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

-Overview and current results of the ImPACT Program-

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ImPACT :Impulsing PAradigm Change through disruptive Technology

**www8.cao.go.jp/cstp/sentan/about-kakushin.html*

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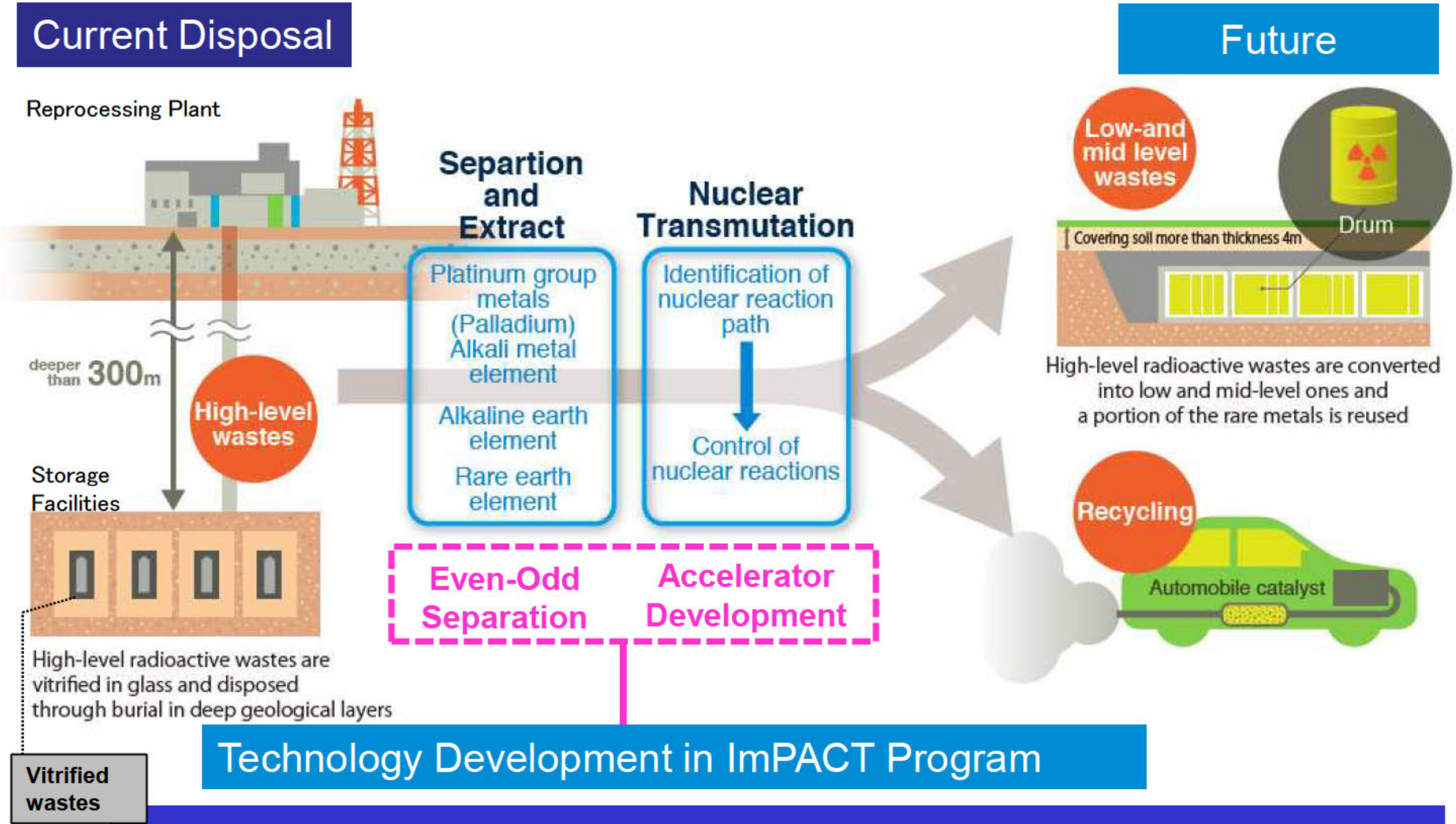
ImPACT* Program



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

2-1. Long Lived Fission Products of Spent Nuclear Fuel

革新的研究開発推進プログラム ImPACT

Ac
TRU
MA

Nuclides	Half life (Billion year)	Radiation conversion coefficient (μ Sv/ kBq)	Content (Spent fuel /ton)
U-235	0.7	47	10 kg
U-238	4.5	45	930 kg

Nuclides	Half life (year)	Radiation conversion coefficient (μ Sv/ kBq)	Content (Spent fuel/ ton)
Pu-238	87.7	230	0.3 kg
Pu-239	24000	250	6 kg
Pu-240	6564	250	3 kg
Pu-241	14.3	4.8	1 kg

Nuclides	Half life (year)	Radiation conversion coefficient (μ Sv/ kBq)	Content (Spent fuel/ ton)
Np-237	2.14Million	110	0.6 kg
Am-241	432	200	0.4 kg
Am-243	7370	200	0.2 kg
Cm-244	18.1	120	60 g

FP

Nuclides	Half life (Million year)	Radiation conversion coefficient (μ Sv/ kBq)	Content (kg/ Spent fuel ton)
Se-79	2.95	2.9	6g
Sr-90	28.8 year	28	0.6
Zr-93	15.3	1.1	1
Tc-99	2.11	0.64	1
Pd-107	65	0.037	0.3
Sn-126	1	4.7	30g
I-129	157	110	0.2
Cs-135	23	2.0	0.5
Cs-137	30.1year	13	1.5

Radiation Conversion Coefficient:
The effect coefficient of nuclides on Human

Citation: Hiroyuki Oigawa, "Tokai Forum lecture 9" (2014)

Previous Studies on Transmutation of MAs and LLFPs in HLW

	Transumutaion targets					Facilities			Issues
	Actinides			Fission Products (FP)		Reactor	ADS	Others	
	U	Pu	MA	Long lived FP					
			Np,Am ,Cm	129I、 99Tc	79Se、93Zr、 107Pd、 135Cs、				
EU			○	○		○	○		Risks of disposal of HLW and LLFPs
US		○	○			○	○		Ibid.
OMEGA (Japan)			○	○		○	○		Risks of LLFPs
SCNES* (Japan)			○	○	○	○			Need of Isotope separation
ADS-MA (JAEA)			○				○		Risks of LLFPs
ImPACT (Japan)			Use of results of ADS-MA	Use of results of OMEGA and SCNES	○			○	Need of Accelarator technology toward zero of HLW without Isotope separation with OMEGA, SCNES and ADS of MA TM

*SCNES: Self-Consistent Nuclear Energy System

Changing the definition of LLFPs on industries and societies

Nuclides	Half-life	Disposal	Resource recycling
Cs(Cesium)-135	2.3million year	**Disposal of Ba (Barium) transmuted from Cs-135**	
Cs(Cesium)-137	30.1year		
Sr(Strontium)-90	28.5 year	Disposal of Rb (Rubidium) -85,87 and Sr-86,88 transmuted from Sr-90	
Pd(Palladium)-107	6.5million year		**Reuse of stable Pd transmuted from Pd -107 for catalysts for vehicles**
Sn(Tin)-126	0.23million year	Transmutation for stable nuclides of Sn(Tin) or Sb(Antimony)	
Zr(Zirconium)-93	1.53 million year		**Reuse of stable Zr transmuted from Zr-93 for Zircalloy cladding and Channel boxes**
Se(Selenium)-79	0.30million yesr	**Stable Se transmuted from Se-79**	
Tc(Technetium)-99	0.21million year	Ru(Ruthenium)-110 transmuted from Tc-99	
I(Iodine)-129	15.70 million year	Xe(Xenon)-130 transmuted from I-129	

- Contribution to safety and security of disposal with converting from High level radioactive wastes to Transuranium (TRU) wastes (ILW) or low level radioactive wastes.
- Most-advanced and only one technology will be established and activate our country's economy.

3-1. Example：Transmutation process of Pd-107

High-level Liquid Waste

↓

Separation-Recovery

Chemical separartion of Pd

Development of separation

↓

Even/Odd Separation

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

Abundance ratio	16	28	27	16	11	0	3
Neutron Number	58	59	60	61	62	63	64
	104	105	106	107	108		110
	Stable	Stable	Stable		Stable	Half life 13hours	Stable

Even/Odd Separation apparatus (RIKEN)

Even isotopes:
All stable nuclides for resource materials

Even: 104, 106, 108, 110

Odd: 105, 107

Pd Ingot

↑

Fabrication Ingot

↑

Transmutation

Recycle process concept

Clearance level proposal

Nuclear reaction data（RIKEN）
Demonstration Test（RIKEN）
Accelerator Development（RIKEN）

Pd-107 transmuted to stable nuclides or short –lived nuclides changing numbers of proton or neutron by beam irradiated

104, 106 — Removed neutron to stable nuclides

105, 107 → To the different stable nuclides or short-lived nuclides by nuclear spallation reaction → Intermediate –level or Low-level wastes

106, 108 — To stable nuclides by adding nuclides

3-2. Outline of this program

ImPACT 革新的研究開発推進プログラム

Automobile catalysts | Magnetic materials,etc | Nuclear medicine | Stable wastes

Palladium | Neodymium | Dysprosium | Xenon | Barium | Cesium

Yttrium | Molybdenum | Rhodium | Zirconium | Strontium | Selenium

Transmutation plant for practical use ～2040

Pilot plant for demonstration ～2030

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 Transmutation system & Accelerator development
- New accelerator development
- Deuteron beam generation system and it's element development

Project 5 Conceptual design
- Decrease of HLW disposal burden
- Proposal of Clearance level
- Conceptual design of total system

New disposal concept WG
✓4 more industries etc.

New accelerator concept WG
✓ 5 industries etc.

4-1. Separation and recovery techniques

Project 1 (Separation & Recovery of LLFP)
①Dissolution of glass solid:
②LLFP Recovery process
③Even/Odd separation (Designated organization : Riken)

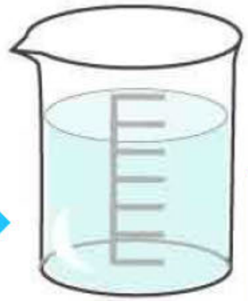
①-1 Pyro (Kyoto Univ./CRIEPI/Toshiba)
①-2 Alkali melt (Fukui Univ.)
①-3 Changing Glass component (Tokyo Univ.)
①-4 Addition of active agent (Ehime Univ.)
①-5 Glass electro-migration (TIT)
①-6 Acid solution (TIT)
①-7 Electrolysis of oxides in molten salts (Doshisha Univ.)

②-1 Volatile flouride (Hitachi)
②-2 Ionique liquid (Keio Univ.)
②-3 Aqueous process (Toshiba/JAEA)

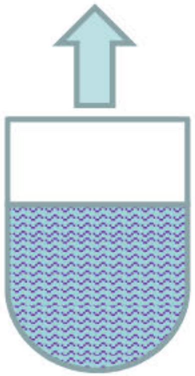
Glass solid

①Dissolution of Vitrified wastes

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

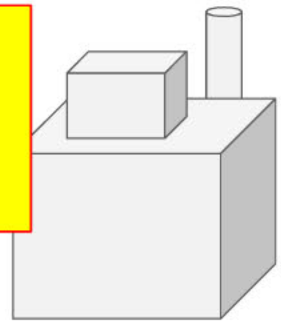


③Even-odd separation by laser technique (Designated organization: Riken)



High Radioactive Waste

② LLFPs Recovery Process from HLLW



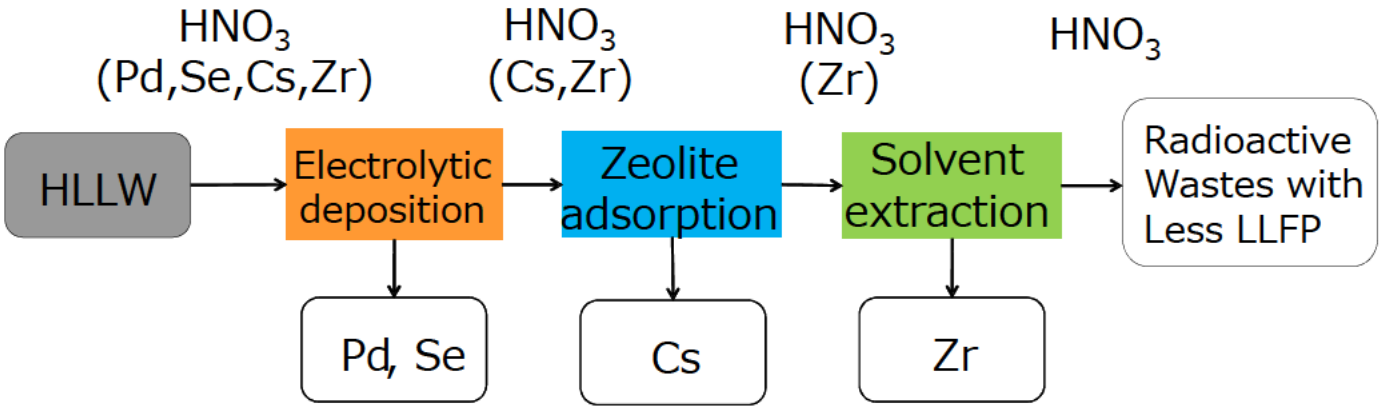
Reprocessing Plant

Separation of even nuclides
(Pd-107 : Limitation to Pd-105 and **Pd-107**)

Current Progress : LLFP Recovery from HLLW

Separating technologies with limited secondary wastes

○no pretreating : use HLLW directly
○no liquid change : separate from nitric acid (HNO_3)
○re-usable : adsorbent, extractant



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

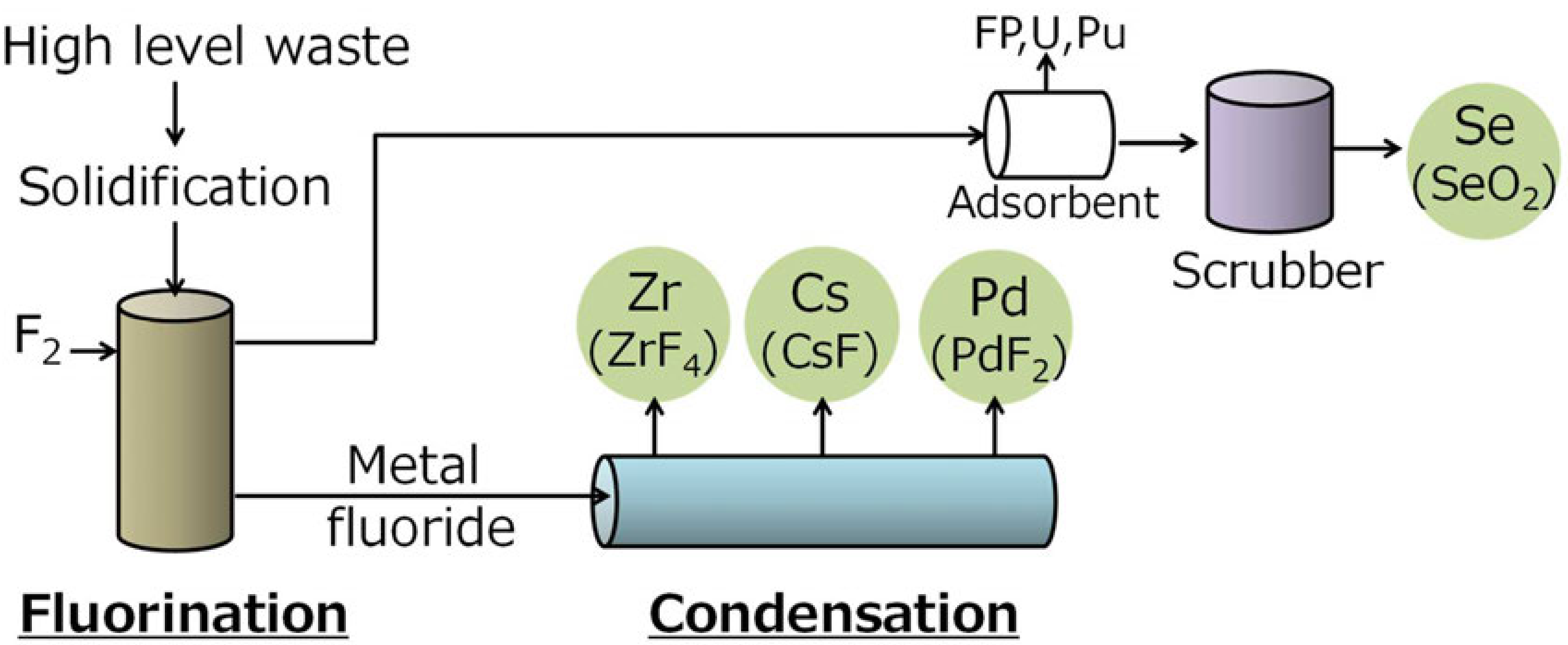
Se: Recovery with noble metal(NM)
Metal Recovery ratio of 20% with Pd

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

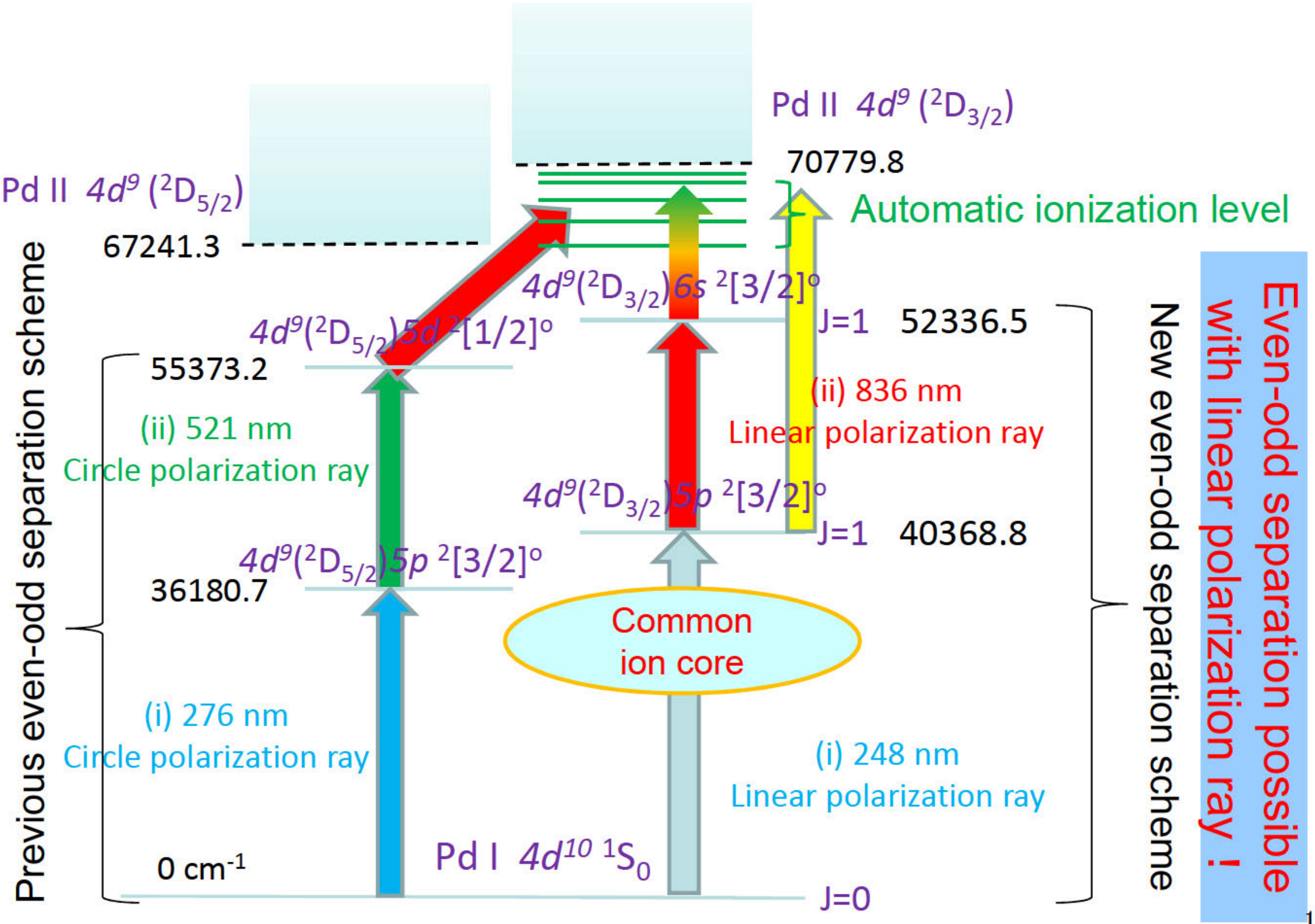
Zr: Separation using solvent extraction
Distribution ratio (D) was confirmed to suggest recovery ratio of ≧90%

Outline of the LLFP separation system

- High level waste is solidified and then reacted with F_2 gas (fluorination) to form metal fluorides
- Metal fluorides are separated based on their vapour pressure difference by "volatilization and condensation"



Current progress : Even/odd separation



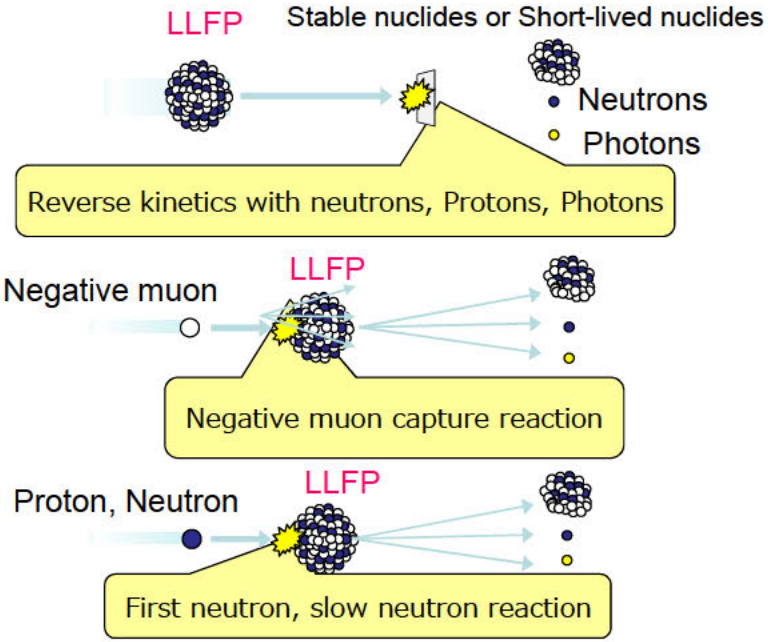
4-2 : Measurement of nuclear reaction data

Target LLFP	Half life
Palladium 107	6.50 M.Y
Zirconium 93	1.53 M.Y
Selenium 79	0.30 M.Y
Cesium 135	2.30 M.Y

Project 2
(Obtained nuclear reaction data)
- Neutron Knockout（RIKEN）
- First 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)
- Nuclear fusion（NIFS/Chubu Univ.）
- Compact cyclotron（Osaka Univ.）
- Muon（Kyoto Univ./JAEA）

- Adaption and overhaul of measure apparatus



- Others, new neutron reaction control methods

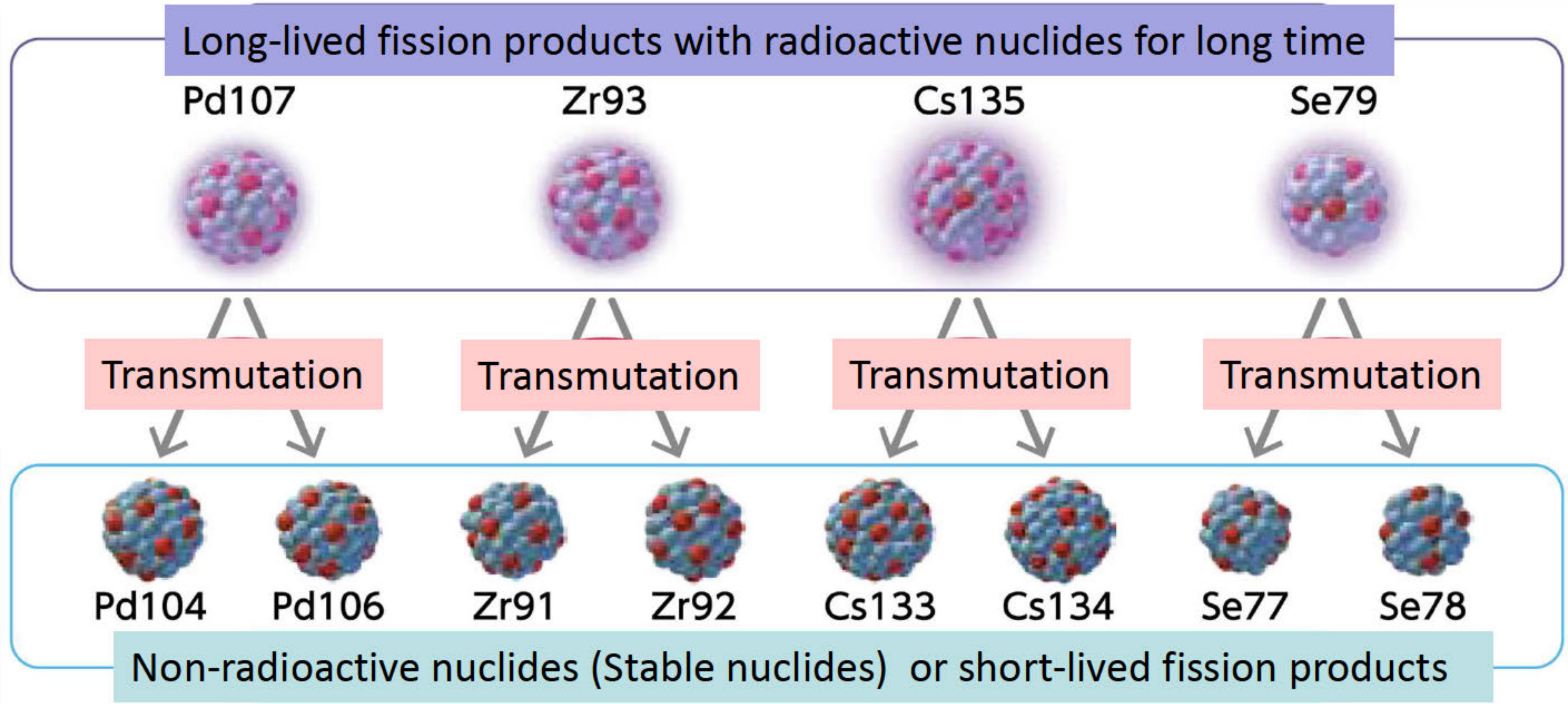
RIKEN RI Beam Factory

Measurement of LLFP nuclear reactions with reverse kinetics

J-PARC
RIKEN RAL

Measurement of LLFP neutron capture reaction

4-2. Transmutation of LLFP

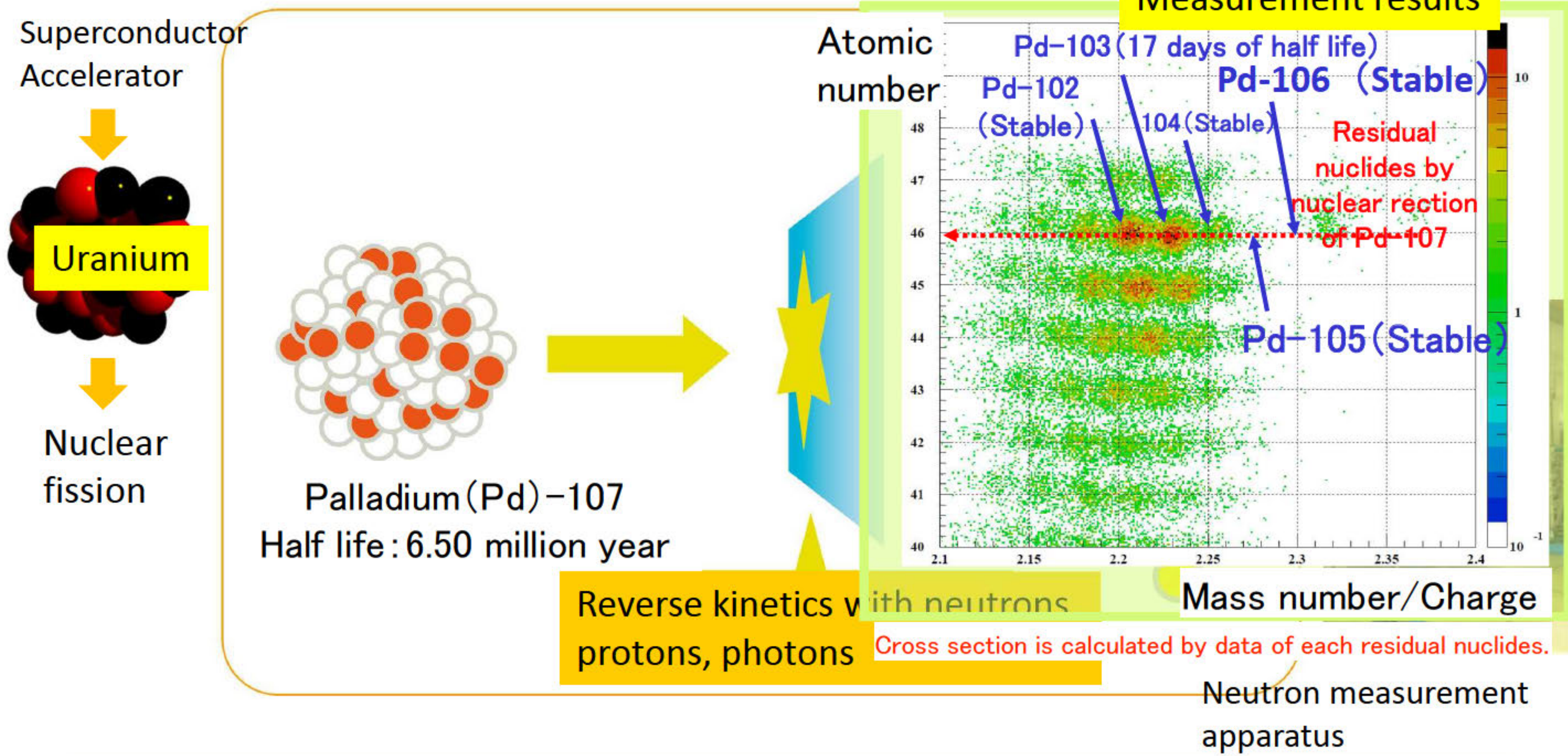


Transmutation: LLFP in HLW converted to short-lived fission products or stable nuclides with nuclear reaction

4-2. Palladium-107 transmutation

Pd-107 transmutation Movie

Current Progress: Measurement of nuclear reaction data



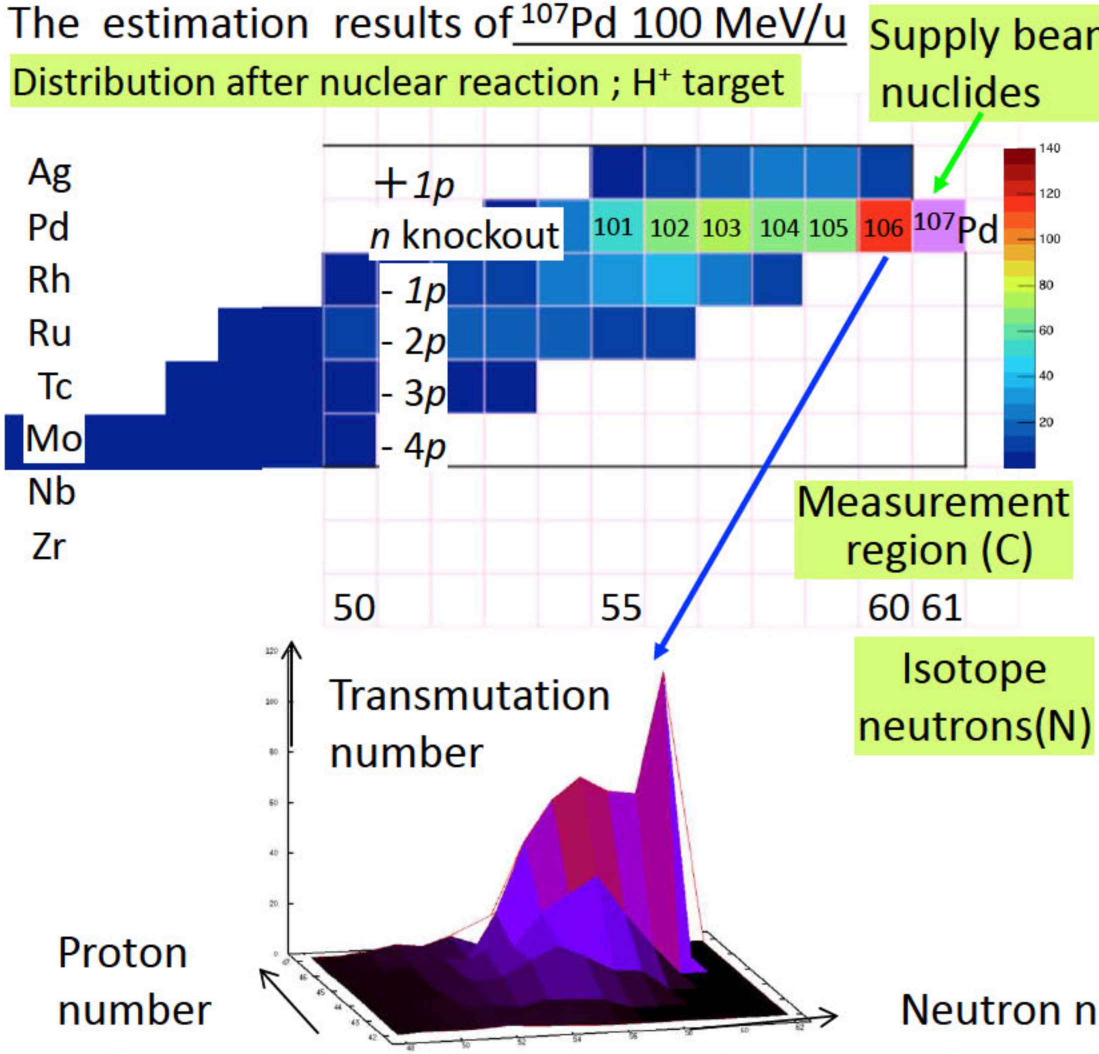
U-238 beam produces non-available long-lived nuclides, which convert to stable nuclides or short-lived nuclides.

Current Progress : High primary beam energy

Pd-107 is transmuted by nuclear reaction with H^+ and estimated of ratio of the nuclear reaction

The estimation results of ^{107}Pd 100 MeV/u

Distribution after nuclear reaction ; H^+ target



- Fragmentation
  ^{107}Pd was transmuted by Neutron knockout reaction

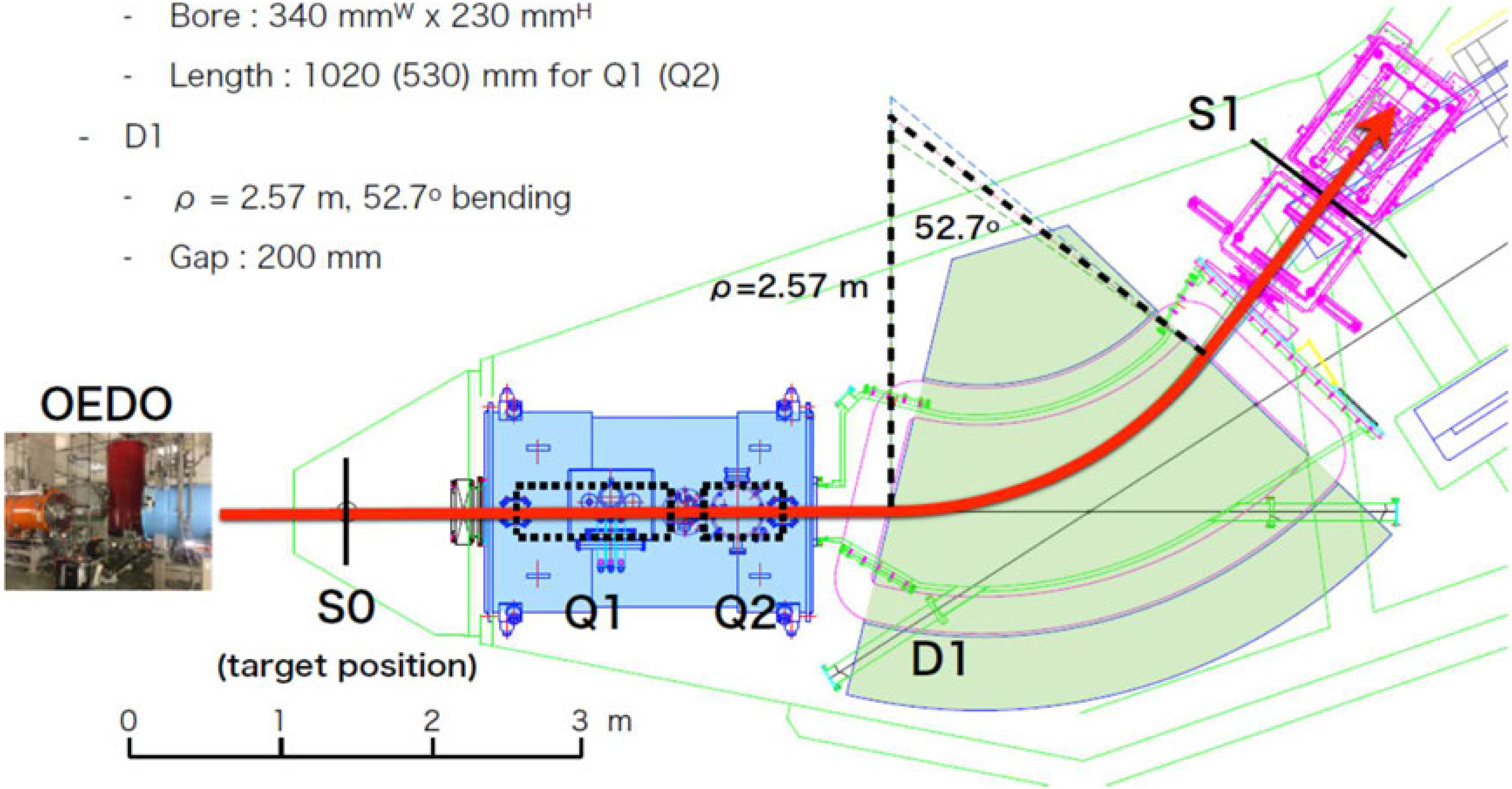
Index of transmutation volume

- Supply number : 107Pd million/hour
- Transmutation number : 10 thousand/ hour（About 1%)

Nuclides	Pd-101	Pd-102	Pd-103	Pd-104	Pd-105	Pd-106	Pd-107
Half life	8.47h	Stable	16.991d	Stable	Stable	Stable	$6.5x10^6$y

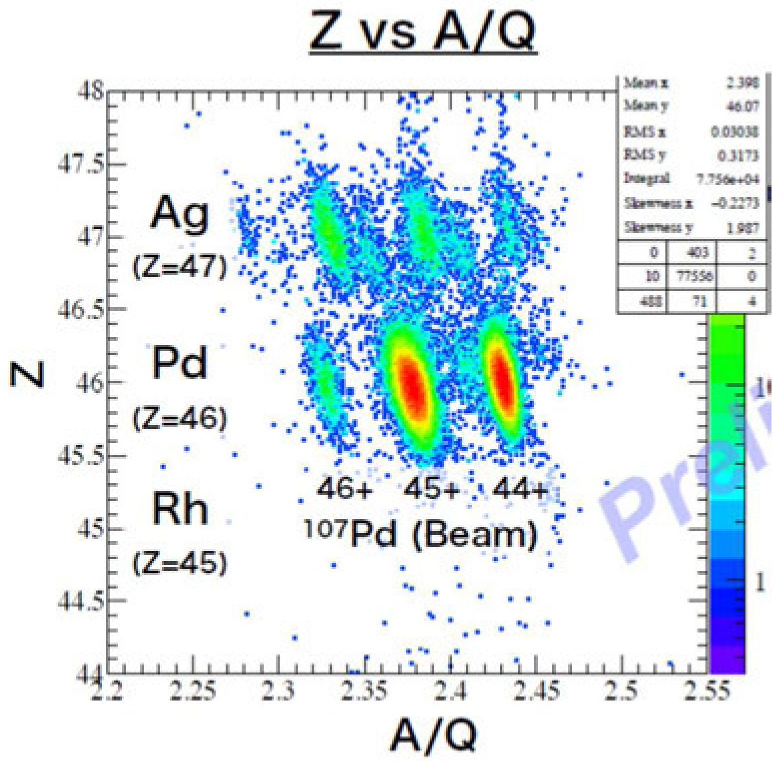
Current Progress : Low primary beam energy

- Q-Q-D magnet configuration (First-half part of SHARAQ spectrometer)
  - Q1, Q2 (Superconducting)
    - Bore : 340 mm^W x 230 mm^H
    - Length : 1020 (530) mm for Q1 (Q2)
  - D1
    - ρ = 2.57 m, 52.7° bending
    - Gap : 200 mm

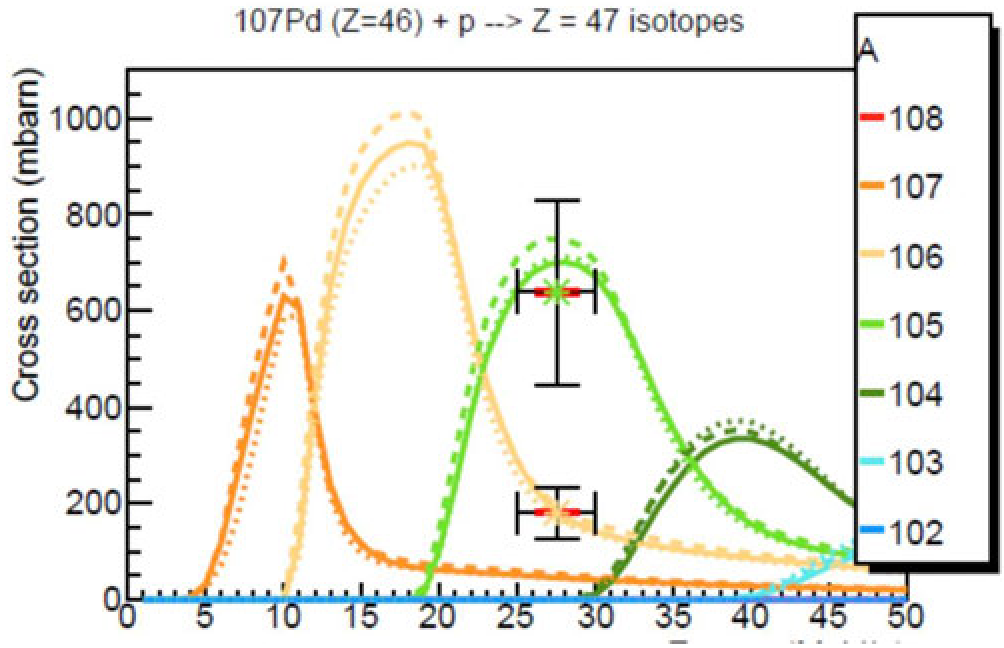


Direct measurement of reaction products from the reaction between low-energy LLFP beam by OEDO and proton/deuteron

Current Progress : Low primary beam energy



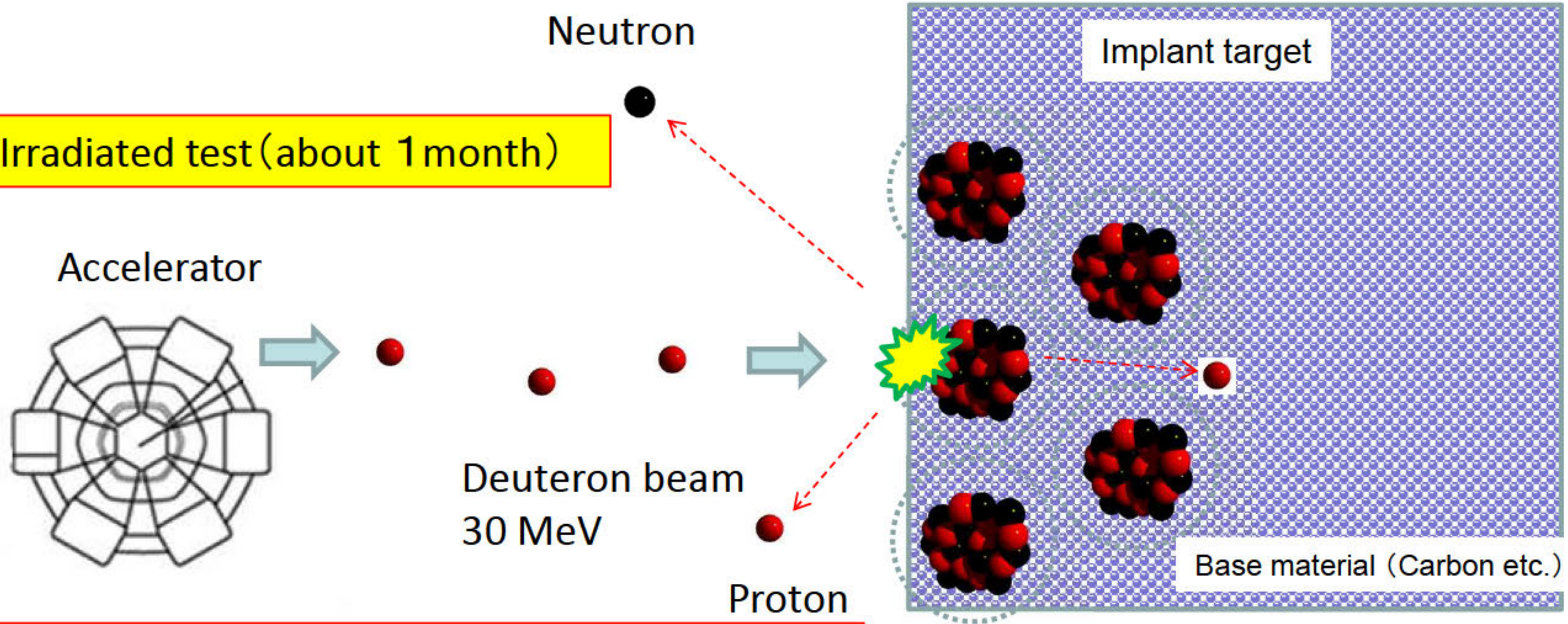
Reaction products of ^{107}Pd+p reaction



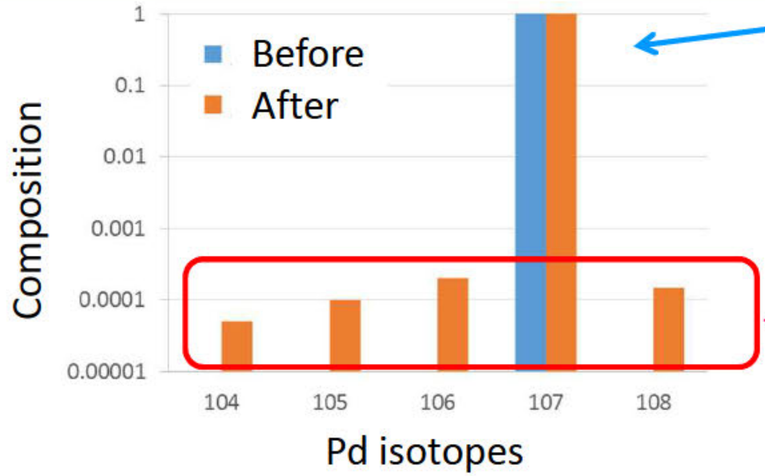
Cross sections of Ag isotopes in ^{107}Pd+p reaction

Current Progress : Demonstration tests

Irradiated test（about 1month）



Produced nuclides by nuclear reactions detected



It is necessary to be irradiated for long time in order to detect amount of nuclear reaction.

If Amount of increase of $^{104\text{-}108}$Pd are detected and measured, it is possible to confirm ^{107}Pd knockout reaction with ratio of isotopes by TIMS.

Current Progress : Demonstration tests

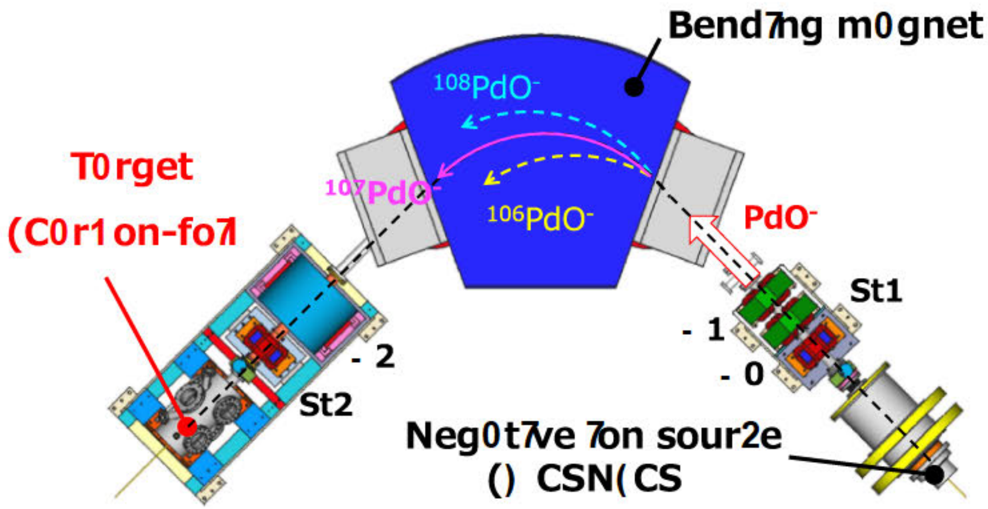


Figure 3 Implantation line

- ^{107}Pd in the sample was electromagnetically-separated and implanted into a carbon foil by the implantation line.

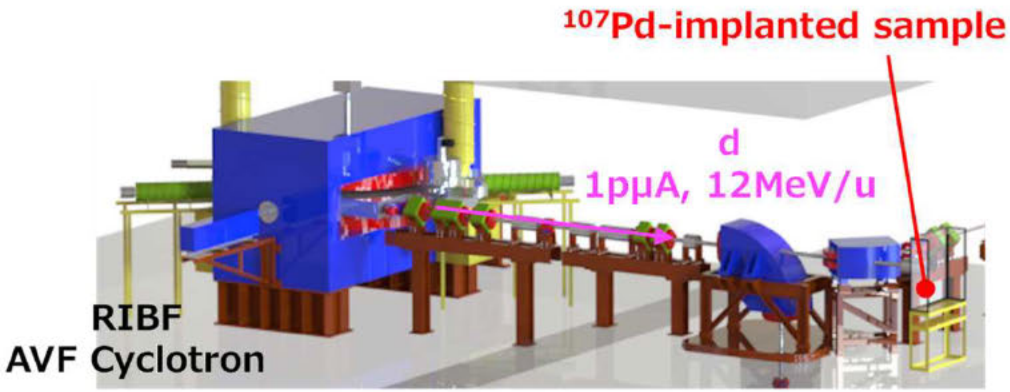


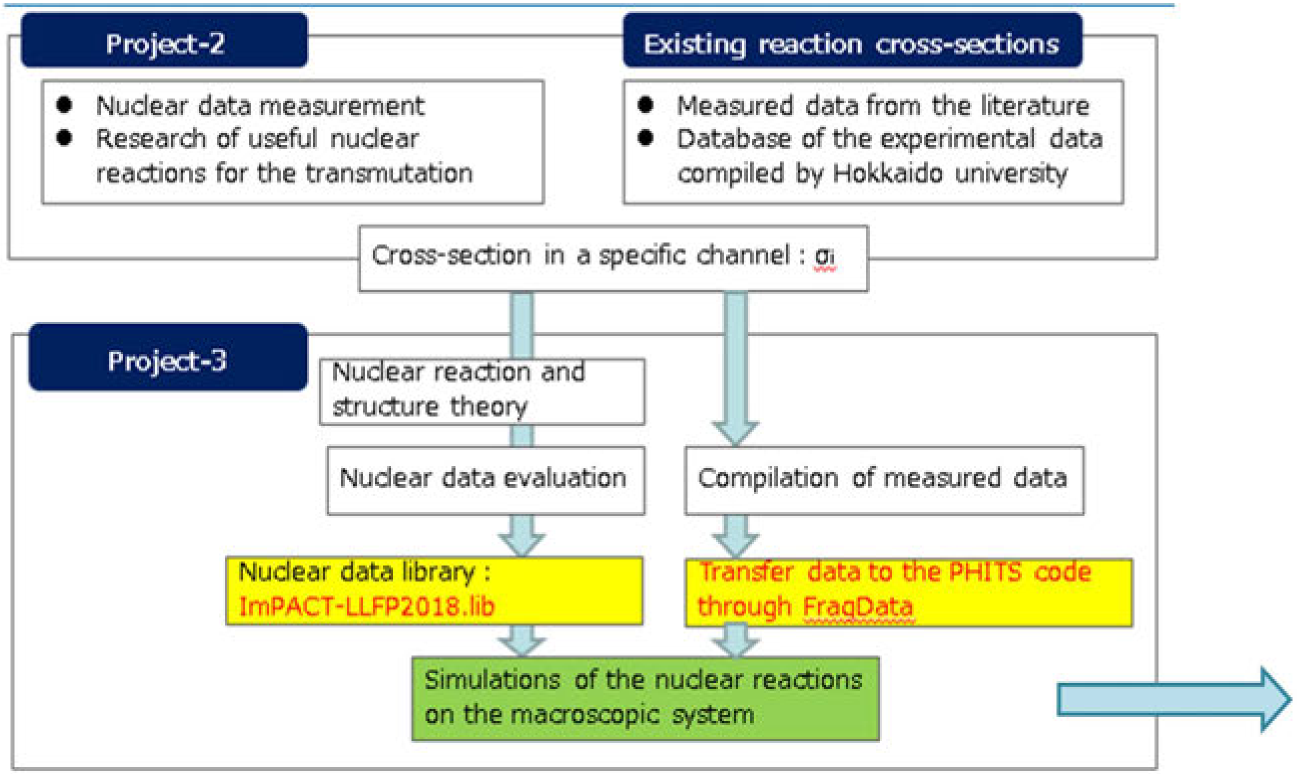
Figure 4 Deuteron irradiation

- ^{107}Pd implanted sample was irradiated by a deuteron at 12 MeV/u produced by the AVF Ring Cyclotron at RIKEN RIBF.

4-3. Improve simulation accuracy of transmutation reaction of long-lived nuclides

- Transmutation reaction database improvement
- Transmutation reaction calculation prediction

→ • Transmutation evaluation tool (PHITS code) improvement

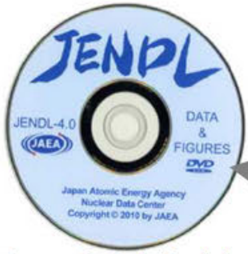


- Estimation of transmutation efficiency on the accelerator-based system
- Evaluation of beam currency, heating, product yield, irradiation damage
- Looking for clues to enhance the transmutation efficiency

Current Progress :New nuclear data library

New nuclear data library :"JENDL/ImPACT-2018"

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



E_n < 20 MeV

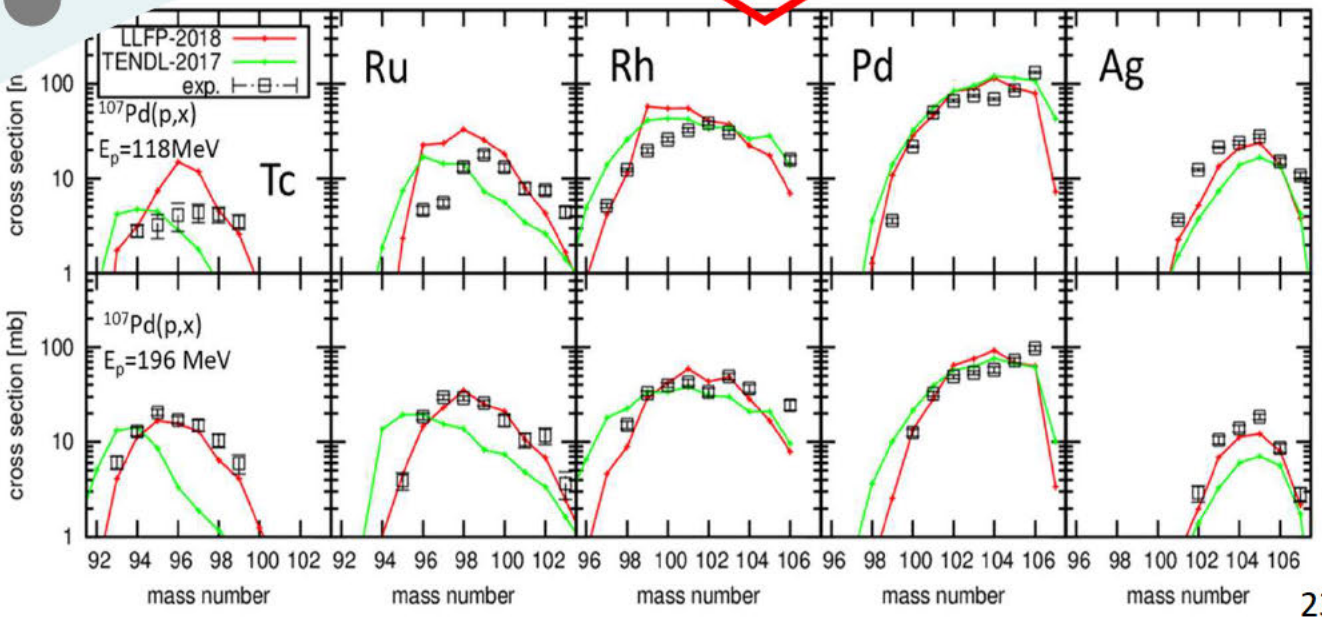
Model-based estimation

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

New measurements

CCONE code
(phenomenology, completeness)

IAEA/EXFOR
(existing measurements)



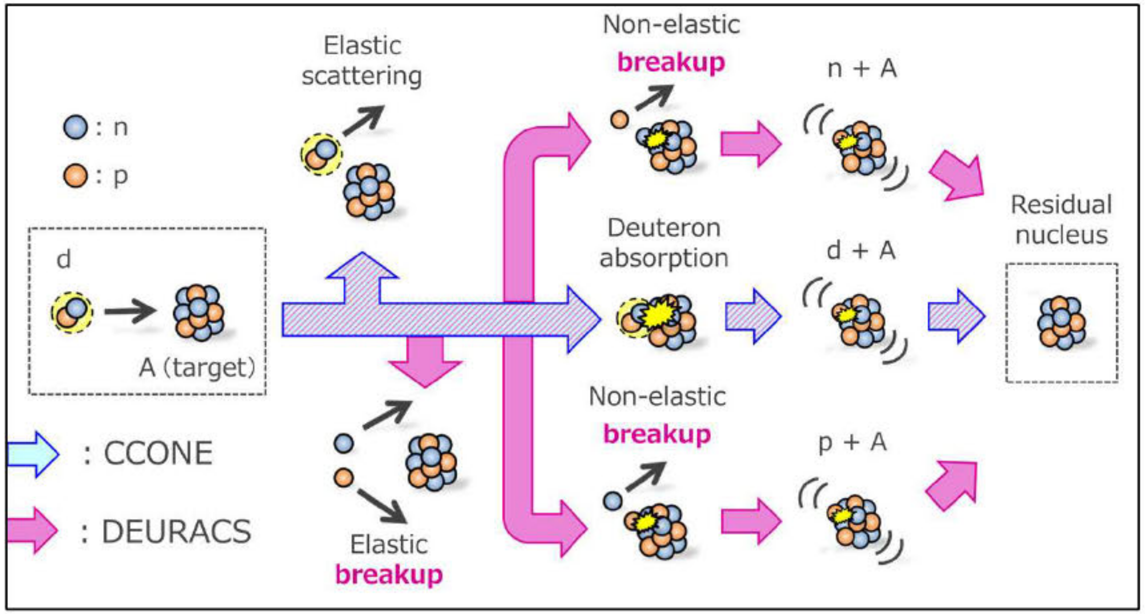
Current Progress : Improve simulation accuracy

- ✓ **Deuteron** is the first priority for a primary beam in the present project.
- → Conventional codes (e.g. TALYS, CCONE) are **NOT** necessarily adequate for deuteron-induced reactions.

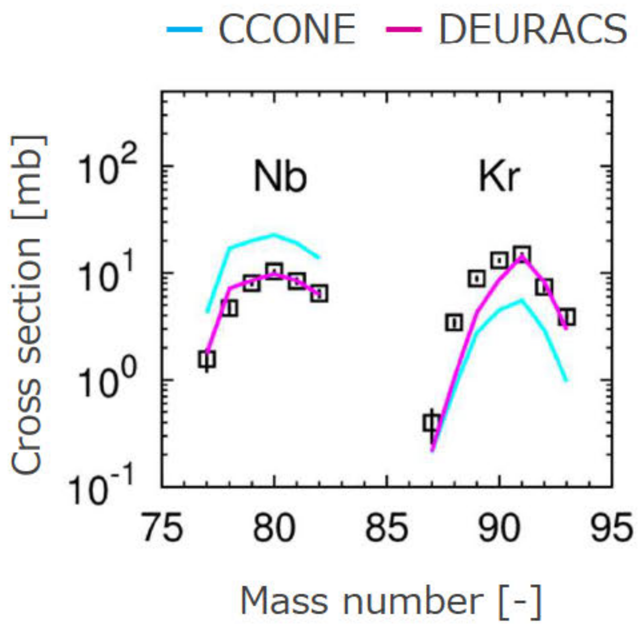
⇨ We have applied **DEURACS** to spallation reactions.

DEURACS – computational code dedicated for deuteron-induced reactions

✓ DEURACS considers **the breakup processes**.



$^{93}Zr + d$ @ 105 MeV/u



4-4 : Accelerator transmutation System and Related Developments for Elemental technologie

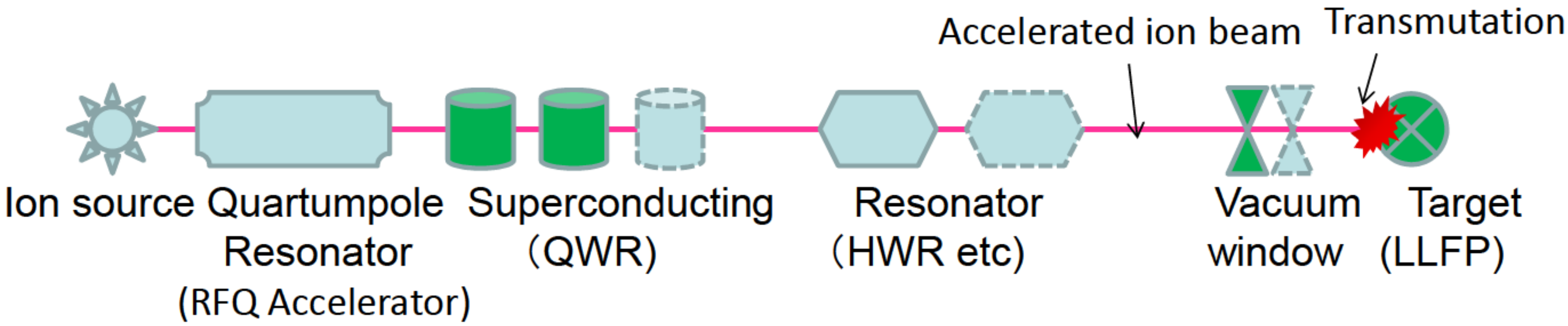
◆ Accelerator and Target development of elemental technologies

・Superconducting Quarter-Wave Resonators

・Vacuum window between accelerator area (High vacuum)and target area (Atmosphere)

・Target development using liquid metal（Heat removal at Transmutation）

[Example of Accelerator and elemental development area]



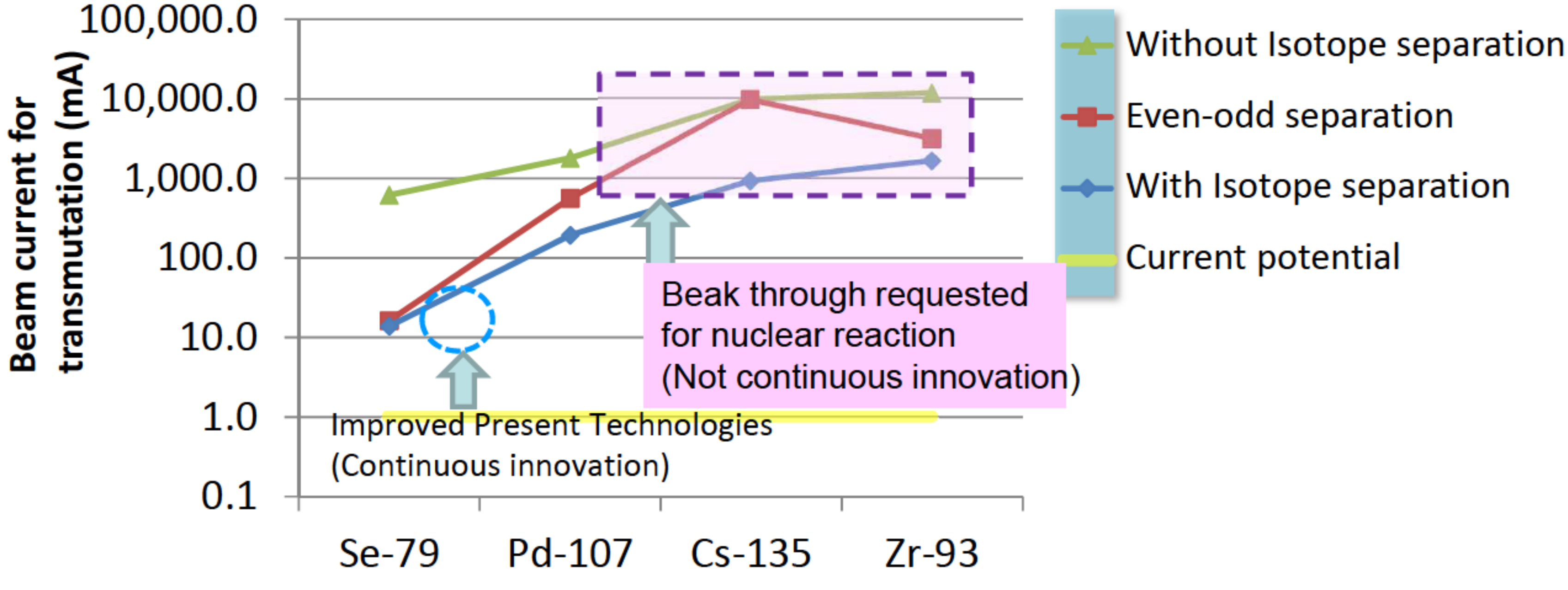
◆ Evaluation of nuclear transmutation system

・Competitive evaluation in nuclear transmutation techniques

・Chosen nuclear transmutation system

Current Progress : Accelerator Transmutation Systen

- 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.

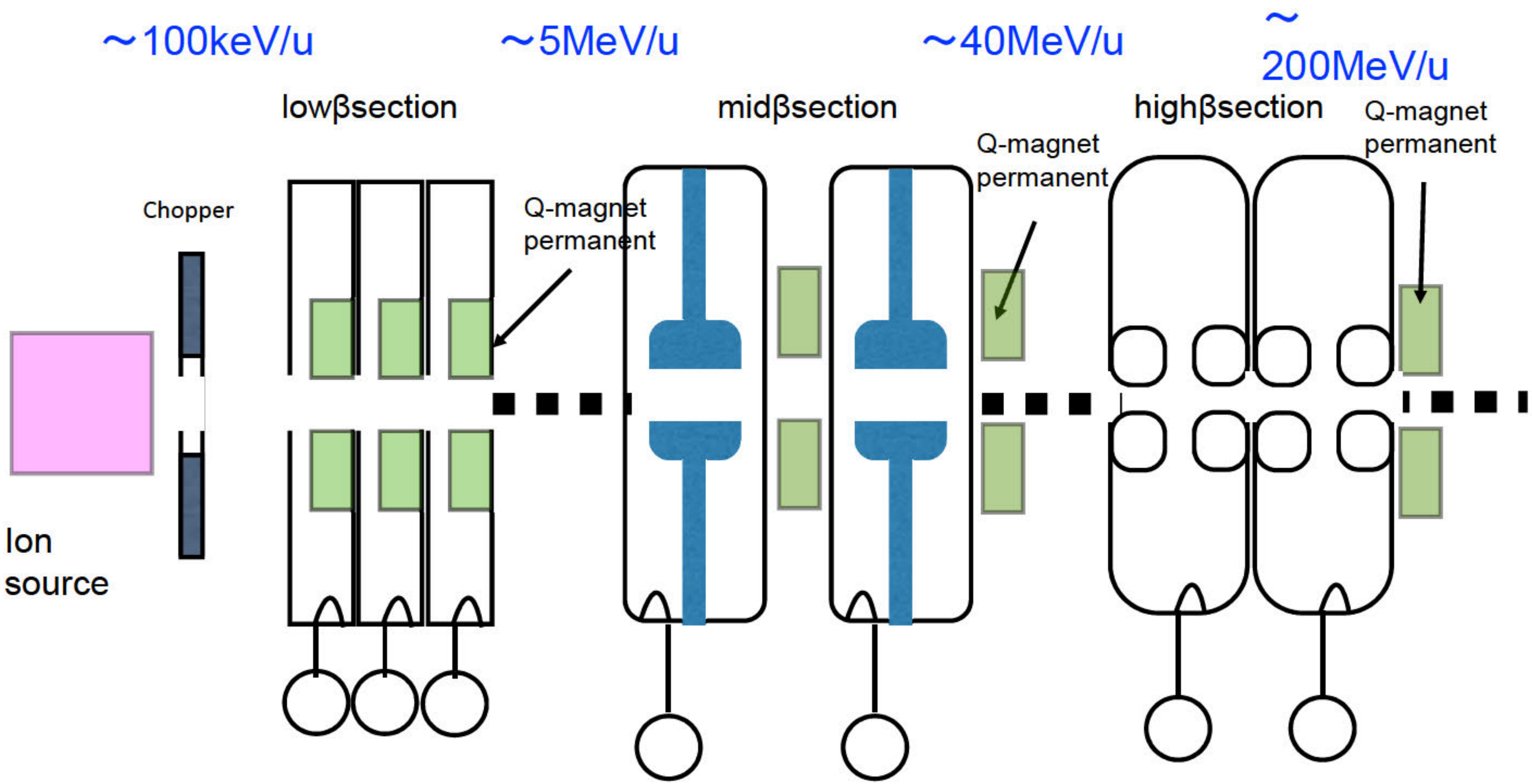


Yield of year (kg/y)	5	250	417	767

* : one year yield of LLFP transmuted by proton beam with 1000 MeV for 3 years of effectual half years

Current Progress : ImPACT2017 Model

Primary beam : deuteron
Energy : 40 MeV/u – 200 MeV/u
Beam current : 1A



4-5 : Disposal and Recycle in ImPACT

Short-lived, Highly radioactive (4%)

To be disposed of after decay for long-term storage (300 years)

Extremely long-lived, Fissionable, α emitter (5%)

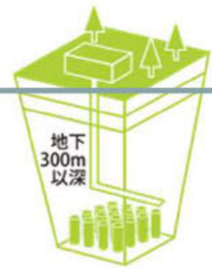
To be transmuted to short-lived by fission reaction in advanced nuclear system and disposed of

Extremely long-lived, Non-fissionable, βγ emitter (20%)

To be recycled and shorted in lifetime in ImPACT program, then disposed of

Stable or short-lived (70%)

To be disposed of as low-level radioactive waste



Content of high-level radioactive waste

All radioactive nuclide is stabilized or shortened in lifetime!

Current Progress : Consideration on disposal concept

Waste from ImPACT process can be disposed of in <u>intermediate depth</u> (several tens meters) or <u>shallow pit.</u>
However, α isotopes and part of is βγ isotopes large for such disposal. Transmutation by advanced nuclear system must be progressed.

Reduction of total radioactivity by ImPACT process

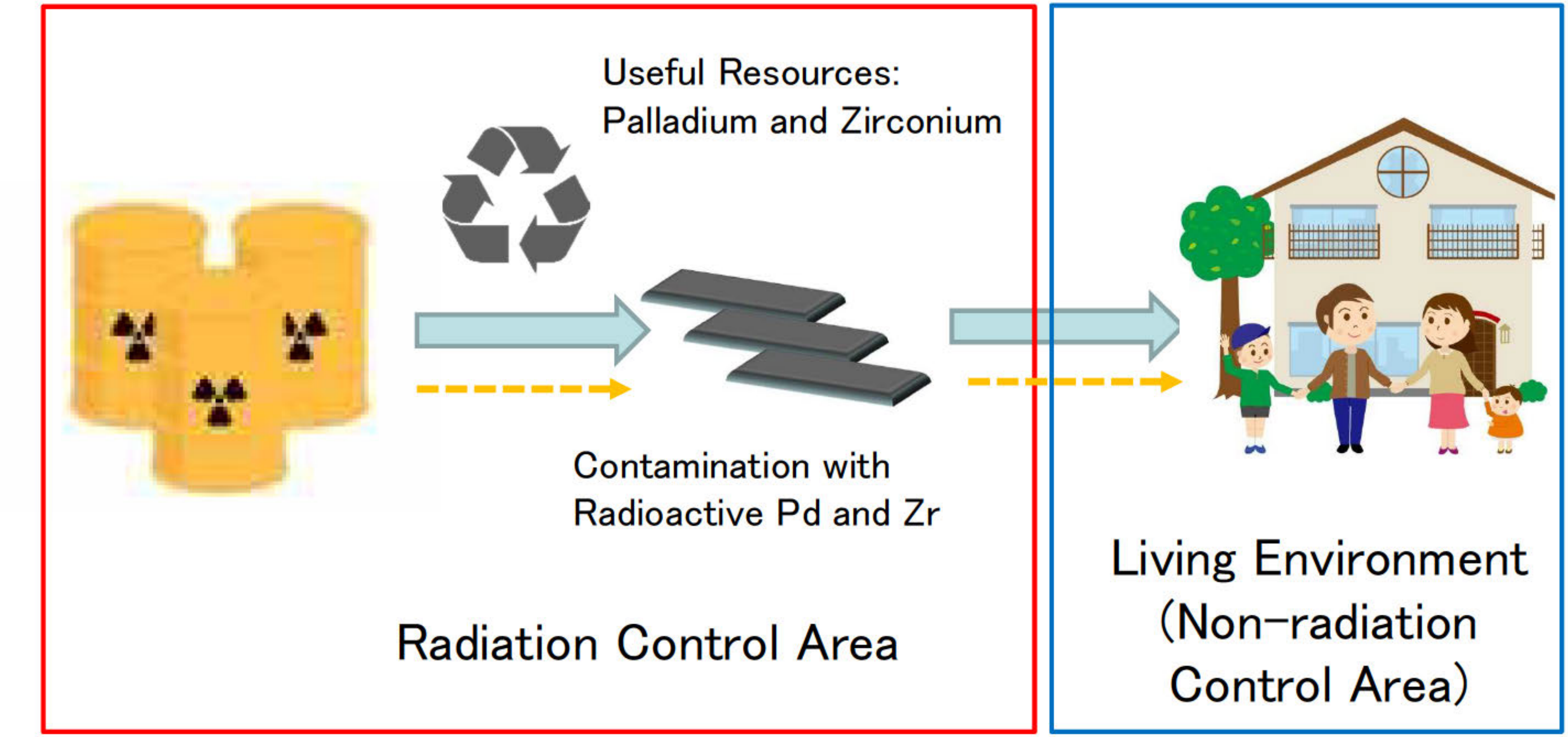
Waste	Disposal concept	βγ (Bq/t)	α (Bq/t)
Original HLW	Deep underground	2E+16	1E+14
Waste from ImPACT process	???	**1E+14**	**1E+12**

Limitation on radioactivity concentration corresponding to disposal concept

Waste	Disposal concept	βγ (Bq/t)	α (Bq/t)
Low and intermediate level waste	Intermediate depth disposal	C-14: 1E+16 Cl-36: 1E+13 Tc-99: 1E+14 **(1E+10)** I-129: 1E+12	Total: 1E+11
	Pit concept	C-14: 1E+11 Co-60: 1E+15 Ni-63: 1E+13 Sr-90: 1E+13 **(2E+13)** Tc-99: 1E+9 **(1E+10)** Cs-137: 1E+14 **(3E+13)**	Total: 1E+10
	Trench concept	Co-60: 1E+10 Sr-90: 1E+7 Cs-137: 1E+8	
Designated Fukushima waste	Isolated-type landfill	Cs-137: 8E+6~1E+8	
Clearance		Cs-137: 1E+5	

*1 Number in parenthesis is concentration after ImPACT process

Current Progress: Estimation of Clearance Levels

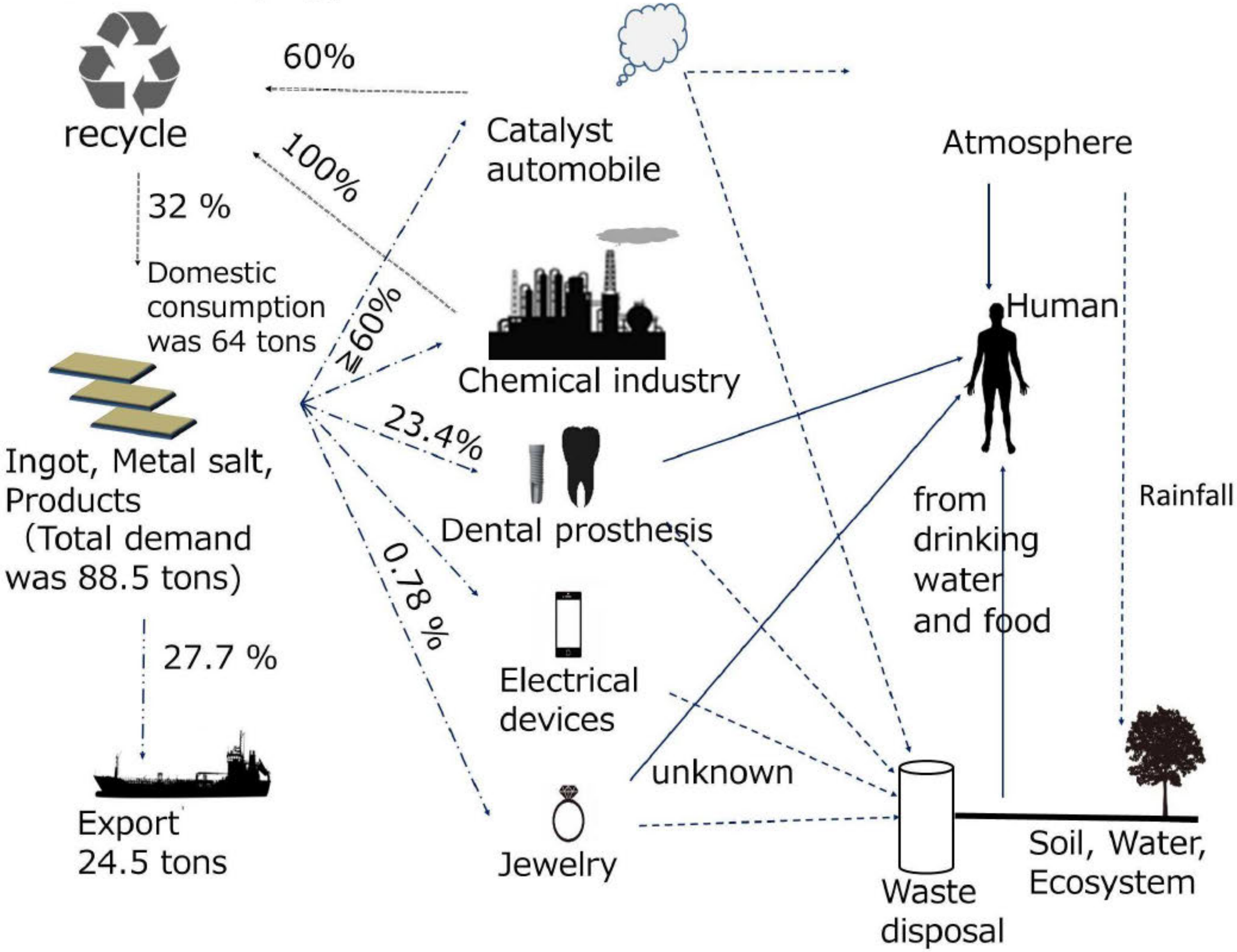


Materials can be transfered from radiation control area to non-radiation control area, if the radioactivity concentration (Bq/g) is less than the clearance level.

Current progress : Evaluating Material flows

Evaluating Material flows of Pd and possible exposure to public in Japan

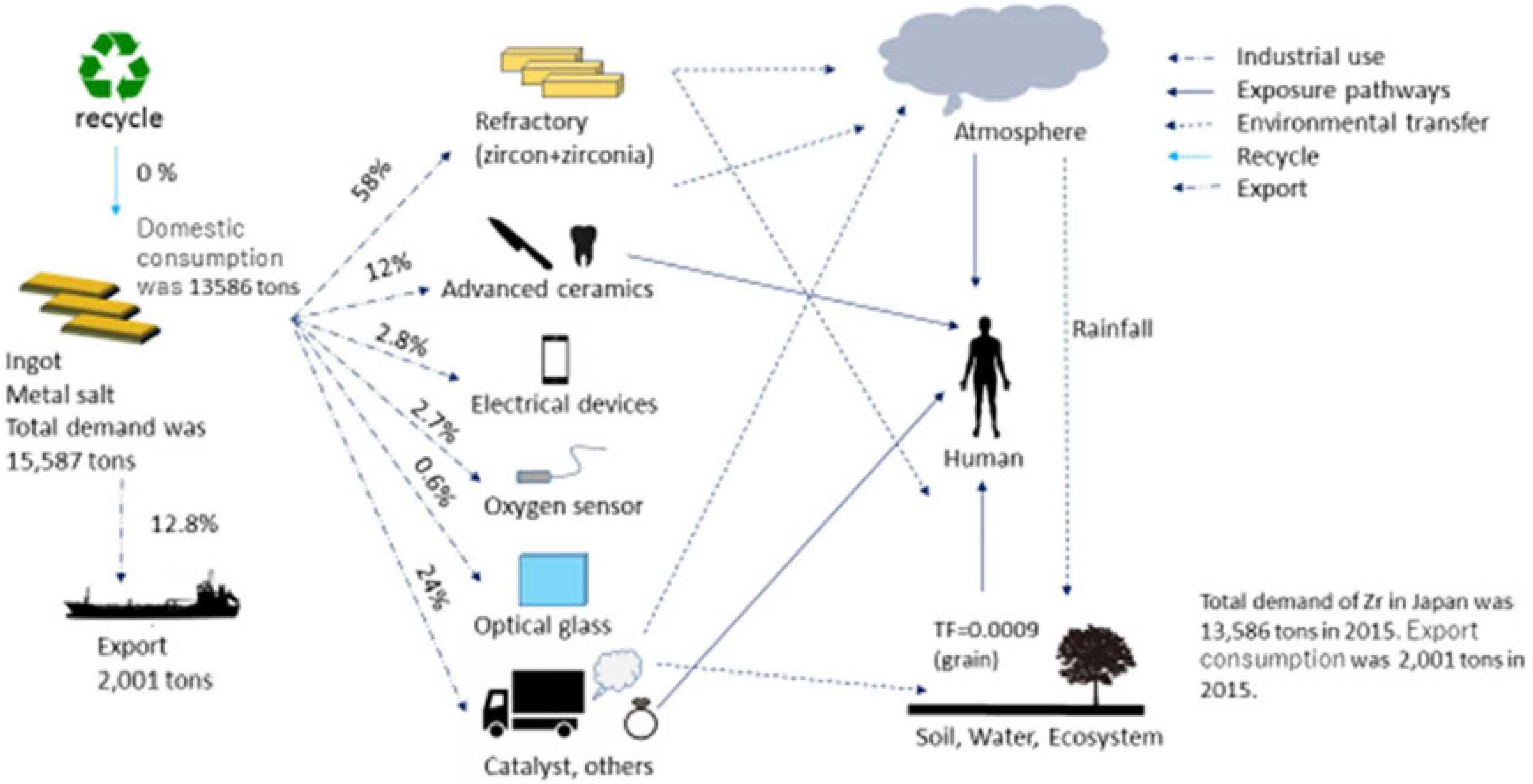
(under research and studying)



←·— Industrial use ←— Exposure pathways ←- - - Environmental transfer ←---- Recycle ←··— Export

Current progress : Evaluating Material flows

Evaluating Material flows of Zr and possible exposure to public in Japan
(under research and studying)



Current Progress : Clearance Levels

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

Exposure Pathways	Highest Dose (mSv/y	Concentration (Bq/g) which may give 10 μSv/y	Specification
Automobile Catalyst	3.3×10^{12}	3.1×10^{9}	Adult, Type S
Food and drinking water	1.3×10^{10}	7.8×10^{7}	1 year age group
Dental appliance	2.0×10^{10}	4.9×10^{7}	Adults, Type S
Occupational inhalation	3.2×10^{6}	3.2×10^{3}	Adults, Type S

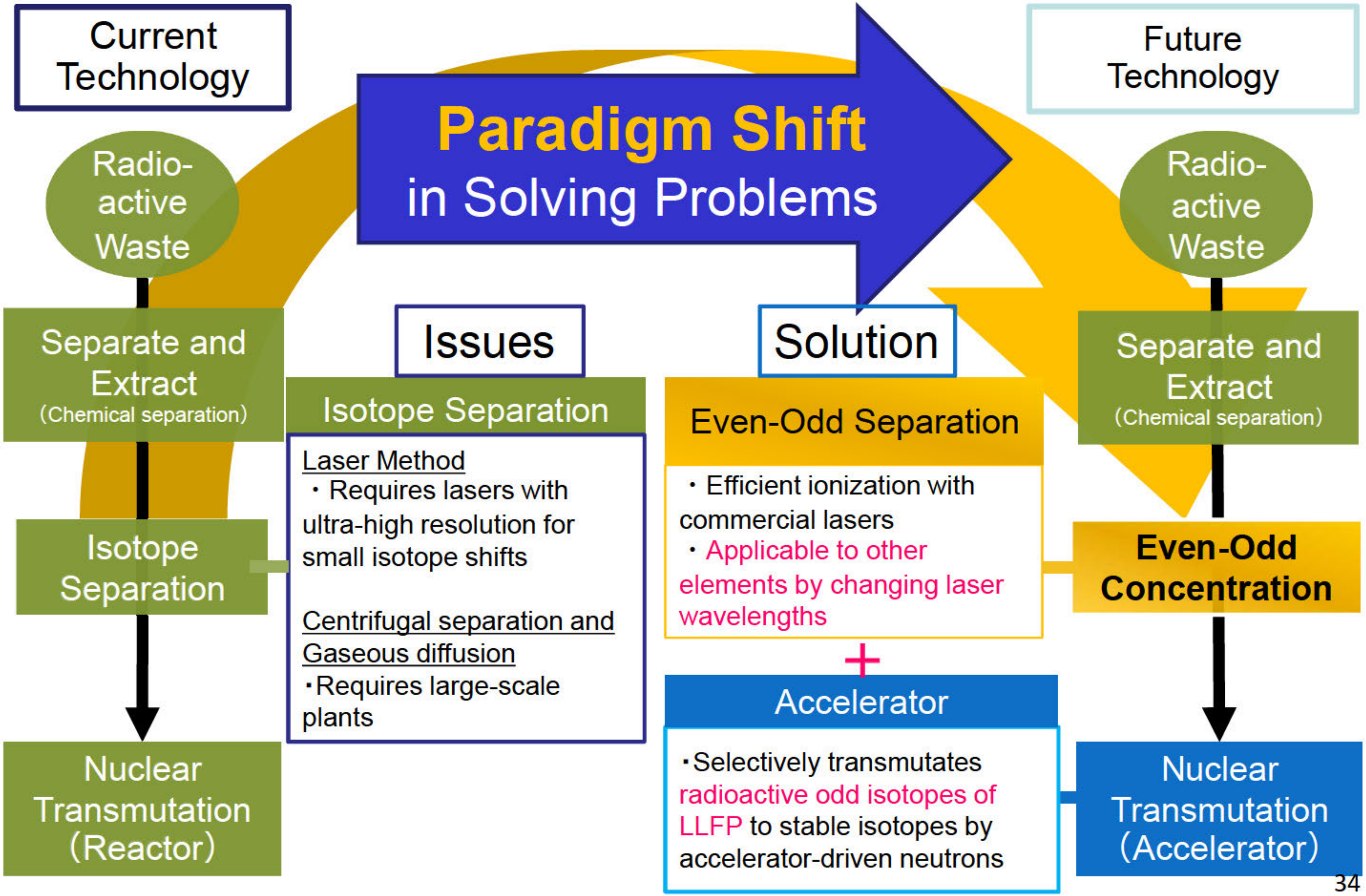
Evaluation of parameters and Estimation of other exposure pathway

↓

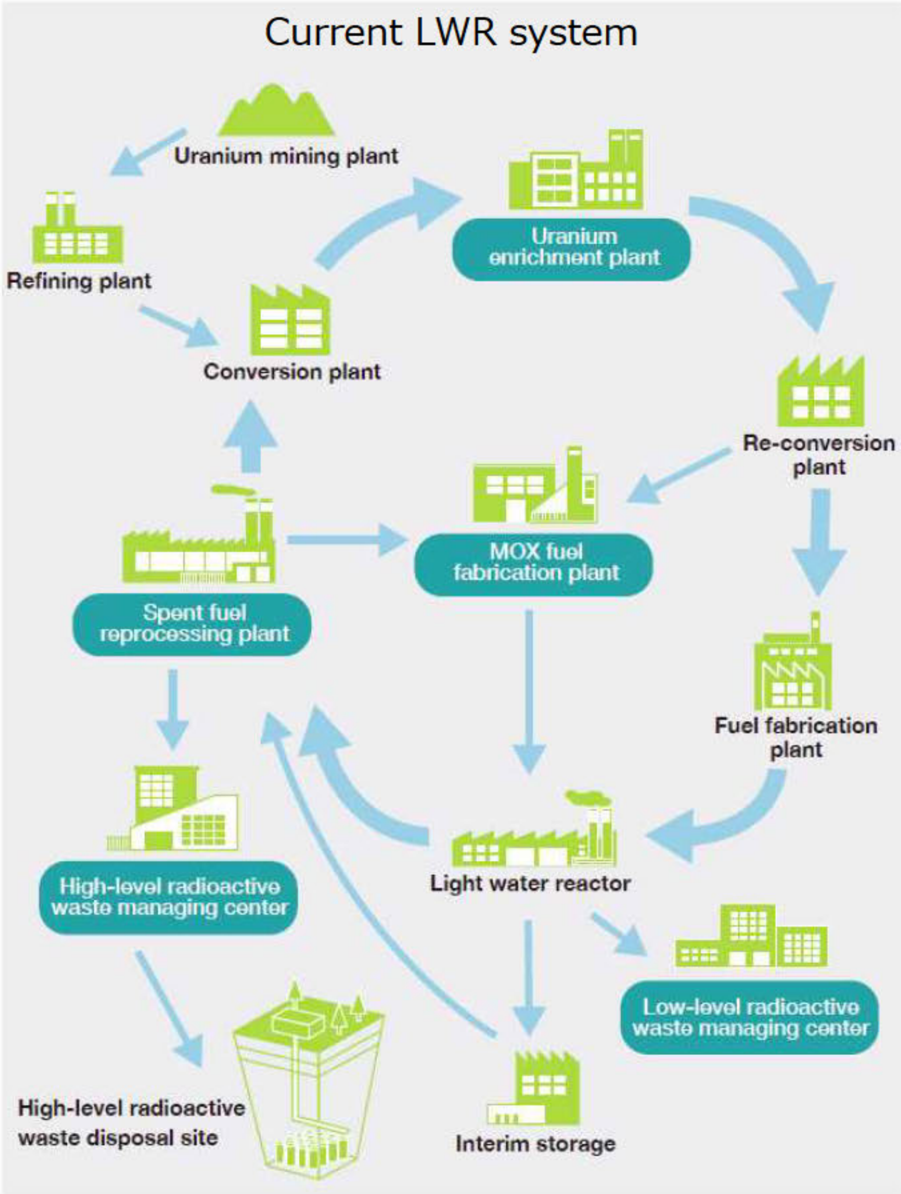
Determination of CLEARANCE LEVELS

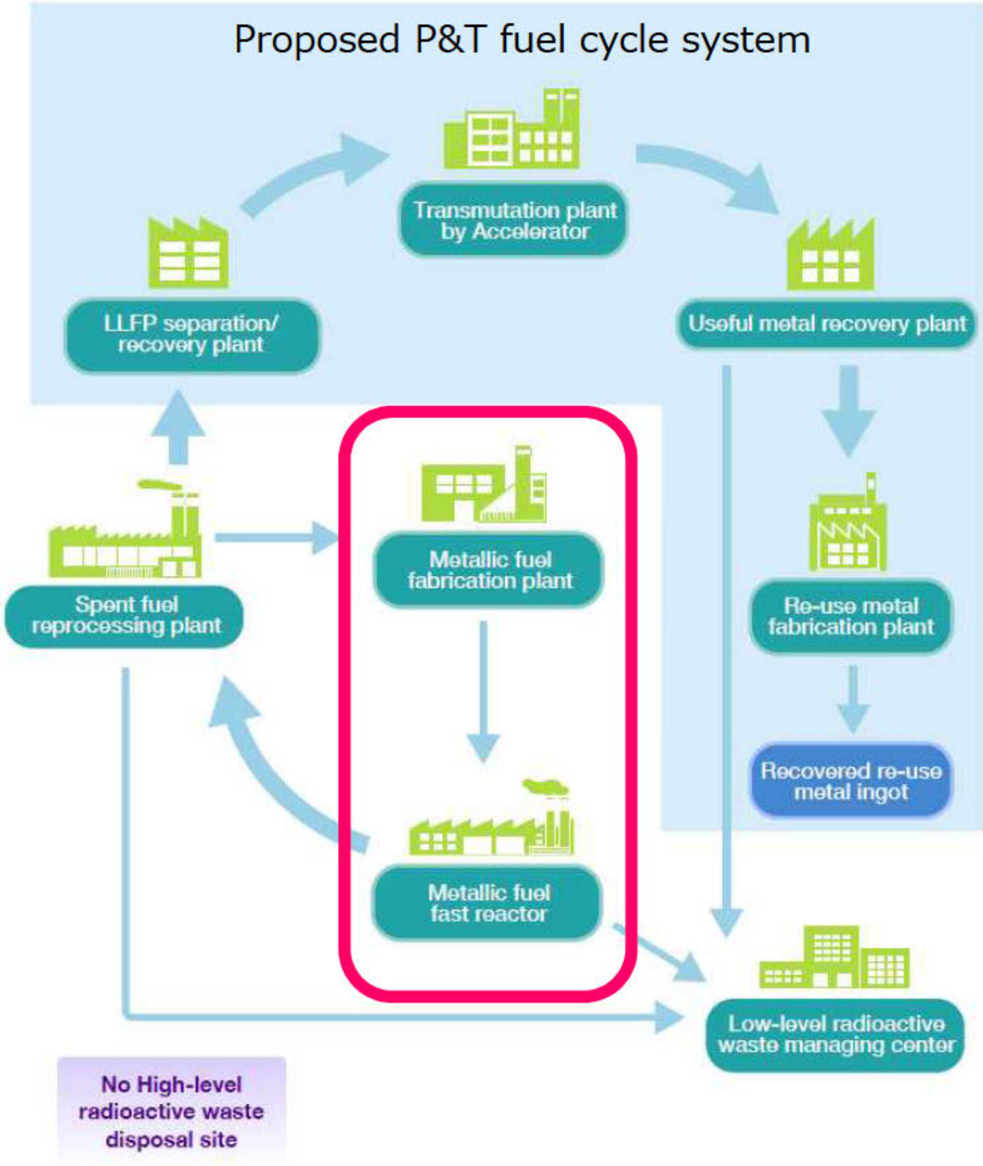
Safety Level for Reuse : Clearance Levels

Our Concept :Paradigm Shift for the Future



Our concept : Future strategy





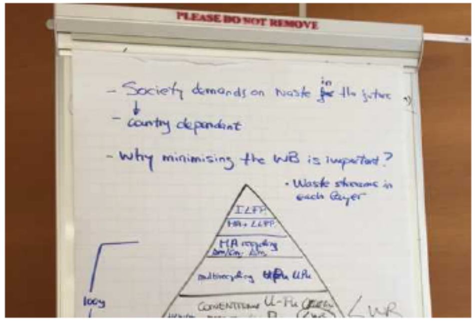
MA : Transmutation with metallic fuel FBR

Future strategy

◆ <u>Global strategy</u>

Talking about IAEA in order to embody our concept of reduction and resource recycling of long-lived fission products (LLFPs) : TECDOC will be published in early 2019 .

Tentative title of the publication: **“Waste Burden Minimisation Strategies for Existing and Innovative Nuclear Fuel Cycles”**



Technical Meeting and Consultancy Meeting on Advanced Fuel Cycle for Waste Burden Minimization in IAEA Headquarters Vienna, Austria on 21-24 June 2016,18-21 April 2017 and 17-19 Oct. 2017
IAEA, top researchers of USA, France, Russia, India, Korea and Japan talked about this concept and made a draft report.

OECD/NEA-P&T： Published a committee of recycling of radioactive wastes and publish reports

◆ <u>Patents strategy</u>

PCT application for global will be got the initiative of LLFPs P&T in the world.

◆ <u>Fusion of fundamental research and engineering technology</u>

Getting rid of the imported fundamental research and starting the first fundamental research for a clear goal in Japan

◆ **<u>The meaning of Japanese cabinet research and strategy of training human resource</u>**

Keeping and expanding all Japan framework which many researchers in various kind of research fields and organizations are joining this program, drawing young researchers with aggressive research issues and continuing the R&D for realization in the society

Summary

- ImPACT Program has started in order to be the first in the world to obtain nuclear reaction data for LLFPs, and to confirm the world's first nuclear reaction path for conversion to short lived nuclides or stable nuclides without isotope separation.

- This program should contribute through proposing innovative feasibilities (by recombining physics and nuclear technology for aiming at resolving nuclear HLW burden problems including waste-resource recycling) and overcome the Valley of Death and the Darwinian Sea by fusion with nuclear physics and nuclear engineering.

- The even/odd separation process and the high beam current accelerator is proposed and being confirmed to be feasible for LLFP P&T system by several experiments.

- It is most important to give young generation the dream which is shown in challenge for difficult research issues and solving steadily ones and overcoming ones.

Thank you for your attention !