LLFP Transmutation in ImPACT project in Japan

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Kenji NISHIHARA
Japan Atomic Energy Agency
Reiko FUJITA
Former Japan Science and Technology Agency

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ImPACT : Impulsing PAradigm Change through disruptive Technology
*www8.cao.go.jp/cstp/sentan/about-kakushin.html
ImPACT (Impulsing PAradigm Change through disruptive Technology)

The Japanese Government fully supported the new R&D Program named “ImPACT” which has started 2014 till March 2019 followed the “First” Program.

- Objectives of the ImPACT Program
  To promote R&D of high risk and high impact for societies or industries with destructive innovations.

- Features of the ImPACT Program
  The DARPA in the US for Defence is the model of the ImPACT Program. The Program Manager is not a researcher but completely managing the whole program with the budget and the authority.
Theme 1. Release from constraints on resources and innovation in manufacturing capabilities

“Japan-Style Value Creation for the New Century”

- Effective utilization of limited resources, highly advanced functionality without the use of expensive resources, and the use of substitute rare resources is difficult.
- There are no realistic methods to making use of unutilized and undiscovered maritime and other such resources.
- If there is no innovation in production technology, the production of high value-added products will rapidly become obsolete.
- The quality, constituents, and production volume of agriculture, forestry, and fishery production are difficult to regulate in response to changing weather conditions. And other such issues.

Theme 2. Realization of an ecologically sound society and innovative energy conservation that changes lifestyles

“Living in Harmony with the World”

- There are no effective methods of achieving large-scale energy conservation while also improving the quality of life (mobile infrastructure, lighting, heating and cooling, information appliances, etc.).
- There are no methods for drastically reducing the volume of waste, which is trending always upward. And other such issues.

Theme 3. Realization of a comfortable living in a society with a declining birthrate and aging population

“Realize Healthy and Comfortable Lives for Everybody”

- Health problems of elderly people, inconveniences of everyday life, and concerns about the healthy growth of children have not been resolved.
- There are no effective ways for people to be freed from the din of motor vehicles, railways, and so on, and to lead lives that bring them healing relief.
- There are no simple, convenient, effective ways for people to protect themselves from the toxins and hazardous substances (viruses, bacteria, explosives, substances impacting food safety, etc.) they find close at hand in their lives. And other such issues.

Theme 4. Provide the world’s most comfortable living environment to society with a declining birthrate and aging population

“Realize Healthy and Comfortable Lives for Everybody”

- There is insufficient capability for prediction of natural phenomena, control of their effects, rapid search, rescue, and transportation when disasters occur, restoration of bridges, roads, and other such infrastructure, and assuming access in times of emergency, dealing with toxic substances, hazardous substances, and other such substances generated by disasters, accidents, or other such events by decontamination or preventing their spread, and other such readiness for dealing with natural disasters.
- There are impediments to advanced mobility in rainstorms, windstorms, nighttime, and other extreme environments, and to increasing the safety and speed of remote demolition of structures and other such heavy work. And other such issues.

Theme 5. Control the impact and minimize the damage from hazards and natural disasters that are beyond human knowing

“Realize a Resilience that is Keenly Felt by Every Individual Japanese”

- There is no effective method of reducing the extent of disasters and their impact, developing a technique for early warning of earthquakes and other natural disasters, and increasing the effectiveness of emergency response agencies and emergency services to reduce their impact.
- There are no effective methods for reconstructing damaged infrastructure and structures, and for increasing the safety and speed of remote demolition of structures and other such heavy work. And other such issues.

“Reduce and Resource Recycling of High-level Radioactive Waste through Nuclear transmutation” was adapted as one of the ImPACT program on June 2014 till March 2019.
Partitioning and Nuclear transmutation are necessary for Resource Recycling and Reduction of the waste volume.

*ImPACT: Impulsing Paradigm Change through disruptive Technology by Cabinet office, Japanese Government*
The 21st Century Invention Prize in 2018

PAT. No.6106892 “Radioactive Waste Treatment Process”
### List of long-lived isotopes in spent nuclear fuel

<table>
<thead>
<tr>
<th>Nuclides</th>
<th>Half life (year)</th>
<th>Radiation conversion coefficient (μ Sv / kBq)</th>
<th>Content (Spent fuel /ton)</th>
</tr>
</thead>
<tbody>
<tr>
<td>U-235</td>
<td>0.7 B.Y.</td>
<td>47</td>
<td>10 kg</td>
</tr>
<tr>
<td>U-238</td>
<td>4.5 B.Y.</td>
<td>45</td>
<td>930 kg</td>
</tr>
<tr>
<td>Pu-238</td>
<td>87.7</td>
<td>230</td>
<td>0.3 kg</td>
</tr>
<tr>
<td>Pu-239</td>
<td>24000</td>
<td>250</td>
<td>6 kg</td>
</tr>
<tr>
<td>Pu-240</td>
<td>6564</td>
<td>250</td>
<td>3 kg</td>
</tr>
<tr>
<td>Pu-241</td>
<td>14.3</td>
<td>4.8</td>
<td>1 kg</td>
</tr>
<tr>
<td>Np-237</td>
<td>2.14 Million</td>
<td>110</td>
<td>0.6 kg</td>
</tr>
<tr>
<td>Am-241</td>
<td>432</td>
<td>200</td>
<td>0.4 kg</td>
</tr>
<tr>
<td>Am-243</td>
<td>7370</td>
<td>200</td>
<td>0.2 kg</td>
</tr>
<tr>
<td>Cm-244</td>
<td>18.1</td>
<td>120</td>
<td>60 g</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Nuclides</th>
<th>Half life (Million year)</th>
<th>Radiation conversion coefficient (μ Sv / kBq)</th>
<th>Content (kg / Spent fuel ton)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Se-79</td>
<td>0.295</td>
<td>2.9</td>
<td>6g</td>
</tr>
<tr>
<td>Zr-93</td>
<td>1.53</td>
<td>1.1</td>
<td>1</td>
</tr>
<tr>
<td>Tc-99</td>
<td>0.211</td>
<td>0.64</td>
<td>1</td>
</tr>
<tr>
<td>Pd-107</td>
<td>6.5</td>
<td>0.037</td>
<td>0.3</td>
</tr>
<tr>
<td>Sn-126</td>
<td>0.1</td>
<td>4.7</td>
<td>30g</td>
</tr>
<tr>
<td>I-129</td>
<td>15.7</td>
<td>110</td>
<td>0.2</td>
</tr>
<tr>
<td>Cs-135</td>
<td>2.3</td>
<td>2.0</td>
<td>0.5</td>
</tr>
</tbody>
</table>

Radiation Conversion Coefficient = dose conversion factor for ingestion

Citation: Hiroyuki Oigawa, “Tokai Forum lecture 9” (2014)
## Previous Studies on Transmutation of MAs and LLFPs in HLW

<table>
<thead>
<tr>
<th>U</th>
<th>Pu</th>
<th>Transmutation targets</th>
<th>Facilities</th>
<th>Issues</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>Actinides: Np, Am, Cm</td>
<td>Fission Products (FP): 129I, 99Tc, 79Se, 93Zr, 107Pd, 135Cs</td>
<td>Reactor</td>
</tr>
<tr>
<td>EU</td>
<td></td>
<td>○ ○ ○ ○</td>
<td>○ ○ ○ ○</td>
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<tr>
<td>US</td>
<td></td>
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<td>○ ○ ○ ○</td>
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<tr>
<td>OMEGA (Japan)</td>
<td></td>
<td>○ ○ ○ ○</td>
<td>○ ○ ○ ○</td>
<td>○ ○ ○ ○</td>
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<tr>
<td>SCNES* (Japan)</td>
<td></td>
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<td>○ ○ ○ ○</td>
<td>○ ○ ○ ○</td>
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<tr>
<td>ADS-MA (JAEA)</td>
<td></td>
<td>○ ○ ○ ○</td>
<td>○ ○ ○ ○</td>
<td>○ ○ ○ ○</td>
</tr>
<tr>
<td>ImPACT (Japan)</td>
<td>Use of results of ADS-MA</td>
<td>Use of results of OMEGA and SCNES</td>
<td>○ ○ ○ ○</td>
<td>○ ○ ○ ○</td>
</tr>
</tbody>
</table>

*SCNES: Self-Consistent Nuclear Energy System

We targeted 4 LLFPs by accelerator.
Recently, the most powerful accelerator, RI beams factory such as 100 times stronger of beam strength in any other facilities at present in the world has been completed, and any kind of nuclear reaction data for LLFPs is possible to be measured by the innovative technique.

The excellent simulation software “PHITS” and evaluated nuclear database “JENDL” have already been in Japan.

The advanced transmutation system becomes possible to be developed.
Example of Pd-107

Key steps are: Chemical separation, Even/odd separation & Nuclear transmutation

Chemical separation of Pd

Even/Odd Separation

Pd-107 and Pd-105 of picking out from recovered Pd

<table>
<thead>
<tr>
<th>Abundance ratio</th>
<th>16</th>
<th>28</th>
<th>27</th>
<th>16</th>
<th>11</th>
<th>0</th>
<th>3</th>
</tr>
</thead>
<tbody>
<tr>
<td>Neutron Number</td>
<td>58</td>
<td>59</td>
<td>60</td>
<td>61</td>
<td>62</td>
<td>63</td>
<td>64</td>
</tr>
</tbody>
</table>

Stable Stable Stable Stable Half life Stable 13hours

Even

Odd

Transmutation

Recycled Pd

To stable nuclides by adding or removing neutron
Structure of this program

Advanced and improvement reprocessing WG ✔
4 more industries etc.

Project 1 Separation & Recovery
- LLFP Chemical separation
- Even/Odd separation by laser technique

Project 2 Nuclear reaction data
- Measurement of new nuclear reaction data
- Confirmation of nuclear reaction

Project 3 Model & Simulation
- Reaction model
- Bulk simulation
- New nuclear reaction database

Project 4 Accelerator development & Transmutation system &
- New accelerator development
- Deuteron beam generation system and its element development

Project 5 Conceptual design for P&T
- Decrease of HLW disposal burden
- Proposal of Clearance level
- Conceptual design of total system

Transmutation plant for practical use ~2040
Pilot plant for demonstration ~2030
New disposal concept WG ✔ 4 more industries etc.
New accelerator concept WG ✔ 5 industries etc.
The program consists of 5 sequential projects
Project 1: Separation and recovery techniques

1. Dissolution of glass solid
2. LLFPs Recovery process
3. Even/Odd separation (Riken)

- Dissolution of vitrified wastes

Target element
(Pd-107, Cs-135, Zr-93, Se-79)
(Pd-107: Mixture of Pd-103~Pd-110)

- Even-odd separation by laser technique
  (Designated organization: Riken)

Extraction from existing glass is considered, too.
Project 1: Chemical recovery from HLLW

Separating technologies with limited secondary wastes

- **no pretreating**: use HLLW directly
- **no liquid change**: separate from nitric acid (HNO₃)
- **re-usable**: adsorbent, extractant

### Chemical recovery processes

- **Electrolytic deposition**
  - HNO₃ (Pd, Se, Cs, Zr)
  - Metal Recovery ratio of 92% at the electrode by potentiostatic electrolysis

- **Zeolite adsorption**
  - HNO₃ (Cs, Zr)
  - Adsorption ratio of 91% by natural zeolite

- **Solvent extraction**
  - HNO₃ (Zr)
  - Distribution ratio (D) was confirmed to suggest recovery ratio of \( \geq 90\% \)

**HLLW**

#### Series test using simulated HLLW
- Resulted in sufficient recovery ratio.

**Pd**: Selectively deposit at high voltage
- Metal Recovery ratio of 92% at the electrode by potentiostatic electrolysis

**Cs**: Recovery using zeolite adsorption
- Adsorption ratio of 91% by natural zeolite

**Se**: Recovery with noble metal (NM)
- Metal Recovery ratio of 20% with Pd

**Zr**: Separation using solvent extraction
- Distribution ratio (D) was confirmed to suggest recovery ratio of \( \geq 90\% \)
Example of HLW Palladium

Odd nuclides (Pd107, Zr93 ..) are ionized, picked up and recovered on cathode.
Project 1: Improvement of even/odd separation

We achieved very high efficiency by $10^5$ times to original scheme.

- Annual production of Pd from 1GWe plant is 27kg/year/GWe.
- To process this amount, 490 Tera laser at 10kHz is required, which is larger by 300 times than current technology.
- Strengthening of laser and irradiation with multiple light paths will solve this gap.
Project 2: Measurement of nuclear data

**Target LLFP**

<table>
<thead>
<tr>
<th>Target</th>
<th>Half life</th>
</tr>
</thead>
<tbody>
<tr>
<td>Palladium 107</td>
<td>6.50 M.Y</td>
</tr>
<tr>
<td>Zirconium 93</td>
<td>1.53 M.Y</td>
</tr>
<tr>
<td>Selenium 79</td>
<td>0.30 M.Y</td>
</tr>
<tr>
<td>Cesium 135</td>
<td>2.30 M.Y</td>
</tr>
</tbody>
</table>

**Project 2**

(Obtained nuclear reaction data)
- Neutron Knockout (RIKEN)
- Fast neutron nuclear spallation (Kyushu University)
- Coulomb breakup (TIT)
- Negative muon capture reaction (RIKEN)
- Neutron capture (JAEA)
- Low-speed RI beam (The university of Tokyo, RIKEN)

(New nuclear reaction control method)

**RIKEN RI Beam Factory**

Measurement of LLFP nuclear reactions with reverse kinetics

**J-PARC RIKEN RAL**

Measurement of LLFP neutron capture reaction

- Nuclear fusion (NIFS/Chubu Univ.)
- Compact cyclotron (Osaka Univ.)
- Muon (Kyoto Univ./JAEA)

Stable nuclides or Short-lived nuclides

- Neutrons
- Photons

Reverse kinetics with neutrons, Protons, Photons

First neutron, slow neutron reaction

- Others, new neutron reaction control methods
Project 2: Reverse kinetics

Superconductor Accelerator

Uranium

Nuclear fission

Palladium (Pd)–107
Half life: 6.50 million years

Proton (H) target

Measurement results

Pd-103 (17 days of half life)
Pd-102 (Stable)
Pd-106 (Stable)
Pd-104 (Stable)
Pd-105 (Stable)

Residual nuclides by nuclear reaction of Pd-107

Mass number/Charge

Atomic number

Cross section is deduced by data of each residual nuclides.

“Reverse kinetics” is key technology for LLFP
Project 2: Low primary beam energy

- Q-Q-D magnet configuration (First-half part of SHARAQ spectrometer)
  - Q1, Q2 (Superconducting)
    - Bore: 340 mm$^w \times 230$ mm$^h$
    - Length: 1020 (530) mm for Q1 (Q2)
  - D1
    - $\rho = 2.57$ m, 52.7° bending
    - Gap: 200 mm

OEDO equipment was installed to decelate LLFP beam to 25MeV/u keeping high resolution.
Project 2: Low primary beam energy

Reaction products of $^{107}$Pd+p reaction

Cross sections of Ag isotopes in $^{107}$Pd+p reaction

LLFP cross section at 25MeV was newly obtained.
Project 2: Verification test of $^{107}$Pd transmutation

The *world’s first* verification test of $^{107}$Pd

- Target fabrication by $^{107}$Pd implantation
- Irradiation test with 12 MeV/u deuteron

![Image of 107Pd sample and target setup]
Project 3: New nuclear data library

New nuclear data library: “JENDL/ImPACT-2018”

- Model-based estimation
  - Updates of phenomenology
  - Microscopic nuclear theory (level density, γ-ray strength function)

- New measurements
- 160 nuclei including $^{79}$Se, $^{93}$Zr, $^{107}$Pd and $^{137}$Cs for neutrons and protons

- $E_n < 20$ MeV

- CCONE code
  (phenomenology, completeness)

- IAEA/EXFOR
  (existing measurements)

- RIBF (SAMURAI)

Project 3: Improve simulation accuracy

Deuteron is the first priority for a primary beam in the present project. Conventional codes (e.g. TALYS, CCONE) are NOT adequate for deuteron-induced reactions. We have applied DEURACS to spallation reactions.

DEURACS – computational code dedicated for deuteron-induced reactions

DEURACS considers the breakup processes.

93Zr + d @ 105 MeV/u

Cross section [mb]

CCONE  DEURACS

Elastic scattering
Non-elastic breakup
Deuteron absorption
Non-elastic breakup
Elastic breakup

Nb  Kr

Mass number [-]
Beam current requested for LLFP transmutation in actual nuclear reaction time* is much higher than present one such as 1mA in every case of isotope separation, even-odd separation and spent fuel without isotope separation.

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Yield of year (kg/y)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Se-79</td>
<td>5</td>
</tr>
<tr>
<td>Pd-107</td>
<td>250</td>
</tr>
<tr>
<td>Cs-135</td>
<td>417</td>
</tr>
<tr>
<td>Zr-93</td>
<td>767</td>
</tr>
</tbody>
</table>

* : one year yield of LLFP transmuted by proton beam with 1000 MeV for 3 years of effectual half years
A new acceleration scheme based on single cell cavities to deliver 1-ampere deuteron beams
Superconducting Quarter-Wave Cavity

Superconducting cavity is essential to achieve low power-consumption and space-saving in linear accelerator. A prototype of QW cavity (75.5 MHz) made of Nb sheets was constructed for low velocity deuteron, to achieve high electric field of 4.5 MV/m and low power loss of 8W. Performance test was successfully organized to obtain the design goals.


A Material-free window for vacuum sealing

A large diameter plasma window was developed for accelerator application. The maximum diameter of the plasma window reached upto 20 mm.


Fig. 1 Developed PW: (a) schematic of the PW; (b) overall view of the PW; (c) internal structure of the intermediate electrode.
Project 5: Treatment of each fraction in HLW

Content of high-level radioactive waste

- **Short-lived, Highly radioactive (4%)**
  - To be disposed of after decay for long-term storage (300 years)
- **Extremely long-lived, Fissionable, α emitter (5%)**
  - To be transmuted to short-lived by fission reaction in advanced nuclear system and disposed of
- **Extremely long-lived, Non-fissionable, βγ emitter (20%)**
  - To be recycled and shortened in lifetime in ImPACT program, then disposed of
- **Stable or short-lived (70%)**
  - To be disposed of as low-level radioactive waste

All radioactive nuclide is stabilized or shortened in lifetime!
Project 5: Decrease of disposal burden

**Number**
- Conventional: 40,000 (#/40years/40GWe)
- ImPACT: 12,000 (#/40years/40GWe)\(\times 0.3\)

**Heat**
- Conventional: 350 W/form, 50 year-storage
- ImPACT: <4 W/form, 300 year-storage\(\times 0.01\)

**Footprint**
- Conventional: Sparse emplacement, Footprint \(\sim 2\text{km}^2\)
- ImPACT: Compact emplacement, Footprint \(\sim 0.01\text{km}^2\)
Project 5: Disposal depth

Conventional

- Isolate highly radioactive waste from living environment.
- Even if it seeps into the groundwater, it is delayed.

Intermediate depth disposal

- Isolate wastes with low radioactive concentration from living environment to some extent.

ImPACT

- More strict safety assessment will be required for intermediate depth
The four LLFPs (Pd, Zr, Se, Cs) studied in this project satisfied the limit values. On the other hand, MAs (Np, Am, etc.), Sn, and Tc, which were not studied, exceeded the limit values. These have been tackled in previous studies, but further reduction is necessary in the future.
Project 5: MA transmutation

Current LWR system

- Uranium mining plant
- Refining plant
- Conversion plant
- Uranium enrichment plant
- Spent fuel reprocessing plant
- MOX fuel fabrication plant
- Light water reactor
- High-level radioactive waste managing center
- Interim storage
- Low-level radioactive waste managing center
- High-level radioactive waste disposal site

Proposed P&T fuel cycle system

- Transmutation plant by Accelerator
- LLFP separation/recovery plant
- Useful metal recovery plant
- Metallic fuel fabrication plant
- Spent fuel reprocessing plant
- Metallic fuel fast reactor
- Re-used metal fabrication plant
- Recovered re-used metal ingot
- Low-level radioactive waste managing center

Reduce High-level Radioactive Waste Disposal site

MA: Transmutation with metallic fuel FBR
Material flow and exposure pathway of Pd107 in Japan

- Industrial use
- Exposure pathways
- Environmental transfer
- Recycle
- Export
### Project 5: Clearance Level of $^{107}\text{Pd}$

Highest effective doses for each of the four exposure pathway and concentrations of $^{107}\text{Pd}$ (1Bq/g) providing a total exposure of 10 $\mu\text{Sv/y}$

<table>
<thead>
<tr>
<th>Exposure Pathways</th>
<th>Highest Dose (mSv/y)</th>
<th>Concentration (Bq/g) which may give 10 $\mu\text{Sv/y}$</th>
<th>Specification</th>
</tr>
</thead>
<tbody>
<tr>
<td>Automobile Catalyst</td>
<td>$3.3 \times 10^{12}$</td>
<td>$3.1 \times 10^{9}$</td>
<td>Adult, Type S</td>
</tr>
<tr>
<td>Food and drinking water</td>
<td>$1.3 \times 10^{10}$</td>
<td>$7.8 \times 10^{7}$</td>
<td>1 year age group</td>
</tr>
<tr>
<td>Dental appliance</td>
<td>$2.0 \times 10^{10}$</td>
<td>$4.9 \times 10^{7}$</td>
<td>Adults, Type S</td>
</tr>
<tr>
<td>Occupational inhalation</td>
<td>$3.2 \times 10^{6}$</td>
<td>$3.3 \times 10^{3}$</td>
<td>Adults, Type S</td>
</tr>
</tbody>
</table>

**Evaluation of parameters and Estimation of other exposure pathway**

**Determination of CLEARANCE LEVELS**
# Project 5: Potential for recycling

<table>
<thead>
<tr>
<th></th>
<th>Unit</th>
<th>Pd</th>
<th>Zr</th>
</tr>
</thead>
<tbody>
<tr>
<td>Annual weight of recycled Pd/Zr</td>
<td>kg/year</td>
<td>1,300</td>
<td>3,100</td>
</tr>
<tr>
<td>(800tHM of SF/year)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Clearance level*</td>
<td>Bq/g</td>
<td>3,300</td>
<td>90</td>
</tr>
<tr>
<td>Impurity ratio of Pd107/Zr93</td>
<td>ppm</td>
<td>170</td>
<td>1.0</td>
</tr>
<tr>
<td>Pd107 / Zr93 impurity in this project</td>
<td>ppm</td>
<td>2,700〜6,900</td>
<td>2,900〜11,700</td>
</tr>
</tbody>
</table>

* Radioactivity concentration equivalent to 10 μSv/year

- Import to Japan is 64 ton/year.
- Recycling can be achieved by reducing the Pd107 content to 15 to 40 times.
- It is necessary to reduce by 3 to 4 digits. The use limited to nuclear power can be considered.
Reduce and Resource Recycling of HLW through Nuclear transmutation was implemented in ImPACT program from 2014 to 2019.

- Recovery of 90% by chemical separation was demonstrated.
- Efficiency of even-odd separation was improved by $10^5$ times.
- New LLFP nuclear data was obtained and new library became available.
- Transmutation of $^{107}$Pd was demonstrated.
- 1A-class deuteron accelerator was designed.
- Accelerator components (cavity, window, target) were developed.
- Possibility of intermediate-depth disposal was shown.
- Clearance level of $^{107}$Pd and $^{93}$Zr was newly estimated.
Summary 2/3: Next Steps

- Expansion to MAs and other LLFPs (I, Tc and Sn) burdening intermediate-depth disposal
- R&D on Cs-135 isotopic separation techniques
- Improvement and demonstration of chemical and even-odd separation using “real” HLLW.
- R&D and trail fabrication of accelerator components (ion source, cavity, target...).
R&D in ImPACT program is corresponding to “Level 6”.

Principle
- Cycle all actinides and all LLFPs, utilizing PGMs.

Theoretical advantage
- Elimination of HLW and geological disposal
- PGMs production

Theoretical disadvantage (=Cost for PT)
- Chemical separation and even-odd isotope separation
- Large accelerator power (1-A class)

Real benefit in the present R&D stage
- Geological disposal is necessary due to MA, Tc, Sn and I.
- 1 ton/year of Pd is recycled in Japanese scale.

Real withdraw = Same to theoretical disadvantage

R&D direction = see “next step” in the previous
Thank you for your kind attention!