TIMS have been employed for measuring ^{236}U, in sensitivities of 10^{-8} and 10^{-10}, respectively, though [1]. The important factors affecting the detection sensitivity of ^{236}Uin ICP–MS and TIMS are molecular ions (e.g., $^{235}UH^+$, $^{234}H_2^+$) and high-abundance isotope peak tail of ^{238}U and ^{235}U on mass 236. To improve the ^{236}U/^{238}U detection sensitivity, multiple-collector ICP–MS (MC–ICP–MS) has been used, with better precision. The outstanding feature of AMS method is the significant suppression of scattering tails due to its high ion energies and the complete elimination of molecular interference. AMS is an effective and robust technique for studying long-lived actinides, e.g., ^{236}U, ^{237}Np and ^{239}Pu. Of the all available detection techniques, AMS is presently the most sensitive technique for ^{236}U measurement, ^{236}U/^{238}U ratios down to the level of 10^{-13} were reported by Wilcken et al. [2]. More and more AMS laboratories performed the actinides research [3–10].

In this paper we present the performance of ^{236}U AMS measurement, developed at the Center for Isotopic Research on Cultural and Environmental heritage (CIRCE) in Caserta, Italy. A 16-strip ion-implanted silicon detector is used to identify actinides by spatial and time resolution, respectively. The pulse height defect of ^{236}U ions in the detector with the energy of 17.26 MeV is analyzed.

2 AMS facility and detection system

CIRCE is a dedicated AMS facility based on a 3-MV tandem accelerator, built by National Electrostatics Corporation. The schematic layout of the present setup is shown in Fig. 1. The AMS facility is an ideal system for actinides measurement, which consists of high-resolution injection magnet, analyzing magnet and electrostatic analyzer. The cesium sputter ion source is a 40-sample MC-SNICS normally biased at −50 kV for ^{236}U measurement. In our case, the sample preparation provides uranium in the form of $U_xO_y + Fe_2O_3$, and an output from a Φ1 mm aluminum cathodes for $^{238}U^{16}O^-$ ions in Faraday Cup 2 (FC-2) is in the range of 50–300 nA with a total injection energy of 50 keV. The injection magnet object and image slits in vertical and horizontal directions are all close to ±1 mm, mass resolution injection system is $M/\Delta M = 500$. High terminal voltage is helpful to obtain lower background and higher ion transmission, but requires a higher field in the analyzing and switching magnet. According to parameters of the analyzer components, $^{236}U^{5+}$ is transmitted at a terminal voltage of 2.900 MV. The double focusing 90° high-energy bending magnet is of $\rho = 1.27$ m, $ME/Z^2 = 176$ and $M/\Delta M = 725$, so that, e.g., $^{236}U^{5+}$ at terminal voltage of 3 MV can be analyzed with a beam spot size of 3.5 mm. The two 45° electrostatic spherical analyzers ($\rho = 2.54$ m and gap = 3 cm) are operated at ±40 kV, and energy resolution is $E/\Delta E = 700$.

The actinides beam line, mounted after the switching magnet, consists of a microchannel plate detector (MCP) and a position-sensitive 16-strip silicon detector. The detector, with an active area of 58 mm × 58 mm, is a partially depleted passivated implanted planar silicon (PIPS) detector (Canberra PF-16CT-58*58-300EB). The 16-strip detector is used to measure the ion energy and the time signal. The ^{236}U, ^{235}U and ^{238}U ions hit different positions of the strip detector due to the different E/q ratios, so the position information can be served as uranium isotopes identification. The time of flight (TOF) detection system with a flight path of 1.5 m is installed to discriminate ^{236}U ions from ^{235}U, ^{238}U and other interferences. Details about the TOF detection system can be found in Ref. [11].

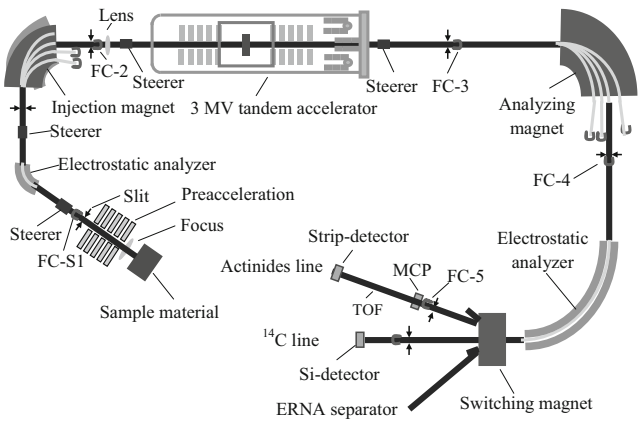

Fig. 1 Schematic layout of the CIRCE AMS system

3 ^{236}U AMS measurement

The principle of AMS and general tuning procedure were described clearly in earlier work [12, 13]. The sensitivity of AMS is mainly limited by the interferences from isobars, isotopes and other backgrounds. The source of interferences was discussed in details, and a program was designed for experimental spectra in AMS measurement [14].

In order to optimize the beam transmission through the various apertures, we tuned the ion-optical components with $^{238}U^{16}O^-$ at terminal voltage of 2.875 MV. The focusing and steering elements were optimized by maximizing the current in FC-4, after the analyzing magnet. According to the currents in FC-2 and FC-4, the stripping yield of +5 charge state is about 3.3%, at 0.93 Pa of the gas stripper pressure. The stripper yield as a function of the

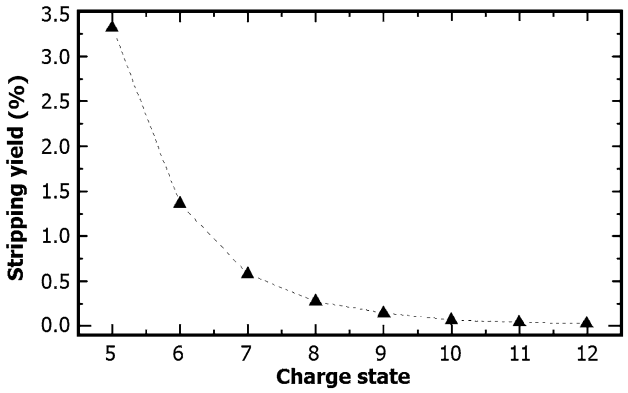

Fig. 2 Charge state distributions of ^{238}U ions passing through the gas stripper at terminal voltage of 2.875 MV

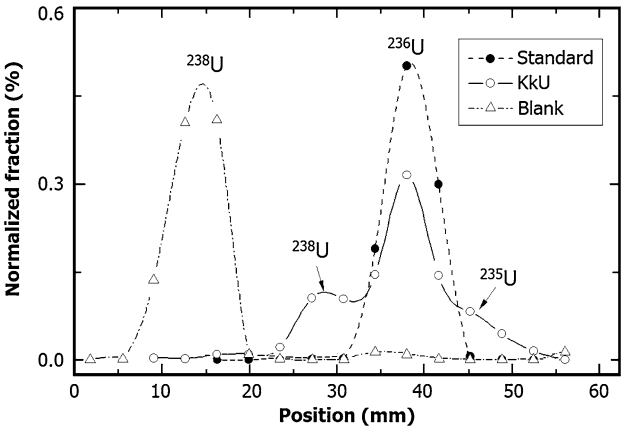

Fig. 3 Normalized fractions (counts per strip over total counts in the 16-strip detector) of uranium isotopes

charge states is shown in Fig. 2. The +5 charge state is the minimum permitted by the bending power of high-energy analyzing magnet. The electrostatic analyzer, the switching magnet and all of the focusing and steering elements were optimized by maximizing the current in FC-5, before the MCP (Fig. 1). The transmission efficiency between FC-4 and FC-5 at the actinides beam line is about 80%. Using a fast beam-switching method, $^{238}U^{16}O^-$ and $^{236}U^{16}O^-$ ions were alternately injected and passed on different components and detected by the Faraday cup or detector. Typical injection time was 120 s for ^{236}U and 10 s for ^{238}U. $^{238}U^{5+}$ at 17.07 MeV (i.e., terminal voltage 2.875 MV) and $^{236}U^{5+}$ at 17.26 MeV (i.e., terminal voltage 2.900 MV) are of the same rigidity; hence, no need of changing the analyzing magnet field. For ^{236}U AMS, two kinds of detection methods were used.

3.1 ^{236}U detected by 16-strip detector

The transmission efficiency from low-energy side FC-2 to detector was measured at 2.5% for $^{236}U^{5+}$ ions. Most of the ^{235}U, ^{238}U and other interference ions were suppressed by analyzing components before they transmitted to the detector. The ^{236}U ions were detected by the 16-strip detector. Two kinds of samples (i.e., KkU and standard) were measured alternately and repeated for several times under the same conditions. The 'standard' with a nominal ratio $^{236}U/^{238}U \sim 1 \times 10^{-8}$, obtained by adding a spike of ^{236}U to the KkU VERA-in-house-U standard, and the KkU itself of $^{236}U/^{238}U = (6.98 \pm 0.32) \times 10^{-11}$ were measured [15]. Due to areal blank sample was not easy to get, the peak of 'blank' sample was almost the same as the KkU, under the same conditions except that the voltage of high-energy electrostatic analyzer was 0.8% less than that of ^{236}U transmission. The ion distributions on the strip detector of different samples are shown in Fig. 3. The *Y* axis is the normalized fraction (counts per strip over total counts of the 16-strip detector). The distance between two adjacent strips is 3.625 mm. The separation between ^{236}U and other isotopes is easy to calculate based on parameters of the electrostatic analyzer. The separation between $^{236}U^{5+}$ and interfering $^{238}U^{5+}$ on the detector is 34 mm, and between $^{236}U^{5+}$ and interfering $^{235}U^{5+}$ is 17 mm, which agree well with the experimental results. As shown in Fig. 3, ^{236}U ions are distributed from strip_8 to strip_12 (31–45 mm), and more than 95% ions are in the region of interest (ROI), from strip_9 to strip_11. Due to the charge exchange process and continuous momentum spectrum of interfering isotopes, part of $^{235}U^{5+}$ and $^{238}U^{5+}$ ions transmitted to the detector, and the interference peaks are marked in Fig. 3. The absolute measurement values of isotopic abundance of 'standard' and KkU sample are $^{236}U/^{238}U = (9.51 \pm 0.36) \times 10^{-8}$ and $^{236}U/^{238}U = (8.23 \pm 0.60) \times 10^{-11}$, respectively. This provides a background level isotopic ratio of $^{236}U/^{238}U$ is about 1×10^{-11}.

3.2 ^{236}U detected by TOF-*E* detection system

A TOF detection system with the flight length of 1.5 m was used to discriminate uranium isotopes. Figure 4 shows the TOF spectrum of the silicon strip_10 with the highest $^{236}U^{5+}$ counts for the KkU standards. From theoretical calculation, the different interference peaks between $^{236}U^{5+}$ ions are $\Delta T(^{236}U^{5+}-^{235}U^{5+}) \sim 1.70$ ns, and $\Delta T(^{238}U^{5+}-^{236}U^{5+}) \sim 3.40$ ns. In Fig. 4 each channel means 160 ps, and the peak of ^{235}U, ^{236}U and ^{238}U is marked. Although the resolution is not good enough to separate ^{235}U and ^{236}U by TOF, combining with the energy signal, the detection limit of 5×10^{-12} was obtained.

3.3 Pulse height defect

Since the energy peak of ^{236}U ions did not agree with the expect result, the pulse height defect (PHD) of the

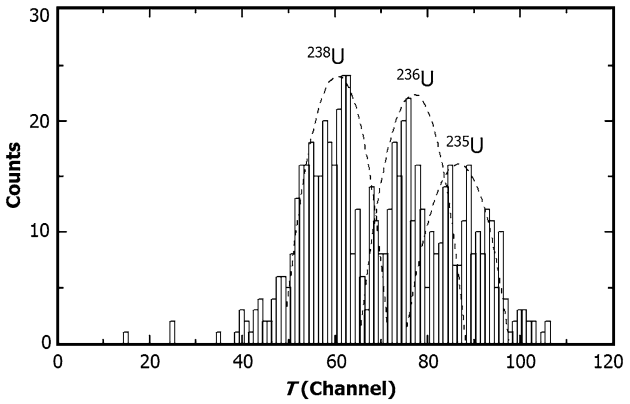

Fig. 4 TOF spectrum showing ^{236}U and the background of ^{238}U (*left peak*) and ^{235}U (*right peak*) in the KkU standard sample

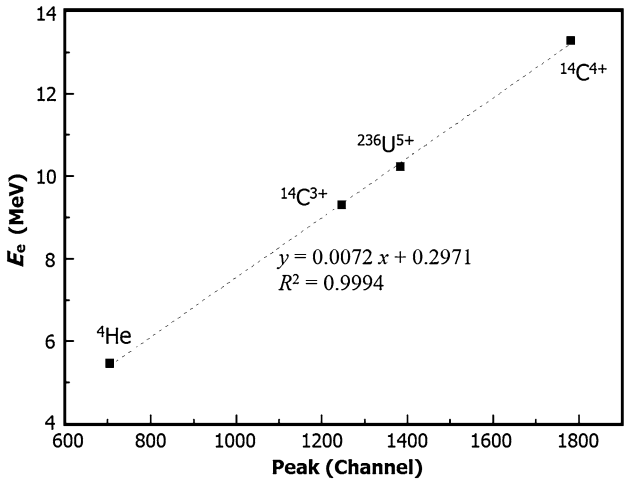

Fig. 5 Channel number versus net energy (E_e) calibration curve for projectiles. E_e is the collected net energy from electronic stopping loss

passivated implanted planar silicon strip detector was measured. The PHD is the difference in detected energy between heavy ions and light ions (e.g., alpha particles) of the same kinetic energy. The PHD of heavy ions in silicon detector consists of three contributions. (1) Window defect E_w that is the energy loss of the heavy ions in the dead layer on the front surface of the detector. (2) The nuclear stopping loss E_N for low-energy nuclear stopping is dominant, the defect is due to non-ionizing collisions of the slow heavy ions with the atoms of the detector. (3) The recombination of electron–hole pairs E_{re}. Along the path of and ionizing heavy particle, a plasma column of high density is formed. The recombination of electron–hole pairs leads to incomplete charge collection and a reduction of detected pulse height.

Pulse height calibration was performed by recording of 5.486 MeV alpha particles from an ^{241}Am source, and 9.535 MeV$^{14}C^{3+}$ and 13.527 MeV$^{14}C^{4+}$ ions from the tandem accelerator. The peak position was determined by fitting a Gaussian to each peak separately. The detail energy losses in dead layer of the detector, the nuclear stopping loss and the electronic stopping loss are given in Table 1. E_w of the above ions passing through the dead layer with the maximum thickness (i.e., 50 nm) of detector was calculated with SRIM 2013. E_N and E_e mean the energy loss caused by nuclear stopping and electronic stopping, respectively. The values of E_N and E_e were obtained by integrate the energy loss curve derived from the SRIM calculation results. For ^{236}U, with high mass number and low incidence energy, the energy loss contribution from nuclear stopping is about 6.336 MeV, only 10.232 MeV, the energy loss by electronic stopping can be collected. Figure 5 shows the calibration curve of energy loss (E_e) versus channel number for alpha, ^{14}C and ^{236}U ions. E_e is the collected net energy from electronic stopping loss. After considering the nuclear stopping loss, there is an excellent agreement between E_e and peak in Fig. 5. In this case, the contribution of the recombination of electron–hole pairs (E_{re}) effect to the PHD can be ignored.

Table 1 Detail energy loss (in MeV) of incident ions

Ions	Energy	Peak (Ch.)	E_W	E_N	E_e
α	5.486	703.5	0.022	0.01	5.454
$^{14}C^{3+}$	9.535	1247	0.165	0.073	9.297
$^{14}C^{4+}$	13.527	1782	0.163	0.078	13.286
$^{236}U^{5+}$	17.26	1348	0.692	6.336	10.232

4 Conclusion

^{236}U AMS measurement was performed at CIRCE by the position sensitivity detection method with a background level of $^{236}U/^{238}U$ is 1×10^{-11}, and the TOF detection method with a background level of $^{236}U/^{238}U$ is 5×10^{-12}, respectively. The pulse height defect is obvious in ^{236}U energy measurement, and the energy difference is 6.336 MeV. The result shows that the contribution of pulse height defect is mainly from the nuclear stopping loss.

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