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JAEA's Activities on Nitride Fuel Research for MA Transmutation

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Nitride Fuel Development in Japan

- **Dedicated fuel for transmutation of long-lived MA**

  R&D on Accelerator-Driven System (ADS) for MA transmutation is in progress in JAEA. The first candidate of the fuel material is U-free nitride fuel, such as (MA,Pu)N-ZrN.

- **Advanced fuel for Gen.-IV type fast reactors (FRs)**

  In the “Feasibility Study on Commercialized Fast Reactor Cycle Systems” (FS) in Japan, (U,Pu,MA)N is considered as a candidate fuel for GFR and LFR.

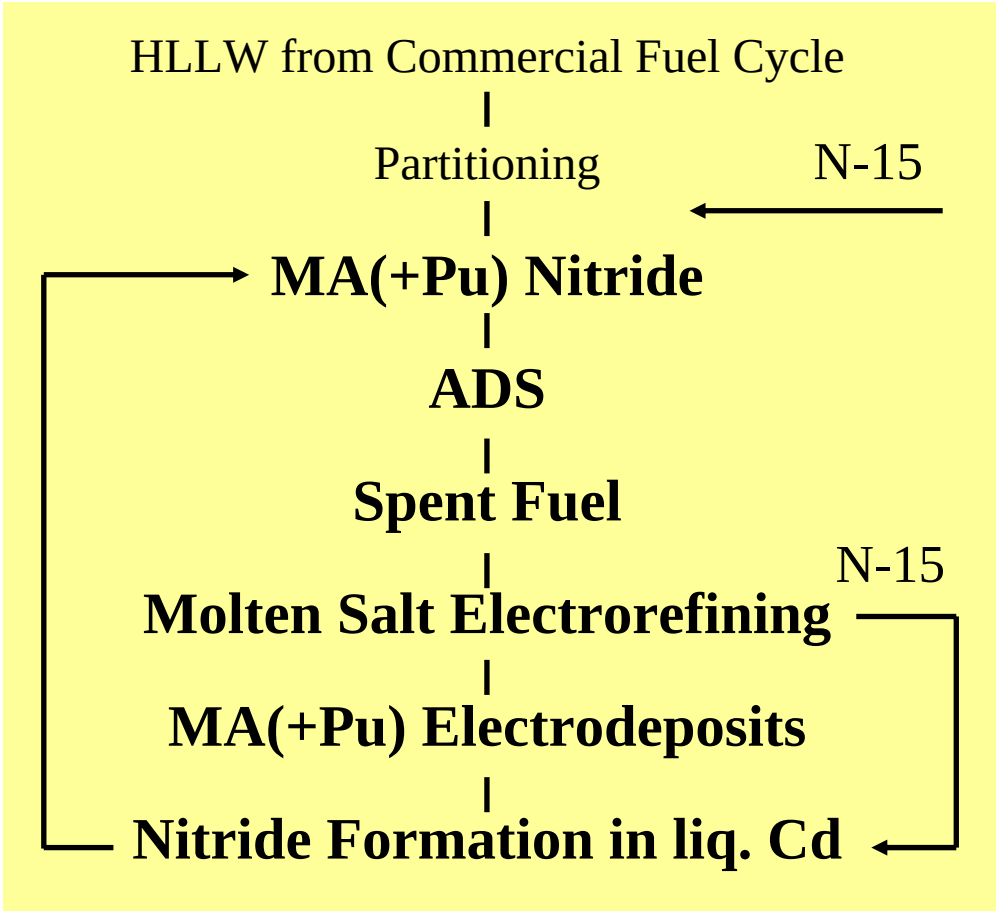
⇩

R&D on nitride fuel has been shifted from FRs to ADS.

Study on MA Transmutation Fuel Cycle in JAEA

Contents of Presentation R&D on

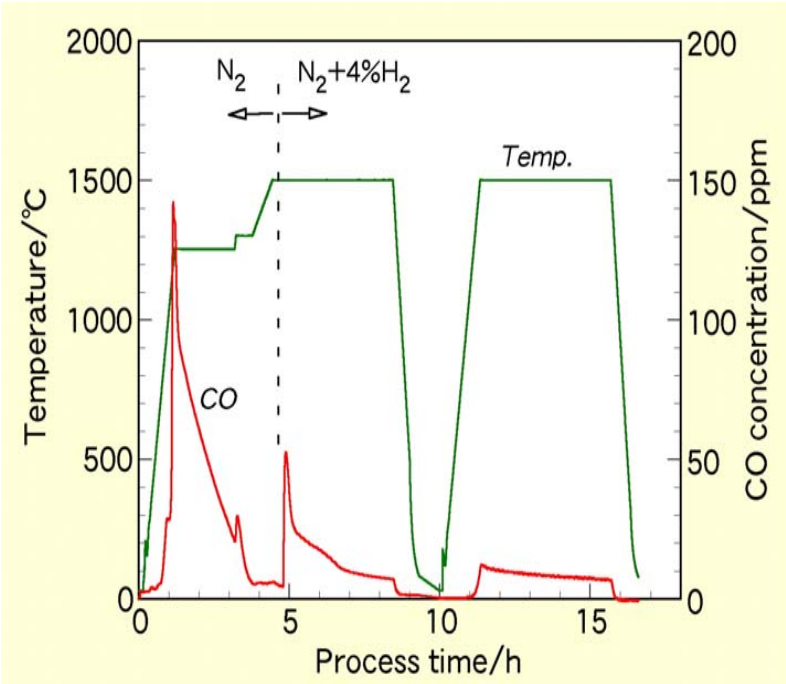
- ✓ **Fabrication of MA bearing nitrides by carbothermic reduction**
- ✓ **Property measurements on MA bearing nitrides and burnup simulated nitrides**
- ✓ **Pyrochemical process for treatment of spent nitride fuel**
- ✓ **Nitride formation behavior of actinides in liquid Cd cathode**



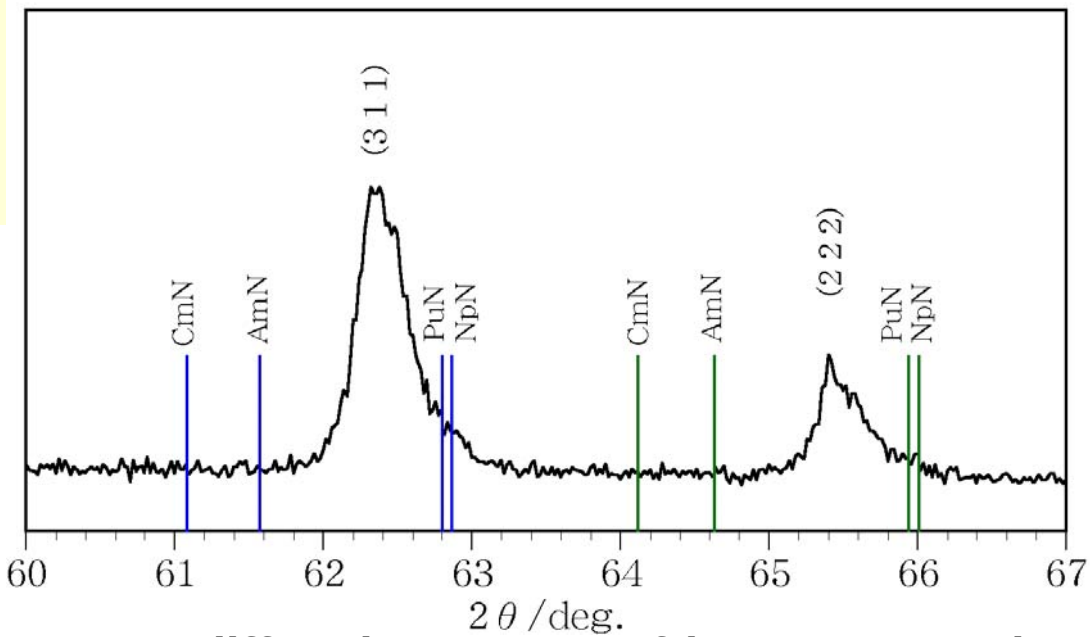
Flowsheet for MA transmutation fuel cycle based on double-strata concept

Preparation of MA Nitride Fuel

- Preparation of MA nitrides by carbothermic reduction -



- ✓ Reduce loss of Am by evaporation
  - ⟹ Lowering temperature of reduction
- ✓ Reduce O and C impurities in nitride
  - ⟹ Controlling C/MO_2 mixing ratio
- ✓ Confirm formation of solid solution
  - ⟹ Single phase of (Np,Pu,Am,Cm)N



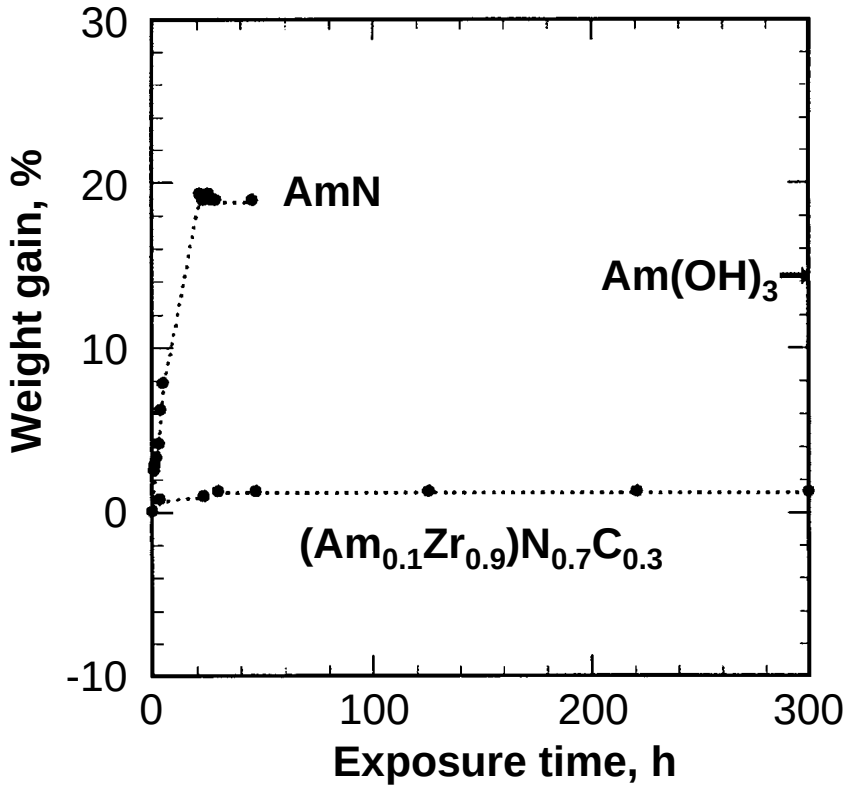
X-ray diffraction pattern of (Np,Pu,Am,Cm)N

Preparation of MA Nitride Fuel with ZrN

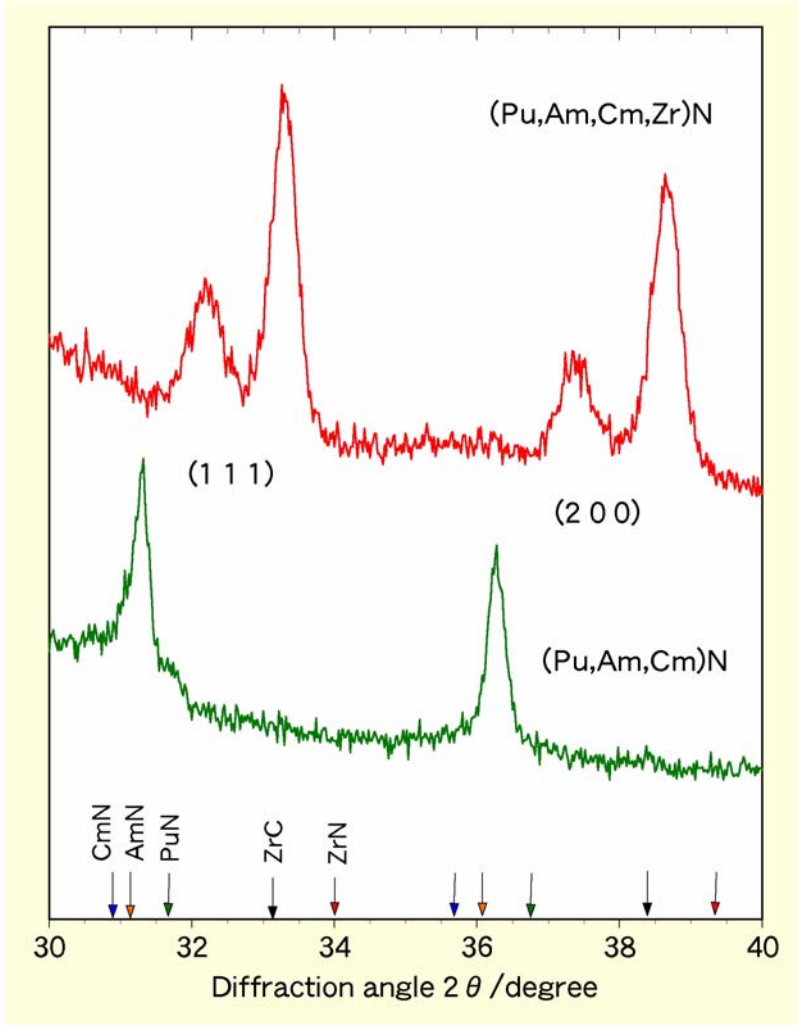
- Effects of ZrN added as a diluent material in (MA,Zr)N-

Formation of two phases ⇒
(Depending on composition)

Improvement of chemical stability ⇓



Weight gain of AmN and (Am,Zr)N exposed in air at room temperature



XRD pattern of (Pu,Am,Cm)N and (Pu,Am,Cm,Zr)N

Preparation of Nitride Pellets without MA

(1) Preparation of (Dy,Zr)N pellets for screening test of sintering aid

- ✓ Sintering temperature of MA nitride pellets should be lower than 1873K for preventing the loss of Am by evaporation.
- ✓ Pd, Ni, YH_2, AlN, Si_3Ni_4, and Y_2O_3 powders were tested for sintering aid.
- ✓ $(Dy_{0.4}Zr_{0.6})N$ pellets with ~90%TD were prepared by sintering at 1823K in N_2 gas stream by use of AlN as sintering aid.

(2) Preparation of UN-based burnup simulated pellets in which Nd, Mo or Pd were added and UN pellets containing ZrN as a diluent material

⇒ (U,Nd)N, UN+Mo, UN+Pd and (U,Zr)N pellets were prepared for thermal, mechanical and electrochemical property measurements.

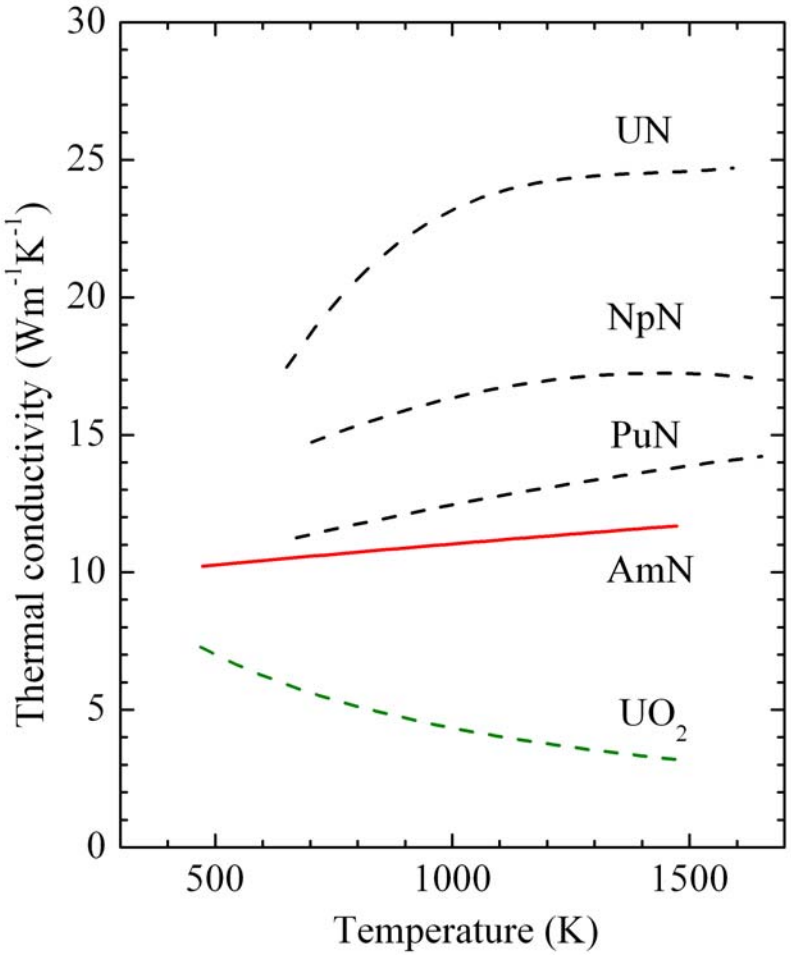
(3) Preparation of PuN pellets containing ZrN or TiN as a diluent material

⇒ (Pu,Zr)N and PuN+TiN pellets were prepared for irradiation tests and electrochemical property measurements.

Property Measurements of MA Nitride (1)

- Determination of thermal conductivity of AmN -

- ✓ AmN disk with 78%TD was prepared by sintering at 1823K in N_2-H_2 stream.
- ✓ Thermal diffusivity of AmN was measured by laser flash method from 373 to 1473K for the first time.
- ✓ Thermal conductivity was determined from thermal diffusivity of AmN together with heat capacity of PuN.
- ✓ Thermal conductivity of actinide mononitrides decreases with atomic number of actinides and gradually increases with temperature in the temperature range investigated.

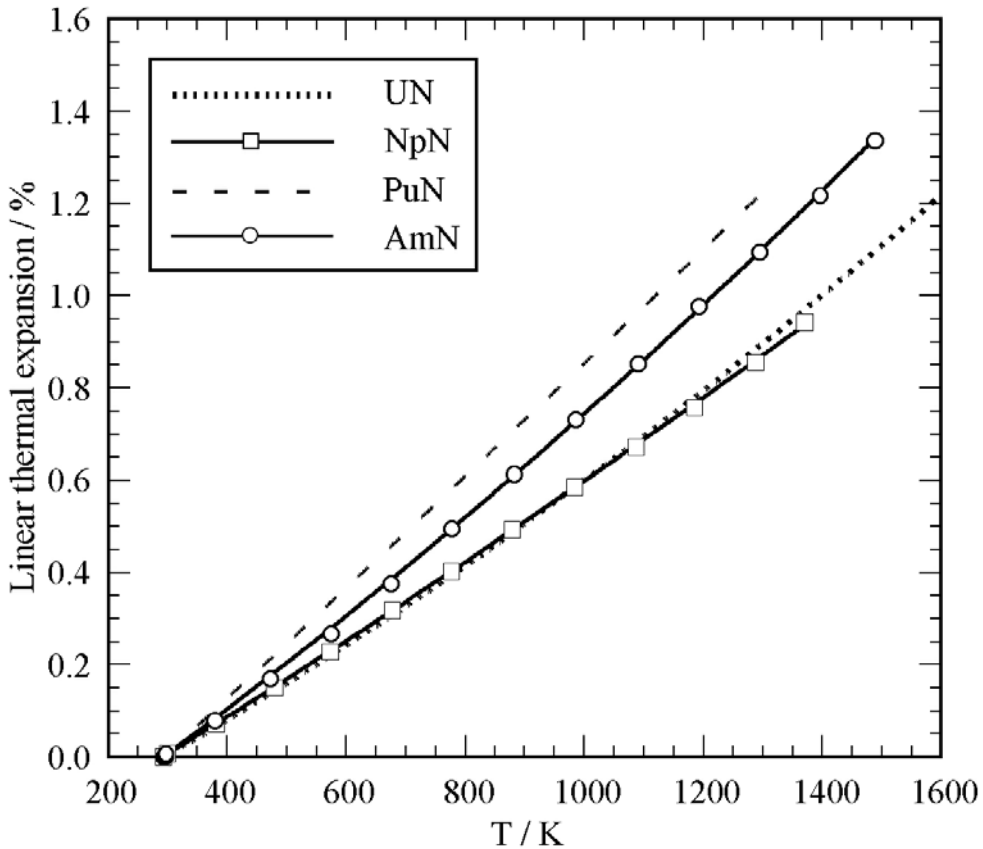


Thermal conductivity of actinide mononitrides corrected to 100%TD

Property Measurements of MA Nitride (2)

- Determination of thermal expansion of NpN and AmN -

- ✓ **Thermal expansion of NpN and AmN was measured by high-temperature X-ray diffractometry.**
- ✓ **Thermal expansion of NpN almost agreed with UN, whereas that of AmN was larger than UN and comparable to PuN.**
- ✓ **Measurements on thermal expansion of (Np,Am)N and (Pu,Am)N are underway.**
- ✓ **Besides thermal diffusivity and thermal expansion, heat capacity measurements of MA nitrides have been started recently.**



Thermal expansion of NpN and AmN in comparison with UN and PuN

Pyrochemical Process of Spent Nitride Fuel

- **Electrodissolution at Anode**

$$AnN = An^{3+} + 3e^{-} + 0.5N_2$$

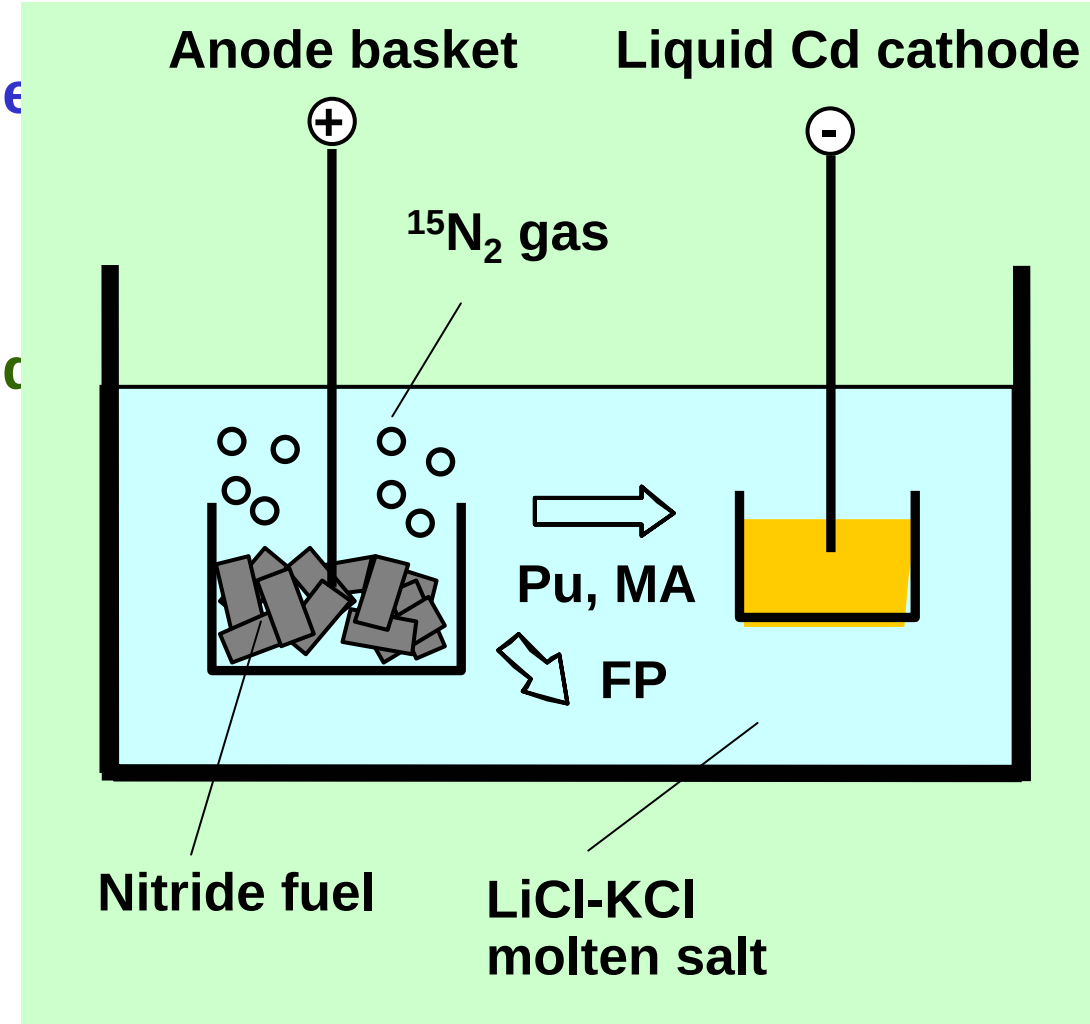
- **Electrodeposition at Cathod**

$$An^{3+} + 3e^{-} + xCd = AnCd_x$$

- **Nitride Formation from Electrodeposits**

$$AnCd_x + 0.5N_2 = AnN + xCd$$

An = (MA, Pu)

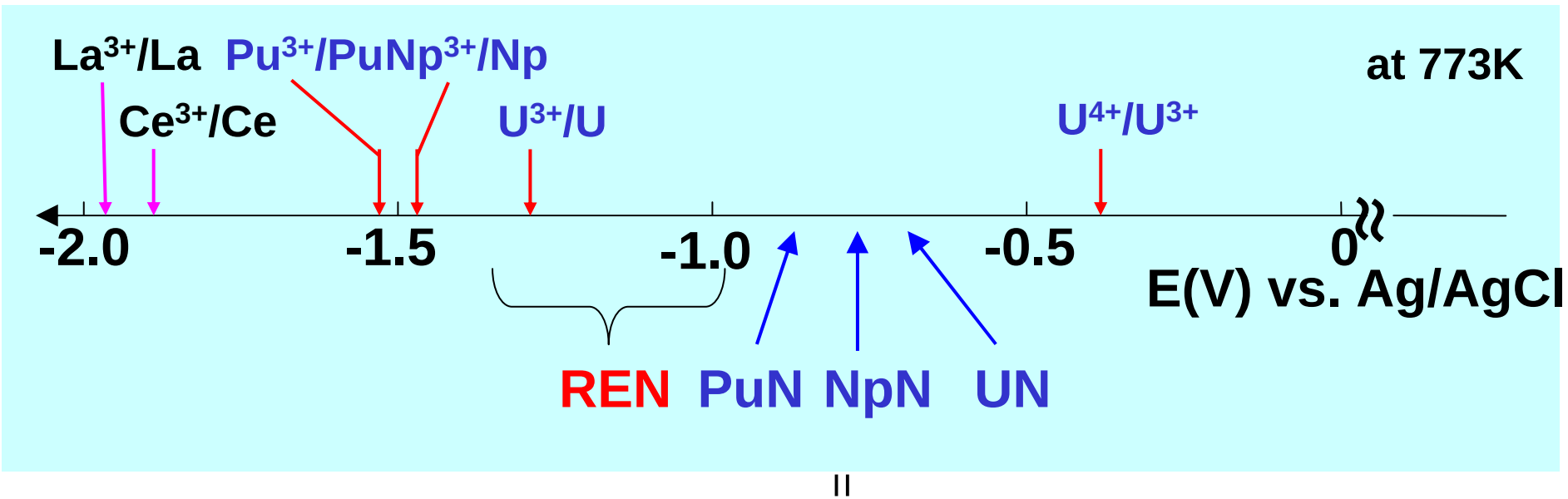


Schematic of Pyroprocess for Spent Nitride Fuel

Electrochemical Behavior of Actinide Mononitrides

- Equilibrium of $LiCl\text{-}KCl\text{-}AnCl_3$ system (An: actinides)

$An = An^{3+} + 3e^-$



- Potential shift owing to the formation of actinide nitride (~0.7V)

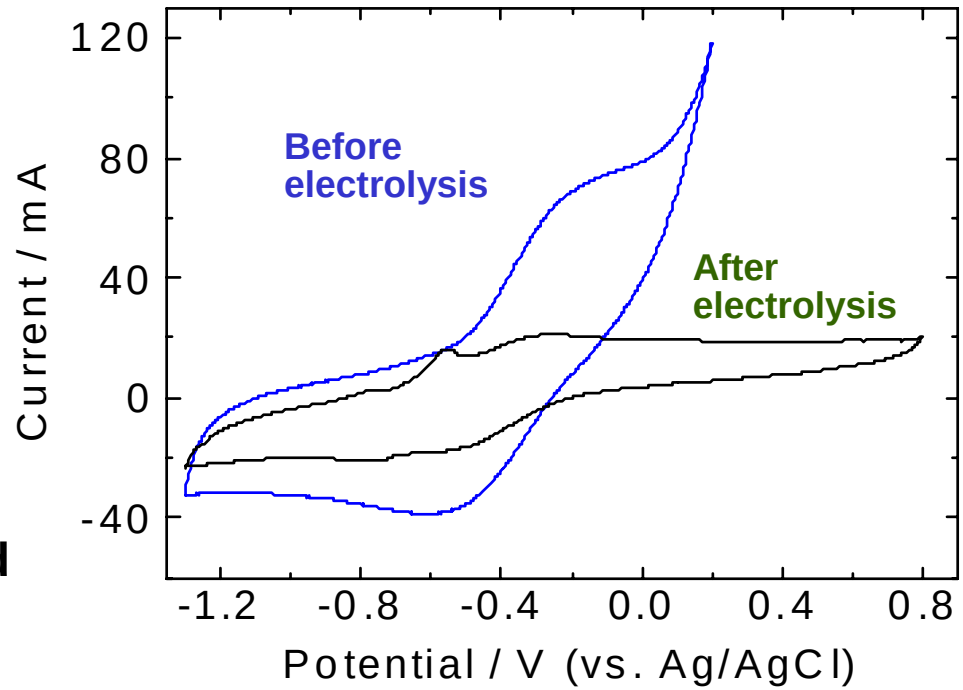
$An = An^{3+} + 3e^-$

$AnN = An + 0.5\ N_2$ … ΔG°_f (Free energy of formation for

Electrochemical Behavior of (U,Zr)N and (Pu,Zr)N

- **Since UN and PuN were stabilized by the formation of (U,Zr)N and (Pu,Zr)N solid solutions, higher anode potential than applied to UN and PuN was necessary for the dissolution. However, U, Pu and Zr could be recovered in liquid Cd cathode by the potential controlled electrolysis.**

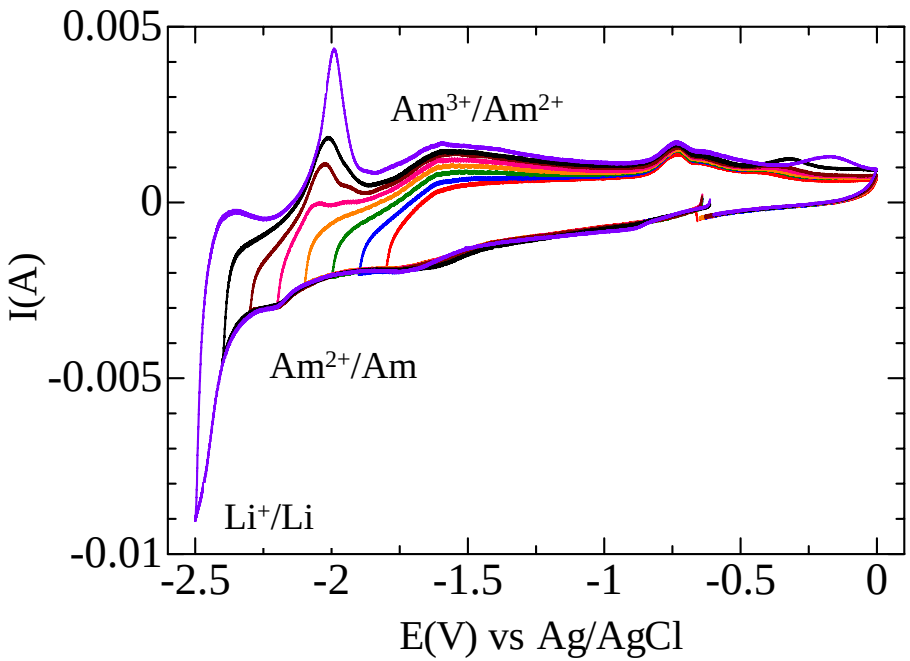
Appearance of (Pu,Zr)N anode used for the cyclic voltammetry and the potential controlled electrolysis

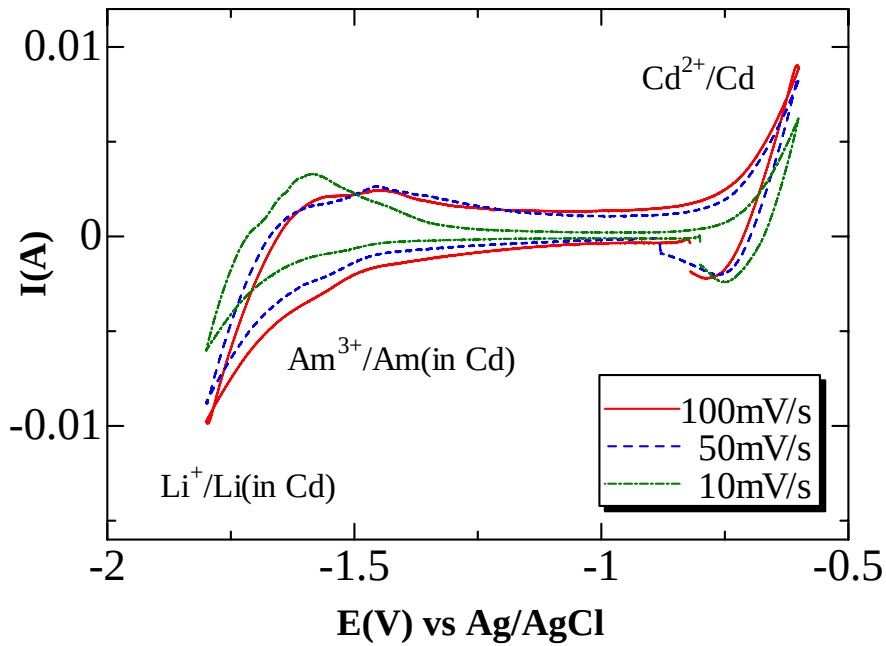


Cyclic voltammograms of (U,Zr)N in LiCl-KCl melt before and after the electrolysis

Electrode Reaction of Am in LiCl-KCl-$AmCl_3$ Melt

- **Electrode reaction of Am in LiCl-KCl-$AmCl_3$ melt was investigated by use of solid Mo and liquid Cd electrodes. Redox reactions of Am(III)/Am(II) and Am(II)/Am were observed at solid Mo electrode, while only redox reaction of Am(III)/Am was observed at liquid Cd electrode.**

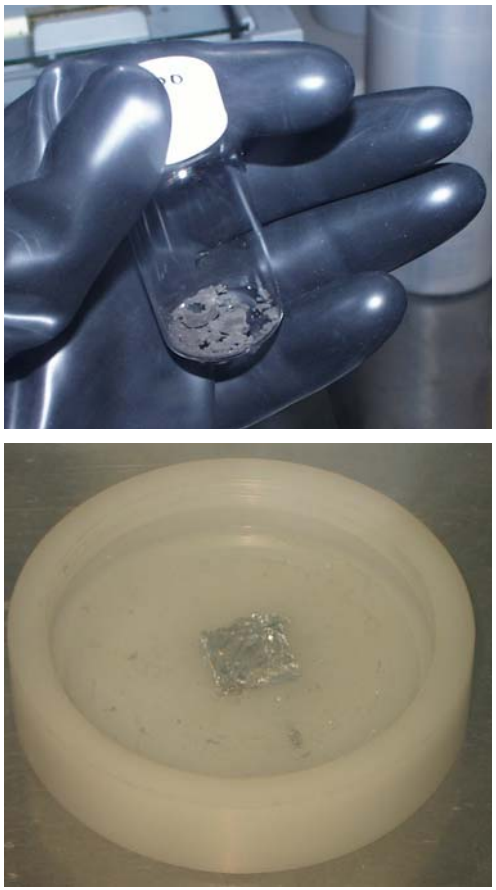




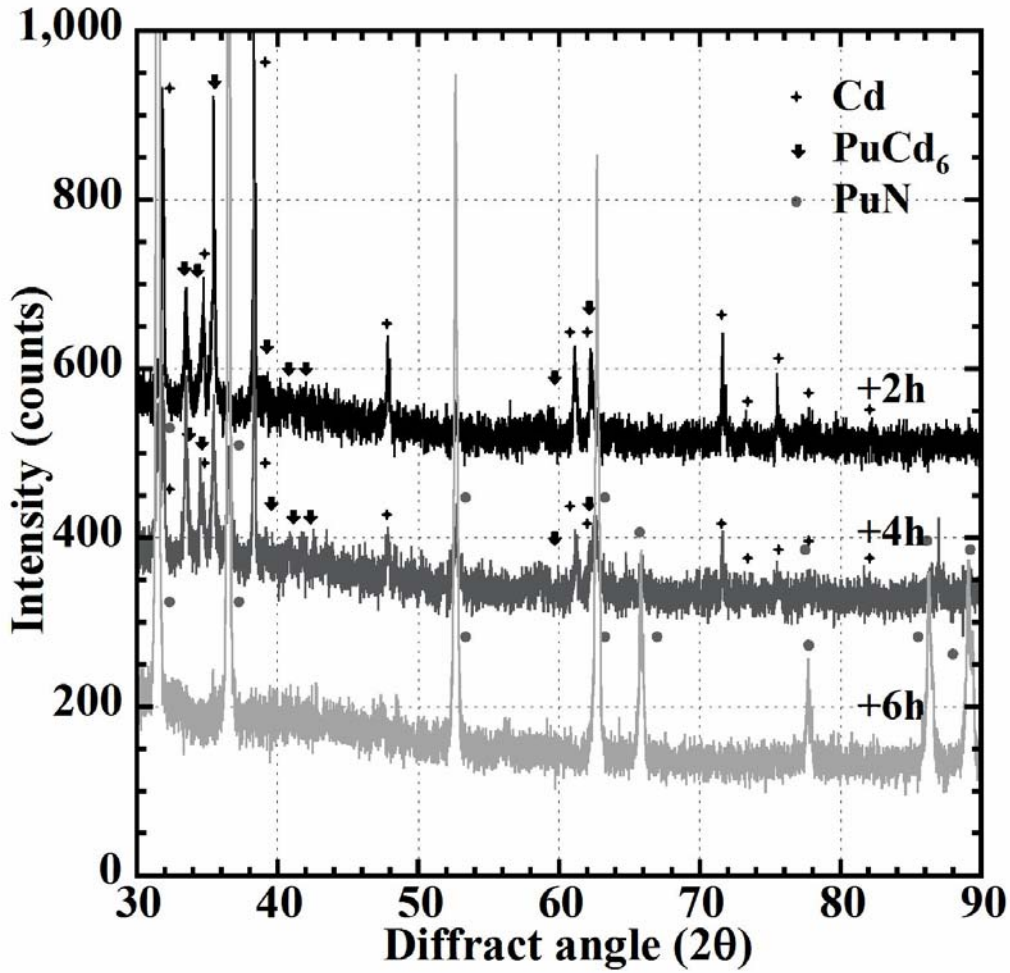
Cyclic voltammograms of LiCl-KCl-$AmCl_3$ with solid Mo (left) and liquid Cd (right) electrodes at 723K

Nitridation-Distillation Reaction for Pu-Cd

- Heating Pu-Cd alloy (Pu; 12wt.%) at 973 K in flowing N_2 gas -



Appearance of Pu-Cd alloy (lower) and PuN powder recovered (upper)



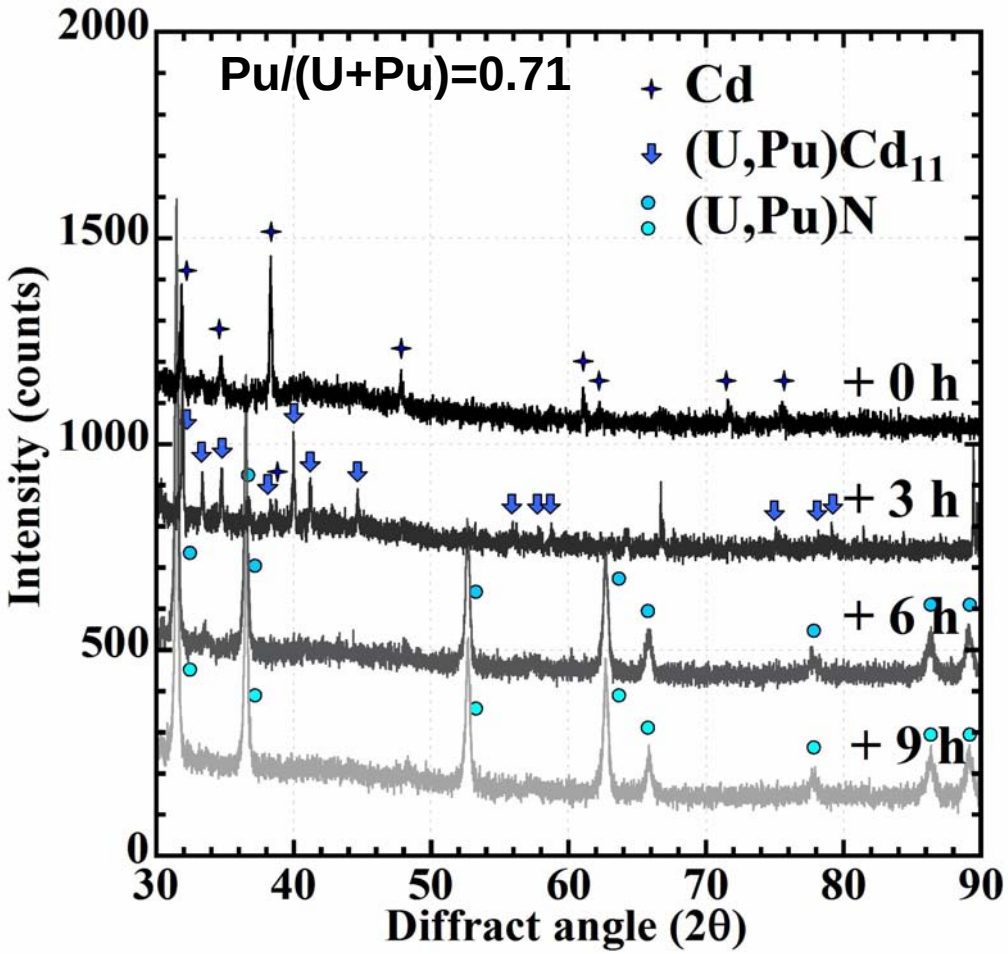
Change of XRD pattern during reaction

Nitridation-Distillation Reaction for Pu-U-Cd

- Heating Pu-U-Cd alloy with small amount of RE at 973 K in flowing N_2 ga

Appearance of (U,Pu,RE)-Cd alloy

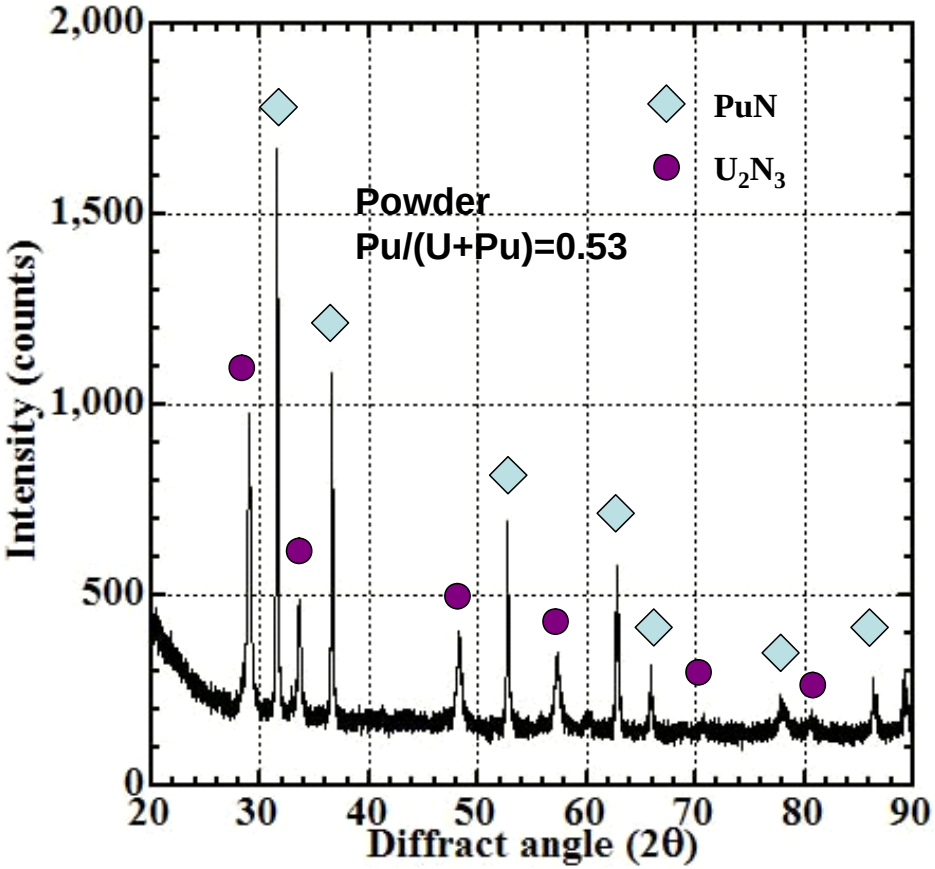
Concentration in Cd (wt.%)	
U	2.88
Pu	6.93
La	0.016
Ce	0.101
Pr	0.105
Nd	0.251



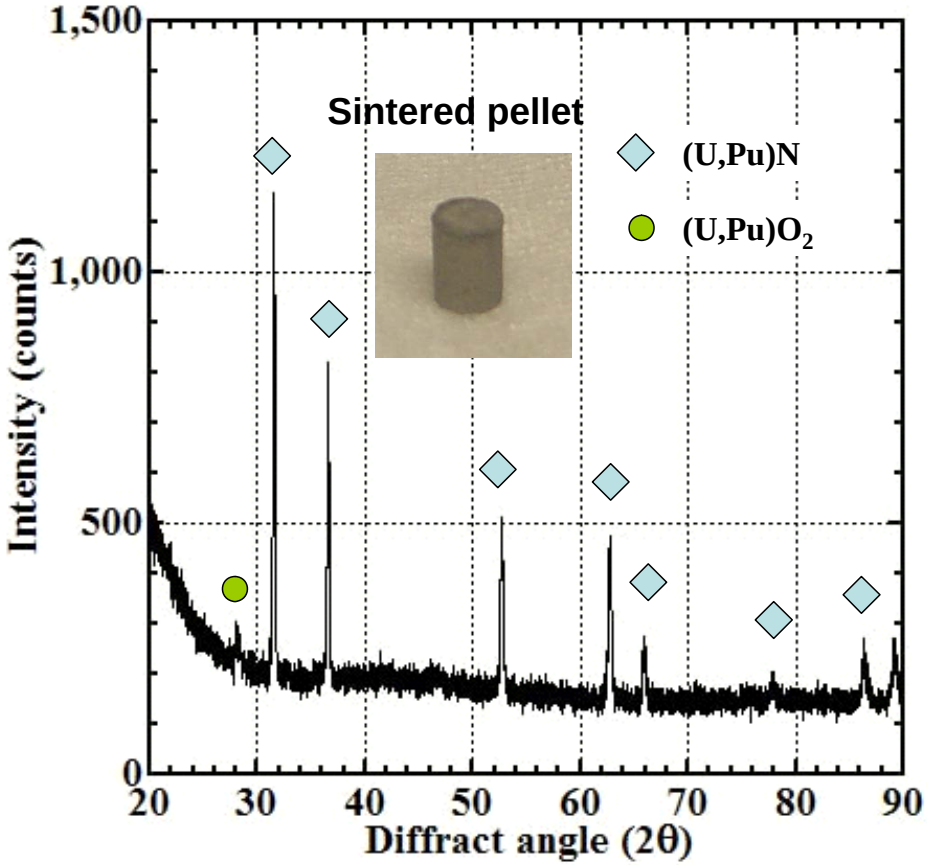
Change of XRD pattern during reaction

Pellet Preparation from Nitride Powder Recovered

- Nitride fuel fabrication in pyrochemical process -



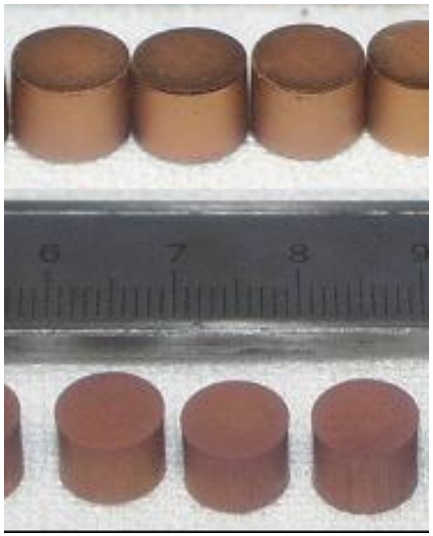
XRD pattern of powder obtained by the nitridation-distillation combined method



XRD pattern of pellet sintered at 1973K in Ar-H_2 mixed gas stream

Irradiation Test of U-Free Nitride Fuel in JMTR

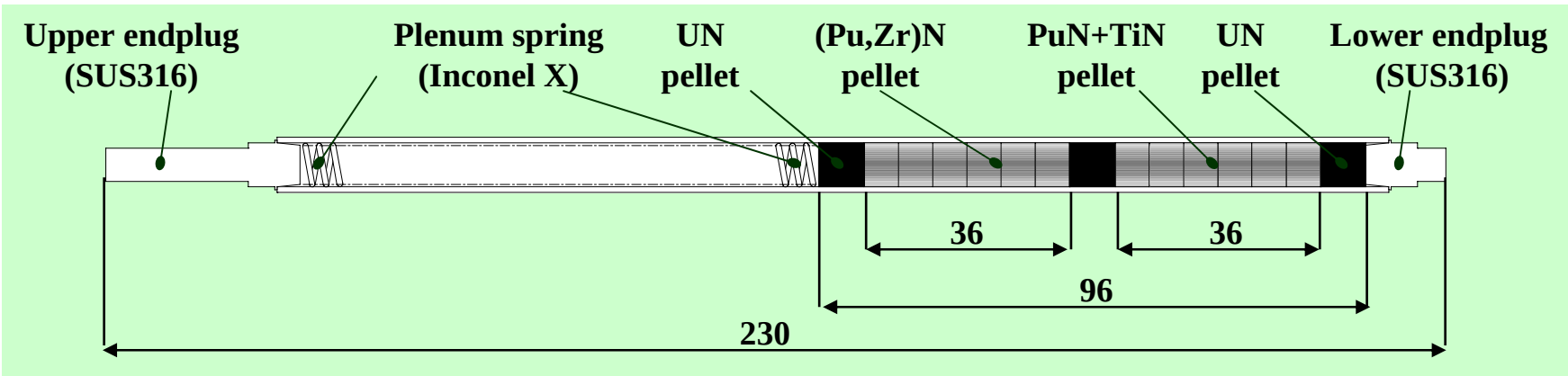
(Pu,Zr)N pellets



PuN+TiN pellets

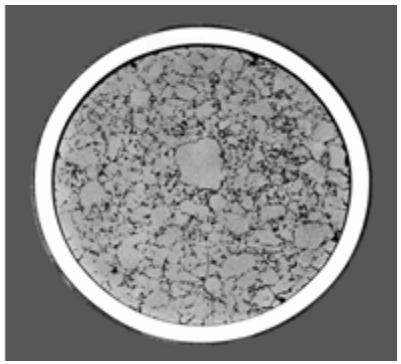
	(Pu,Zr)N	PuN+TiN
Irradiation period	May 2002 ~ Nov. 2004 (246 EFPD)	
Av. linear power	408 W/cm	355 W/cm
Burnup	14.7 at%-Pu	17.0 at%-Pu
Max. fuel Temp. (Estimation)	1273 K	1083 K
Cladding tube	SUS316, 9.40 mm∅, 0.51 mmt	

- PIEs have been completed in JAEA's hot cells.-

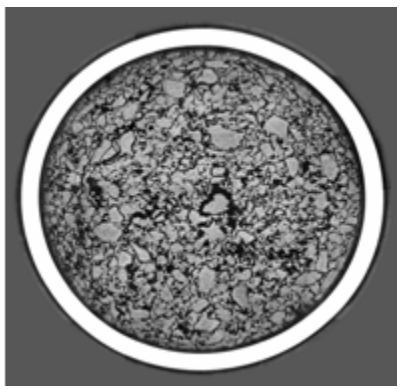


Results of PIEs for U-Free Nitride Fuel (1)

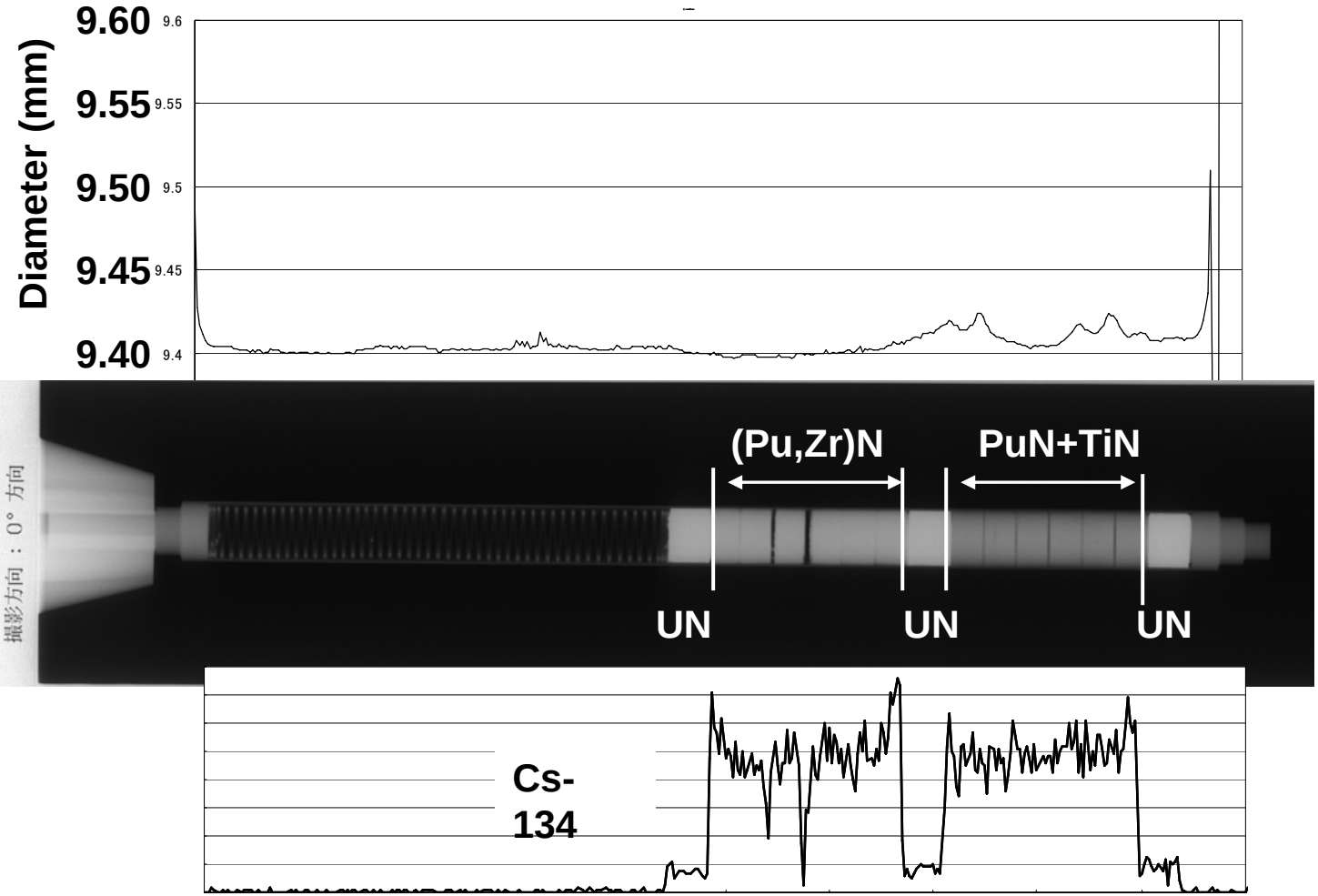
- Cross sections, profilometry, X-ray radiography and gamma scanning-



(Pu,Zr)N



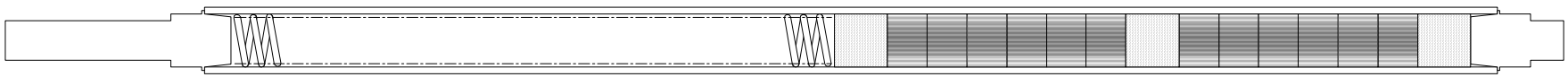
PuN+TiN

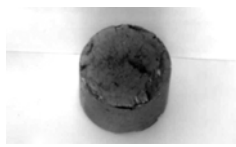


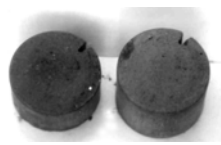
Results of PIEs for U-Free Nitride Fuel (2)

- Results of swelling and fission gas release of pellets -

	(Pu,Zr)N	PuN+TiN
Density before irradiation	7.22 g/cm^3 90.4 %TD	5.14 g/cm^3 86.6 %TD
Density after irradiation	6.91 g/cm^3 86.4 %TD	4.92 g/cm^3 83.7 %TD
Volume increase rate $\Delta V/V$	3.6 %	2.6 %
FP gas release rate ((Xe+Kr) released/(Xe+Kr) produced)	1.6%	







Conclusion

- ✓ **R&D on nitride fuel cycle for transmutation of MA in JAEA has been progressed by experiments on fuel fabrication, property measurements, pyrochemical process and irradiation test.**
- ✓ **Thermal and electrochemical properties of MA bearing nitrides have been investigated, mainly focusing on AmN and nitrides containing ZrN as a diluent material. Property measurements of UN-based burnup simulated nitrides are also in progress.**
- ✓ **PIEs of U-free nitride fuel, (Pu,Zr)N and PuN+TiN, irradiated in JMTR have been completed. For the moment, no detrimental effect on irradiation behavior has been observed by the addition of such diluent materials.**

mestic and international collaboration is essential for

Acknowledgement

Part of this study was carried out within the task "Technological development of a nuclear fuel cycle based on nitride fuel and pyrochemical reprocessing" entrusted from the Ministry of Education, Culture, Sports Science and Technology (MEXT) of Japan.