Nuclear reaction study for high-level radioactive waste:

Cross section measurements for proton- and deuteron-induced spallation reactions of long-lived fission products

He Wang

RIKEN Nishina Center

15th Varenna conference on NRM, Varenna, Italy, June 11 - 15, 2018
Content

• Motivation

• Experiment details

• Results and discussion on $^{137}\text{Cs}$, $^{90}\text{Sr}$, $^{107}\text{Pd}$

• Summary
Motivation

High-level radioactive waste

- Long-lived fission products e.g. $^{137}$Cs, $^{90}$Sr, $^{107}$Pd, $^{93}$Zr...
- 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
Further Reprocessing on LLFP nuclei

How about accelerator system to reduce radioactivity of LLFP?

Lack of nuclear reaction data for LLFP (so far, n-capture only)

Ref: Lecture by Dr. Oigawa, CNS summer school, 2015
### 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>$^{107}\text{Pd}, {}^{93}\text{Zr}, {}^{90}\text{Sr}, {}^{135}\text{Cs}$</td>
<td>Spallation/Coulomb breakup</td>
</tr>
<tr>
<td></td>
<td></td>
<td>$^{93,94}\text{Zr}, {}^{79,80}\text{Se}$</td>
<td>Spallation/Exclusive cross section</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>Low energy</td>
</tr>
</tbody>
</table>

- Half-life distributions of fragments from production cross section
- RIBF provides a unique opportunity to get reaction data
Nuclear Reaction study on LLFP at Varennna

- $^{137}$Cs, $^{90}$Sr
  First targets for study spallation
- $^{107}$Pd
  Recovery of palladium metal
- $^{93}$Zr by Y. Watanabe on June 15th
- $^{136}$Xe by X.H. Sun on June 15th

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>Half-life [year]</th>
<th>$(n, \gamma)$ [b]</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{137}$Cs</td>
<td>30.1</td>
<td>0.27</td>
</tr>
<tr>
<td>$^{90}$Sr</td>
<td>28.8</td>
<td>0.01</td>
</tr>
<tr>
<td>$^{107}$Pd</td>
<td>6.5x10^6</td>
<td>9.2</td>
</tr>
</tbody>
</table>
Nuclear Reaction Study via Inverse Kinematics Method

Inverse kinematics

- Proton and deuteron target
- Energy dependence

LLFP beams

Target

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

$^1\text{H} (p)$

Reaction products

neutron

gamma
BigRIPS/ZeroDegree

RI Beam Factory

Super-heavy Element Science

113\textsuperscript{th} Nh “Nihonium”

RI production

Materials
Biology

RILACII (construction)

28GHzECRIS (construction)

SAMURAI (construction)

e-RI scattering with SCRIT (construction)

SAMURAI (construction)

SLOWRI (R&D)

ZDS

Rare RI ring (R&D)

SHARAQ (construction)

BigRIPS/ZeroDegree

RI production

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Biology

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e-RI scattering with SCRIT (construction)

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SLOWRI (R&D)

ZDS

Rare RI ring (R&D)

SHARAQ (construction)

BigRIPS/ZeroDegree
RI Beam Factory

World’s strongest beam intensity
Experimental setup

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

**Production target** $^{238}\text{U}$

**Target** $^{238}\text{U}$, $^{239}\text{Pu}$, $^{240}\text{Pu}$, $^{241}\text{Pu}$

**Secondary target** $\text{CD}_2$, $\text{CH}_2$, $\text{C}$

**BigRIPS**

**ZeroDegree spectrometer**
Experimental setup

- **BigRIPS**
- **ZeroDegree spectrometer**

**Production target**
- \(^{238}\text{U}\)

**Tagging**
- Target
- Secondary target: \(\text{CD}_2, \text{CH}_2, \text{C}\)

- FP (\(^{90}\text{Sr}/^{137}\text{Cs}/^{107}\text{Pd}\)) beams production
- Identification for products: \(B_\rho, \Delta E, \text{TOF}, \text{Total kinetic energy}\)
The first challenge: $^{137}\text{Cs}$
Isotopic distribution cross section

\[ \sigma = \frac{N_{\text{products}}}{N_{\text{beam}} \times n_{\text{target}}} \]
Products from $^{137}$Cs

Intra-nucleon cascade and evaporation by PHITS

\[ Z = 56 \quad Z = 55 \quad Z = 54 \quad Z = 53 \quad Z = 52 \quad Z = 51 \]

\( d \quad p \)

Products from $^{90}$Sr

Comparison with PHITS

Energy dependence for light-mass ions (\(^{107}\)Pd)

\[ \sigma_d \text{ at 118 MeV/u is similar to } \sigma_p \text{ at 196 MeV/u} \]

Potential of spallation for LLFP transmutation

- **$d$-induced spallation reaction**
  - Total cross section: 1300 mb
  - $^{90}$Sr: 998 mb
  - $^{137}$Cs: 1100 mb

- **$p$-induced spallation reaction**
  - Total cross section: 785 mb
  - $^{90}$Sr: 420 mb
  - $^{137}$Cs: 365 mb

- $(n_{th}, \gamma)$
  - Total cross section: 270 mb
  - $^{90}$Sr: 10 mb
  - $^{137}$Cs: 260 mb
Total cross section for $^{107}$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,γ)</td>
<td>9.2</td>
</tr>
</tbody>
</table>

Transmutation rate depends on Cross section and Flux
Summary

• Spallation reactions for LLFP nuclei using inverse kinematics at RIBF

• Cross sections on $p$ and $d$ for $^{137}$Cs, $^{90}$Sr and $^{107}$Pd
  Target dependence
  Energy dependence

• Comparison with spallation models

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

• Collaboration with nuclear engineering
Collaborators

RIKEN Nishina Center

Kyushu University
Y. Watanabe, S. Kawase, K. Nakano, S. Araki, T. Kin

Tokyo Institute of Technology

Miyazaki University
Y. Maeda, S. Kawakami, T. Yamamoto

University of Tokyo

CNS, Univ. of Tokyo
S. Michimasa, M. Matsushita, S. Shimoura

Hokkaido University
A. Makinaga, M. Aikawa
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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Nucleus-Nucleus Collisions 2018

Omiya, Japan (not far from Tokyo)
4-8, December 2018

Hosted by RIKEN Nishina Center
Supported by IUPAP

You can get full information at
http://nn2018.riken.jp
International Conference on

Nucleus-Nucleus Collisions

- Organized every 3 years since the first meeting in 1982 at MSU
- 11th at Texas (USA), 12th at Catania (Italy), 13th at Omiya (Japan)
- 350+ participants in the 12th
- 350—400 participants in NN2018

Nucleosynthesis

- Fusion & fission
- Superheavy elements
- Unstable nuclei
- EoS and strangeness
- Quark-gluon plasma

Low to Ultra high Energy

+ Applications (Cancer therapy, nuclear transmutation, RI for other fields... )
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Abstract submission is now open!
New deadline: June 30th
Banquet (in the evening of Dec. 7)

Paradise for railway geeks
Thank you

Looking forward to seeing you in NN2018