Separation and recovery of LLFPs from high level wastes

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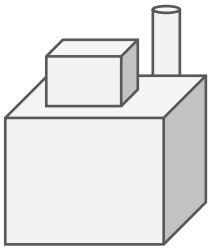
- Project 1 Separation and recovery of Pd, Se, Cs and Zr from the wastes from reprocessing plant
- Proposed separation process of the LLFPs from HLLW
- R&D status of the separation technique for the LLFPs
- Summary

Project 1 Separation and Recovery Technique

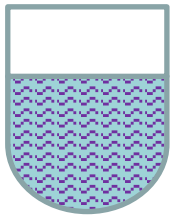
Purpose：Efficient separation and recovery of LLFPs techniques

Target LLFP
(Se-79, Zr-93, Pd-107, Cs-135)
(FP Pd: mainly 104, 105, 106, 107*, 108, 110)
***β-radioactive**

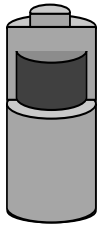
Current process :
ImPACT new process :



Reprocessing Plant



High-level Liquid Waste (HLLW)



Vitrified radioactive waste (VRW)

Insoluble residue

①Recovery LLFP form HLLW
【Concept】
○Use HLLW directly
○Recycle reagents

②Recovery LLFP form VRW
【Concept】
○Dry process
:Reduction-melting using molten salt
○Wet process
: acid dissolution in an air atmosphere

③Recovery LLFP from UR
【Concept】
○Separate alloy and oxides (fluoride et al.,)

Laser even/odd separation

Trans-mutation or recycling

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Simulated solution(Sim. HLLW)

Simulated High-level Liquid Radioactive Wastes (sim. HLLW)

- Based on the composition of the concentrated High-level Liquid Radioactive Wastes from commercial reprocessing plant

Compositions of simulated HLLWs
(29 elements, 2M HNO_3)

Elements	Concentration (g/L)	Elements	Concentration (g/L)
SeO_2	0.17	Cs_2O	5.29
Rb_2O	0.84	BaO	4.15
SrO	1.99	La_2O_3	3.01
Y_2O_3	1.27	CeO_2	15.26
ZrO_2	10.48	Pr_6O_{11}	2.83
MoO_3	10.49	Nd_2O_3	9.97
MnO	2.47	Sm_2O_3	1.86
RuO_2	5.90	Eu_2O_3	0.30
Rh_2O_3	1.07	Gd_2O_3	9.86
PdO	3.11	P_2O_5	0.18
Ag_2O	0.15	Cr_2O_3	0.06
CdO	0.24	Fe_2O_3	1.22
SnO_2	0.22	NiO	0.12
Sb_2O_3	0.04	Na_2O	24.62
TeO_2	1.22	-	-

sim. HLLW

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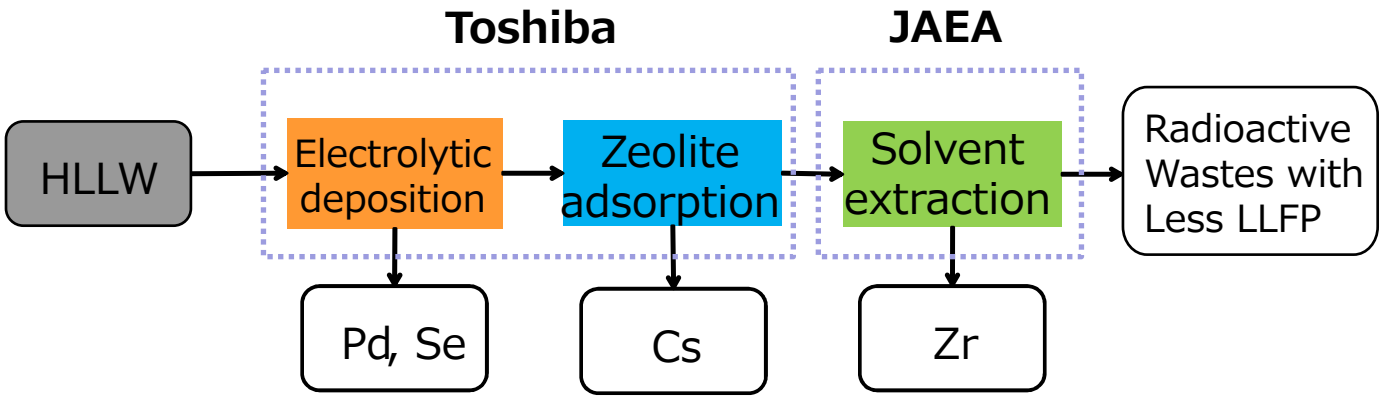
Separating technologies with limited secondary wastes

Proposed process

○no pretreating : use HLLW directly
○no liquid change : separate from nitric acid media
○re-usable : electrode, 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%

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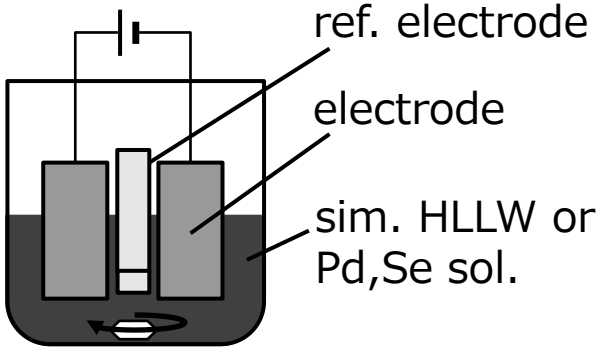
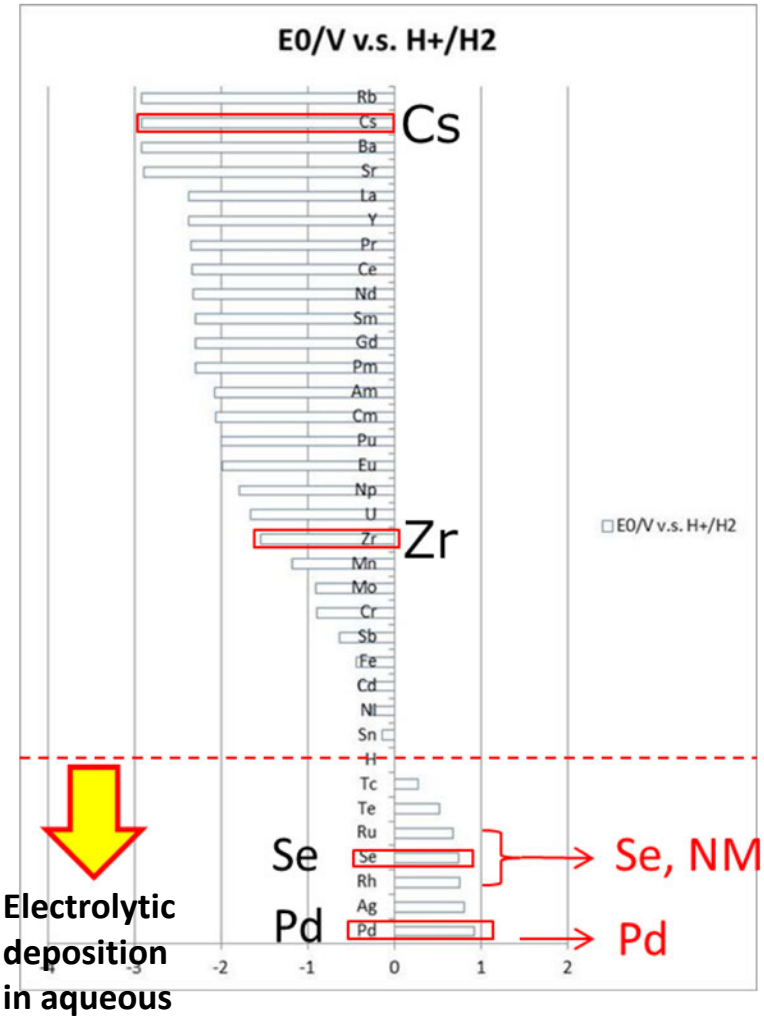
Electrolytic deposition ～Experimental conditions



Electrolysis

Electrolysis condition

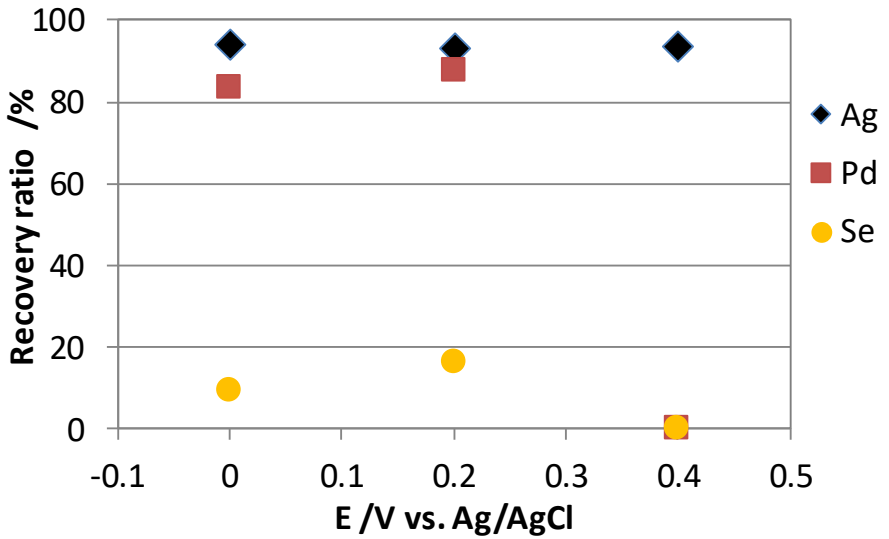
Concentration of HNO_3: 2M
Solution : sim. HLLW
Potential : 0V～0.4V vs. Ag/AgCl
Rotation : 600 rpm
Liquid volume : 10 ml
Electrode : Pt 2 cm^2



Standard electrode potential of each species in HLLW

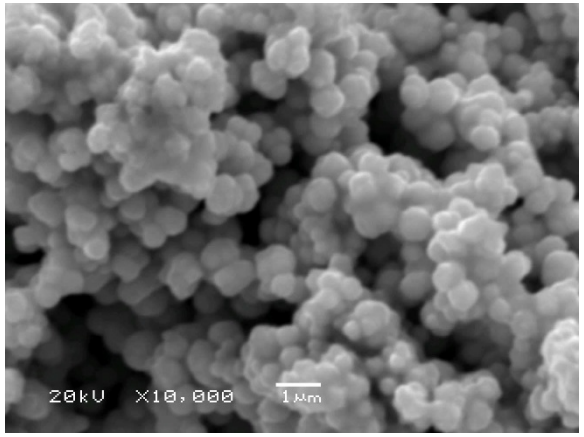
Electrolytic deposition～Results and discussion

・Constant potential electrolysis

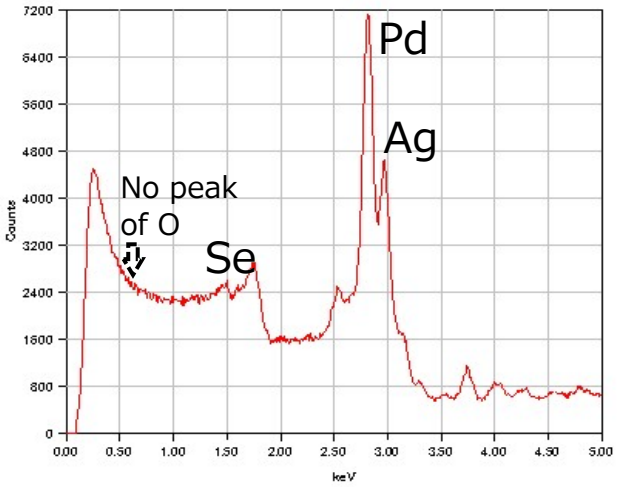


Recovery ration of metals on potential

SEM



EDS



SEM/EDS of deposition

Pd, Se and Ag were deposited as metal by electronically

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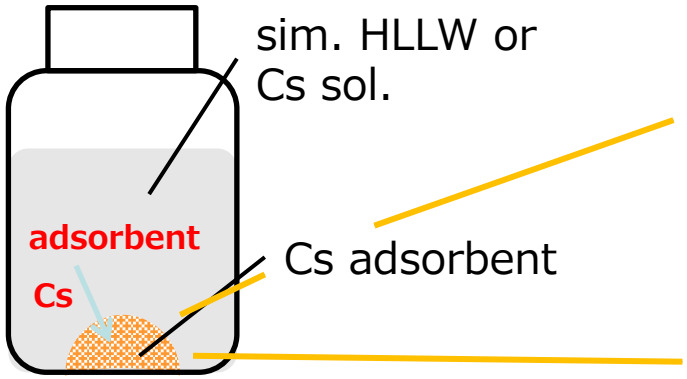
Zeolite adsorption～Experimental conditions



Cs adsorption test

Cs adsorbent

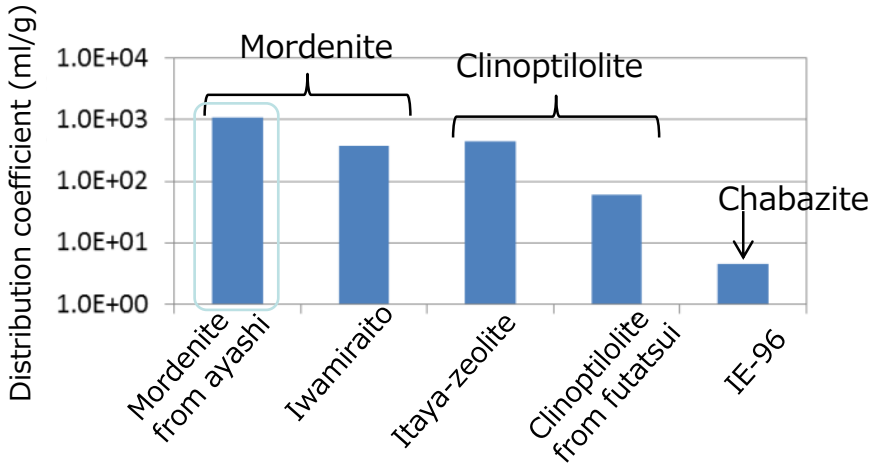
Distribution coefficient

$$Kd = \frac{C_L - C_e}{C_e} \times \frac{m}{V} \times 10^{-3}$$

Kd: distribution coefficient (m^3/kg)
m: weight of adsorbent (g),
V: liquid volume (ml),
C_e: achieved Cs concentration in liquid phase (μg/ml),
C_L: initial Cs concentration in liquid phase(μg/ml)

Adsorption condition

Concentration of HNO_3: 2M
Solution : sim. HLLW, Cs solution
adsorbent : 90-100 ml/g
Rotation : 100 rpm
Liquid volume : 10 ml



Distribution coefficient of Cs adsorbent

Mordenite > Clinoptilolite » Chabazite

※selected the best adsorbent

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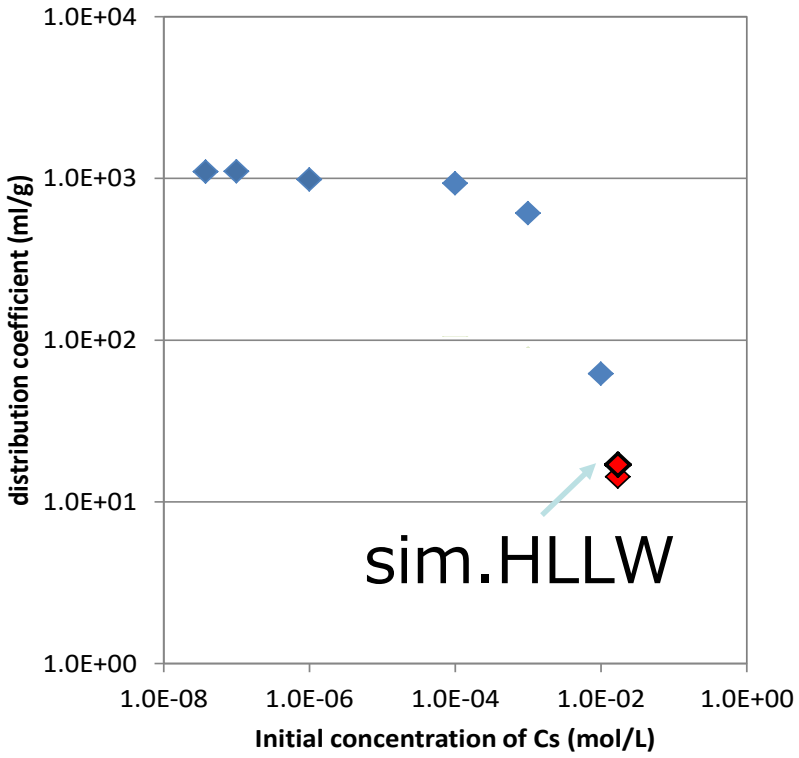
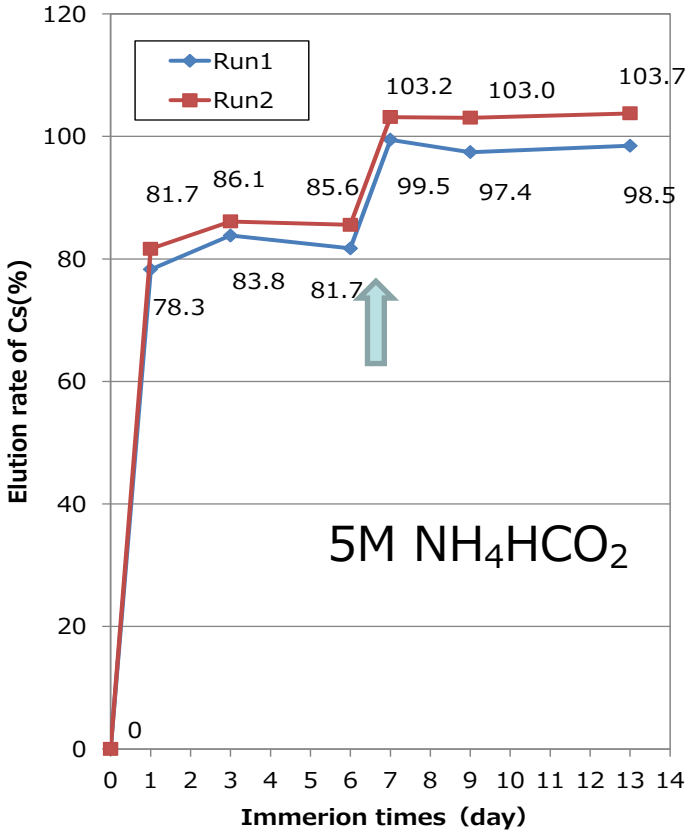
Zeolite adsorption～Results and discussion



Absorption of Cs by mordenite.



Elution of Cs from mordenite.

Cs can be absorbed and eluted using mordenite

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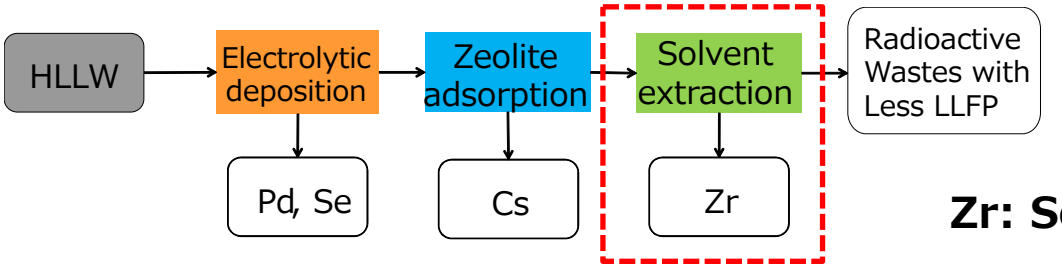
Solvent Extraction with New extractant for Zr

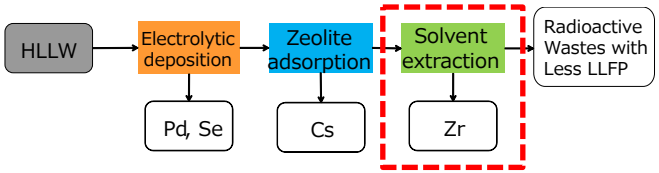


Zr: Separation using solvent extraction

HAA: New extractant for Zr separation

- One of the radiolytic products from DGA extractants
- Bidentate Ligand with O donors
- Soluble in dodecane
- CHON principle molecule
- Simple synthesizing method

Solvent Extraction with New extractant for Zr – Extraction Behavior



Zr: Separation using solvent extraction

- Extractability
  Zr > Mo > Ag ≈ Sb > Sn > Lns > Fe
- High selectibiliy for Zr、Mo

- Extracted Complex Zr:HAA=1:3 with NO_3^-
- High selectibily for Zr from Mo in high Nitric acid concentration

Summary

- **Proposed separation process for LLFPs from HLLW**
  - The separation process for Pd, Se, Cs and Zr are based on electrolytic deposition, adsorption and solvent extraction technique.
  - Pd and Se are recovered by electrolytic deposition.
  - Cs are adsorbed by Zeolite.
  - Zr are separated by solvent extraction using new extractant, HAA.
- **Current status of R&D for the separation process**
  - Pd and Se are recovered as metal deposition
  - Cs are recovered with Zeolite. Zr was adsorbed on mordenite and eluted effectively.
  - New extractant for Zr separation, HAA, was developed. The condition of the solvent extraction of Zr from HLLW are established.
  - LLFPs, Sn and Se, and Mo, which is the interference element for Vitrification process, are recovered with HAA extractant
- **Future work for the LLFP separation process development**
  - There is possibility of a rational LLFP separation process with HAA extractant.
  - The integrated separation process for all actinide elements and LLFPs will be discussed for future advanced fuel cycle with P&T.