Activities of Nuclear Data Measurements using the ANNRI in J-PARC/MLF

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• Introduction
• Our instrument, “ANNRI”
  o J-PARC and MLF
  o Detector system
• Example of Measurements
  o Experiment of Cm-244 and 246 (MAs)
  o Experiment of Zr-93 (LLFP)
  o Experiment of Sn-112 (stable isotope)
• Summary
Introduction(1)

~Motivations of our study~

- To construct new types of reactors,
- To reduce High-Level radioactive Waste (Transmutation),
- To evaluate source terms from the Fukushima reactors,

Neutron Capture Cross Sections of
  - minor actinides (MAs) and
  - long-lived fission products (LLFPs)

are very important.

However, Experimental data of MAs and LLFPs is not sufficient both in quality and in quantity. For example.....
Present status of nuclear data (1)

Experimental results and evaluated neutron capture cross sections of $^{244}$Cm (half-life: 18.1 years) and $^{246}$Cm (4730 years). These are very important MAs.

However, there is only one experimental data set.

This previous measurement was performed using a nuclear explosion in 1969.
There are big differences between evaluated data JENDL 4.0 and ENDF/B-VII. This is because evaluators in JENDL 4.0 adopted Pomerance’s result and evaluators in ENDF/B-VII adopted Nakamura’s result.
Experimental data for MAs and LLFPs are not sufficient both in quality and in quantity. This is because...

- It is not easy to prepare enough amount of MA and LLFP sample with a high purity.
- MAs and LLFPs are radio active nuclides.

To overcome these problems,

1) High Intense pulsed neutron source in J-PARC/MLF
   - Small amount of sample can be used
   - Influence due to decay $\gamma$-rays can be reduced.

2) High energy resolution $\gamma$-ray detector system
   - Background due to impurities can be removed.

were applied to the TOF measurement.

We have constructed new TOF instrument named “ANNRI”.
In our project, we have been measuring these isotopes. Red: Already Published. Blue: Already Measured. Black: Future Plan
In this talk, as examples of our experiments, measurements for

- Cm-244, Cm-246, (MAs)
- Zr-93 (LLFP)
- Sn-112 (Stable)

will be demonstrated.

**Cm-244,246: A. Kimura (JAEA)**

**Zr-93 (preliminary): J. Hori (KURRI)**

**Sn-112 (Very Preliminary results)**
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Our instrument, “ANNRI” ~ J-PARC~

An aerial photograph of J-PARC

- **50-GeV synchrotron**
- **rapid cycle 3-GeV synchrotron**
- **LINAC**
- **MLF**

Protons are accelerated to 3 GeV and injected to a mercury target in the **Materials and Life Science Experimental Facility (MLF)**.
Our instrument, “ANNRI”

top view and side view of the ANNRI.

Main spectrometer which consists of Ge detectors was installed at the flight length of 21.5m.

The neutrons were collimated to 7, 3, or 22 mm in diameter at the sample position.
Our instrument, “ANNRI” ~ Beam Intensity ~

By Dr. Kino

Only under 120kW operation, the neutron intensity is more than 7 times higher than the other instruments. MLF is operated under beam power of 300kW. (Furthermore, In future, the beam power will increase to 1MW.)

Very Strong pulsed neutron source!!


Neutron intensity of ANNRI under 120kW operation comparison to other facilities.

<table>
<thead>
<tr>
<th>energy-integrated intensities</th>
<th>1MW (Future)</th>
<th>120kW</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.5-25 meV</td>
<td>$4.3 \times 10^7$ n/s/cm$^2$</td>
<td>$4.5 \times 10^6$ n/s/cm$^2$</td>
</tr>
<tr>
<td>0.9-1.1 keV</td>
<td>$6.3 \times 10^6$ n/s/cm$^2$</td>
<td>$6.6 \times 10^5$ n/s/cm$^2$</td>
</tr>
</tbody>
</table>
However, neutron energy resolution was not so good!!

- The moderator is large (14cm) and flight length is not so long (21.5m)
- The pulsed protons usually consist of two bunches, each with a width of 100 ns, at intervals of 600 ns (called “double-bunch mode”),

MLF (Materials and Life Science Experimental Facility) has 23 beam-lines, and most beam-lines are diffractometers, scattering spectrometers or reflectometers. So, most users require only “neutron intensity”.
Our instrument, “ANNRI”

~Detector system~

Our spectrometer has
- 2 cluster-Ge detectors
  (7 Ge crystals are installed in the detector)
- 8 coaxial-Ge detectors
  ⇒ 22 Ge Crystals.

Energy resolution for 1.33MeV γ-rays:
- 5.8keV (for 200kevents/s),
- 2.4keV (for 20kevents/s) [1]

Peak efficiency for 1.33MeV γ-rays:
- 3.64 ± 0.11 %

ANNRI was extensively damaged by the earthquake.

The $\gamma$-ray shields made of heavy metals for the large Ge spectrometer leaned to the walls. One of the shields was stopped by breaking an air-conditioner, and the other by a signal cable ruck installed along the walls. At the same time, several of the annular shaped BGO scintillators fell onto the floor, and their PMTs were damaged.
The restoration of ANNRI was almost finished in this March. Our project and user program in ANNRI has restarted.

In this 2012 JFY, ANNRI has been used in user programs for more than 70 days.
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  o Experiment of Sn-112 (stable isotope)

• The Great East Japan Earthquake

• Summary
Experiments of $^{244,246}$Cm

~Samples and Measurement conditions~

Samples:

- **Cm-244 ($T_{1/2}=18.1\,y$: MA)**
  - Net weight = 0.6 mg
  - Activity = 1.8 GBq
  - Measurement Periods: 64 h

- **Cm-246 ($T_{1/2}=4753\,y$: MA)**
  - Net weight = 2.1 mg
  - Activity = 12.1 MBq
    ($^{244}$Cm: 1.7 GBq)
  - Measurement Periods: 94 h

Because of regulation of MLF, the samples are sealed in Al capsuel (278mg).

Table 1 The isotopic composition of the $^{244}$Cm sample or the $^{246}$Cm sample.[1]

<table>
<thead>
<tr>
<th></th>
<th>$^{244}$Cm sample</th>
<th>$^{246}$Cm sample</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>TIMS (mole%)</td>
<td>TIMS (mole%)</td>
</tr>
<tr>
<td>$^{244}$Cm</td>
<td>90.1±1.7</td>
<td>27.5±0.5</td>
</tr>
<tr>
<td>$^{245}$Cm</td>
<td>2.71±0.34</td>
<td>1.06±0.28</td>
</tr>
<tr>
<td>$^{246}$Cm</td>
<td>7.22±0.34</td>
<td>59.4±1.3</td>
</tr>
<tr>
<td>$^{247}$Cm</td>
<td>N.D.</td>
<td>2.9±0.4</td>
</tr>
<tr>
<td>$^{248}$Cm</td>
<td>N.D.</td>
<td>9.10±0.24</td>
</tr>
</tbody>
</table>
Experiments of $^{244,246}$Cm

$\sim$TOF Spectrum $\sim$

This graph shows TOF spectra of the $^{244}$Cm, the $^{246}$Cm sample, and the dummy case. Many resonance peaks were observed.

In more than 100eV region, because of “double-bunch mode”, resonance peaks were formed double peaks.

In Double-Bunch Mode

$600 \text{ ns}$
Experiments of $^{244,246}$Cm

$\sim\gamma$-ray Spectrum at the 1$^{\text{st}}$ resonance of $^{244}$Cm $\sim$

The 252.4- and 380.8-keV $\gamma$-rays have already been studied in $\alpha$ decay of $^{249}$Cf, electron capture decay of $^{245}$Bk, and $\beta$-decay of $^{245}$Am. The other $\gamma$-rays were previously unknown $\gamma$ rays.
Experiments of $^{244,246}$Cm ~γ-ray Spectrum at the 1$^{\text{st}}$ resonance of $^{246}$Cm ~

The all observed γ-rays were previously unknown γ rays.
Experiments of $^{244,246}$Cm

~Analysis~

The data were analyzed with this procedure.

- Dead-Time Correction
- Background Estimation and Subtraction
- Self-Shielding and Multiple-Scattering Correction
- Normalization (Using the 1\textsuperscript{st} resonance of $^{240}$Pu)
- Evaluation and Subtraction of Influence of Fission Events
- Evaluation and Subtraction of Influence of Impurities

The obtained neutron-capture cross sections are....
Experiments of $^{244,246}$Cm

~Cross Section of $^{244}$Cm~

Only one neutron-capture cross-section data of $^{244}$Cm (n,g) was reported in 1969[1].

The results of the resonance peaks under 20-eV are the first experimental results in the world.

The previous measurement was performed using the nuclear explosion “Physics 8” as a pulsed neutron source in 1969.

Only one neutron-capture cross-section data of $^{246}\text{Cm} (n,g)$ was made in 1971[1].

The results of the resonance peaks under 20-eV are the first experimental results in the world.

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Experiments of $^{93}\text{Zr}$

~ Sample and Measurement conditions ~

By Dr. Hori

<table>
<thead>
<tr>
<th>LLFP</th>
<th>Activity (Bq)</th>
<th>Net weight (Total weight)</th>
<th>Measurement Periods</th>
<th>Size (mm)</th>
<th>Manufacture</th>
</tr>
</thead>
<tbody>
<tr>
<td>Zr-93</td>
<td>47 M</td>
<td>470mg (3.31g)</td>
<td>52H</td>
<td>20$\phi \times 5.3$</td>
<td>USA ORNL</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Zr Isotopes</th>
<th>purity[%]</th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Zr-90</td>
<td>5.04 ± 0.06</td>
<td></td>
</tr>
<tr>
<td>Zr-91</td>
<td>12.36 ± 0.04</td>
<td></td>
</tr>
<tr>
<td>Zr-92</td>
<td>15.53 ± 0.03</td>
<td></td>
</tr>
<tr>
<td><strong>Zr-93</strong></td>
<td><strong>18.86 ± 0.04</strong></td>
<td></td>
</tr>
<tr>
<td>Zr-94</td>
<td>22.53 ± 0.02</td>
<td></td>
</tr>
<tr>
<td>Zr-96</td>
<td>25.67 ± 0.09</td>
<td></td>
</tr>
</tbody>
</table>

Isotopic purities were determined with a Thermal Ionization Mass Spectrometer (TIMS) at KURRI. Purity is not enough. Contamination with impurities are very large.
Resonances of $^{93}\text{Zr}$ and stable Zr isotopes were observed.

TOF spectrum of Zr-93

G1

G2

$^{93}\text{Zr} 110\text{eV}$

$^{93}\text{Zr} 225\text{eV}$

$^{91}\text{Zr} 182\text{eV}$

$^{91}\text{Zr} 292\text{eV}$

$^{86}\text{Zr} 301\text{eV}$

$^{91}\text{Zr} 240\text{eV}$

Counts / 100ns

Neutron Energy [ eV ]

G1(Zr–93): 1491–1550ch.(60ch.)
G2(Zr–91,96): 936–990ch.(55ch.)
G3(off resonance): 1391–1490ch.(100ch.)
P. H. Spectra gated by TOF regions

Black: Zr-93, Red: Zr-91 and Zr-96 resonances, Green: Off resonance

Counts/2.5keV
Gamma-ray Energy [ keV ]

Counts/2.5keV
Gamma-ray Energy [ keV ]
We have observed 5 ground-state, 12 primary and 13 cascade transitions for $^{93}$Zr.

Neutron capture cross sections were derived with gating on these 5 ground-state transitions.

<table>
<thead>
<tr>
<th>Initial state (keV)</th>
<th>Final state (keV)</th>
<th>Energy (keV)</th>
<th>Relative intensities</th>
</tr>
</thead>
<tbody>
<tr>
<td>919</td>
<td>0</td>
<td>919</td>
<td>82.9±4.1</td>
</tr>
<tr>
<td>1671</td>
<td>0</td>
<td>1671</td>
<td>9.1±0.5</td>
</tr>
<tr>
<td>2846</td>
<td>0</td>
<td>2846</td>
<td>6.0±1.2</td>
</tr>
<tr>
<td>2908</td>
<td>0</td>
<td>2908</td>
<td>1.0±0.2</td>
</tr>
<tr>
<td>8219</td>
<td>0</td>
<td>8219</td>
<td>1.0±0.2</td>
</tr>
<tr>
<td>Total</td>
<td></td>
<td></td>
<td>100</td>
</tr>
</tbody>
</table>
We have obtained absolute neutron cross sections of $^{93}$Zr by TOF and ground-state transition methods in the energy range from 0.01 eV to 5 keV.

At thermal neutron energy, our results supported Nakamura’s data and evaluated value of ENDF-B/VII. (There is an obvious disagreement with JENDL-4.0.)

We have found a new resonance at 14 eV.
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Experiment of $^{112}$Sn

~Very Preliminary Cross Section ~

Sample: $^{112}$Sn(99.6%) 83.8mg, Beam Power: 210kW, Measurement Periods: 36 hours

Resonance parameters in JEFF 3.1 are the same as those in JENDL 4.0, except for negative resonance.

Resonances at 21, 46, and 151 eV were not observed, (although they are listed in both JENDL 4.0 and ENDF B-VII).

Resonance at 240 eV was observed. This is listed in ENDF B-VII but not listed in JENDL 4.0.
Experiment of $^{112}$Sn

Very Preliminary $\gamma$-ray spectrum

Prompt $\gamma$-ray spectra from $^{112}$Sn resonances are obtained. There are many differences between the resonances.
Experiment of $^{112}$Sn

~Check the origin of the 240-eV resonance~

The 332- and 498-keV γ-rays are clearly observed in all resonances. They have already been studied in other reactions. ($^{110}$Cd(α,nγ), $^{114}$Sn(p,d)...) This result indicates that “the 240-eV resonance is clearly one of the $^{112}$Sn resonances”.

This 240-eV resonance is listed in ENDF B-VII but not listed in JENDL 4.0.
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SUMMARY

~Disadvantages of ANNRI~

- Neutron energy resolution is not good!!
  - The moderator is large (14cm) and flight length is not so long (21.5m).
  - MLF usually operated with “double-bunch mode”. (with a width of 100 ns, at intervals of 600 ns)

- Non-sealed RI and nuclear fuel materials (U, Pu, and Th) cannot be used, because of regulation of MLF.
SUMMARY

~Advantages of ANNRI~

- Strong pulsed neutron with High speed DAQ.
  - A small amount (less than 1 mg) sample can be used.

- High $\gamma$-ray energy resolution with Ge detectors.
  - Prompt $\gamma$-ray spectrum from each resonance are obtained.
  - Using these spectra, an origin of each resonance can be checked.
  - (The target nuclide or impurities)
  - Spin assignments may be checked.

- Rich machine time.
  - In this 2012 JFY, ANNRI has been used in “user program” for more than 80 days.
  - Everybody can apply this program.
  - (We have other 60 days for our research, 30 days for microanalysis and 30 days for maintenance.)
SUMMARY

We have built a new experimental instrument (ANNRI), and obtained preliminary neutron-capture cross sections of MAs and LLFPs.

• Results of these experiments show that neutron-capture cross sections are deduced with a small amount (less than 1 mg) and high radioactive sample.
• From JFY 2011, ANNRI has been started “user program”. Everyone can be used ANNRI using this program.

→ http://is.j-parc.jp/uo/index_e.html J-PARC User office
OR Dr. Harada harada.hideo@jaea.go.jp
Acknowledgment

Thank you for your kind attention!!
Seven nuclei were selected as candidate nuclei for transmutation.

First priority nuclei are Tc-99 and I-129. Other LLFPs are also important in future.
Introduction (4)

Present status of nuclear data (3)

- Se-79, Sn-126
  - No experimental data

![Graphs showing cross-sections for Se-79 and Sn-126 in JENDL-4.0](image)
Experiments of LLFPs

~ Ground-state transition method ~

Ground-state transition method was applied for low purity samples.

**Ground-state transition method**
Cross section was obtained by summing up partial cross section for each ground-state transition.

**Advantage**
- The true capture events can be separated from the background due to impurities by using unique γ rays from target.
- **Lower limit** of capture cross sections or absolute yields of the γ rays can be obtained. (Capture cross sections were obtained by normalizing sum of the yields using relative intensities of the unique γ rays)
Experiments of LLFPs
\(^{107}\text{Pd}\)~

By Dr. Nakamura

- We have obtained neutron cross sections of \(^{107}\text{Pd}\) by TOF and ground-state transition methods in the energy range from 0.1 eV to 300 eV.
- The present results supported Nakamura’s data at thermal neutron capture cross section and JENDL 4.0.
Poisoned Decoupled moderator (PM) for high resolution

Decoupled moderator (DM) for balanced performance

- Adoption of Ag-In-Cd (AIC) alloy for high decoupling energy at 1 eV
- Optimized decouple coverage for lower pulse tail
- Adoption of Cd poison

Proton beam

- Optimized for 100% para-hydrogen (all)
TOF spectrum gated at 919keV gamma-ray peak

Resonances due to Zr stable isotopes

Black line: TOF for all region of P.H.
Red line: Net TOF for 919-keV γ ray

Resonances due to other impurities

Counts / 100ns

Extraction of background components due to impurities was done successfully.
Experimental set up
~ Data acquisition system~

The DAQ were stored
- γ-ray pulse height
- TOF

Time resolution (For TOF):
10 ns (Max 16.7ms)

Maximum event rate:
More than 200k events/s.

For dead-time correction, pulses from the random-pulse Generator were input and measured with the DAQ.

This DAQ is a VME-based.
Each channel has
- analog circuits for pulse shaping,
- a high-speed ADC, and
- a high-power Digital Signal Processor.