The 3rd International Pyroprocessing Research Conference (IPRC2010)
November 29 – December 3, 2010
RIAR, Dimitrovgrad, Russia

Recent Progress on Development of the Pyrochemical Process of Spent Nitride Fuels for ADS

H. Hayashi[1)], T. Satoh[1)], H. Shibata[1)], T. Iwai[1)], K. Nishihara[1)], Y. Arai[1)],
H. Kofuji[2)], M. Myochin[2)]

[1)] Nuclear Science and Engineering Directorate
[2)] Advanced Nuclear System Research and Development Directorate
Japan Atomic Energy Agency (JAEA)

R&D Activity on Pyrochemical Process in JAEA

Pyrochemical Reprocessing Development in FaCT*

* Fast Reactor Cycle Technology Development

Advanced Nuclear System Research and Development Directorate, H. Funasaka, M. Myochin, H. Kofuji, et al.

CPF in Nuclear Fuel Cycle Engineering Laboratories, JAEA Tokai

Fundamental Research on Pyrochemical Process Including Treatment of Spent Nitride Fuel for ADS

Nuclear Science and Engineering Directorate,

K. Minato, Y. Arai, H. Hayashi, et al.

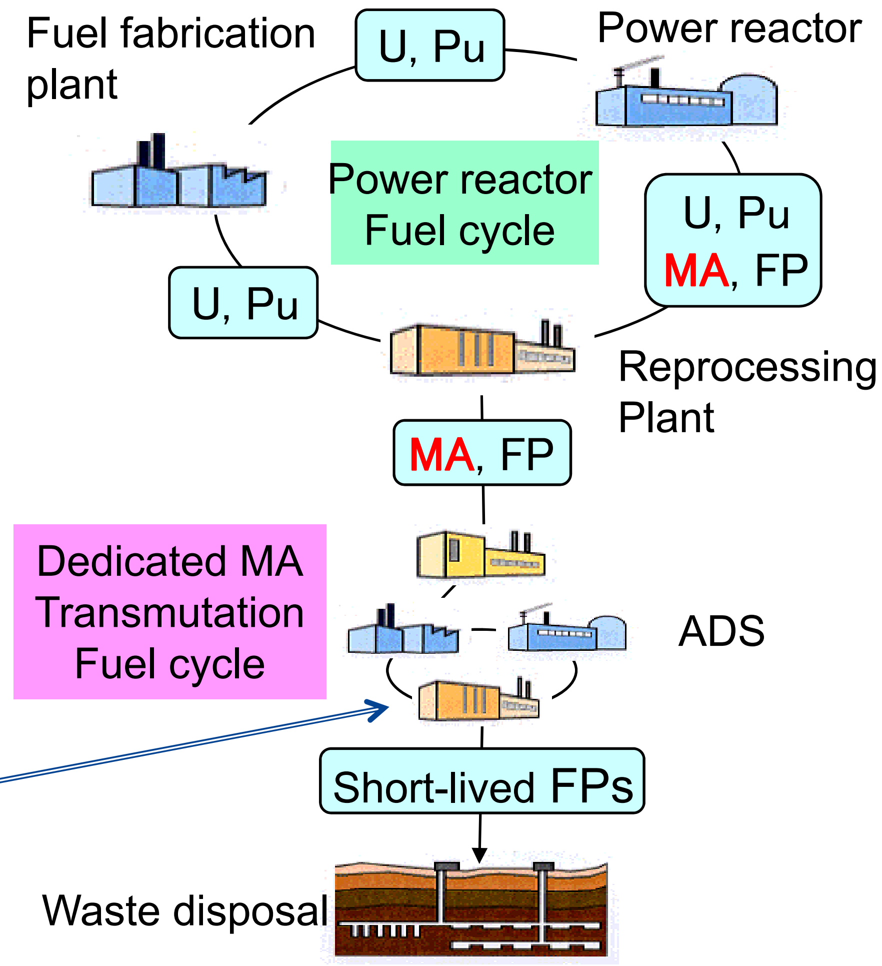
http://nsed.jaea.go.jp/en_index.html

NUCEF in Nuclear Science Research Institute, JAEA Tokai

R&D has been promoted by the joint research with Central Research Institute of Electric Power Industry (CRIEPI)

Double Strata Fuel Cycle Concept for Transmutation of MA

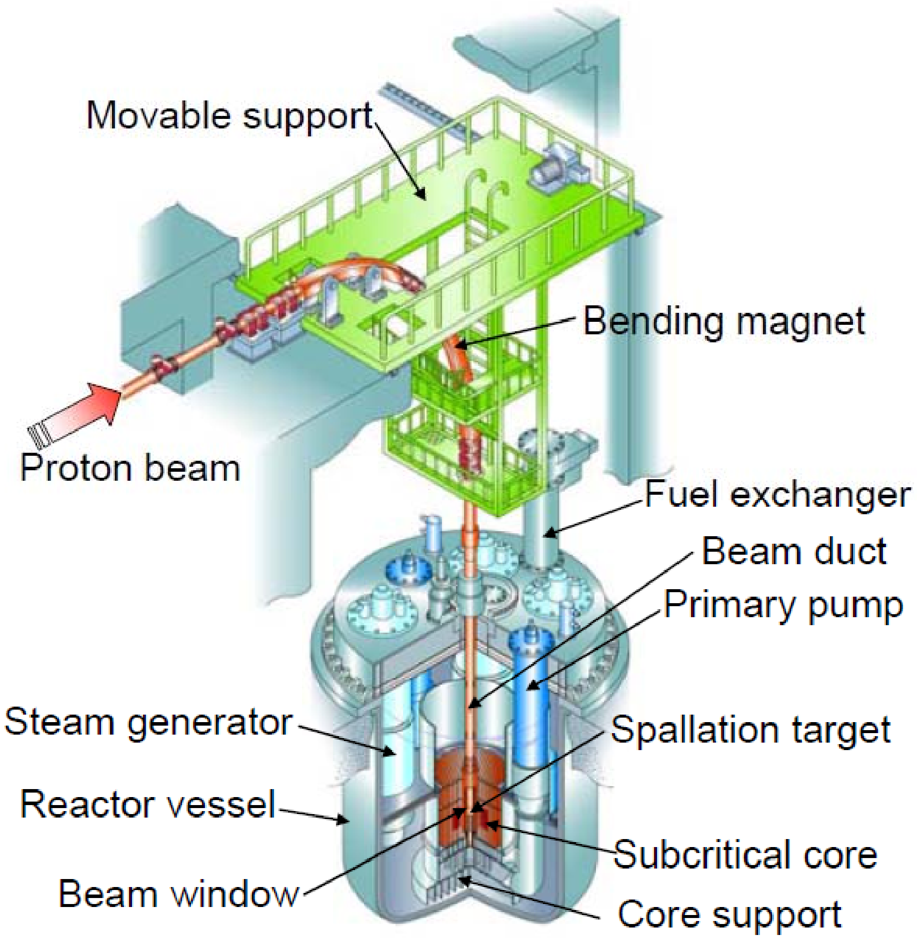
- Each fuel cycle pursues safety and economy of the fuel cycle independently.
- Performance of the power reactor fuel cycle is not disturbed by MA.
- Hazardous MA are confined in the dedicated MA transmutation fuel cycle with a small throughput.



- **Pyrochemical process is proposed for the treatment of spent nitride fuel for ADS***

* Accelerator-driven system

Design Study of ADS and MA Nitride Fuel



Conceptual view of 800 MWth LBE-cooled ADS

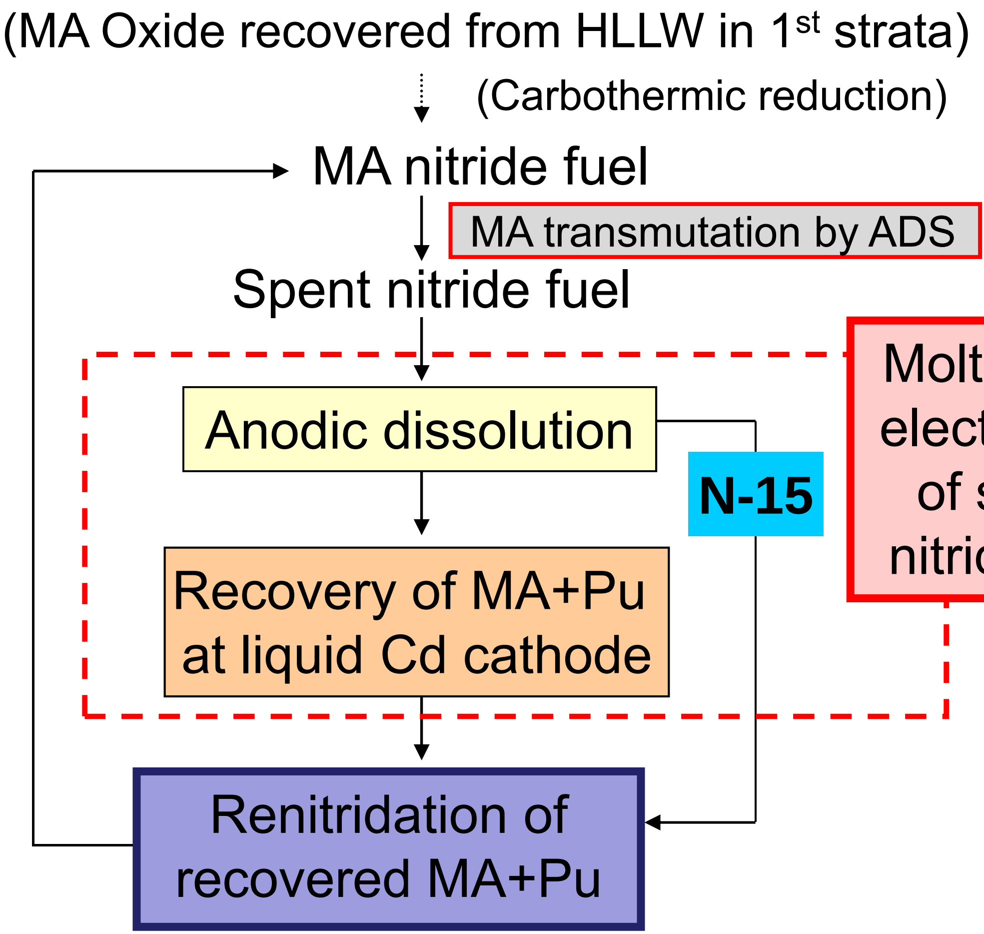
Basic design parameters of ADS

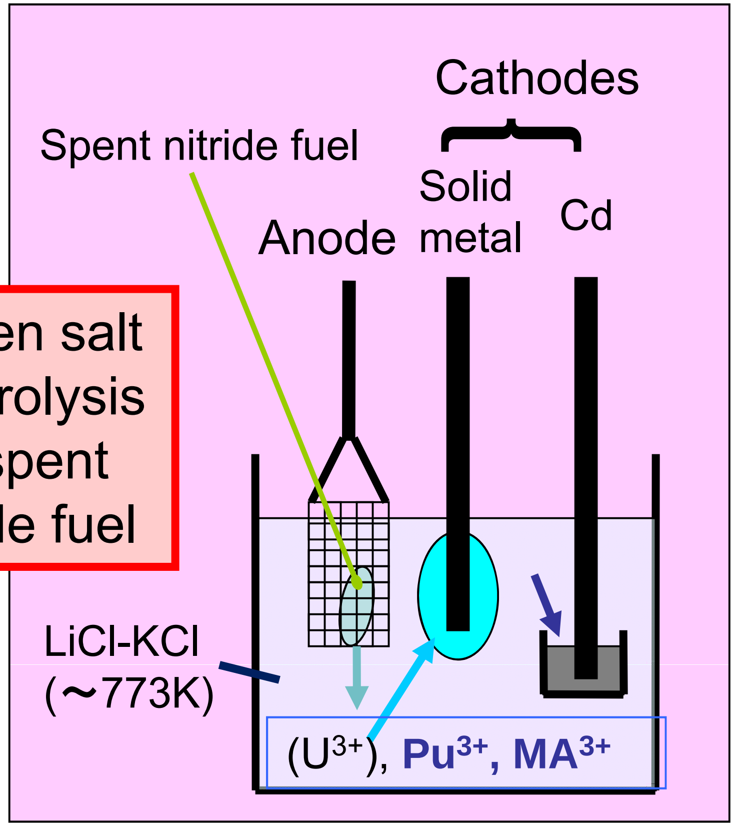
Parameters	Specification
Thermal power	800 MW
Electric power	270 MW
Coolant (average inlet / outlet temperature)	LBE (300 ^oC / 407 ^oC)
Spallation target	LBE (window-type)
Accelerator	Superconducting LINAC
Proton beam energy	1.5 GeV
Active core diameter	234 cm
Active core height	100 cm
Fuel	(MA, Pu)N
Inert matrix	ZrN
Effective multiplication factor (k_{eff})	Max. : 0.970
Cycle length	600 EFPD
Transmutation rate	500 kgMA/cycle

<u>MA nitride fuel for ADS</u>

- MA is contained as a principal component in the fuel
- Diluent material is contained in place of U
  (The weight ratio in the fuel is MAN/PuN/ZrN=0.3/0.2/0.5)

Pyrochemical Process of Spent Nitride Fuel

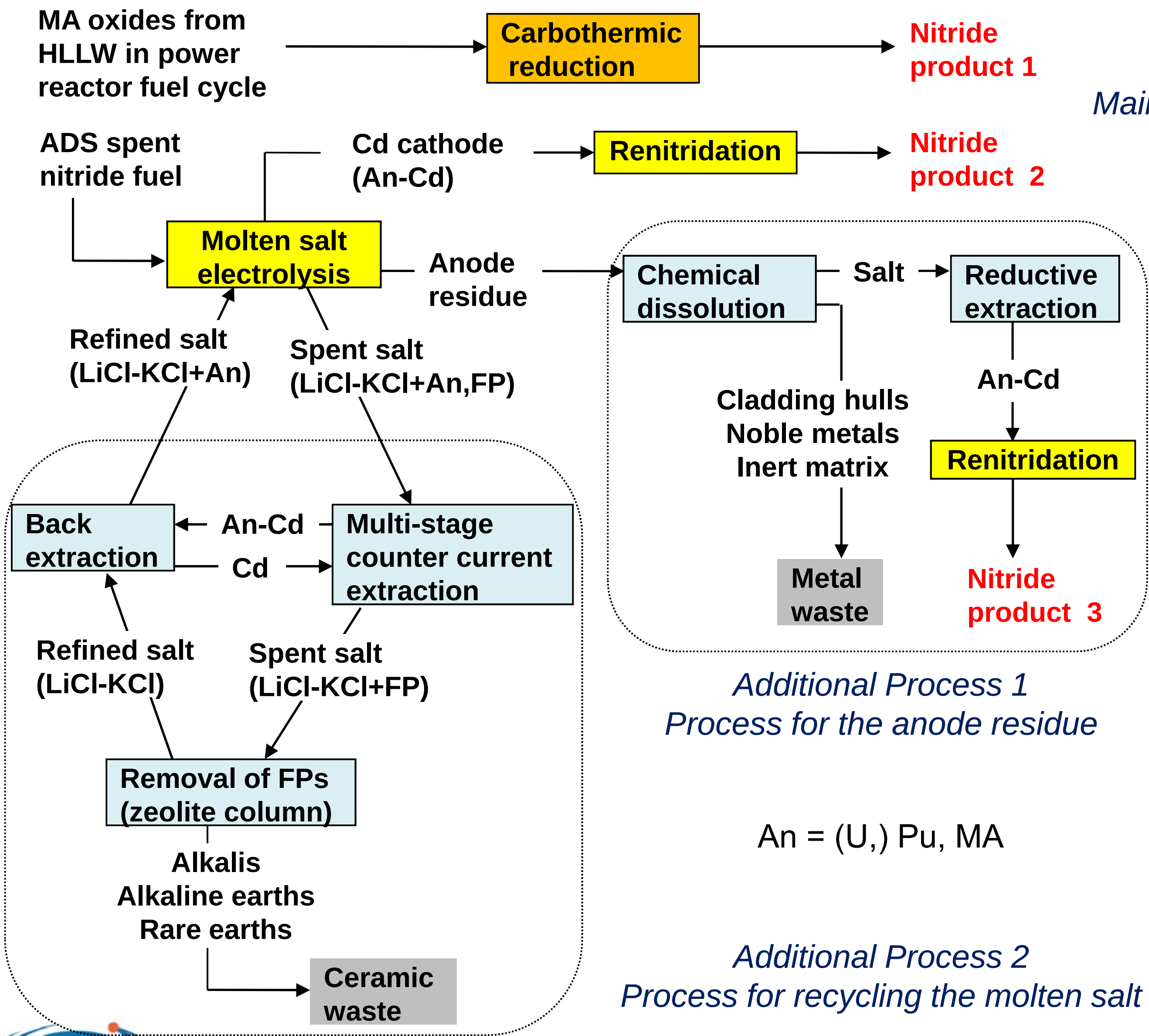




Schematic of molten salt electrolysis

This study aims at proving that the pyrochemical process is applicable to nitride fuel for the transmutation of MA by ADS

Process Flow Diagram of MA Nitride Fuel Cycle for ADS



Criteria from design study of ADS:

- → Recovery yield of MA+Pu in the reprocessing >99.95%
- → REs contamination in the recycled fuel <3.0wt%

⇩

- ✓ Two additional processes are included.
- ✓ Recycled fuel is fabricated from the nitride products1, 2 and 3.

Additional Processes

- To maximize the recovery ratio of actinides from the spent fuel,
- To minimize the ratio of lanthanides impurity in the recovered actinides as low as possible.

"*Process for the anode residue***"** *(Additional process 1)*

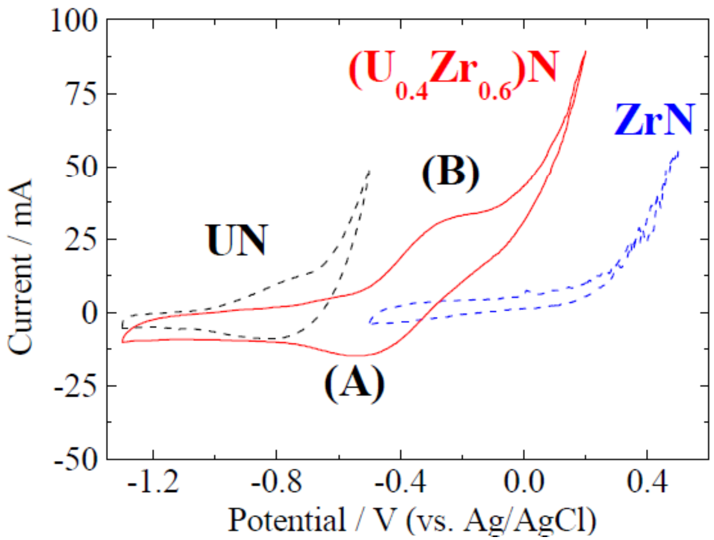
- Anode residue from the molten salt electrorefining is treated to recover actinides existing in the residue by reductive extraction.

"*Process for recycling the molten salt* **"** *(Additional process 2)*

- The ratio of lanthanides in recovered actinides is reduced by lowering the concentration of lanthanides in the molten salt bath.
- Removal of lanthanides from the molten salt bath is carried out by multi-stage extraction of molten salt/liquid metal system followed by zeolites treatment, and the treated salts are recycled to electrorefiner.

These additional processes were originally developed for metal electrorefining process for fast reactor fuel cycle.

Electrolysis of Nitride Fuel Containing ZrN



■ Addition of ZrN to nitride fuel
→ Improving chemical stability
→ Improving thermal conductivity
→ Facing difficulty in reprocessing

Cyclic voltammograms for the dissolution of (U,Zr)N

Results of potential-controlled electrolyses of (An,Zr)N (An=U or Pu)

		$(U_{0.4}Zr_{0.6})N$		$(Pu_{0.1}Zr_{0.9})N$	
		U	Zr	Pu	Zr
Metal in initial sample (mg)		149	86	20	72
Metal in the salt (wt.%)	Initial	0.51	ND	1.10	ND
	Final	0.42	ND	1.09	ND
Metal left in the anode basket (%)		No sample left (dropped out)			
Metal recovered in the LCC (mg)		129	20	7.3	4.0
Recovery yield (%)		86	23	37	5.6

Actinides are selectively recovered in Cd cathode

Further study on the electrolyses of ZrN-containing nitride fuel is necessary for proving the technological feasibility.

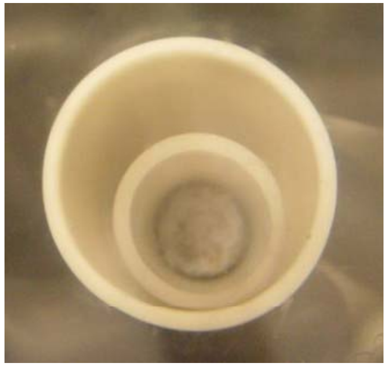
A Series of Experiments for (U,Pu)N

(U,Pu)N pellet
in Mo crucible

Electro-
refining

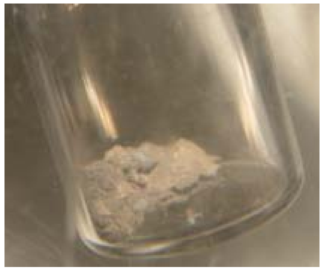
Liquid Cd cathode
in Al_2O_3 crucible

Cutting



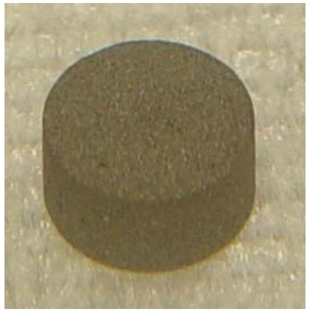
U-Pu-Cd alloy in
Y_2O_3 crucible

Renitridation

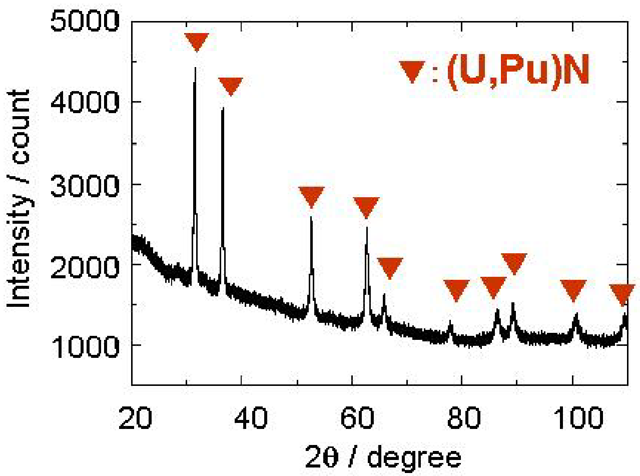


(U,Pu)N powder

Sintering

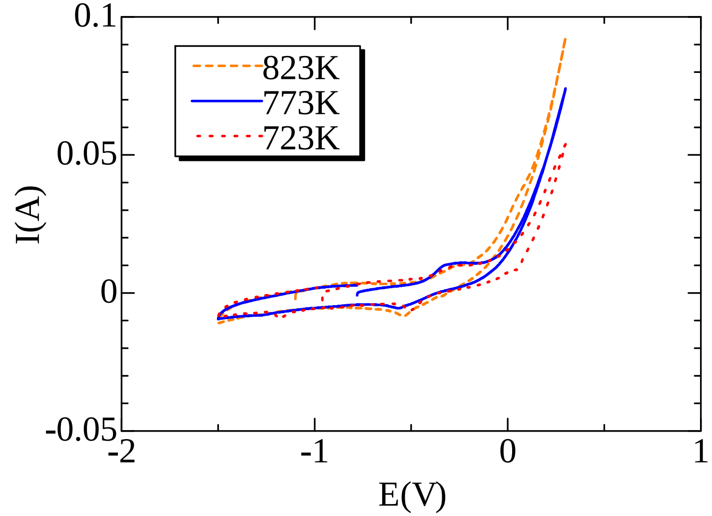


(U,Pu)N pellet



- ✓ Single phase of (U,Pu)N
- ✓ Density: ~84%TD
- ✓ O_2 impurity: 0.1~0.2wt%

Experimental Study on Americium



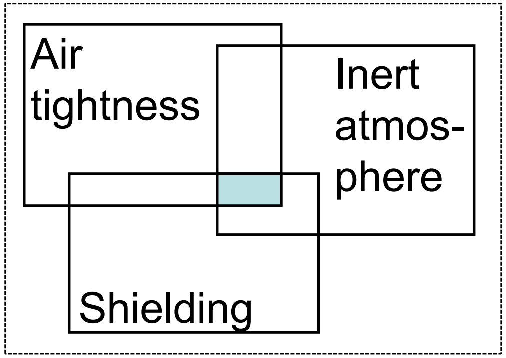
Cyclic voltammograms of AmN

Electrolysis of AmN → Am recovered in Cd cathode (Am-Cd alloy) 28.9mg

973K,5h | N_2 100cm^3/min

↓

Fine black powder 10.4mg



Fine black powder 7.2mg

673K,3h | in vacuum

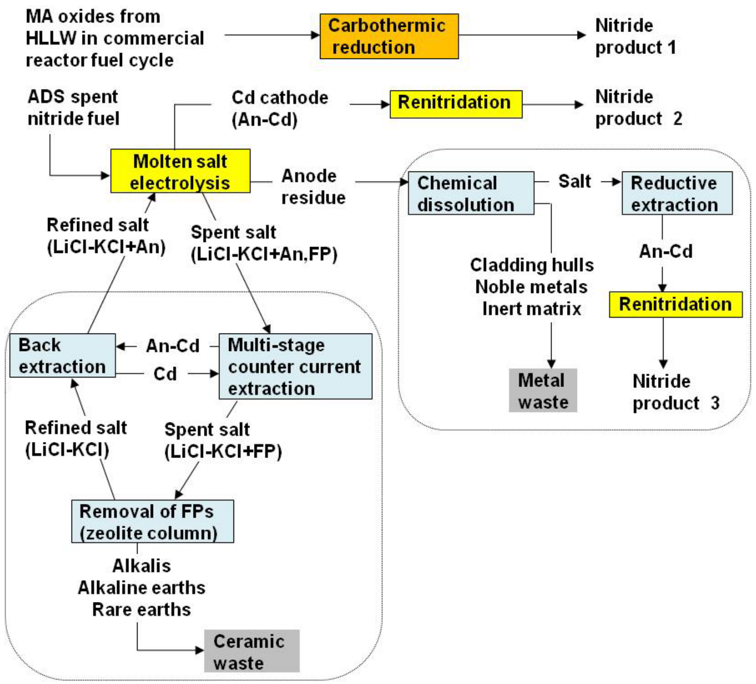
↓

Heated sample 6.3mg

Appearances of Module for TRU High Temperature Chemistry, TRU-HITEC, with high-purity Ar gas atmosphere and electrorefiner for Am and Cm samples

AmN powder

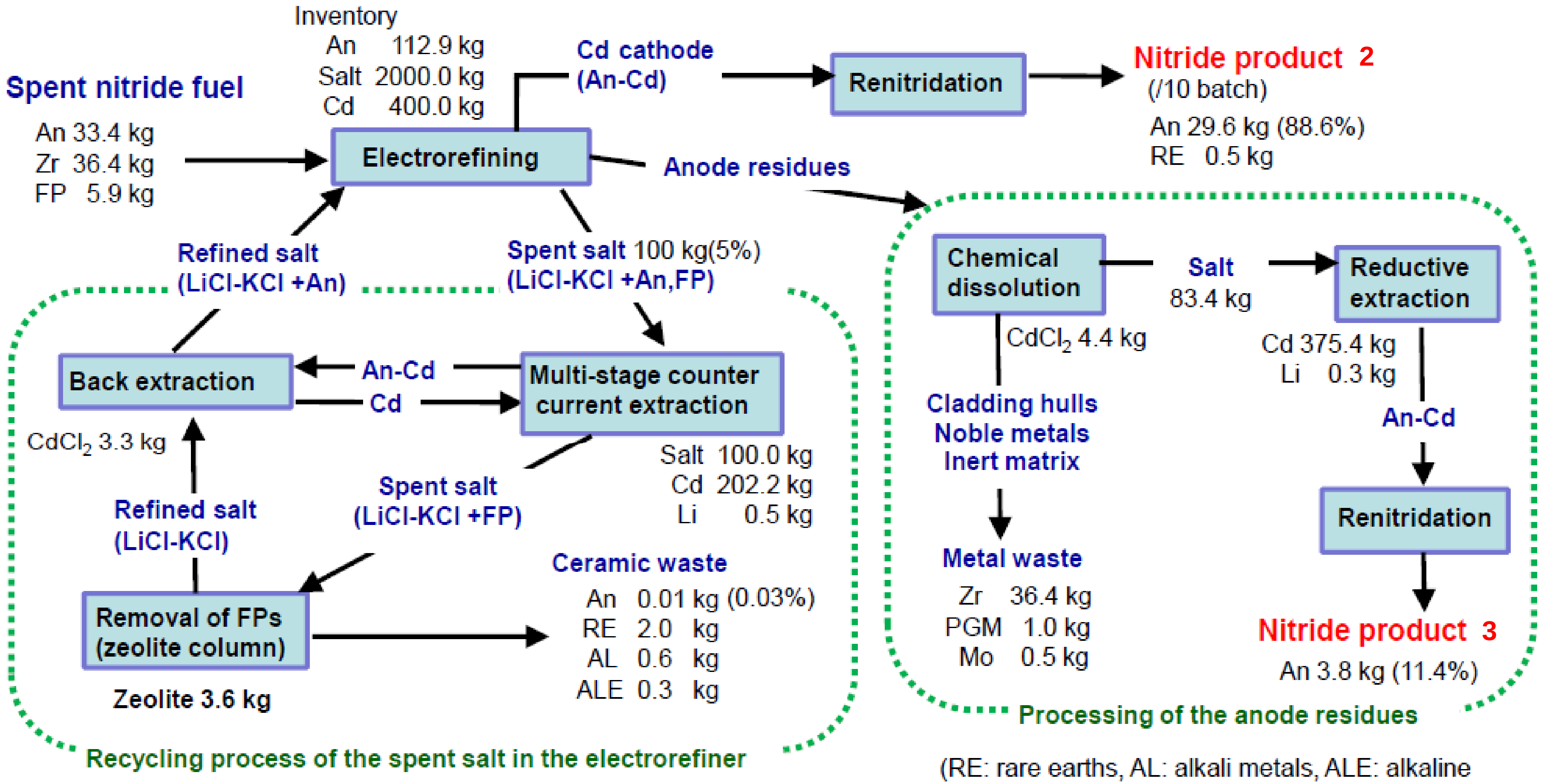
Material Balance Calculation



Calculation conditions

- 1000 kg/y of MA from HLLW of the 1st strata (which can be transmuted by 4 ADS plants)
- The number of days in operation: 200 days/year
- MA from the commercial cycle: 5.0 kg/day
- Actinides from spent fuel of ADS: 33.4 kg/day
- 5% of molten salt in the electrorefiner is treated by the additional process per day to remove REs.
- 10% of actinide elements remains as an anode residue after electrorefining process
- All of the attached salt and Cd are assumed to be returned to the process without loss after distillation.

Throughputs per Day after One Year Operation



(RE: rare earths, AL: alkali metals, ALE: alkaline earths, PGM: platinum group metals)

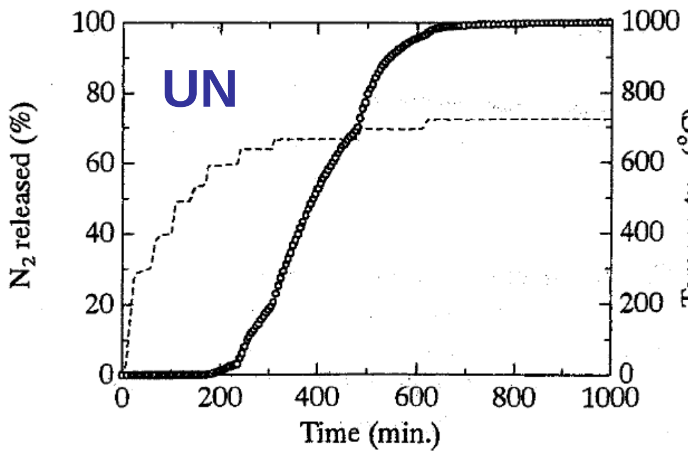
- ☐ Recovery yield of MA and Pu is 99.97% (>99.95%)
- ☐ Concentration of REs in the nitride products is 2.6wt% (<3.0wt%)

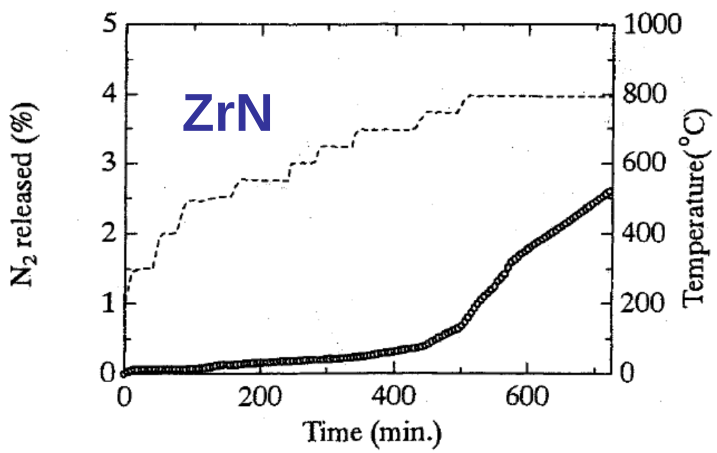
→ Satisfy the criteria of the design study of ADS

Treatment of Inert Matrix Material ZrN

- The anode residue is considered to have ZrN as a main component.
- Removal of ZrN from AnN in the anode residue is possible by chemical process.
  - ✓ ZrN is more stable than actinide nitrides.

$$2AnN + 3CdCl_2 = 2AnCl_3 + 3Cd + N_2\uparrow$$





Dissolution behaviour of nitrides by the reaction with $CdCl_2$ in LiCl-KCl

Summary

- A process flow diagram of the pyrochemical process of spent nitride fuel for ADS was established.

- Elemental technology on each process was developed by laboratory-scale experiments.

- Material balance of actinides, fission products and diluent material was calculated for the process by use of the reported data including metal electrorefining process.

- It was elucidated from the material balance calculation that the proposed pyrochemical process can satisfy the criteria of the design study of ADS.

Thank you !

Спасибо !