AMS and its Measurements for Long-lived Heavy and Transactinide Nuclides at CIAE

JIANG Shan, HE Ming, DONG Kejun, WANG Wei, HE Guozhu, SHEN Hongtao, WANG Xianggao, WU Shaoyong, YUAN Jian

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Collaborators

Tsukuba University, Japan
Taipei Central Institute, China
Guangxi University, China
Beijing Normal University, China
China Institute of Atomic Energy

Birth place of China nuclear science and technology
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1. Introduction

Accelerator Mass Spectrometry (AMS) is a kind of Mass Spectrometry which is based on ion accelerator and detector techniques.

AMS has a extremely high sensitivity, it also be called super-sensitive MS.

- abundance sensitivity -- $10^{-15}$
- detection limit — $10^{-18}\text{g/g}$, or $10^4\text{atoms}$
Why AMS has so high sensitivity?

What can AMS do for measurements and applications?
Why AMS has a very high sensitivity

♣ Can eliminate molecular interferences by tandem accelerator stripping.
♣ Has ability to identify and eliminate isobaric interferences by particle detection techniques.
♣ Has a very high efficiency\(10^{-2}\).
The principle of MS and AMS

36Cl measurement with MS
Molecular: $^{35}\text{ClH, }^{18}\text{O}_2, \; ^{19}\text{F}^{17}\text{O}, ^{24}\text{Mg}^{12}\text{C}$
Isobar : 36S
### Sensitivities between AMS and conventional methods

<table>
<thead>
<tr>
<th>Method</th>
<th>Detection Limit (g/g)</th>
<th>Abundance Sensitivity</th>
</tr>
</thead>
<tbody>
<tr>
<td>MS</td>
<td>$10^{-12} (10^{10} \text{ atoms})$</td>
<td>$10^{-8}$</td>
</tr>
<tr>
<td>NAA</td>
<td>$10^{-11} (10^{11} \text{ atoms})$</td>
<td>$10^{-6}$</td>
</tr>
<tr>
<td>AMS</td>
<td>$10^{-18} (10^4 \text{ atoms})$</td>
<td>$10^{-15}$</td>
</tr>
</tbody>
</table>
What can AMS do for measurements and applications?

<table>
<thead>
<tr>
<th>Measurements</th>
<th>Applications</th>
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</thead>
<tbody>
<tr>
<td>Cosmogonies nuclides, $^{10}\text{Be}$,  $^{14}\text{C}$, $^{26}\text{Al}$, $^{36}\text{Cl}$….</td>
<td>Geology, Archaeology, Environment, Biology….</td>
</tr>
<tr>
<td>Nuclides which have extremely low content and radioactivity</td>
<td>Nuclear physics for determination of very long <strong>Half-life</strong>, very small <strong>cross section</strong> and the measurement of <strong>heavy and transactinide nuclides</strong></td>
</tr>
</tbody>
</table>
Principle of determination of half-life and cross section using AMS

\[ T_{1/2} : \]

\[ \frac{dN}{dt} = -N\lambda \]

\( \lambda = \ln 2 / T_{1/2} \)

\( \frac{dN}{dt} \)—decay rate, by using LSS

\( N \)– number of atoms, by using AMS
\[ N_p = N_t \Phi \sigma t \]

- \( N_t \) -- number of target atoms
- \( \sigma \) -- cross section, \( \Phi \) -- beam current
- \( N_p \) -- number of atoms from reaction by AMS
**Half-life determined with AMS**

<table>
<thead>
<tr>
<th>Element</th>
<th>Half-life</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>10Be</td>
<td>$(1.43\pm0.1)\times10^6$ yr</td>
<td>David Fink, Nucl Instr and Meth, 2007</td>
</tr>
<tr>
<td>32Si</td>
<td>$101\pm18$ yr</td>
<td>Kutschera W, Phys Rev Lett, 1980</td>
</tr>
<tr>
<td></td>
<td>$108\pm18$ yr</td>
<td>Elmore D, Phys Rev Lett, 1980</td>
</tr>
<tr>
<td></td>
<td>$133\pm9$ yr</td>
<td>Hofmann H J, Nucl Instr and Meth, 1990</td>
</tr>
<tr>
<td></td>
<td>$162\pm12$ yr</td>
<td>Thomsen M S, Nucl Phys, 1991</td>
</tr>
<tr>
<td>41Ca</td>
<td>$(103\pm7)\times10^3$ yr</td>
<td>Clein K, Earth Plan Sci Lett, 1991</td>
</tr>
<tr>
<td></td>
<td>$(110\pm10)\times10^3$ yr</td>
<td>Kutscher W, Radiocarbon, 1992</td>
</tr>
<tr>
<td>44Ti</td>
<td>$54\pm21$ yr</td>
<td>Frekers D, Phys Rev, 1983</td>
</tr>
<tr>
<td></td>
<td>$67\pm16$ yr</td>
<td>Alburger D E, Phys Rev, 1990</td>
</tr>
<tr>
<td></td>
<td>$59.2\pm0.6$ yr</td>
<td>Ahmad I, Phys Rev Lett, 1998</td>
</tr>
<tr>
<td>60Fe</td>
<td>$(1.5\pm0.3)\times10^6$ yr</td>
<td>Kutscher W, Nucl Instr and Meth, 1984</td>
</tr>
<tr>
<td>79Se</td>
<td>$(280\pm36)\times10^3$ yr</td>
<td>He Ming, Nucl Instr and Meth, 2002(CIAE)</td>
</tr>
<tr>
<td>126Sn</td>
<td>$(207\pm21)\times10^3$ yr</td>
<td>Haas P, Nucl Instr and Meth, 1996</td>
</tr>
</tbody>
</table>
Cross sections determined with AMS

$^{26}\text{Mg}(p,n)^{26}\text{Al}$  

$^{16}\text{O}(n,x)^{14}\text{C}$, $^{36}\text{Ar}(n,p)^{35}\text{Cl}$, $^{16}\text{O}(p,x)^{10}\text{Be}$, $\text{Fe}(p,x)^{26}\text{Al}$, $\text{Ti}(p,x)^{41}\text{Ca}$  

$^{27}\text{Al}(n,2n)^{26}\text{Al}$  
Zhao Qiang, Chin Phys Lett, 1998

$\text{natFe}(p,x)^{53}\text{Mn}$, $\text{natNi}(p,x)^{53}\text{Mn}$, $\text{natNi}(p,x)^{60}\text{Fe}$  
S. Merchel, Nucl Instr and Meth, 2000

$^{40}\text{Ca}(\alpha,\gamma)^{44}\text{Ti}$  
C. Vockenhuber, Nucl Instr and Meth, 2007

$^{25}\text{Mg}(p,\gamma)^{26}\text{Al}$  
Arazi A, Nucl Instr and Meth, 2007

$^{54}\text{Fe}(n,\gamma)^{55}\text{Fe}$  

$^{79}\text{Se}$ (s-process)  

$^{14}\text{N}(16\text{O},\alpha)^{26}\text{Al}$  
Ming He, Nucl Instr and Meth, 2007 (CIAE)

$^{58}\text{Ni}(n,\gamma)^{59}\text{Ni}$, $^{78}\text{Se}(n,\gamma)^{79}\text{Se}$  
G. Rugel, Nucl Instr and Meth, 2007

$^{60}\text{Ni}(n,2n)^{59}\text{Ni}$  
A. Wallner, Intern. conference on nuclear data, 2007

$^{62}\text{Ni}(n,\gamma)^{63}\text{Ni}$  
D. Robertson, Nucl. Instr. and Meth., 2007

$^{40}\text{Ca}(n,\gamma)^{41}\text{Ca}$, $^{54}\text{Fe}(n,\gamma)^{55}\text{Fe}$  
A. Wallner, Nucl Instr and Meth, 2007

$^{209}\text{Bi}(n,\gamma)^{210}\text{Bi}$  
Stan-Sion C, Nucl Instr and Meth, 2007

$^{9}\text{Be}(n,\gamma)^{10}\text{Be}$, $^{13}\text{C}(n,\gamma)^{14}\text{C}$  

$^{62}\text{Ni}(n,\gamma)^{63}\text{Ni}$, $^{64}\text{Ni}(\gamma,n)^{63}\text{Ni}$  
I. Dillman, Nucl Instr and Meth, 2009
Actinide and Superheavy Nuclides

$^{236}\text{U},^{244,242,240}\text{Pu},^{237}\text{Np}$....Superheavy nuclides

ASM labs. at Canada, Switzerland, Israel, U.S, Australia, Germany, Austria, China...
2. AMS measurements

Beijing National Tandem Accelerator Laboratory, CIAE

AMS
AMS < 10%
250 - 300 h

L20 -- AMS
L30 -- General Chamber
L40 -- On Line Y-ray Measurement
L50 -- Radioactive Nuclear Beam
L60 -- Heavy Ion Scanning
R20 -- Q3D
R30 -- VUV Monochromator
R60 -- Fission Chamber
R70 -- Fast Neutron TOF
Tv=13MV, R= 35cm, ME=15, M/ΔM=80

The layout of original CIAE’s AMS system, 13 MV
AMS Measurements in CIAE

<table>
<thead>
<tr>
<th>Element</th>
<th>Target material</th>
<th>Injected ion</th>
<th>Terminal voltage</th>
<th>Analyzed ion</th>
<th>Interference ion</th>
<th>Detector</th>
<th>Backgr. in ratio</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{10}$Be</td>
<td>BeO+Nb</td>
<td>BeO$^-$</td>
<td>8.50 MV</td>
<td>$^{10}$Be$^{+3}$</td>
<td>$^{10}$B$^{+3}$</td>
<td>ΔE-E</td>
<td>$10^{-14}$</td>
</tr>
<tr>
<td>$^{36}$Cl</td>
<td>AgCl+Ag</td>
<td>Cl$^-$</td>
<td>8.30 MV</td>
<td>$^{36}$Cl$^{+8}$</td>
<td>$^{36}$S$^{+8}$</td>
<td>ΔE-E</td>
<td>$10^{-15}$</td>
</tr>
<tr>
<td>$^{26}$Al</td>
<td>Al$_2$O$_3$+Cu</td>
<td>AlO$^-$</td>
<td>8.50 MV</td>
<td>$^{26}$Al$^{+7}$</td>
<td>$^{26}$Mg$^{+7}$</td>
<td>ΔE-Q3D</td>
<td>$10^{-15}$</td>
</tr>
<tr>
<td>$^{41}$Ca</td>
<td>CaF$_2$+Ag</td>
<td>CaF$^-$, CaF$_3^-$</td>
<td>8.70 MV</td>
<td>$^{41}$Ca$^{+8}$</td>
<td>$^{41}$K$^{+8}$</td>
<td>ΔE+Q3D</td>
<td>$10^{-15}$</td>
</tr>
<tr>
<td>$^{79}$Se</td>
<td>SeO$_2$+Ag</td>
<td>SeO$_2^-$</td>
<td>8.05 MV</td>
<td>$^{79}$Se$^{+9}$</td>
<td>$^{79}$Br$^{+9}$</td>
<td>ΔE-E</td>
<td>$10^{-12}$</td>
</tr>
<tr>
<td>$^{129}$I</td>
<td>AgI</td>
<td>I$^-$</td>
<td>8.05 MV</td>
<td>$^{129}$I$^{+11}$</td>
<td>$^{127}$I$^{+11}$</td>
<td>TOF</td>
<td>$10^{-13}$</td>
</tr>
</tbody>
</table>

$^{99}$Tc, $^{182}$Hf, $^{126}$Sn, $^{92}$Nb, $^{236}$U, $^{237}$Np ...are being carried out in CIAE.
AMS Measurements in CIAE

Common interesting -- $^{36}$Cl, $^{26}$Al, $^{10}$Be, $^{41}$Ca……
Fission products – $^{99}$Tc, $^{93}$Zr, $^{79}$Se, $^{151}$Sm……
Heavy nuclides – $^{182}$Hf, $^{236}$U and transactinide nuclides.
Applications

Geology\textsuperscript{36}Cl, \textsuperscript{10}Be, \textsuperscript{26}Al...

Environment\textsuperscript{14}C, \textsuperscript{129}I...

Biology\textsuperscript{14}C, \textsuperscript{79}Se, \textsuperscript{41}Ca for OP

Nuclear science
3. Applications in nuclear science

3.1. Half-life determination for $^{79}\text{Se}$, $^{32}\text{Si}$

3.2. Cross section for $^{93}\text{Nb}(n, 2n)^{92}\text{Nb}$

3.3. Fission products $^{126}\text{Sn},^{151}\text{Sm},^{93}\text{Zr}$ measurements

3.4. $^{236}\text{U}$ and $^{237}\text{Np}$ measurements

3.5. Search for superheavy nuclides in nature
3.4. Cross section for $^{238}\text{U}(n, 3n)^{236}\text{U}$

$^{236}\text{U}$ and $^{237}\text{Np}$ measurements
The determination of $^{238}$U(n,3n)$^{236}$U cross section Needs:

High purified $^{238}$U target ($^{236}$U content less than $10^{-9}$)

High intensity neutron beam current for getting more $^{236}$U

High mass resolution and efficiency in AMS measurement
238U(n,3n)236U Reaction

Neutron source: 600 kV Neutron Generator at (CIAE)
Reaction: T(d, n)⁴He
Deuterium beam current : ~ 0.5mA
Deuterium beam energy Ed: 300KeV
TiT₁.₅ : ~ 1mg/cm²
neutron yield : ~ 3 × 10¹⁰ n.s⁻¹
Irradiation time : 198h

The relative neutron flux was monitored by accompanying α-particles
Primary result for $^{236}$U standard measurement

(a) $^{236}$U dilution standard sample (Sample 2) with a $^{236}$U/$^{238}$U ratio of $(4.6\pm0.4) \times 10^{-8}$.  
(b) Natural uranium sample (Sample 3) with a $^{236}$U/$^{238}$U ratio of $(4.8\pm0.7) \times 10^{-10}$.
Spectrum for $^{236}\text{U}$ measurement

$^{236}\text{U}/^{238}\text{U}=(1.79\pm0.08)\times10^{-9}$, $\sigma=556.7\pm43.4\text{ mb}$
Spectrum for $^{237}\text{Np}$ measurement

$^{237}\text{Np}$ comes from $^{238}\text{U}(n,2n)$ $^{237}\text{U}$, $^{237}\text{Np} / ^{238}\text{U} = (2.08 \pm 0.24) \times 10^{-9}$
3.5. Search for superheavy nuclides in nature
The road for searching of superheavy nuclides in nature with MS and AMS as below. All of the results were negative

(1) 1960’ MS $10^{-7}$.


(3) 1978, Pennsylvania, AMS Pt sample, Z=110, A=294. $1\times10^{-11}$.

(4) 2008, Marinov, et, al.

Evidence for a long-lived superheavy nucleus with atomic mass number A=292 and atomic number Z$\equiv$122 in natural Th.

(5) 2008-2011, AMS re-measured (4) and in Pt,Pb et al in nature by Austria and Germany.

The results were negative under detection limit of $10^{-14}$ ($10^6$ atoms).
Questions are:

How about the result if the detection limit is lower than $10^{-14}$?
How about the result in different places and elements in the world?
Our plan:

- AMS detection limit lower than $10^{-14}$ ($10^{-14} - 10^{-16}$ g/g).
  - by high beam current ion source
  - high mass resolution injector
  - thin stripper foil
- Samples from different places and different elements.
  - such as U, Th, Pt…
CIAE dedicate AMS injector system of high mass resolution, 400
Fig. An injector mass scan spectrum of a HfF$_4$ sample.
A new dedicated 6MV+0.4MV AMS facility at CIAE
Summary

AMS measurements based on HI-13 tandem accelerator in CIAE were developed. Many of interesting nuclides were measured and got very good results.

Its applications especially in nuclear physics were and are being carried out. Such as determination of hafe-life, cross-section and content. It is shown that AMS is an important tool in nuclear science.

In order to develop AMS applications, a new dedicate AMS system was proposed. Many interesting applications especial for transactinide element will be carried out based on the new AMS system.
Thank you !