# PROGRESS OF AN ACCELERATOR MASS SPECTROMETRY SYSTEM ON THE TSUKUBA 12UD PELLETRON TANDEM ACCELERATOR*

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*Abstract*

The 12UD Pelletron tandem accelerator was installed at the University of Tsukuba in 1975. In recent years, the main research field of the 12UD Pelletron tandem accelerator has shifted to accelerator mass spectrometry (AMS) research from nuclear physics. AMS is an ultra-sensitive technique for the study of long-lived radioisotopes, and stable isotopes at very low abundances. The high terminal voltage is an advantage in the detection of heavy radioisotopes. It is important for sensitive measurements of heavy radioisotopes that background interference of their stable isobars are suppressed by AMS measurements. With the multi-nuclide AMS system at the University of Tsukuba (Tsukuba AMS system), we are able to measure long-lived radioisotopes of $^{14}$C, $^{26}$Al, $^{36}$Cl and $^{129}$I by employing a molecular pilot beam method that stabilize the terminal voltage with 0.1% accuracy. Much progress has been made in the development of new AMS techniques for the Tsukuba AMS system. As for $^{36}$Cl AMS, $^{36}Cl^{9+}$ at 100 MeV is used for AMS measurements. The standard deviation of the fluctuation is typically ± 2%, and the machine background level of $^{36}$Cl/Cl is lower than $1 \times 10^{-15}$. This report presents the overview and progress of the Tsukuba AMS system.

## INTRODUCTION

The 12 UD Pelletron tandem accelerator was manufactured by National Electrostatic Corp. (NEC), USA and was installed at the University of Tsukuba, Tandem Accelerator Complex (UTTAC) in 1975 [1]. A maximum terminal voltage of 12 MV is available for various ion beam applications. Figure 1 shows a layout of the 12UD Pelletron tandem accelerator facility. In its early stages, the 12 UD Pelletron tandem accelerator was principally used for nuclear physics research. In recent years, the focus of nuclear physics research has shifted to high energy accelerators. The beam time for accelerator mass spectrometry (AMS) research has increased to about 42% of the total operation time. Figure 2 shows a percentage of the experimental beam time for one year on the 12UD Pelletron tandem accelerator. At present, AMS research projects are conducted on more than 50 days per annum. 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 $^{14}$C, $^{26}$Al, $^{36}$Cl and $^{129}$I.

*Work supported by Grants-in-Aid for Scientific Research Programs of the Ministry of Education, Culture, Sports, Science and Technology, Japan.
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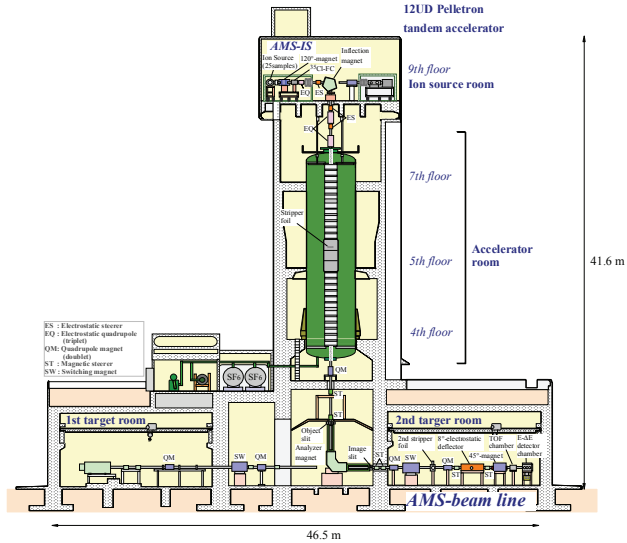


Figure 1: Layout of the 12UD Pelletron tandem accelerator facility at the University of Tsukuba.

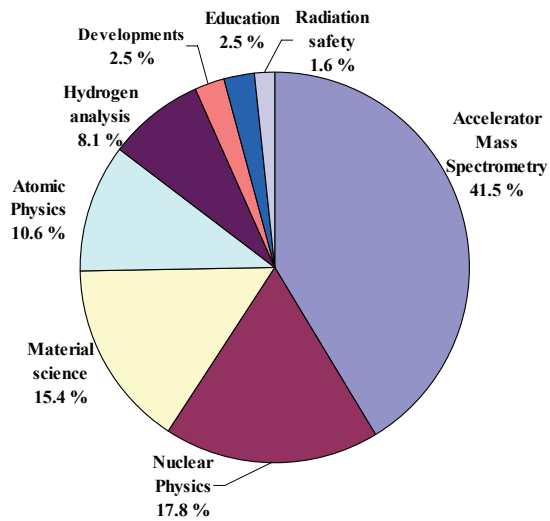


Figure 2: Percentage of the experimental beam time for the 12UD Pelletron tandem accelerator.

## DESCRIPTION OF THE TSUKUBA AMS SYSTEM

Figure 3 shows a schematic diagram of the Tsukuba AMS system. The AMS beam line is equipped with a 25-sample sputter ion source and a 120° mass separator on the low energy side and a spectrometer consisting of a 90°

analyzing magnet (ρ = 1.28 m, ME/$q^2$ = 200 MeV amu, p/Δp = 7.9 × $10^3$), a second stripper foil, a 45° magnetic momentum analyzer, an 8° electrostatic analyzer (ρ= 10 m, E/q = 10 MeV), TOF and a gas ΔE – SSD E detector on the high energy side. The beam currents of the major stable nuclei are measured by offset Faraday cups just behind the 120° mass separator. The terminal voltage of the 12UD Pelletron tandem accelerator is controlled by a slit current feedback system in which the slit current is generated by a pilot beam. This method enables the terminal voltage to be kept stable within ± 0.1% [3, 4]. A 5 μg/$cm^2$ carbon foil with a diameter of 16 mm is used to strip the ions at the terminal. A second stripper carbon foil of 11 μg/$cm^2$ with a diameter of 20 mm is used to eliminate the pilot ions from the mass separator beam line.

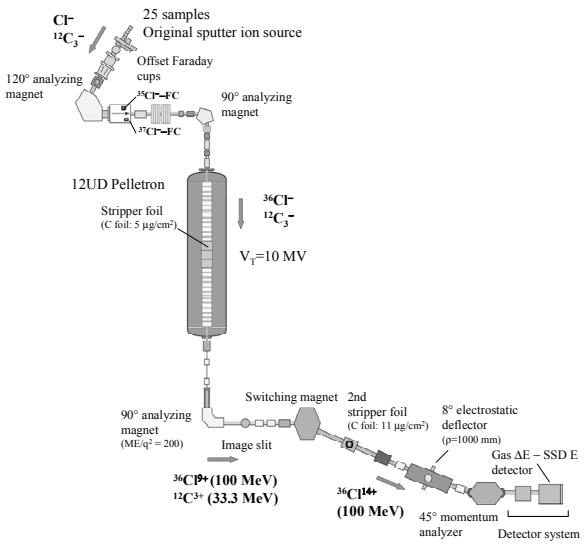


Figure 3: Schematic diagram of the Tsukuba AMS system. Labels indicate the main components along the AMS beam line and the experimental setup for AMS measurements of $^{36}$Cl.

In the case of $^{36}$Cl-AMS, a $^{12}C_3^-$ pilot beam is used for the slit current feedback system. The experimental setup for $^{36}$Cl measurements is also indicated in Figure 1. $^{36}Cl^{9+}$ at 100 MeV and $^{12}C^{3+}$ at 33.3 MeV have the same mass energy product ME/$q^2$ after acceleration with a terminal voltage of 10 MV. The beam current of the $^{12}C^{3+}$ ions measured by the image slit is applied for slit current feedback to stabilize the terminal voltage. After the second stripper carbon foil, $^{36}Cl^{14+}$ and interference ions are selected and detected by the gas ΔE – SSD E detector, which consists of two gas ΔE sections [4]. For our AMS system, a standard sample is measured prior to measuring an unknown sample to reduce the system uncertainties.

## RECENT PROGRESS OF THE TSUKUBA AMS SYSTEM

### *AMS Ion Source*

A dedicated AMS sputter ion source with 25 sample cathodes has been developed at the University of Tsukuba. The AMS ion source is installed on a 100 kV platform and is controlled remotely by a computer through fiber optic links. We converted the Cs ionizer to a spherical type manufactured by NEC. A focused electrode was also installed to focus the Cs beam in front of the target. As a result, we can precisely control the focusing of the Cs beam onto a 1-mm-diameter sample holder. The Cs beam spot is evenly distributed on the target. The maximum beam current from the ion source is 50 μA for $^{35}Cl^-$.

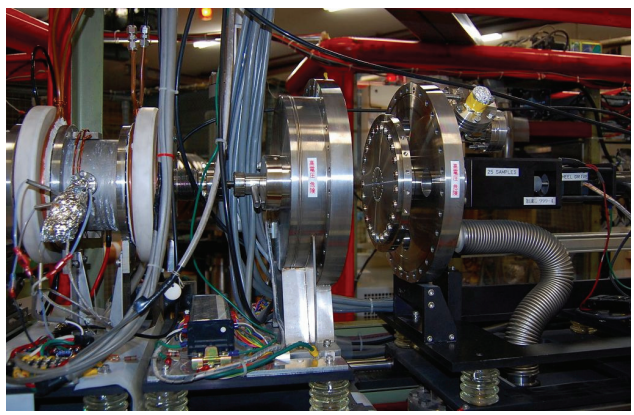

Figure 4: A 25-sample holder of an original Cs sputtering ion source for AMS.

### *AMS Data Integration System*

Major stable beam currents are measured on the low energy side of our AMS system. Two offset Faraday cups are installed behind the 120° mass separator, as shown in Figure 3. A data integration system for AMS was developed that consists of a new beam-current monitoring system and a new data acquisition system (DAQ). The beam-current monitoring system for the offset Faraday cups can record the currents of multiple beams and automatically transfer the current integrated data into a spreadsheet. The DAQ is formed by the NIM ADCs and the CAMAC crate controlled by a FreeBSD operating system. The data integration system is combined with the DAQ in the beam-current monitoring system, which includes start and stop functions for AMS measurements. AMS analysis software has been developed at the University of Tsukuba and it permits AMS spectra to be displayed and analyzed on the same PC control system. Figure 5 shows a display of the data integration system for the Tsukuba AMS system.

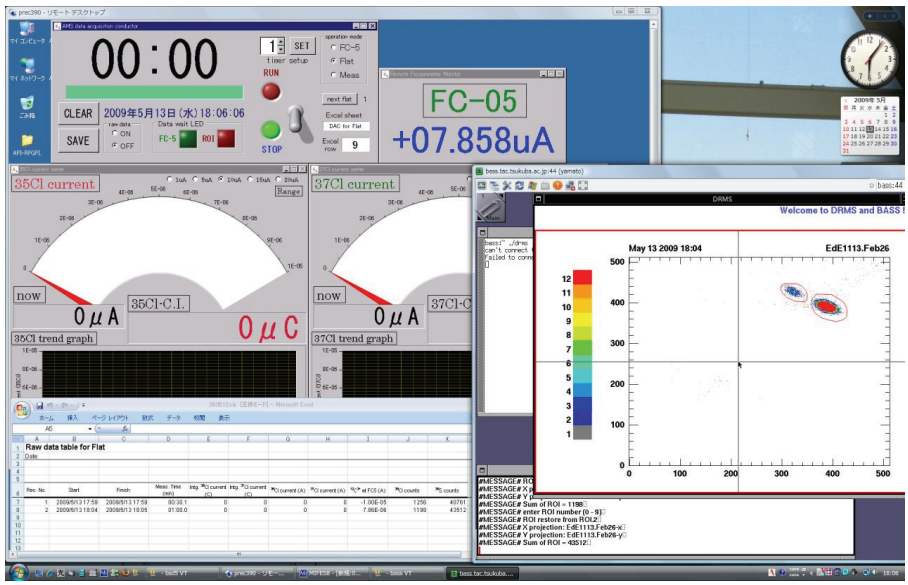


Figure 5: A display on the monitor of the data integration system for the Tsukuba AMS system. Beam current monitor system of the offset-Faraday cups located on the low energy side. The screen displays the diagrammatic representation of AMS spectrum by the PC control system.

### *New Gas ΔE – SSD E Detector*

We developed a new gas ΔE – SSD E detector which consists of two gas ΔE sections and an ion-implanted silicon surface-barrier detector with a 45 × 45 $mm^2$ active area (Hamamatsu Photonics K.K.). The tail of the isobaric interference in the detector was reduced by introducing two ΔE gates. A 4 μm aramid film (Toray Industries, Inc.) is used as the entrance window of the detector. Pure iso-butane gas with a pressure of 670 Pa is applied in the gas section for the AMS measurement. Figure 6 shows a cross sectional view of the new detector.

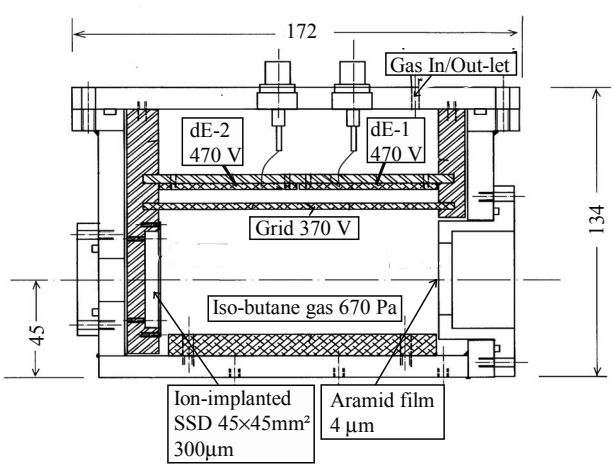


Figure 6: A new gas ΔE – SSD E detector.

## PERFORMANCE OF THE TSUKUBA AMS SYSTEM

Table 1 gives a summary of the AMS performance for multiple nuclides. We have developed the pilot beam method for various nuclides. Over the last three years, we have improved the performance of $^{36}$Cl-AMS in order to measure low $^{36}$Cl concentrations in the Dome Fuji ice core [5].

### *$^{14}$C-AMS*

A terminal voltage of 7 MV is selected and 35 MeV $^{14}C^{4+}$ is accelerated with a $^{7}Li^{2}$ pilot beam. After the second stripper foil, 35 MeV $^{14}C^{5+}$ is detected by the gas ΔE – SSD E detector. The measurement precision was approximately ± 2% [6].

### *$^{26}$Al-AMS*

We use $AlO^-$ molecular ions instead of $Al^-$. 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 by the slit current feedback system. The maximum beam current of $AlO^-$ extracted from $Al_2O_3$ samples was more than 1.5 μA. The beam current of $^{27}Al^{16}O^-$ is measured by an offset Faraday cup simultaneously while measuring $^{26}$Al counted by the gas ΔE – SSD E detector. 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. The pilot beam current of $^{26}Mg^{7+}$ is measured at the image point of 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. The beam transmission of fully stripped $Al^{13+}$ ions from $AlO^-$ is up to 10%. The detection limit for the $^{26}Al/^{27}Al$ ratio is lower than $1 \times 10^{-15}$ [4].

Table 1: Summary of $^{14}C$, $^{26}Al$, $^{36}Cl$ and $^{129}I$ AMS by the Tsukuba AMS System.

| **Trace isotope** | **$^{14}C$** ($T_{1/2}$= 5,730 yr) | **$^{26}Al$** ($T_{1/2}$= 7.20×10$^5$ yr) | **$^{36}Cl$** ($T_{1/2}$= 3.01×10$^5$ yr) | **$^{129}I$** ($T_{1/2}$= 1.57×10$^7$ yr) |
|---|---|---|---|---|
| **Target material** | $C+Li_2O$ | $Al_2O_3 + {}^{26}MgO_2 + Ag$ | $AgCl + C_{60}$ | $AgI+MoO_2+Nb$ |
| **Injection ion** | $^{14}C^-$ | $^{26}AlO^-$ | $^{36}Cl^-$ | $^{129}I^-$ |
| **Pilot beam** | $^{7}Li_2^-$ | $^{26}MgO^-$ | $^{12}C_3^-$ | $^{97}MoO_2^-$ |
| **Reference ion** | $^{12}C^-$ | $^{27}AlO^-$ | $^{35}Cl^-$ & $^{37}Cl^-$ | $^{127}I^-$ |
| **Typical current of reference ion** | 10 μA | 1.5 μA | ~20 μA & 5 μA | 10 μA |
| **Injection energy** | 103 keV | 115 keV | 103 keV | 103 keV |
| **Terminal voltage** | 7 MV | 10.2 MV | 10 MV | 9.68 MV |
| **Particle energy** | 35 MeV ($^{12}C^{4+}$) | 78 MeV ($^{26}Al^{7+}$) | 100 MeV ($^{36}Cl^{9+}$) | 125.8 MeV ($^{129}I^{12+}$) |
| **Detected ion** | $^{12}C^{5+}$ | $^{26}Al^{13+}$ | $^{36}Cl^{14+}$ | $^{129}I^{26+}$ |
| **Background** | $^{14}C/^{12}C < 2 \times 10^{-14}$ | $^{26}Al/^{27}Al < 1 \times 10^{-15}$ | $^{36}Cl/Cl < 1 \times 10^{-15}$ | $^{129}I/^{127}I < 1 \times 10^{-13}$ |
| **Typical precision** | ≤ 2 % | 5 – 10 % | ≤ 2 % | ≤ 8 % |

## *$^{36}$Cl-AMS*

The performance of $^{36}$Cl-AMS was enhanced by improving the AMS technique and system, including modifying the sample preparation technique [7], upgrading the ion source and installing a new data acquisition system. Beam currents of $^{35}Cl^-$ and $^{37}Cl^-$ could be measured by the offset Faraday cups with the beam-current monitoring system (see Figure 5). A typical beam current for $^{35}Cl^-$ was up to 20 μA. $^{36}Cl^{14+}$ with an energy of 100 MeV is detected by the gas ΔE – SSD E detector.

In 2007, we changed the standard reference sample to the KN standards [8] instead of using internal standards. Figure 7 shows $^{36}$Cl spectra for the KN standard of $^{36}Cl/Cl = 1.60 \times 10^{-12}$ and a halite sample from the Himalayas, which was mined from a layer that was several hundreds of million years old, to confirm the system background level. The $^{36}$Cl spectrum of the KN standard sample exhibits complete separation from $^{36}$S in the detector. The AMS system achieved complete discrimination between $^{36}$Cl and $^{36}$S up to a counting rate of ~5 kHz. The background level measured for the halite sample was lower than $1 \times 10^{-15}$ for the $^{36}Cl/Cl$ ratio, as shown in Figure 7(b). The precision for the $^{36}$Cl-AMS system was typically ±2% which was determined from the reproducibility of standard sample measurements.

## *$^{129}$I-AMS*

A $^{97}Mo^{16}O_2$ molecular pilot beam is applied to $^{129}$I–AMS [9]. $^{127}I^-$ ions are measured by the offset Faraday cup simultaneously while measuring $^{129}$I ions in the detector. An electric beam deflector has been installed in front of the offset Faraday cup at the 120º magnet. The purpose of this installation is to increase the separation between the $^{129}I^-$ ion trajectory and the $^{127}I^-$ ion trajectory. We anticipate that the $^{127}I^-$ current can be measured more precisely using this deflector and consequently more

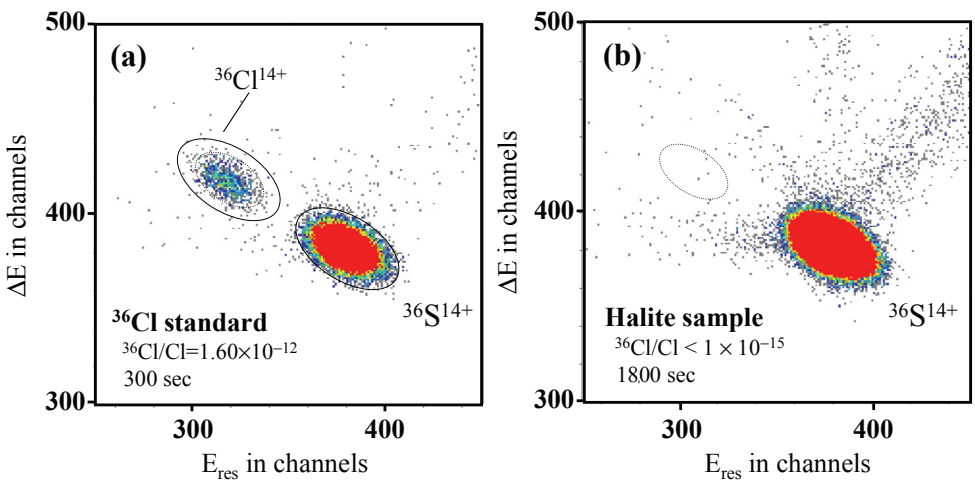


Figure 7: (a) $^{36}$Cl spectrum of a standard sample for $^{36}Cl/Cl = 1.60 \times 10^{-12}$. (b) $^{36}$Cl spectrum of a halite sample for the system background check. Total 3 counts of $^{36}$Cl are detected for 30-minute measurement. The background level is lower than $^{36}Cl/Cl = 1 \times 10^{-15}$.

accurate measurement should be realized. $^{129}I^-$ 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. $^{97}Mo^{9+}$ ions pass through about 5 mm from the image point of the magnet and produce the slit current for the slit current feedback used to stabilize the terminal potential. 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 lower than $1 \times 10^{-13}$.

## SUMMARY

In recent years, the 12 UD Pelletron tandem accelerator is principally used for accelerator mass spectrometry (AMS) research. The Tsukuba AMS system is currently capable of measuring environmental levels of long-lived radioisotopes of $^{14}C$, $^{26}Al$, $^{36}Cl$ and $^{129}I$. Especially for the $^{36}Cl$-AMS, the machine background level of $^{36}Cl/Cl$ has been achieved lower than $1 \times 10^{-15}$.

We replaced the old point-to-plane corona needle system with a resistor-based potential grading system manufactured by NEC in the spring of 2009. The replacement is expected to improve the stability of the terminal voltage. We have developed a GVM control system for the terminal voltage. A second injection beam line has been designed for the AMS system. The 40-sample multiple cathode MC-SNICS manufactured by NEC was installed at the UTTAC in 2008. The new injection beam line with a high current ion source will enhance the ability to efficiently perform routine AMS measurements.

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