Irradiation Experiments on $^{237}$Np and $^{241}$Am by Accelerator-Driven System at Kyoto University Critical Assembly

Cheol Ho Pyeon and Masao Yamanaka
Institute for Integrated Radiation and Nuclear Science, Kyoto University
25th September, 2019
Contents

- Objectives

- Accelerator-Driven System (ADS) at Kyoto University Critical Assembly (KUCA)
  - KUCA facility
  - Experimental settings
  - Core configuration
  - Neutron spectrum
  - Experimental results of MA irradiation by ADS
  - Comparative study on Critical and Subcritical cores

- Summary

- Supplement: Joint research of Pb-Bi with JAEA

Objectives

- Conduct feasibility study on MA nuclear transmutation by ADS
- Investigate neutron characteristics through the experiments and the MCNP analyses
# Experimental facilities in the world

<table>
<thead>
<tr>
<th>Project or Facility</th>
<th>Country</th>
<th>Fuel</th>
<th>Reflector or Coolant</th>
<th>Spectrum</th>
<th>Accelerator (target)</th>
<th>Power</th>
<th>Remarks</th>
</tr>
</thead>
<tbody>
<tr>
<td>MUSE</td>
<td>France</td>
<td>MOX</td>
<td>Na</td>
<td>Fast</td>
<td>14 MeV – n</td>
<td>Zero</td>
<td>Finished</td>
</tr>
<tr>
<td>YALINA</td>
<td>Belarus</td>
<td>LEU</td>
<td>Solid metal</td>
<td>Fast &amp; Thermal</td>
<td>14 MeV – n</td>
<td>Zero</td>
<td>Finished</td>
</tr>
<tr>
<td>VENUS-F</td>
<td>Belgium</td>
<td>LEU (MOX)</td>
<td>Pb</td>
<td>Fast</td>
<td>14 MeV – n</td>
<td>Zero</td>
<td>Finished</td>
</tr>
<tr>
<td>KUCA</td>
<td>Japan</td>
<td>HEU</td>
<td>Polyethylene (Graphite, Pb &amp; Pb-Bi)</td>
<td>Thermal</td>
<td>14 MeV – n 100 MeV – p (W, Pb-Bi…)</td>
<td>Zero</td>
<td>Being</td>
</tr>
<tr>
<td>CLEAR-1</td>
<td>China</td>
<td>UO₂</td>
<td>Pb</td>
<td>Fast</td>
<td>14 MeV – n</td>
<td>Zero</td>
<td>Planned</td>
</tr>
<tr>
<td>TEF</td>
<td>Japan</td>
<td>LEU + MA</td>
<td>Pb-Bi</td>
<td>Fast</td>
<td>400 MeV – p (Pb-Bi)</td>
<td>500 Wth</td>
<td>Planned</td>
</tr>
<tr>
<td>MYRRHA</td>
<td>Belgium</td>
<td>MOX + MA</td>
<td>Pb-Bi</td>
<td>Fast</td>
<td>600 MeV – p (Pb-Bi)</td>
<td>100 MWth</td>
<td>Planned</td>
</tr>
</tbody>
</table>

Table: Specification of ADS facilities in the world
### Main parameters in the FFAG accelerator

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td># of sectors</td>
<td>12</td>
</tr>
<tr>
<td>Energy</td>
<td>100 MeV</td>
</tr>
<tr>
<td>Repetition rate</td>
<td>20 Hz</td>
</tr>
<tr>
<td>Average beam current</td>
<td>1 nA</td>
</tr>
<tr>
<td>Width</td>
<td>50 ns</td>
</tr>
<tr>
<td>Field index</td>
<td>7.5</td>
</tr>
<tr>
<td>Closed orbit radius</td>
<td>4.4 - 5.3 m</td>
</tr>
</tbody>
</table>

**Electron LINAC**

11 MeV, 5 mA (Max.)

**Charge convert**

H⁻ -> H⁺

**Main ring (FFAG acc.)**

100 MeV H⁺

**Proton beam current**

1 nA

**Max. power**: 100 W  
**N Yield**: $1 \times 10^9$ 1/s

**KUCA**

**ADS composition in KURRI**

- Electron LINAC
- Charge convert
  - H⁻ -> H⁺
- Main ring (FFAG acc.)
- 100 MeV H⁺
- Proton beam current
  - 1 nA
- Max. power: 100 W
- N Yield: $1 \times 10^9$ 1/s
ADS composition at KUCA

D + T target = 14 MeV neutrons

100 MeV protons + W target = S palliation neutrons

Beam line of D

Beam line of protons

KUCA core

Pulsed neutron generator

W or Pb-Bi target

FFAG accelerator
KUCA core (Solid-moderated core)

- KUCA core -
A solid-moderated and -reflected core

Fig. KUCA core

Fig. Image of KUCA core and fuel assembly loaded
**Fission and Capture reactions**

---

**Ex. ) Fission reactions of MA**

Np-237  
(T<sub>1/2</sub>=214 M year)

Np-237 (T<sub>1/2</sub>=214 M year) → Neutrons → Fission

- Mo-102  
(T<sub>1/2</sub>=21 h)
  - β-ray
- Tc-102  
(T<sub>1/2</sub>=5 s)
  - β-ray
- Ru-102 (stable)

Production of LLFP about 10%

---

**Ex. ) Capture reactions of MA**

Np-237 (T<sub>1/2</sub>=214 M year) → Neutrons → Capture

- Np-238  
(T<sub>1/2</sub>=2 d)
  - β-ray
- Pu-238  
(T<sub>1/2</sub>=88 y)
  - β-ray

Next transmutation by capture or fission reactions

---

**Ex. ) Capture reactions of LLFP**

I-129  
(T<sub>1/2</sub>=15.70 M year)

I-129 (T<sub>1/2</sub>=15.70 M year) → Neutrons → Capture

- I-130  
(T<sub>1/2</sub>=12 h)
  - β-ray
- Xe-130 (stable)

T<sub>1/2</sub>: Half-life

---

Source: Dr. K. Tsujimoto, JAEA
Core configuration (KUCA A core: EE1 core)

Fig. Top view of KUCA EE1 core
BTB chamber and Neutron spectrum

Reaction rate ratio by BTB

- Fission reactions (Np-237 or Am-241/U-235)
  - Test foil: Np-237 (Mass 89 µg, Purity 99.99%)
  - Am-241 (Mass 15 µg, Purity 99.99%)
  - Reference foil: U-235 (Mass 10 µg, Enrich. 99.91 wt%)

- Capture reactions (Np-237/Au-197)
  - Test foil: Np-237 (as same above)
  - Reference foil: Au-197 (Size: 8 mmϕ*50 µmm)

Fig. Spectrum of spallation neutrons (Injection of 100 MeV protons)
Nuclear transmutation of MA by ADS

Core (Subcriticality) and Protons
- Subcriticality: $225 \pm 10$ pcm (PNS method)
  $(215 \pm 9$ pcm; $\alpha$-fitting method)
- Neutron flux: $(1.82 \pm 0.09) \times 10^7$ 1/s/cm$^2$
- Reactor power: $1.35 \pm 0.07$ W
- Irradiation time: 4 hours
- Protons: 100 MeV (Energy)
  0.5 nA (Intensity)
  20 Hz (Frequency)
  100 ns (Beam width)

Fig. Pulsed-height signals of Np-237 and U-235 fission reactions

Fig. $\gamma$-ray spectrum of Np-237 capture reactions

Fig. Pulsed-height signals of Am-241 and U-235 fission reactions

Validity of experimental results (Fission reactions)

Comparison between “Critical and ADS” for variation of reactor power

\[ ^{235}U + \frac{1}{0}n \rightarrow ^{139}_{56}Ba + ^{94}_{36}Kr + 3\frac{1}{0}n \]

\[ ^{237}_{93}Np + \frac{1}{0}n \rightarrow ^{144}_{53}I + ^{102}_{42}Mo + 3\frac{1}{0}n \]

**U-235 and Np-237**

*(High-energy fission)*

Critical irradiation


Kyoto Univ., C. H. Pyeon 11
Comparative study (Critical vs. ADS)

Table Comparison between Critical and ADS (Fission)

<table>
<thead>
<tr>
<th>Reaction rate ratio</th>
<th>ADS</th>
<th>Critical</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{237}\text{Np} / ^{235}\text{U}$</td>
<td>0.048 ± 0.001</td>
<td>-</td>
</tr>
<tr>
<td>$^{241}\text{Am} / ^{235}\text{U}$</td>
<td>0.035 ± 0.003</td>
<td>0.034 ± 0.001</td>
</tr>
</tbody>
</table>

Table Comparison between Critical and ADS (Capture)

<table>
<thead>
<tr>
<th>Reaction rate ratio</th>
<th>ADS</th>
<th>Critical</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{237}\text{Np} / ^{197}\text{Au}$</td>
<td>1.88 ± 0.28</td>
<td>1.88 ± 0.08</td>
</tr>
</tbody>
</table>

- **Source efficiency (Accelerator) : $\mathcal{E}$**

\[ P \sim F = \mathcal{E} S \left(1 + k + k^2 + \cdots \right) = \frac{\mathcal{E} S}{1 - k} \]

- **Current ADS exp. condition**

For $P = 1.35$ W, $\rho = 225$ pcm ($k = 0.99775$), $S (= I) = 0.5$ nA, $\mathcal{E} = \text{"0.61\%"}$

- **Ideal ADS experiments at KUCA**

- Assumption:
  
  $P = 5$ W (Experience at KUCA)
  
  $\rho = 3000$ pcm ($k = 0.97$; Operating mode of actual ADS)

- For $\mathcal{E} = 0.61\%$ and 100 MeV protons,

**Best solution: ($S =$) “24.60 nA” intensity (Impossible)**

(Max. intensity of protons at KUCA is “1nA.”)

Review (at KUCA)

- Implementation of MA irradiation by ADS
- Limitation caused by regulations:
  
  proton beams and MA mass

However,

- Successful demonstration of MA irradiation by ADS


Critical irradiation

Investigation of experimental accuracy

Fig. Spectrum of EE1 core

Fig. Spectrum of U-Pb fuel cell

MCNP analyses (Critical irradiation)

Table Comparison (Fission) between Exp. and MCNP (Np-237/U-235)

<table>
<thead>
<tr>
<th>Region</th>
<th>MCNP</th>
<th>Experiment</th>
<th>C/E</th>
</tr>
</thead>
<tbody>
<tr>
<td>U-Pb</td>
<td>0.067 ± 0.001</td>
<td>0.071 ± 0.004</td>
<td>0.95 ± 0.06</td>
</tr>
</tbody>
</table>

Table Comparison (Fission) between Exp. and MCNP (Am-241/U-235)

<table>
<thead>
<tr>
<th>Region</th>
<th>MCNP</th>
<th>Experiment</th>
<th>C/E</th>
</tr>
</thead>
<tbody>
<tr>
<td>U-Pb</td>
<td>0.079 ± 0.003</td>
<td>0.069 ± 0.001</td>
<td>1.14 ± 0.05</td>
</tr>
<tr>
<td>EE1</td>
<td>0.036 ± 0.001</td>
<td>0.034 ± 0.001</td>
<td>1.08 ± 0.02</td>
</tr>
</tbody>
</table>

Table Comparison (Capture) between exp. And MCNP (Np-237/Au-197)

<table>
<thead>
<tr>
<th>Region</th>
<th>MCNP</th>
<th>Experiment</th>
<th>C/E</th>
</tr>
</thead>
<tbody>
<tr>
<td>U-Pb</td>
<td>2.09 ± 0.10</td>
<td>2.15 ± 0.33</td>
<td>0.97 ± 0.16</td>
</tr>
<tr>
<td>EE1</td>
<td>1.88 ± 0.08</td>
<td>2.02 ± 0.08</td>
<td>0.93 ± 0.05</td>
</tr>
</tbody>
</table>

- **Calculation**
  - MCNP6.1 with ENDF/B-VII.1
  - Total number of history: 1E+08
  - Error (Standard deviation): < 3%

- **Discussion**
  - Experimental error: < 8%
  - Variation of neutron spectrum: U-Pb vs. EE1
  - Fission: Np-237/U-235 -> Good accuracy (5%)
    Am-241/U-235 -> Further investigation
  - Capture: Np-237/Au-197 -> Good accuracy (around 5%)

For numerical analyses of MA irradiation by ADS,

- **Future works**
  - Suitable modeling of spallation source in high-energy regions (over 20 MeV) by PHITS
  - Fine combination of PHITS (Spallation) and MCNP (Reaction rates) codes
Summary

- MA irradiation by ADS at KUCA
  - Demonstration of world’s first nuclear transmutation of MA by ADS

- Comparative study on MA irradiation
  - Comparison between Critical and ADS irradiation of MA
  - Almost same accuracy of measurements

Future works

- Suitable modeling of spallation source in high-energy regions (over 20 MeV)
- Fine combination of PHITS (Spallation) and MCNP (Reaction rates) codes
- MA irradiation at a critical state for spectrum variation in KUCA
Solid Pb-Bi Study
(collaboration with KUCA and JAEA)

Uncertainties of x-secs of Pb and Bi isotopes

- **Motivation**
  - Discrepancy between JENDL-3.3 and JENDL-4.0 of Pb and Bi x-secs through numerical simulations of JAEA ADS model (Pb-Bi coolant model)

- **Experiments at KUCA (critical state)**
  - Sample worth (reactivity) of Pb and Bi plates at a critical state

<table>
<thead>
<tr>
<th>Reactivity (pcm)</th>
<th>JENDL-3.3</th>
<th>JENDL-4.0</th>
<th>ENDF/B-VII.0</th>
<th>JEFF-3.1</th>
</tr>
</thead>
<tbody>
<tr>
<td>94 ± 7</td>
<td>1.63 ± 0.13</td>
<td>1.13 ± 0.10</td>
<td>0.79 ± 0.08</td>
<td>0.89 ± 0.09</td>
</tr>
<tr>
<td>110 ± 6</td>
<td>1.53 ± 0.10</td>
<td>1.07 ± 0.08</td>
<td>0.85 ± 0.07</td>
<td>0.97 ± 0.07</td>
</tr>
<tr>
<td>145 ± 6</td>
<td>1.65 ± 0.08</td>
<td>1.12 ± 0.06</td>
<td>0.94 ± 0.05</td>
<td>1.00 ± 0.05</td>
</tr>
<tr>
<td>156 ± 7</td>
<td>1.76 ± 0.08</td>
<td>1.13 ± 0.06</td>
<td>0.94 ± 0.05</td>
<td>0.98 ± 0.05</td>
</tr>
</tbody>
</table>


- **Further investigation**
  - Uncertainty quantification of Pb and Bi isotopes based the KUCA experiments