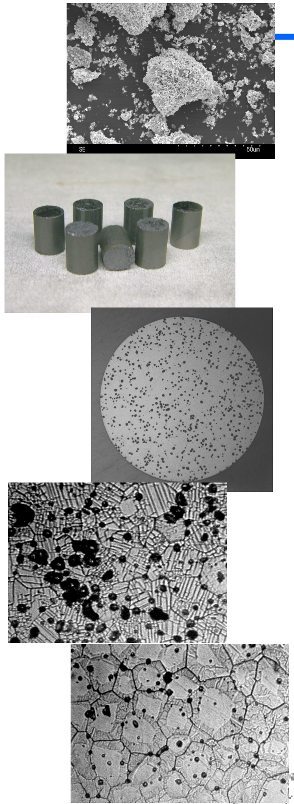

Development of Np and Am bearing MOX fuels for Japan sodium cooled fast reactors

Masato KATO

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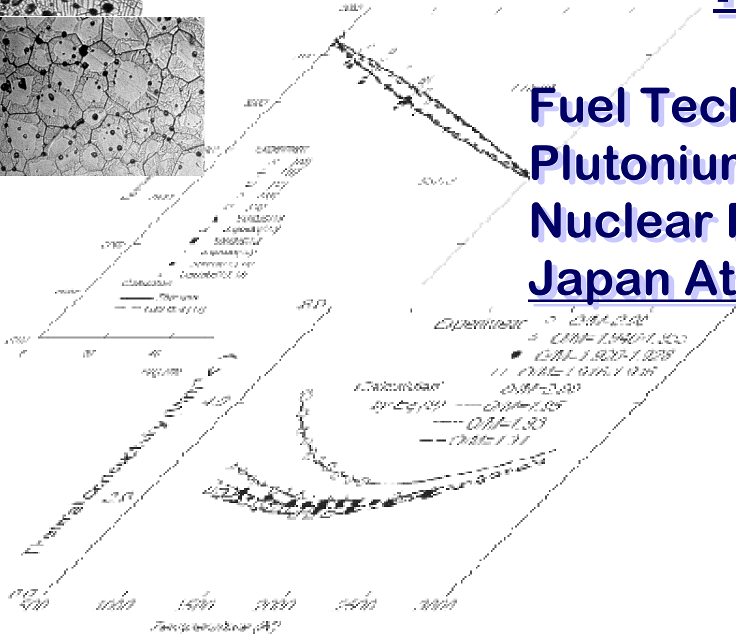
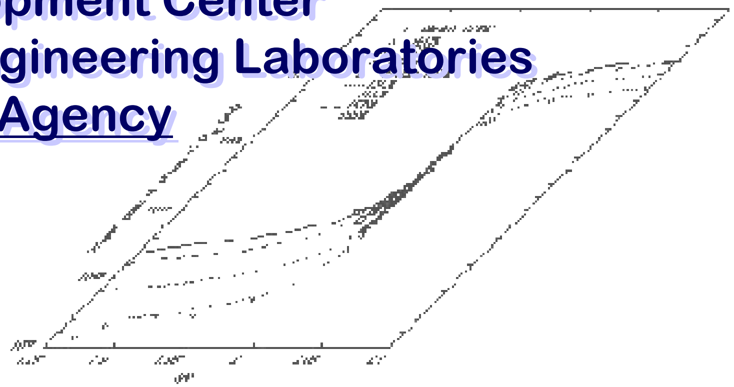
Japan Atomic Energy Agency





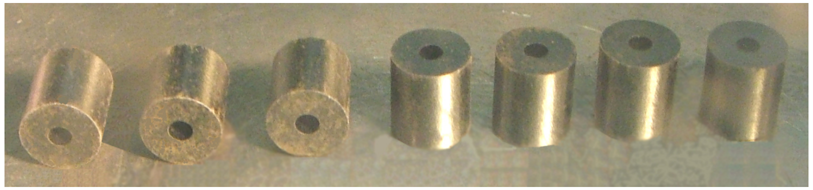
Background

— Development of MOX containing MA —

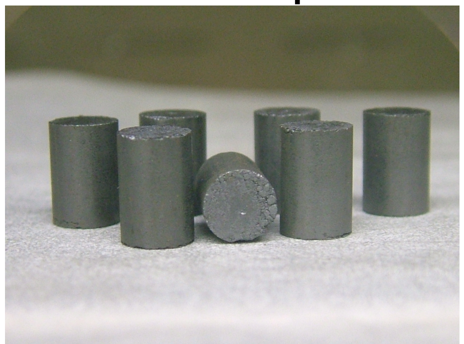
- **Japan Atomic Energy Agency has developed homogeneous mixed oxide containing minor actinides (MA-MOX) for Japan sodium-cooled fast reactors.**

Subjects to be carried out

1. Database and models for properties
2. Advanced fabrication process
3. Analysis code of irradiation behaviour of low O/M MA-MOX pellet



Hollow MOX pellets



Pellets of $(Np_{0.02}Am_{0.02}Pu_{0.3}U_{0.64})O_2$

Specification of the fuel pellet

Type : Hollow type
Pu content : 20 - 30%
MA content : ~ 5%(Np+Am+Cm)
Density : 95%TD
O/M : <1.97

Background
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1. Database and models for properties
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Experimental evaluation

1. Property Measurements
   - Thermodynamic data
   - Thermal properties
   - Chemical properties
2. Pellet Fabrication Test
   - Homogenization
   - O/M adjustment Technology
   - Sintering behavior
3. Irradiation test
   - Actinide redistribution
   - Microstructure change
   - FCCI

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Experimental evaluation

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   - Actinide redistribution
   - Microstructure change
   - FCCI

Purpose of this work

To analysis PIE results by using the measured data to develop an analysis code.

Contents

1. ***Irradiation Test and PIE Results***
   - Irradiation condition
   - Microstructure changes
   - Actinides redistribution
2. ***Properties of the fuel pellets***
   - Evaluation of properties

     Lattice parameter, Meting temperature

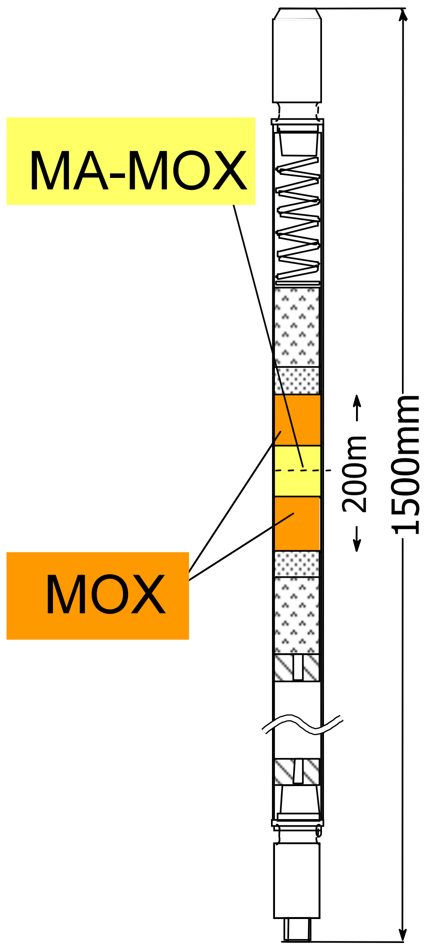
     Thermal conductivity, Oxygen potential
3. ***Analysis of Irradiation behaviour***
   - O/M redistribution
   - Relation between Pu redistribution and vapour pressure
   - Evaluation of microstructure change

Irradiation test

Condition 1

First Test

Pin No.1-1

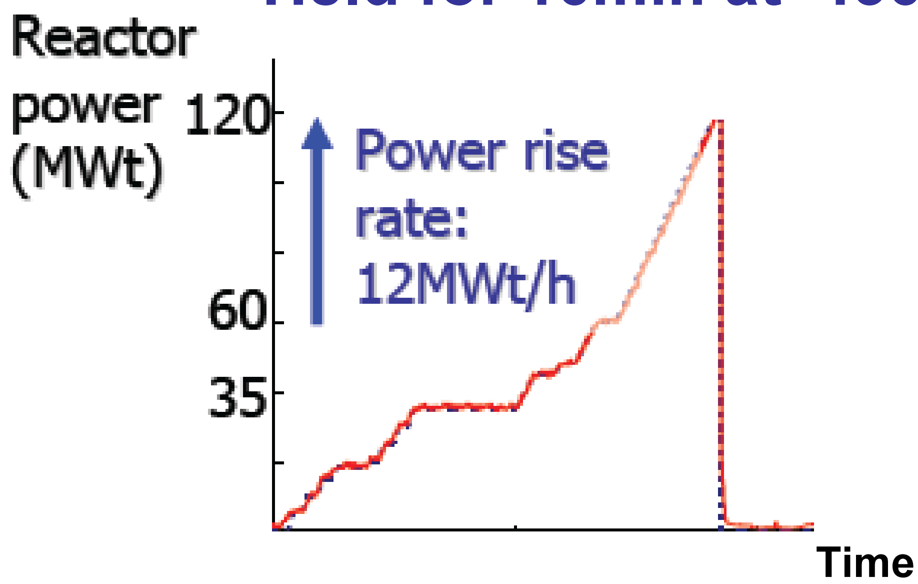


O/M=1.98

Fuel Pin

Pin No.	Name	Composition	O/M
No.1-1	MA-MOX	$(Np_{0.016}Am_{0.016}Pu_{0.3}U_{0.668})O_y$	1.98
	MOX	$(Pu_{0.3}U_{0.7})O_y$	1.98

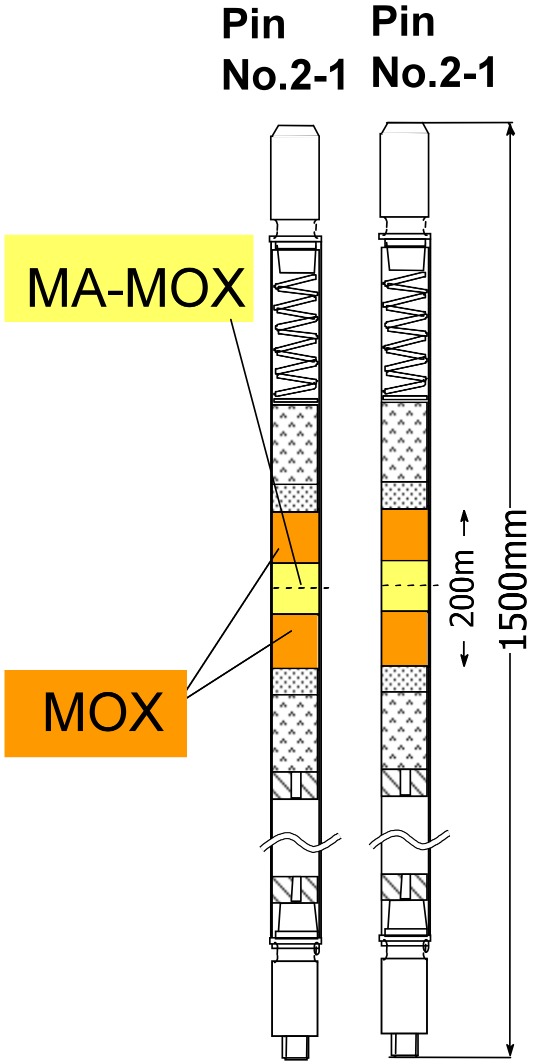
Held for 10min at 430W/cm



Irradiation pattern

Irradiation test

Condition 2



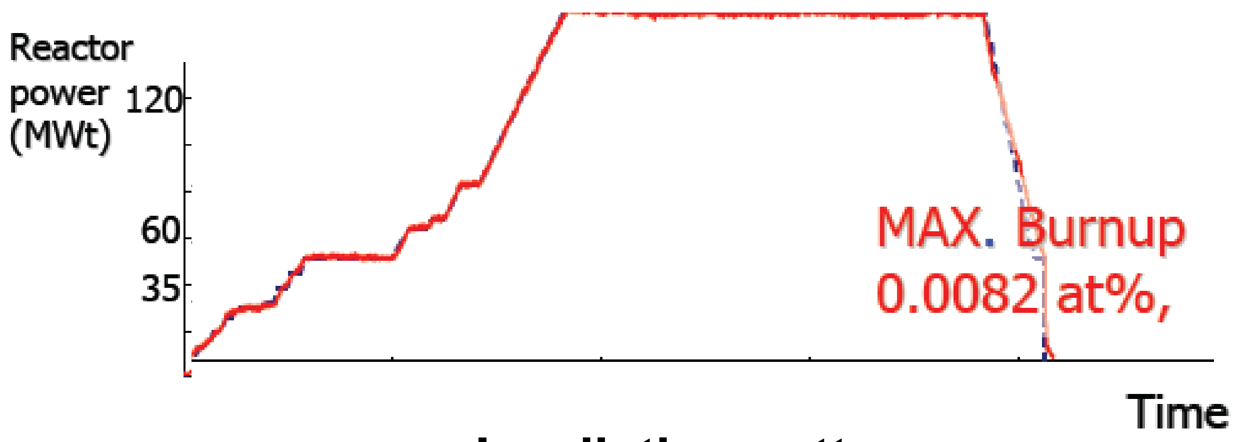
O/M=1.98 , 1.96

Fuel Pins

Second Test

Pin No.	Name	Composition	O/M
No.2-1	MA-MOX	$(Np_{0.016}Am_{0.016}Pu_{0.3}U_{0.668})O_y$	1.98
	MOX	$(Pu_{0.3}U_{0.7})O_y$	1.98
No.2-2	MA-MOX	$(Np_{0.016}Am_{0.016}Pu_{0.3}U_{0.668})O_y$	1.96
	MOX	$(Pu_{0.3}U_{0.7})O_y$	1.96

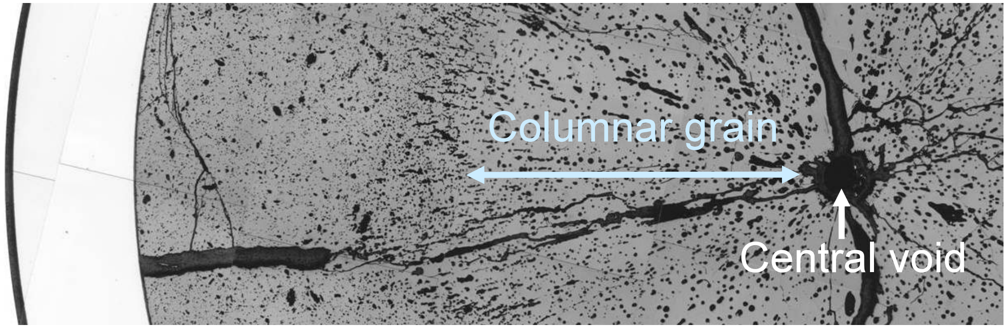
Held for 24h at 430W/cm



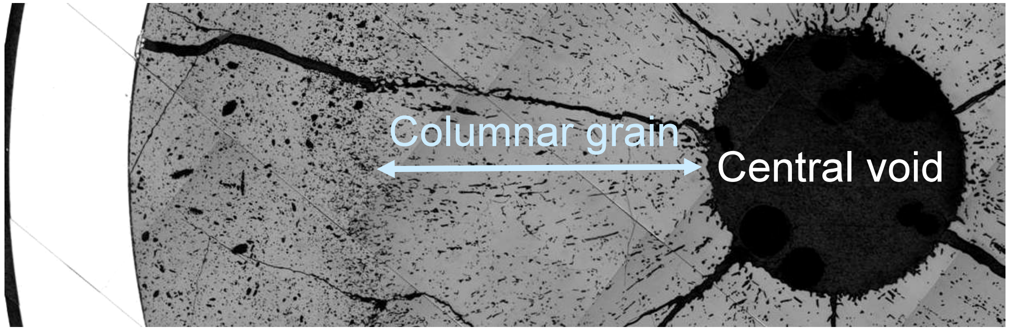
Irradiation pattern

PIE Results

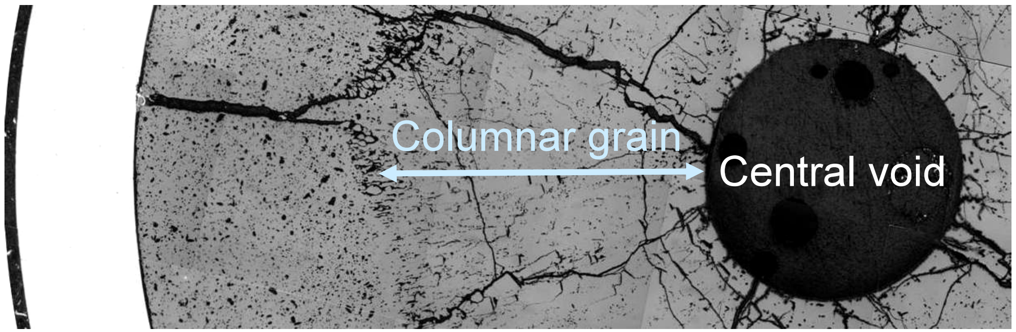
Microstructure Change



No.1-1 MA-MOX, O/M=1.98, at 427W/cm for **10 min**



No.2-1 MA-MOX, O/M=1.98, at 432W/cm for **24h**



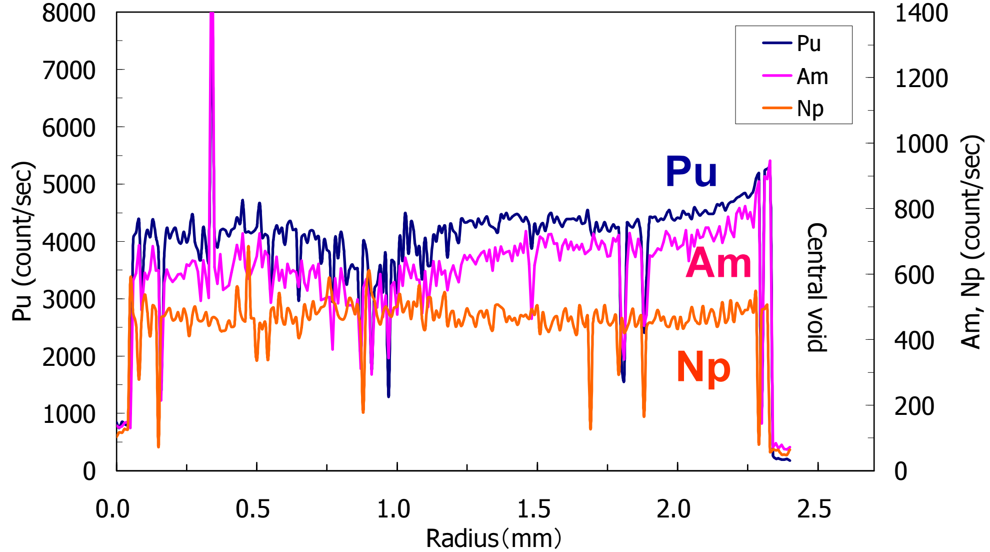
No.2-2 MA-MOX, O/M=1.96, at 429W/cm for **24h**

Observation results

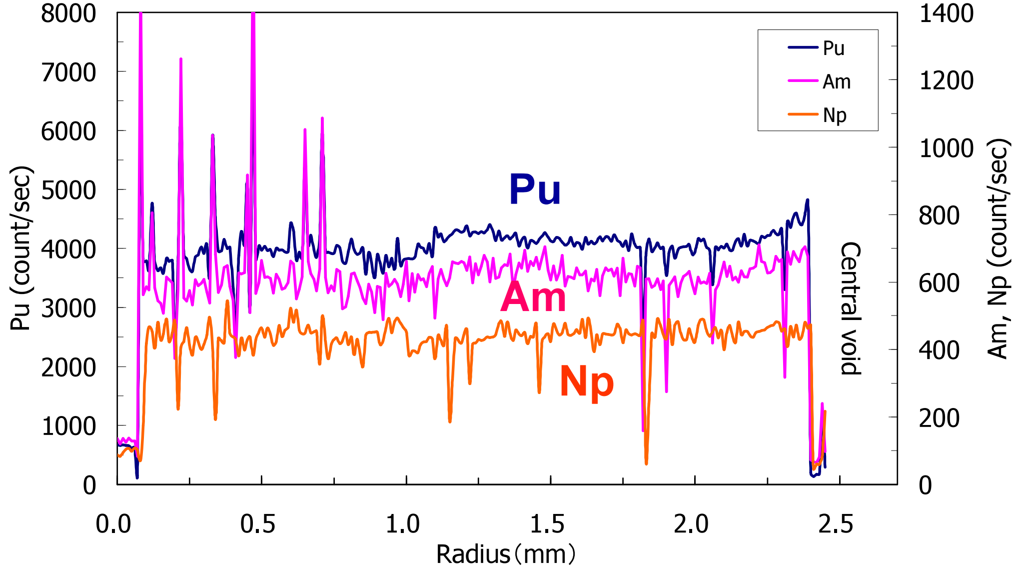
1. The heat rate of the fuels attained about 430W/cm.
2. The pellets were unmelted.
3. Pores migrated to the center.
4. Columnar grain and central void were observed

PIE Results

Actinide Redistribution



No.2-1 MA-MOX, O/M=1.98, at 432W/cm for 24h



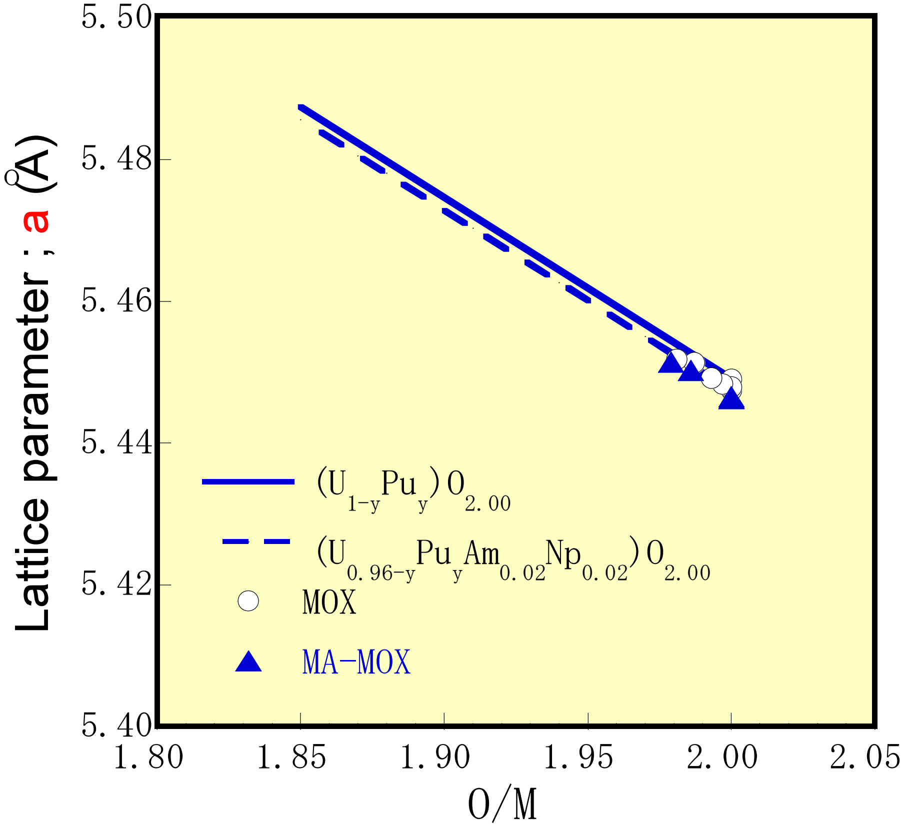
N0.2-2 MA-MOX, O/M=1.96, at 429W/cm for 24h

Observation results

1. The content of Pu and Am increased at pellet center.
2. The increment of Pu and Am decreased with decreasing O/M.
3. The contents of Np and U were flat.

Properties of fuel pellets (1/4)

Lattice Parameter



Lattice Parameter vs. O/M ratio

$$a = 4/\sqrt{3}(r_c(1+0.112x)+r_a)$$

$$r_c = (r_U(1-z-y'-y'')+r_{Pu}z+r_{Am}y'+r_{Np}y'')$$

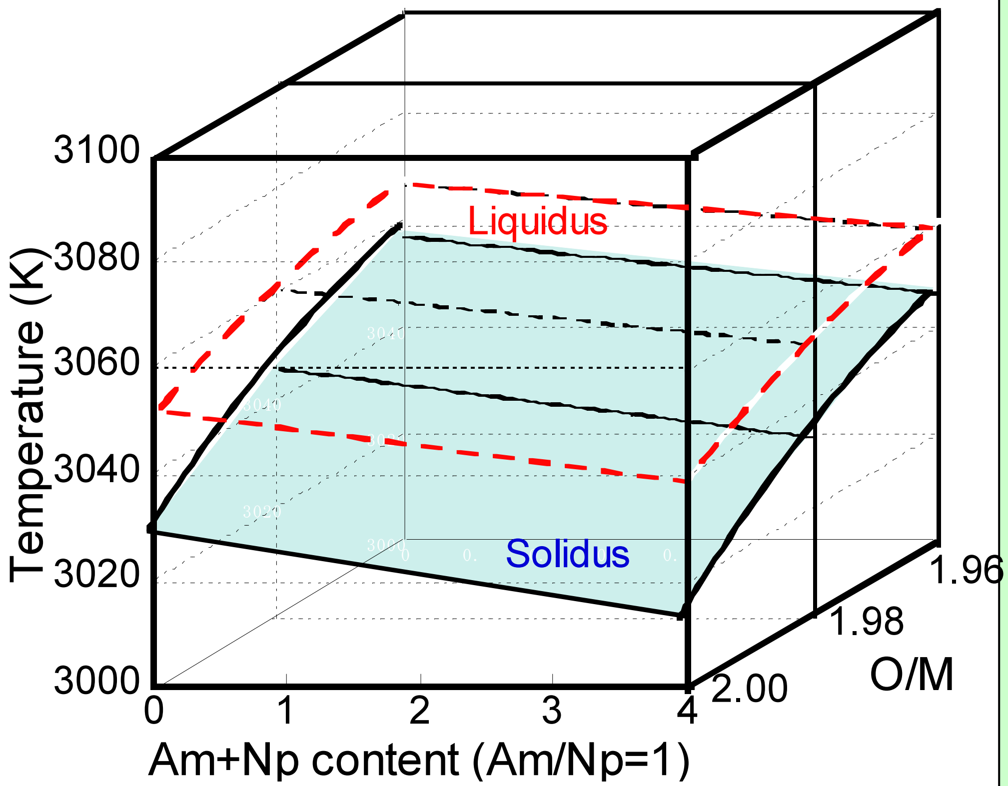
M.Kato, et al.,J.N.M.385 (2009) 117

Results

1. The model to represent the lattice parameter was derived.
2. MA content slightly caused the lattice parameters to decrease.
3. The difference of theoretical density is 0.01-0.02g/cm^3.

Properties of fuel pellets (2/4)

Melting Temperature



Variation of Solidus and Liquidus Temp.

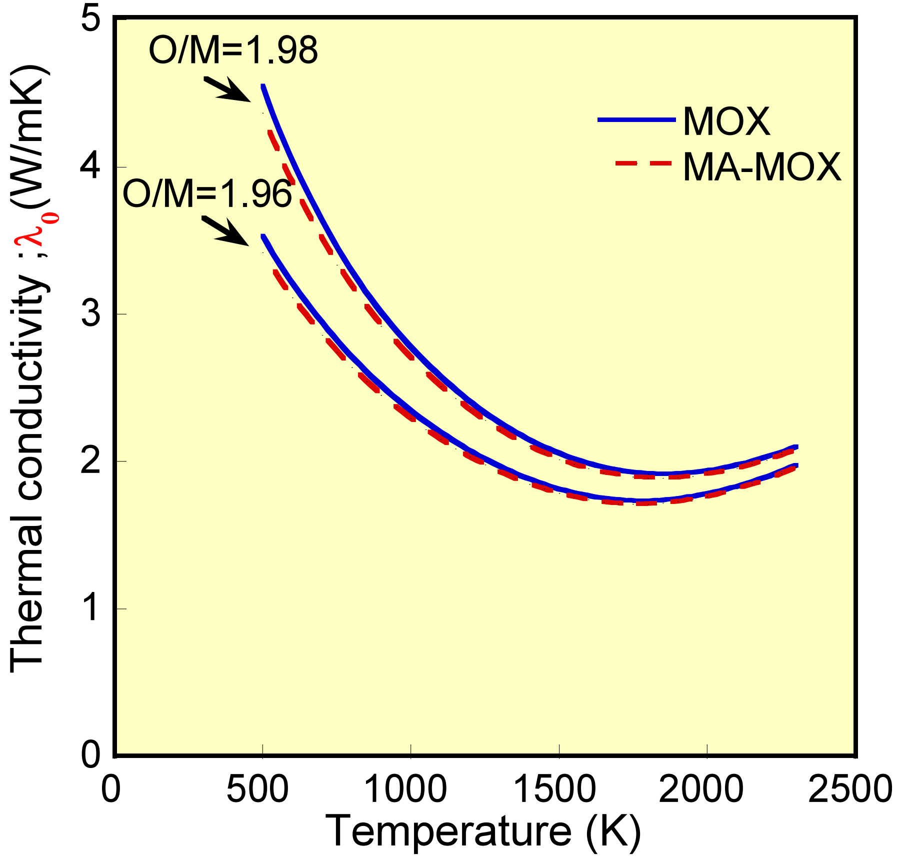
M.Kato, et al.,ICAPP '09

Results

1. The data were measured and analyzed by the ideal solid solution model.
2. MA content caused to decrease by 2-3K /%MA.
3. The solidus temperature of MA-MOX is over 3000K.

Properties of fuel pellets (3/4)

Thermal conductivity



Thermal conductivity

Results

1. The data were measured as functions of MA content, density, O/M and temperature.
2. MA content caused to decrease slightly in temperatures of less than 1000K.

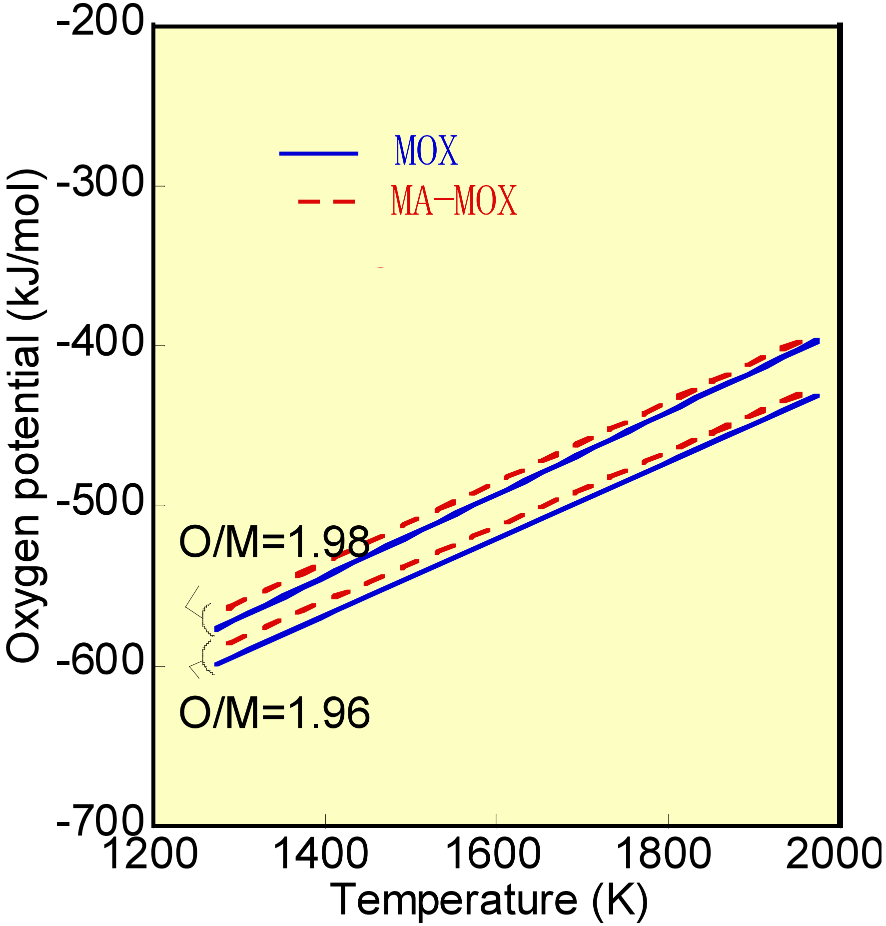
$$\lambda_0 = \frac{1}{\left(2.713x + 3.583\times10^{-1}\times z_1 + 6.317\times10^{-2}\times z_2 + 1.595\times10^{-2}\right) + \left(-2.625x + 2.493\right)\times10^{-4}T} + \frac{1.541\times10^{11}}{T^{5/2}}\exp\left(\frac{1.522\times10^{4}}{T}\right)$$

z_1: Am content
z_2: Np content
x:Deviation x in MO_{2-x}

M.Kato, et al., OECD/NEA, Oct. 6-10, 2008, Japan

Properties of fuel pellets (4/4)

Oxygen potential



Oxygen potential

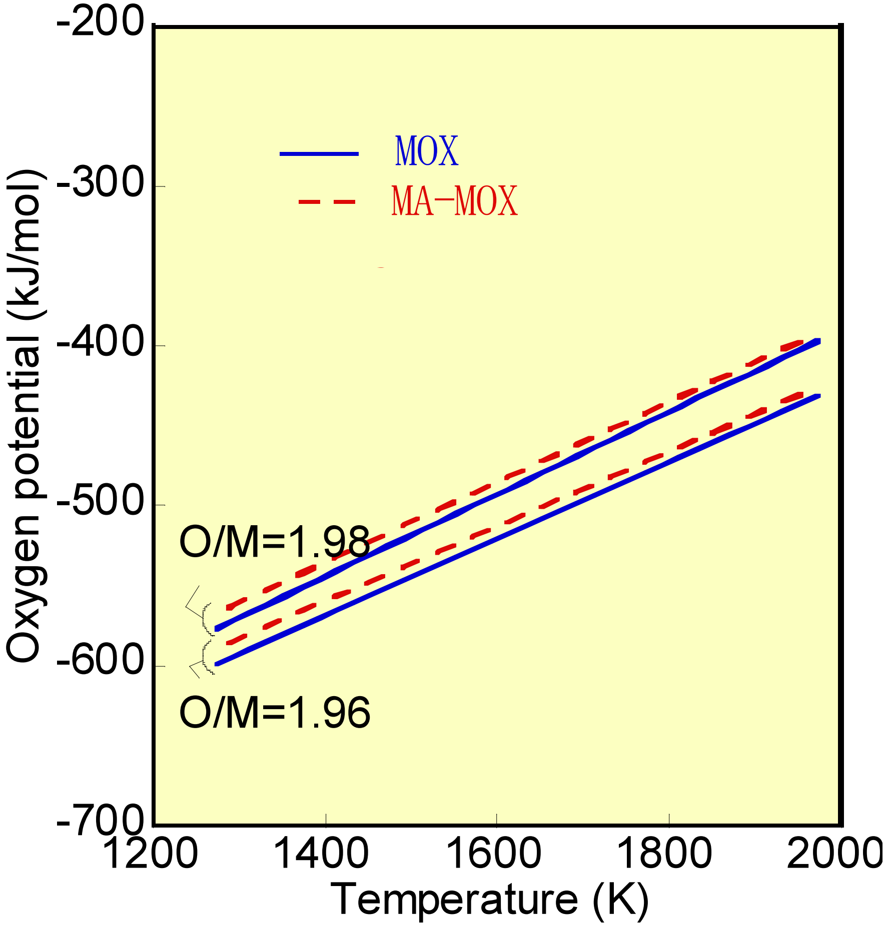
M.Kato, et al.,J.N.M. 385 (2009) 419

Results

1. The data were measured by gas equilibrium method.
2. MA content caused the oxygen potential to increase slightly.

Properties of fuel pellets (4/4)

Oxygen potential



Oxygen potential

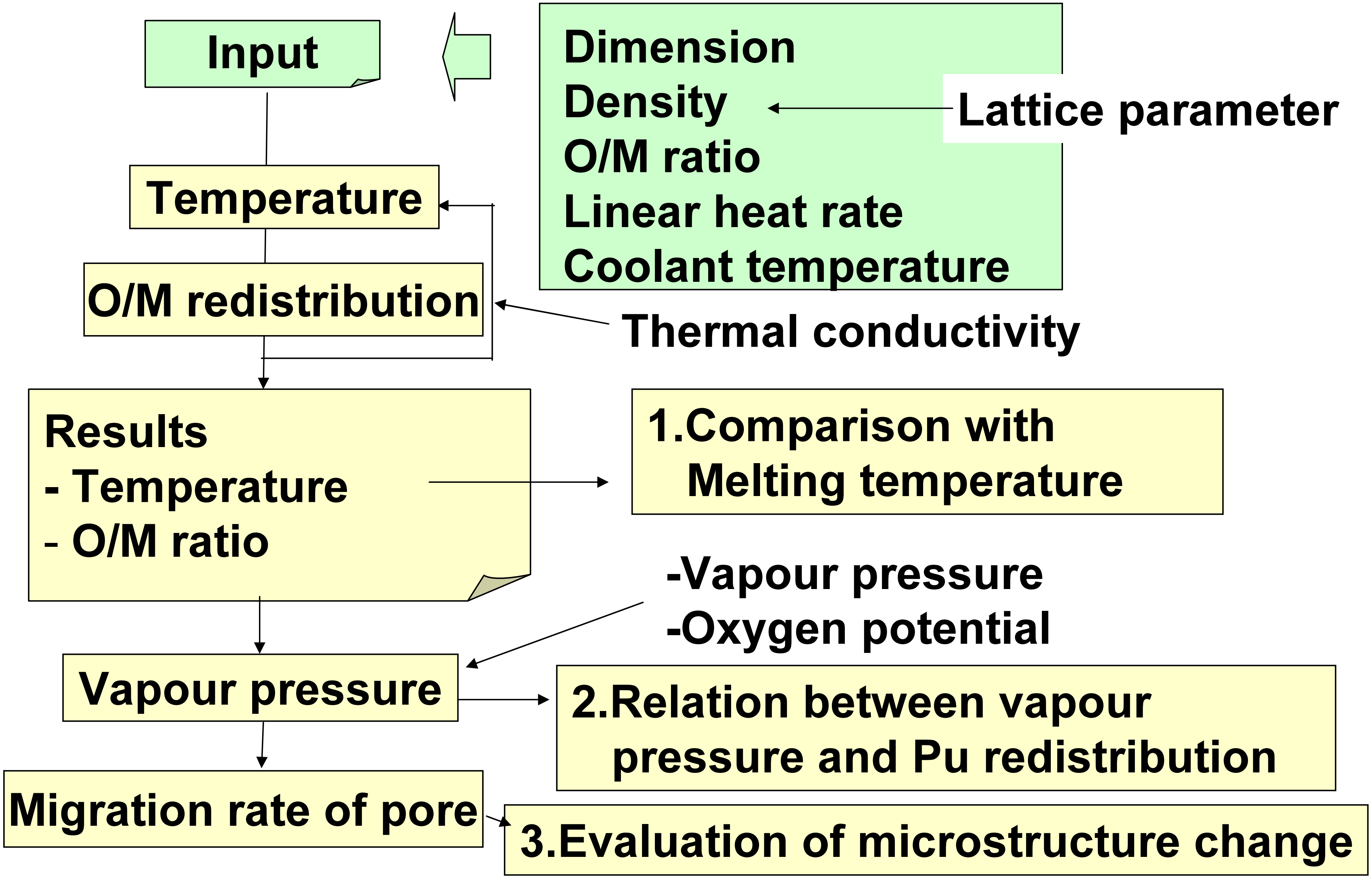
Results

1. The data were measured by gas equilibrium method.
2. MA content caused the oxygen potential to increase slightly.

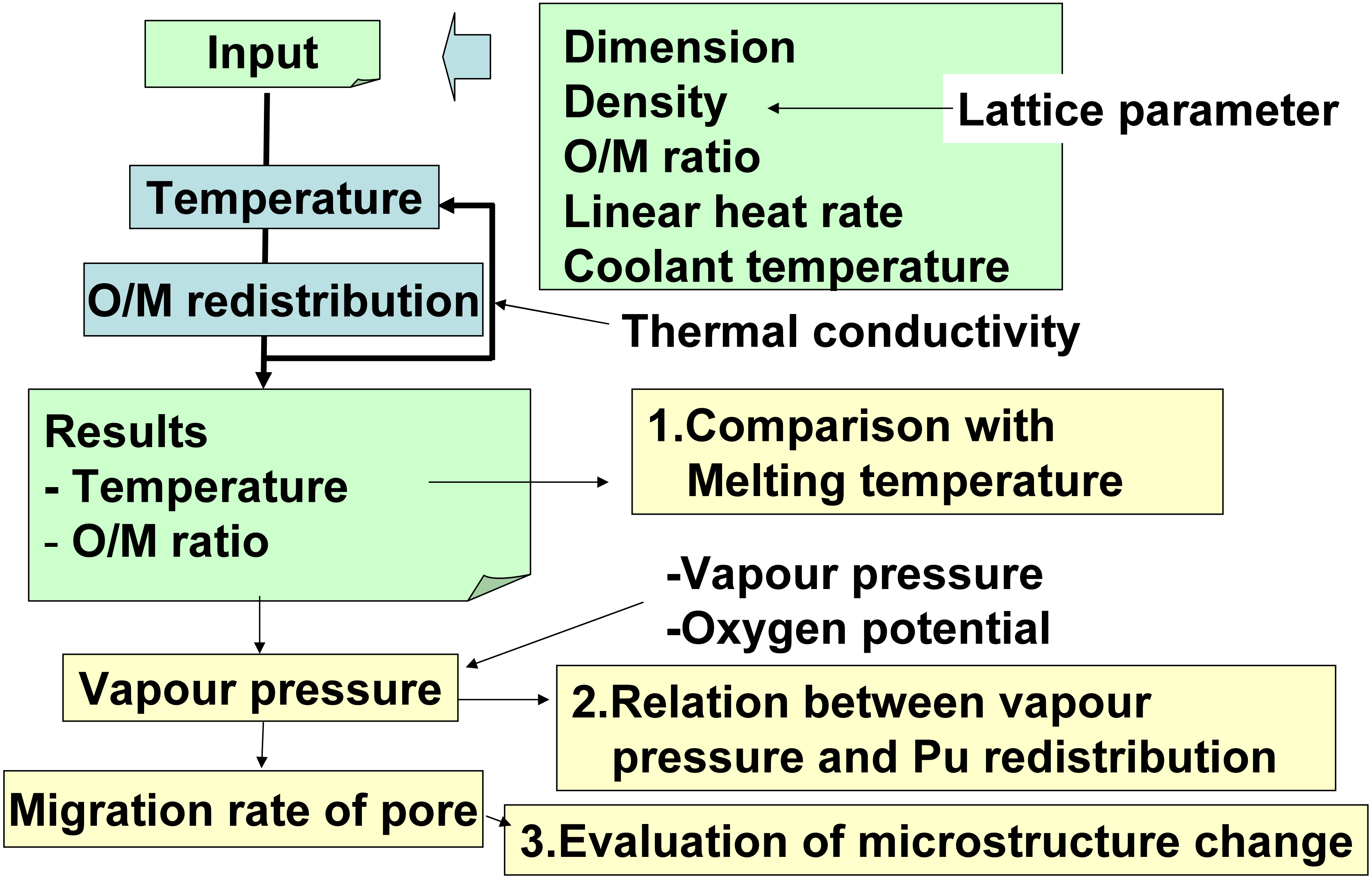
Summary

The effect of MA addition on the properties is negligibly small in the operation temperature range of FR fuels

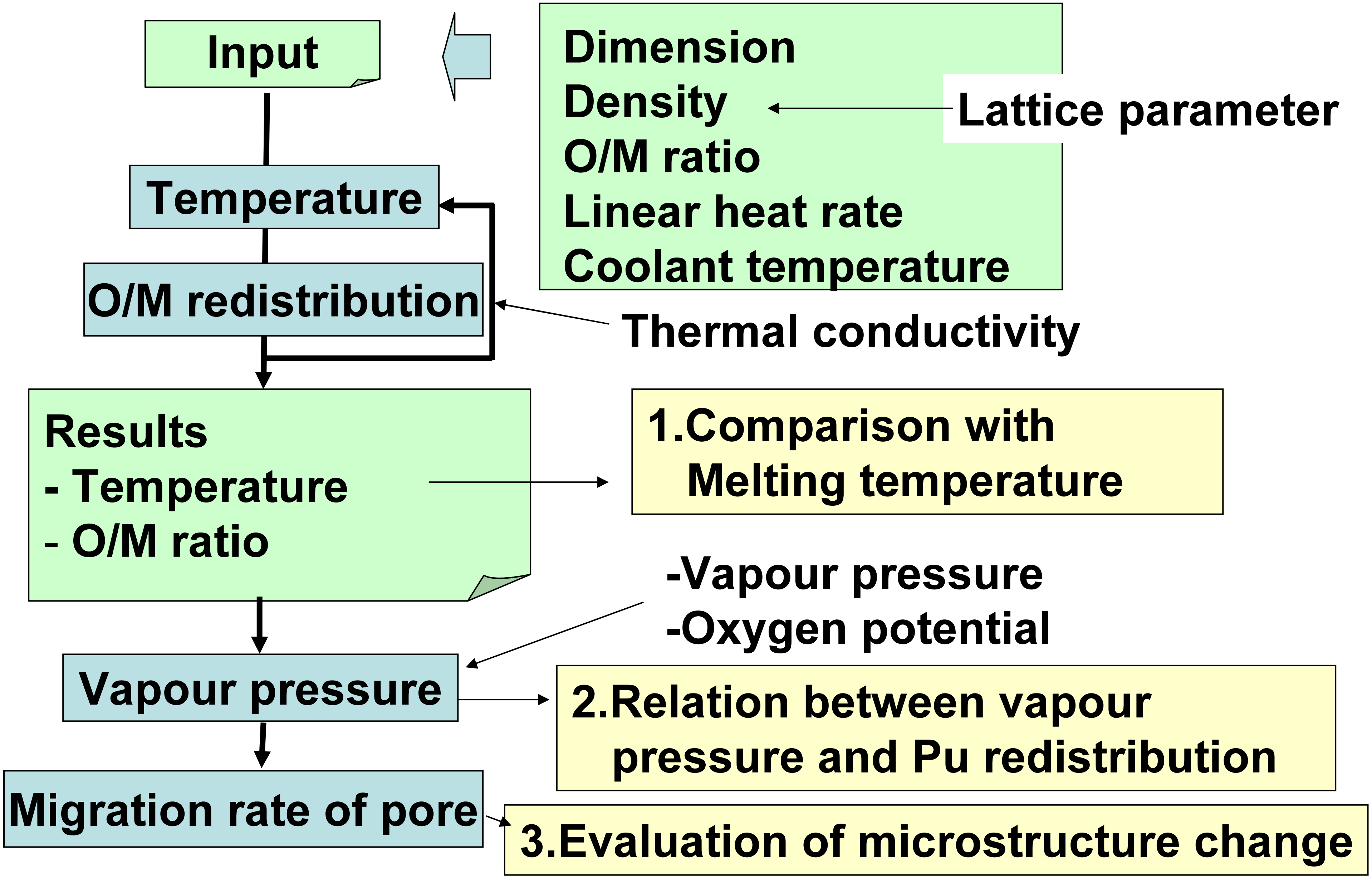
Analysis of irradiation behaviour
Analysis Procedure



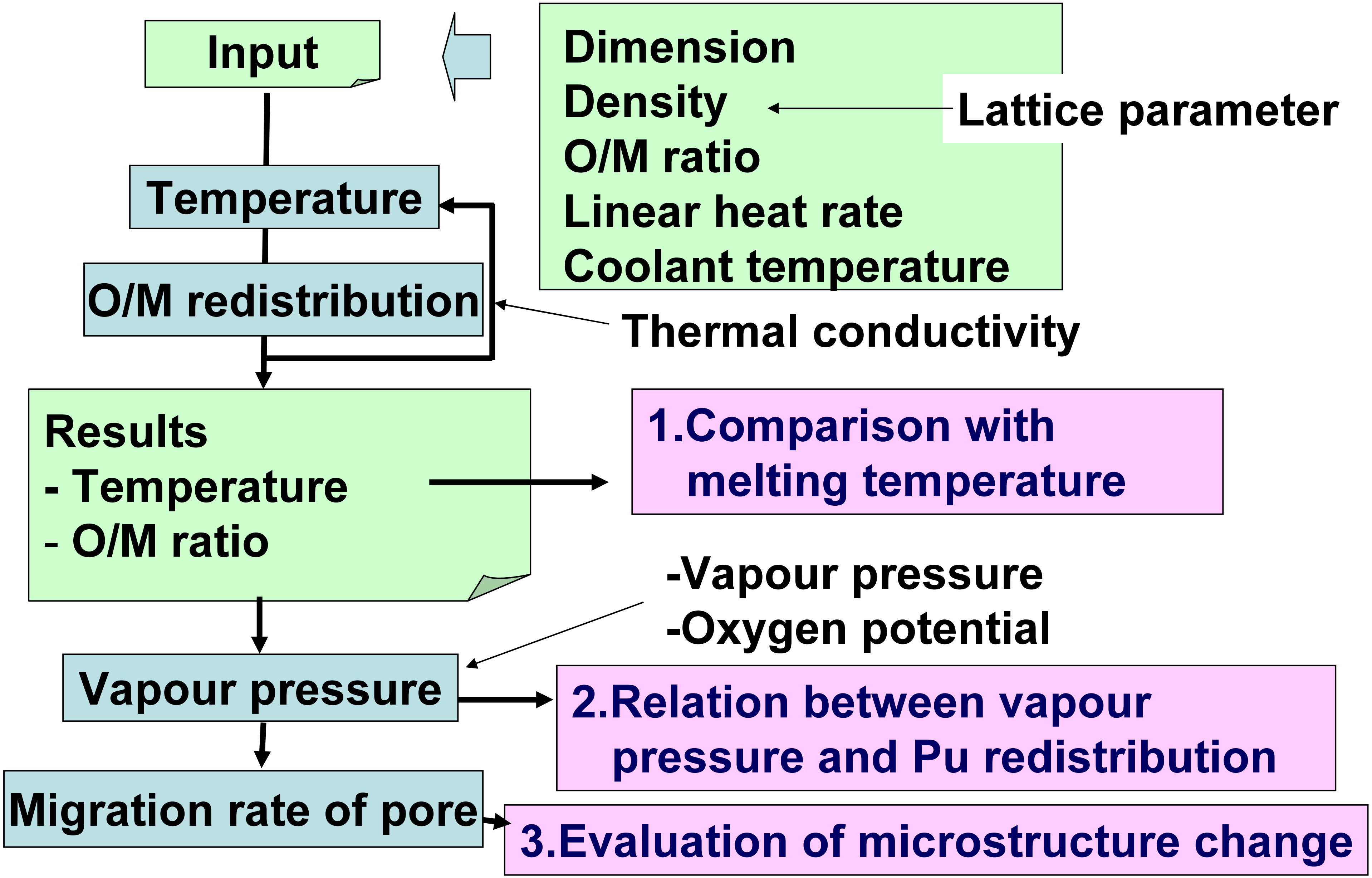
Analysis Procedure



Analysis Procedure



Analysis Procedure



Analysis Procedure

Input

- Dimension
- Density ← Lattice parameter
- O/M ratio
- Linear heat rate
- Coolant temperature

Temperature

O/M redistribution ← Thermal conductivity

Results
- Temperature
- O/M ratio

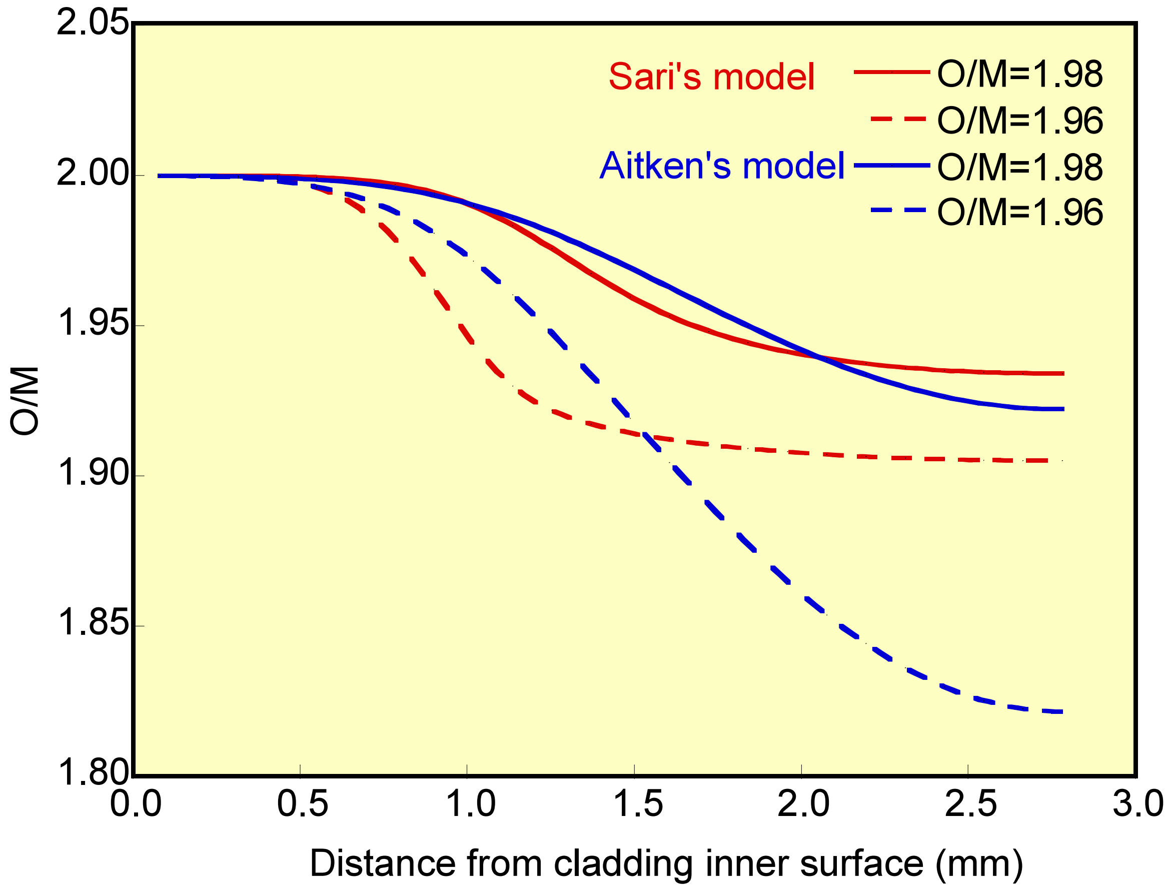
→ 1.Comparison with Melting temperature

Vapour pressure ← -Vapour pressure, -Oxygen potential

— 2.Relation between vapour pressure and Pu redistribution

Migration rate of pore → 3.Evaluation of microstructure change

O/M Redistribution

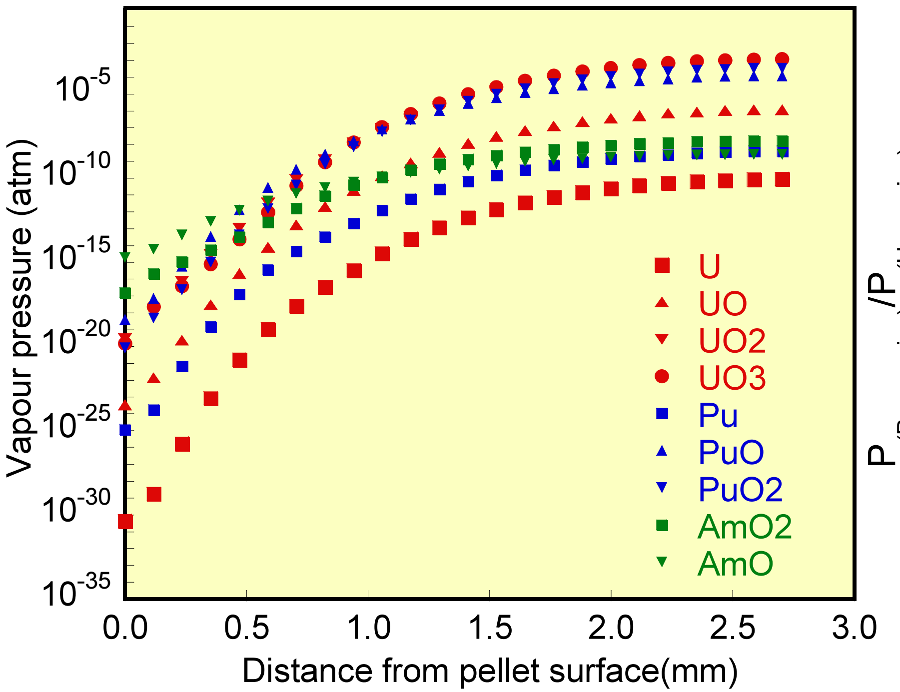


O/M ratio distribution in the radial direction

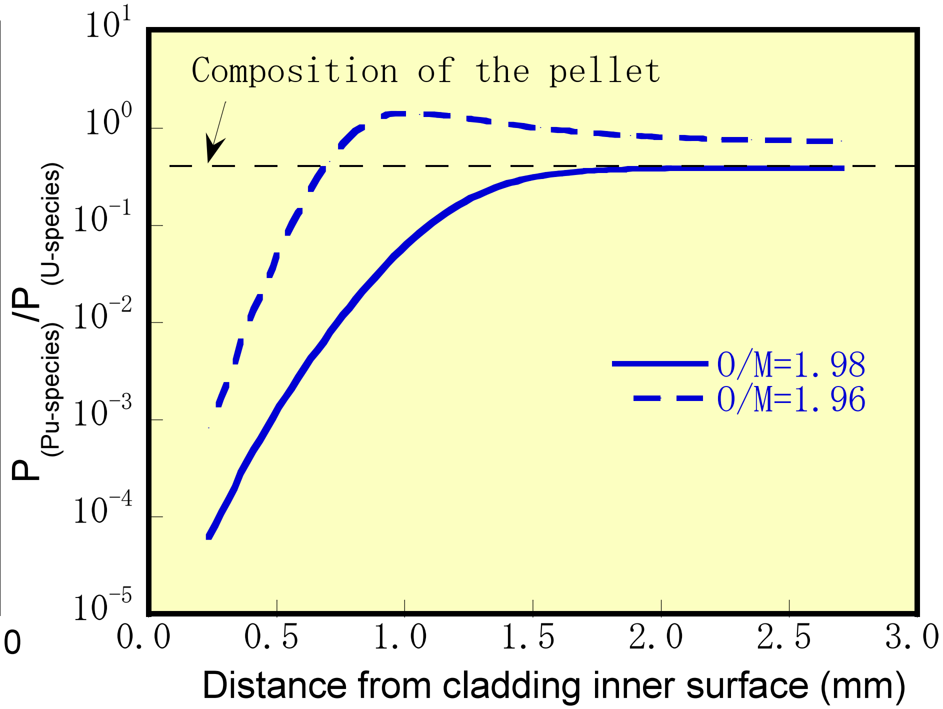
Results

1. Both models gave almost the same result in high O/M pellets.
2. The results of low O/M pellets differed according to models.
3. Sari’s model was employed.

Evaluation of vapour pressure



Variation of vapor pressure

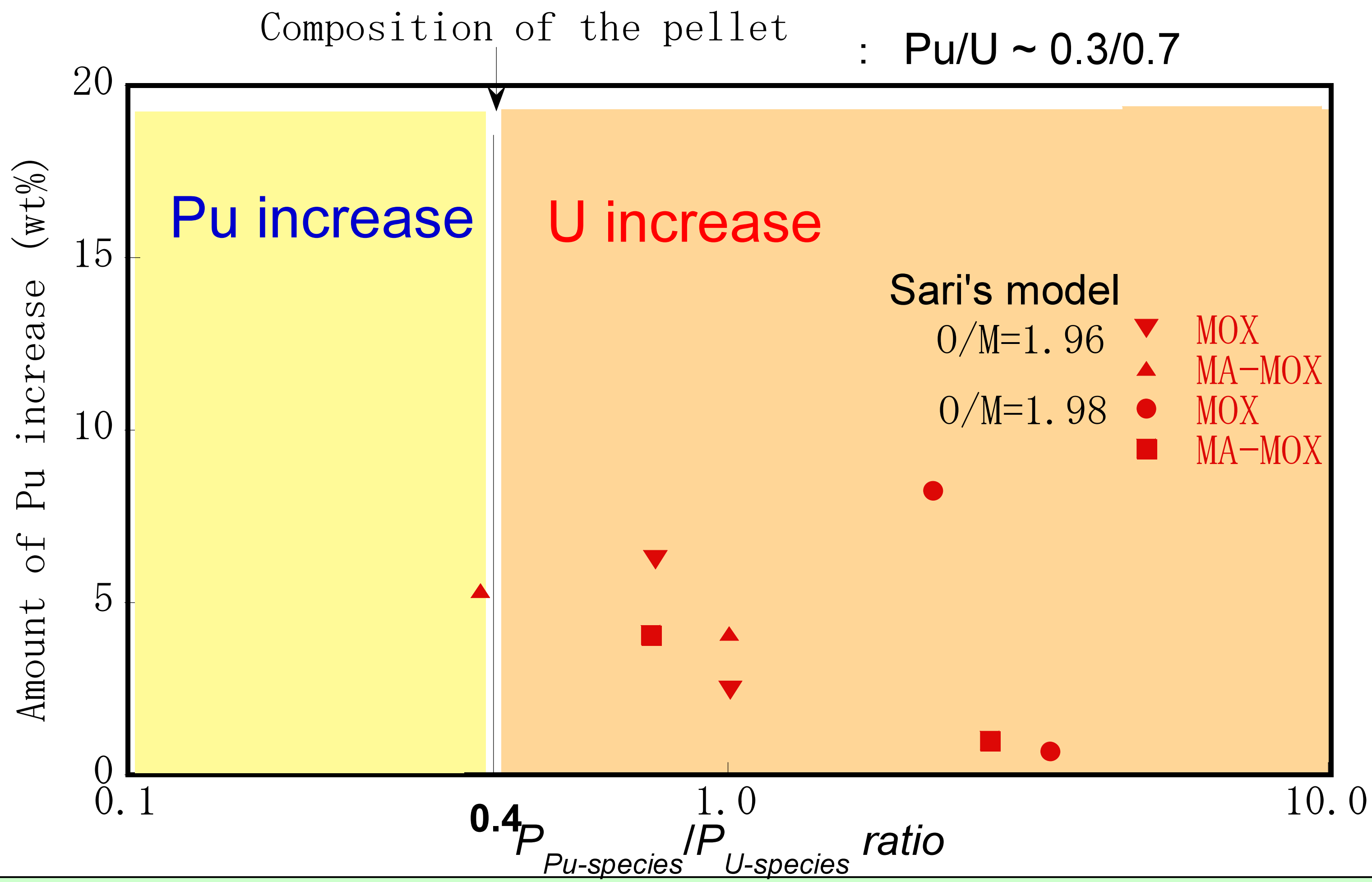


Variation of Pu-/U- species

Result

1. Variation of vapor pressure ratio of Pu/U-species was evaluated using Rand –Markin model.

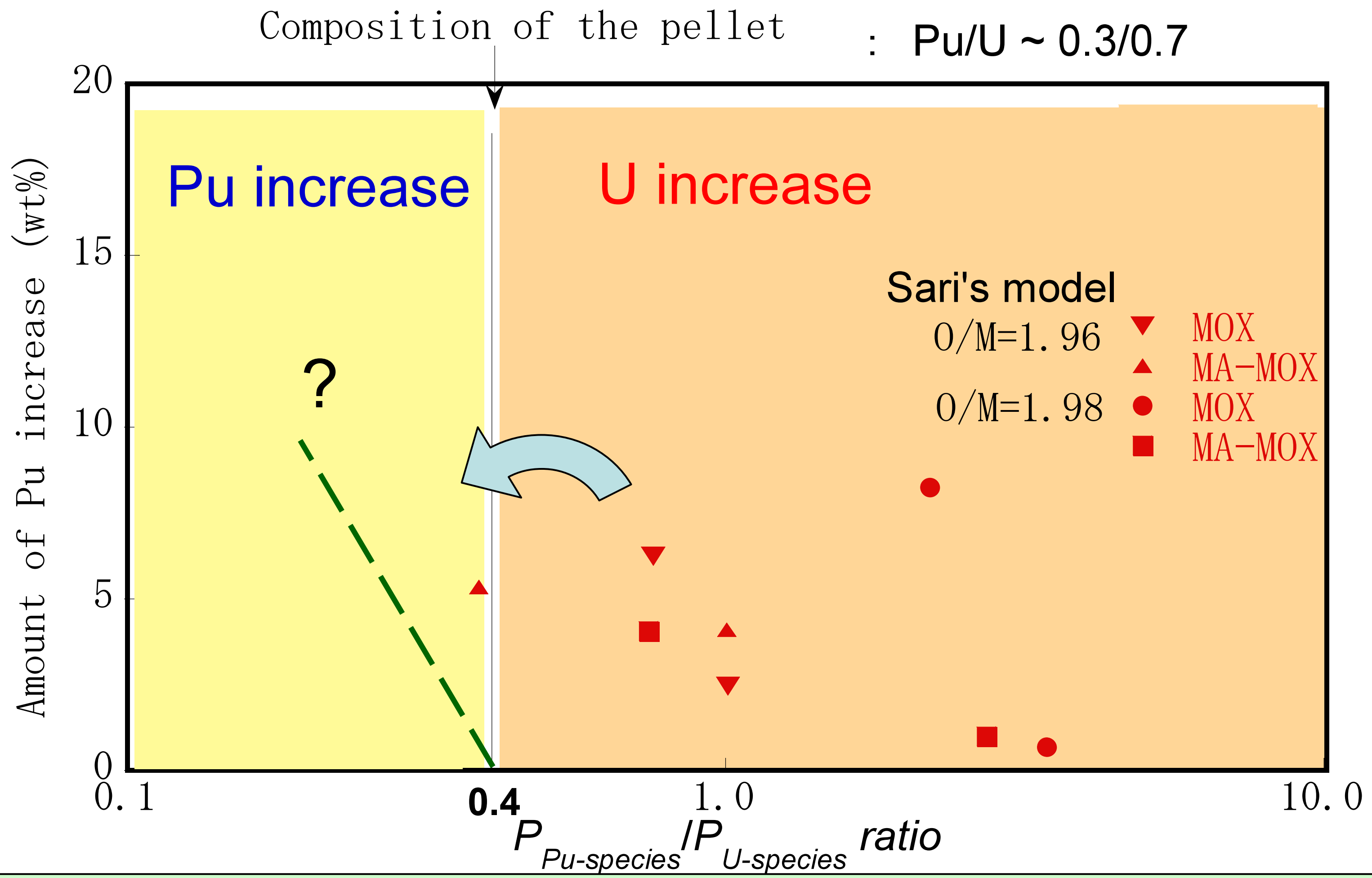
Evaluation of Pu redistribution



Result

1. Calculation result shows that U content increases at pellet center.

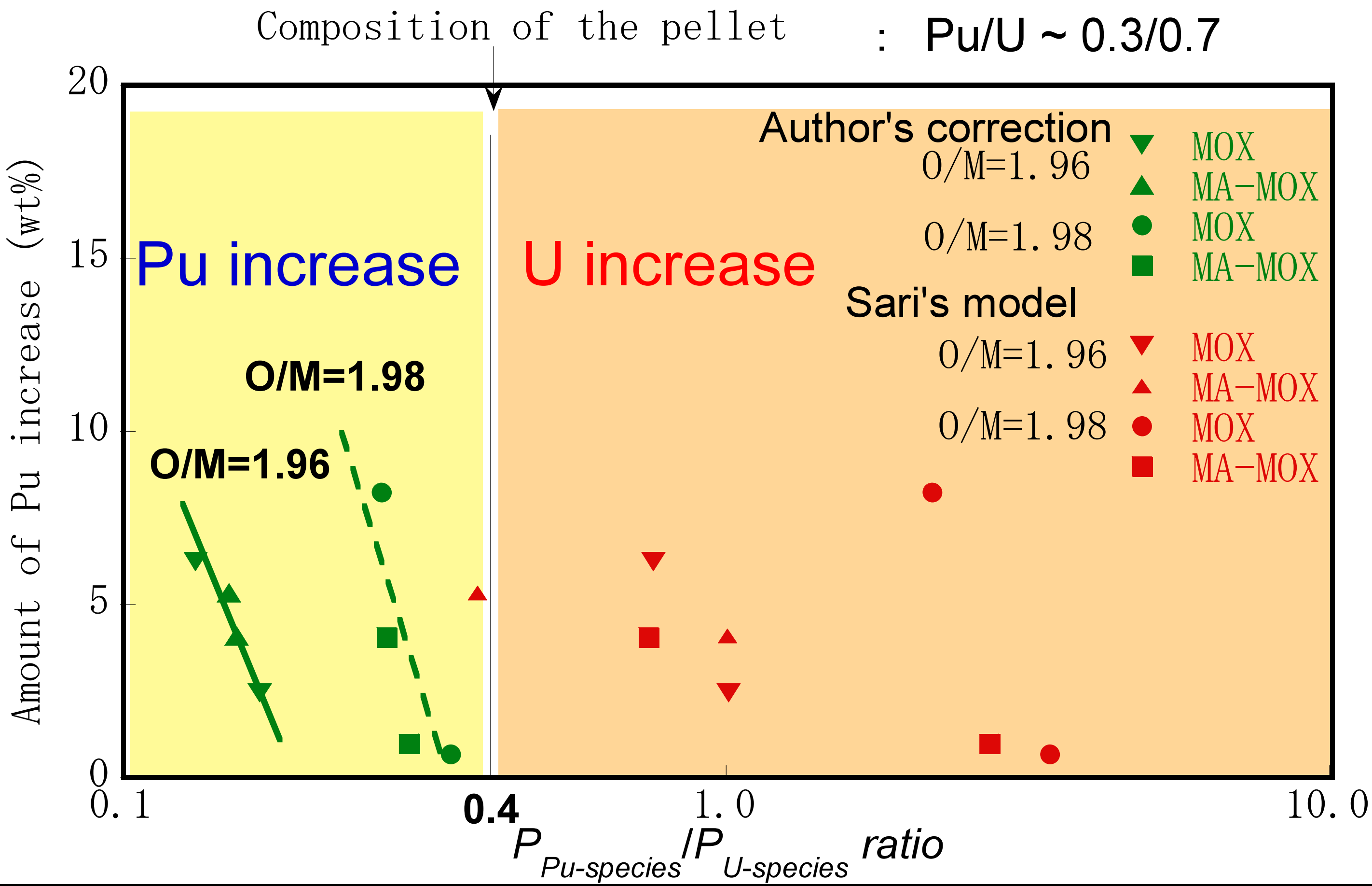
Evaluation of Pu redistribution



Expectation

Increment of Pu should be increased with decreasing the $P_{Pu\text{-}species}/P_{U\text{-}species}$.

Evaluation of Pu redistribution



Results

1. Calculation results show that Pu content increase at pellet centre.
2. Two lines were obtained depending on O/M ratio.

Reviewing of the model for O/M redistribution

Sari's model for O/M redistribution

$$\ln\left(x_1 / x_2\right) = \frac{Q*}{R}\left(\frac{1}{T_2} - \frac{1}{T_1}\right)$$

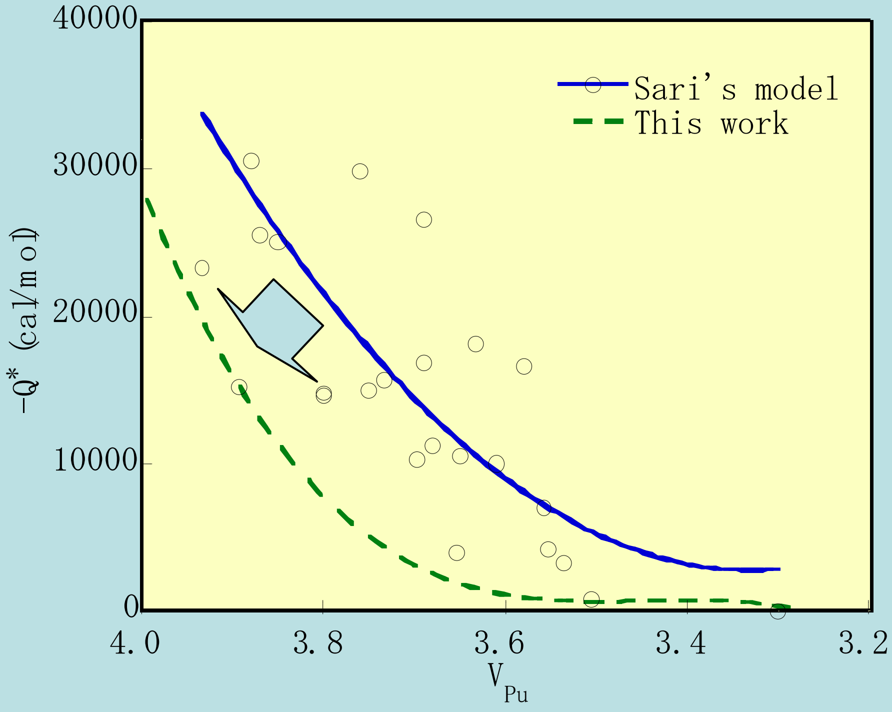
$$Q* = -9.45\times10^5 + 5.66\times10^5 V_{Pu} - 8.5\times10^4 V_{Pu}^2$$

X: deviation in $(U,Pu)O_{2-x}$
T: temperature
R : gas constant,
Q* : the molar effective heat of transport
V_{Pu} : Pu valence

Evaluation of equation for Q*

Calculation of O/M Redistribution, temperature and vapour pressure

Evaluation of the relationship between the $P_{Pu\text{-}species}/P_{U\text{-}species}$ and the Pu increase



Corrected Eq.

$$Q* = -7.01\times10^6 + 6.1012\times10^6 V_{Pu} - 1.7705\times10^6 V_{Pu}^2 - 1.7122\times10^5 V_{Pu}^3$$

Reviewing of the model for O/M redistribution

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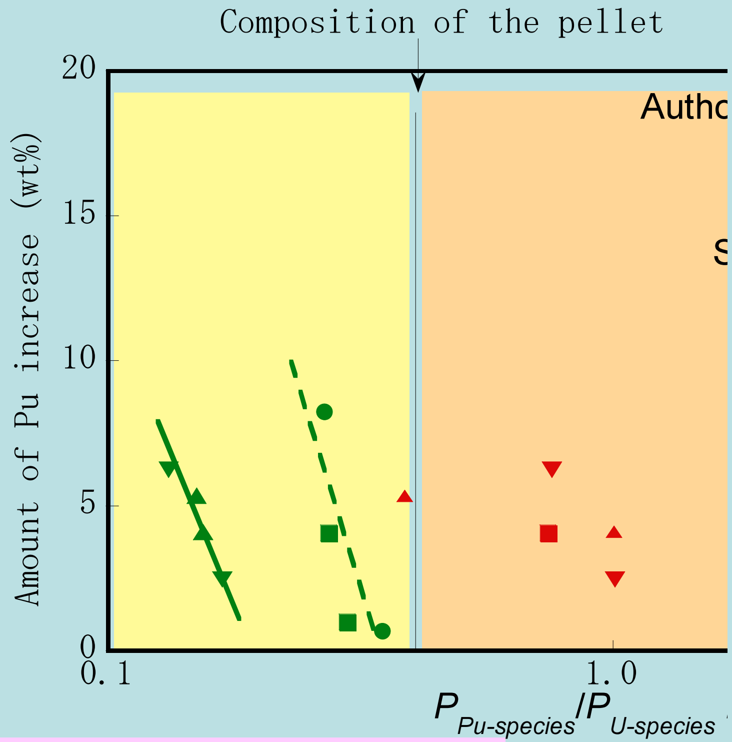
T: temperature
R: gas constant,
Q*: the molar effective heat of transport
V_{Pu}: Pu valence

Evaluation of equation for Q*
↓
Calculation of O/M Redistribution, temperature and vapour pressure
↓
Evaluation of the relationship between the $P_{Pu\text{-}species}/P_{U\text{-}species}$ and the Pu increase
↓ (loop back to Evaluation of equation for Q*)



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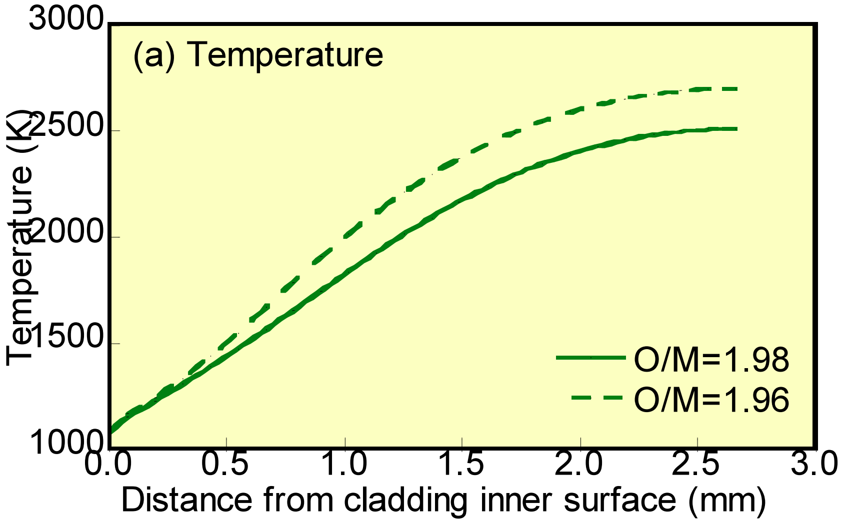
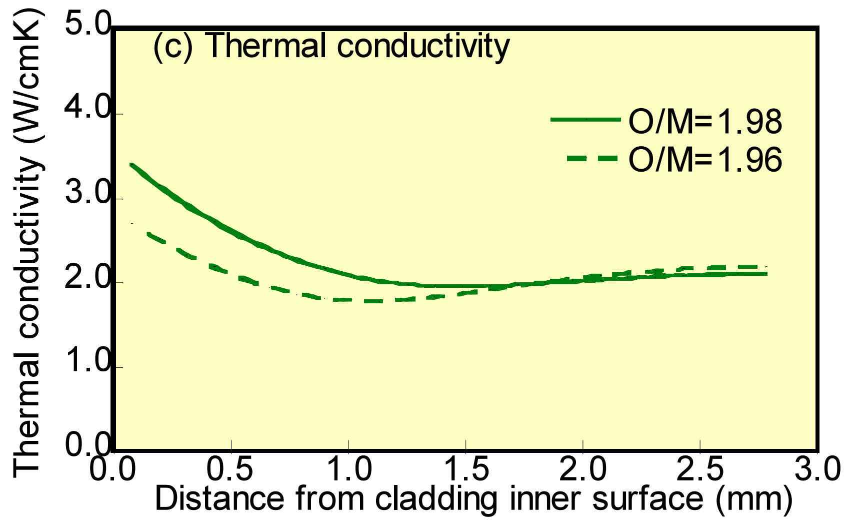
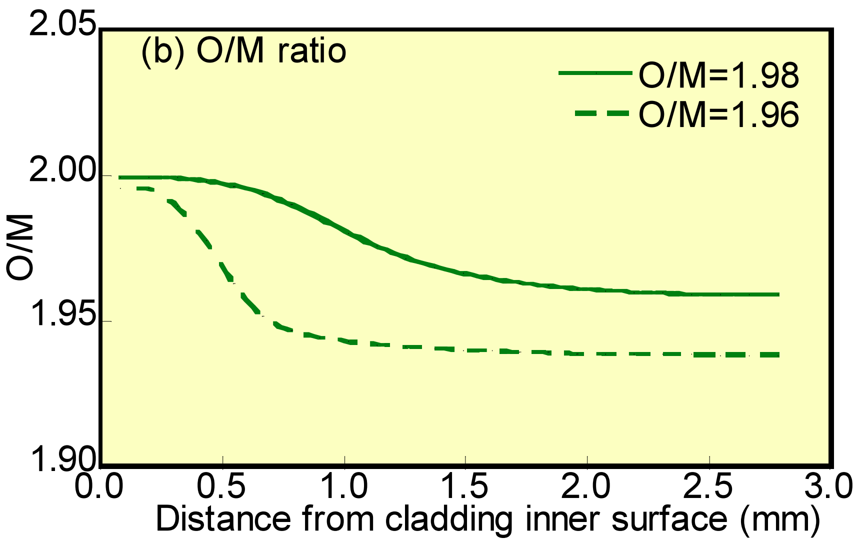
Calculation results in MA-MOX pellets



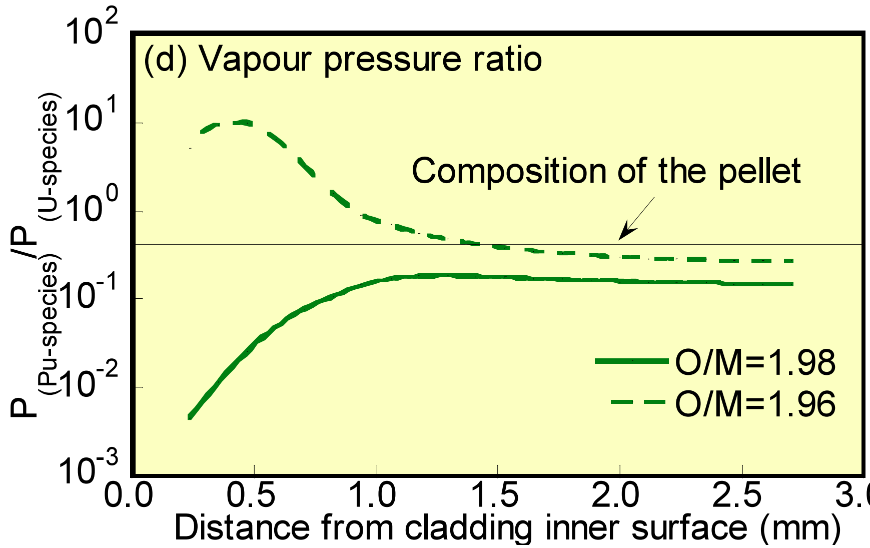
Temperature



Thermal conductivity

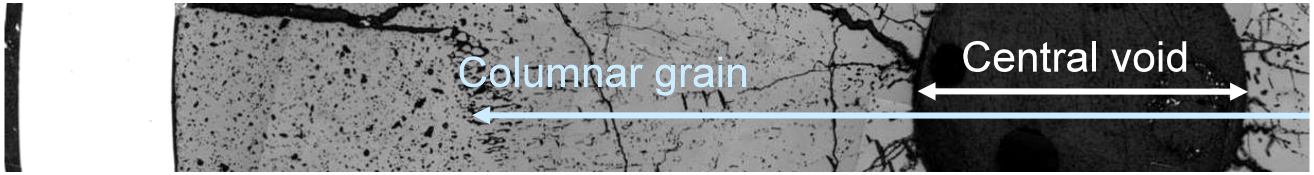


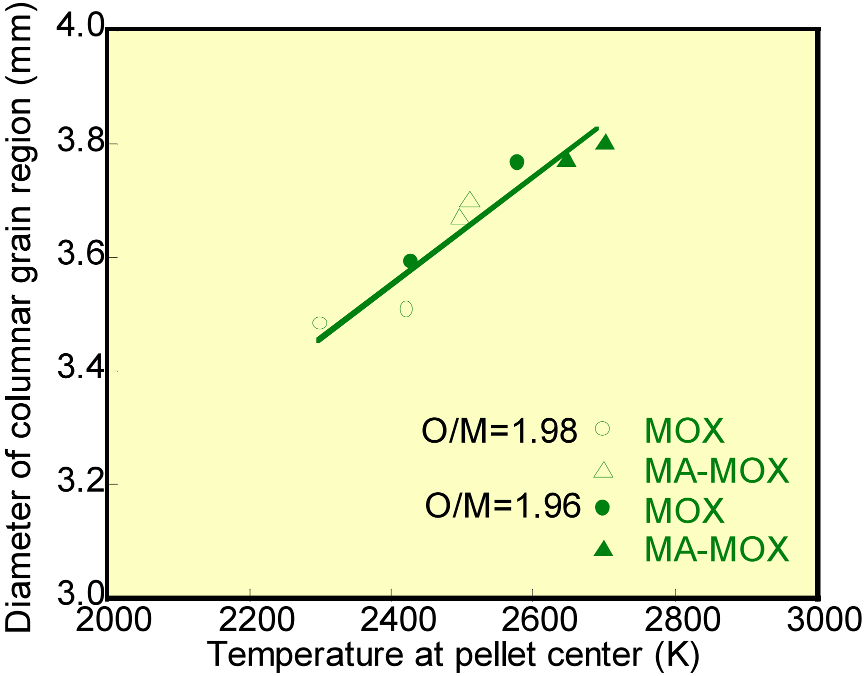
O/M ratio



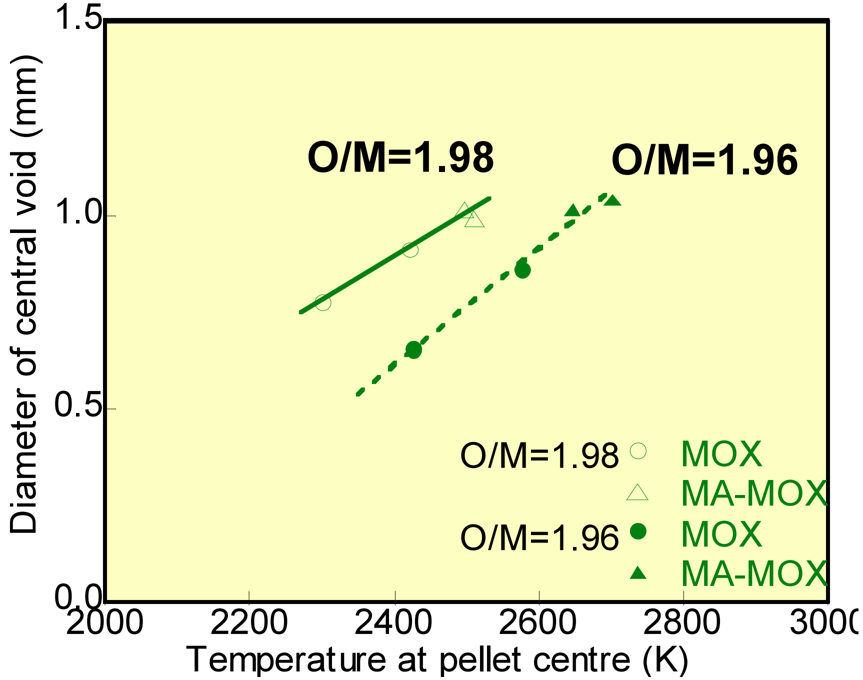
Vapour pressure ratio of Pu-/U-species

Microstructure change





Columnar grain region



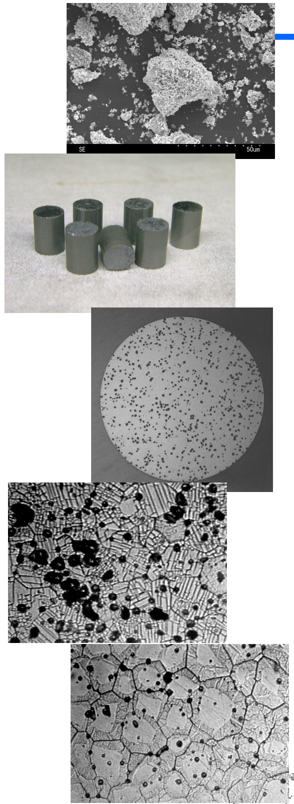
Central void

Results

1. The both region grew with temperature.
2. The central void of high O/M pellet is larger than that of low O/M pellets.

Summary

1. ***The physical properties of the Np/Am-MOX were evaluated, and the effect of Np/Am addition was negligibly small.***
2. ***The relation between the Pu redistribution and the vapor pressure was described by correcting Sari's model for O/M redistribution.***
3. ***The low O/M pellet attained higher temperature, however, the diameter of the central void was small as compared with the high O/M pellet.***
4. ***It is needed to measure experimental data and to derive advanced models for describing irradiation behavior of low O/M fuel.***



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