Progress of an accelerator mass spectrometry system on the Tsukuba 12UD Pelletron tandem accelerator

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Outline of presentation

• Introduction
  - 12UD Pelletron tandem at the University of Tsukuba
  - AMS and facilities

• AMS system on the 12UD Pelletron tandem
  - Description of the Tsukuba AMS system
  - Recent progress
  - Performance of $^{26}$Al, $^{36}$Cl and $^{129}$I AMS

• Summary and future plans
• Introduction
  - 12UD Pelletron tandem at the University of Tsukuba
  - AMS and facilities
Japan

University of Tsukuba, Tandem Accelerator Complex

Proton Medical Research Center: PMRC

- 250 MeV Proton Synchrotron (2001)

Tandem Accelerator Complex: UTTAC

- 12UD Pelletron Tandem Accelerator (1975)
- 1MV Tandetron Accelerator (1987)

University of Tsukuba, Tsukuba science city

60 km from Tokyo in the northeast

2 accelerator facilities

Proton Beam Radiotherapy
12UD Pelletron tandem accelerator (1975)

- AMS: 42 %
- Nuclear Physics: 17.8 %
- Material science: 15.4 %
- Atomic Physics: 10.6 %
- Hydrogen analysis: 8.1 %
- Education: 2.5 %
- Radiation safety: 1.6 %

3000 hours/yr

Now

- AMS
- IBA
- Material science

University of Tsukuba, Tsukuba, Japan.

- Model: Vertical Tandem Van de Graaff
- Terminal voltage range: 1 - 2 MV
- Insulation Gas: SF₆ pressure: 0.6 MPa
- Accelerator Tank:
  - Height: 17.9 m
  - Diameter: 4.8 m
  - Volume: 350 m³
  - Total weight: 120 ton
- Analyzing Magnet:
  - Magnetic field: 2.2 T
  - Acceleration voltage: 55 - 60 kV
  - Beam current: 67 - 70 nA
- Beam energy: 35 MeV

Nuclear physics
- AMS
- IBA
- Material science

AMS-beam line
Upgrade of the 12UD Pelletron tandem

12UD at Tsukuba University, 1975

2009 Divided resistor system
We replaced the old corona needles with the divided resistor system.

Variable terminal voltage
(No shorting column)

\[ V_t = 1 \sim 12 \text{ MV} \]
Beam energy for the 12UD Pelletron

Terminal voltage: 1 - 12 MV
Accelerator Mass Spectrometry

Targets of AMS

- $^{10}\text{Be}$ ($T_{1/2} = 1.36 \times 10^6$ yr)
- $^{14}\text{C}$ (5730 yr)
- $^{26}\text{Al}$ ($7.1 \times 10^5$ yr)
- $^{36}\text{Cl}$ ($3.0 \times 10^5$ yr)
- $^{129}\text{I}$ ($1.57 \times 10^7$ yr)
- ...

Isobar, Isobaric Molecular suppression

Accelerator

- High energy part
- Momentum analysis
- Mass analysis
- Energy analysis
- Detectors

$E/Q \quad \frac{M}{Q} \frac{E}{Q} = \text{const.}$

Magnetic analyzer

Electrostatic analyzer

$E/Q = \text{const.}$

interested ion

$0 \quad M/Q$
AMS facilities

**14C-AMS**

- 3MV
- 1MV
- 500 kV

Oxford, Groningen, Kiel, Arizona, NOSAMS, Nagoya, SNU, ...

LLNL, KIGAM...

**Multi Nuclides AMS**

- 10Be, 14C, 26Al, 36Cl, 129I, ...
- 5MV and higher (← originally for nuclear physics)

LLNL(10MV), PRIME-Lab(8MV), ANSTO(9MV), ANU(14MV), ETH(6MV), Tokyo(5MV), Tsukuba(12MV), SWERC(5MV)

**3MV**

VERA, LUND, JAEA-Mutsu, ...
• AMS on the 12UD Pelletron tandem
  - Description of the Tsukuba AMS system
  - Recent progress
  - Performance of $^{26}$Al, $^{36}$Cl and $^{129}$I AMS
## Progress of the Tsukuba AMS system

<table>
<thead>
<tr>
<th>Year</th>
<th>Development</th>
</tr>
</thead>
<tbody>
<tr>
<td>1993-96</td>
<td>Trial AMS measurement for $^{14}\text{C}$.</td>
</tr>
<tr>
<td>1996-98</td>
<td>Development of AMS system $^{14}\text{C}$-AMS</td>
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<tr>
<td></td>
<td>AMS ion source (original)</td>
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<tr>
<td></td>
<td>Mass separator beam line</td>
</tr>
<tr>
<td>1999-</td>
<td>Development of $^{26}\text{Al}$, $^{36}\text{Cl}$-AMS</td>
</tr>
<tr>
<td></td>
<td>Pilot beam methods (Instead of GVM control)</td>
</tr>
<tr>
<td>2002-</td>
<td>Development of $^{129}\text{I}$-AMS</td>
</tr>
<tr>
<td>2007-</td>
<td>$^{36}\text{Cl}$ AMS 9 MV $\rightarrow$ 10 MV (Improved beam line)</td>
</tr>
<tr>
<td></td>
<td>Background: $^{36}\text{Cl}/\text{Cl} &lt; 1 \times 10^{-15}$</td>
</tr>
<tr>
<td></td>
<td>Repetition accuracy: $\pm 3%$</td>
</tr>
<tr>
<td>2009-</td>
<td>Upgrade of the 12UD Pelletron (Resister system)</td>
</tr>
<tr>
<td></td>
<td>GVM terminal control system</td>
</tr>
</tbody>
</table>
A pilot beam method is used to stabilize the terminal voltage. The terminal voltage is kept stable within 0.1%.

26Al, 36Cl, 129I - AMS

Pilot beams (isobar)

\[ \text{26Al}^\text{O}^- \rightarrow \text{26Mg}^\text{O}^- \]
\[ \text{36Cl}^- \rightarrow \text{12C}_3^- \]
\[ \text{129I}^- \rightarrow \text{97MoO}_2^- \]

AMS-beam line 500 samples/year.
**36Cl-AMS by the Tsukuba AMS system**

**Target material**: AgCl + C$_{60}$

- **Cl$^-$**: $\sim$ 20 $\mu$A
- **$V_T$**: 10 MV
- **Pilot beam**: $^{12}$C$_3^-$
- **Detection ion**: $^{36}$Cl$^{14+}$
- **Particle energy**: 100 MeV
- **Detection range**: $^{36}$Cl/Cl = $10^{-10}$ ~ $10^{-14}$
- **Back-ground**: $^{36}$Cl/Cl < $10^{-15}$
- **Repetition accuracy**: $\pm$ 3 % (36Cl/Cl $\sim$ $10^{-12}$)

**AMS-beam line**

- $^{36}$Cl$^{9+}$: 100 MeV
- $^{12}$C$_3^+$: 33.3 MeV
AMS Cs sputtering ion source

Sputtering Ion Source [Original]
(25 samples)

AMS-IS

120°-magnet

15 kV
EL
ES

12U D Pelletron tandem accelerator

Inflection magnet
ME/q²=30

1st Stripper foil

Terminal voltage
Vt ≤ 11 MV

ES
EQ
EL

100 kV

Object slit

ST

Analyzer magnet
ME/q²=200

Slit current feedback controller

GVM

FC

Original 25-sample changer

AMS-beam line

EL: Einzel lens
ES: Electrostatic steerer
EQ: Electrostatic quadrupole
FC: Faraday cup
GVM: Generating volt meter
QG: Quadrupole magnet
ST: Magnetic steerer
SW: Switching magnet
TPS: Terminal potential stabilizer

8°-electrostatic deflector

45°-magnet

TOF chamber

E-AE detector chamber
AMS Cs sputtering ion source

**AMS-IS**

- 120°-magnet
- 100 kV
- 15 kV
- EL
- ES
- AMS-beam line
- Automatic sample changer
- Sputtering ion source
- Slit & ES 120°-magnet
- Offset FC
- EQ

**12UD Pelletron tandem accelerator**

- Terminal voltage
  - Vt ≤ 11 MV
- 1st Stripper foil
- Inflection magnet
- ME/q²=30
- Object slit
- Analyzer magnet
- ME/q²=200
- Image slit
- TOF chamber
- 45°-magnet
- E-AE detector chamber
- 8°-electrostatic deflector
- 45°-magnet
- 8°-electrostatic deflector

**AMS-beam line**

- Slit current feedback controller
- GVM
- TPS
AMS Cs sputtering ion source

**AMS-IS**

12UD Pelletron tandem accelerator

Terminal voltage
Vt ≤ 11 MV

AMS-beam line

**AMS-Beam line**

- **EL**: Einzel lens
- **ES**: Electrostatic lens
- **EQ**: Electrostatic quadrupole (triplet)
- **FC**: Faraday cup
- **GVM**: Generating volt meter
- **QM**: Quadrupole magnet (doublet)
- **ST**: Magnetic steerer
- **SW**: Switching magnet
- **TPS**: Terminal potential stabilizer
AMS Cs sputtering ion source

**AMS-IS**

- AMS-beam line
  - Sputtering Ion Source [Original] (25 samples)
  - 120°-magnet
  - 15 kV
  - EL
  - ES
  - 100 kV
  - FC
  - EQ
  - ES
  - Inflection magnet
  - ME/q=200
  - Analyzer magnet
  - Image slit
  - ST
  - 2nd stripper foil
  - QM
  - SW
  - QM
  - ST
  - 45°-magnet
  - TOF chamber
  - E-AE detector chamber
  - 8°-electrostatic deflector

**12UD Pelletron tandem accelerator**

- Terminal voltage
  - Vt ≤ 11 MV

- Slit current feedback controller

**AMS beam line**

- 10.63 μA
- 1886.433 μC
- 3.482 μA
- 618.294 μC

**Inflection magnet**

- 120°

**Sputtering Ion Source [Original]**

- 100 kV

**First Stripper foil**

- 15 kV

**EQ Analyzer magnet**

- 100 kV

**Object slit**

- 120°

**ES**

- Electrostatic steerer

**EQ**

- Electrostatic quadrupole (triplet)

**EL**

- Einzel lens

**FC**

- Faraday cup

**GVM**

- Generating volt meter

**QM**

- Quadrupole magnet (doublet)

**ST**

- Magnetic steerer

**SW**

- Switching magnet

**TPS**

- Terminal potential stabilizer

**AMS Cs sputtering ion source**

- 100 kV

- 15 kV

- Inflection magnet

- 120°

- Magnet
Upgrade of LEBT for 12UD Pelletron tandem accelerator

12UD Pelletron tandem accelerator

Beam transport
Injection energy
\[ ^{36}\text{Cl} : 103 \text{ kV} \]
\[ \text{H}: 120 \text{ kV} \]
Terminal voltage: 10 MV

Electrostatic quadrupole (triplet)

Einzel lens was replaced with electrostatic quadrupole (triplet).

Object point of the Inflection magnet

Electrostatic quadrupole (triplet)

#1-1 tube
Tsukuba 12UD first unit (2009)

Lower resistance at the entrance.

Adjustment for the focusing effect

Ion beam

First unit

4/6 gap short

3/6 gap short

gap full

0.37 G

0.55 G

1.1 G

1/2 gradient

1/3 gradient
Terminal section (Charge exchange)

Terminal section was modified to the large aperture canal (φ20) in 2004.

2 foil units

12UD Pelletron tandem accelerator

Negative ⇒ Positive ions

Carbon stripper foil

ES : Electrostatic steerer
EQ : Electrostatic quadrupole (triplet)
QM: Quadrupole magnet (doublet)
ST : Magnetic steerer
SW : Switching magnet

Negative ions (25 samples) pass through ion source to stripper foil, and are accelerated by 12UD Pelletron tandem accelerator. The ions exit to negative and positive ion chambers after passing through the terminal section. 2 foil units are used to modify the aperture to large, and the ion beam is further accelerated by Pelletron accelerator.
Terminal section (Charge exchange)

Terminal section was modified to the large aperture canal (φ20) in 2004.

2 foil units

Foil unit A (115 foils)
Foil unit B (115 foils)
Focal point
Terminal section (Charge exchange)

Terminal section was modified to the large aperture canal (φ20) in 2004.

Carbon foil for AMS: 5 μg/cm²
Terminal section (Charge exchange)

Terminal section was modified to the large aperture canal (φ20) in 2004.

Carbon foil for AMS: 5 μg/cm²

Foil unit A (115 foils)
Foil unit B (115 foils)

Focal point
Beam transport of the 12UD Pelletron

Object point
12UD accelerator tubes

Electrostatic Q-triplet

Terminal Carbon stripper foil

Acceptance
\(~ 5 \text{ mm mrad}\)

LEBT

Injection energy: 103 kV

V_T: 10 MV

Magnification: 1.5 times on the Terminal stripper.

Beam spot on the stripper foil: \(\phi 6 \text{ mm}\)
Mass separator beam line

- Switching magnet
- 8° electrostatic deflector
- 45° magnet
- TOF
- 2nd stripper foil
- Gas ΔE - E detector
Gas E-ΔE detector

100 MeV $^{36}$Cl in the gas detector.
$^{36}\text{Cl}$-AMS (2-dimensional spectrum)

(a) $^{36}\text{Cl}^{14+}$
36Cl standard
$^{36}\text{Cl}/^{35}\text{Cl}=1.60 \times 10^{-12}$
300 sec
$^{35}\text{Cl}$ current: 1929 μC (6.5 μA)

(b) Halite sample
$^{36}\text{Cl}/^{35}\text{Cl} < 1 \times 10^{-15}$
1800 sec
$^{35}\text{Cl}$ current: 11492 μC (6.5 μA)

$^{36}\text{Cl}/^{35}\text{Cl}$
Background: $\sim 1 \times 10^{-15}$

Standard sample
$^{36}\text{Cl}/^{35}\text{Cl}=1.60 \times 10^{-12}$

Blank sample
\( ^{26}\text{Al-AMS (2-dimensional spectrum)} \)

- Full stripping technique
  - Pilot beam: \( ^{26}\text{MgO}^- \)
  - Beam current of \( \text{AlO}^- \) from \( \text{Al}_2\text{O}_3 \) sample: \( \sim 1.5 \mu\text{A} \)

- \( ^{26}\text{Al is very clearly separated from } ^{26}\text{Mg.} \)
- Background of the \( ^{26}\text{Al-AMS: } < 1 \times 10^{-15}. \)
A pilot beam method is used to stabilize the terminal voltage.

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

<table>
<thead>
<tr>
<th>Target material</th>
<th>$V_T$</th>
<th>Injection ion</th>
<th>Pilot beam</th>
<th>Detection ion</th>
<th>Particle energy</th>
<th>Background</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\text{Al}_2\text{O}_3+^{26}\text{MgO}_2+\text{Ag}$</td>
<td>10.2 MV</td>
<td>$^{26}\text{AlO}^-$</td>
<td>$^{26}\text{MgO}^-$</td>
<td>$^{26}\text{Al}^{13+}$</td>
<td>78 MeV</td>
<td>$&lt; 1 \times 10^{-15}$</td>
</tr>
</tbody>
</table>

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

<table>
<thead>
<tr>
<th>Target material</th>
<th>$V_T$</th>
<th>Injection ion</th>
<th>Pilot beam</th>
<th>Detection ion</th>
<th>Particle energy</th>
<th>Background</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\text{AgCl}+\text{C}_6\text{O}$</td>
<td>10 MV</td>
<td>$^{36}\text{Cl}^-$</td>
<td>$^{12}\text{C}_3^-$</td>
<td>$^{36}\text{Cl}^{14+}$</td>
<td>100 MeV</td>
<td>$&lt; 1 \times 10^{-15}$</td>
</tr>
</tbody>
</table>

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

<table>
<thead>
<tr>
<th>Target material</th>
<th>$V_T$</th>
<th>Injection ion</th>
<th>Pilot beam</th>
<th>Detection ion</th>
<th>Particle energy</th>
<th>Background</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\text{AgI}+\text{MoO}_2+\text{Nb}$</td>
<td>9.7 MV</td>
<td>$^{129}\text{I}^-$</td>
<td>$^{97}\text{MoO}_2^-$</td>
<td>$^{129}\text{I}^{26+}$</td>
<td>126 MeV</td>
<td>$&lt; 1 \times 10^{-13}$</td>
</tr>
</tbody>
</table>
Applications by the Tsukuba AMS system

Mainly for earth and environmental sciences.

<table>
<thead>
<tr>
<th>Nuclear safety research</th>
<th>Soil</th>
<th>Rock</th>
<th>Groundwater, rain, ice</th>
<th>Biological sample</th>
</tr>
</thead>
<tbody>
<tr>
<td>Atomic bomb, neutron fluence</td>
<td>sediment</td>
<td>meteorite</td>
<td>ice</td>
<td>Human hair</td>
</tr>
</tbody>
</table>

- Hiroshima A-bomb sample
- Soil
- Limestone
- Meteorite
- Rain water
- Ice core
- Human hair
Summary and future plans

• **12UD Pelletron tandem at the University of Tsukuba**
  We have upgraded the 12UD Pelletron tandem.
  LEBT, divided resister system, terminal stripper.
  The beam time for AMS research has increased to about 42% of the total operation time.

• **Tsukuba AMS system**
  We are able to measure long-lived radioisotopes of $^{26}$Al, $^{36}$Cl and $^{129}$I by employing a molecular pilot beam method that stabilize the terminal voltage with 0.1% accuracy.
  Main research fields are earth and environmental sciences.

Future plans

• **GVM control system**
• **New injection beam line (MC-SNICS)**
Thank you for your kind attention.
Appendix
- Full stripping technique: $^{26}\text{Al}^{13+}/^{27}\text{AlO}^-$ [counts/μC]
- Pilot beam: $^{26}\text{MgO}^-$
- Beam current of AlO$^-$ from Al$_2$O$_3$ sample: $\sim$1.5μA

The beam transmission of full stripped Al$^{13+}$: 10%. 

 AMS ion source

$^{26}\text{MgO}^-$

$^{26}\text{AlO}^-$

120° mag.

$^{26}\text{Al}^{13+}$

$V_t=10.2\text{MV}$

$^{27}\text{AlO}^-$

$^{26}\text{Mg}^n+$

$^{26}\text{Al}^m+$

$\Delta E-E$ detector

1st Stripper foil

2nd stripper foil (40μg/cm$^2$)

Switching mag.

Deflector (8°)

$^{26}\text{Mg}^n+$ (m$'$~12)

$^{26}\text{Al}^m+$ (n$'$~13)

$^{26}\text{Mg}^7+$

$^{26}\text{Al}^7+$

(77.7 MeV)

Analyzing mag.

Slit current feedback

1st Stripper foil

$^{26}\text{MgO}^-$
AMS facilities in JAPAN

10 AMS facilities (12 accelerators)

Paleo Labo Co., Ltd.
Tandem (500 k V): $^{14}$C

JAEA Tono
Tandem (5MV): $^{14}$C

Kyoto University
Tandem (8MV): $^{14}$C

Kyushu University
Tandem (10MV): $^{14}$C

JAEA Mutsu
Tandetron (3MV): $^{14}$C, $^{129}$I

Institute of Acceleratory Analysis Ltd.
Tandem (3MV): $^{14}$C
Tandem (500kV): $^{14}$C

NIES-TERRA
Tandem (5MV): $^{14}$C

University of Tsukuba
Tandem (12MV): $^{26}$Al, $^{36}$Cl, $^{129}$I

Nagoya University
Tandetron (3MV): $^{14}$C
Tandetron (2.5MV): $^{14}$C

MALT, The University of Tokyo
Tandem (5MV): $^{10}$Be, $^{14}$C, $^{26}$Al, $^{36}$Cl, $^{129}$I
AMS data acquisition and control system
Replicated $^{36}\text{Cl}$ standard measurements

It takes 3 minutes for each measurement. The standard deviation of the fluctuation of the $^{36}\text{Cl}/^{35}\text{Cl}$ ratio is kept within $\pm 3\%$.

Standard sample:
$^{36}\text{Cl}/^{35}\text{Cl} = 5.90 \times 10^{-11}$
$^{36}\text{Cl}$ production

- Primary Cosmic Ray

$^{36}\text{Cl} (T_{1/2}=301 \text{ kyr}): \beta$-decay $^{36}\text{Ar} (98 \%)$

- Natural Cosmic rays
  - $^{40}\text{Ar}(p, n\alpha)\ 36\text{Cl}$
  - $^{36}\text{Ar} (n, p)\ 36\text{Cl}$

- Neutron capture (U, Th)
  - $^{35}\text{Cl} (n, \gamma)\ 36\text{Cl}$

- Human activities
  - $^{35}\text{Cl} (n, \gamma)\ 36\text{Cl} (44 \text{ barn})$
  - Nuclear explosion tests in the sea (1952-1958)
  - Atomic power plant,
  - Nuclear fuel cycle facilities, etc.
Blank sample: $^{36}\text{Cl} / ^{35}\text{Cl} = 2.4 \times 10^{-14}$
5 minutes measurement

$^{36}\text{Cl-AMS}$ (Vt:10MV)
Beam energy: $^{36}\text{Cl}^{14+}$ 100MeV
Background: $^{36}\text{Cl}/^{35}\text{Cl} = \sim 1 \times 10^{-15}$
Detection range: $^{36}\text{Cl}/^{35}\text{Cl} = 10^{-10} \sim 10^{-14}$
Accuracy: $\pm 3\%$ ($^{36}\text{Cl}/^{35}\text{Cl} = 10^{-12}$)

Reproduction in replicated measurements

Standard sample ($^{36}\text{Cl} / ^{35}\text{Cl} = 6.5 \times 10^{-12}$)

$^{36}\text{Cl}$ 32 counts
$^{36}\text{S}$

$\Delta E$

$E - \Delta E$
Charge state distribution of Cl ions

Charge state distribution of $^{35}$Cl and $^{37}$Cl ions after the passage through a carbon foil. [by Dr. Shima et al.]

1st stripper foil

Charge fraction $F(q)$ of $^{35}$Cl$^{9+}$ increased 1.5 times from 20% to 30%.

2nd stripper foil

The optimum thickness of 2nd stripper foil: 12-15μg/cm² [F(14+)]
Sample preparation

<table>
<thead>
<tr>
<th>Sample preparation</th>
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</thead>
<tbody>
<tr>
<td>Sample (10–20 mL)</td>
<td>← 30% H₂O₂ (1 mL)</td>
</tr>
<tr>
<td></td>
<td>← 13 M HNO₃ (1 mL)</td>
</tr>
<tr>
<td></td>
<td>← 0.3 M AgNO₃ (1 mL)</td>
</tr>
<tr>
<td>Centrifugation (2500 rpm, 10 min)</td>
<td></td>
</tr>
<tr>
<td>Supernatant</td>
<td>Precipitate (AgCl)</td>
</tr>
</tbody>
</table>

**Sulfur Reduction (2 times)**

| Filtration (Filter Paper) |  |
| Filtrate ([Ag(NH₃)₂]⁺ Cl⁻) | Precipitate (BaSO₄) |
| ← 3 M NH₄OH (2 mL)       | ← sat. Ba(NO₃)₂ (3 mL) |
| ← 13 M HNO₃ (0.5 mL)     |  |
| Centrifugation (2000 rpm, 15 min) |  |
| Supernatant             | Precipitate (AgCl)  |

- Washing (with 0.01 M HNO₃, 2 times)
- Washing (with 99.5% C₂H₅OH)
- Drying (130°C, 3 h)

Sample: AgCl 1~5mg

A benzene saturated solution of fullerene (C₆₀) for ¹²C₃⁻ pilot beam. [7.6 μl / AgCl: 1 mg]