ACCELERATOR MASS SPECTROMETRY AT THE TSUKUBA 12 MV PELLETRON TANDEM ACCELERATOR*

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Abstract

Accelerator Mass Spectrometry (AMS) is a highly sensitive mass spectrometric method for measuring rare isotopes. The technique is mainly applied in chronology, earth and environmental sciences to date samples using long-lived radioisotopes. With a multi-nuclide AMS system on the 12 MV Pelletron tandem accelerator at the University of Tsukuba (Tsukuba AMS system), we are able to measure environmental levels of long-lived radioisotopes of ^{26}Al, ^{36}Cl and ^{129}I by employing a molecular pilot beam method. The present status of the Tsukuba AMS system is reported in this paper.

INTRODUCTION

The Tsukuba AMS system is based on the NEC 12UD Pelletron tandem accelerator which can supply a maximum terminal voltage of 12 MV. The 12 MV Pelletron tandem accelerator was installed at the University of Tsukuba, Tandem Accelerator Complex (UTTAC) in 1975 [1]. In its early stages, the 12 MV Pelletron tandem accelerator was principally used for nuclear physics research. In recent years, the main research field of the 12 MV Pelletron tandem accelerator has shifted to AMS from nuclear physics. In 2009, the beam time for AMS research reached to about 30% of the total operation time. It is important for sensitive measurements of heavy radioisotopes that background interference of their stable isobars are suppressed by AMS measurements. The high terminal voltage is an advantage in the detection of heavy radioisotopes because the relative differences in the energy loss in the detector for the isobaric interference and target radioisotopes are clearly separated. The Tsukuba AMS system has been continually developed since 1993 [2] and it is currently capable of measuring environmental levels of long-lived radioisotopes of ^{26}Al, ^{36}Cl and ^{129}I.

THE 12 MV PELLETRON TANDEM ACCELERATOR AT THE UTTAC

Figure 1 shows a schematic view of the 12 MV Pelletron tandem accelerator facility at the University of Tsukuba. It is a vertical tandem Van de Graaff accelerator. Three ion sources are set on the 9th floor. Two experimental rooms on the 1st floor are located on opposite sides of the accelerator tower. In 2009, we replaced the old point-to-plane corona needle system with a resistor-based potential grading system. The divided resister system allows the terminal voltage to vary from 1 to 12 MV without shorting column units. The divided resister system is also expected to improve the stability of the terminal voltage.

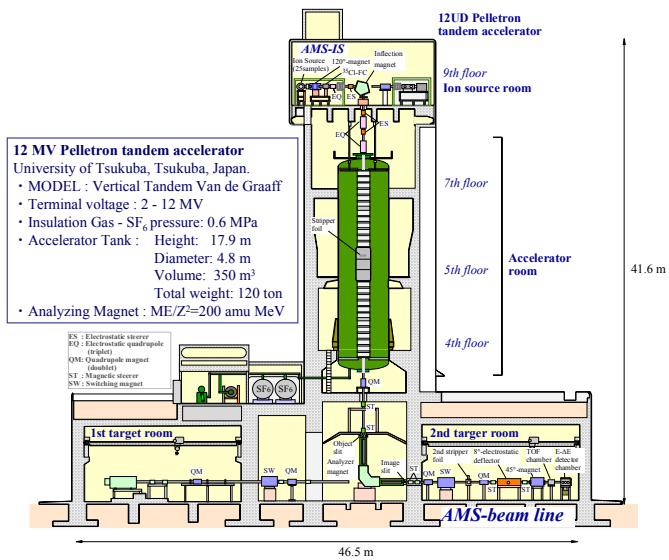

Figure 1: Schematic view of the 12 MV Pelletron tandem accelerator facility at the University of Tsukuba.

THE TSUKUBA AMS SYSTEM

Figure 2 shows a schematic layout of the Tsukuba AMS system. An original Cs sputtering ion source has an automatic changer of 25 samples on a rotating wheel. The ions extracted from the ion source are analyzed by a 120° mass separator. Beam currents of the major stable nuclei are measured by off-set Faraday cups just behind the 120° mass separator. A carbon stripper foil with a thickness of 5 μg/cm^2 and a diameter of 16 mm is used to change the ion charge state from negative to positive at the terminal. An AMS mass separator beam line after the 90° analyzing magnet (ρ= 1.28 m, ME/q^2= 200 MeV amu, p/Δp= 7.9 × 10^3) consists of a second carbon stripper foil, an 8° electrostatic energy analyzer(ρ= 10 m, E/q= 10 MeV), a 45° magnetic momentum analyzer, a TOF detector and a gas ΔE – E counter telescope. A second stripper carbon foil with a thickness of 15 μg/cm^2 and a diameter of 20 mm is used to eliminate the pilot ions from the mass separator beam line. The gas ΔE – E counter telescope which consists of two gas ΔE sections and an ion

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implanted silicon surface barrier detector with a 45 × 45 mm^2 active area. A 4 μm aramid film is used as the entrance window of the gas detector. Pure iso-butane gas with a pressure of 750 Pa is applied in the gas section for AMS measurements. Figure 3 shows the AMS mass separator beam line.

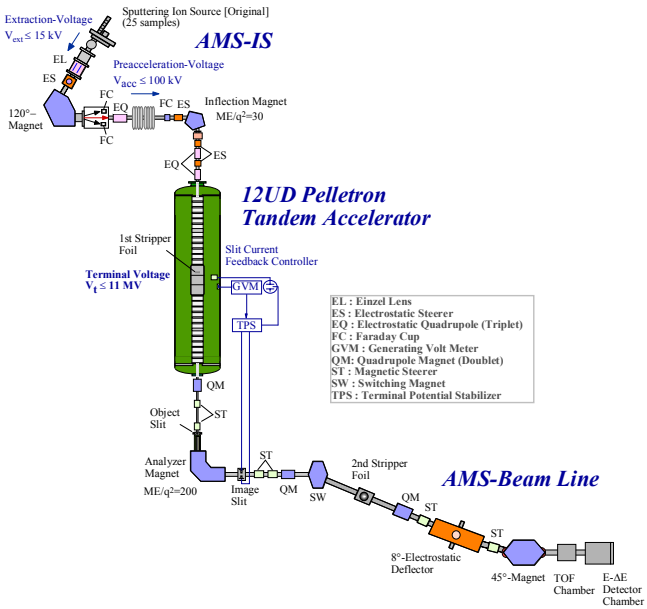

Figure 2: Schematic layout of the Tsukuba AMS system.

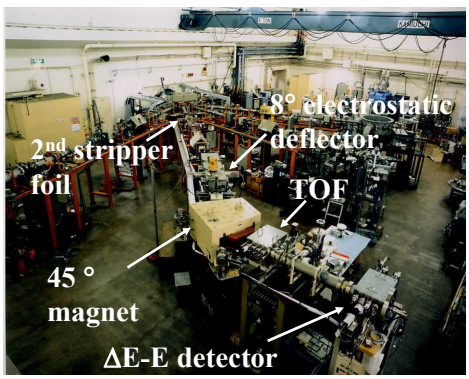

Figure 3: AMS mass separator beam line.

A pilot beam method is used to stabilize the terminal voltage through slit current feedback instead of GVM stabilization for the Tsukuba AMS system. For our system, ions of the long-lived radioisotopes and a molecular pilot beam with the same particle mass are extracted simultaneously from the ion source and injected into the accelerator. The terminal voltage is precisely controlled by the slit current feedback system with the slit current generated by the pilot beam. The terminal voltage is kept stable within an accuracy of 0.02 % by this method [2]. When we perform ^{36}Cl–AMS measurements (see Figure 4), $^{12}C_3^-$ tri-molecular ions are accelerated simultaneously with $^{36}Cl^-$ ions. After acceleration with a terminal voltage of 10 MV, $^{36}Cl^{9+}$ at 100 MeV and $^{12}C^{3+}$ at 33.3 MeV have the same mass energy product ME/q^2 = 44.4 MeV amu, and therefore, they are able to pass through the same orbit in the 90° analyzing magnet. The beam current of the $^{12}C^{3+}$ ions measured by the image slit is used for the slit current feedback system. After the second stripper carbon foil, $^{36}Cl^{14+}$ and interference ions are selected and detected by the gas ΔE – E counter telescope.

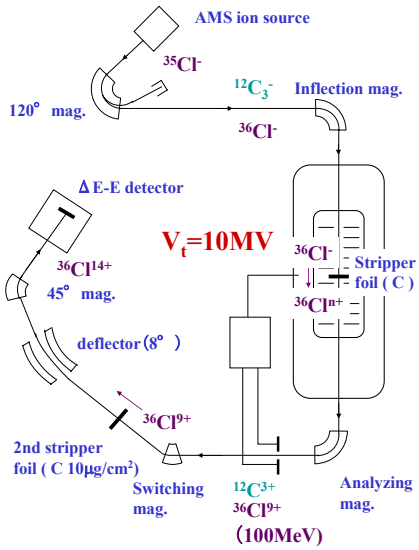

Figure 4: Schematic diagram of ^{36}Cl–AMS.

CURRENT PERFORMANCE OF THE TSUKUBA AMS SYSTEM

Table 1 shows the current performance of the Tsukuba AMS system. The Tsukuba AMS system is currently capable of measuring environmental levels of long-lived radioisotopes of ^{26}Al, ^{36}Cl and ^{129}I.

^{26}Al-AMS

In the pilot beam method, the target sample is prepared as an Al_2O_3 powder mixed with silver and enriched $^{26}MgO_2$. $^{26}MgO^-$ molecular ions are used as the pilot beam to control the stability of the terminal voltage. The maximum beam current of AlO^- extracted from an Al_2O_3 sample is more than 1.5 μA. The accelerator is operated at a terminal voltage of 10.2 MV, and $^{26}Al^{7+}$ and $^{26}Mg^{7+}$ ions with energies of 78 MeV are selected by the 90° analyzing magnet. $^{26}Al^{7+}$ ions are fully stripped to $^{26}Al^{13+}$ ions by a second carbon stripper foil and then ^{26}Al and ^{26}Mg are clearly separated by the subsequent spectrometer. Figure 5 shows a ^{26}Al spectrum of a standard sample. The beam transmission of fully stripped Al^{13+} ions from AlO^- is up to 10%. The detection limit for the ^{26}Al/Al ratio is better than 5×10^{-15} [3].

^{36}Cl-AMS

A typical beam current for $^{35}Cl^-$ is up to 20 μA. $^{36}Cl^{14+}$ with an energy of 100 MeV is detected by the gas ΔE – E counter telescope. Schematic diagram for ^{36}Cl-AMS is already shown in Figure 4. Figure 6 shows a ^{36}Cl spectrum for a standard sample of $^{36}Cl/Cl = 1.60 \times 10^{-12}$.

Table 1: Current performance of ^{26}Al, ^{36}Cl and ^{129}I AMS by the Tsukuba AMS system

Nuclide	^{26}Al ($T_{1/2}$=7.05 × 10^5 yr)	^{36}Cl ($T_{1/2}$=3.01 × 10^5 yr)	^{129}I ($T_{1/2}$=1.57 × 10^7 yr)
Target material	Al_2O_3 + $^{26}MgO_2$ + Ag	AgCl + C_{60}	AgI+MoO_2+Nb
Injection ion	$^{26}AlO^-$	$^{36}Cl^-$	$^{129}I^-$
Pilot beam	$^{26}MgO^-$	$^{12}C_3^-$	$^{97}MoO_2^-$
Reference ion	$^{27}AlO^-$	$^{35}Cl^-$ & $^{37}Cl^-$	$^{127}I^-$
Typical current of reference ion	1.5 μA	10 μA & 2.5 μA	7 μA
Injection energy	115 keV	103 keV	103 keV
Terminal voltage	10.2 MV	10 MV	9.68 MV
Particle energy	78 MeV ($^{26}Al^{7+}$)	100 MeV ($^{36}Cl^{9+}$)	125.8 MeV ($^{129}I^{12+}$)
Detected ion	$^{26}Al^{13+}$	$^{36}Cl^{14+}$	$^{129}I^{26+}$
Background	$^{26}Al/^{27}Al < 5 \times 10^{-15}$	$^{36}Cl/Cl < 1 \times 10^{-15}$	$^{129}I/^{127}I < 1 \times 10^{-13}$
Typical precision	≤ 5 %	≤ 2 %	≤ 8 %
Number of targets	20 /year	450 /year	20 /year

The Tsukuba AMS system achieves complete discrimination between ^{36}Cl and ^{36}S up to a counting rate of ~5 kHz. The background level measured for a blank sample is better than 1 × 10^{-15} for the ^{36}Cl/Cl ratio [4]. The precision for the ^{36}Cl–AMS system is typically 2% which is determined from the reproducibility of standard sample measurements.

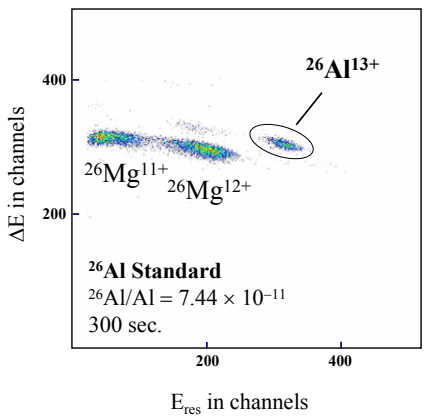

Figure 5: ^{26}Al spectrum of a standard sample for $^{26}Al/Al = 7.44 \times 10^{-11}$.

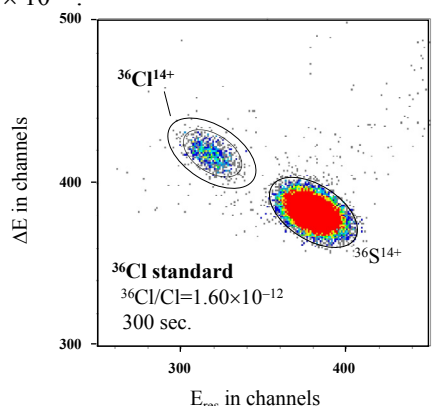

Figure 6: ^{36}Cl spectrum of a standard sample for $^{36}Cl/Cl = 1.60 \times 10^{-12}$.

^{129}I-AMS

A $^{97}Mo^{16}O_2$ molecular pilot beam is applied to ^{129}I–AMS [4]. $^{129}I^-$ and $^{97}Mo^{16}O_2^-$ ions are accelerated concurrently and $^{129}I^{12+}$ and $^{97}Mo^{9+}$ ions can pass through the 90° analyzing magnet. After passing through the second stripper foil, $^{129}I^{26+}$ is selected as the detection particle. $^{129}I^{26+}$ ions are clearly detected by a silicon surface barrier detector. The terminal voltage is set to 9.68 MV. The background level of $^{129}I/^{127}I$ is better than 1 × 10^{-13}.

SUMMARY

In recent years, the 12 MV Pelletron tandem accelerator is principally used for AMS research. For ^{26}Al–AMS, the ions extracted from the ion source are AlO^- by using the Al_2O_3 target sample. Also, the fully-stripping AMS technique is used to achieve an effective detection limit of better than 5 × 10^{-15} for the $^{26}Al/Al$ ratio. The terminal voltage of 10 MV is used for ^{36}Cl–AMS, thereby the energy of the $^{36}Cl^{9+}$ is achieved to 100 MeV. This energy increment helps us to get a clearer separation between ^{36}Cl and ^{36}S in the gas ΔE – E counter telescope. The standard deviation of the fluctuation for the $^{36}Cl/Cl$ ratio is within 2 %, and the effective detection limit is better than 1 × 10^{-15}. For ^{129}I–AMS, the background level is better than 1 × 10^{-13}. We have measured over 500 samples in 1 year by the Tsukuba AMS system, including samples for earth and environmental sciences and nuclear safety research.

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