Nuclear Reaction Data for Long-Lived Fission Products

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This work was funded by ImPACT Program of Council for Science, Technology and Innovation (Cabinet Office, Government of Japan).
Background for new reaction data

- Nuclear reactions which transmute Long-Lived Fission Products (LLFP) to stable or short lived RI

- Recent world-best accelerators (such as RIBF, J-Parc) in Japan enable us to obtain good nuclear data by using new technology in nuclear science.

- Good simulation software and database of evaluated nuclear data in Japan

Development of new transmutation system
Nuclear reactions for nuclear transmutation by Accelerator

- Nuclear reactions which transmute Long-Lived Fission Product (LLFP: $^{107}$Pd, $^{93}$Zr, $^{79}$Se, $^{135}$Cs, $^{126}$Sn, ($^{129}$I, $^{99}$Tc)) to stable or short lived RI

Candidates
- Neutron induced reaction
  - Neutron capture
  - Neutron knockout
- Negative muon capture reaction
- Fragmentation/Spallation reaction
- Proton/deuteron-induced fusion-like reaction
Nuclear reactions for nuclear transmutation

- Nuclear reactions which transmute Long-Lived Fission Product (LLFP) to stable or short lived RI Candidates
- Neutron induced reaction
  - Neutron capture \((n,\gamma)\) \(A^Z \rightarrow A^{+1}Z\)
  - Neutron knockout \((n,2n)\) \(A^Z \rightarrow A^{-1}Z\)

\[\begin{align*}
  \text{p/n induced reaction} \\
  (p,xn) \\
  (n,2n),(p,\gamma),(\gamma,n) \\
  \text{spallation (n/p,xpyn)} \\
  \end{align*}\]
Neutron capture cross section (Term./Res.)

ANNRI (Accurate Neutron-Nucleus Reaction measurement Instrument)
LLFP targets ($^{135}$Cs, $^{137}$Cs)
Nuclear reactions for nuclear transmutation

- Nuclear reactions which transmute Long-Lived Fission Product (LLFP) to stable or short lived RI Candidates
  - Negative muon capture reaction
    - Populate highly excited state followed by neutron(s) emission
A picture of Muon Nuclear Capture Reactions on $^{107}$Pd Target

(The neutron and $\gamma$ ray emissions are prompt events: DC muon)

Compound nuclear states (10-20 MeV ?)

Muonic atom

$^{107}$Pd($\mu^-$, $\gamma$) $^{107}$Pd

$^{107}$Pd($\mu^-$, 2$n$)$\gamma$

$^{107}$Pd($\mu^-$, 3$n$)$\gamma$

$^{107}$Pd($\mu^-$, 4$n$)$\gamma$

$^{105}$Rh

$^{104}$Rh

$^{103}$Rh

$^{106}$Rh

$^{107}$Rh

$^{107}$Rh

Stable

$\beta^-\gamma$ decay

$\beta^-\gamma$ decay

$\beta^-\gamma$ decay

$\beta^-\gamma$ decay

Delayed $\gamma$-ray spectrum from $\mu^-$-$^{107}$Pd

(The $\beta^-$ decay and the associated $\gamma$ ray emissions are delayed events: pulsed muon.)
RI Beam Factory at RIKEN

3 injectors + cascade of 4 cyclotrons
⇒ several to 345 MeV/nucleon
A variety of primary beams (d(pol) to U)
World highest-intensity RI beams
Experiment in Inverse Kinematics

LLFP: $^{107}\text{Pd}$, $^{93}\text{Zr}^{(+90}\text{Sr})$, $^{135(+137)}\text{Cs}$, $^{79}\text{Se}$ produced as secondary beam

Tagging in event by event and bombard on secondary target @ F8

Reaction Residues measured @ ZD

Secondary Target CD$_2$, CH$_2$, C, Empty Pb,C,Empty

Gas D$_2$, H$_2$, 4atm Empty

Bp+TOF+$\Delta$E +Total Energy

RIBF facility

BigRIPS

ZeroDegree
Fragmentation/Spallation reaction

93Zr@100MeV/u

107Pd@200MeV/u
## Reaction data with LLFPs by RIBF-ImPACT

<table>
<thead>
<tr>
<th>Experiments</th>
<th>Beam lines</th>
<th>Settings</th>
<th>Purpose</th>
<th>Energy [MeV/u]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pre-ImPACT</td>
<td>BigRIPS+ZeroDegree</td>
<td>$^{137}$Cs $^{90}$Sr</td>
<td>Fragmentation/spallation</td>
<td>190</td>
</tr>
<tr>
<td>ImPACT in 2015 spring</td>
<td>BigRIPS+ZeroDegree</td>
<td>$^{107}$Pd $^{93Zr}/^{90}$Sr $^{135}$Cs</td>
<td>Fragmentation/Spallation/Coulomb</td>
<td>100/200</td>
</tr>
<tr>
<td>ImPACT in 2015 autumn</td>
<td>BigRIPS+SAMURAI</td>
<td>$^{93,94}$Zr $^{79,80}$Se</td>
<td>Exclusive measurements</td>
<td>100/200</td>
</tr>
<tr>
<td>ImPACT in 2016 autumn</td>
<td>BigRIPS+ZeroDegree</td>
<td>$^{107}$Pd $^{93}$Zr $^{126,127}$Sn</td>
<td>Spallation/Coulomb</td>
<td>100/200</td>
</tr>
<tr>
<td>ImPACT in 2017 autumn</td>
<td>BigRIPS+OEDO/SHARAQ</td>
<td>$^{107}$Pd $^{93}$Zr $^{79,77}$Se</td>
<td>p/d induced reaction $(d,p)$ for $(n,\gamma)$ surrogate</td>
<td>24/30, 30, 20</td>
</tr>
</tbody>
</table>
Neutron induced reaction

- Neutron capture
  - Direct measurements for thermal to resonance region
  - Surrogate reactions \((d,p), (\gamma'',n)\) for higher energy
- Neutron knockout

Evaluated Data from JENDL4

Eval. From \((p,pxn)\)
RIKEN/UT/…

J-Parc ANNRI
(JAEA)

Coulex (“\(\gamma''\),n)
TITech/RIKEN/…

Surrogate \((d,p)\)
UT/RIKEN/…

Evaluated Data from JENDL4
Coulomb dissociation [\(\gamma, n\) reaction]
(Beam of fission fragments) + (Pb targets)

**Coulex cross sections**

<table>
<thead>
<tr>
<th>Nucleus</th>
<th>present</th>
<th>systematics</th>
<th>Berman</th>
<th>Cb-TDHFB</th>
<th>HFB+QRPA</th>
</tr>
</thead>
<tbody>
<tr>
<td>(^{94}\text{Zr})</td>
<td>403 ± 26 ± 31</td>
<td>388 ± 1</td>
<td>317 ± 1</td>
<td>380</td>
<td>462</td>
</tr>
<tr>
<td>(^{93}\text{Zr})</td>
<td>374 ± 29 ± 30</td>
<td>424 ± 1</td>
<td>-</td>
<td>402</td>
<td>475</td>
</tr>
</tbody>
</table>

Fitting relative energy spectrum assuming response function using TALYS code

\(\sigma_f\): previous result by Berman

**Level density: Back-shifted Fermi Gas model**
Low-energy beam below 50 MeV/u

OEDO Beam-line

Construction was completed in Mar 2017

**OEDO RFD**
- $f_{RF} = 18.25$ MHz
- $V_{max} = 400$ kV
- $\text{Gap(H)} = 200$ mm
- $L \ (Z) = 1200$ mm
- $W(V) = 400$ mm

**OEDO**

**STQ**
- Bore Radius: 120 mm
- Max. gradient: 14.1 T/m
- Combination: 500-800-500 mm
- Total length: 2700 mm
Low-energy nuclear reaction data for LLFP

- Surrogate reactions (d,p)
- Evaluation of (n,xn) from proton/deuteron induced knockout
- Proton/deuteron-induced fusion-like reaction

- New energy-degrading system at RIBF
**Energy compaction by mono-energetic degrader**

- **107Pd, 79Se (BigRIPS)**
- **107Pd, 79Se (SHARAQ)**

**RF HV:** 250kV  
**Phase:** 80 deg.

- **107Pd** 33 MeV/u  
- **79Se** 45 MeV/u

Energy measurement from TOF

Energy compaction by mono-energetic degrader

**Deg FE9**
- Mom. acc. was set to be 0.1% at F1.  
- E was also controlled by D magnets.

**Deg F5**
- 107Pd 33 ± 0.5 MeV/u

**Fig. 11** Energy compaction from 172 MeV/u to 45 MeV/u by a wedged-shape degrader.
- The red (black) histogram shows the energy distribution of the 79Se beam with (without) the FE9 degrader. The thickness and angle of the degrader was set to be 6 mm and 20 mr, respectively.

**Energy loss** from 172.8 ± 3.4 (w/o FE9 deg.) to 46.3 ± 2.7 (w/ FE9 deg.) MeV/u.

- Generally it is hard to tune actual ion transport exactly to the designed one and also a manufacturing of a completely mono-energetic degrader matching to real ion optics is arduous due to an accuracy of energy-loss estimation in the degrader. However, the angle-variable degrader system can be controlled as a mono-energetic degrader with matching to the real ion-optical situation. Therefore, by this degrader system, the ion-optical condition as designed was satisfied against a mismatching of the ion-optical design and the actual setting for the experiment. The performance of this degrader system are detailedly described in Ref. [38].

**4.2. Beam focusing**

The FE11 focusing through the RFD is demonstrated in Fig. 12.

- The secondary beam was set for 75Se at 50 MeV/u, but the angle-tuning of the FE9 degrader was not optimized.

**Deg FE9**
- 79Se 45 ± 1.9 MeV/u

- Mom. acc. was set to be 2% at F1.  
- E controlled by AT degrader system.
Effects of RF Deflector

- $^{79}\text{Se}$ at 170 MeV/u
- $^{107}\text{Pd}$ at 170 MeV/u (BigRIPS production)

Focusing (FE12)

- RF HV: 250 kV
- Phase: 80 deg.

Beam focusing

- $^{79}\text{Se}$ at 45 MeV/u
- $^{107}\text{Pd}$ at 33 MeV/u (FE12)

- TOF (F3-FE10) [ns]
- $X_{\text{FE11}}$ [mm]
- $X_{\text{FE12}}$ [mm]

S0X 1D

- RF ON
- RF OFF

Entries: 14987
Mean: -3.46
RMS: 27
Underflow: 13
Overflow: 15
Integral: 1.484e+04

20 mm (FWHM)
Setup of experiments at low energy

- \(^{107}\text{Pd}, \, ^{79}\text{Se}\) (BigRIPS)
- \(^{107}\text{Pd}, \, ^{79}\text{Se}\) (SHARAQ)

- RF HV: 250kV
- Phase: 80 deg.
- Degrader

- Ag, Pd, Rh, Ru, …
  - \(~25\text{ MeV/u}\)

- PI of the outgoing particles; TOF-dE-Range
  - \(~10\text{ mg/cm}^2\)

- SHARAQ QQE mode
$^{93}\text{Zr},^{107}\text{Pd}$ p/d induced reaction

M. Dozono

$^{107}\text{Pd}$ (Beam)

$^{45}\text{Rh}$

(Z=45)

$^{44,45,46+}$

Ag

(Z=47)

Pd

(Z=46)

107Pd (Beam)

A/Q

Cross section (mbarn)

Energy (MeV/u)
(n,\(\gamma\)) vs. (d,\(p\))

\[ Q_{\text{Se}^80 + \text{Se}^{79}} = 9.91 \text{ MeV} \]

\[ Q_{\text{Se}^80 + \text{d}} = 7.69 \text{ MeV} \]


\[
\sigma_{\text{Se}^{79}(n,\gamma)^{80}\text{Se}}(E_n) = \frac{\sigma_{\text{CN}}^{\text{Se}^{80}}(E_n)}{\text{P}_{\text{decay}}^{\text{Se}^{80} \rightarrow \gamma + \text{Se}^{79}}(E^*)}
\]

determined by the optical model potential

determined by (d,p)

Weisskopf-Ewing approximation
V. Weisskopf, DH. Ewing, Phys. Rev. 57, 472('40)
Experimental Setup at OEDO

Recoil particles: TiNA, SSD-CsI (CNS/RCNP/RIKEN)
reaction products: detectors at final focal plane
target: CD$_2$ 4mg/cm$^2$
Beam int~ $10^4$ pps at on CD$_2$

coincidence measurement of recoil particles + outgoing particles.
Surrogate for $^{79}\text{Se}(n,\gamma)$ w/o measuring $\gamma$ rays

$^{79}\text{Se}(d,p)^{80}\text{Se}$

$^{79}\text{Se}$

$^{80}\text{Se}$

$^{78}\text{Se}^*$

$^{80}\text{Se}^*$

4mg/cm$^2$ CD$_2$

SSD(16chx6)+CsI

coincidence meas. of recoil particles & outgoing particles.

$^{79}\text{Se}(n,\gamma)^{80}\text{Se}$

$^{77}\text{Se}(n,\gamma)^{78}\text{Se}$

$\sigma$ (mb)

$E_n$ (MeV)

Compound by TALYS

ENDF/B-VII.1
Summary

- Nuclear reactions which transmute Long-Lived Fission Product (LLFP) to stable or short lived RI are measured
- Fragmentation/Spallation reactions on p/d were measured at 200, 100, 50 MeV/u @ BigRIPS+ZDS/SAMURAI
- Proton/deuteron-induced fusion-like reaction
- Lower energy LLFP beam is now ready at OEDO beamline
- $^{77,79}$Se(d,p) were measured successfully with a newly developed recoil particle tracker TiNA.