Pyropartitioning Experiment to Recover TRUs from High Level Liquid Waste

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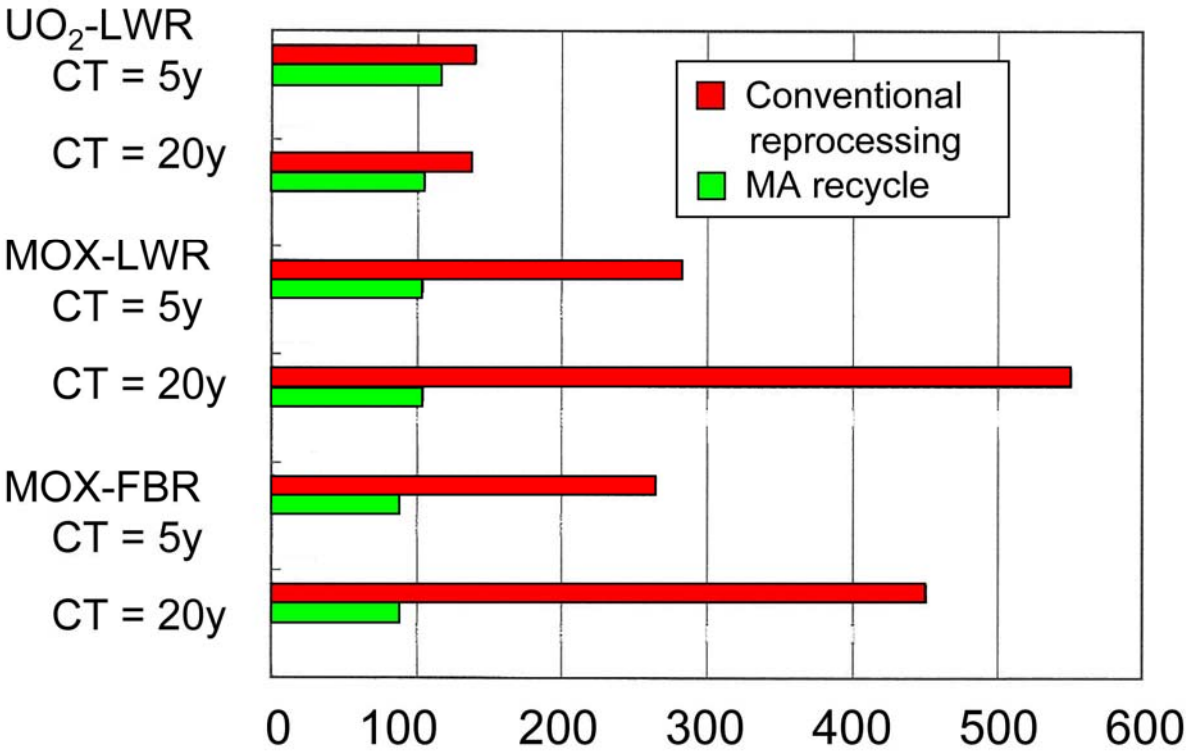
Benefits of MA recycle

- ***Reduction of potential toxicity of high level waste***

- ***Reduction of heat generation of high level waste***

⇨ ***Prolongation of repository's life time***



Emplacement area necessary for geologic disposal (m^2/TWh)

Burn-up
- UO_2-LWR : 43GWd/t
- MOX-LWR: 43GWd/t
- MOX-FBR : 79GWd/t

CT : Cooling time before reprocessing

from
"Status and perspective of partitioning and transmutation technology", *J. Atomic Energy Society of Japan*, **50** [3], 158 (2008).

P-T concepts in Japan

Organization	Material	Separation	Transmutation	Fuel
CRIEPI	HLLW from Purex	Pyro	Commercial FBR	Metal
JAEA	HLLW from Purex	Aqueous& Pyro	ADS	Nitride
JAEA	Spent fuel	Aqueous	Commercial FBR	Oxide

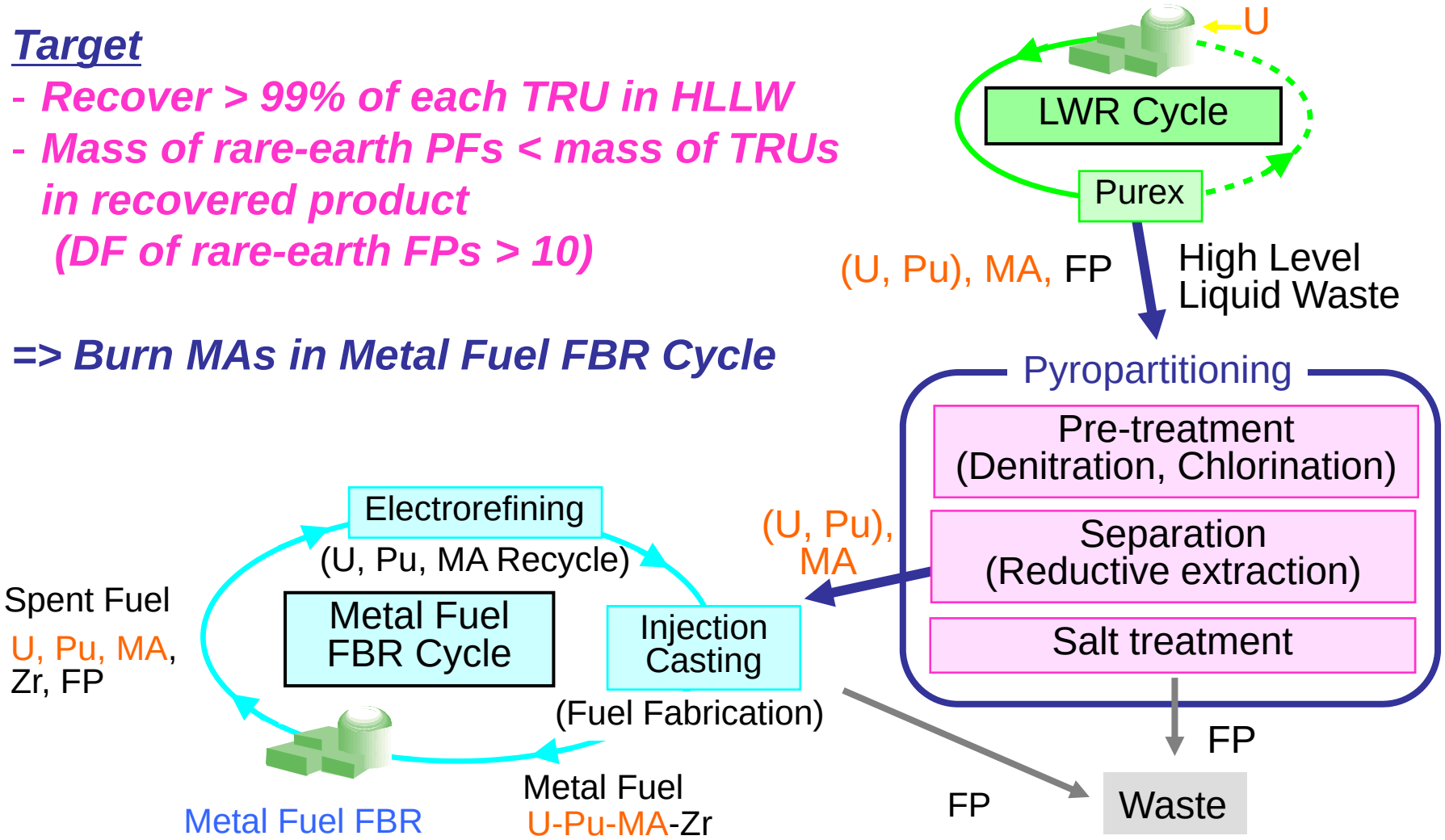
"Subcommittee of Partitioning and Transmutation Technology" set by the "Atomic Energy Commission in Japan" issued a report on R&D of P-T technologies in April/2009.

CRIEPI's concept on pyropartitioning

Target

- ***Recover > 99% of each TRU in HLLW***
- ***Mass of rare-earth PFs < mass of TRUs in recovered product (DF of rare-earth FPs > 10)***

=> Burn MAs in Metal Fuel FBR Cycle



Irradiation of MA-containing metal fuel

MA-containing alloys were irradiated in Phénix.

3 metal fuel pins & 16 oxide fuel pins were arranged in an capsule.

Pin No.1 : U-19Pu-10Zr

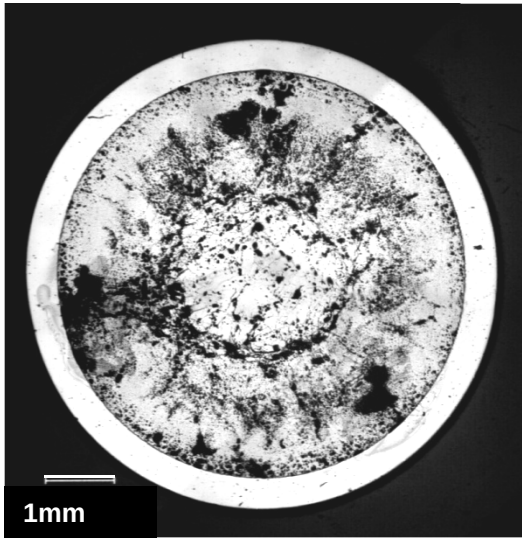
Pin No.2 : U-19Pu-10Zr-2MA-2RE

Pin No.3 : U-19Pu-10Zr-5MA / -5MA-5RE

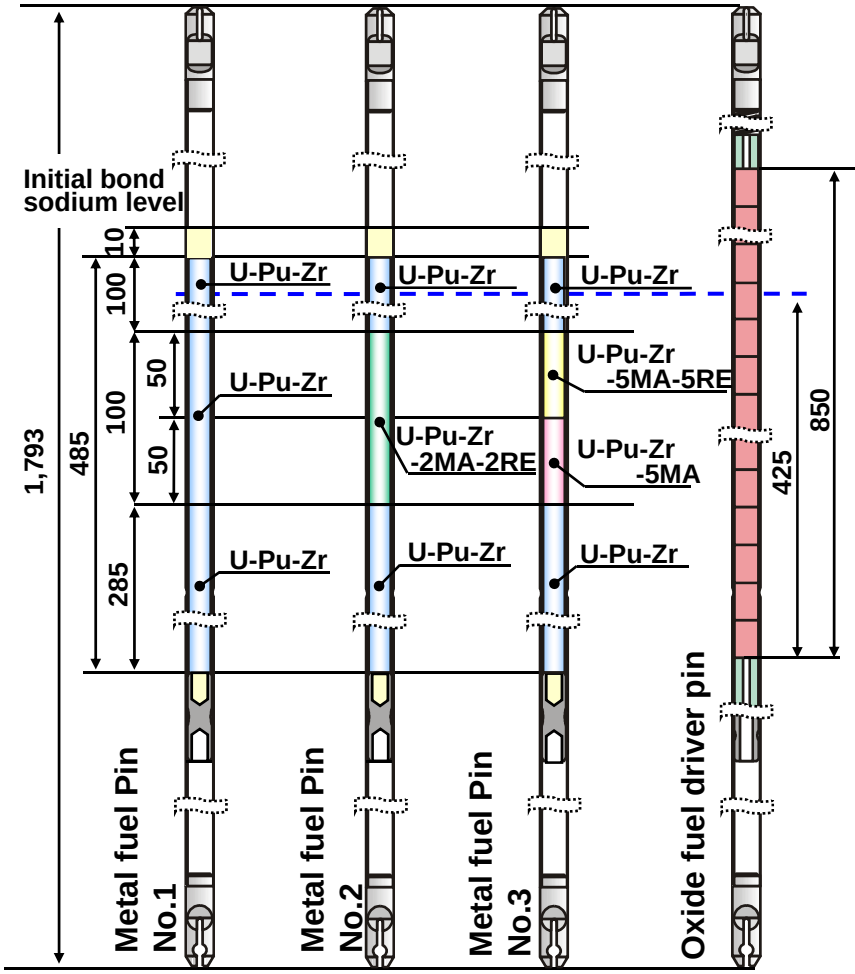
Cladding material : 15-15Ti

Burnup goals ~2.5at.% (METAPHIX-1), ~ 7at.% (METAPHIX-2), ~10at.% (METAPHIX-3).

PIE is under way



Cross-Sectional Overview(U-19Pu-10Zr-5MA-5RE)



Schematic views of irradiated fuel pins.

Merits of pyropartitioning

- *Pu and MAs are recovered together.*
  => It is difficult to separate pure Pu.

- *MAs/rare-earth FPs separation is probable.*

- *Solvents (molten salt and liquid metal) are radiation resistant.*
  => Secondary waste is less.

- *Better compatibility with metal fuel FBR.*
  => Better transmutation of MAs due to hard neutron spectrum.

CRIEPI's pyropartitioning process

Denitration

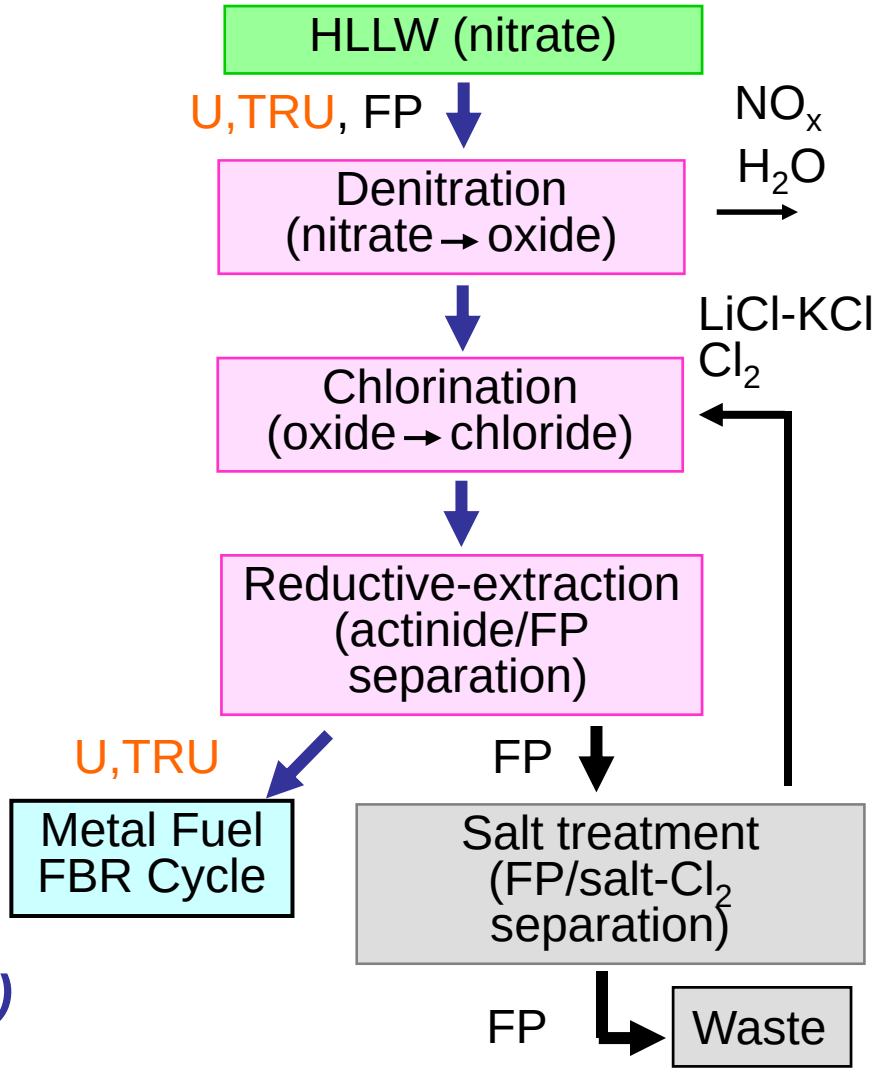
- *Convert elements in HLLW into oxides by calcination in air.*
  ex) $UO_2(NO_3)_2 \rightarrow UO_3 + NO_x$

Chlorination

- *Convert denitrated material into chloride by using* Cl_2 *gas and carbon.*
  ex) $UO_3 + 1.5Cl_2 + 3C \rightarrow UCl_3$ *(in LiCl-KCl)* $+ 3CO$

Reductive-extraction

- *Separate actinide elements from FPs by molten salt/liquid metal reductive-extraction.*
  ex) UCl_3 *(in LiCl-KCl)* $+ 3Li \rightarrow$ *U (in Cd)* $+ 3LiCl$ *(in LiCl-KCl)*



Situation of research on pyropartitioning -1

Achieved (since 1986)

- ***Obtaining thermodynamic data for actinide/FP separability study using U and TRUs***
- ***Series experiments of denitration and chlorination using U and simulating FPs***
- ***Demonstration of actinide/FP separation using simulated chlorination product containing U, TRUs, and simulating FPs***

ex) Separation factor values (SF) of TRUs and rare-earth elements vs. U in LiCl-KCl/Cd and LiCl-KCl/Bi (500^{o}C)

Element	SF in LiCl-KCl/Cd	SF in LiCl-KCl/Bi
U (basis)	1	1
Np	1.9	1.1 x 10
Pr	4.2 x 10	9.22×10^{2}
Nd	4.5 x 10	9.33×10^{2}
Ce	4.9 x 10	8.34×10^{2}
La	1.31×10^{2}	2.53×10^{3}
Gd	1.79×10^{2}	1.04×10^{4}
Y	5.32×10^{3}	5.84×10^{5}

Situation of research on pyropartitioning -2

Must be solved

- ***Behaviors of TRUs and Tc***
- ***Evaporation of U and Tc during chlorination***

  (cf. 5% of U and 40% of Re (surrogate of Tc) evaporated at chlorination of 700°C, 10hours with direct introduction of Cl_2 gas into molten salt.)
- ***Any difference between actual material and simulating material?***

Pyropartitioning experiment using real HLLW is necessary!

Pyropartitioning experiment using real HLLW

Objective

- *Recover all of actinide elements as reductive-extraction product.*
- *Confirm separation behavior at reductive-extraction.*

  ⇨ *Demonstration of pyropartitioning process*

Procedure

- *Use real HLLW as starting material of denitration.*
- *Series experiment of denitration, chlorination, and reductive-extraction*
- *Evaporated material during denitration and chlorination is analyzed, as much as possible.*

Preparation of real HLLW

- Raffinate from Purex reprocessing of irradiated PWR-MOX fuel.
- To observe behaviors of Np and Pu, concentrated solutions of Np and Pu were added to the Raffinate.

Composition of HLLW

U: 8400 µg/g

TRU: 600 µg/g

(Np-237: 105 µg/g, Pu-239: 54 µg/g, Am-243 + Cm-243: 66 µg/g)

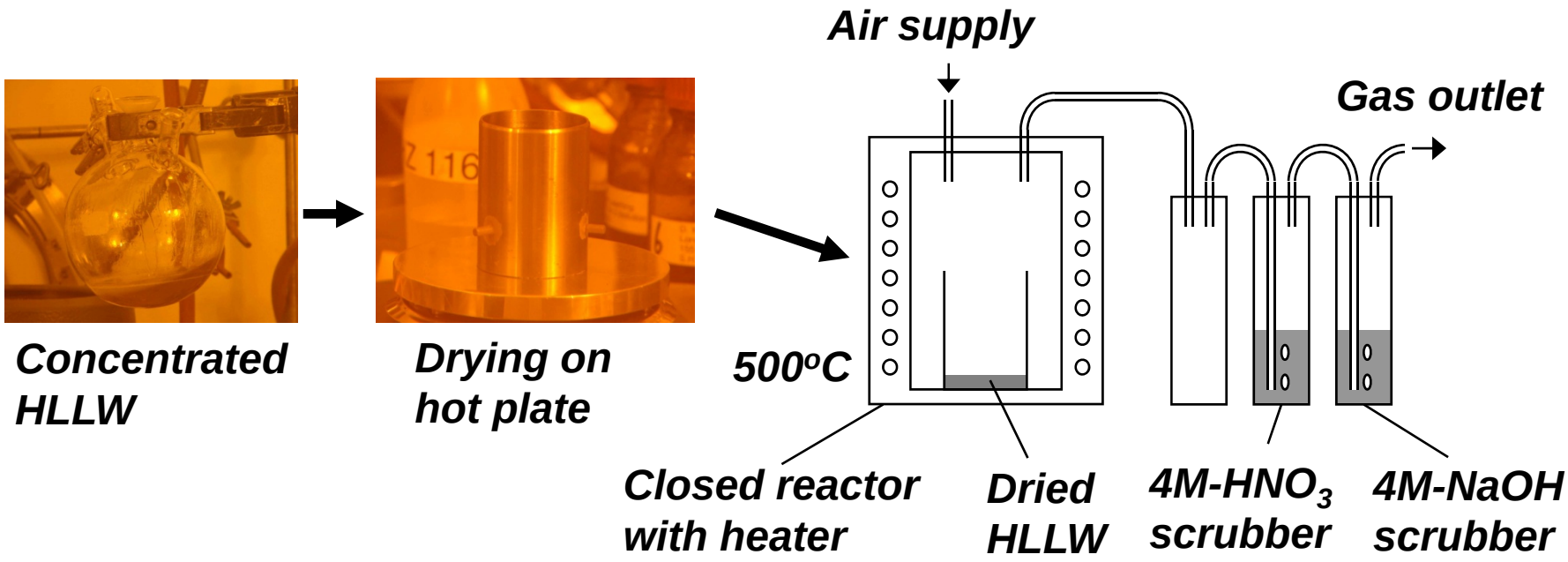
Fission products: 2000 µg/g

(rare-earth: 870 µg/g, alkaline-earth: 290 µg/g,
alkaline: 170 µg/g, noble metal: 260 µg/g, Tc-99: 15 µg/g)

- 520g of the HLLW was used.

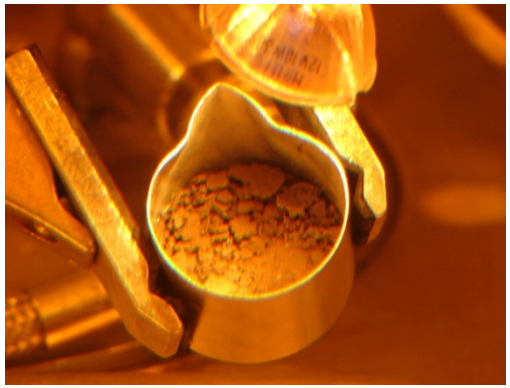
Experimental : Denitration

- HLLW was concentrated for volume reduction (90-120°C, 3 hours).
- Concentrated solution was dried (100-140°C, 20 hours).
- Dried material was calcined under air flow (500°C, 84 hours).
- Volatile material and NO_x were trapped at scrubbers.

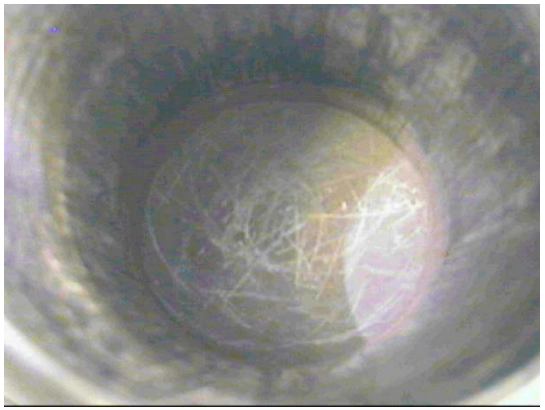


Results : Denitration

- *The calcined material was taken out easily from the crucible.*
- *The mass of the calcined material (7.32g) almost agreed with theoretical value (6.91g).*
- *No damage was found on inner surface of SS crucible used for dying and calcination.*



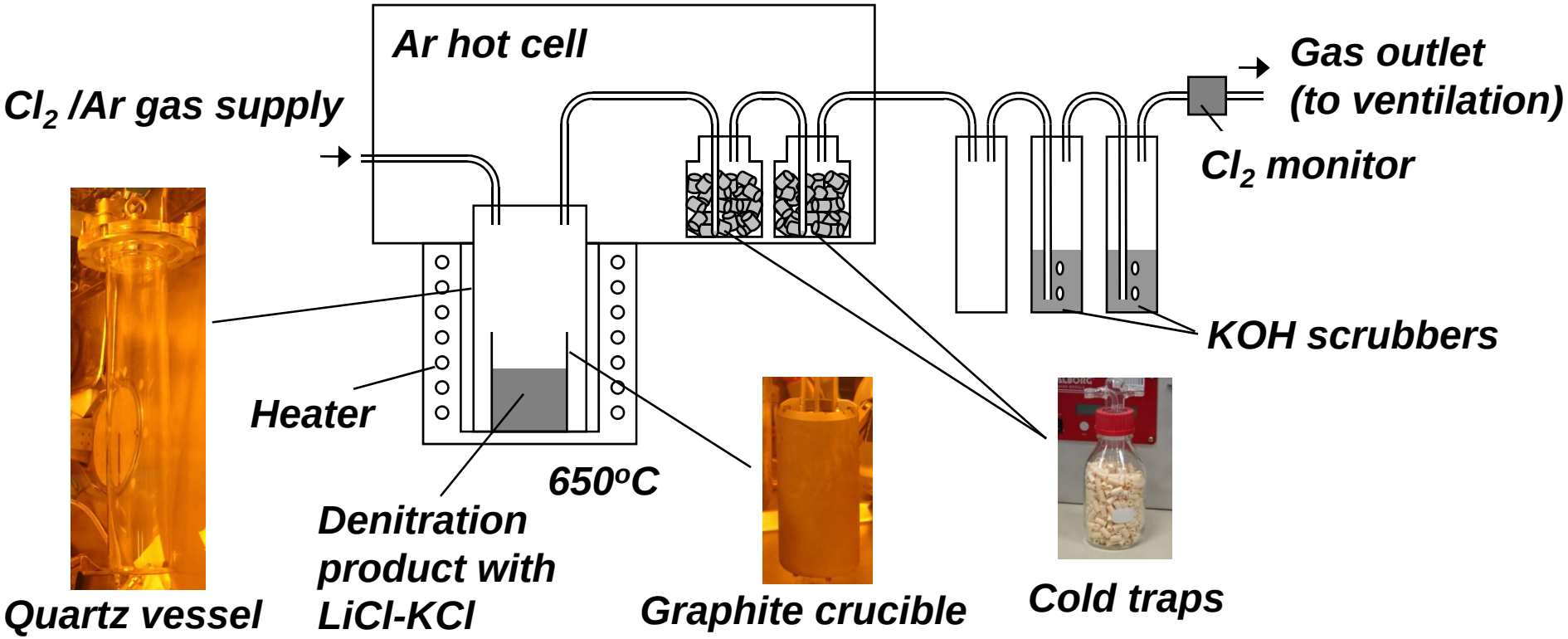
Calcined material (7.32g)



Inside of SS crucible

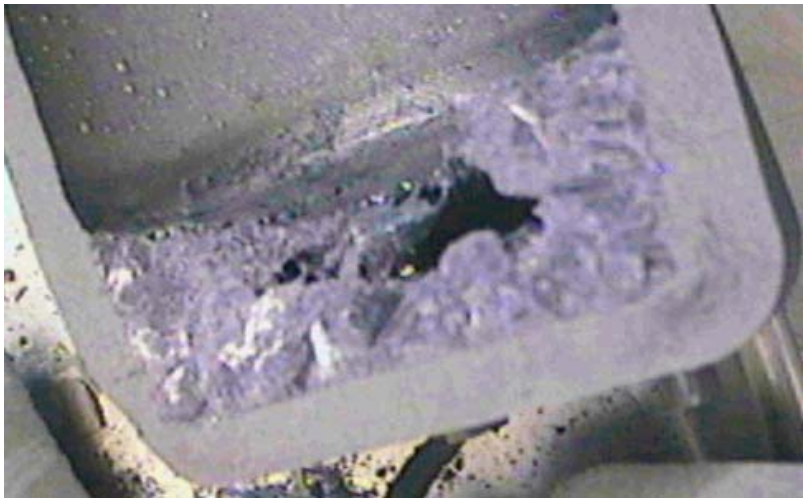
Experimental : Chlorination

- *Whole denitration product (7.3g) was heated to 650°C together with 97.5g of LiCl-KCl salt in a graphite crucible.*
- *The vessel was filled with chlorine gas for 32 hours.*

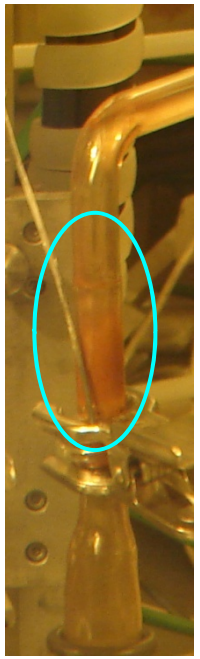


Results : Chlorination -1

- *Uniform salt with no precipitate was recovered.*
- *Some brown deposit was observed.*



Chlorination product



Deposit at Cl_2 gas outlet

Results : Chlorination -2

Mass balances of elements at denitration and chlorination

Element/Group	Evaporated at denitration	Evaporated at chlorination	Chlorination product	Total
U	0.0%	0.0%	113%	113%
Np	0.0%	0.0%	109%	109%
Pu	0.0%	0.0%	99%	99%
Am	0.0%	0.0%	113%	113%
Cm	0.0%	0.0%	105%	105%
Tc	0.0%	0.6%	82%	82%
Rare-earth FP	0.0%	0.1%	101%	101%
Alkaline-earth FP	0.0%	1.9%	106%	108%
Transition metal FP (Tc excluded)	0.0%	20.4%	23.7%	44%
Noble metal FP	0.1%	0.0%	128%	128%
Other FP (Cd excluded)	0.0%	0.4%	102.0%	104%

Results : Chlorination -3

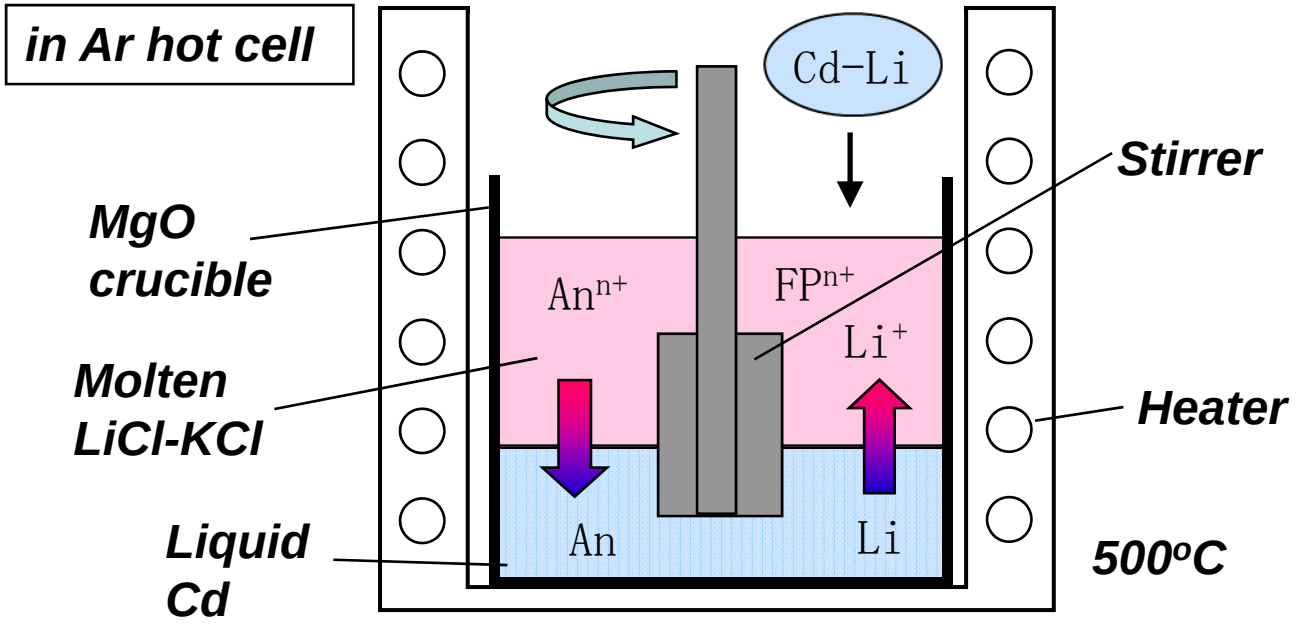
- *Actinide elements in chlorination product salt were 99-113%.*
- *No actinide elements evaporated.*

- *The evaporated material contained Mo, Zr, Sn etc., as expected.*
- *Almost all of Tc remained in chlorination product.*

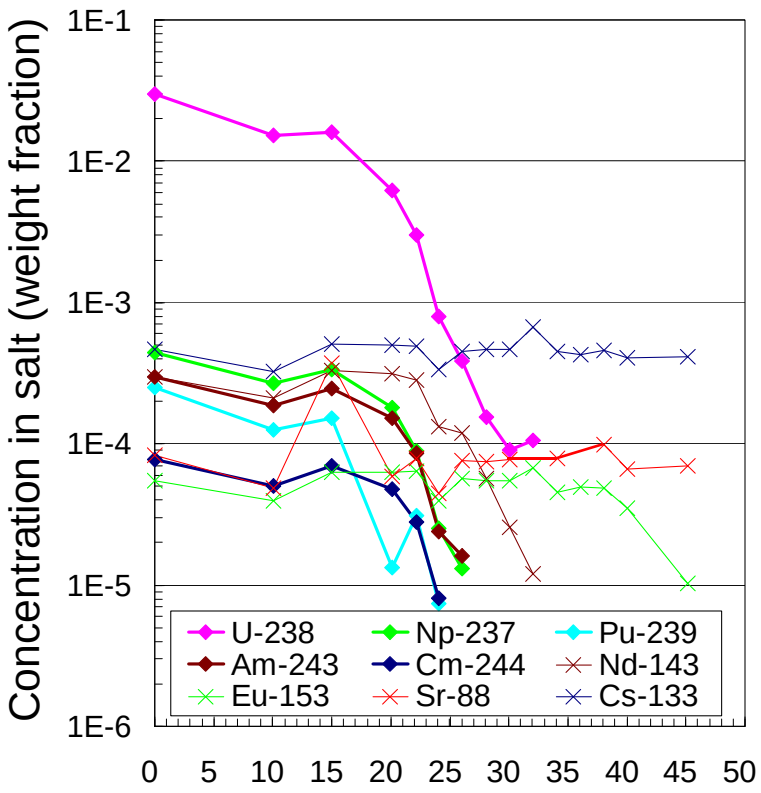
The conditions (temperature, way of Cl_2 gas introduction, duration) seemed suitable to avoid evaporation of U and Tc.

Experimental : Reductive-extraction

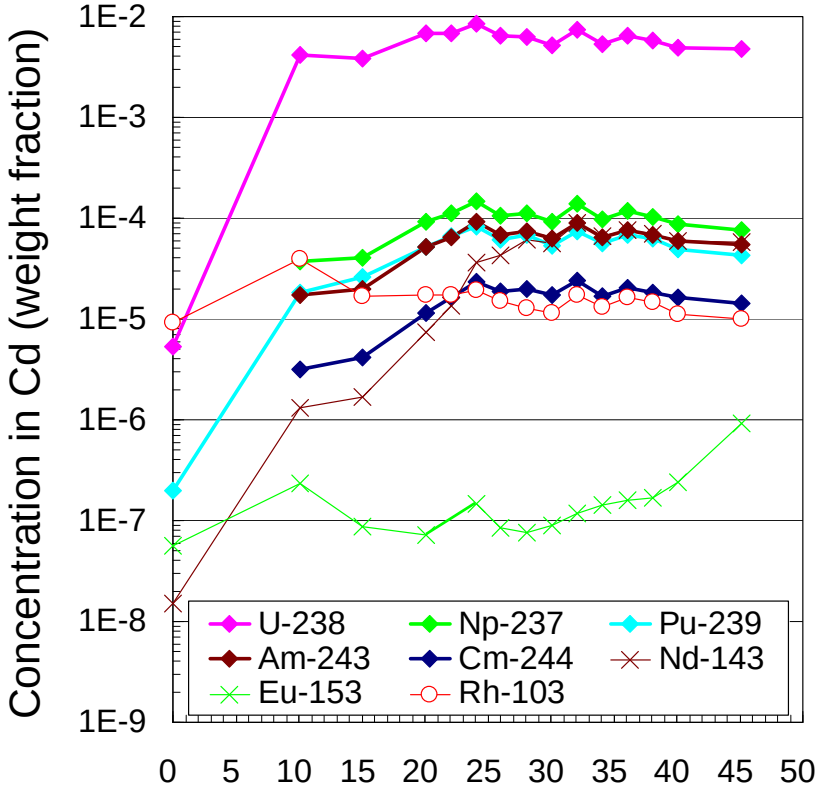
- *Half of chlorination product (47.3g) was heated to 500°C together with 204.5g of Cd metal in a MgO crucible.*
- *Elements in salt were gradually reduced and extracted to Cd phase by Cd-Li alloy addition.*
- *Distribution of each element was evaluated.*



Results : Reductive-extraction -1



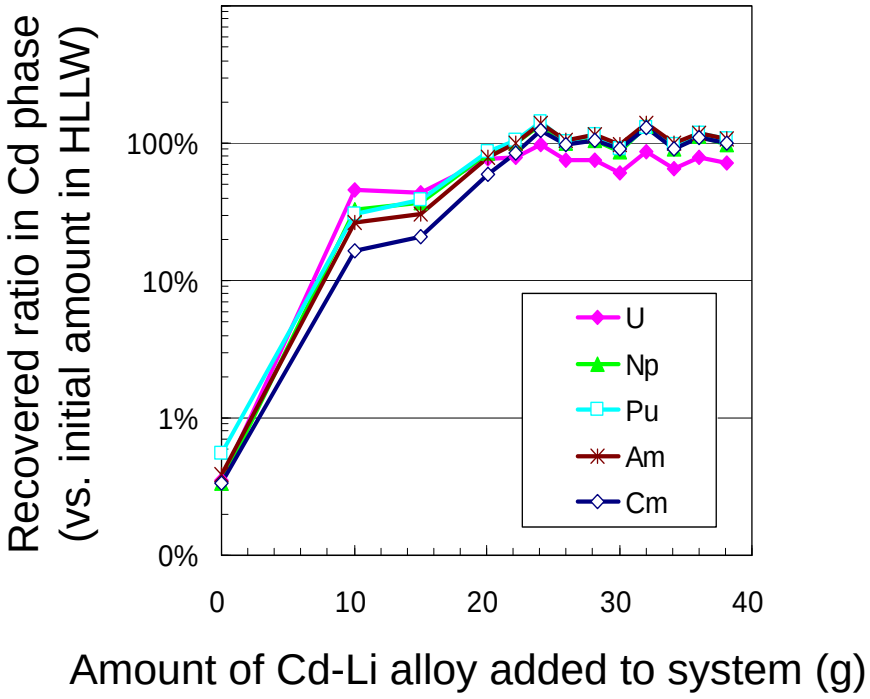
Amount of Cd-Li alloy added to system (g)



Amount of Cd-Li alloy added to system (g)

- TRUs and U were completely removed from salt phase and recovered in Cd.

Results : Reductive-extraction -2



All of TRUs were recovered in Cd.

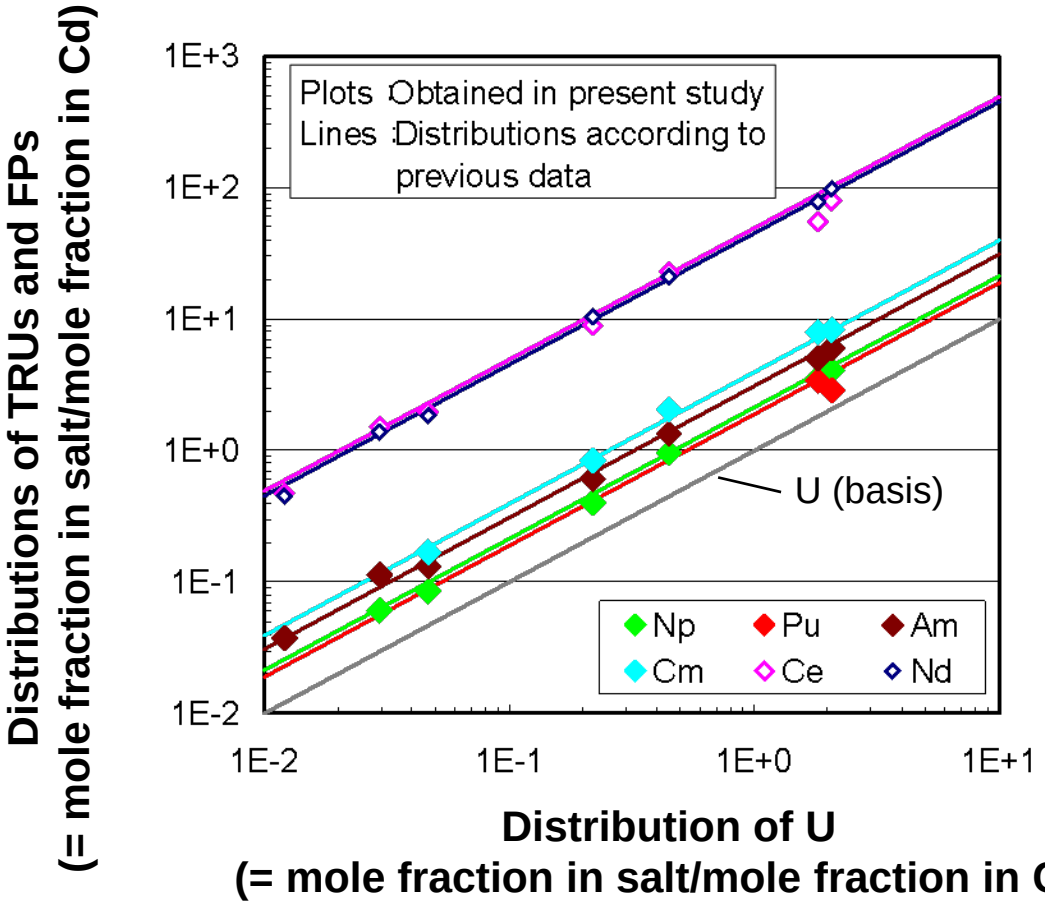
➡ ***No mass loss and perfect reaction at denitration, chlorination, and reductive-extraction***

Results : Reductive-extraction -3

Reductive-extraction

- ***Separation behaviors of TRUs and rare-earth FPs vs. U were similar to previous data.***

=> *Separation must be performed as expected*.



Conclusion

- *Almost 100% of each TRU in real high level liquid waste was recovered as reductive-extraction product.*
- *Separation behaviors of actinide elements from FPs were similar to previous data obtained by un-irradiated material.*
- *Each FP behaved as expected.*

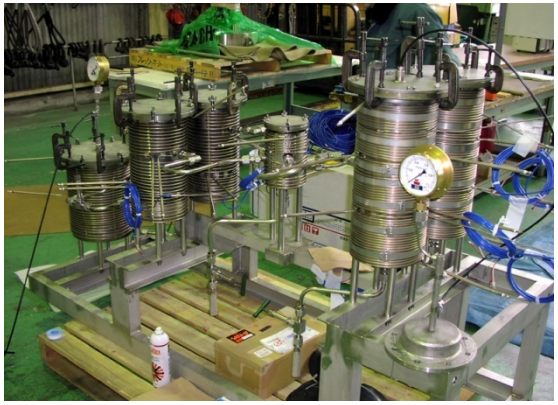
- *Whole pyropartitioning process (denitration, chlorination, and reductive-extraction) was successfully demonstrated.*
- *Since difference of SFs between MAs and rare-earth FPs is not large enough, multi-stage countercurrent reductive-extraction system is necessary to attain separation target.*

Development with metal fuel FBR cycle

- *CRIEPI continues developing both "pyropartitioning" and "metal fuel FBR cycle" technologies, including process installation development, as integrated one.*

ex)

- *3 stage counter-current extraction equipment was installed. Demonstration was performed using lanthanide elements as simulants.*



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