Development of Nitride Fuel and Pyrochemical Process for Transmutation of Minor Actinides

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Content of Presentation

(1) Introduction of nitride fuel cycle development for transmutation of minor actinides (MA)

(2) Progress of thermal property measurements on nitride fuel

- Thermal expansion, thermal conductivity, and so on
- MA nitrides and their solid solutions
- Burnup simulated nitrides
- ZrN or TiN-containing nitrides

(3) Progress of pyrochemical process for treatment of spent nitride fuel

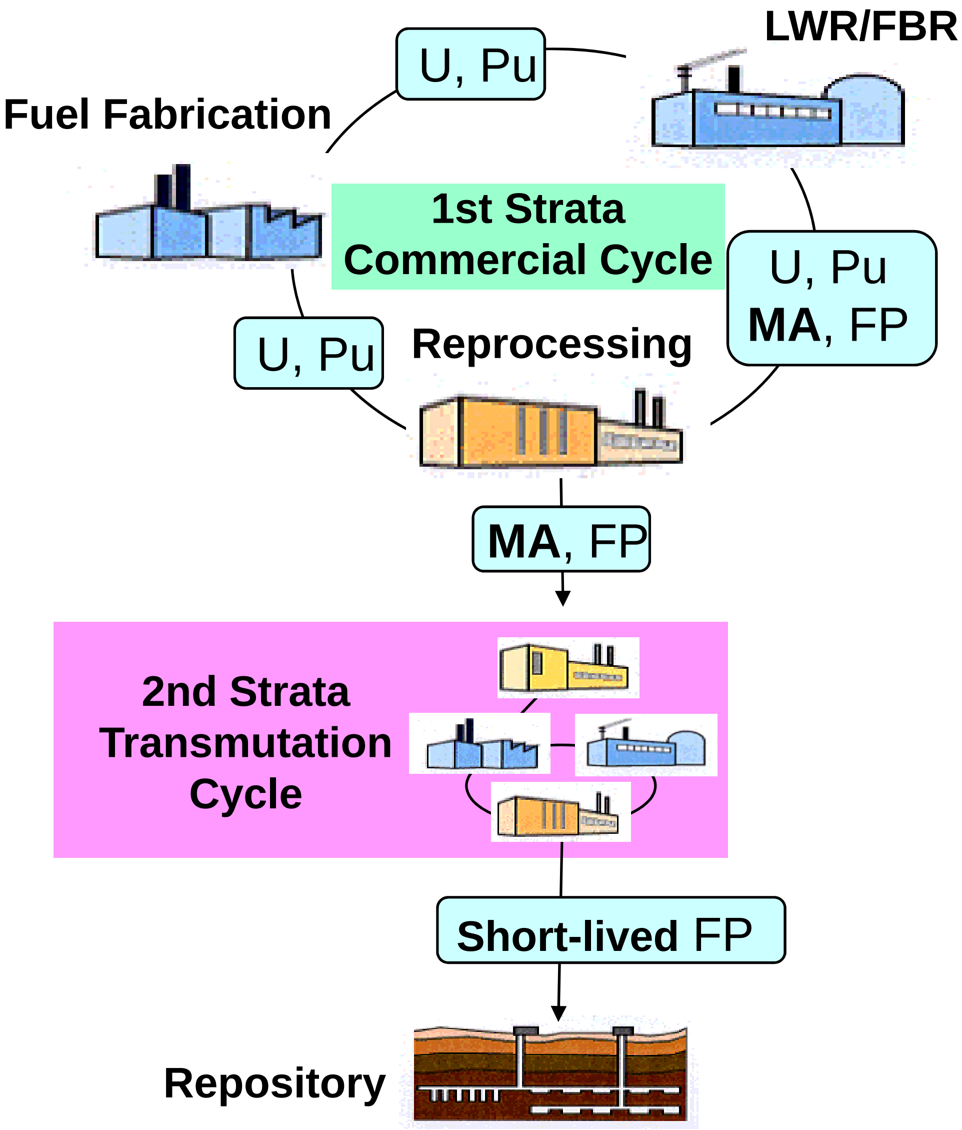
- Electrochemical behavior of nitrides in LiCl-KCl melt
- Formation of nitrides from electrodeposits in cathode

(4) Summary

Double-Strata Cycle Concept and Nitride Fuel

➢ Double-strata fuel cycle concept

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

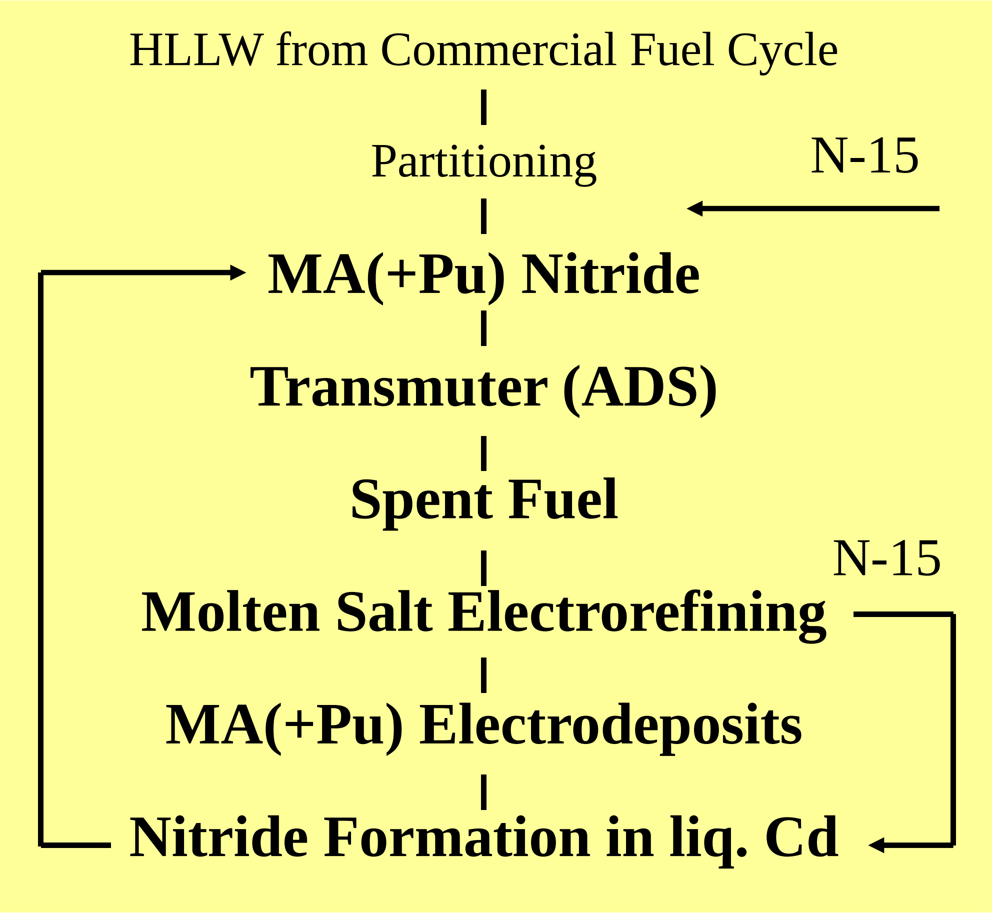


➢ Nitride fuel for MA transmutation

- High thermal conductivity, high melting point and high heavy metal density
- Mutual solubility among actinide mononitrides

R&D Activities on Nitride Fuel and Pyrochemical Process

- ✓ Preparation of MA bearing nitrides by carbothermic reduction
- ✓ Measurement of thermal properties on MA nitrides and burnup simulated nitrides
- ✓ Molten salt electrolysis for treatment of spent nitride fuel
- ✓ Formation of nitrides from electrodeposits in cathode
- ✓ Irradiation behavior of U-free nitride fuel
- ✓ N-15 related issues



Flowsheet for MA transmutation fuel cycle

Preparation of Nitride Fuel for ADS

- ✓ Nitride fuel for ADS is a so-called U-free fuel. In this case, MA are contained as a principal component and a diluent material such as ZrN is added in place of U. A composition of nitride fuel proposed by JAEA is $(MA_{0.24}Pu_{0.16}Zr_{0.6})N$.
- ✓ There are two routes for preparation of nitride fuel for ADS. One is the carbothermic reduction of MA oxides partitioned from HLLW, and the other is the nitride formation of MA and Pu recovered in liquid Cd cathode by pyrochemical reprocessing.

1) MA partitioned from HLLW

↓ *Sol-gel method*

(MA oxide + carbon) sphere

↓ *Carbothermic reduction*

MA nitride sphere →

2) (MA + Pu) in liquid Cd cathode

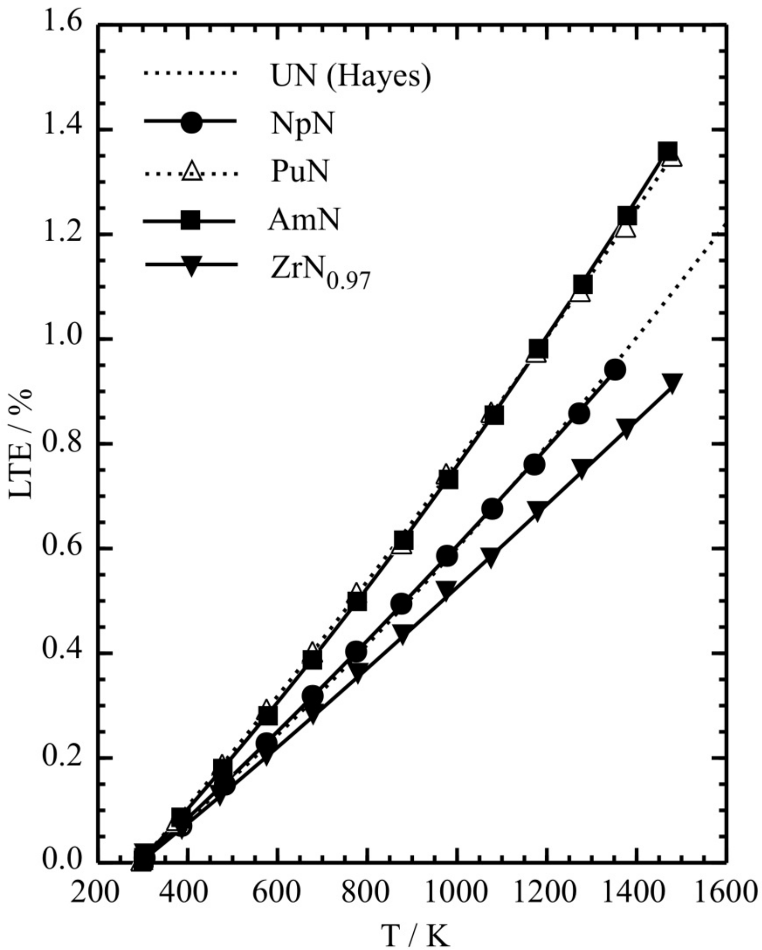
↓ *Nitridation/distillation combined reduction*

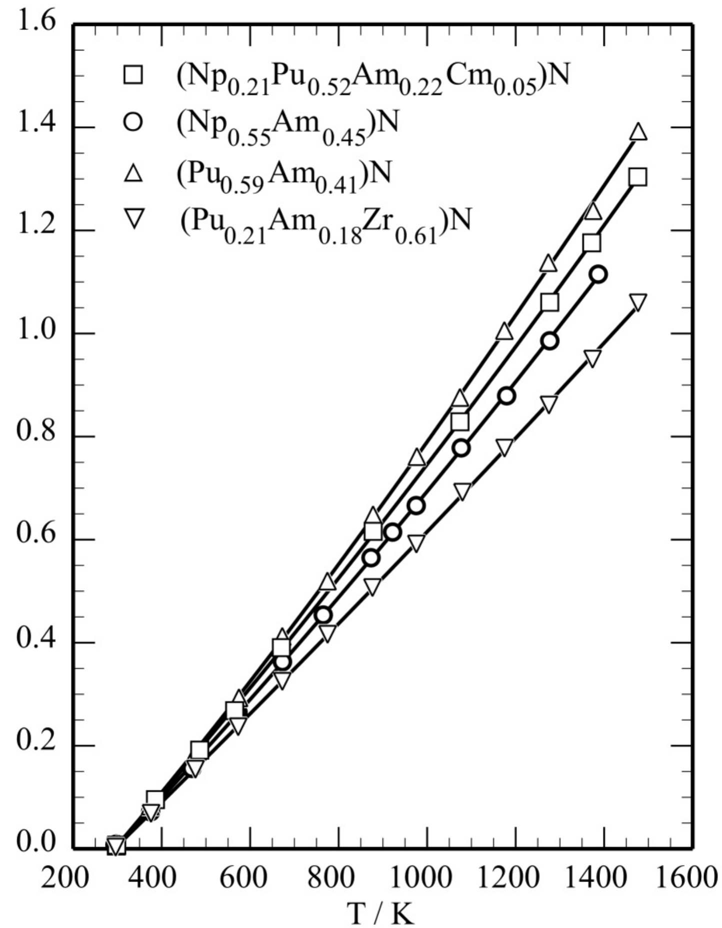
(MA + Pu) nitride powder

↓

(MA,Pu,Zr)N fuel for ADS ← ZrN

Thermal Expansion of MA Nitrides and Their Solid Solutions

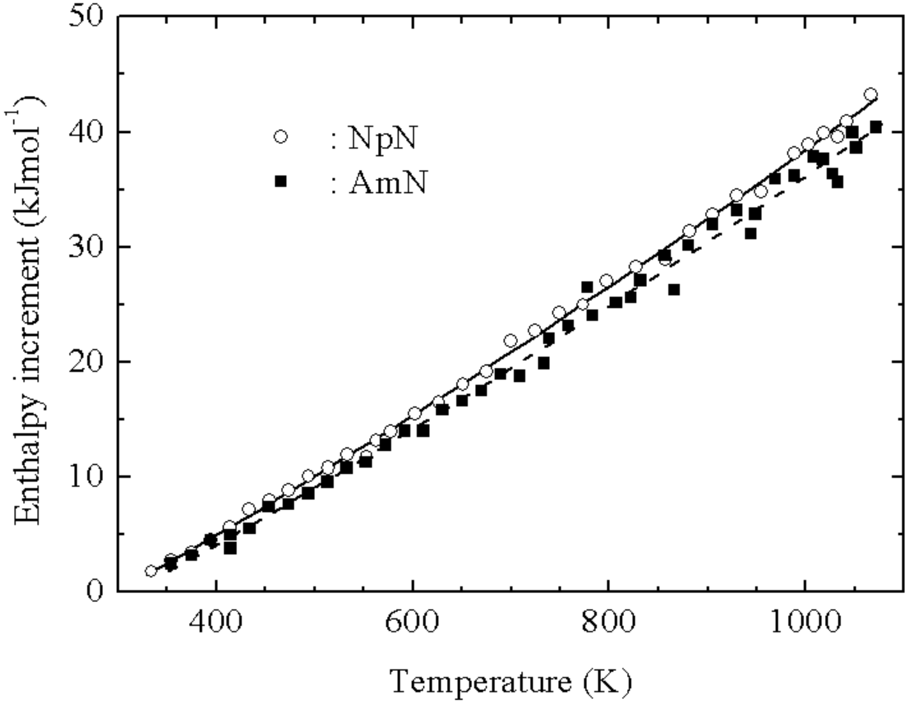


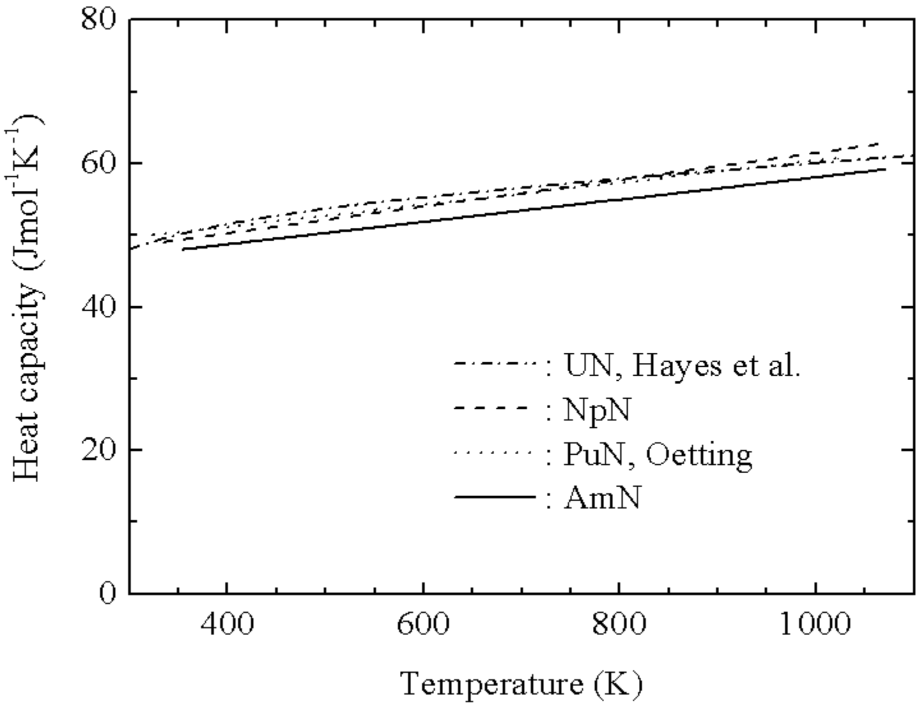


- Thermal expansions of MA nitrides and their solid solutions were measured from room temperature to 1478K by high temperature X-ray diffractometry.
- Average thermal expansion coefficients of the solid solutions could be approximated by the linear mixture rule of respective mononitrides.

Ref. M. Takano, et al (2008)

Heat Capacity of NpN and AmN

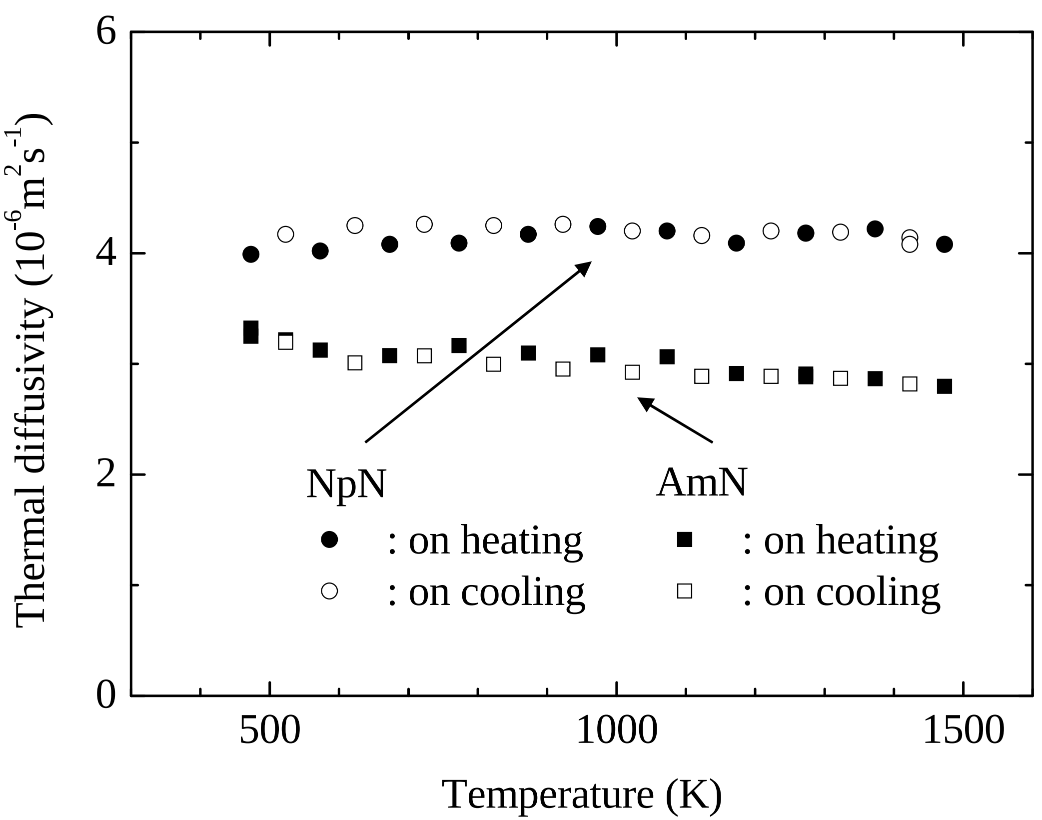


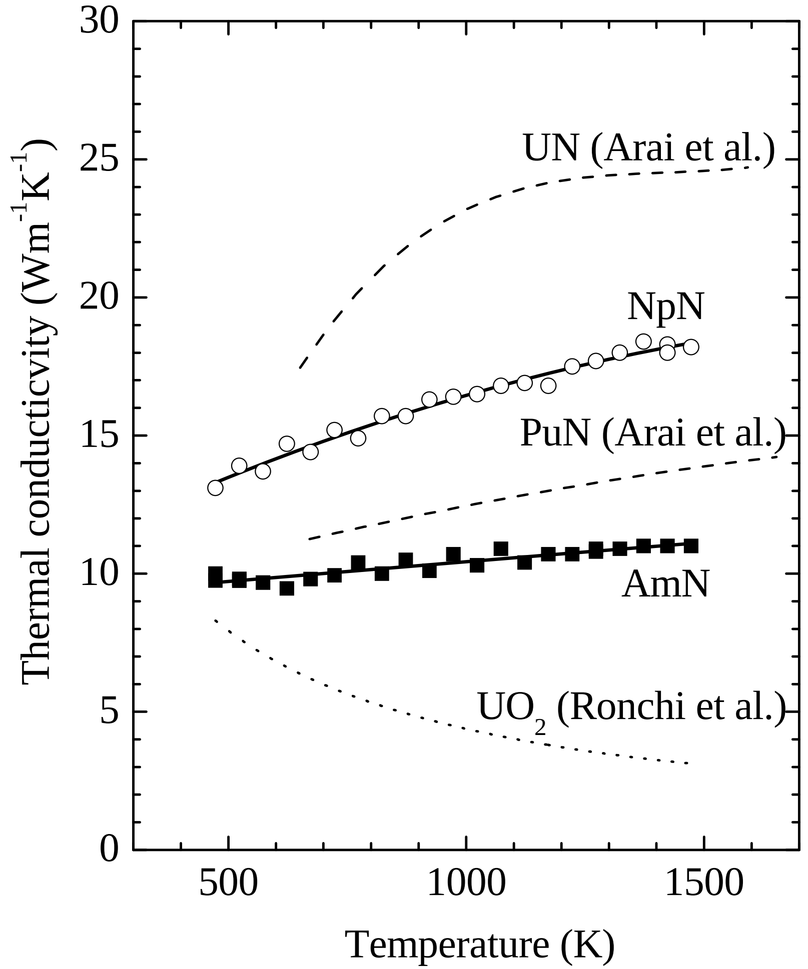


- Heat capacities of NpN and AmN were measured from 374 to 1071K by drop calorimetry and compared with literature values for UN and PuN.
- Heat capacities of actinide mononitrides had similar temperature dependence, although that of AmN was slightly smaller than those of UN, NpN and PuN.

Ref. T. Nishi, et al (2008)

Thermal Conductivity of NpN and AmN

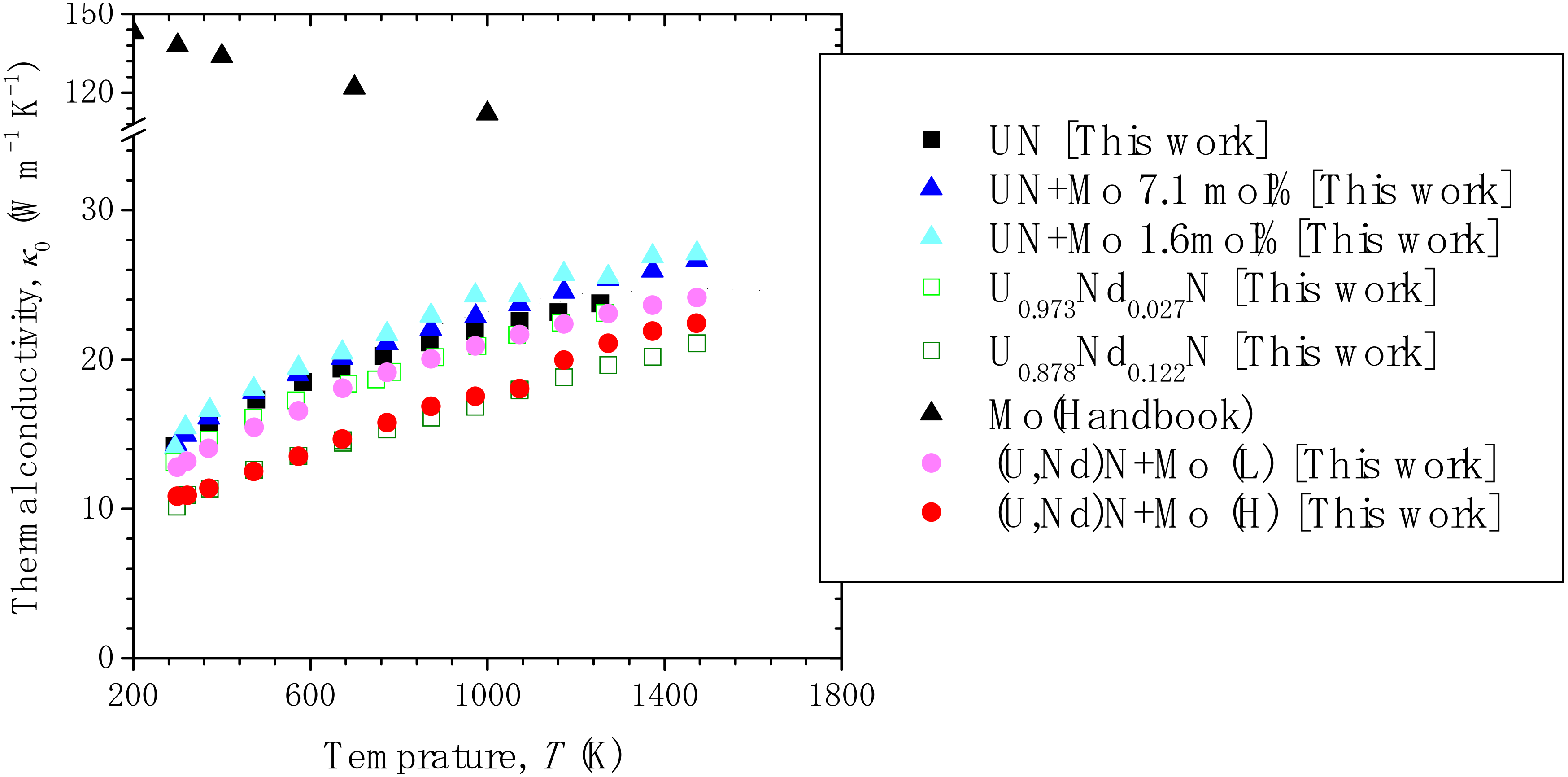




- Thermal diffusivities of NpN and AmN were measured from 473 to 1473K by laser flash method, from which thermal conductivities were derived.
- Thermal conductivities of actinide mononitrides decreased with the atomic number of actinides, although they had similar temperature dependence.

Ref. T. Nishi, et al (2008)

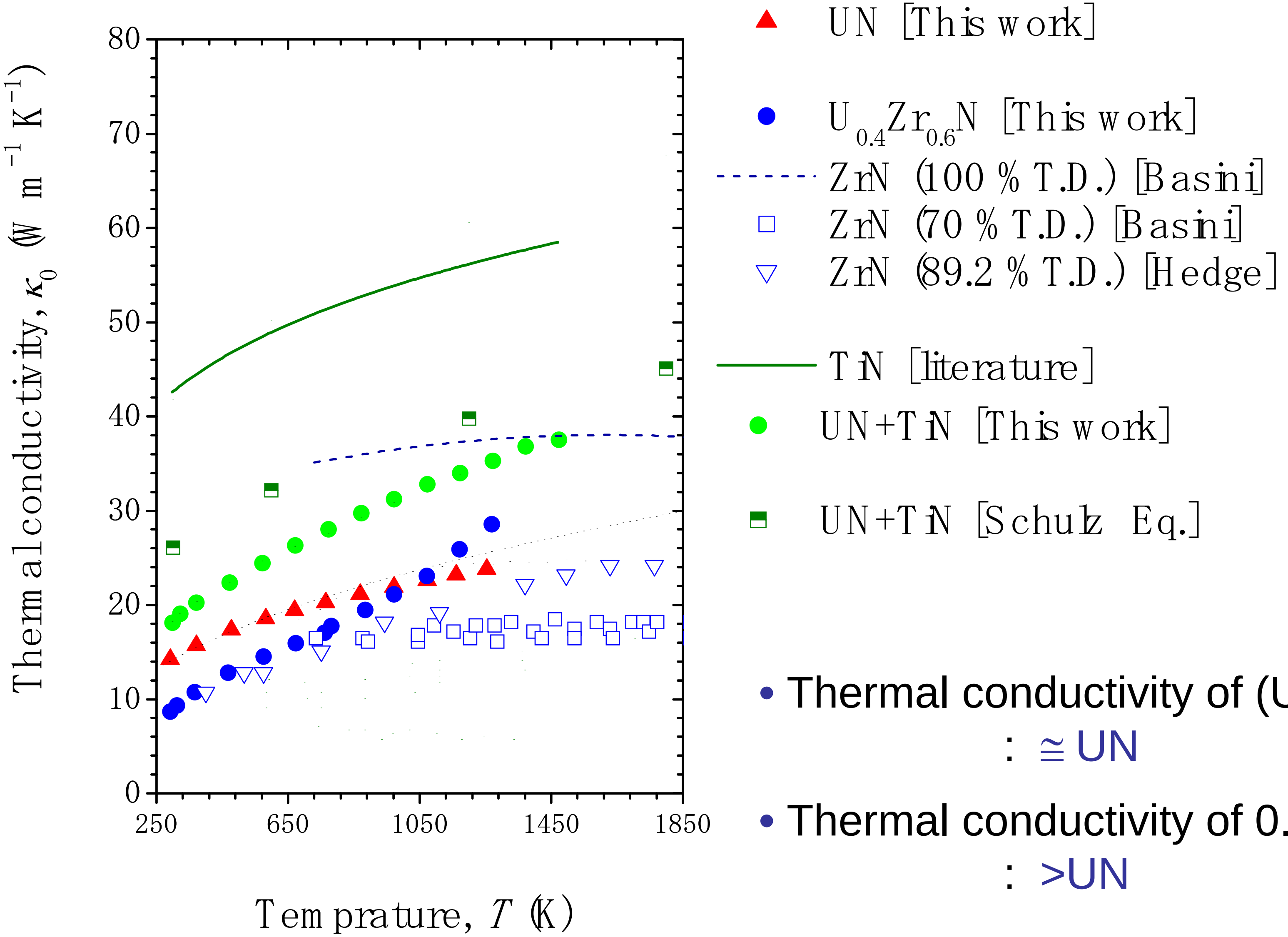
Thermal Conductivity of Burnup Simulated Nitrides



- Thermal conductivity of UN+Mo : Almost independent of Mo content
- Thermal conductivity of (U,Nd)N : Decrease with Nd content
- Thermal conductivity of (U,Nd)N+Mo : Decrease with Nd content but independent of Mo content

Ref. M. Uno, et al (2007)

Thermal Conductivity of Nitrides with Diluent Materials

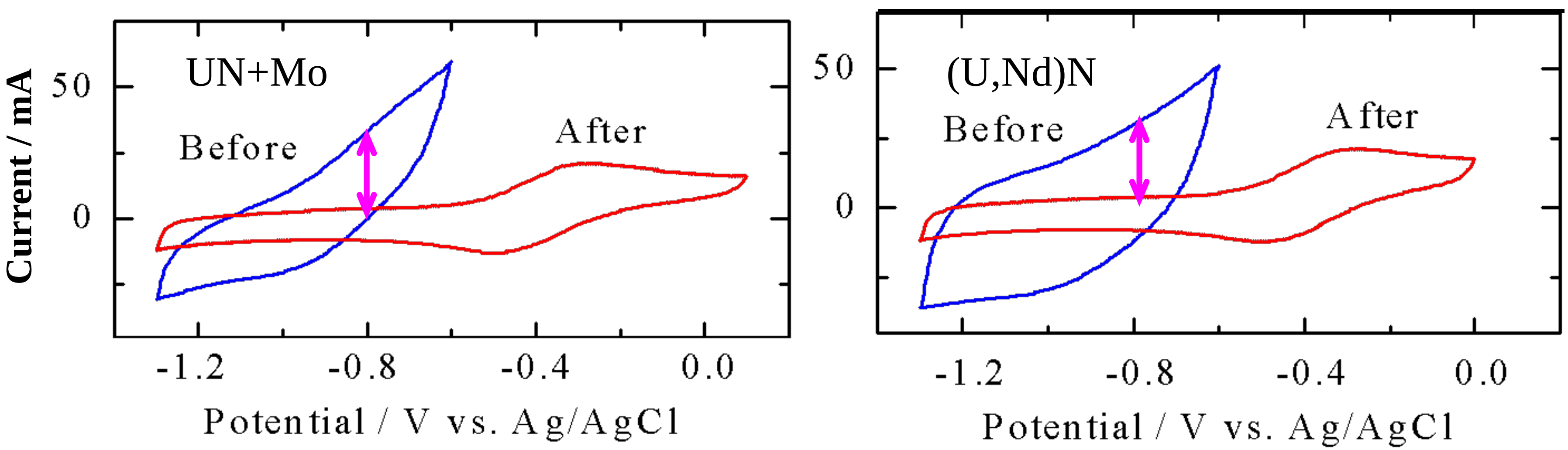


- Thermal conductivity of $(U_{0.4}Zr_{0.6})N$
  : ≅ UN
- Thermal conductivity of 0.4UN+0.6TiN
  : >UN

Ref. M. Uno, et al (2007)

Molten Salt Electrolysis of Burnup Simulated Nitrides

- ✓ **Electrochemical measurement in LiCl-KCl-UCl_3: CV and EMF**
- ✓ **Potential-controlled electrolysis using liquid Cd cathode**
  - ➢ **UN in UN+Mo and (U,Nd)N: Dissolution in LiCl-KCl at the similar potential as pure UN and recovery of U in liquid Cd cathode**
  - ➢ **Mo in UN+Mo: Remain in anode undissoved**
  - ➢ **NdN in (U,Nd)N: Dissolution but Nd almost stay in the salt phase**

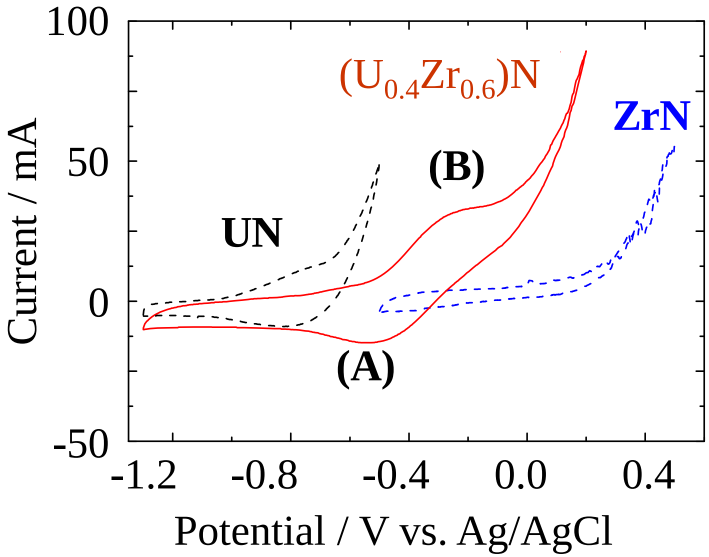


Comparison of CV before and after the electrolysis for UN+Mo and (U,Nd)N

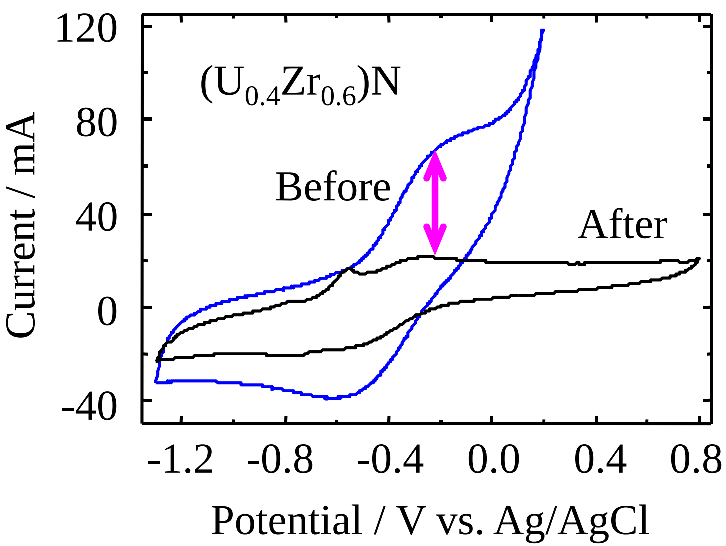
Ref. T. Satoh, et al (2007)

Molten Salt Electrolysis of Nitrides with Diluent Materials

- ✓ Electrochemical measurement in LiCl-KCl-UCl_3: CV and EMF
- ✓ Potential-controlled electrolysis using liquid Cd cathode
  - ➢ UN in (U,Zr)N: Dissolution in LiCl-KCl at more positive potential (~by 1V) than pure UN and recovery of U in liquid Cd cathode
  - ➢ UN in UN+TiN: Dissolution in LiCl-KCl at the similar potential as pure UN and recovery of U in liquid Cd cathode



Comparison of CV between UN, $(U_{0.4}Zr_{0.6})N$ and ZrN



Comparison of CV before and after the electrolysis for $(U_{0.4}Zr_{0.6})N$

Ref. T. Satoh, et al (2008)

AmN Formation Behavior in Molten Cd

Prepared by carbothermic reduction of AmO_2

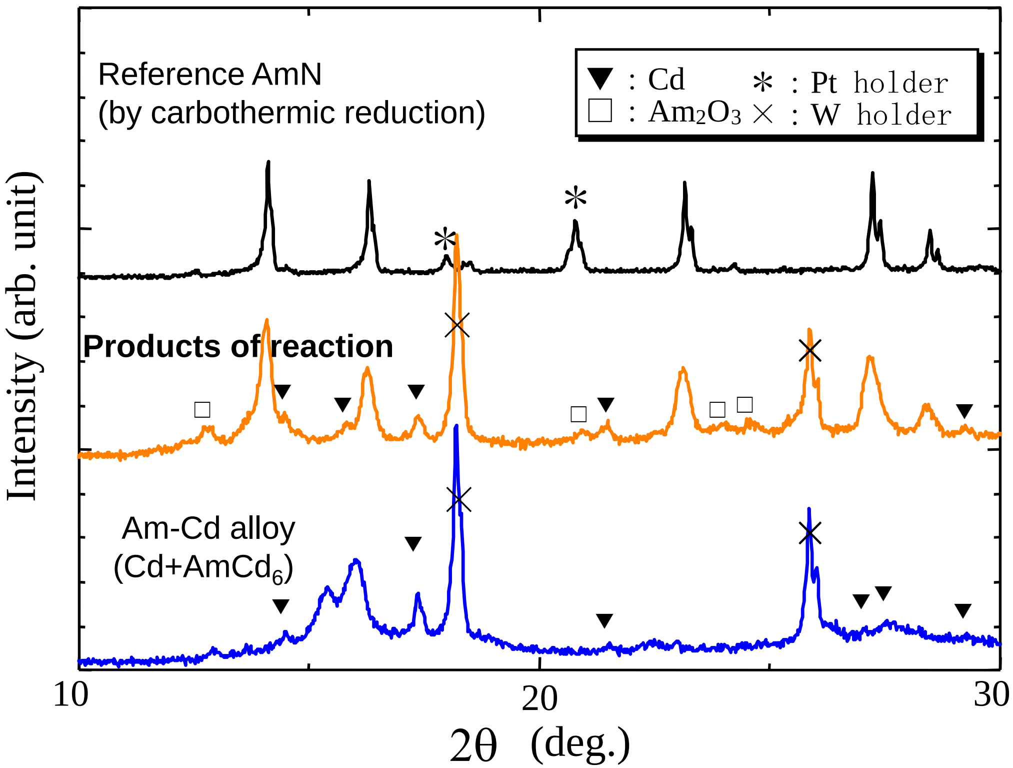
Prepared by solid-solid reaction of AmN and $CdCl_2$

(1) Electrolysis of AmN in LiCl-KCl-$AmCl_3$ melt with liquid Cd cathode

(2) Nitridation/distillation combined reaction of the electrodeposits in N_2 stream at 973K

(3) Vacuum heating at 723K for removal of residual Cd

Constituted by Cd and $AmCd_6$



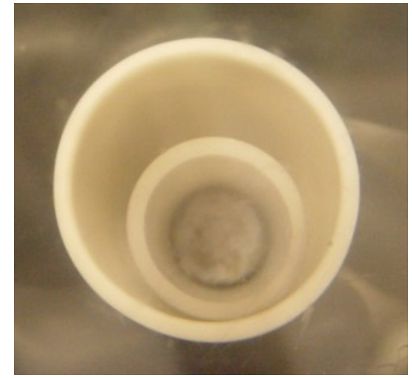
Appearance of AmN powder after heating the product in vacuum at 723K

Ref. H. Hayashi, et al (2008)

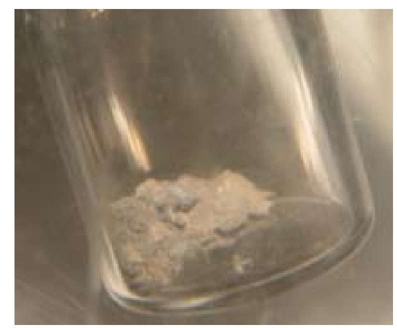
Nitride Pellet Preparation from Pyrochemical Process

(U,Pu)N pellet in Mo crucible before electrolysis

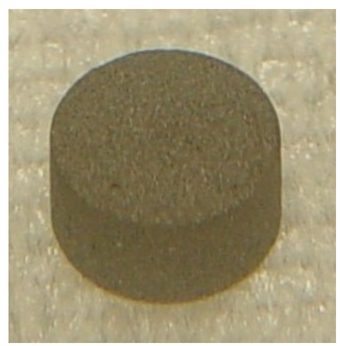
Liquid Cd cathode in Al_2O_3 crucible after electrolysis



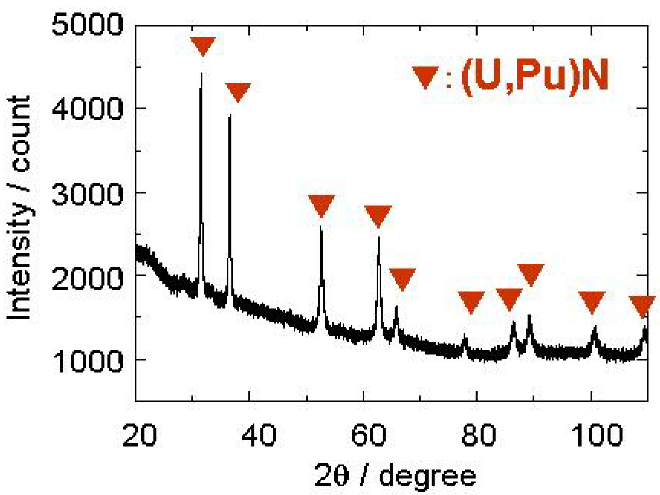
U-Pu-Cd in Y_2O_3 crucible before nitridation/distillation combined reaction



(U,Pu)N powder obtained by nitridation/distillation combined reaction



(U,Pu)N pellet prepared by sintering in flowing Ar-H_2 atmosphere at 2023K



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

Ref. Y. Arai, et al (2008)

Summary

- ✓ For demonstrating technical feasibility of nitride fuel cycle for transmutation of MA, fundamental study has been carried out continuously by use of MA, Pu and burnup simulated nitrides.
- ✓ Thermal properties, such as thermal expansion, heat capacity and thermal conductivity, of MA nitrides and burnup simulated nitrides have been almost clarified. Measurements on MA nitrides with a diluent material such as ZrN or TiN are ongoing.
- ✓ As for pyrochemical process for the treatment of spent nitride fuel, molten salt electrolysis and actinide-renitridation process have been investigated. Material balance of actinides throughout the process is to be clarified in a laboratory scale.
- ✓ Further study shall include the preparation of thermal and thermodynamic database on MA nitride fuel cycle in addition to ongoing experimental study, which will contribute to the progress of design study and evaluation of fuel behavior.

Acknowledgement

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