Nuclear reaction study for long-lived fission products in high-level radioactive waste:

Cross section measurements for proton- and deuteron-induced spallation reactions

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Content

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- Experiment details
- Results and discussion on $^{107}\text{Pd}$, $^{137}\text{Cs}$ ($^{136}\text{Xe}$), $^{90}\text{Sr}$
- Summary
Motivation

Spent fuel recycling

LLFP  MA
Motivation

High-level radioactive waste

- Long-lived fission products e.g. $^{107}$Pd, $^{93}$Zr, $^{126}$Sn, $^{137}$Cs...
- Minor Actinide e.g. $^{241,243}$Am, $^{237}$Np...

![Diagram showing LLFP, MA, and Spent fuel recycling]
Motivation

High-level radioactive waste

- Long-lived fission products e.g. $^{107}$Pd, $^{93}$Zr, $^{126}$Sn, $^{137}$Cs...
- Minor Actinide e.g. $^{241,243}$Am, $^{237}$Np...

In Japan, ~800t U / year (~75% of 50 LWR)

1t MA and 39t LLFP in spent fuel
Partitioning and transmutation

Spent fuel recycling

LLFP → MA

MA (Np, Am) → 1t
Platinum (Pt, Rh, Pd) → 4t
Heat generator (Cs, Sr) → 5t
Other FP → 30t

Ref: Lecture by Dr. Oigawa, CNS summer school, 2015
Partitioning and transmutation

Spent fuel recycling → LLFP → MA

- MA (Np, Am) 1t → ADS, FBR
- Platinum (Pt, Rh, Pd) 4t → re-used, except for $^{107}$Pd (6.5x10^6 y)
- Heat generator (Cs, Sr) 5t → Disposal
- Other FP 30t

Ref: Lecture by Dr. Oigawa, CNS summer school, 2015
Accelerator Transmutation System for LLFP

How about accelerator system to reduce radioactivity of LLFP?  
(R. Fujita: WeAM2Pl.2@NN2018)

At reactors,
• transmutation reactions are limited to neutron-induced reactions at energy of thermal to MeV
• LLFP is further produced

At accelerator system,
• A variety of reactions can be applied  
  e.g. Spallation reaction, fusion, muon capture, \((n,2n)\) ...

Lack of nuclear reaction data for LLFP (so far, \(n\)-capture only)
### A challenge at RIKEN

<table>
<thead>
<tr>
<th>Year</th>
<th>Energy [MeV/u]</th>
<th>LLFP</th>
<th>Purpose</th>
</tr>
</thead>
<tbody>
<tr>
<td>2014</td>
<td>190</td>
<td>$^{137}\text{Cs}/^{136}\text{Xe}$, $^{90}\text{Sr}$</td>
<td>Spallation</td>
</tr>
<tr>
<td>2015</td>
<td>100/200</td>
<td>$^{107}\text{Pd}$, $^{93}\text{Zr}$, $^{90}\text{Sr}$, $^{135}\text{Cs}$, $^{93,94}\text{Zr}$, $^{79,80}\text{Se}$</td>
<td>Spallation/Coulomb breakup</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>2016</td>
<td>50</td>
<td>$^{107}\text{Pd}$, $^{93}\text{Zr}$</td>
<td>Spallation</td>
</tr>
<tr>
<td></td>
<td>100/200</td>
<td>$^{126,127}\text{Sn}$</td>
<td>Spallation/Coulomb breakup</td>
</tr>
<tr>
<td>2017</td>
<td>30</td>
<td>$^{107}\text{Pd}$, $^{93}\text{Zr}$, $^{79}\text{Se}$</td>
<td>Fusion-like</td>
</tr>
</tbody>
</table>

- RI Beam Factory provides a unique opportunity to get reaction data.
- Half-life distributions of fragments from production cross section.
Inverse Kinematics Method

- Proton and deuteron target
- Energy dependence
- Direct measurement on product

LLFP beams

Reaction products
- neutron
- gamma

$^2\text{H} (n \text{ and } p)$
$^1\text{H} (p)$
Experimental setup

- FP ($^{107}$Pd/$^{137}$Cs/$^{90}$Sr) beams production
- Identification for products $B\rho$, $\Delta E$, TOF

BigRIPS

- Production target
- Secondary target $CD_2$, $CH_2$, $C$

ZeroDegree spectrometer
Experimental setup

**BigRIPS**

**ZeroDegree spectrometer**

**Production target**

$^{238}\text{U}$

**Tagging**

**Target**

**Secondary target**

$\text{CD}_2$, $\text{CH}_2$, $\text{C}$

- $^{90}\text{Sr}/^{137}\text{Cs}/^{107}\text{Pd}$ beams production
- Identification for products $B\rho$, $\Delta E$, TOF, Total kinetic energy

**Diagram:**

- Beam Production
- BigRIPS
- Tagging
- Target
- $\text{CD}_2$
- Products PID
- Secondary target $\text{CD}_2$, $\text{CH}_2$, $\text{C}$
PID example: $^{107}\text{Pd}@100\text{MeV/u}$

**Projectile PID@BigRIPS**

- Atomic number $Z$
- Mass-to-charge ratio $A/Q$

**Products @ZeroDegree**

- Atomic number $Z$
- Mass-to-charge ratio $A/Q$

- $^{102}\text{Pd}$

**Beam Production**

**BigRIPS**

**Tagging**

**ZeroDegree spectrometer**

**Target**

$\text{CD}_2$
Cross section for product

\[ \sigma = \frac{N_{\text{products}}}{N_{\text{beam}} \times n_{\text{target}}} \]
$^{107}$Pd results @ 100 MeV/u

- $\sigma_p > \sigma_d$ for $\Delta Z = +1$
- $\sigma_p \sim \sigma_d$ for Pd, Rh
- $\sigma_p < \sigma_d$ for light-mass ions
- Total cross section $\sim 1$ barn

Cross section: Energy dependence

- **Ru (Z=44)**
- **Tc (Z=43)**
- **Mo (Z=42)**

- **$p@200$ MeV/u**
- **$p@100$ MeV/u**

Cross section: Energy dependence

\[ \sigma_d \text{ at } 100 \text{ MeV/u is similar to } \sigma_p \text{ at } 200 \text{ MeV/u} \]

\[ \sigma_d \text{ at } 100 \text{ MeV/u is similar to } \sigma_p \text{ at } 200 \text{ MeV/u} \]

\[ \sigma_d \text{ at } 50 \text{ MeV/u is similar to } \sigma_p \text{ at } 100 \text{ MeV/u} \]
Intra-nucleon cascade and evaporation by PHITS

• INCL4.6 + GEM

Cross section: LLFP and neighboring stable nucleus

\[ 137\text{Cs} \]

**Figure:**

- Panel a): Ba (Z=56)
- Panel b): Cs (Z=55)
- Panel c): Xe (Z=54)
- Panel d): I (Z=53)
- Panel e): Te (Z=52)
- Panel f): Sb (Z=51)

The figure shows the cross section for different elements as a function of neutron number (N) and proton number (Z). The data points are indicated by black circles for protons (p) and red squares for deuterons (d).

**Legend:**

- Stable
- Short-lived
- Long-lived

**References:**

Cross section: LLFP and neighboring stable nucleus

- $^1_1$H ($p$)
- $^2_1$H ($d$)

$^{137}\text{Cs}$

$^{136}\text{Xe}$

Cross section [mb]

Proton number $Z$

Neutron number $N$

Stable

Short-lived

Long-lived

[Graph showing cross sections for various elements and isotopes, including Ba (Z=56), Cs (Z=55), Xe (Z=54), I (Z=53), Te (Z=52), and Sb (Z=51), with data points and error bars for different isotopes, such as $^{136}\text{Ba}$, $^{137}\text{Ba}$, $^{138}\text{Ba}$, $^{135}\text{Cs}$, $^{136}\text{Cs}$, $^{137}\text{Cs}$, $^{134}\text{Xe}$, $^{135}\text{Xe}$, and $^{136}\text{Xe}$]
Potential of spallation for LLFP transmutation

- $d$-induced spallation reaction
- $p$-induced spallation reaction
- $(n_{th}, \gamma)$

Total cross section [mb]

- $^{90}\text{Sr}$
- $^{137}\text{Cs}$
Total cross section for $^{107}\text{Pd}$

<table>
<thead>
<tr>
<th></th>
<th>Cross section [barn]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Spallation</td>
<td>1</td>
</tr>
<tr>
<td>(n,\gamma)</td>
<td>9.2</td>
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</tbody>
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Potential of spallation for LLFP transmutation I

Total cross section for $^{107}$Pd

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<td>$(n,\gamma)$</td>
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Transmutation rate depends on
Cross section and Flux
Potential of spallation for LLFP transmutation II

Half-life distributions of products

$^{107}\text{Pd} + p$ 200 MeV/u

- Stable: 64.8%
- $>30000\text{y}$: 6.41%
- 1000–30000y: 1.79%
- 30–1000y: 1.03%
- 30y–1y: 7.88%
- <1y: 18.0%
Potential of spallation for LLFP transmutation II

$^{107}\text{Pd} + p$ 100MeV/u

- Stable: 63.2%
- $>30000y$: 2.03%
- 30y~1y: 9.45%
- $<1y$: 25.0%

$^{107}\text{Pd} + p$ 200MeV/u

- Stable: 64.8%
- $>30000y$: 6.41%
- 1000~30000y: 1.79%
- 30~1000y: 1.03%
- 30y~1y: 7.88%
- $<1y$: 18.0%
Potential of spallation for LLFP transmutation II

$^{107}\text{Pd} + p$ 50MeV/u

- Stable: 66.2%
- $<1\text{y}$: 31.1%
- 30y~1y: 2.74%

$^{107}\text{Pd} + p$ 100MeV/u

- Stable: 63.2%
- $<1\text{y}$: 25.0%
- 30y~1y: 9.45%
- $>30000\text{y}$: 2.03%

$^{107}\text{Pd} + p$ 200MeV/u

- Stable: 64.8%
- $<1\text{y}$: 18.0%
- 30y~1y: 7.88%
- 1000~30000y: 1.79%
- 30~1000y: 1.03%
Summary

• Spallation reactions for LLFP nuclei using inverse kinematics at RIBF
  Target dependence
  Energy dependence

• Comparison with spallation models

• Potential for the transmutation on LLFP
  Total reaction cross section
  Production of other radioactive isotopes at different reaction energies

• Collaboration with nuclear engineering
Collaborators

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Reduction and Resource Recycling of High-level Radioactive Wastes through Nuclear Transmutation

http://www.jst.go.jp/impact/en/program/08.html

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Thank you