

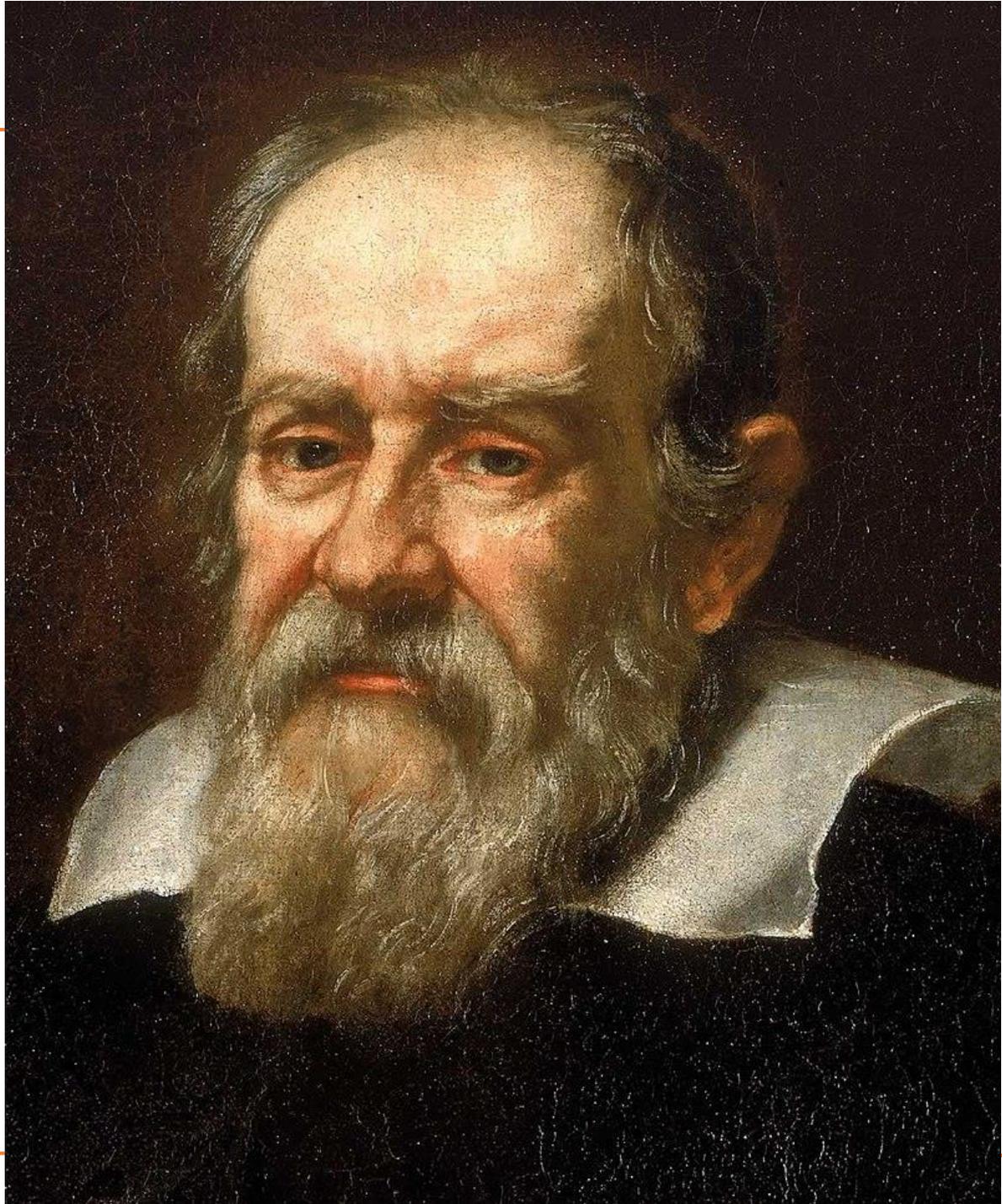
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# Multi-recycling of TRU, Tc & I with the Stabilization of Cs & Sr

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Seoul National  
University

ImPACT Symposium  
University of Tokyo  
2018. 12. 2-3.

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# World Energy Challenge

환경단체, 원전수출반대, **No Nuclear Power !**

회견 **No Wind or Solar Power !**

문재인 대통령 26일 UAE 바라카 원전 1호기 완공식 참석에 맞춰 월요일 광화문서 개최

안희민 기자 | 2018-03-24 16:17:27



환경단체가 정부의 원전수출 정책에 반대하는 기자회견을 26일 서울 광화문 이순신 장군 동상 앞에서 오전 10시 30분 개최한다. 사진=픽사베이 제공

# World Energy Challenge

Nov 24, 2018, 02:54pm

## Pro-Nuclear Activists Win Landslide Electoral Victory In Taiwan



**Michael Shellenberger** Contributor ⓘ

Energy

*I write about energy and the environment*



28 Sep 2018, 12:24 pm | [Sven Egenter](#)

## Auditors: German govt must manage Energiewende better, need CO2 price

[#Cost & Prices](#) [#Energiewende](#)

[world nuclear news](#)

[Energy & Environment](#) | [New Nuclear](#) | [Regulation & Safety](#) | [Nuclear Policies](#) | [Corporate](#) | [Uranium & Fuel](#) | \

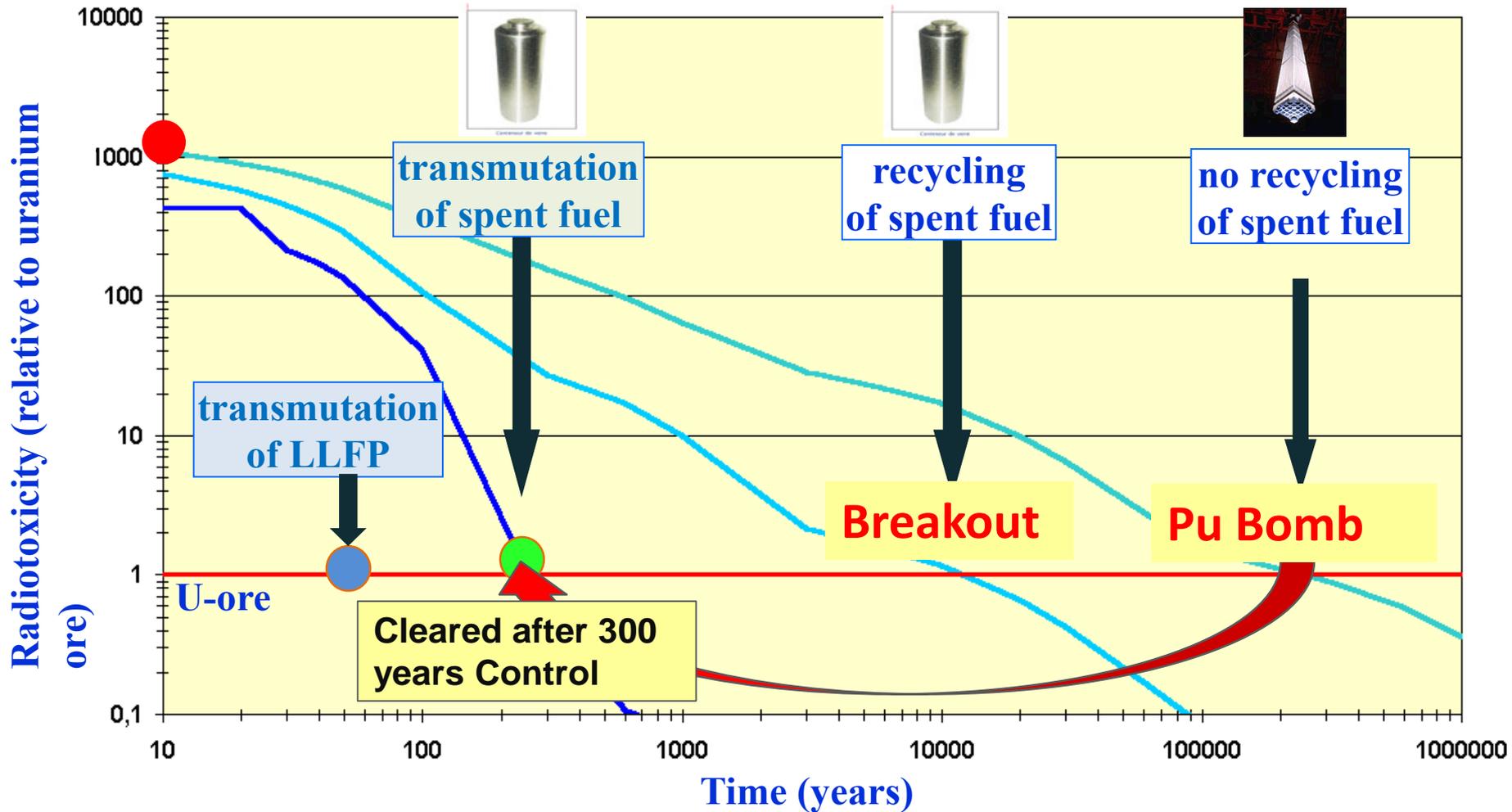
## Union of Concerned Scientists calls for policy to preserve nuclear

09 November 2018



The Union of Concerned Scientists (UCS) has called for federal and state policies in the USA to help preserve safely operating nuclear power plants that are at risk of premature closure to ensure their low-carbon energy is not replaced by fossil fuels.

# Innovative SNF Waste Burden Minimization





# SNF Repository as Potential Pu Mine

- Spent fuels, bury or eliminate? : safeguards problems

## Solutions to safeguards problems

### 1. “DIV issue”

- Provision of Design information through the whole life (all phases) of the repository
  - Inspectorates shall have access to the site to verify the Design-as-built anytime

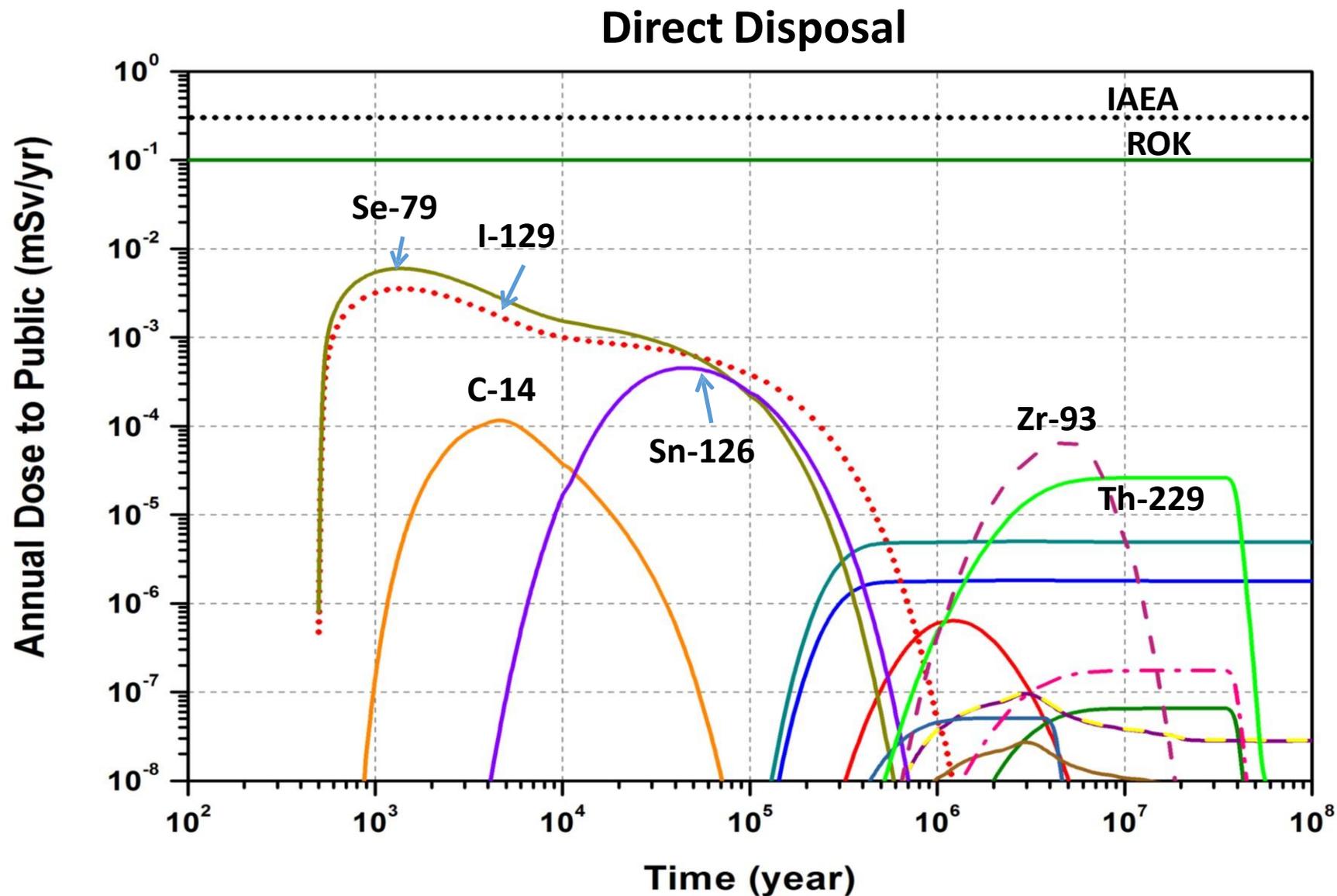
### 2. “Graveyard” issue

- It is imperative not to dispose any NM without verifying it
- Verification of all fuel and C-o-K of NM before encapsulation
- Maintaing the C-o-K of Canisters until final disposal
- R&D needed

### 3. Long term “Plutonium mine” issue

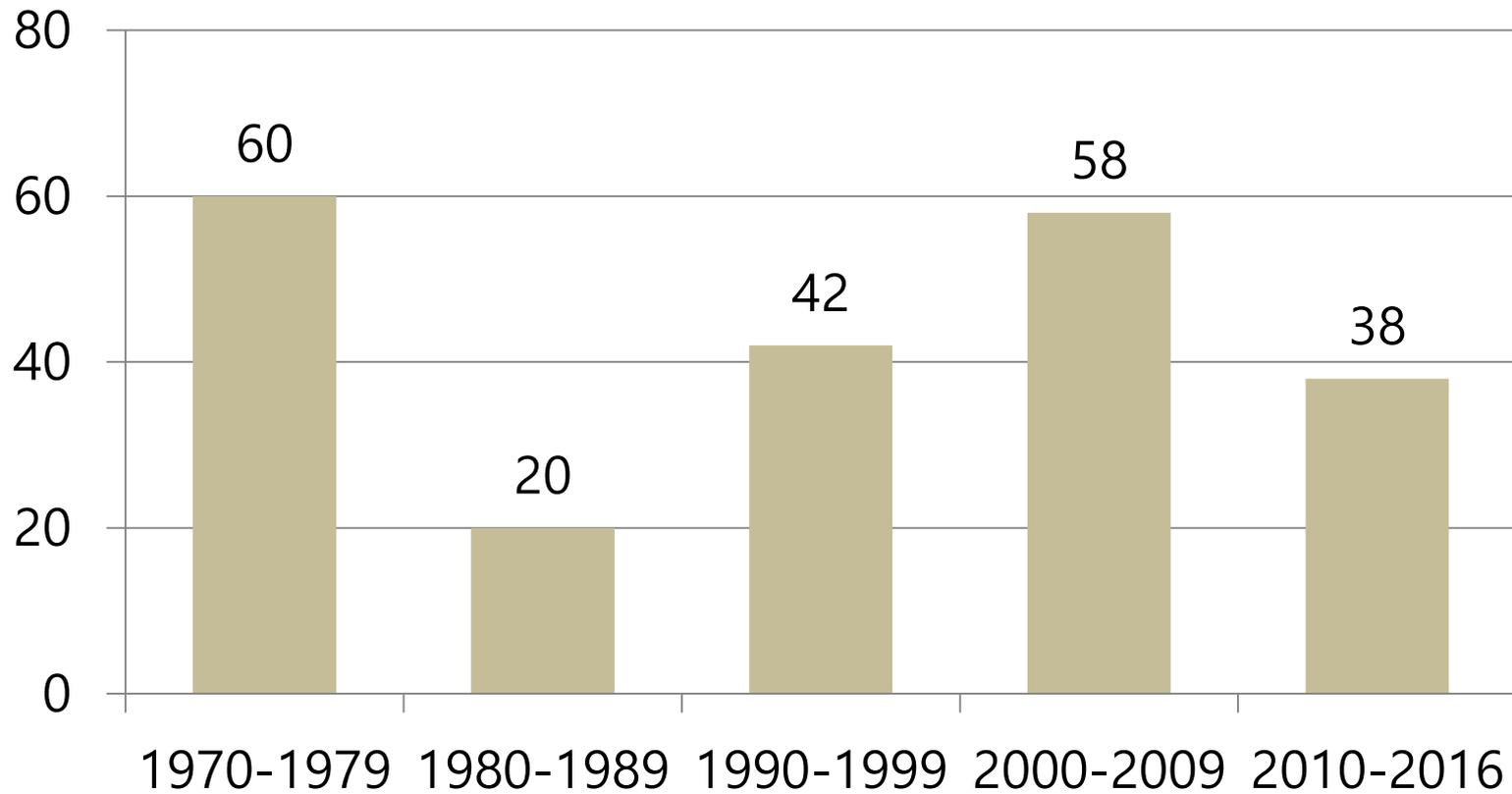
- Detecting potetial undeclared activities at later stages
  - Time scales of 10 000y !!
  - Remains for burden for next generations and societies - we need to raise our kids in a way they can take the responsibility..

# Environment-friendliness predicted by GoldSim®



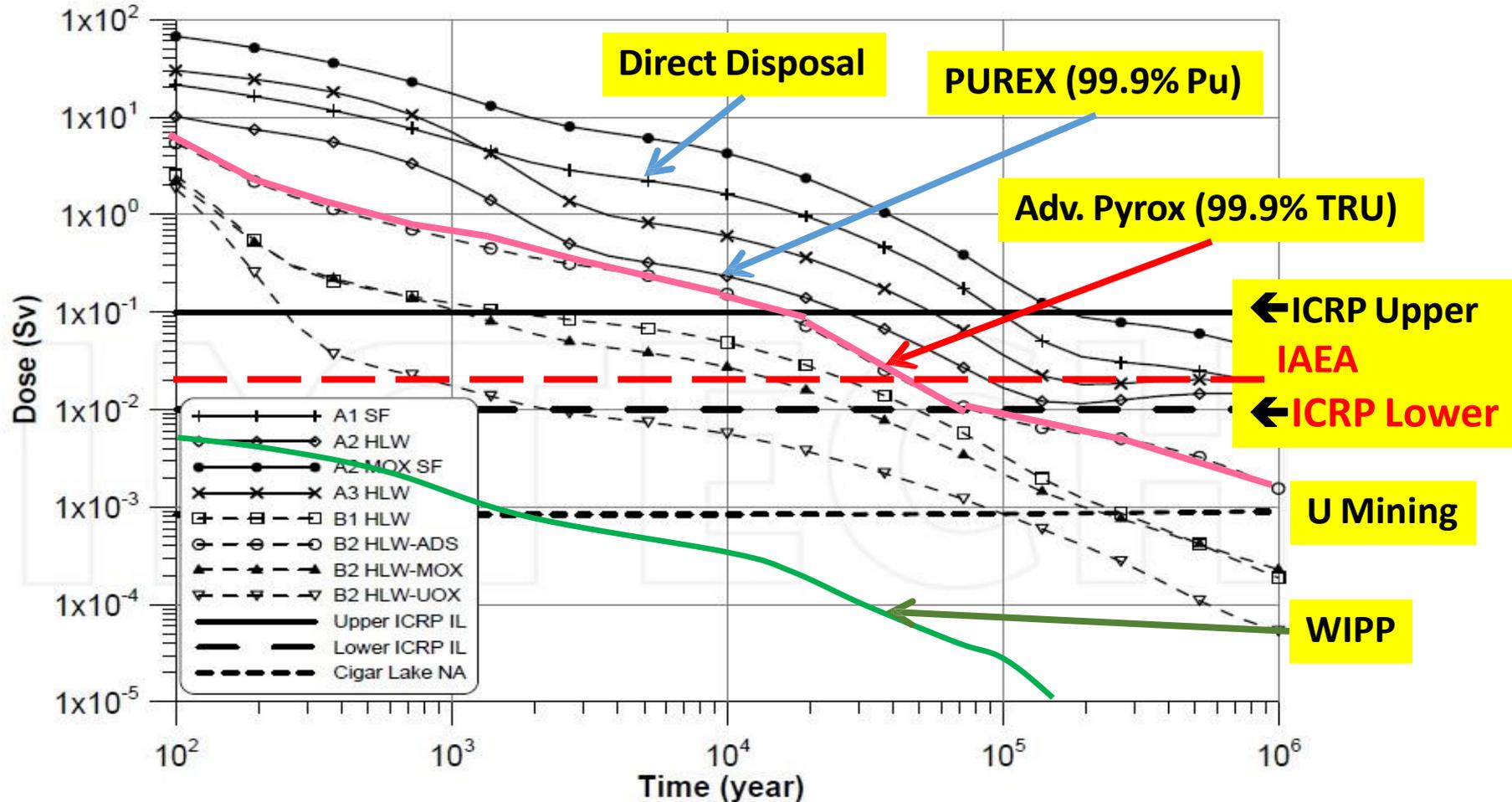
# Natural Intrusion

**The number of world earthquake ( $M \geq 7$ )**



# Human Intrusion

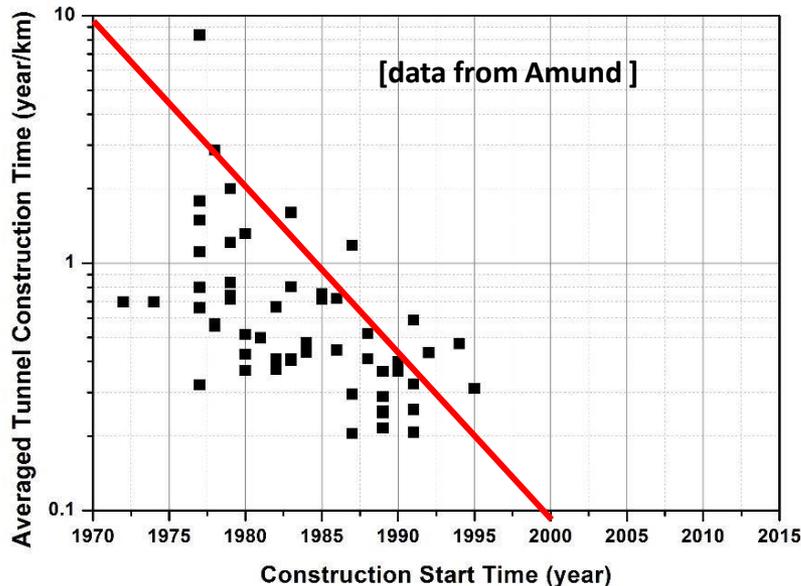
## Dose Upon Human Intrusion after the Institutional Control Period (from EU-Red Impact Study Results & SNU Study on WIPP)



# SNF Repository as Potential Pu Mine

## ➤ Geological repository: practical, sustaining barrier?

Scenarios	INFCE, 1970		Peterson, 1996
	Repository in granite	Repository in salt	Yucca Mountain
Shaft drilling	<ul style="list-style-type: none"><li>• 4 months</li><li>• 25 M\$</li></ul>	<ul style="list-style-type: none"><li>• 2~6 months</li></ul>	<ul style="list-style-type: none"><li>• 6~8 weeks</li></ul>
Tunnel excavation	<ul style="list-style-type: none"><li>• 12~18 months</li><li>• 100 M\$</li></ul>	<ul style="list-style-type: none"><li>• 12~18 months</li></ul>	<ul style="list-style-type: none"><li>• 6 ~ 12 months</li><li>• 2.5~7.4 M\$</li></ul>



## World drilling speed record:

### 1) Vertical Drill

1.43 km/day in 1997 on the Satun A-17 well by Unocal Thailand.

### 2) Horizontal Drill

2.23 km/day in 2014 by Baker

# Multi-recycling of TRU, Tc & I with the Stabilization of Cs & Sr (Level 5)

## Approach

Adv. Partitioning  
of TRU, Tc, I, Cs, Sr  
TRU Recycling  
Cs & Sr Stabilization

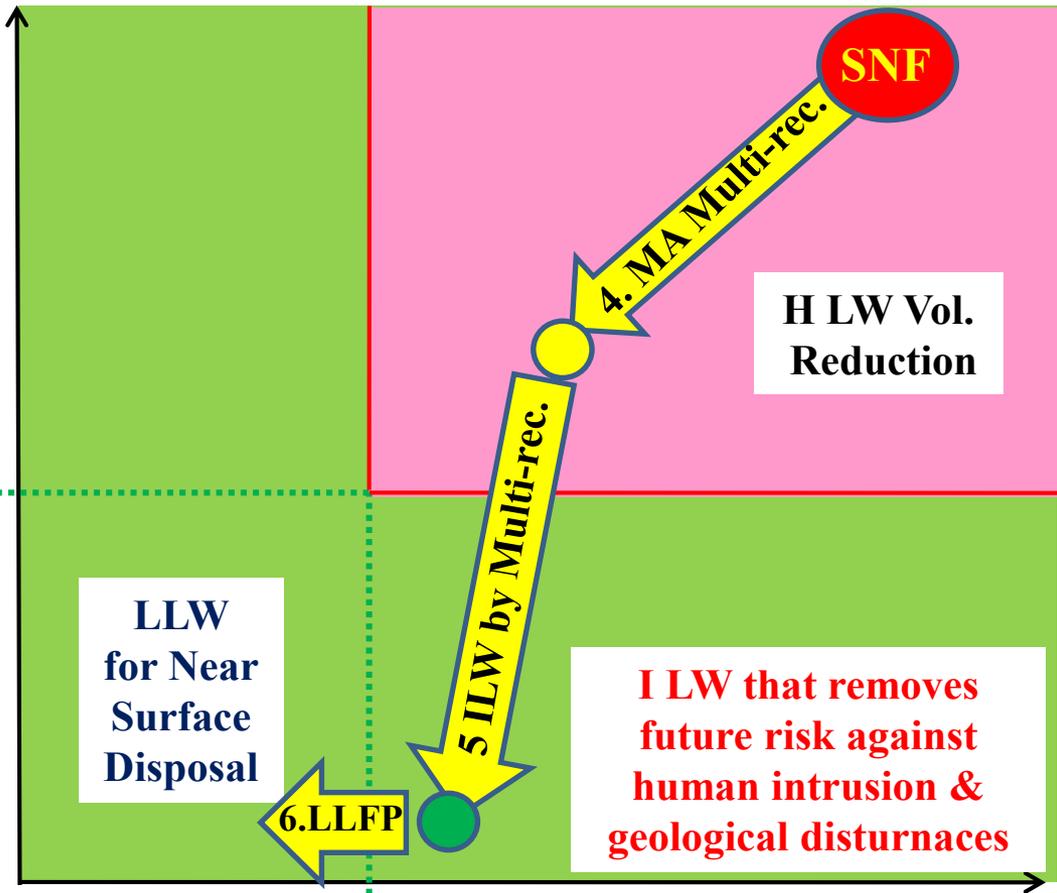
## Goal

Develop innovative pyro-partitioning technology for  $2\text{kW}/\text{m}^3$  decontamination of HLW (Level 5)

Stabilize Cs & Sr

Supply LLFP targets to innovative technology for minimizing LLFP (Level 6)

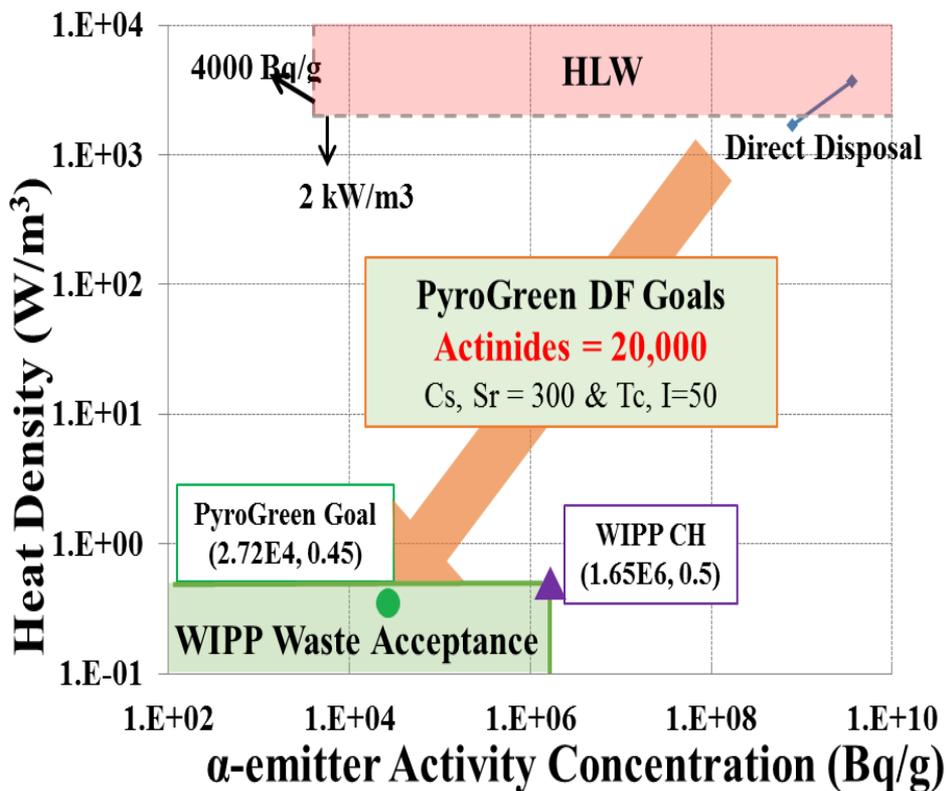
Heat  
Density



$\sim 4,000 \text{ Bq/g}$  Long-living Alpha Emitter Concentration ( $t_{1/2} > 20\text{yrs}$ )

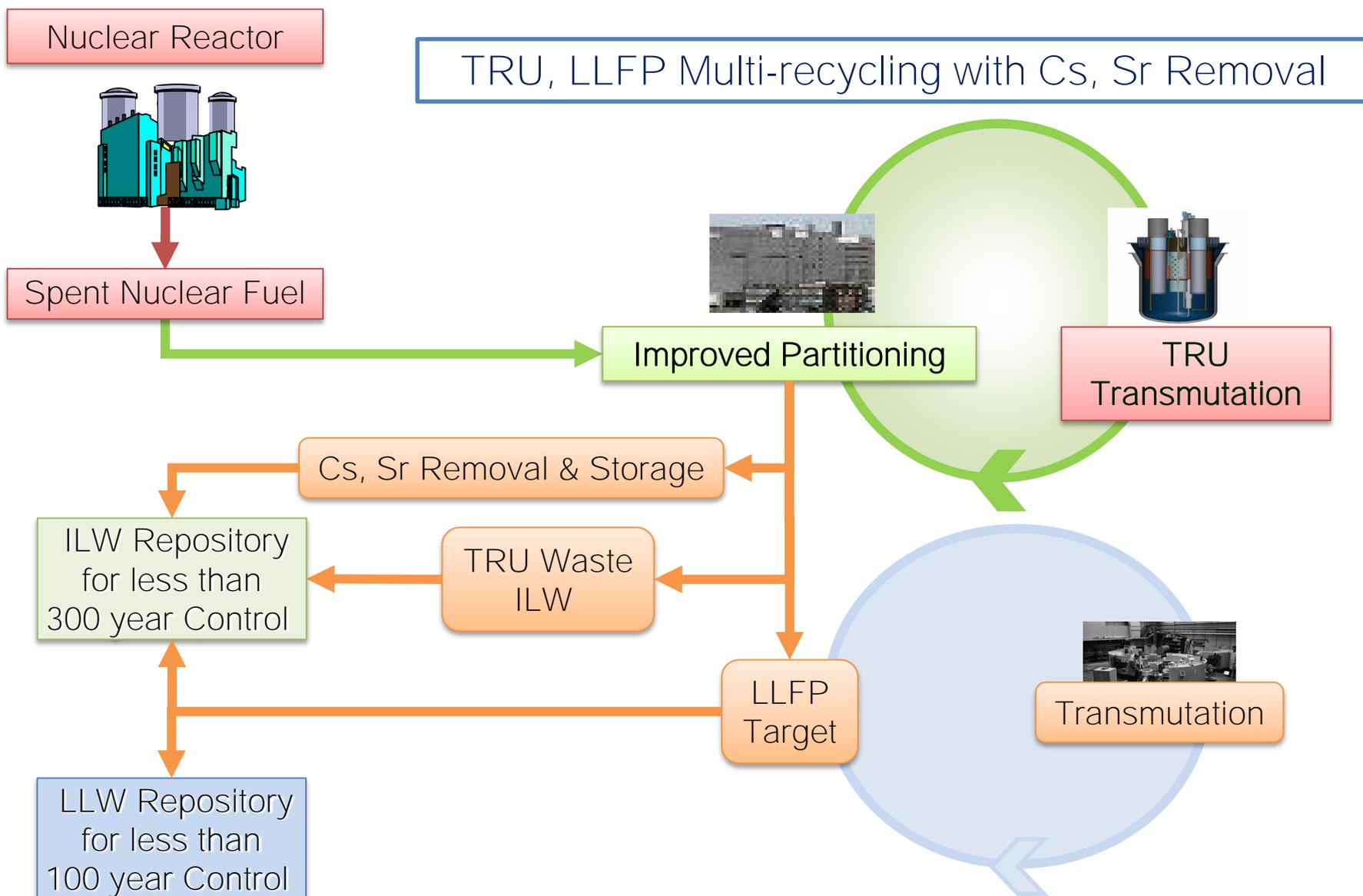
# Immediate Goals: Eliminating HLW

- Intermediate Level Waste (ILW) which can satisfy WIPP waste acceptance criteria can remove the risk



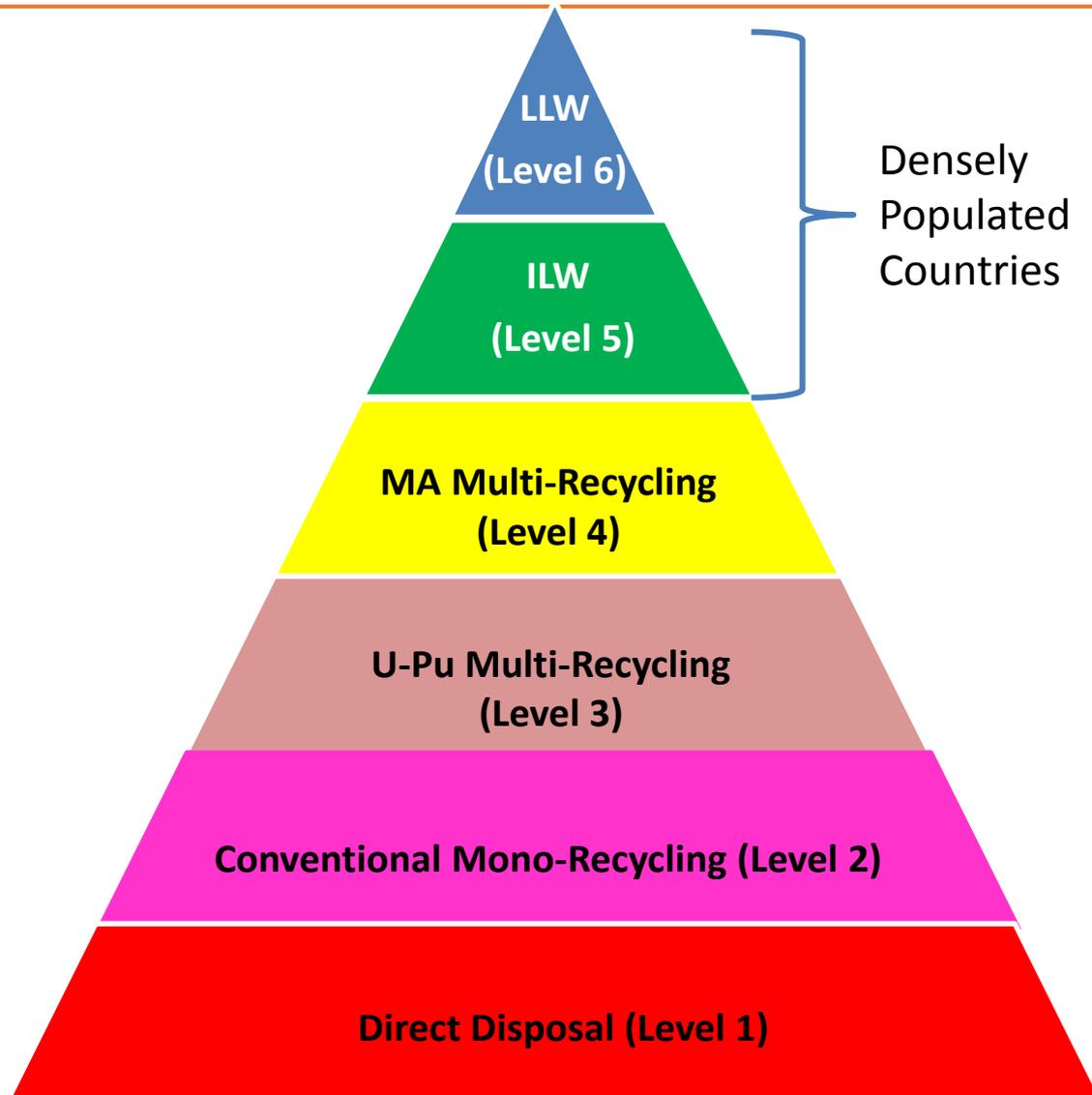
Uncertainties	Nation	Criteria	Safety and Security in Densely Populated Region		
			Direct Disposal	Pyro-Green	Adv. PUREX/Pyrox
Safety (leaching and migration)	IAEA	0.3 mSv/yr or $10^{-5}/yr$	Satisfy	Satisfy	Satisfy
	US EPA	<10,000 years : 15 mRem	Satisfy	Satisfy	Satisfy
	ROK	0.1 mSv/yr	Satisfy	Satisfy	Satisfy
Safety (IHI)	IAEA	20 mSv	Not Satisfy	Satisfy	Not Satisfy
	ROK	$10^{-6}$	Not Satisfy	Satisfy	Not Satisfy
Security (CHI)	IAEA	No burden for future	Not Satisfy	Satisfy	Satisfy

# Global Partnership for Spent Nuclear Fuels : Nonproliferation and Security



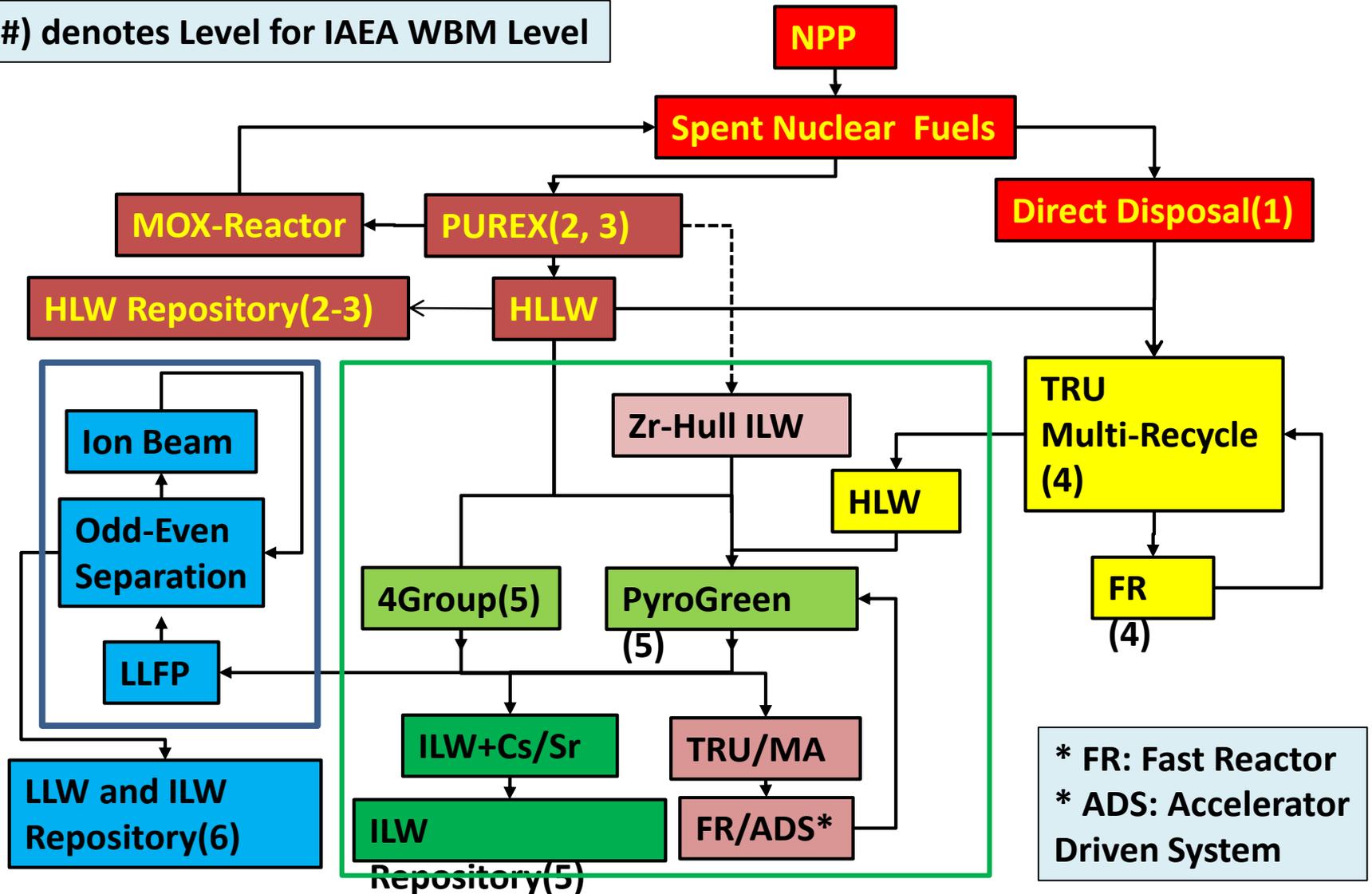
# Innovative SNF Waste Burden Minimization

6	Innov. P&T of LLFP & Turning into ILW or LLW
5	Imp. P&T of TRU with Cs, Sr, Tc, I Separation for Turning into ILW
4	Adv. Aqu.-Pyro. & FR & Small HLW
3	Multi-cycle PUREX & Reduced HLW
2	Once through PUREX & Reduced HLW
1	Direct Disposal of HLW. One through cycle



# Levelized Approach for SNF WBM (IAEA)

(#) denotes Level for IAEA WBM Level

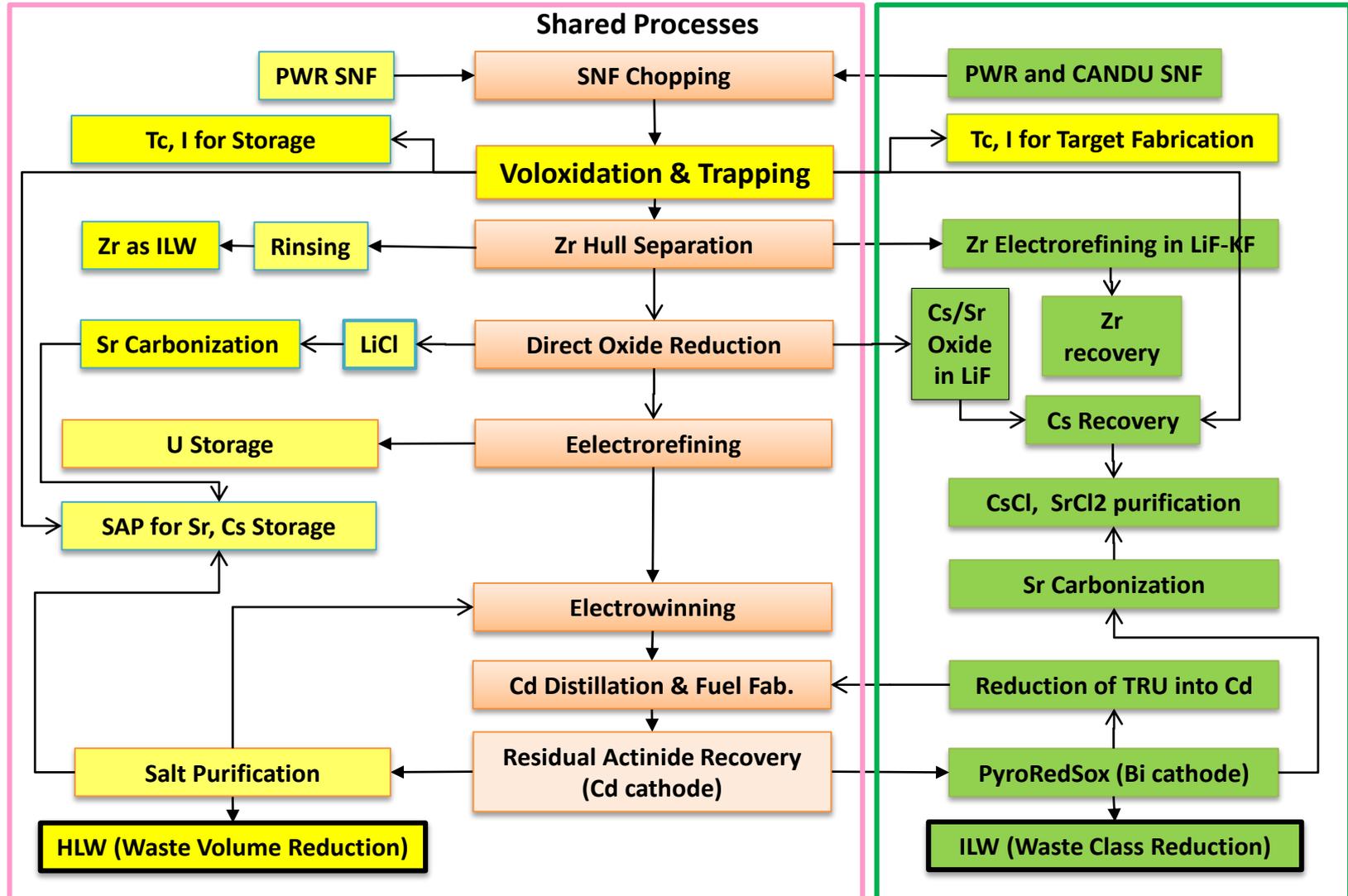


\* FR: Fast Reactor  
 \* ADS: Accelerator Driven System

# Pyroprocess Level 4 & 5: Tc & I Voloxidation

## Level 4 Pyroprocessing : KAERI Case

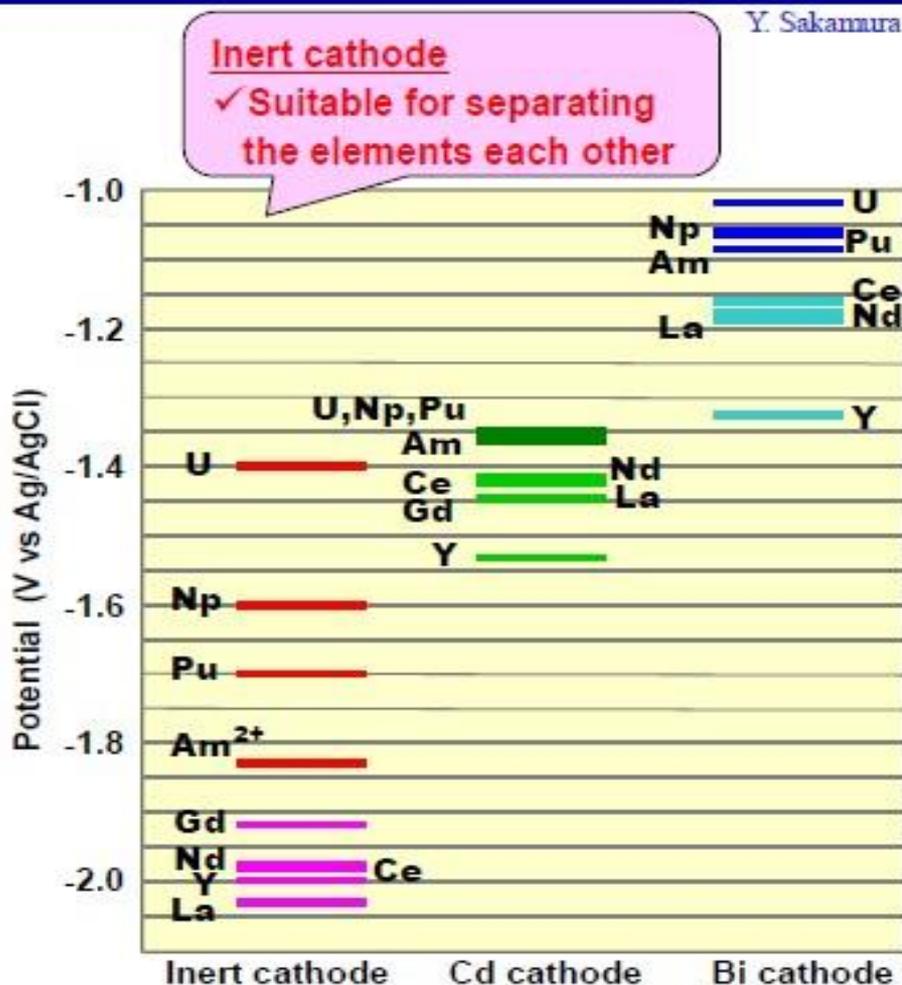
## Level 5 Pyroprocessing: PyroGreen Case



# Cd for Pyro Level 4 vs. Bi for Pyro Level 5

## Redox Potential at Various Cathodes

Y. Sakamura *et al.*, Proc. GLOBAL '99, August 29-Sept. 3, 1999, Jackson Hole, USA.



### Cd cathode

- ✓ Capable to collect all actinides together.
- ✓ Easy to remove Cd by distillation.
- ✓ Compatible with Fe material.

Table Separation factor with respect to Nd

	Inert cathode	Cd cathode	Bi cathode
U	$2.0 \times 10^{11}$	25	1300
Np	$2.1 \times 10^7$	16	200
Pu	$2.7 \times 10^5$	18	130
Am	710	10	67
Y	0.37	0.0053	0.0012
La	0.090	0.28	0.59
Ce	0.78	0.76	2.0
Nd	1	1	1
Gd	14	0.25	

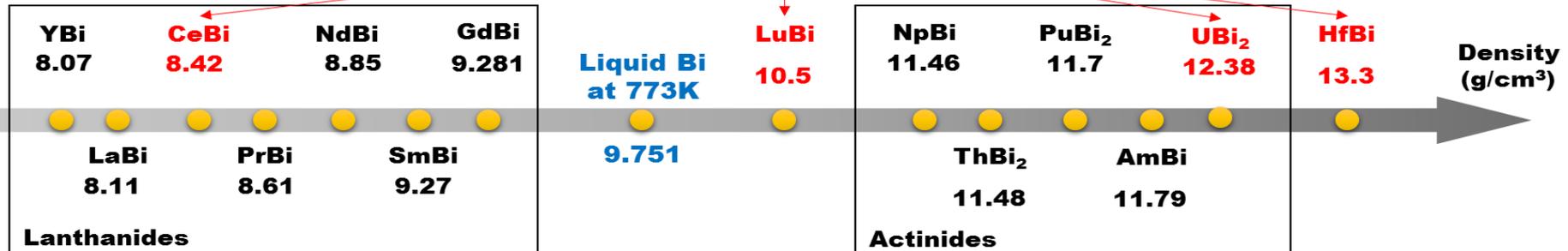
### Bi cathode

- ✓ Separation of actinides from lanthanides is easier for Bi than for Cd.

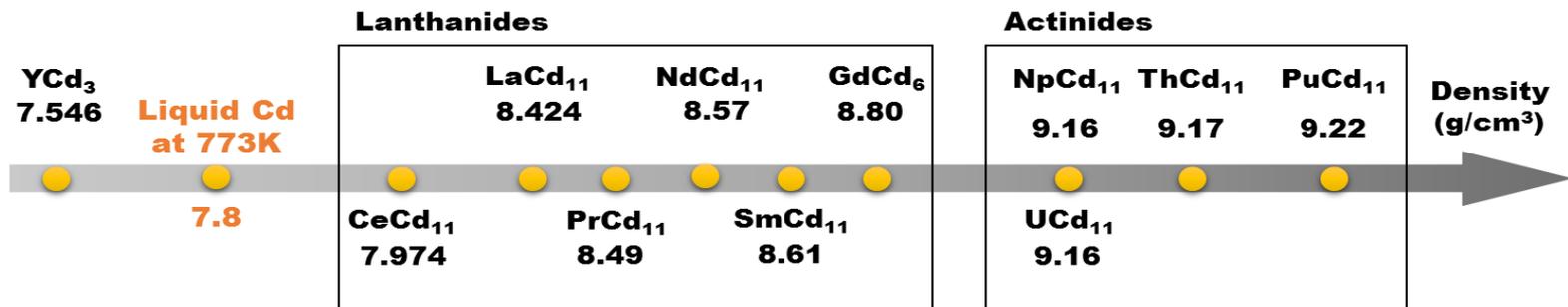
Fig. Redox potential of  $M^{3+}/M$  couple in LiCl-KCl eutectic at  $500^\circ\text{C}$ ,  $X_{M^{3+} \text{ in Salt}} = X_{M \text{ in Cd}} = X_{M \text{ in Bi}} = 0.001$ .

# Densities of Bi, Actinides and Lanthanide

Investigated to show density separation in this study

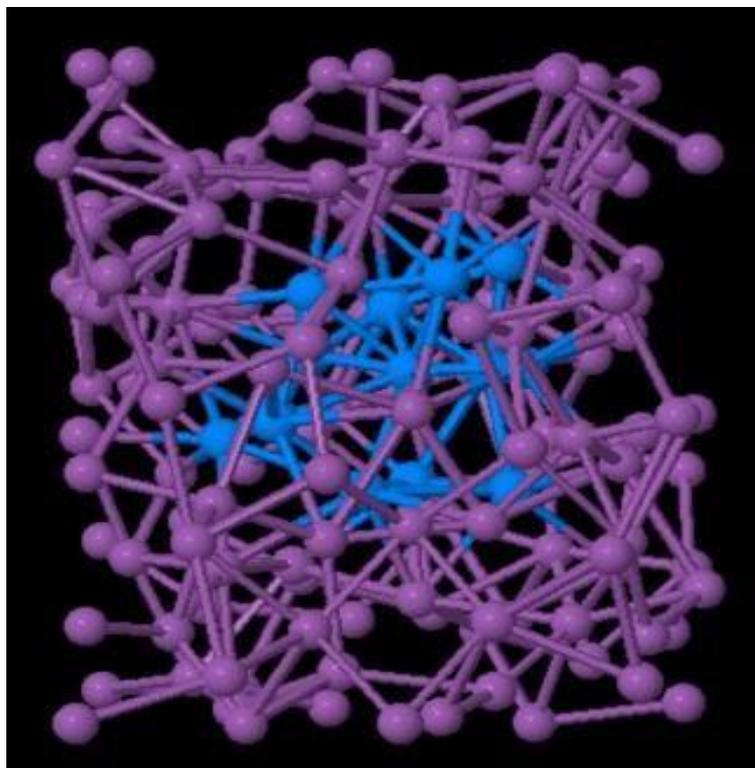
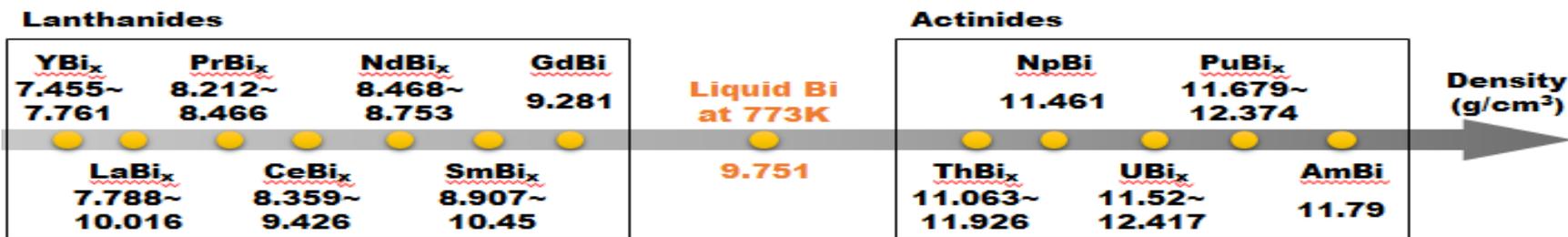


- Density distribution of Cd and Cd-M intermetallics (M=An and Ln)



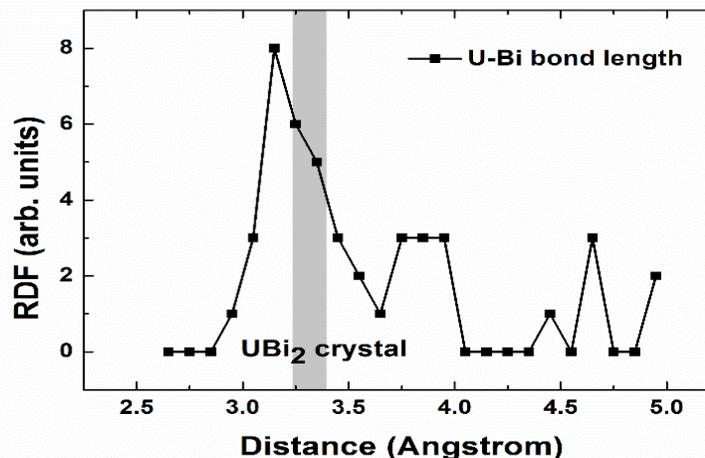
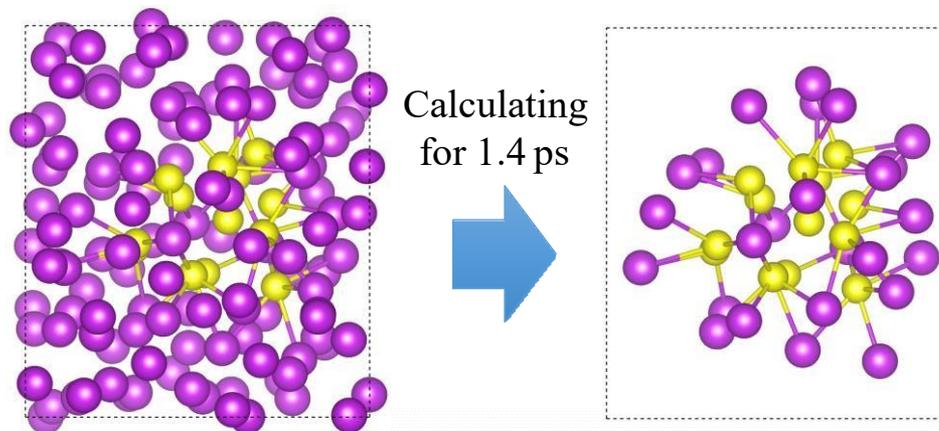
- Unlike Cd, the density of Bi at 500°C lies between lanthanides and actinides.
- Solubility of An and Ln is low enough (~1.2 at.%), to form IMC

# Densities of Bi, Actinides and Lanthanide

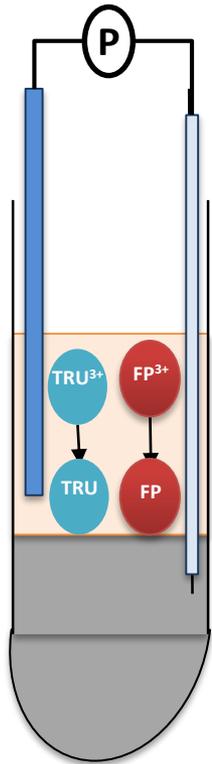


**U<sub>13</sub> nanoparticle formation**

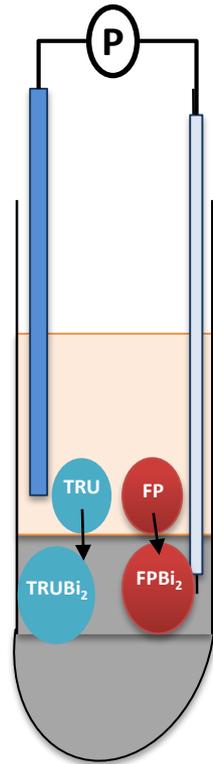
- Ab-initio modeling on Bi-U formation



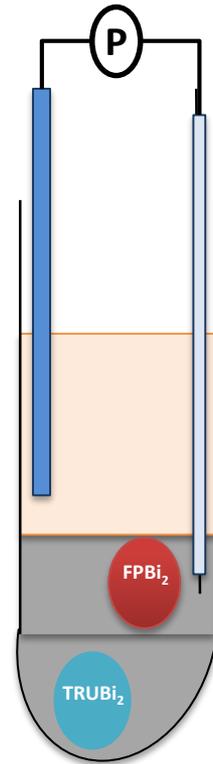
# Densities of Bi, Actinides and Lanthanide



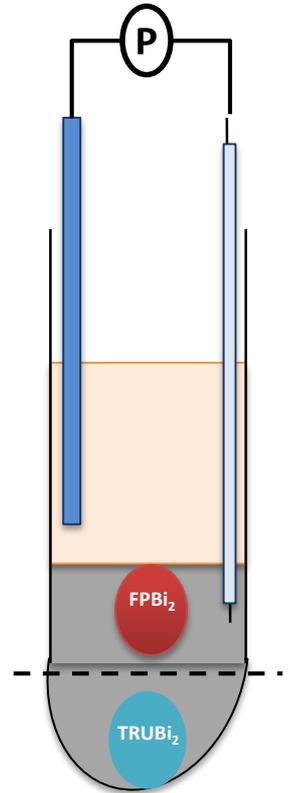
Electrodeposition at Bi surface



Dissolution and intermetallic formation



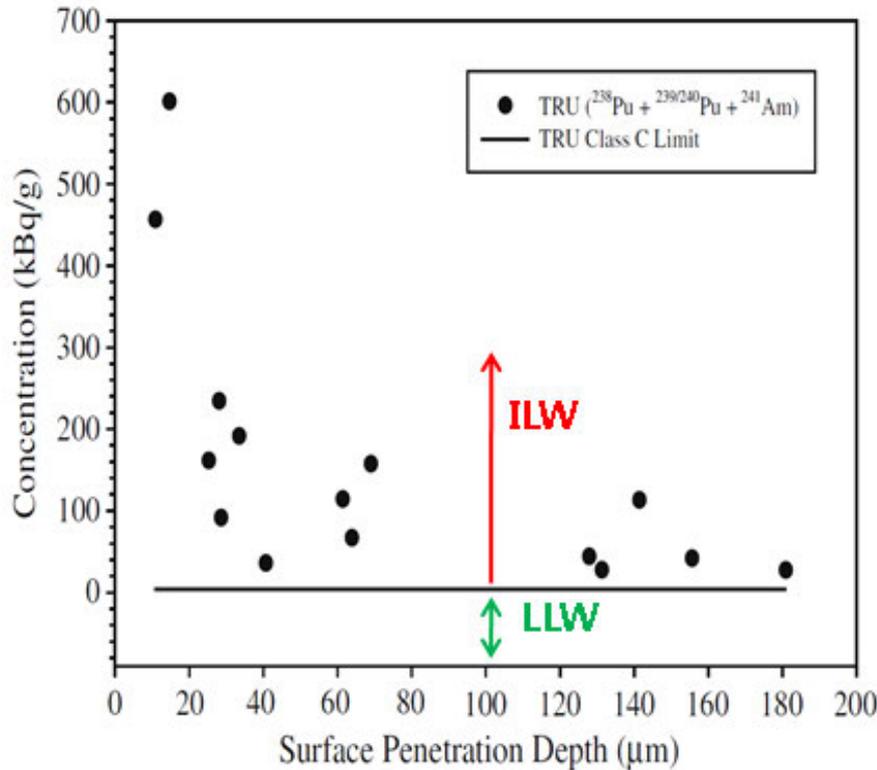
Intermetallic density based distribution



Intermetallics Separation by Density

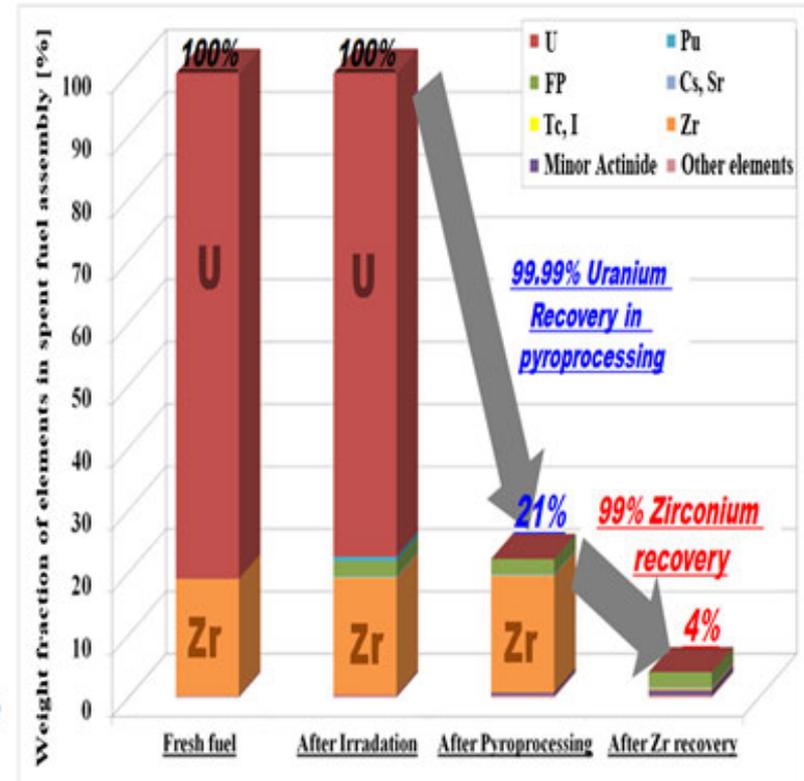
# Zr-Hull Electrorefining for ILW Volume Minimization

## TRU and FP inside irradiated cladding



(Tracy S. Rudisill, 2009)

## ILW Reduction



# Pioneering Study by Dr. Fujita Group

## ❖ Fujita et. Al- Toshiba Corp.- Study (2005) – BWR Channel Box [1]

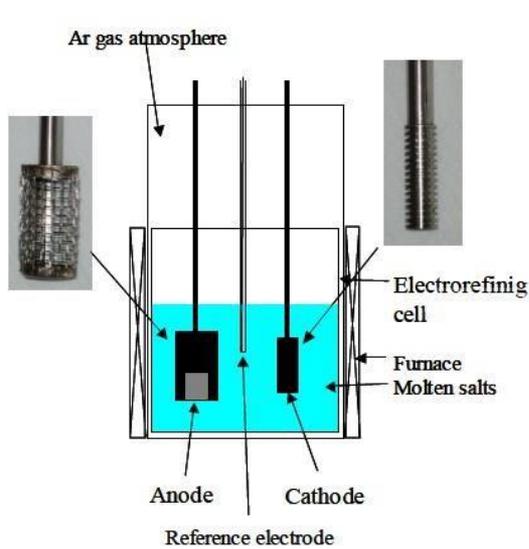


Fig1. The Schematic experimental apparatus for electrorefining

**Table 1** Electrorefining test conditions

Molten salts	LiCl-KCl-ZrCl <sub>4</sub> -LiF
Anode	Stainless steel
cathode	Low carbon steel
Temperature (K)	923
Anode current density (A/cm <sup>2</sup> )	0.1

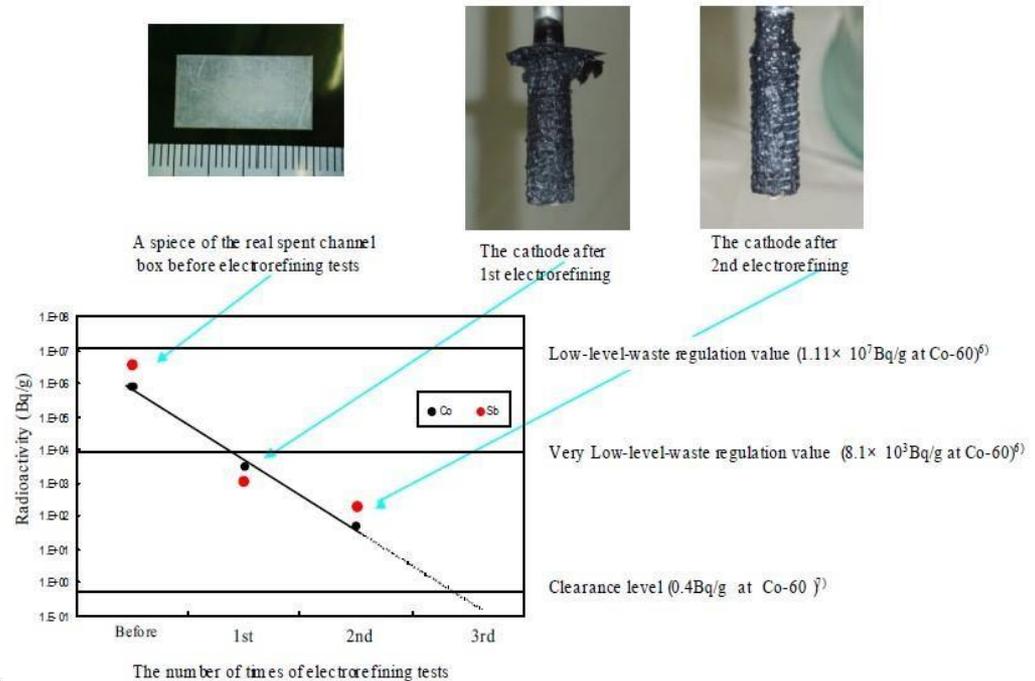


Fig2. The results of electrorefining tests in molten salts

- After the Electrorefining, DF of the Co  $\cong$  200/Step

- **Operating temperature is still high and fluoride corrosion could also occur.**

Ref [1] R.Fujita, et al., DEVELOPMENT OF ZIRCONIUM RECOVERY PROCESS FOR ZIRCALOY CLADDINGS AND CHANNEL BOXES FROM BOILING WATER REACTORS BY ELECTROREFINING IN MOLTEN SALTS, ICAPP 2005

# Cs & Sr Separation and Stabilization

- U.S.A. R&D on Cs & Sr Separation from Aqueous Processes
  - Chlorinated Cobalt Dicarbolide.Polyethylene Glycol (ChCoDiC/PEG)
    - 99% recovery
  - Fission Product Extraction (FPEX)
    - 99.9% recovery

- Russia's Pilot Facility (UE-35) for Cs & Sr Extraction at RT-1

- ChCoDiC extractant
- 50,000 kCi removed
- Recovery not reported

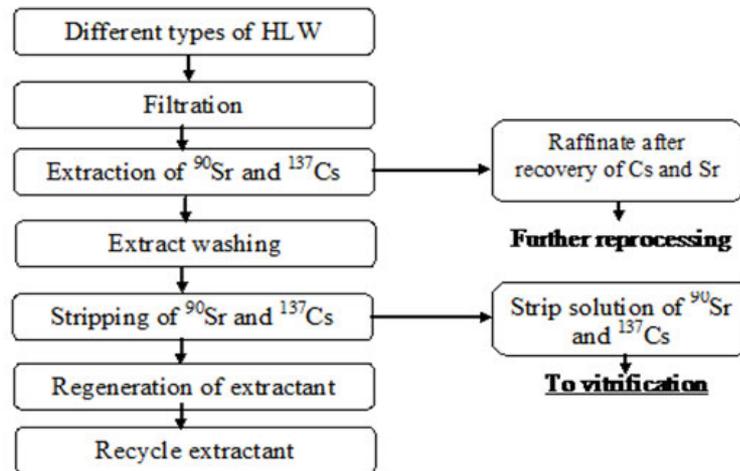


Fig.1. Key extraction flowsheet for reprocessing of HLW with the use of ChCoDiC at industrial facility UE-35 in "Mayak" PA.

# Cs & Sr Separation and Stabilization

## ➤ India's Post PUREX Process

- HLLW Extraction into heat-generating group and alpha-emitters
- Cs & Sr Stabilization for Long-term Storage
- Vitrification of 200,000 Ci of Cs in Borosilicate & Interim Stabilization
- Cs-pensil for Blood Irradiation Sources
- Sr recovered and used for Clinical Trials

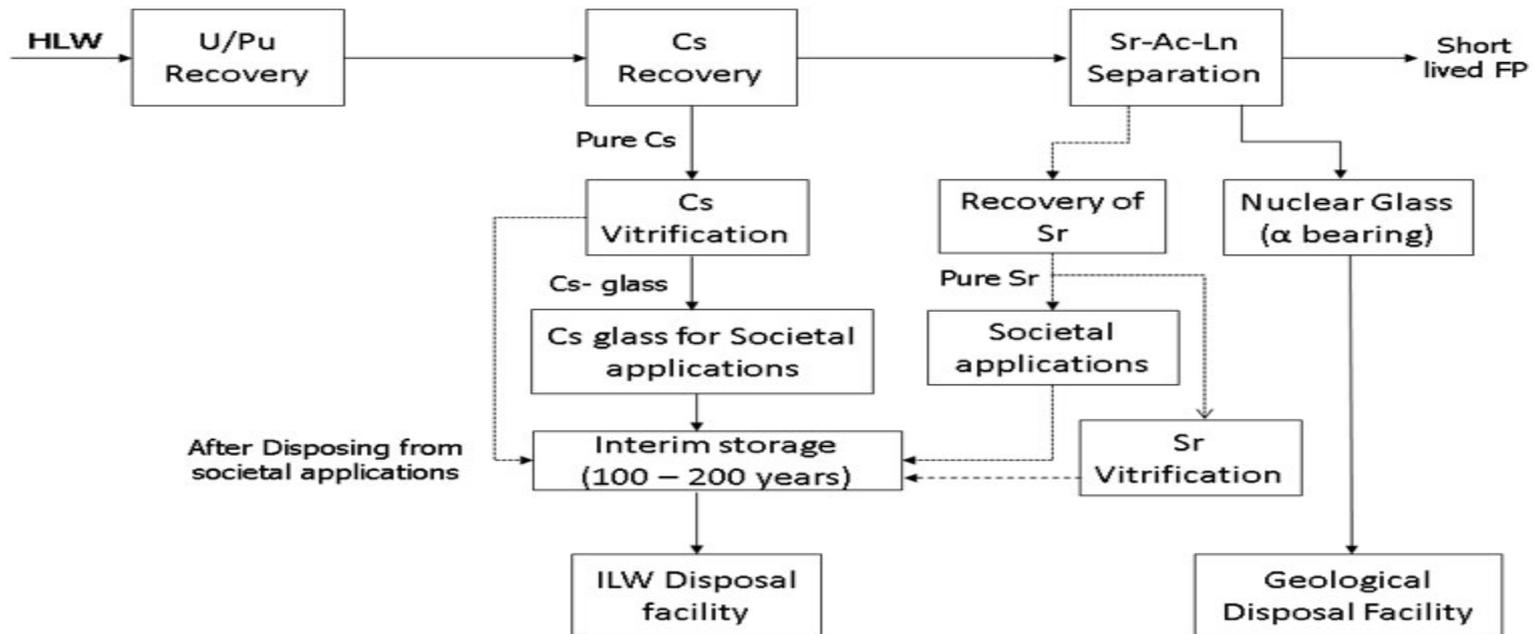
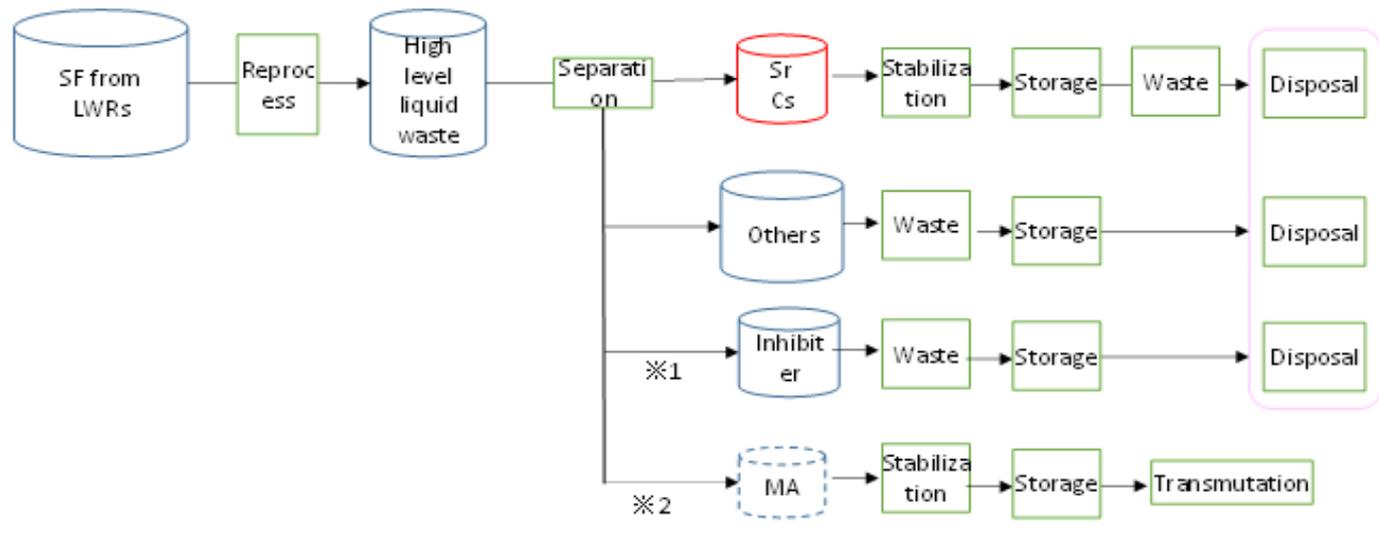


Fig 2. Reference flow sheet for fission product partitioning

# Cs & Sr Separation and Stabilization

## ➤ JAEA's Post-PUREX Process for Cs & Sr

- Cs & Sr Extraction from HLLW
- Stabilization Vitrification in Borosilicate & Interim Stabilization
- Vitrification in Heat-resistant Calcined Zeolite for Long-term Storage



## ➤ JAEA's 4-Group Extraction for MA, Cs & Sr (Improvement)

(Ref. 4. OECD/NEA NSC/WPFC/DOC(2012)15, SNF Flowsheet, June 2012)

# Cs & Sr Separation and Stabilization

- ROK – US Pyroprocess
  - Cs from Voloxidation up to 98%
  - Sr from Carbonate process up to 98.4%
- PyroGreen (SNU)
  - Additional “Salt Zone Refining-Multi-Pass”
  - Cs and Sr up to 99.7%,
- Summary: Cs & Sr

Recovery, %	TRU	Tc	I	Cs	Sr
JAEA 4-Group : Level 5 Hydro (Decontamination Factor)	99.998 (50,000)	96.6 (30)		99.99 (10,000)	99.99 (10,000)
SNU PyroGreen :Level 5 Pyro (Decontamination Factor)	99.995 (20,000)	98 (50)	98 (50)	99.7 (300)	99.7 (300)
ROK-US Pyroprocess : Level 4 (Decontamination Factor)	99.9 (1,000)	98 (50)	99 (100)	>99 (>100)	98.4 (100)
US Aqueous (FPEX) (Decontamination Factor)				99.9 (1,000)	

# Summary

Type		Recovery, %(DF)				
		TRU	Tc	I	Cs	Sr
Hydro-process	Industrial PUREX (1995+)	>99.9 Pu (>1,000)				
	UREX+3(US)	99.997 (30,000)			high	High
	ACSEPT (France)	>99.9 (>1,000)	>99 (>100)			
	ACSEPT(UK)-continuous	>99.99 Pu (>14,000)				
	4-Group (Japan)	99.998 (50,000)	96.6 (30)		99.99 (10,000)	99.99 (10,000)
Pyro-process	Pyroprocessing (ROK)	99.9 (1,000)		99 (100)	98 (50)	99 (100)
	PyroGreen (ROK)	99.995 (20,000)	98 (50)	98 (50)	99.7 (300)	99.7 (300)

# Deep Isolation in Shale or Welded Tuff Layers

Professor Richard Muller <https://www.deepisolation.com/>

