

Primary result of ^{236}U measurement with accelerator mass spectrometry at CIAE*

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Abstract The rare isotope ^{236}U has a half-life of $2.342(3)\times 10^7$ years, and is produced principally by thermal neutron capture on ^{235}U . The isotopic atom ratio of $^{236}\text{U}/^{238}\text{U}$ depends on the integral thermal neutron flux received by the material of interest. ^{236}U is potentially useful as a “fingerprint” for indicating the presence of neutron-irradiated uranium usually originating from nuclear activity. By extracting negative molecular ion UO^- from the uranium oxide target, simulating the $^{236}\text{U}^{16}\text{O}^-$ beam transport with $^{238}\text{U}^{16}\text{O}^-$ and $^{208}\text{Pb}_2^{16}\text{O}^-$ pilot molecular ion beam, transporting the ^{236}U -containing ion beam with a high resolution injection magnet analyzer and electrostatic analyzer system, and finally identifying and detecting ^{236}U with a time-of-flight detector (TOF), a method for AMS (Accelerator Mass Spectrometry) measurement of ^{236}U was established on the HI-13 Accelerator AMS system at China Institute of Atomic Energy.

Key words ^{236}U , accelerator mass spectrometry, isotopic ratio

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1 Introduction

The rare isotope ^{236}U has a half-life of $2.342(3)\times 10^7$ years, and is produced principally by thermal neutron capture on ^{235}U . The thermal neutron capture cross-section of ^{235}U ($\sigma[^{235}\text{U}(n, \gamma)]=98$ b) is about one sixth of its fission cross-section ($\sigma[^{235}\text{U}(n, f)]=583$ b). The $^{236}\text{U}/^{238}\text{U}$ (atom number ratio) is around 10^{-10} – 10^{-14} in nature, depending on the integral thermal neutron flux received by the material. The $^{236}\text{U}/^{238}\text{U}$ could be used as an integrating neutron monitor over the last 100 M years [1]. Therefore, ^{236}U is potentially useful as a “fingerprint” for indicating the presence of neutron-irradiated uranium usually originating from nuclear activity.

The measurement of naturally occurring ^{236}U is difficult or even impossible without AMS (Accelerator Mass Spectrometry). AMS offers significant advantages over conventional mass spectroscopic meth-

ods in high sensitivity, smaller sample size, relatively free from isomer and molecular-ion interferences, and shorter measurement time. X. L. Zhao et al. of Toronto group [2] first reported that the $^{236}\text{U}/^{238}\text{U}$ is lower than 5×10^{-10} in a natural uranium ore from Cigar Lake deposit in Canada by using AMS technique. The natural $^{236}\text{U}/^{238}\text{U}$ has also been measured by several groups [3–7], with typical from 10^{-12} to 10^{-8} .

In this paper, a method for the measurement of ^{236}U with AMS on the HI-13 tandem accelerator at China Institute of Atomic Energy (CIAE) is reported.

2 Experimental method

Since the installation of the AMS facility at CIAE in 1989, ^{26}Al , ^{36}Cl , ^{41}Ca , ^{55}Fe , ^{64}Cu , ^{79}Se , ^{99}Tc , ^{126}Sn , ^{129}I , ^{151}Sm and ^{182}Hf have been measured, and applications in the fields of biomedicine, nuclear physics,

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nuclear astrophysics and environmental science [8–12] have also been carried out. The accelerator mass spectrometry system on the HI-13 tandem accelerator at CAIE with a newly upgraded injector system is depicted schematically in Fig. 1.

2.1 Ion source

The so-called Multi-Cathode Source for Negative Ions with a Cesium Sputter (MC-SNICS) [13] has been used in the CIAE AMS system. The MC-SNICS source contains a target wheel holding up to 40 samples. Under normal running conditions, each sample can be sputtered with the cesium beam for several hours.

In order to characterize the source performance for ^{236}U measurement, experiments on extracting multiform of negative ^{238}U -containing molecular ions were carried out. The typical results of different targets and different molecular ions are listed in Table 1, where the beam currents were measured

directly with the Faraday cup following the injection magnet. The results show that the beam currents of all the candidate extracting ions tested, UO^- , UF_2^- and UF_5^- , are very small. Compared with UO^- , UF_5^- has much lower transmission and lower charge state after passing through the stripper foil. So, extracting UF_5^- is not the best choice. When $^{236}\text{U}^{19}\text{F}_2$ ions are extracted, $^{238}\text{U}^{17}\text{O}^{19}\text{F}^-$ ions will be accompanied. If $^{236}\text{U}^{16}\text{O}^-$ ions are extracted, the more abundant $^{235}\text{U}^{17}\text{O}^-$ and $^{234}\text{U}^{18}\text{O}^-$ ions will be accompanied. Considering the natural abundances of ^{238}U ($\sim 99.28\%$), ^{235}U ($\sim 0.72\%$) and ^{17}O ($\sim 0.037\%$), the interference from $^{238}\text{U}^{17}\text{OF}^-$ ($99.28\% \times 0.037\% = 2.7 \times 10^{-5}$ in theory) is larger than that from $^{235}\text{U}^{17}\text{O}^-$ ($0.7\% \times 0.037\% = 2.7 \times 10^{-7}$ in theory). Furthermore, the UF_2^- 's transmission efficiency is also lower than that of UO^- . Taking what is mentioned above into consideration, UO^- may be the most suitable form of ions to be extracted from the target.

Table 1. Negative ion beam currents from different types of uranium targets.

target	ion: beam current/nA		
$\text{UO}_2+\text{Nb}(1:1)$	$^{238}\text{UO}^-$: 18.1 nA	$^{238}\text{UO}_2^-$: 5.5 nA	$^{238}\text{UO}_3^-$: 1.4 nA
$\text{UF}_4+\text{Nb}(1:1)$	$^{238}\text{UF}^-$: 2 nA	$^{238}\text{UF}_2^-$: 19.3 nA	$^{238}\text{UF}_3^-$: 5 nA
	$^{238}\text{UF}_4^-$: 8 nA	$^{238}\text{UF}_5^-$: 32.2 nA	$^{238}\text{UF}_6^-$: 0.7 nA
$\text{UO}_2+\text{C}(1:1)$	$^{238}\text{UC}^-$: 2 nA	$^{238}\text{UC}_2^-$: ~ 1 nA	$^{238}\text{UC}_3^-$: ~ 1 nA
	$^{238}\text{UC}_4^-$: ~ 0.5 A		

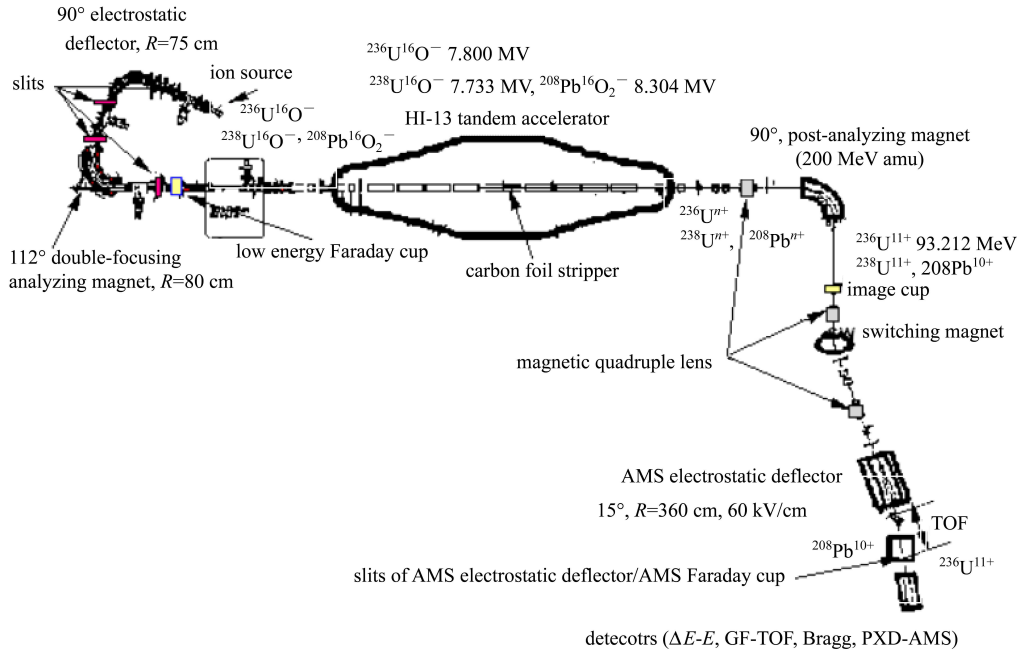


Fig. 1. Schematic layout of the accelerator mass spectrometer based on HI-13 tandem accelerator after the upgrade of the injector system for heavy ions.

Different conductor powders, Nb, Al and Ag, were tested for UO_2 sample with UO^- as the form of ions to be extracted, the beam current from UO_2+Nb (1:1) target turned out to be the largest, very stable for over three hours and suitable for alternated measurements in AMS.

2.2 Injection system

The original setup with the mass resolution of $M/\Delta M \approx 220$ could not satisfy the requirement for AMS measurement of heavy isotopes (e.g., ^{236}U , ^{182}Hf , ^{151}Sm). Therefore, a high resolution achromatic beam line that consists of a 90° electrostatic analyzer (ESA) and a 112° double-focusing analyzing magnet (MA) (as shown in Fig. 1) was installed for AMS. This spherical 90° ESA with 75 cm radius, has a maximum electric field strength of 4 kV/cm and provides both horizontal and vertical focusing. The 112° double-focusing MA with a bending radius of 80 cm, has maximum magnetic field of 1.3 T. The energy resolution of the electrostatic deflector is $\frac{E}{\Delta E} = \frac{D\rho}{S_2 - MS_1}$. The Mass resolution of the analyzing magnet is $\frac{M}{\Delta M} = \frac{D\rho}{S_2 - MS_1}$, where $D\rho$ is coefficient of chromatic dispersion (3000 for ESA, 3360 for MA), M (-1 for ESA, -1.14 for MA) is the coefficient of magnifier, S_2 and S_1 are the slits of object and image. The new injector system was tested by using the sample material of HfO_2 . When the ESA's slits and MA's object slits are opened up, the image point slits (x direction) of MA are 1 mm, the mass resolution of the injector system is about 430. If the object point and image point slits of MA are respectively set at ± 2.5 mm and ± 1 mm, the mass resolution can be increased to 630, which leads to a decrease of beam current only by 5%, compared with the beam current of slits completely opened up. ^{236}U AMS measurements require a high resolution injector system to clip the tail from ^{235}U ions. The negative ions are pre-accelerated to the maximum injection energy $E_{\text{inj}} = 120$ keV.

2.3 The accelerator and beam transport

The terminal voltage was carefully chosen for ^{236}U measurement on the HI-13 tandem accelerator mass spectrometer, with an upper limit set by the post-analyzing magnet (i.e., 200 MeV·amu). High terminal voltages have advantages of lower background due to energy straggling and higher accelerator transmission. On the other hand, higher energy requires a higher field in the post-analyzing and switching magnets. For $^{236}\text{U}^{11+}$ ions, the maximum usable termi-

nal voltage was 7.8 MV, which corresponds to a final energy of 93.2 MeV, with a charge state fraction of about 14%. The terminal voltage is stabilized through the current modulated by corona probe. The signals from a generation voltmeter measuring (GVM) the terminal voltage are used for voltage stabilization. The terminal voltage can be precisely controlled within an accuracy of 0.1% for the ^{236}U AMS measurement. In the terminal of the HI-13 tandem accelerator, a carbon foil of $3 \mu\text{g}\cdot\text{cm}^{-2}$ thickness was used as a stripper. At the high-energy side, $^{236}\text{U}^{11+}$ ions were analyzed.

At the beginning of our work, the ^{236}U beam transport was simulated with the sample material of UO_2 and extracting ions of $^{238}\text{U}^{16}\text{O}^-$. Due to the small beam current (20–100 nA) and the significant scattering induced by the carbon foil and Coulomb explosion, the beam current in high-energy side was too small to be tuned for beam transport simulation. In order to tune the accelerator, the transportation simulation was divided into two steps by using $^{238}\text{UO}_2$ and $^{208}\text{Pb}^{16}\text{O}_2$ pilot beams, respectively. An outline of the beam transport simulation in the ^{236}U -AMS with a $^{208}\text{Pb}^{16}\text{O}_2$ molecular ion pilot beam method developed at CIAE is depicted in Fig. 1. Firstly, in order to stabilize the terminal voltage by using the GVM and ensure optimum optics state for ^{236}U determination, $^{238}\text{U}^{16}\text{O}^-$ ion beam was extracted from the UO_2 sample and relevant parameters of the ionic optical system were adjusted to optimize the beam transport from lower energy system to the image of the post-analyzing magnet, where $^{238}\text{U}^{11+}$ was selected with the same magnetic rigidity as for $^{236}\text{U}^{11+}$ at 7.8 MV. Secondly, the sample material of PbO_2 , instead of UO_2 , was used and $^{208}\text{Pb}^{16}\text{O}_2^-$ ions were extracted from the ion source to simulate the $^{236}\text{U}^{11+}$ transport from the image of post-analyzing magnet to the AMS Faraday cup, while keeping all the parameters already optimized in the first step unchanged. At this step, $^{208}\text{Pb}^{10+}$ was selected with the same magnetic rigidity as for $^{236}\text{U}^{11+}$ at 7.8 MV. The experiment showed that the two-step simulation method for the optimization of ^{236}U beam transportation makes the adjustment much easier.

The transmission efficiency from the low-energy side Faraday cup to the detector system was measured to be about 2.5×10^{-4} for $^{238}\text{U}^{11+}$. This poor beam transmission limits the efficiency of ^{236}U measurements. In the future, we intend to investigate methods to improve the overall transmission.

2.4 Detector system

For ^{236}U AMS measurement, only the time of

flight (TOF) detector has sufficient resolution to distinguish the neighboring isotopes which happen to pass the ion optical filters. Fig. 2 shows the TOF detector system for the ^{236}U measurement. The TOF system consists of a micro-channel plate (MCP) and a gold-silicon surface barrier detector (SBD), separated by a flight path of 1.8 m. The secondary electrons induced by the incoming ions at a $7\ \mu\text{g}\cdot\text{cm}^{-2}$ carbon foil are reflected by an electrostatic mirror, collected

and multiplied by MCP. In order to avoid the noise coincidence, the signals from SBD have a rise time of about 30 ns at the bias voltage of 300 V, used as the start signal of TAC567 (time-to-amplitude converter). The SBD detector is also used as energy detector for mass/charge discrimination, since the nuclide of interest (^{236}U) has an energy gain different from that of interfering isotopes (isobars) in the acceleration process.

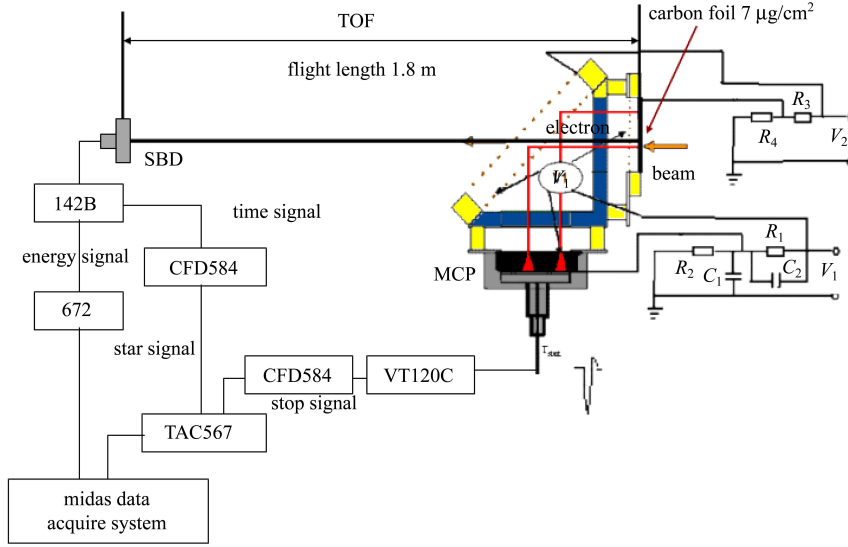


Fig. 2. Schematic setup of the TOF detection system.

In order to get a good time resolution of the TOF, the threshold and constant fraction were set carefully to optimize the CFD584 (constant fraction discrimination). The length of the $50\ \Omega$ coaxial cable in CFD584 for constant-fraction shaping delay was calculated according to the formula $L = \frac{t_{d(\text{Ext})}}{\lambda} \approx \frac{1.1t_r - 0.7}{\lambda}$ ($\lambda = 5\ \text{ns}$, delay time of 1 m $50\ \Omega$ -coaxial cable; t_r is the rise time).

3 Results

Scaling the main components by changing the voltage of AMS electrostatic deflector to the mass we want to measure. Fig. 3 shows the scan spectra of electrostatic deflector. The isotopes were counted directly with SBD. The image slits of the AMS electrostatic deflector were set to $4\ \text{mm} \times 7\ \text{mm}$ for clipping the tail of $^{235}\text{U}^{11+}$ and $^{238}\text{U}^{11+}$ ions. The AMS electrostatic deflector rejection of isotopes (abundance sensitivity) is $\sim 6 \times 10^2$, at which point abundance sensitivity is expected to be limited by the factors such

as the energy stability of the accelerator, the formation of $^{235}\text{U}^{17}\text{O}$ and $^{238}\text{U}^{14}\text{N}$ ions, and the scattering in the high energy spectrometer.

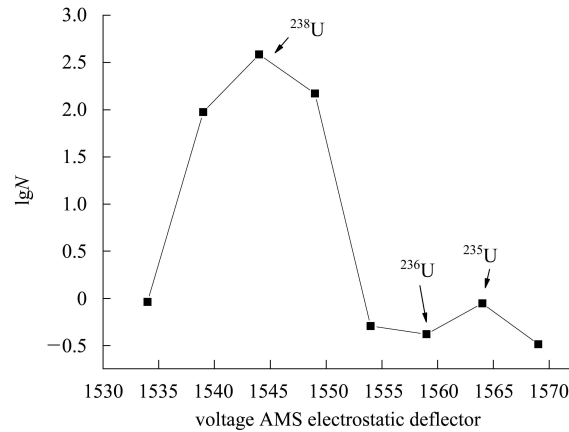


Fig. 3. The scan spectrum of the AMS electrostatic deflector, $N = \text{counts/s}$.

We test the system using 3 depleted uranium samples. Fig. 4 shows the TOF spectra and TOF vs the

energy two dimension spectra of ^{236}U ions from sample 1 and sample 2. The TOF spectrum is gated through a logical window of energy. The TOF resolution (FWHM) is about 800 ps, which depends on the quality and the thickness of the MCP foil ($3\ \mu\text{g}\cdot\text{cm}^{-2}$) and the rise time of the SBD (30 ns). The total transmission of uranium ions at 93.2 MeV from the “start” MCP to the SBD is measured to be $\sim 45\%$, resulting from the angular spread of the heavy-ion beam

after the TOF carbon foils and the shadow of the wire grids. For ions having the same rigidity and the same charge state, the TOF is proportional to the ion mass and so the TOF mass resolution for ^{236}U is $M/\Delta M \approx 220$. The main background ions near the $^{236}\text{U}^{11+}$ are $^{235,238}\text{U}^{11+}$, which enter the accelerator as molecular ions of $^{235}\text{U}^{17}\text{O}^-$, $^{238}\text{U}^{12}\text{CH}_2^-$ and $^{238}\text{U}^{14}\text{N}$ with the mass number of 252. These interfering ions are unresolved from $^{236}\text{U}^{16}\text{O}^-$.

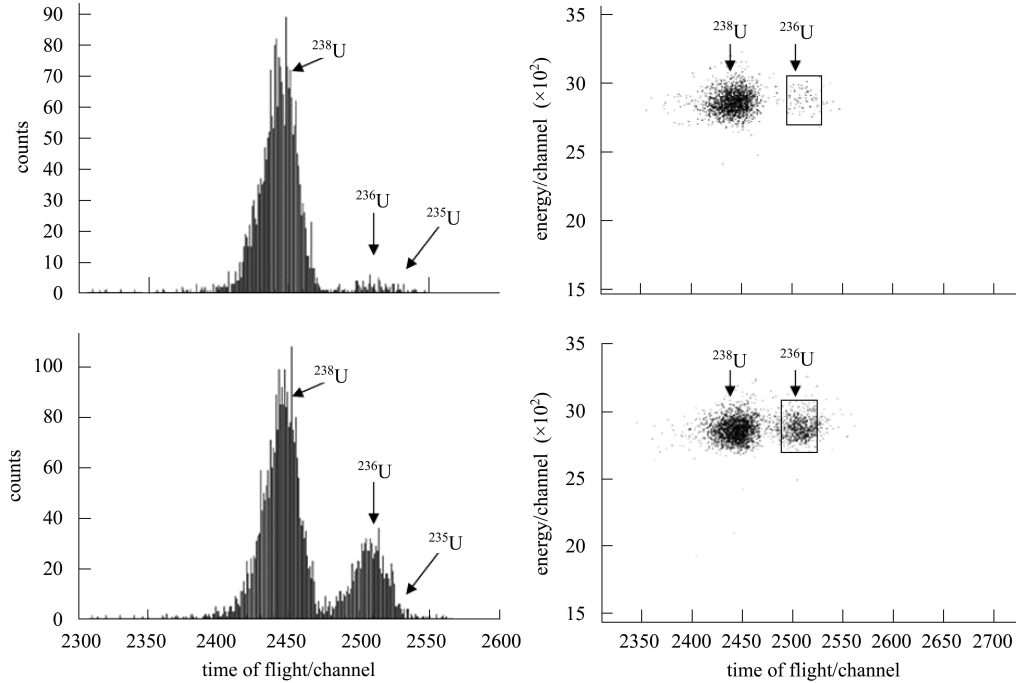


Fig. 4. The TOF spectra and TOF vs the energy two dimension spectra of ^{236}U ions for sample 2 and sample 3.

Table 2. Results of $^{236}\text{U}/^{238}\text{U}$ ratios and ^{235}U abundances for 3 samples.

sample	$^{236}\text{U}/^{238}\text{U}$ ratio	^{235}U (atoms. %)
1 ^{a)}	$(2.710 \pm 0.008) \times 10^{-5}$	~ 0.3
2	$(8.9 \pm 0.9) \times 10^{-9}$	~ 0.72
3	$(4.6 \pm 0.4) \times 10^{-8}$	~ 0.72

a) measured with Thermal ionisation mass spectrometry, and used for normalization.

The first measurement results of ^{236}U are presented in Table 2. The $^{236}\text{U}/^{238}\text{U}$ of sample 1 is $(2.710 \pm 0.008) \times 10^{-5}$, measured with TIMS and used for normalization. sample 2 (UO_2) was purchased by CIAE in 1960s. sample 3 is from sample 2 contaminated by sample 1 during the sample preparation. The $^{236}\text{U}/^{238}\text{U}$ for sample 2 and sample 3 is $(8.9 \pm 0.9) \times 10^{-9}$ and $(4.6 \pm 0.4) \times 10^{-8}$, respectively. The main contribution to the uncertainties is the poor counting statistics due to the small number of ^{236}U

atoms recorded in the detector. The sensitivity is lower than 10^{-9} for $^{236}\text{U}/^{238}\text{U}$ in present work.

4 Discussion

By extracting negative molecular ions UO^- from the sample $\text{UO}_2 + \text{Nb}$ (1:1), simulating the transport of ^{236}U ions with the newly proposed $^{208}\text{Pb}^{16}\text{O}_2^-$ molecular ions, identifying the interference isotopes by a high resolution injection magnet analyzer, an electrostatic analyzer and a TOF detector, a method for AMS measurement of ^{236}U has been established on the HI-13 Accelerator at CIAE. The sensitivity is lower than 10^{-9} for $^{236}\text{U}/^{238}\text{U}$ in present work.

There is still room for further improvement in the AMS measurement of ^{236}U on our AMS system. The following efforts are being made in dealing with the major limiting factor for highly sensitive measurement of ^{236}U :

1) Improvement in stripper system

Carbon foil stripper was used in our measurement system that would increase the yield for high charge-states (high energy) ions. But the accelerator transmission is poor, because the beam quality (small divergence and narrow energy distribution) decreases due to Coulomb explosion in the foil, especially for heavy ion $^{236}\text{U}^{11+}$. If gas stripper is used, the chance of Coulomb explosion will be reduced. However, that will lead to lower charge-states (lower energy) of ^{236}U ions and limit the maximum magnetic rigidity of the post analyzing magnets (200 MeV·amu). Solid stripper is different from the gas stripper [14]. In the former, the dissociation process undergoes roughly two separated steps, the first being the fast removal of few outer electrons and the second being a violent Coulomb explosion, usually resulting in well resolved rings on a three dimensional plot of intensity-energy-angle. Based on the above discussion, a combination of gas + foil stripper may reduce the three dimensional intensity-energy-angle plots and improve the beam quality.

2) Improvement in vacuum system

Interactions of the ions of interest with residual gas in the accelerator tubes may change the charge state and movement direction of the ions, and lead to the unwanted transmission of interfering ions with either the wrong mass or the wrong energy. This is a small effect, but the $^{236}\text{U}/^{238}\text{U}$ is also very small and

the effect ultimately becomes the main background in the detection. The best possible vacuum and higher pre-accelerated voltage are needed in the accelerator tubes to minimize the charge-exchange interactions and the elastic scattering of the ions to be determined, especially the heavy ions of ^{236}U .

3) Improvement in detection system

The TOF system currently used in our measurement does not have sufficiently high resolution to identify isotope ^{235}U . Besides, the transmission of the TOF system is also poor. We are planning to establish a new TOF system with two MCPs, one energy detector, and using the ultra-thin diamond-like carbon foil [15] (DLC foils) as the entry window. The new TOF system may have a resolution of ~ 600 ps, sufficient to identify the isotope ^{235}U for ^{236}U AMS measurement.

With the improvements mentioned above, a sensitivity of or better than 10^{-11} is expected for the AMS measurement of $^{236}\text{U}/^{238}\text{U}$. Besides, blank samples with the lowest possible ^{236}U content are needed to test the system. In the near future, we plan to analyze water samples with extremely low ^{236}U concentration. The method of ^{236}U AMS measurement can be applied to other long-lived actinide isotopes, such as ^{237}Np , ^{239}Pu , ^{240}Pu , ^{242}Pu and ^{244}Pu .

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