Observation of Nuclear Transmutation
Reactions induced by D₂ Gas Permeation
through Pd Complexes

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Features of the Present Method

D$_2$ gas permeation through the Pd complex

Cross Section of Pd Complex

- Pd substrate
- CaO 2nm
- Pd 40nm
- vacuum
- D$_2$ gas
- D Permeation
- Cs, Ba, etc.
**Fabrication of Pd Complex**

1. **Washing a Palladium Sample with Acetone**
2. **900 °C 10H Annealing under Vacuum Condition (< 10⁻⁷ Torr)**
3. **Washing the Sample with Aqua Regia (100sec)**
4. **5 times Alternatingly Sputtering of CaO(20 Å) and Pd(180 Å)**
5. **Ion Beam Sputtering of Pd only (400 Å)**

Additional Details:
- **CaO/Pd 1000 Å**
- **Pd 400 Å**
- **Pd 0.1mm**
- **25mm × 25mm**
Schematic View of the Experimental Apparatus

- X-ray Gun
- Photoelectron Energy Analyzer
- Pd Complex Test Piece
- Chamber A
- Evacuation
- Chamber B
- Ge Detector
- D2 Gas
- Evacuation
- XPS
- Q-Mass
Photograph of the Experimental Setup

- Photoelectron Energy Analyzer
- X-ray Gun
- D₂ Inlet
- Ge Detector
- Main Chamber
Decrease of Cs and Emergence of Pr
Identification of Pr by TOF-SIMS

D$_2$ Gas Permeation

TOF-SIMS device (TRIFT™ II; ULVAC-PHI)
Decrease of Sr and Emergence of Mo

Number of atoms ($10^{14}/\text{cm}^2$)

Time (h)

- Sr 1st
- Mo 1st
- Sr 2nd
- Mo 2nd
- Sr 3rd
- Mo 3rd
Relation of Isotopic Composition between Sr and Mo

![Graph showing isotopic composition of Sr and Mo](image)

- **Given Sr**
  - Mass number: 84, 86, 87, 88
  - Isotope ratio (%): 0, 20, 40, 60, 80, 100

- **Detected Mo**
  - Mass number: 92, 94, 95, 96, 97, 98, 100
  - Isotope ratio (%): 0, 20, 40, 60, 80, 100

- **Mass N. +8**
  - $^{88}_{38} Sr \rightarrow ^{96}_{42} Mo$
  - Atomic N. +4

- **Mass N. +8**
  - $^{133}_{55} Cs \rightarrow ^{141}_{59} Pr$
  - Atomic N. +4
Recent Results; Part 1

Transmutation of $^{138}\text{Ba}$ into $^{150}\text{Sm}$ and $^{137}\text{Ba}$ into $^{149}\text{Sm}$
Transmutation of Ba into Sm;
Natural Ba

Natural Ba ($^{138}\text{Ba}$ major)

D

Pd
CaO

$^{150}\text{Sm}$ was detected after D permeation on the Pd complex
Schematic View of the Ex-situ Measurement Apparatus

Pd Complex with an Element (Cs, Sr, Ba, etc)

D2 Gas

Chamber A

Chamber B

Ex-situ Measurement;
XPS, SIMS, ICP-MS, etc.
XPS Spectra for detected Sm

Counts

Counts(Ba1st)

Counts(Ba2nd)

Sm 3d $\frac{5}{2}$

Sm 3d $\frac{3}{2}$

Binding Energy (eV)
Full Spectrum

Counts

Binding Energy (eV)

Natural Ba
$^{138}$Ba major

Pd

Ba

Sm 3d

Sm 4d

C

D

Pd

CaO
Sm Natural Abundance

<table>
<thead>
<tr>
<th>Mass Number</th>
<th>144 Sm</th>
<th>147 Sm</th>
<th>148 Sm</th>
<th>149 Sm</th>
<th>150 Sm</th>
<th>152 Sm</th>
<th>154 Sm</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>3.2%</td>
<td>15.1%</td>
<td>11.3%</td>
<td>13.8%</td>
<td>7.5%</td>
<td>26.6%</td>
<td>22.5%</td>
</tr>
</tbody>
</table>

Ratio to $^{152}\text{Sm}$

Sm Natural Abundance

Mass Number: 144, 145, 146, 147, 148, 149, 150, 151, 152, 153, 154
SIMS Spectra for Given and Detected Elements

Sm Natural Abundance

138Ba

150Sm

138BaO

Pd\(^{40}\)Ca

BaO

Counts

Mass Number
### Examination of Molecular Ions

<table>
<thead>
<tr>
<th>Pd</th>
<th>Pd$^{40}$Ca</th>
<th>Ba</th>
<th>Ba$^{16}$O</th>
</tr>
</thead>
<tbody>
<tr>
<td>102(1%)</td>
<td>142</td>
<td>130(0.1%)</td>
<td>146</td>
</tr>
<tr>
<td>104 (11%)</td>
<td>144</td>
<td>132(0.1%)</td>
<td>148</td>
</tr>
<tr>
<td>105 (22%)</td>
<td>145</td>
<td>134(2.4%)</td>
<td>150</td>
</tr>
<tr>
<td>106 (27%)</td>
<td>146</td>
<td>135(6.6%)</td>
<td>151</td>
</tr>
<tr>
<td>108 (26%)</td>
<td>148</td>
<td>136(7.8%)</td>
<td>152</td>
</tr>
<tr>
<td>110 (12%)</td>
<td>150</td>
<td>137(11.3%)</td>
<td>153</td>
</tr>
<tr>
<td></td>
<td></td>
<td>138(71.7%)</td>
<td>154</td>
</tr>
</tbody>
</table>

No Molecular Ions for 149.

$^{110}$Pd(12%)Ca and $^{134}$Ba(2.4%)O for mass 150, however their effects should be lower than $^{106}$Pd(27%)Ca and $^{138}$Ba(71.7%)O
Transmutation of Natural Ba into Sm

- XPS analysis showed Sm signal.
- SIMS analysis showed the increase of mass 150.
- Natural Sm isotopic distribution did not match with SIMS mass data.
- These facts strongly suggest that $^{150}\text{Sm}$ exists on the Pd complex after D$_2$ gas permeation.
Transmutation of Ba into Sm; mass 137 Enriched Ba

$^{137}\text{Ba}$ (Enriched Ba)

$^{149}\text{Sm}$ were possibly detected after D permeation on the Pd complex
SIMS Spectra for #1Experiment

Counts

Mass Number

137Ba

149Sm

Pd^{40}Ca

BaO

FG.

BG.

Sm Natural Abundance

Ratio to 152Sm

151

152

153

154

144 145 146 147 148 149 150 151 152 153 154

Mass Number
SIMS Spectra for #2 Experiment

Counts

1000
100
10
1

Mass Number

135 140 145 150 155

137\text{Ba}

149\text{Sm}

Pd^{40}\text{Ca}

BaO

FG.

BG.

Ratio to \text{u/Sm}

0 0.2 0.4 0.6 0.8 1

144 145 146 147 148 149 150 151 152 153 154

Sm Natural Abundance
Transmutation of $^{137}$Ba into Sm

- SIMS analysis showed the increase of mass 149.
- Natural Sm isotopic distribution did not match with SIMS mass data.
- XPS analysis showed very weak Sm spectra. Now we are trying to obtain clear XPS signals.
- These facts suggests that $^{149}$Sm exists on the Pd complex if we consider that Sm spectra were obtained by XPS using natural Ba.
Mass Correlation between Given and Detected Elements

138\text{Ba} \rightarrow 150\text{Sm}

137\text{Ba} \rightarrow 149\text{Sm}

FG.
BG.
FG.BG.
The Aim of Ba Transmutation Experiments

$^{137}_{56} \text{Ba} \rightarrow 6d(3\alpha) \quad ^{149}_{62} \text{Sm}$

Experimental Results

$^{149}\text{Sm}$ is a Mossbauer Isotope

Excitation Energy: 22.5keV

If we measure the Mossbauer effect of $^{149}\text{Sm}$, we will obtain clear evidence of generation of $^{149}\text{Sm}$. And the information on the ultra fine structure relating to the electronic state and phonon of the generated $^{149}\text{Sm}$ will be obtain.
Recent Results; Part 2

Pr Confirmation by XRF and Experiments for in-situ Measurement at SPring-8
Identification of Pr by X-ray Fluorescence

Detection of Pr using SOR X-ray at Spring-8, Harima, Japan (FG1, FG2: Signals from Samples after D2 Permeation, BG: Signals from the sample before Permeation)
Experimental Set-up for *in-situ* Measurement located at SPring-8
Photograph of the Experimental Set-up

BL-37XU

Detector

Chamber

SOR X-ray
An Example of Pr Detection by the Experiments at SPring-8
Recent Results; Part 3

Measurement and Experiments relating to the role of CaO
TEM Photograph of the Pd Complex

- Thin Pd Film: 400 Å
- CaO and Pd Layers: 1000 Å
- Bulk Pd: 0.1 mm

25mm × 25mm

400 Å

Pd

CaO

Pd Substrate
Depth Profile of Cs and Pr by TOF-SIMS

![Graph showing the depth profile of Cs and Pr with TOF-SIMS data. The x-axis represents sputtering time in seconds, ranging from 0 to 1000, and the y-axis represents counts in arbitrary units. The graph displays two sets of data: No D and After D. For Cs, the counts peak at around 200 seconds and decrease significantly after 600 seconds. For Pr, the counts increase sharply at 200 seconds and remain higher than Cs after 600 seconds. The graph also indicates a 100 Å scale on the side.](image-url)
Depth Profile of Cs and Pr by XPS (1)
D⁺ Ion Bombardment Experiment
Performed at Tohoku Univ.

Experimental Apparatus

D⁺ Ion beam bombardment on metal target
Deuterium Density measured by $D^+$ Ion Bombardment Experiment

Density of Pd complex (Pd/CaO) indicated one order larger than normal Pd.
**MgO cannot work instead of CaO**

<table>
<thead>
<tr>
<th>MgO/Pd 100nm</th>
<th>Cs</th>
<th>No Pr; Two cases out of two experiments.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pd 40nm</td>
<td></td>
<td>ICP-MS measurements show no Pr(&lt;0.01ng).</td>
</tr>
<tr>
<td>Pd 0.1mm</td>
<td></td>
<td>D₂ gas Flow rate enough(2-3sccm)</td>
</tr>
<tr>
<td>25mm × 25mm</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>CaO/Pd 100nm</th>
<th>Cs</th>
<th>Almost every time Pr were detected.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pd 40nm</td>
<td></td>
<td>More than 60 cases.</td>
</tr>
<tr>
<td>Pd 0.1mm</td>
<td></td>
<td></td>
</tr>
<tr>
<td>25mm × 25mm</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
Consideration on the Role of CaO

- Increase of Deuterium Density?
- Modify the Electronic State of Surface Pd?

Depth Profile Measurement of D
By a Resonance Nuclear Reaction

\[ \frac{7}{3}Li(\frac{2}{1}d, \gamma)\frac{9}{4}Be \]
1. Transmutations of Ba into Sm were observed both for the case of giving natural Ba on Pd complex samples and for the case of giving mass 137 enriched Ba. It means that we obtained mass distribution of Sm depending on given isotopic distribution of Ba by our experimental method.

2. One of our experimental apparatus was carried to SPring-8 for the purpose of in-situ measurement and we obtained some Pr signals by the X-ray Fluorescence method.

3. According to a D+ ion beam bombardment experiment performed at Tohoku University, deuterium density of our Pd complex indicated one order larger than normal Pd.