

Experimental Evaluation of Am and Np Bearing Mixed Oxide Fuel Properties

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Background

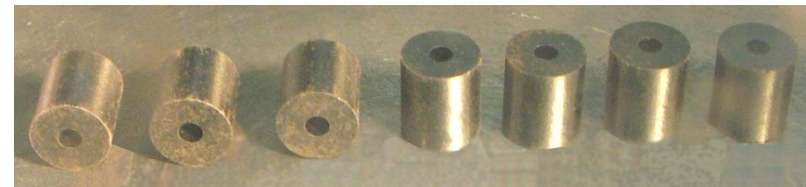
- Development of MOX containing MA -



- Japan Atomic Energy Agency has developed homogeneous mixed oxide containing minor actinides (MA-MOX) as a fuel of the advanced fast reactor.

Specification of the fuel pellet

Type	: Hollow type
Pu content	: 20 - 30%
MA content	: - 5%(Np+Am+Cm)
Density	: 93%TD
O/M	: 1.95
Burn-up	: 150GWd/t



Hollow pellets of MOX



Pellets of $(\text{Np}_{0.02}\text{Am}_{0.02}\text{Pu}_{0.3}\text{U}_{0.64})\text{O}_2$

- Physical property measurements of Am and Np bearing MOX
- The effect of MA addition on the physical properties



Outline

- **Sample preparation**
- **Phase state and Phase separation**
- **Lattice parameters**
- **Oxygen potentials**
- **Melting temperatures**
- **Thermal conductivities**

Sample preparation

UO_2 , $(\text{U,Pu})\text{O}_2$, $(\text{U,PuNp})\text{O}_2$, $(\text{Pu,Am})\text{O}_2$

Starting materials

Ball mill

Press

Sintering

Annealing for $\text{MO}_{2.00}$

Annealing to adjust O/M ratio

Measurement



at 1923-1973K for 3-4 h in 5% H_2 /Ar flowing gas added moisture

at 1123K for 4 h in the atmosphere of $\Delta G = -400$ -420kJ/mol

at 1123-1973K for 3-25 h in an appropriate atmosphere



Sample preparation

List of samples

Sample Name	Composition				O/M
	U(%)	Pu(%)	Np(%)	Am(%)	
MOX-1	70.3	29		0.7	2.00-1.909
MOX-2	69.5	30		0.5	2.00-1.924
6%Np-MOX	64.3	29	6	0.7	2.00-1.914
12%Np-MOX	58.3	29	12	0.7	2.00-1.909
2.4%Am-MOX	67.6	30		2.4	2.00-1.951
2%Np/Am-MOX	66	30	2	2	2.00-1.923
1.8%Np/Am-MOX	66.4	30	1.8	1.8	2.00-1.919
12%Pu-MOX	87.9	11.8		0.3	2.00-1.971
20%Pu-MOX-1	79.7	19.9		0.4	2.00-1.942
20%Pu-MOX-2	78.7	19.8		1.5	2.00-1.947
MOX-3	69.6	29.8		0.6	2.00
40%Pu-MOX-1	59.6	39.7		0.7	2.00-1.916
40%Pu-MOX-2	58.5	39.6		1.9	2.00-1.961
40%Pu-MOX-3	58.4	38.3		3.3	2.00
43%Pu-MOX	53.7	42.8		3.5	2.00
46% Pu-MOX	51.4	46.3		2.4	2.00-1.718
60% Pu-MOX	37.7	60		2.3	2.00
2%Am-PuO ₂		97.9		2.1	2.00
6%Am-PuO ₂		93.6		6.4	2.00
7%Am-PuO ₂		94		7.2	2.00



Pu : Z = 0 – 97.9%

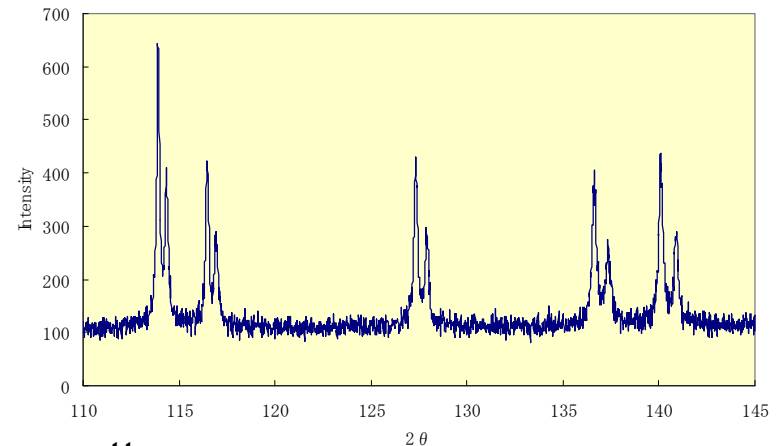
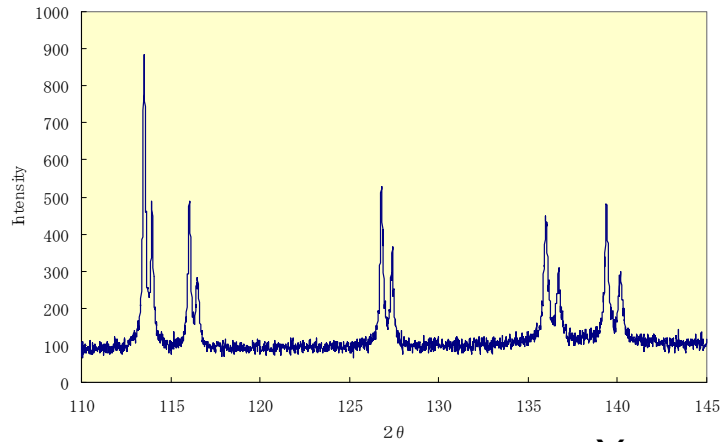
Am : y' = 0.5 – 7.2%

Np : y'' = 0 – 12%

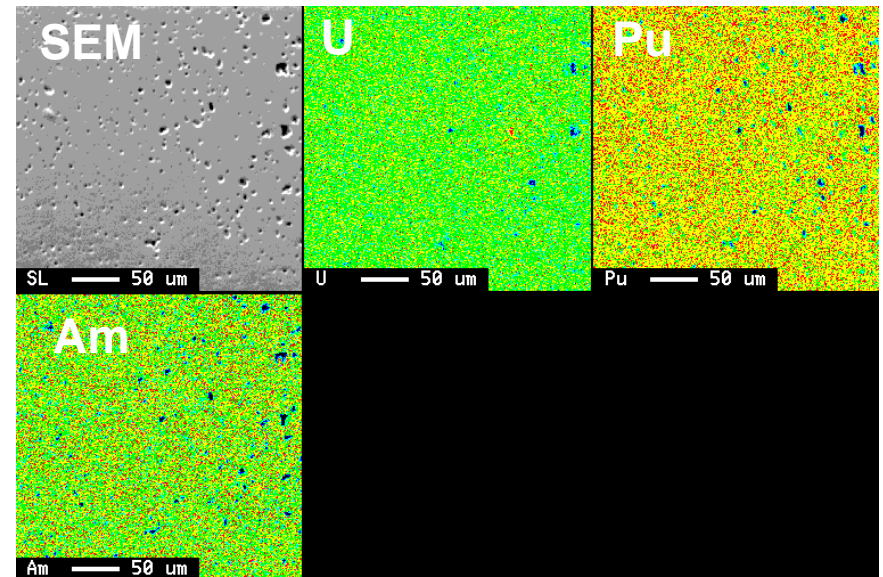
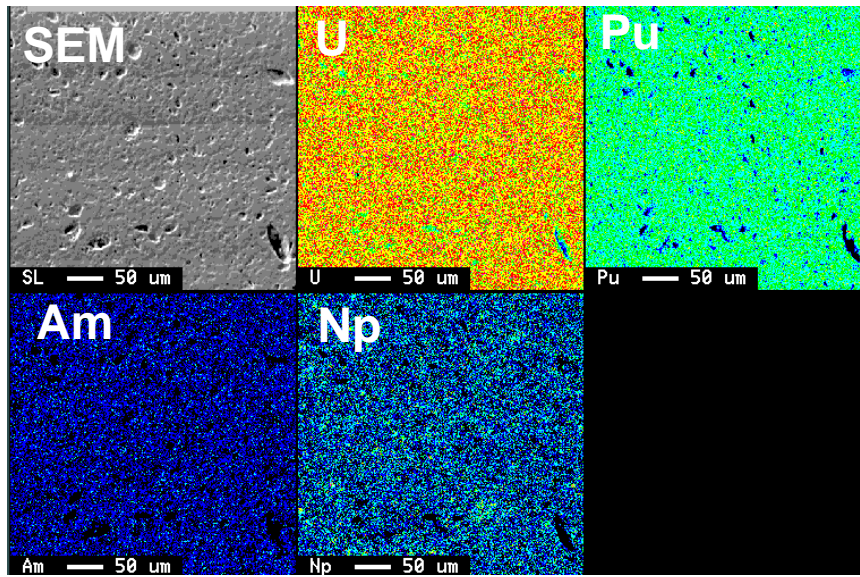
x : x = 0 – 0.282

(O/M=2.00 – 1.718)

Homogeneity of samples



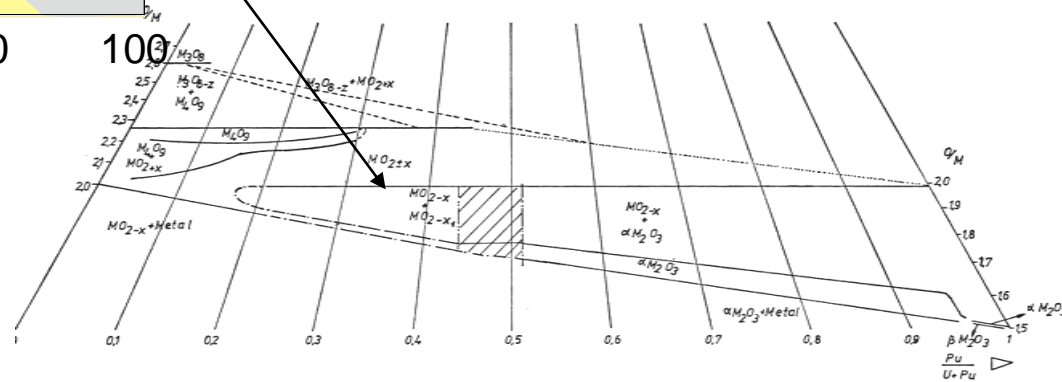
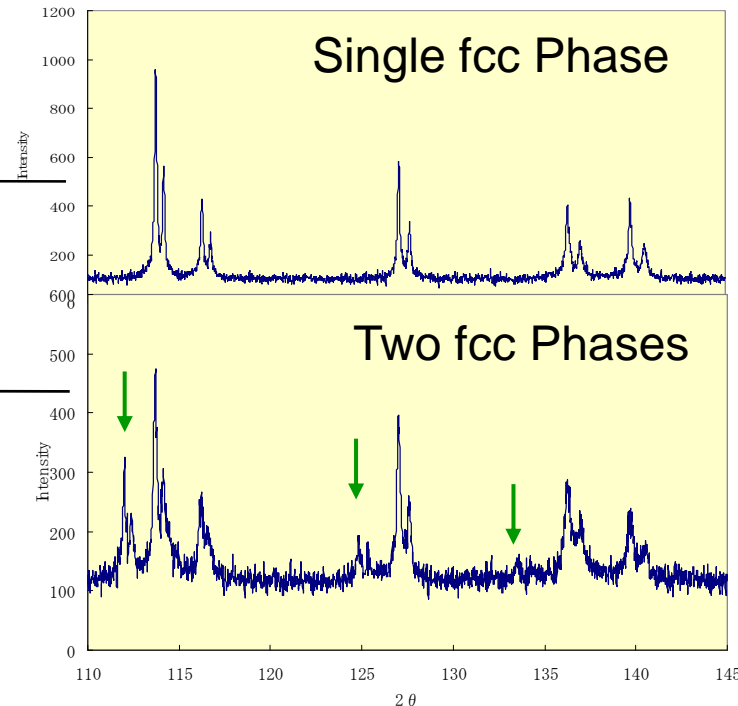
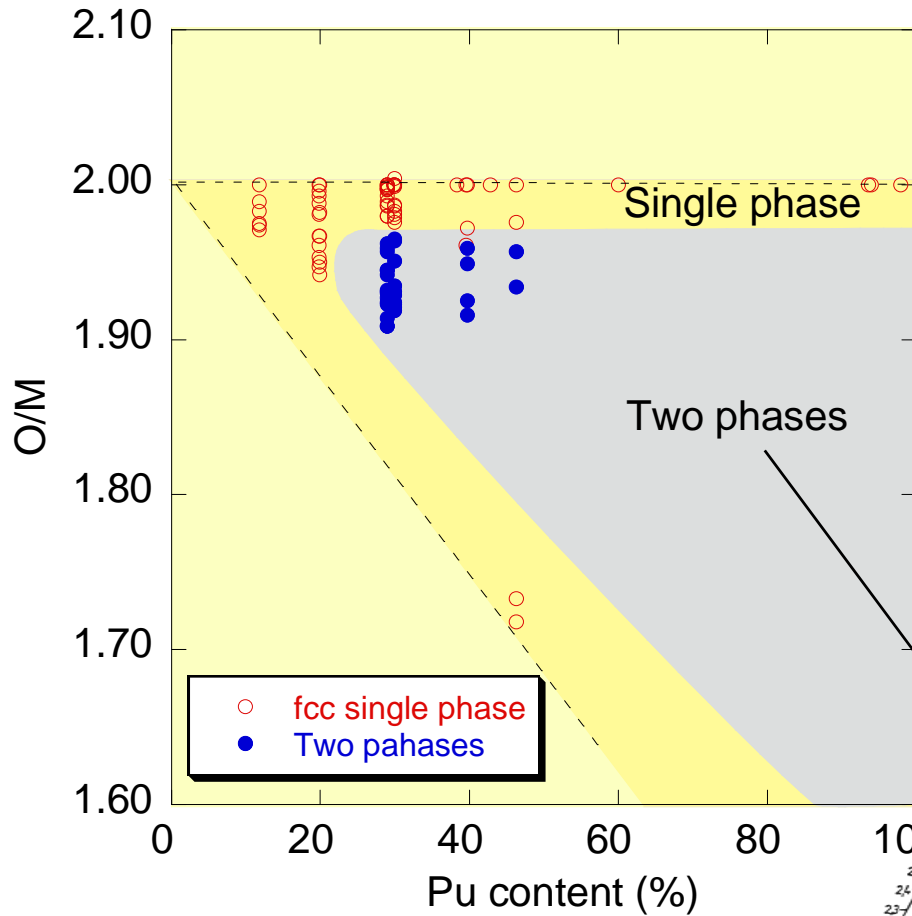
X-ray diffraction pattern



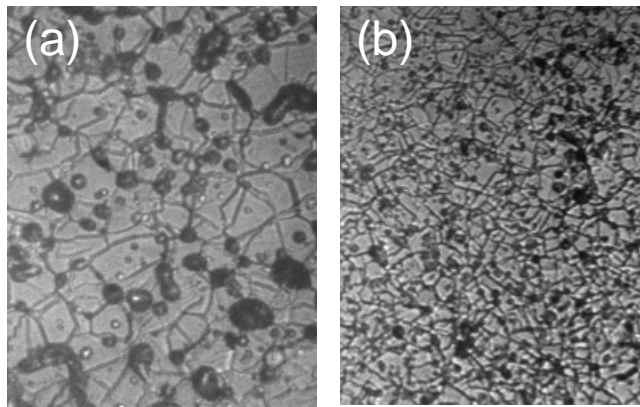
EPMA analysis on cross section



Phase State at room temperature

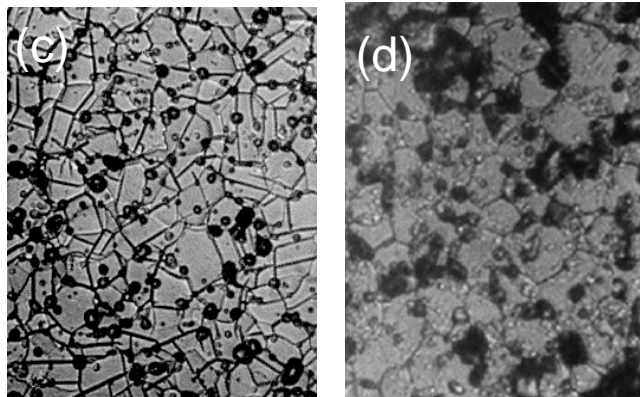


Phase Separation Behavior



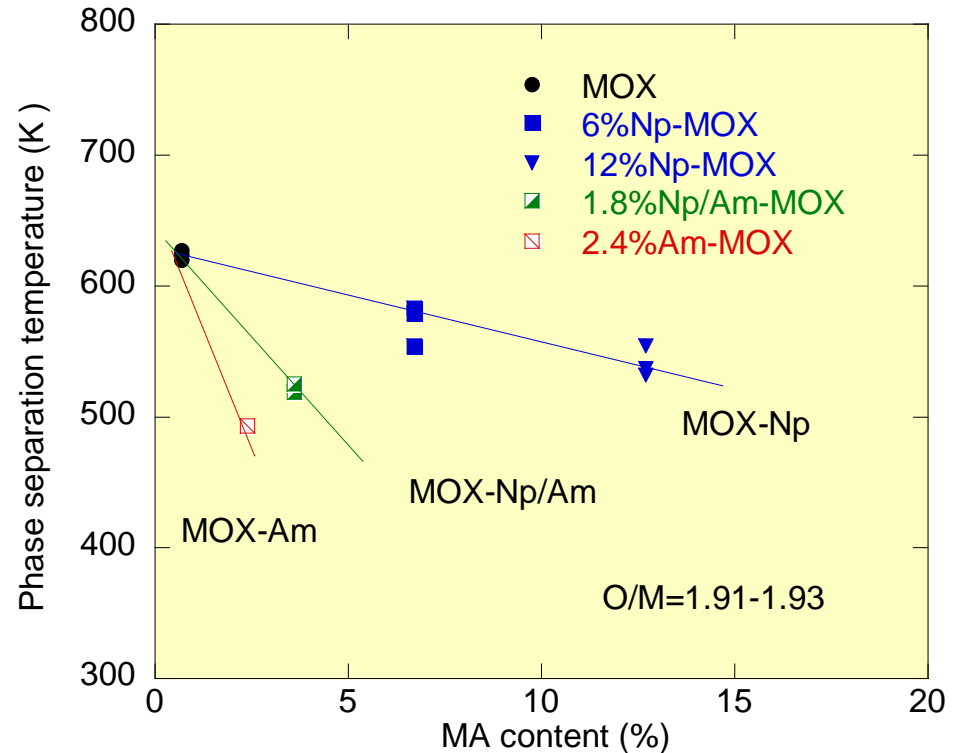
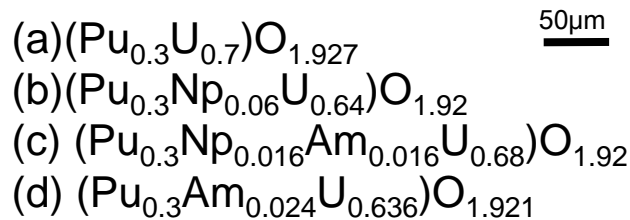
MOX

6%Np-MOX



Am/Np-MOX

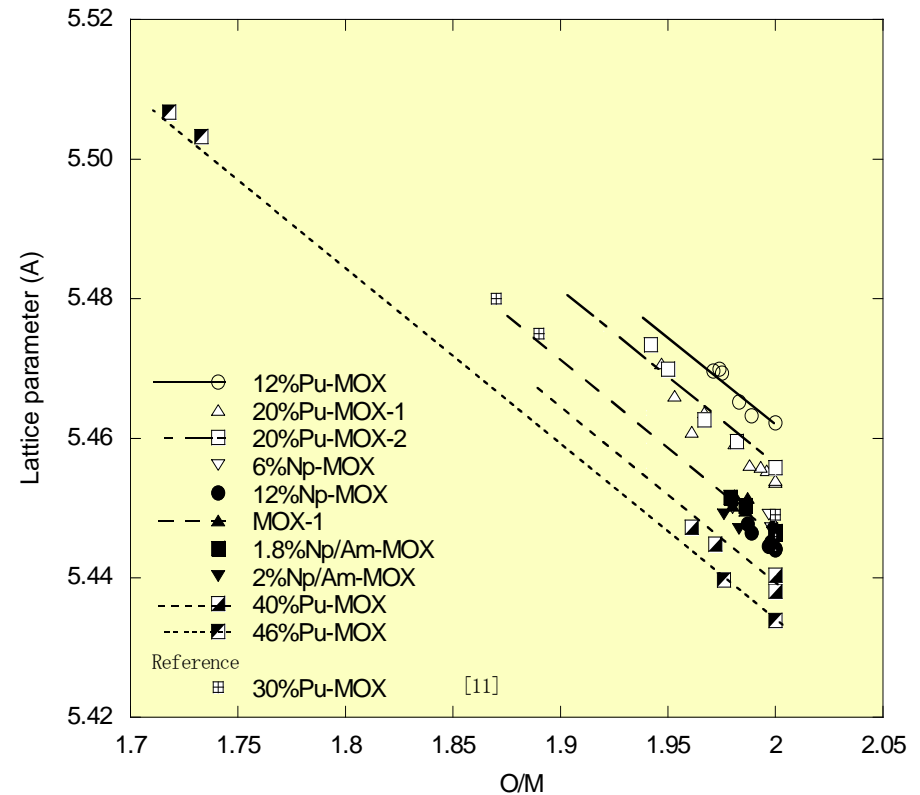
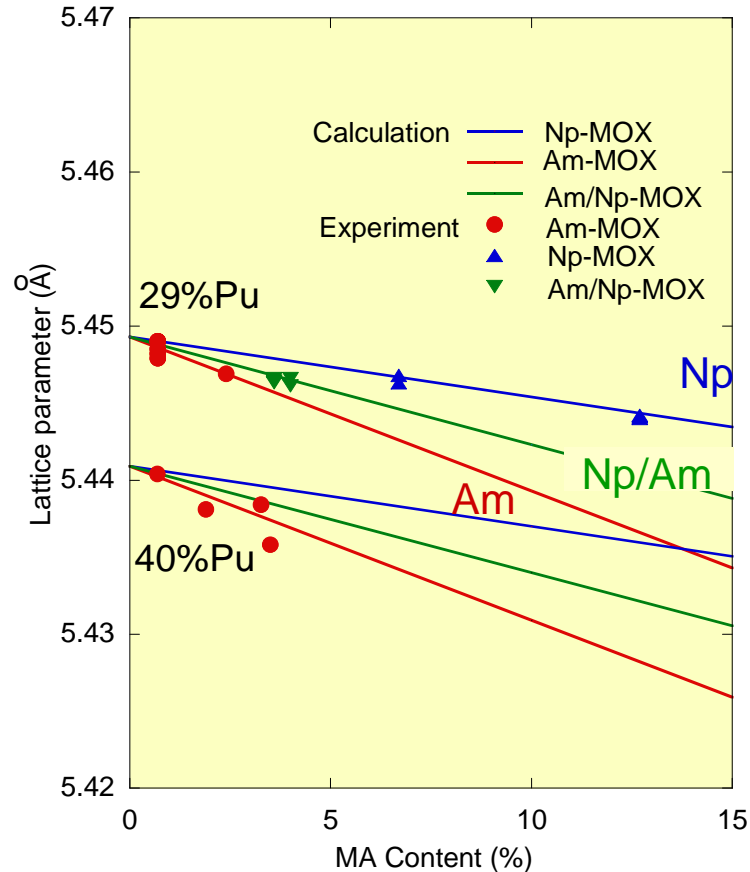
Am-MOX



The effect of MA content on the phase separation temperature.

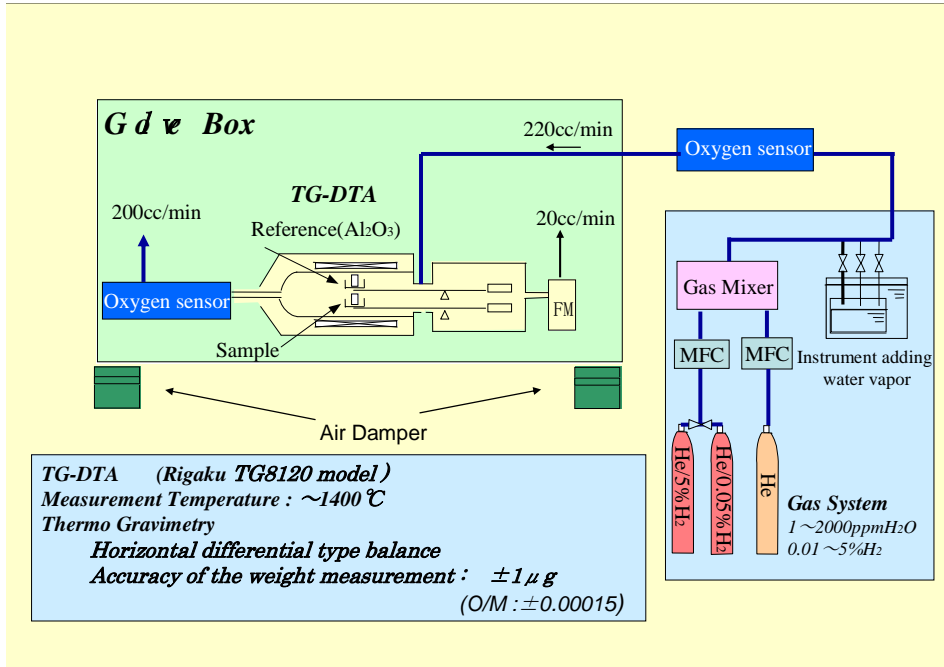
➤ No effect of phase separation on the fuel properties.

Lattice parameters

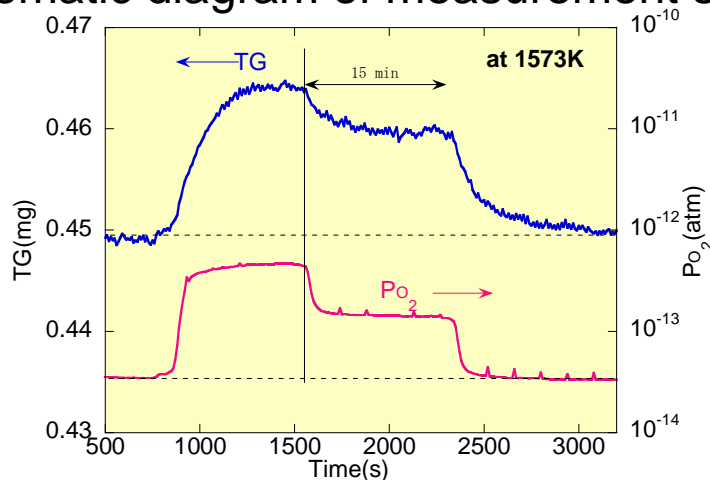


- The lattice parameters decrease with MA content, and increase with decreasing the O/M.
- The derived model represent the experimental data very well.

Oxygen potentials



Schematic diagram of measurement system

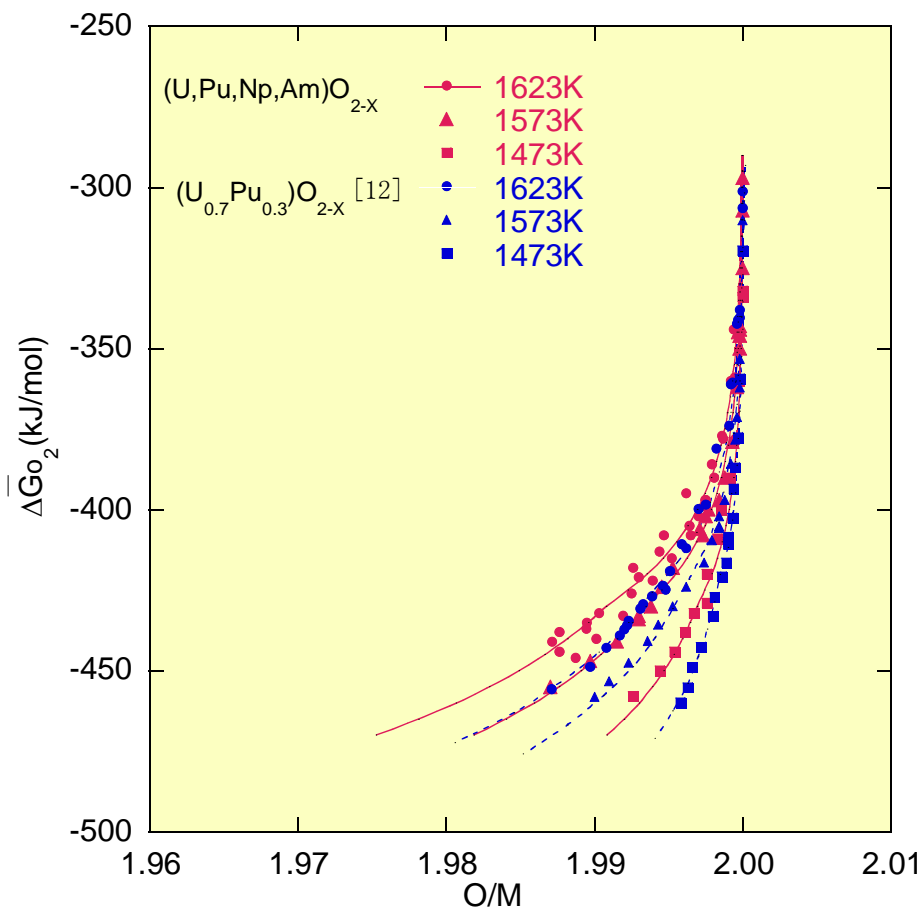


Measurement results

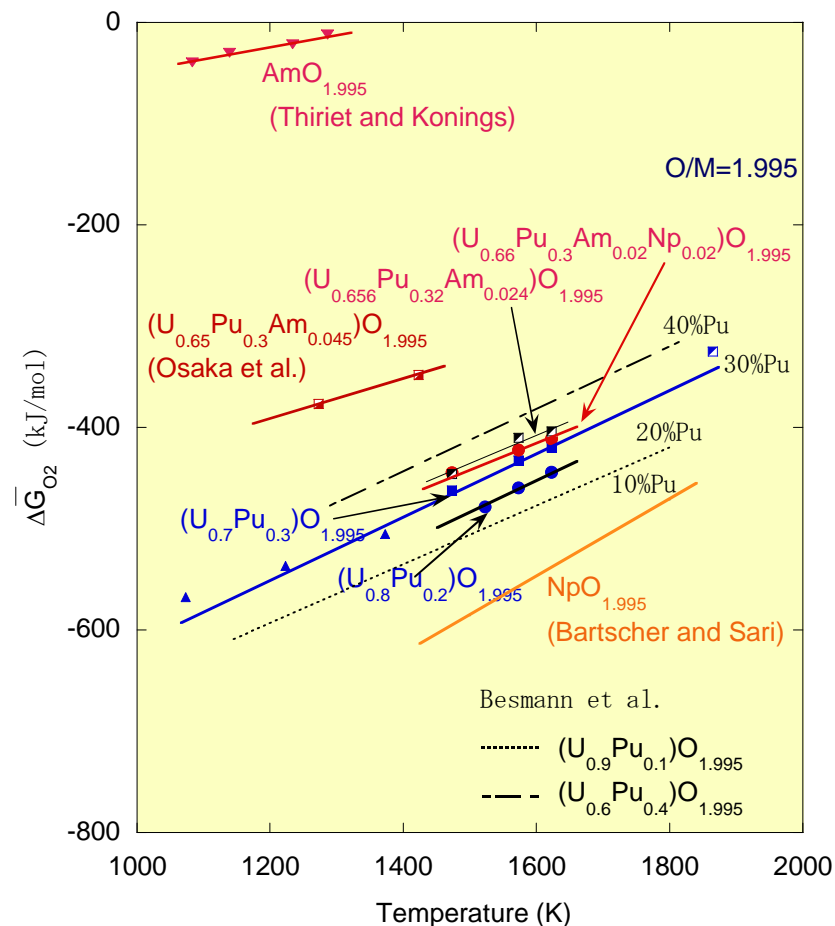
- ◆ Horizon type TG-DTA was employed in the measurement.
- ◆ The oxygen partial pressure was controlled by controlling the ratio of $P_{\text{H}_2}/P_{\text{H}_2\text{O}}$ and measured with stabilized ZrO_2 oxygen sensors.
- ◆ The change of TG attained to equilibrium condition for about 15min.
- ◆ The O/M ratio was calculated from the weight change.
- ◆ The O/M ratio and P_{O_2} were measured with good repeatability and precision.

Oxygen potentials

Measurement Results



The change of the ΔG_{O_2}

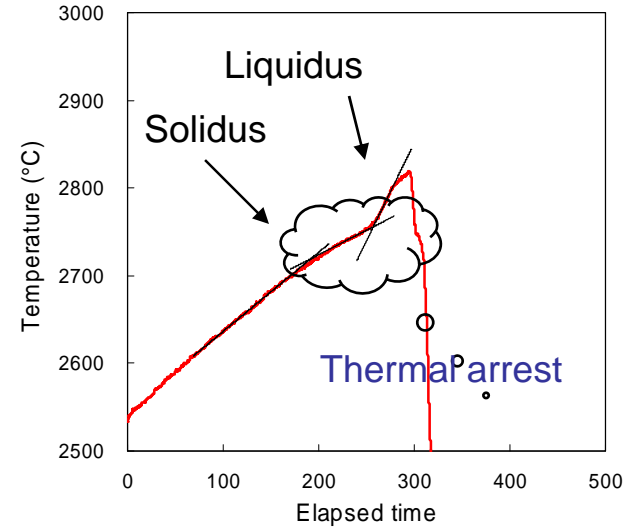
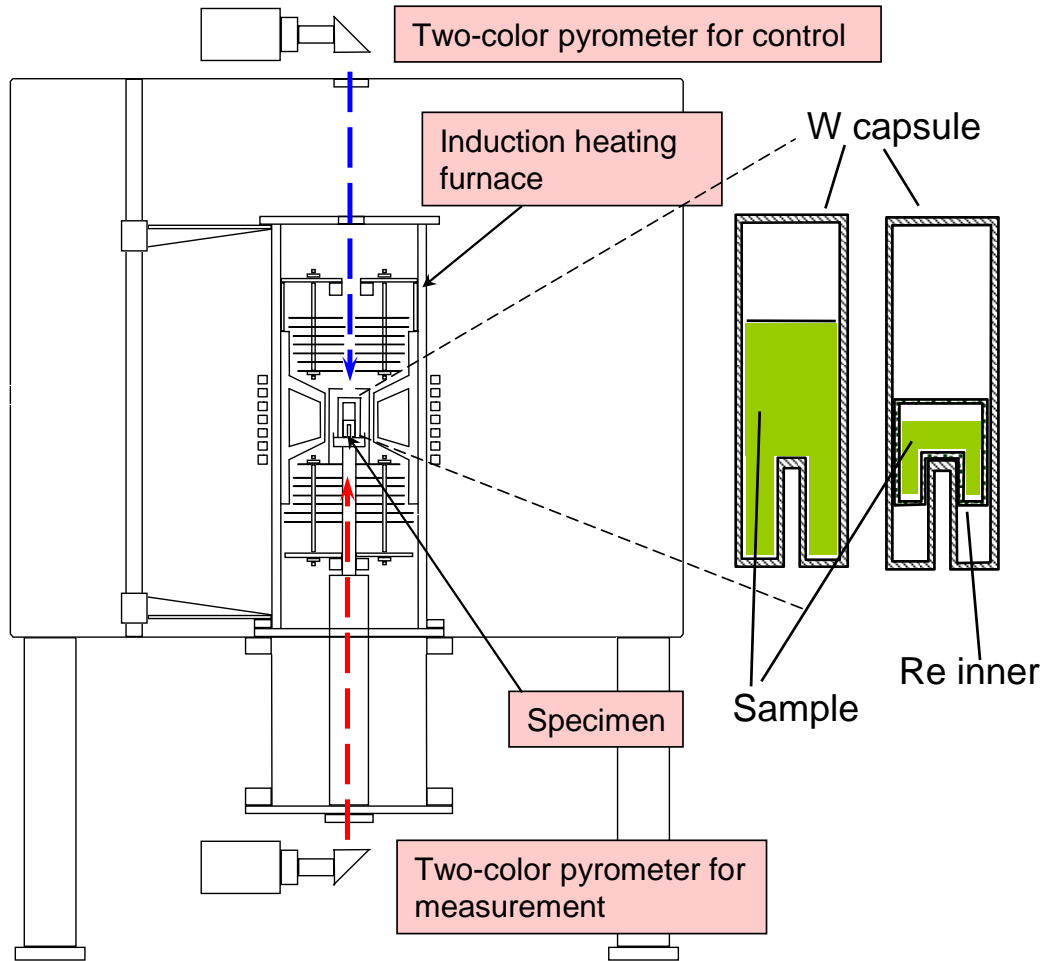


Comparison of the ΔG_{O_2} of $MO_{1.995}$

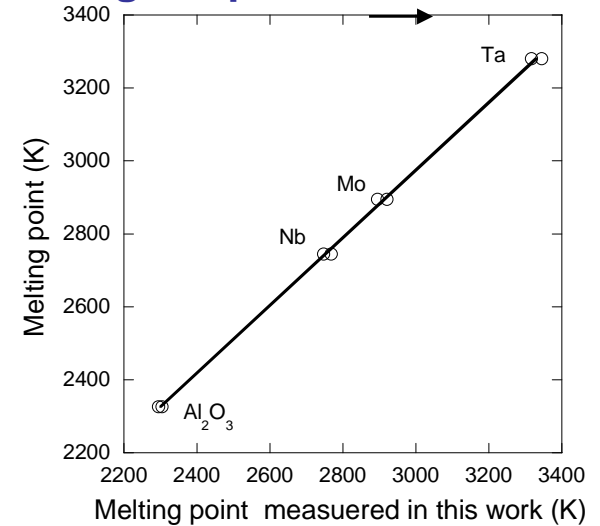
- The ΔG_{O_2} of $(U,Pu,Am,Np)O_{2-x}$ are slightly higher than those of MOX without MA.
- The slightly higher ΔG_{O_2} is caused by Am content.

Melting temperatures

Experiments



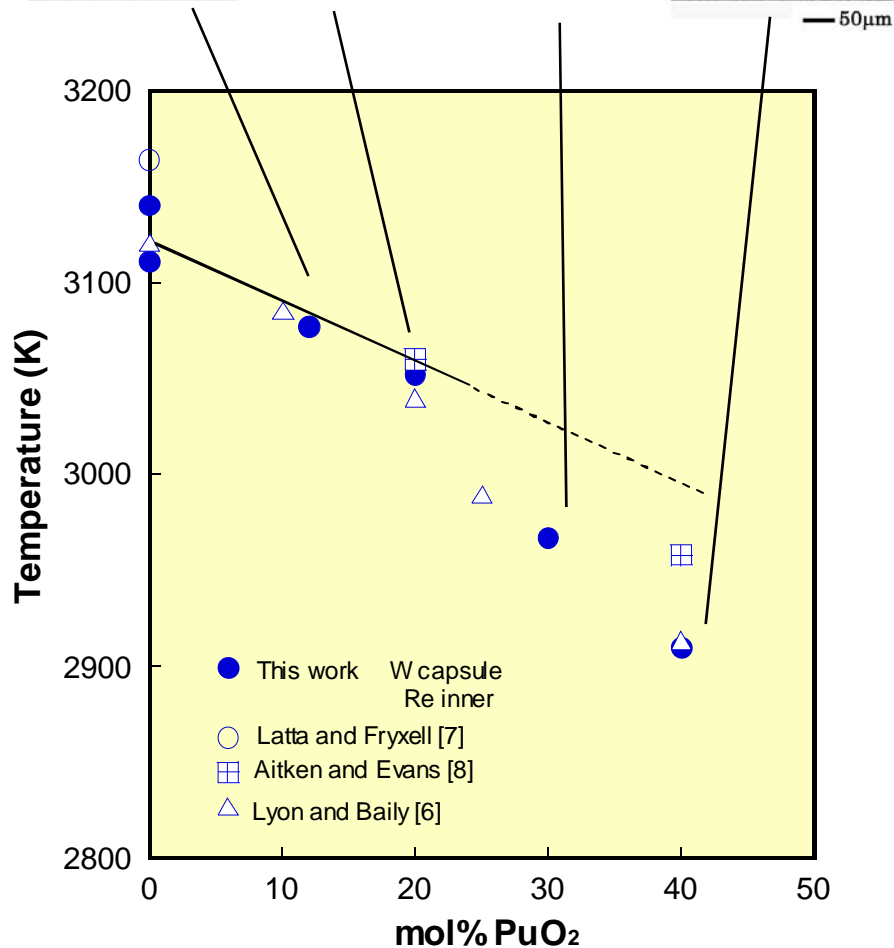
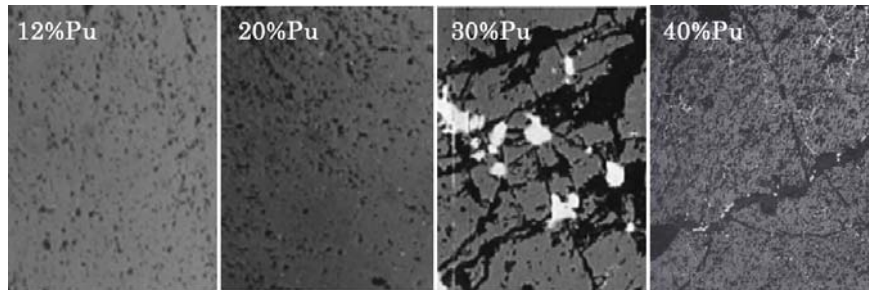
Heating temperature curve of MOX



Temperature calibration by standard samples

Melting Temperatures

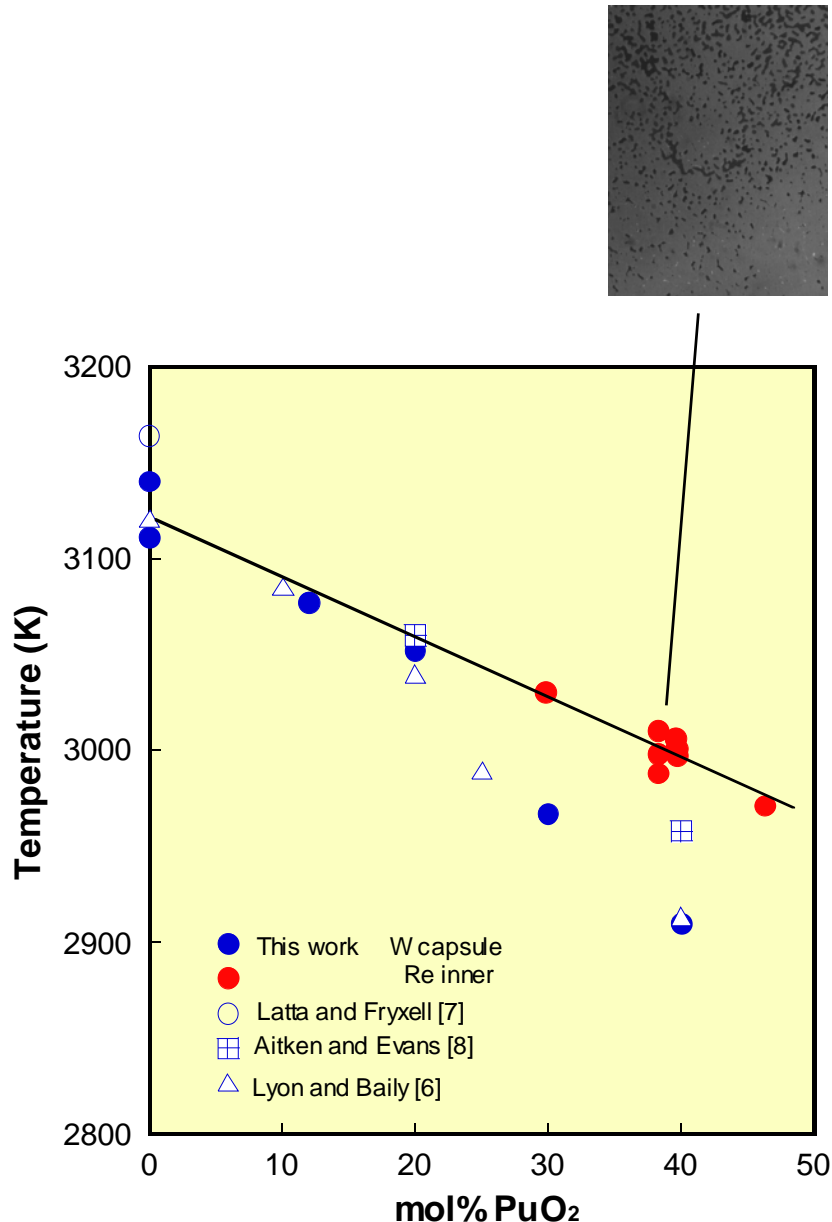
Solidus Temperatures depending on Pu content



- The data measured by conventional method were in good agreement with other data, and the measured value fell off at about 30%Pu –MOX.
- The reaction between MOX and W was observed in the measured samples with 30% and 40%Pu content.

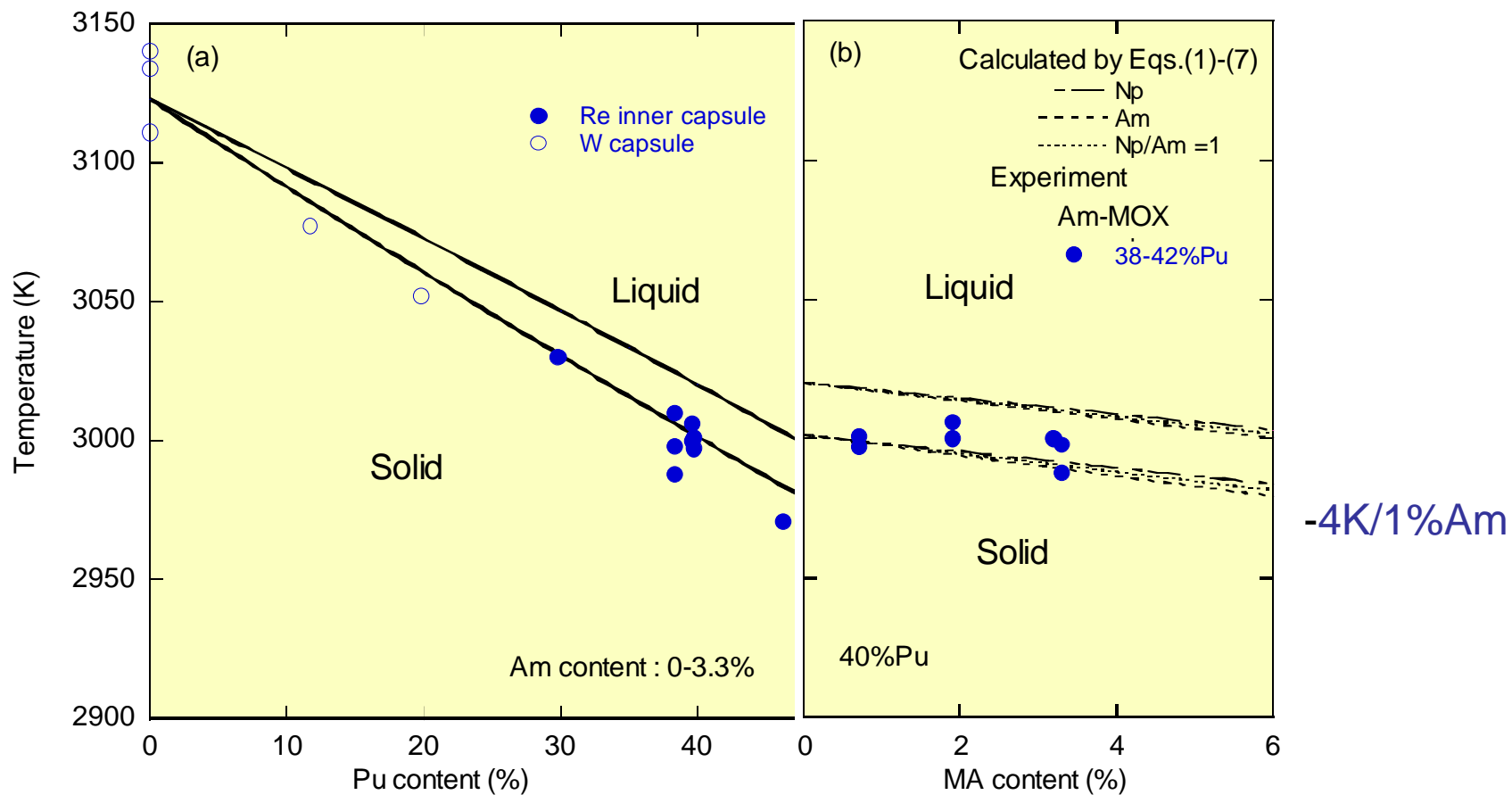
Melting Temperatures

Solidus Temperatures depending on Pu content



- The data measured by conventional method were in good agreement with other data, and the measured value fell off at about 30%Pu –MOX.
- The reaction between MOX and W was observed in the measured samples with 30% and 40%Pu content.
- The solidus measured with Re-capsule are consistent with that of MOX with Pu content of less than 20%.
- It can be concluded that the solidus temperature measured by using Re inner is true melting temperature of MOX

Melting Temperatures

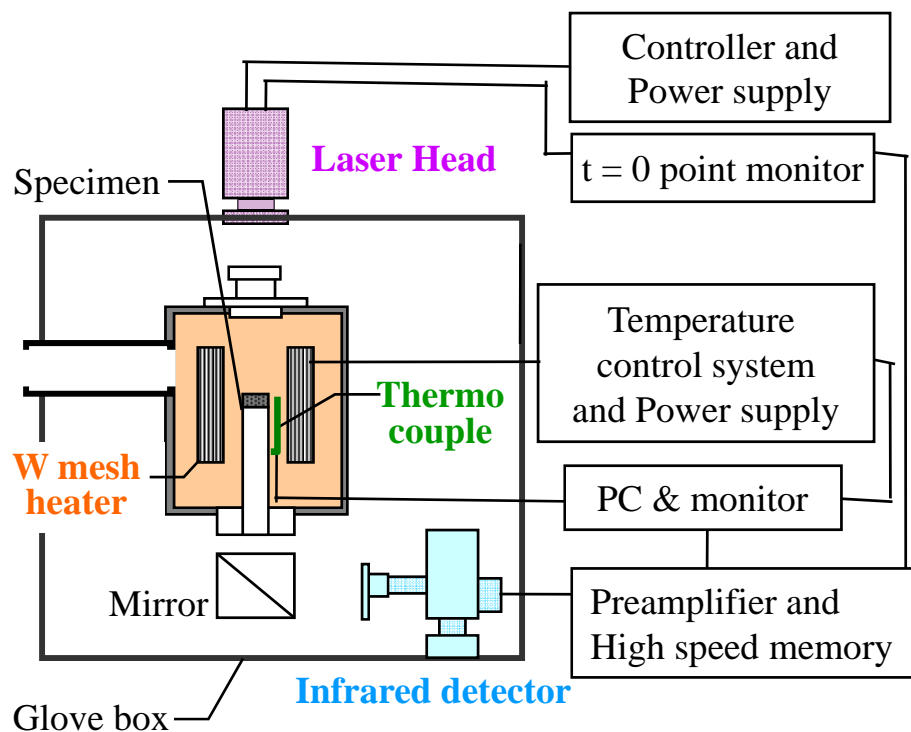
UO₂-PuO₂ system and effect of MA addition

Thermal conductivity

Measurement apparatus



✓ Thermal diffusivity (α) measurement : Laser flash method



• High temperature furnace

Resistance heating furnace

• Temperature-measuring device

W-Re thermo couple

• Laser source

Nd glass Laser

Maximum output 17 J / Pulse

• Infrared detector

InSb sensing device (~1673 K)

(Hamamatsu Photonics K.K.)

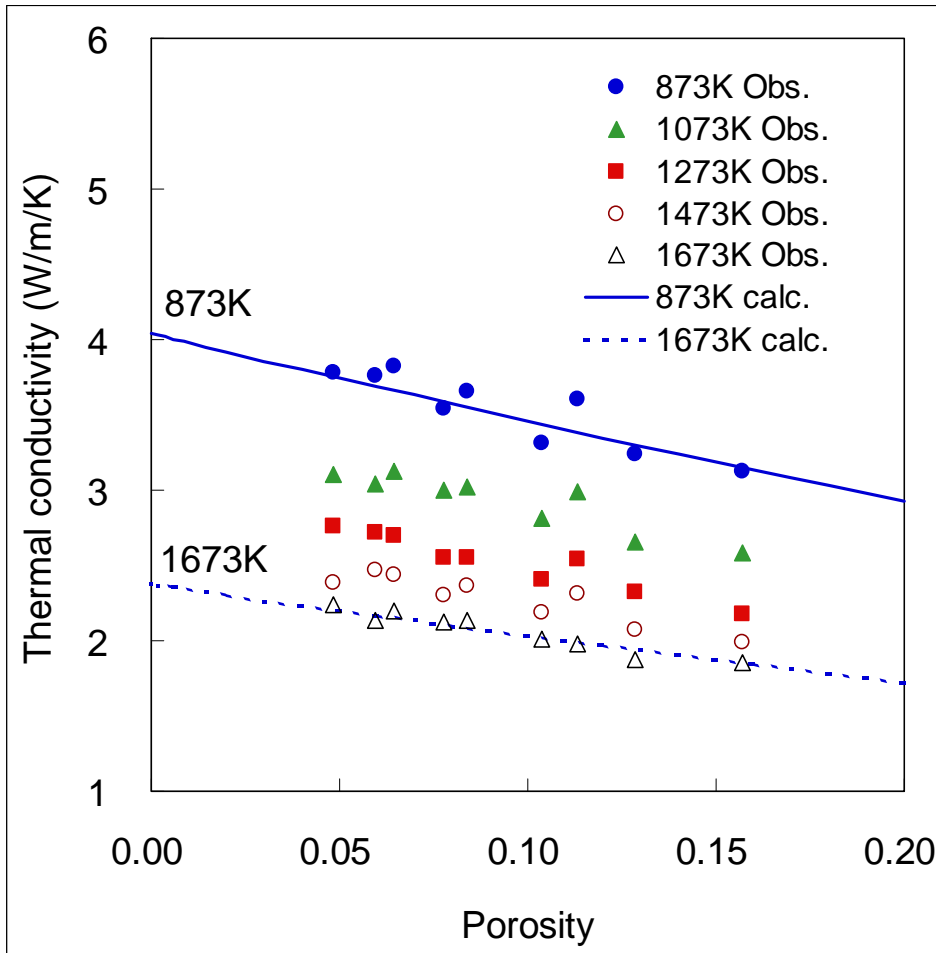
Si sensing device (1673 K ~)

(Tokyo Seiko co.)

Fig.1 Thermal diffusivity measurement apparatus.

Thermal conductivity

Dependency of density



Porosity dependency of thermal conductivities on 2%Am-MOX

- ✓ Porosity (density) correction equation of λ is described as follows.
- ✓ In this section, correction coefficient (β) is determined.

$$\lambda = F \times \lambda_0$$

F : Maxwell-Eucken equation

$$F = \frac{(1-p)}{(1+\beta \times p)} \quad \beta = 0.5$$

λ_0 : The value of MOX specimen with theoretical density (100%TD)

λ : The experimental value of MOX specimen with porosity p

F : Porosity correction equation (Maxwell-Eucken equation)

β : Correction coefficient

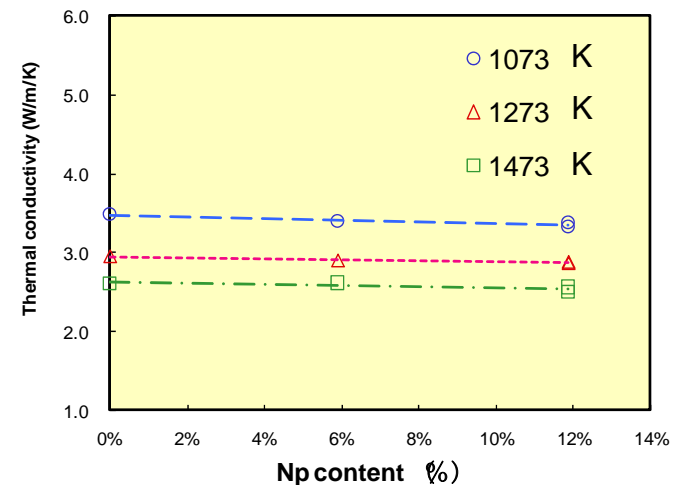
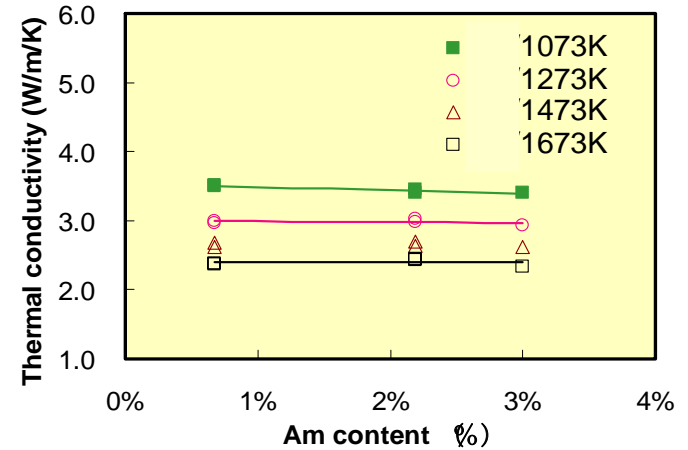
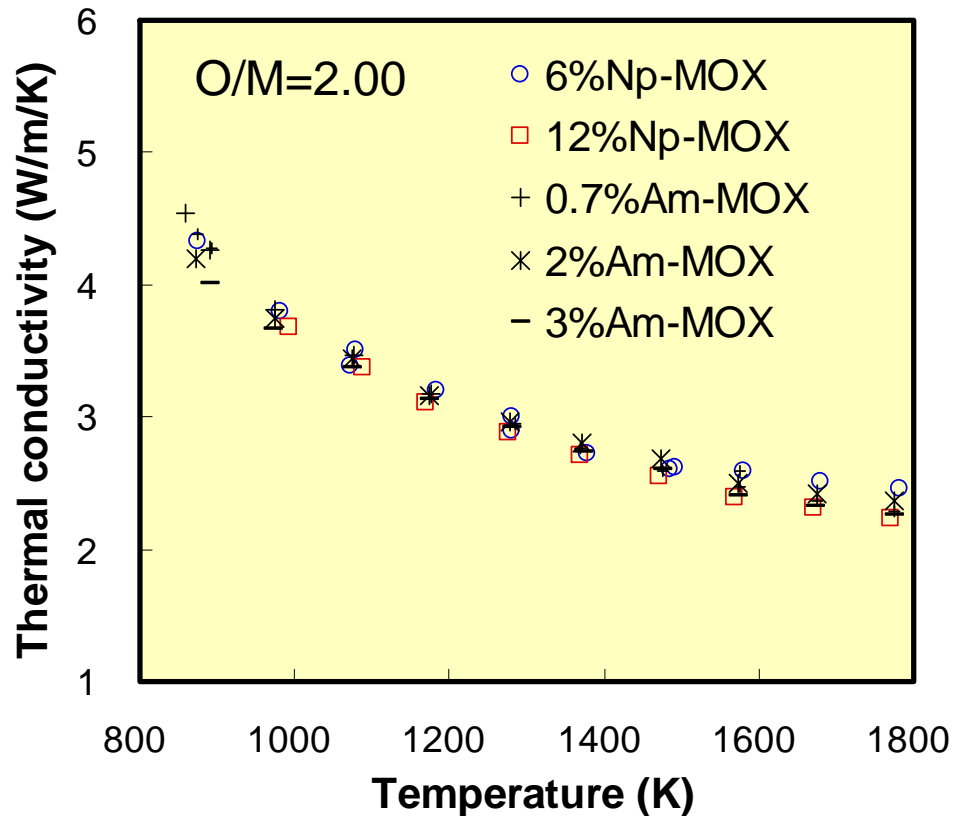
p : Porosity ($p = 1 - (\rho / \rho_{th})$)

ρ : Density of specimen

ρ_{th} : Theoretical density of specimen

Thermal conductivity

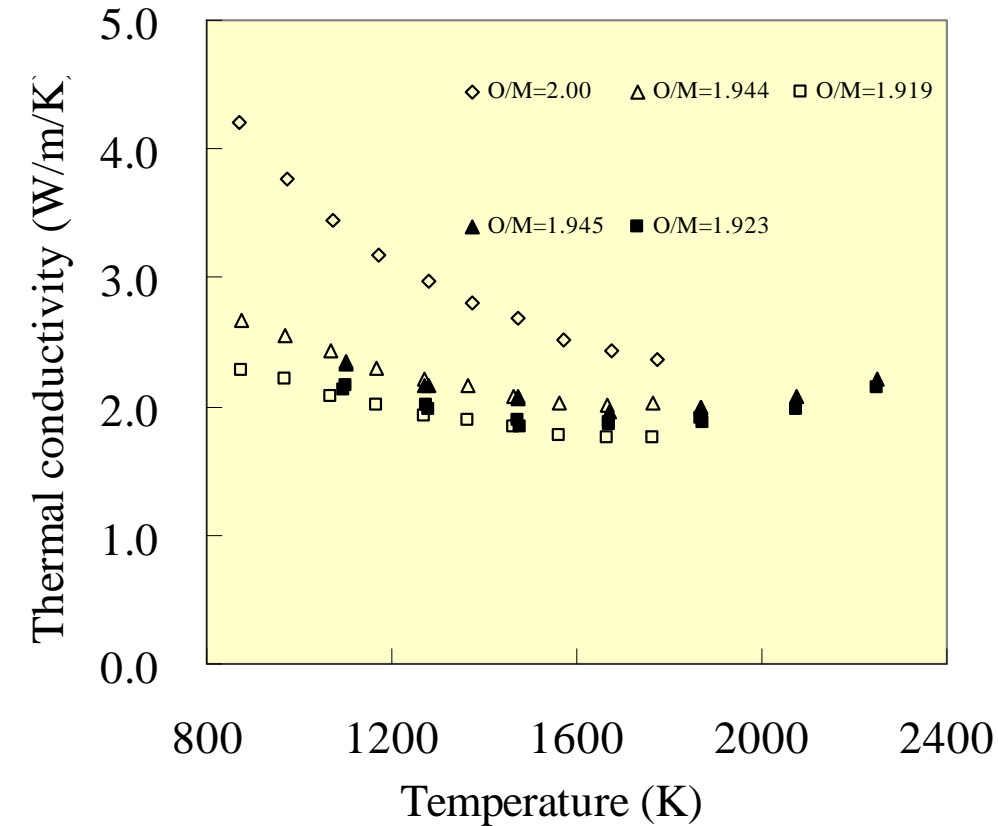
Effect of MA content



- The addition of MA caused to decrease slightly the thermal conductivities in the temperature range of less than 1000K.

Thermal conductivity

Effect of O/M ratio



- The thermal conductivities decrease significantly with decreasing O/M ratio.
- The equation was derived as functions of temperatures, MA contents and O/M ratios.

$$\lambda_0 = \frac{1}{(2.713x + 3.583 \times 10^{-1} \times z_1 + 6.317 \times 10^{-2} \times z_2 + 1.595 \times 10^{-2}) + (-2.625x + 2.493) \times 10^{-4} T} + \frac{1.541 \times 10^{11}}{T^{5/2}} \exp\left(\frac{1.522 \times 10^4}{T}\right)$$

z_1 : Am content

z_2 : Np content

x : Deviation x in MO_{2-x}

- It is expected that the equation will be revised by expanding data in the high temperature region.



Summary

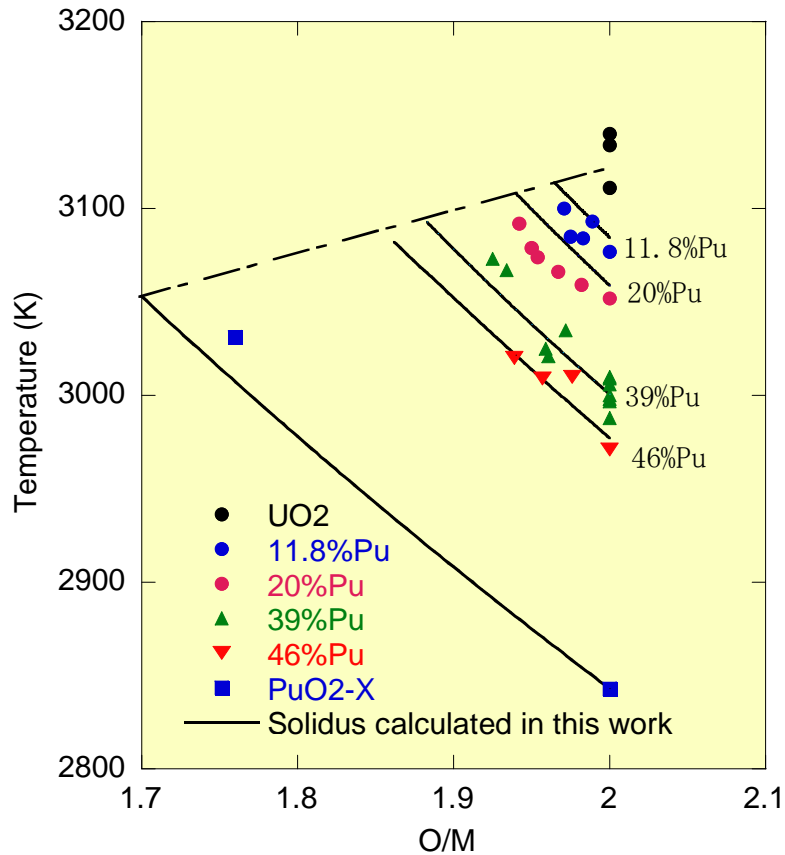
- The physical properties, lattice parameters, phase diagram, oxygen potentials, melting temperatures and thermal conductivities, of Am and Np-bearing MOX were measured.
- The effects of MA addition on the physical properties are small. No MA addition affects fuel properties significantly.
- We obtained the basic data for conducting the irradiation tests of MA-bearing MOX fuel.



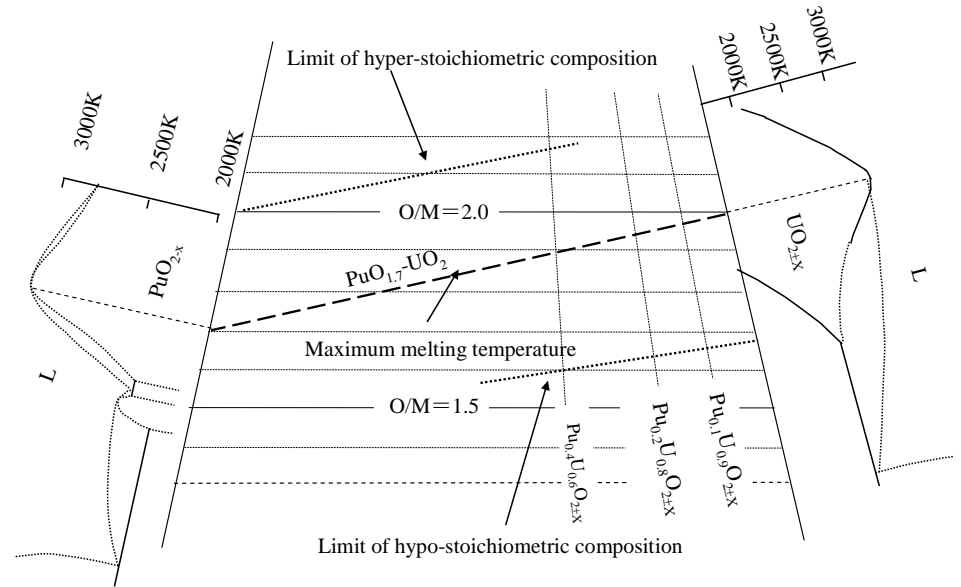


Melting Temperatures

Effect of O/M ratio



The effect of O/M ratio



The expected phase diagram in U-Pu-O ternary system



Database and Models

Thermal conductivity

Lattice parameter

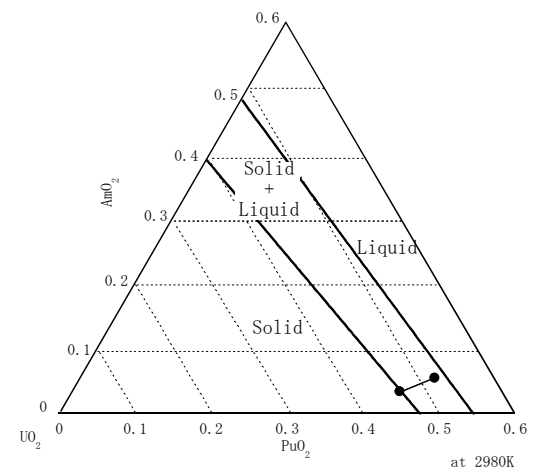
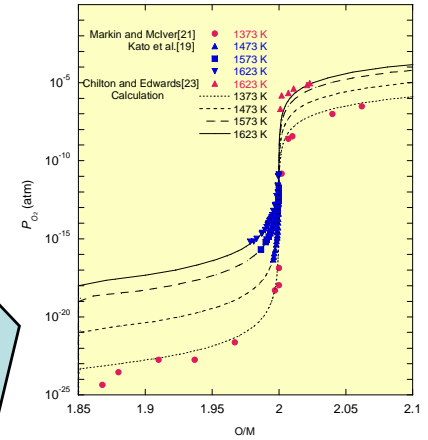
Sample Name	Composition(%)					Result of X-ray diffraction	
	U	Pu	Np	Am	O/M	Lattice Parameter(Å)	Phase
MOX-1	70.3	29	0	0.7	2	5.449	fcc
	70.2	29	0	0.7	2	5.449	fcc

Oxygen potential

	U	Pu	Np	Am	O/M	T	ΔG	
						K	kJ/mol	
2.0%Pu		80	20	0	0	2.0000	1623	-349.5
2.0%Pu		80	20	0	0	1.9916	1623	-466.0
2.0%Pu		80	20	0	0	2.0001	1623	-362.4
2.0%Pu		80	20	0	0	1.9911	1623	-466.0

Melting temperature

Sample Name	Composition				Type of Cell	Melting Temp.		
	U	Pu	Am	O/M		Solidus	Liquidus	
	(%)	(%)	(%)			(K)	(K)	
UC ₂₀₀	100	0	0	2	A	3140	3145	
	100	0	0	2	A	3111	3130	
	100	0	0	2	A	3134	3173	
12%Pu-MOX	87.9	11.8	0.3	2	A	3077	3117	
	87.9	11.8	0.3	1.989	A	3093	3135	
	87.9	11.8	0.3	1.983	A	3084	3105	
	87.9	11.8	0.3	1.975	A	3085	3107	
	87.9	11.8	0.3	1.974	A	3054	3069	
20%Pu-MOX	87.9	11.8	0.3	1.971	A	3100	3124	
	79.7	19.9	0.4	2	A	3052	3090	
	79.7	19.9	0.4	1.982	A	3059	3089	
	79.7	19.9	0.4	1.967	A	3066	3079	
	79.7	19.9	0.4	1.954	A	3074	3109	
31%Pu-MOX	79.7	19.9	0.4	1.95	A	3079	3097	
	79.7	19.9	0.4	1.942	A	3092	3118	
	59.6	39.7	0.7	2	C	3030	3074	
	0.7%Am-41%Pu-MOX	59.6	39.7	0.7	2	C	2997	3029
	0.7%Am-42%Pu-MOX	59.6	39.7	0.7	2	C	3009	3020



Modeling