

# DEVELOPMENT OF MINOR ACTINIDE TRANSMUTATION BY CRIEPI

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# Metal Fuel FBR & Pyro-process

Innovative fuel cycle system is under development in CRIEPI

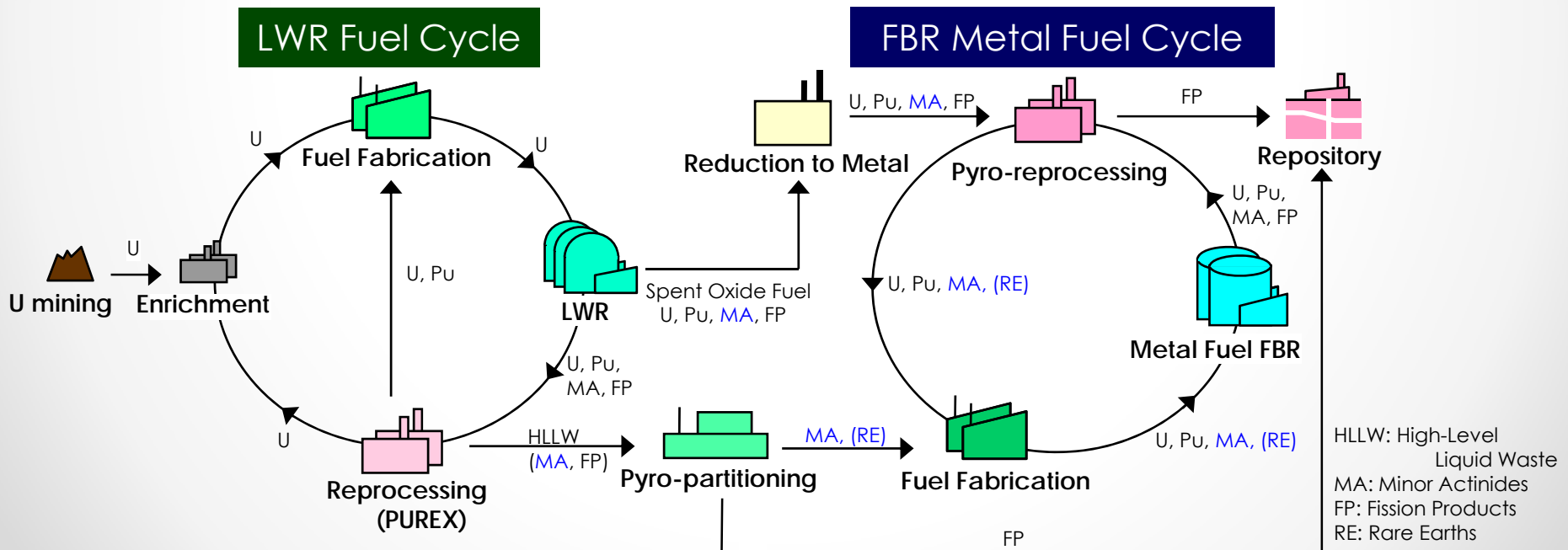
U-Pu-Zr Fuel FBR : Excellent nuclear performances & safety features

Pyro-reprocessing : Simultaneous recovery of MA with U and Pu

Injection Casting Fuel Fabrication : Simple remote-control operation

Goals:

Security of the long-term energy supply,  
Reduction of the amount and the toxicity of radioactive waste,  
Improvement of the proliferation resistance.



# MA-Containing Metal Fuel Development

- How much content of MA should be loaded in metal fuel FBR?

## *Evaluation of expected fuel compositions*

Burnup and recycle calculations of MA & RE in metal fuel FBR cycle  
Mass flow analysis based on the future fuel cycle scenario

- How about the effect of MA addition in metal fuel?

## *Development of MA- and RE-containing U-Pu-Zr alloys*

Ex-reactor experiments  
Irradiation experiment  
postirradiation examinations



Characterization of U-Pu-Zr-MA-RE

**Various experimental studies on U-Pu-Zr-MA(-RE) alloys are performed in cooperation with JRC-ITU.**

# MA Burnup Performance in Metal Fuel FBR

Large-scale & high-burnup metal fuel core design is assumed as a model of commercial FBR.

Output	1,500MWe / 3,900MWt
Core residence time	6years
Coolant temp. inlet / outlet	355 / 510°C
Max. cladding temp.	650°C
Max. linear power	500W/cm
Ave. discharge burnup	150GWd/t

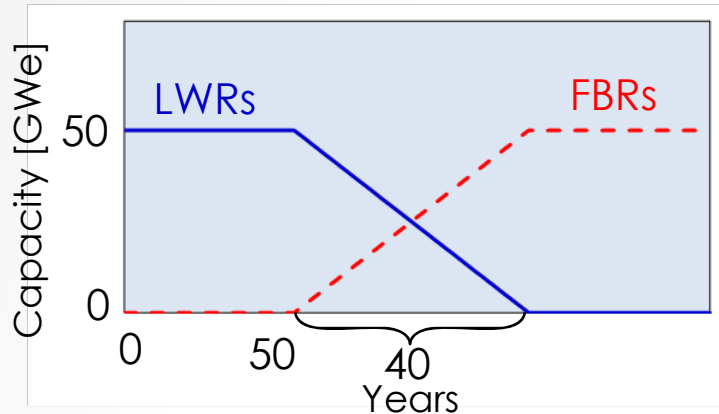
Feed compositions and core performance parameters

	No-MA-makeup	MA-enriched	
MA content, wt%	0.8	2.0	5.0
RE content <sup>1</sup> , wt%	0.5	1.5	3.5
Pu enrichment, wt%	16.6	16.4	15.8
Makeup MA ratio <sup>2</sup> , %	-	37	53
Doppler const., $Tdk/dT$	$-2.3 \times 10^{-3}$	$-2.2 \times 10^{-3}$	$-2.1 \times 10^{-3}$
Coolant coeff., $\rho/^\circ C$	0.254	0.273	0.310

<sup>1</sup>: D.F. = 10, <sup>2</sup>: (MA from LWR) / (MA in FBR fuel)

# Mass Flow Evaluation (1)

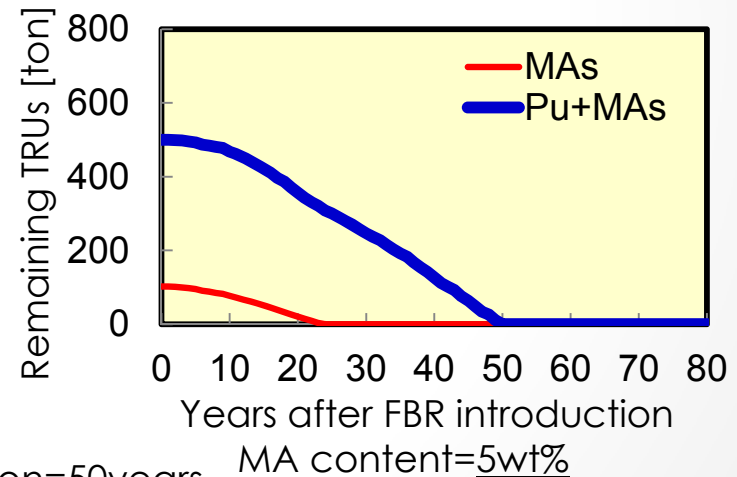
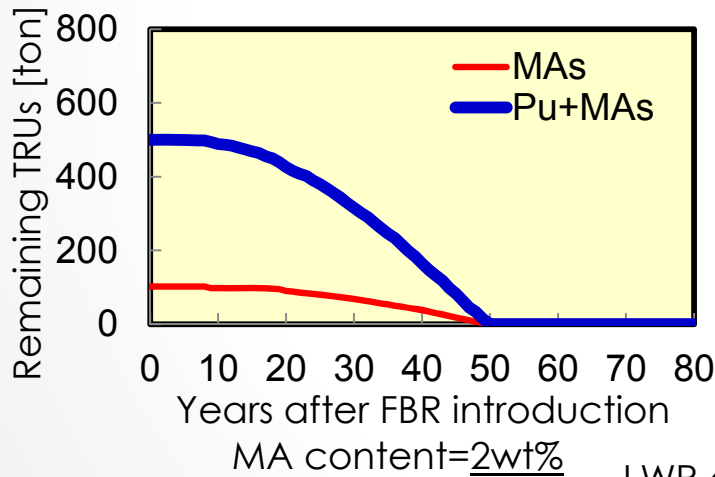
## Transition Scenario from LWRs to FBRs



### Assumptions

50GWe of LWRs are operated for 50years before FBR introduction,  
 Reactor lifetime is 40 years,  
 MA content in the feed is 2 or 5wt%.

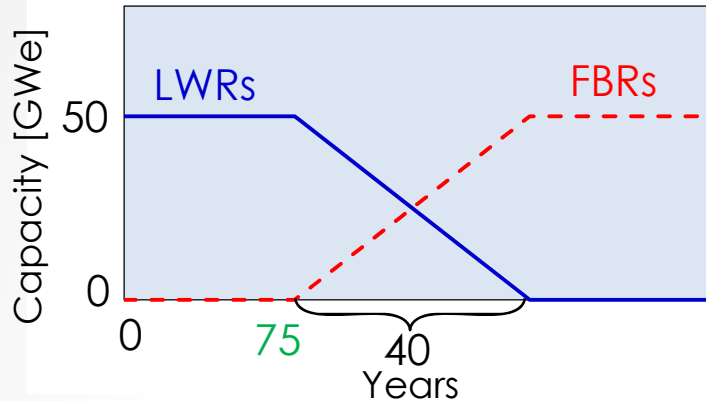
## Mass Balance of Pu & MA



MA content of 2wt%: MA & Pu are recycled at the almost same time,  
 5wt%: MA can be consumed in shorter-term.

# Mass Flow Evaluation (2)

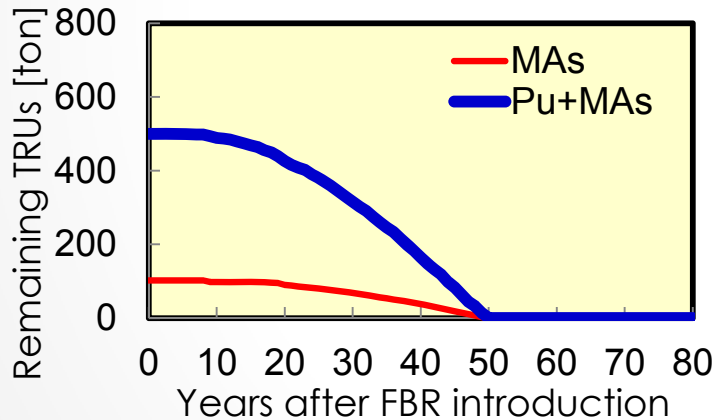
## Transition Scenario from LWRs to FBRs (2)



### Assumptions

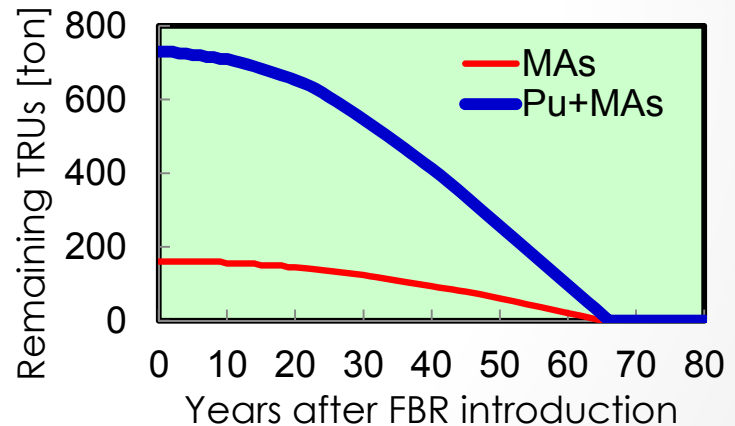
50GWe of LWRs are operated for 75years before FBR introduction,  
 Reactor lifetime is 40 years,  
 MA content in the feed is 2wt%.

## Mass Balance of Pu & MA



LWR operation=50years

MA content=2wt%



LWR operation=75years

MA content of 2wt%: MA & Pu are balanced.

# Miscibilities among U-Pu-Zr-MA-RE

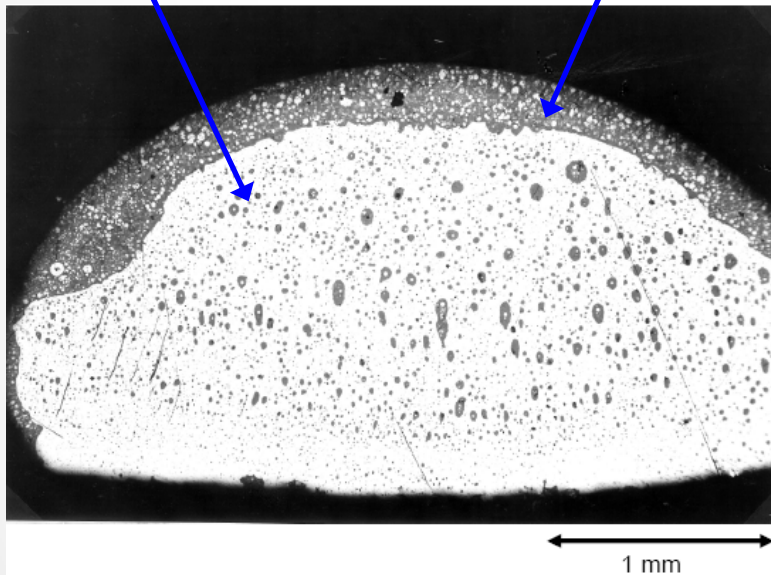
U-Pu-Zr-MA-RE alloys of different compositions were mixed by arc-melting.

→ U-Pu-Zr-MA alloys without RE can be blended homogeneously.

44U-18Pu-10Zr-9Np-5Am-3Ce-10Nd (wt%)

U-Pu-Zr-Np phase

Pu-Am-RE phase

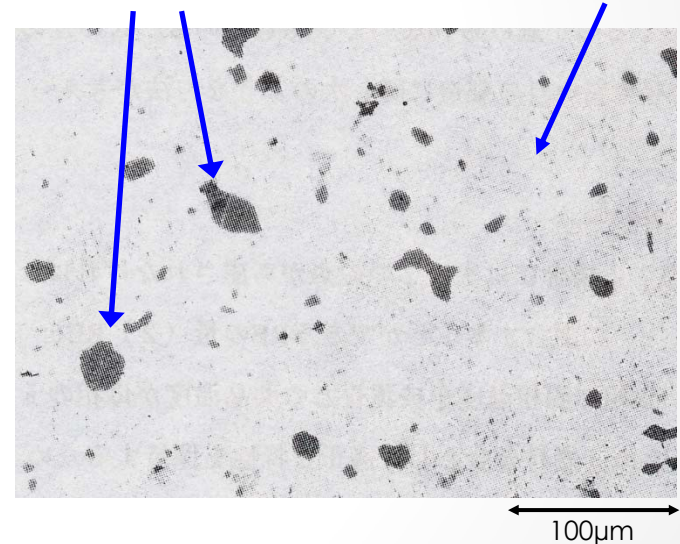


In the alloys of high RE content,  
→ Matrix segregates into upper  
and lower parts.

39U-22Pu-12Zr-15Np-10Am-0.6Ce-1.8Nd

Pu-Am-RE precipitates

U-Pu-Zr-Np phase



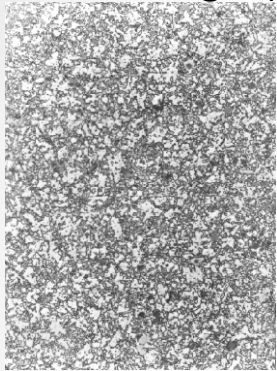
In the alloys of low RE content ( $\leq 5\%$ ),  
→ RE-rich precipitates were  
uniformly dispersed.

RE  $\leq 5\%$  can be mixed in U-Pu-Zr-MA matrix.

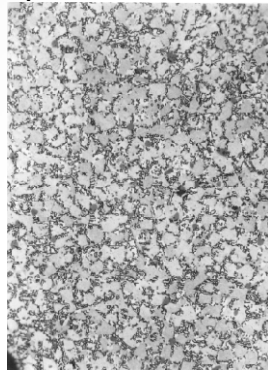
# Phase Structures of annealed U-Pu-Zr-MA-RE

U-Pu-Zr-MA-RE alloys were annealed and quenched.

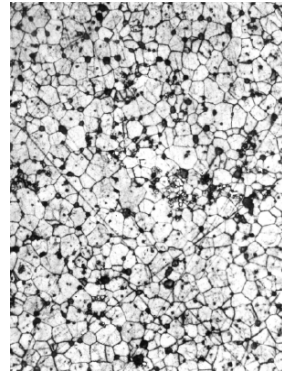
Metallography of U-Pu-Zr-2MA-2RE.



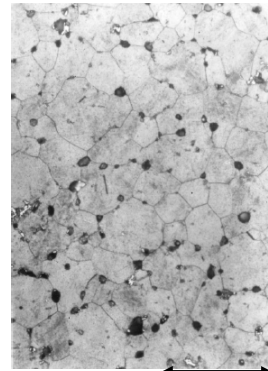
500°C



600°C



700°C

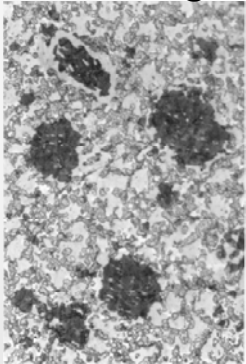


50mm  
850°C

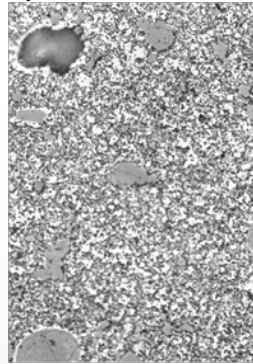
Matrix phase

- ≤ 600°C: Two phase structures  
ζ+δ at 500°C  
γ+δ (or ζ+δ) at 600°C
- ≥ 700°C: Single γ-phase

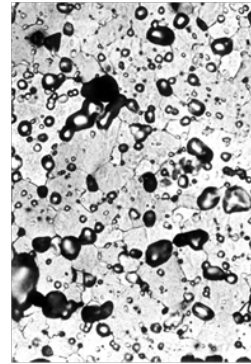
Metallography of U-Pu-Zr-5MA-5RE.



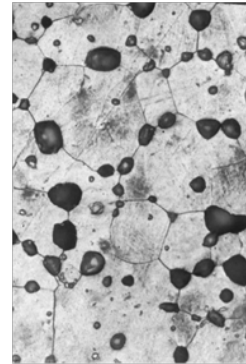
500°C



600°C



700°C



50mm  
850°C

Am & RE-rich precipitates

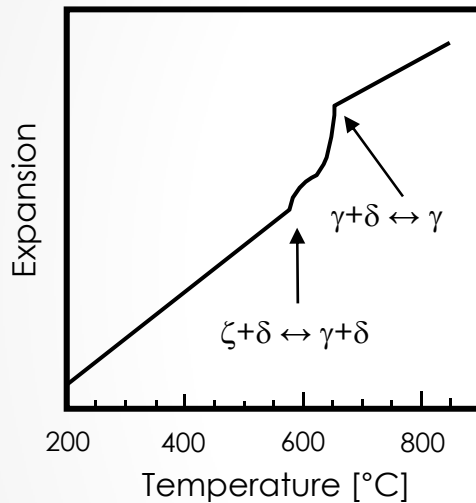
- Uniformly dispersing
- Cohesion at grain boundary (≥700°C)
- ~3μm (-2MA-2RE),
- ≥10μm (-5MA-5RE)



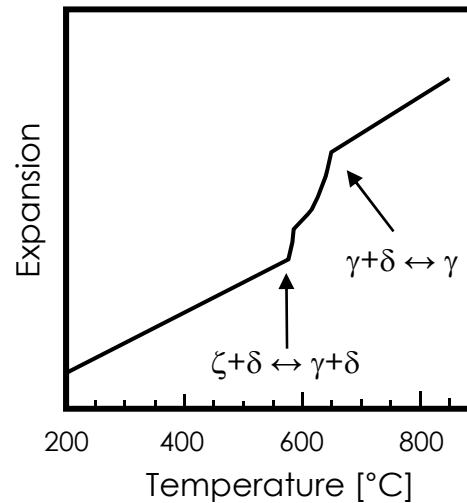
# Phase transition temperature

Phase transition temperature of U-Pu-Zr(-MA-RE) were measured by dilatometry method.

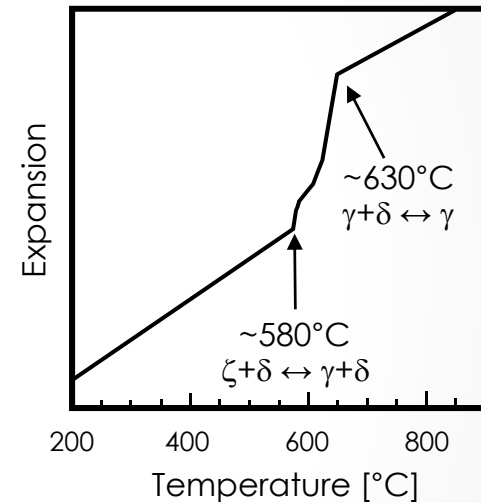
Dilatometric curves



(a) U-Pu-Zr-2MA-2RE



(b) U-Pu-Zr-5MA-5RE



(c) U-Pu-Zr

For all samples,

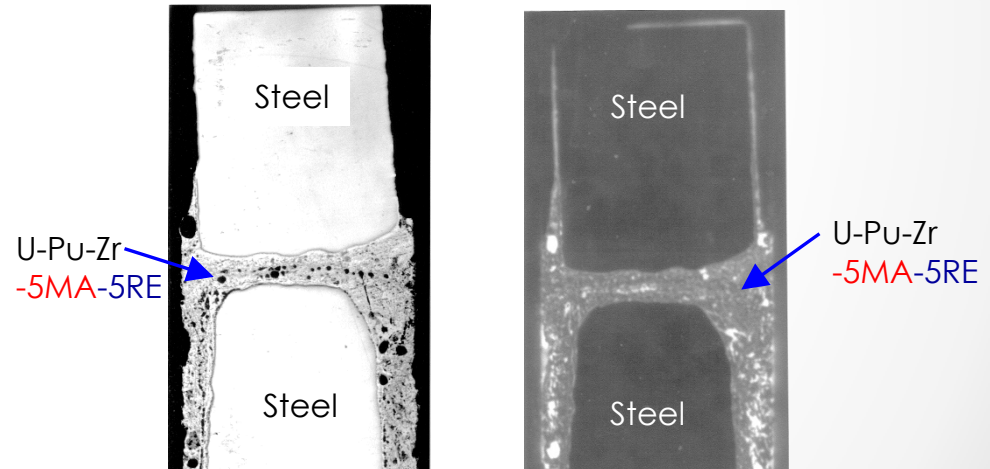
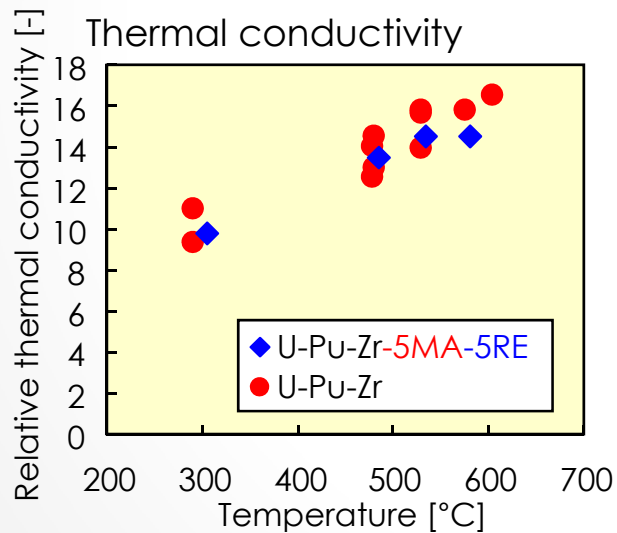
two distinctive phase transition temperatures at  $\sim 580^\circ\text{C}$  &  $\sim 630^\circ\text{C}$

→ Insignificant influence of MA and RE addition up to 5wt%

# Other properties

	U-19Pu-10Zr -5MA-5RE	U-19Pu-10Zr	Reported U-19Pu-10Zr
Melting point [°C]	1207±10	1217±10	1214±75 [2]
Elasticity			
Young's modulus [GPa]	93.31	85.22	
Shear modulus [GPa]	35.39	32.65	
Poisson's ratio	0.32	0.31	
Compatibility with SS * [1]	920-960	970-990	

\*: Metallurgical reaction temperature between the alloy and stainless steel.



Influence of MA and RE addition  $\leq 5\text{wt}\%$  is not significant.

# Fabrication of MA-containing Metal Fuel

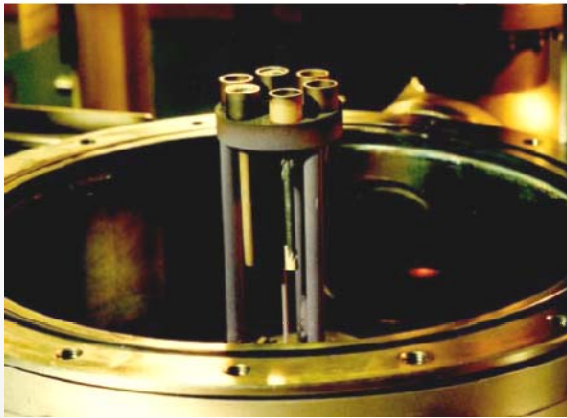
## Fuel Fabrication:

U-19Pu-10Zr-2MA-2RE, U-19Pu-10Zr-5MA,  
U-19Pu-10Zr-5MA-5RE and U-19Pu-10Zr

MA=60Np-30Am-10Cm, RE=10Y-10Ce-70Nd-10Gd.

Fuel Rod diameter	4.9 mm
Fuel Rod lengths	20-50 mm
Density	
U-19Pu-10Zr-2MA-2RE	14.73 g/cm <sup>3</sup>
U-19Pu-10Zr-5MA-5RE	14.66 g/cm <sup>3</sup>
U-19Pu-10Zr-5MA	15.31 g/cm <sup>3</sup>
U-19Pu-10Zr	15.77 g/cm <sup>3</sup> *

\*: Reported value =15.8g/cm<sup>3</sup>,  
J.H.Kittel, et al., N.E.D. 15 (1971)



Ytria molds used for fuel rod casting



Cast fuel rods

# Irradiation Experiment

MA-containing alloys were irradiated in Phénix.

3 metal fuel pins & 16 oxide fuel pins  
were arranged in an capsule.

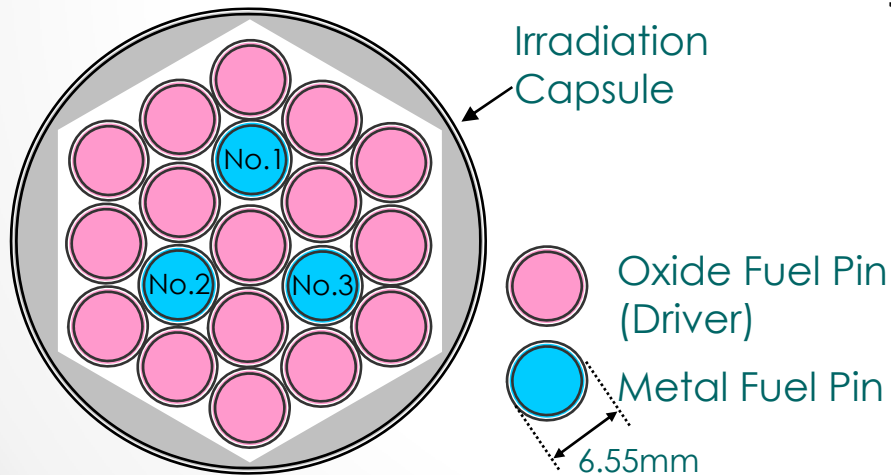
Pin No.1 : U-19Pu-10Zr

Pin No.2 : U-19Pu-10Zr-2MA-2RE

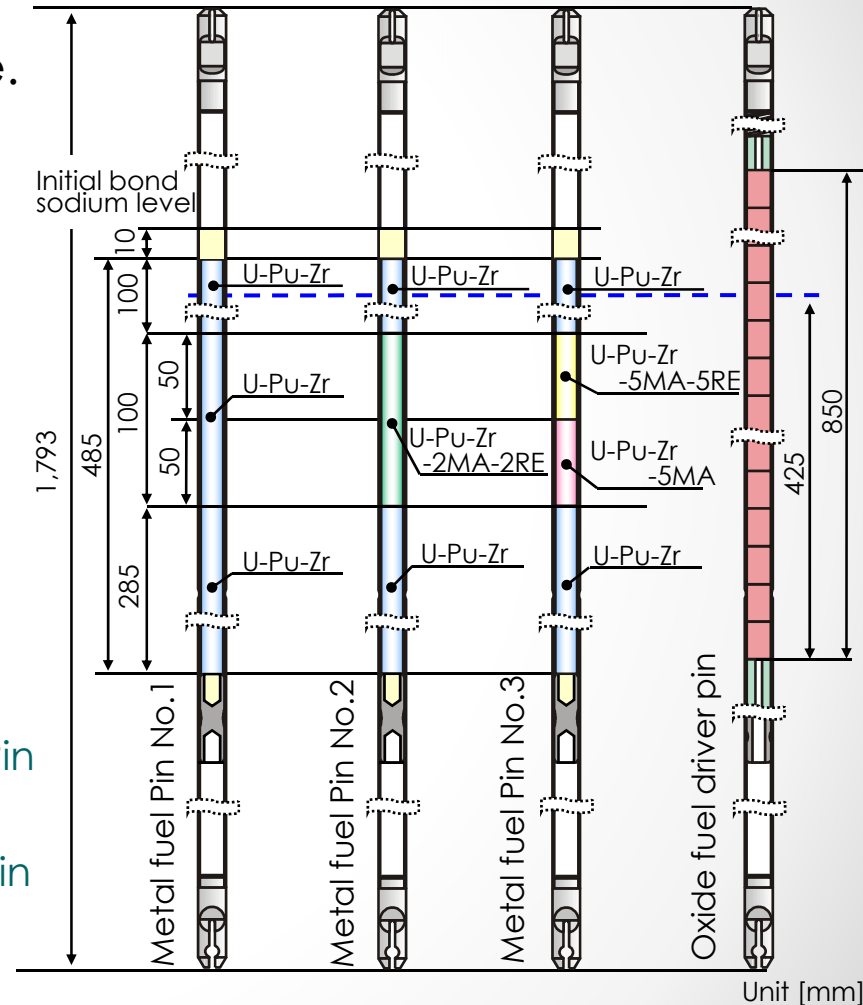
Pin No.3 : U-19Pu-10Zr-5MA / -5MA-5RE

Cladding material : 15-15Ti

Burnup goals ~2.5at.% (METAPHIX-1),  
~7at.% (METAPHIX-2),  
~10at.% (METAPHIX-3).

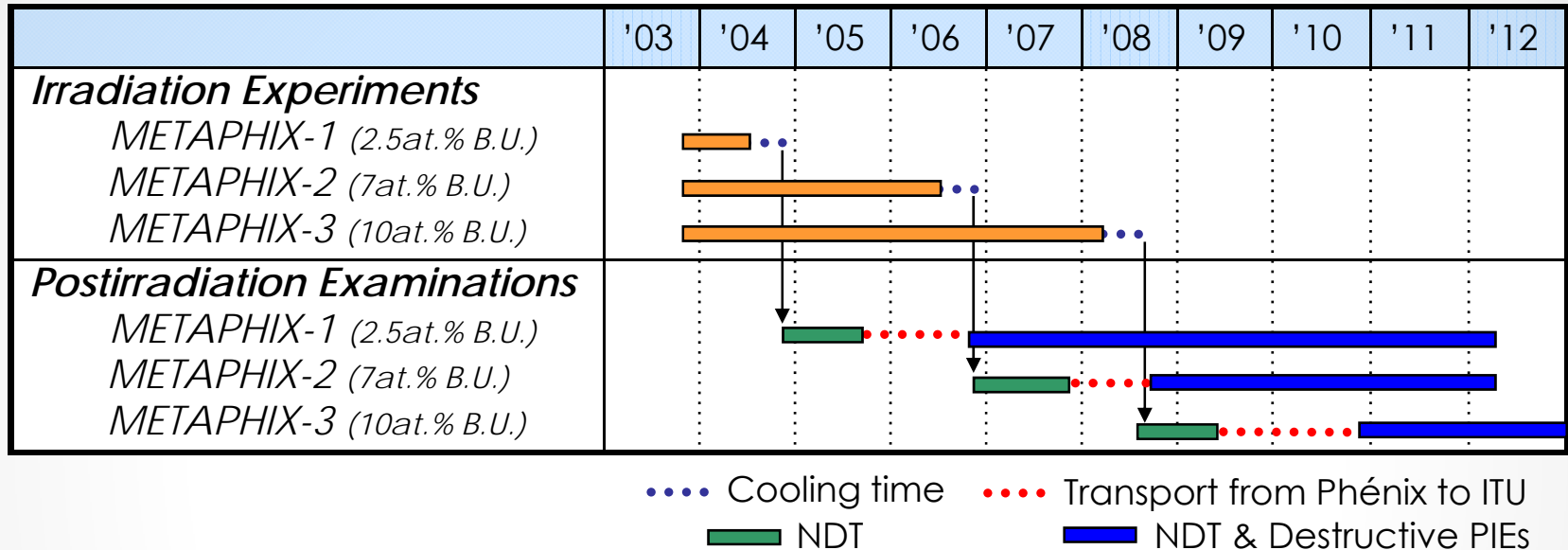


Fuel pin arrangement in irradiation capsule.



Schematic views of irradiated fuel pins.

# METAPHIX Program



- Irradiation experiments were carried out from Dec. 2003 to May 2008 in Phénix.
- After cooling, NDT were carried out.  
*No excessive damage due to neutron irradiation was observed.*
- Irradiated fuel pins are transported to ITU for nondestructive & destructive PIE.
- After the PIE, pyro-reprocessing experiment is planned.

# Irradiation Conditions

Irradiation parameters were analyzed taking account of the operation diagram of the Phénix reactor.

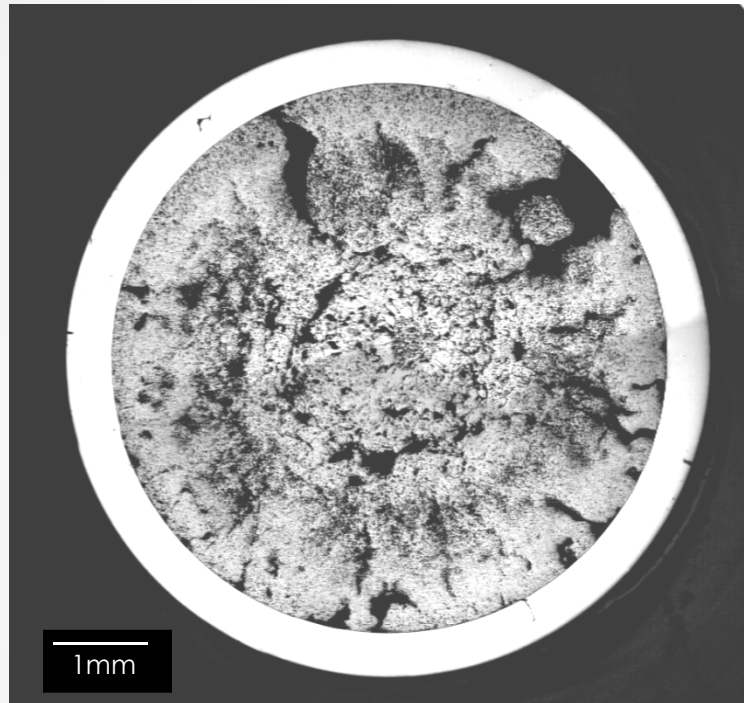
Projected Irradiation Conditions for METAPHIX Experiment

	<i>Pin No.1</i> <i>U-19Pu-10Zr</i>	<i>Pin No.2</i> <i>U-19Pu-10Zr</i> <i>+2MA+2RE</i>	<i>Pin No.3(lower)</i> <i>U-19Pu-10Zr</i> <i>+5MA</i>	<i>Pin No.3(upper)</i> <i>U-19Pu-10Zr</i> <i>+5MA+5RE</i>
<b><i>Begin of Irradiation</i></b>				
<i>Max. Linear Power</i> <sup>1</sup> [W/cm]	350	327	343	332
<i>Max. Cladding Temp.</i> <sup>2</sup> [°C]	581	581	581	←
<b><i>End of METAPHIX-1 (120EFPD</i><sup>3</sup>)</b>				
<i>Max. Linear Power</i> <sup>1</sup> [W/cm]	330	308	325	313
<i>Max. Cladding Temp.</i> <sup>2</sup> [°C]	572	572	572	←
<i>Max. Burnup</i> [at.%]	2.4	2.5	2.4	2.6
<b><i>End of METAPHIX-2 (360EFPD</i><sup>3</sup>)</b>				
<i>Max. Linear Power</i> <sup>1</sup> [W/cm]	295	276	294	282
<i>Max. Cladding Temp.</i> <sup>2</sup> [°C]	556	556	556	←
<i>Max. Burnup</i> [at.%]	6.9	7.1	7.0	7.5
<b><i>End of METAPHIX-3 (900EFPD</i><sup>3</sup>)</b>				
<i>Max. Linear Power</i> <sup>1</sup> [W/cm]	268	251	269	256
<i>Max. Cladding Temp.</i> <sup>2</sup> [°C]	543	543	543	←
<i>Max. Burnup</i> [at.%]	10.9	11.2	11.2	11.9

<sup>1</sup>: Top of the test alloy, <sup>2</sup>: Top of the fuel stack, <sup>3</sup>: EFPD=Effective Full Power Days.

# Metallography (1)

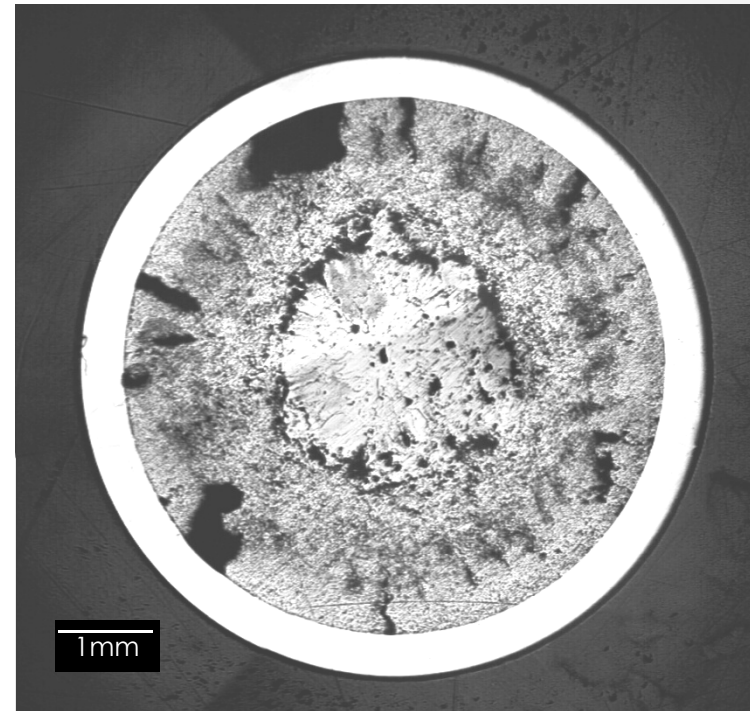
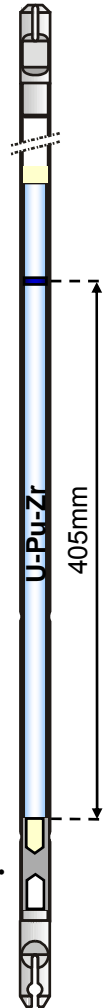
METAPHIX-1, U-19Pu-10Zr



(a)

Three concentric regions are formed.

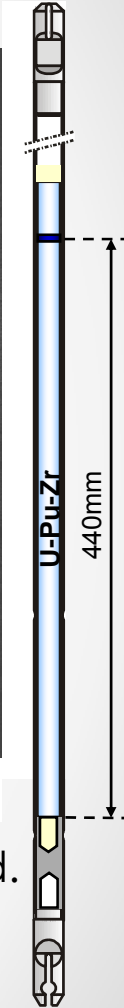
$\gamma \leftrightarrow \gamma + \zeta \leftrightarrow \zeta + \delta$   
(center)  $\longleftrightarrow$  (periphery)



(b)

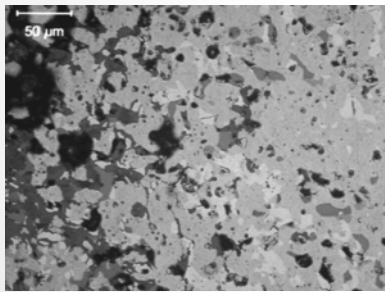
Two concentric regions are formed.

( $\gamma$ -phase is not observed.)  
→ Irradiation temperature  $< 630^\circ\text{C}$

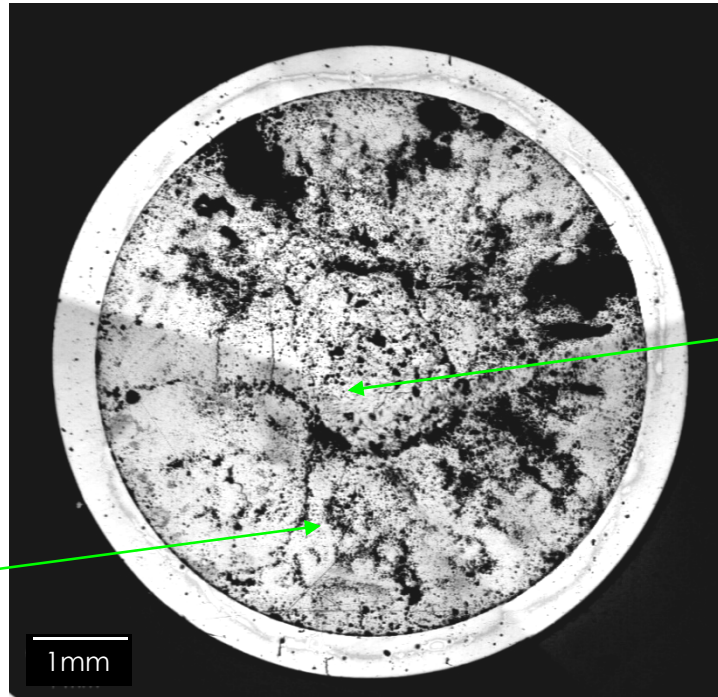


# Metallography (2)

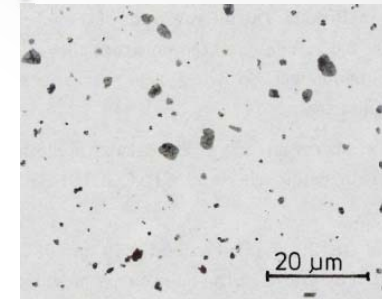
METAPHIX-1, U-19Pu-10Zr-2MA-2RE



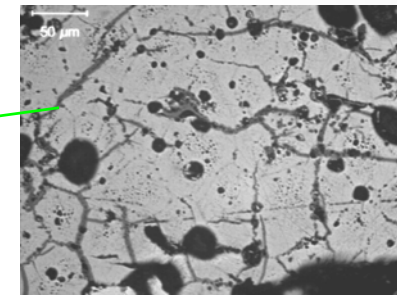
Intermediate Zone  
100μm



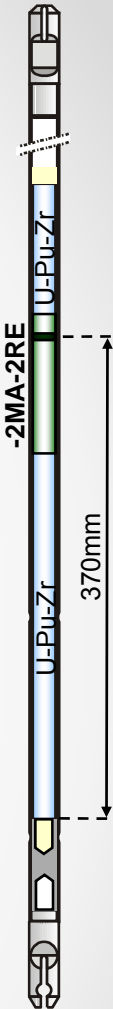
Cross-Sectional Overview



Unirradiated U-Pu-Zr-2MA-2RE



Central Zone 100μm  
Dense phase  
→ (γ+ζ)-phases

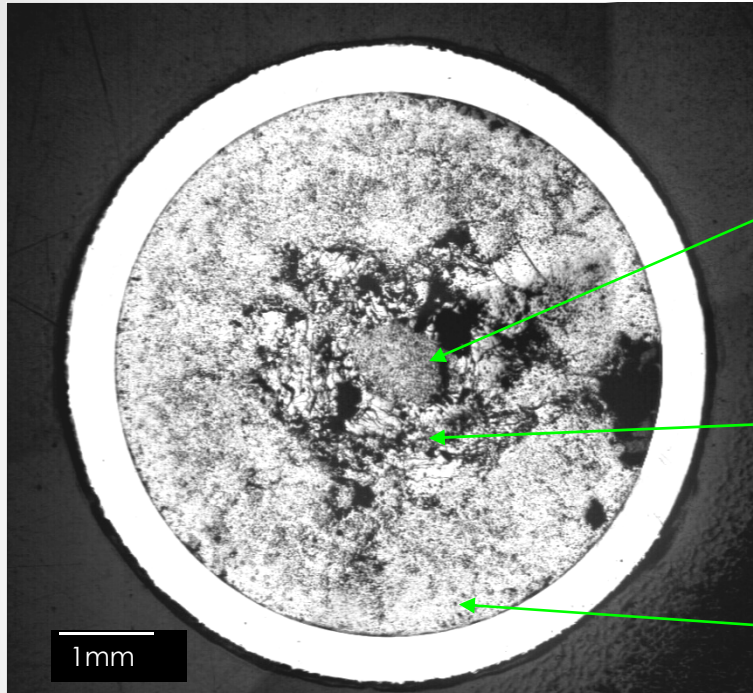


Matrix morphology is similar to that of U-Pu-Zr fuel (b).  
Some narrow layered phases (MA-RE inclusions) spread along grain boundaries in  $\gamma+\zeta$  zone.  
In low-temperature region, some dark spots (MA and RE inclusions) are visible.

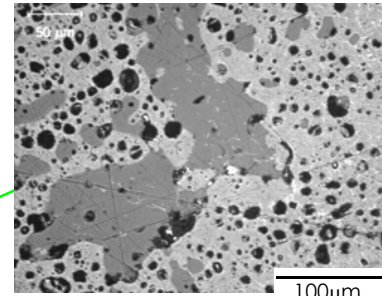


# Metallography (3)

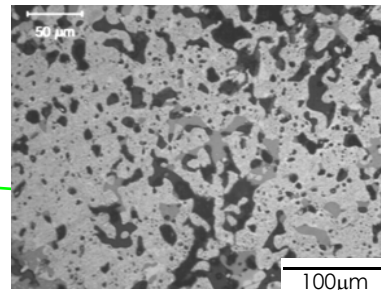
METAPHIX-1, U-19Pu-10Zr-5MA-5RE



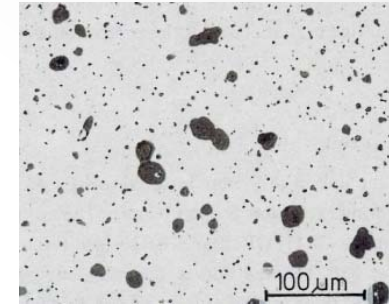
Cross-Sectional Overview



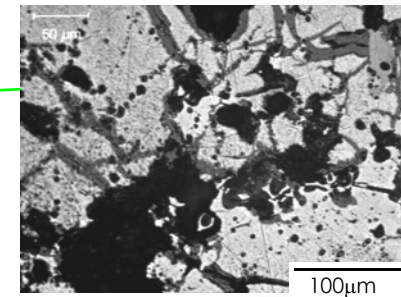
Central Zone  
Porous phase  
→  $\gamma$ -phase



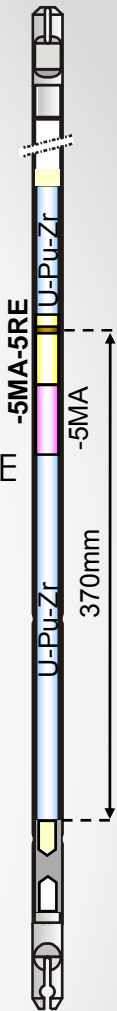
Peripheral Zone



Unirradiated U-Pu-Zr-5MA-5RE



Intermediate Zone  
Dense phase  
→  $(\gamma+\zeta)$ -phases



Matrix morphology is similar to that of U-Pu-Zr fuel (a).  
Large precipitates (MA and RE inclusions) appear in  $\gamma$  phase zone.  
Some narrow layered phases (MA-RE inclusions) spread along grain boundaries in  $\gamma+\zeta$  zone.  
In low-temperature region, small dark spots (MA and RE inclusions) are observed.

# Characteristics of Irradiated MA-Containing Metal Fuel

1. The radial distribution of fuel matrix morphology is similar to that of U-Pu-Zr ternary fuels.
2. Some large precipitates (MA and RE inclusions) appear in the high-temperature phase.
3. In the dense matrix zone, some narrow layered phases (MA and RE inclusions) spread along grain boundaries.
4. In low-temperature region, some dark spots (MA and RE inclusions) are visible.

# Summary

Mass flow of Pu and MA was analyzed for future LWR-FBR transition scenario.

MA content in the FBR fuel was estimated to be 2wt%.

With using 5wt% MA content fuel, MAs recycling from LWRs can be accelerated for several decades.

Relevant characteristics of U-Pu-Zr-MA-RE were examined.

In the case of  $\leq 5\text{wt}\%$  MA and  $\leq 5\text{wt}\%$  RE additions,

- Am-RE-rich precipitates are dispersed almost uniformly in the alloy,
- Basic properties are practically unchanged.

MA-containing U-Pu-Zr alloys were irradiated in Phénix.

- o Compositions: U-19Pu-10Zr, U-19Pu-10Zr-2MA-2RE, U-19Pu-10Zr-5MA(-5RE)
- o Peak burnups:  $\sim 2.5\text{at}\%$ ,  $\sim 7\text{at}\%$  and  $\sim 10\text{at}\%$ .

NDT of the METAPHIX-1, -2 & -3 pins

- No critical damage had occurred during irradiation.

Metallography of METAPHIX-1

- Matrix structure is similar to that of U-Pu-Zr fuels.
- Large precipitates appear in  $\gamma$ -phase zone.
- Some layered phase spread along grain boundaries in  $\gamma+\zeta$  phase region.

Quantitative analyses are being carried out.

- Fuel constituent redistribution,
- MA transmutation performance

*Thank you for your attention!!*

# Compositions of Metal Fuel Alloys

4 types of metal fuel alloy were prepared.

Average Compositions of Fabricated Metal Fuel Alloys [wt%]

Target	71U-19Pu-10Zr	67U-19Pu-10Zr +2MA+2RE	66U-19Pu-10Zr +5MA	61U-19Pu-10Zr +5MA+5RE
U	71.00	66.85	66.30	63.50
Pu	18.93	19.80	19.35	19.75
Zr	10.19	9.46	8.97	8.19
MA	0.03	2.08	4.74	4.78
<i>Np</i>	-	1.23	2.97	3.04
<i>Am</i>	0.03	0.67	1.45	1.52
<i>Cm</i>	-	0.18	0.32	0.31
RE	-	1.73	-	3.40
Y	-	0.12	-	0.31
Ce	-	0.20	-	0.45
Nd	-	1.25	-	2.30
Gd	-	0.16	-	0.32

Impurities < 0.3wt%

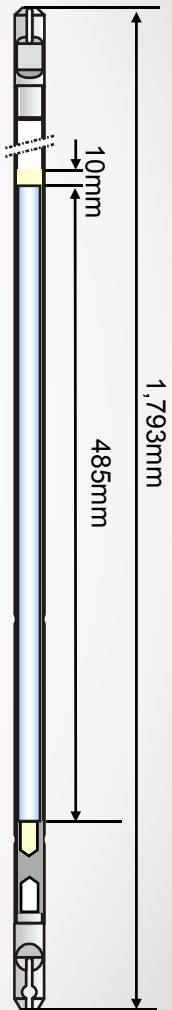
# Specifications of Metal Fuel Pins

Fuel pins were manufactured according to Phénix geometry.

Fuel Pin Specifications in this Irradiation Experiments

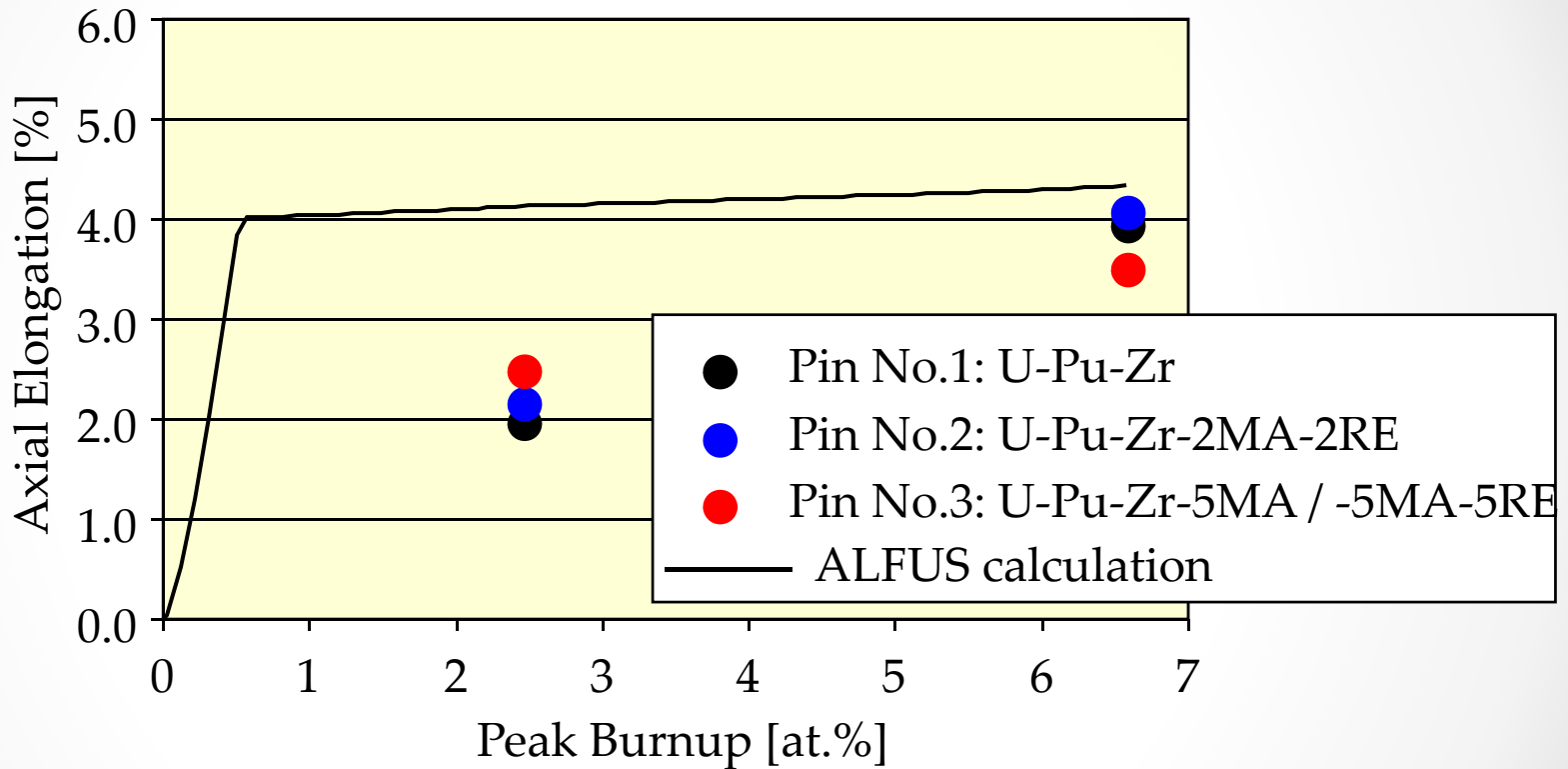
<i>Pin length [mm]</i>	1,793
<i>Outer cladding diameter [mm]</i>	6.55
<i>Cladding material</i>	15-15 Ti
<i>Fuel length [mm]</i>	485
<i>Fuel diameter [mm]</i>	4.9
<i>Initial fuel-cladding gap [mm]</i>	0.375
<i>Fuel smear density [%]</i>	75.2
<i>Sodium level above fuel* [mm]</i>	~ 10
<i>Plenum length [mm]</i>	464

\* : Sodium is filled into fuel-cladding gap as thermal bonding.



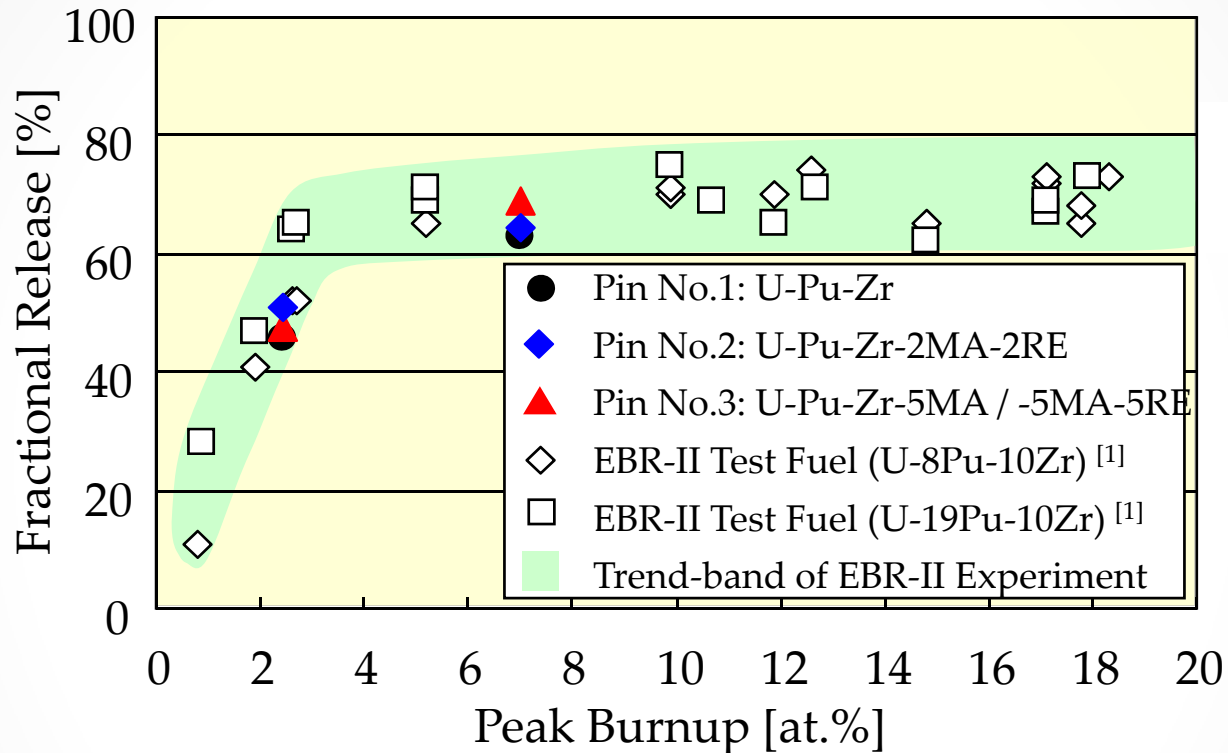
# Axial Swelling of Fuel alloy

Fuel stack position was estimated by axial gamma-ray distribution from  $^{106}\text{Ru}$ .



Fuel elongation behavior is independent of MA and RE additions.  
Axial swelling of METAPHIX fuels is within the range of the prediction.

# Fission Gas Release



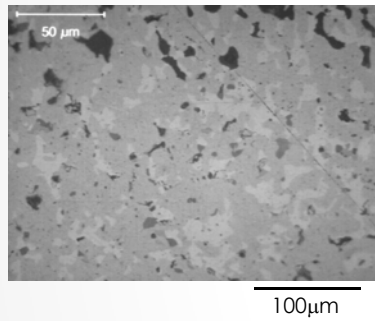
FP gas release fraction of MA & RE-containing fuel pins is the same level as that of U-Pu-Zr alloy fuel pins, and consistent with EBR-II ternary test fuel data.

[1]: R. G. Pahl, et al., *Proc. Int. Fast Reactor Safety Meeting*, Session 2 Vol. IV, 129 (1990).

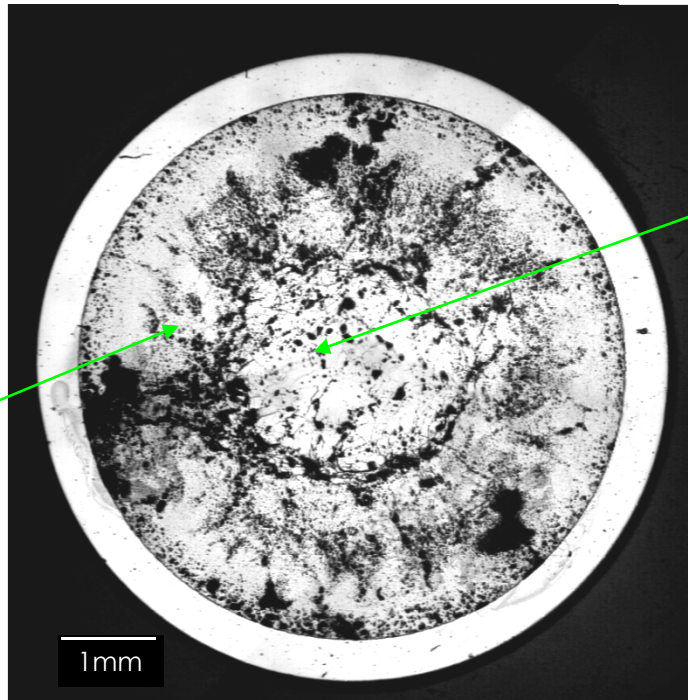


# Metallography (3')

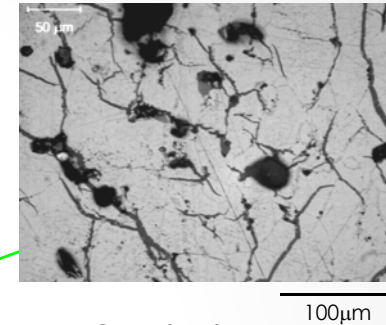
METAPHIX-1, U-19Pu-10Zr-5MA



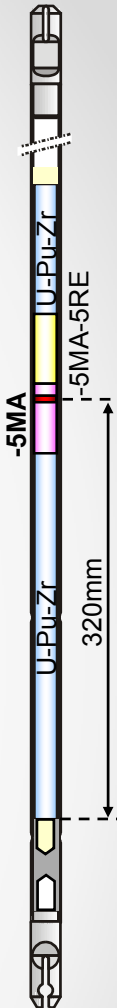
Intermediate Zone



Cross-Sectional Overview



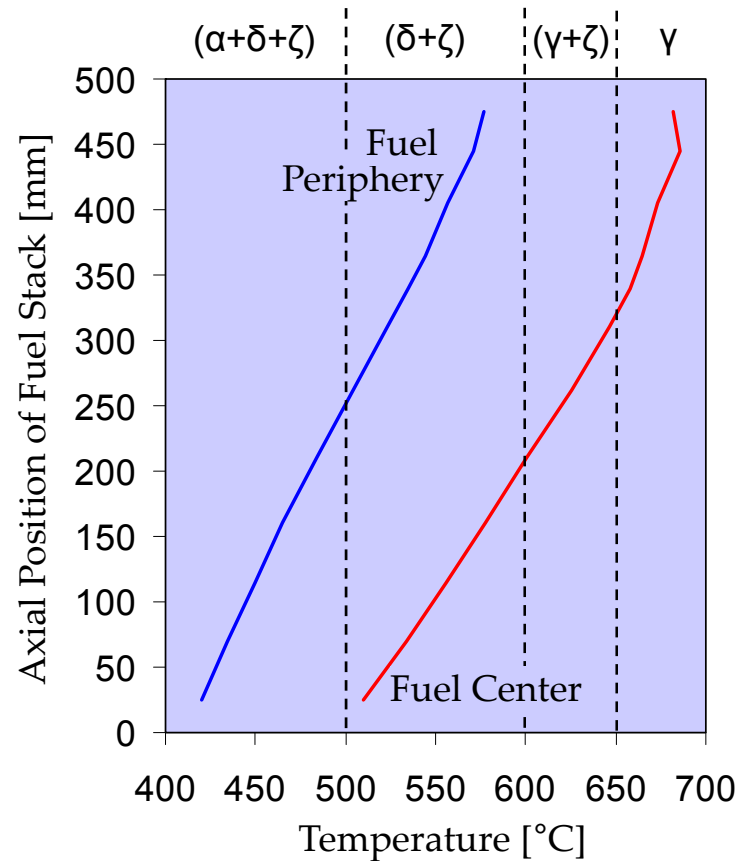
Central Zone  
Dense phase  
→ (γ+ζ)-phases



Matrix morphology is similar to that of U-Pu-Zr fuel (b).  
Some narrow layered phases (MA-RE inclusions) spread along grain boundaries in  $\gamma+\zeta$  zone.  
In low-temperature region, small dark spots (MA and RE inclusions) are dispersed.

# Irradiation Behavior Analysis

## -Fuel Temperature Distribution-



420°C < Temp. < 685°C

Fig. Evaluated irradiation temperature for METAPHIX-1 fuel pin at EOI (Pin No.1: U-19Pu-10Zr).

# Discussion - Irradiation Temperature -

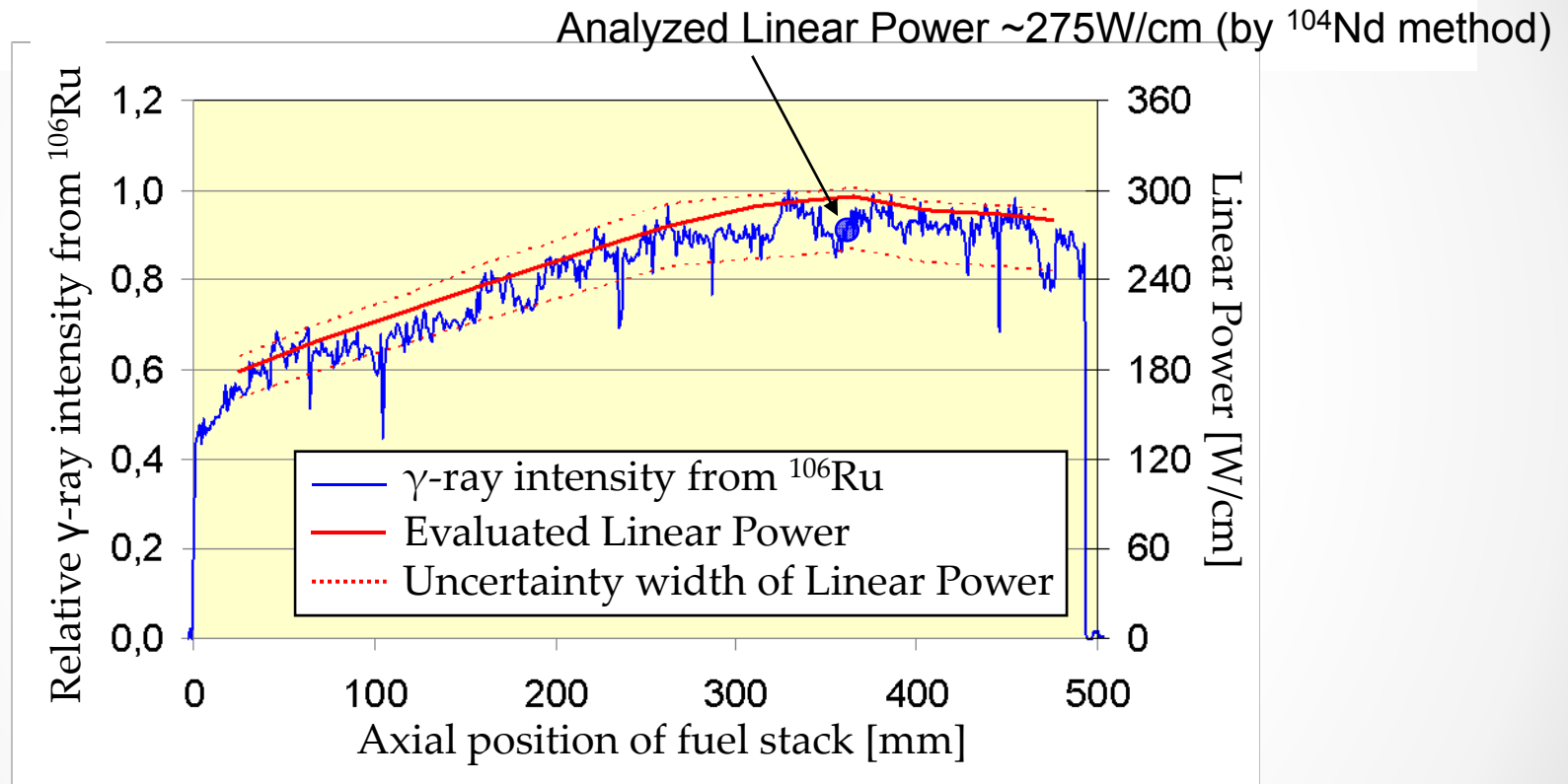
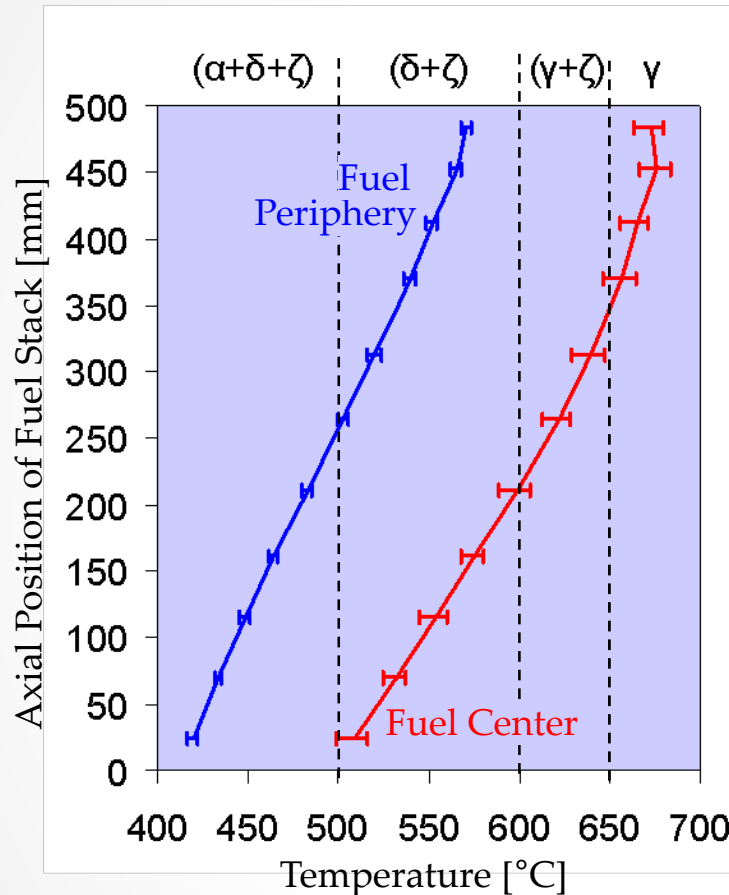


Fig. Relative  $\gamma$ -ray intensity emitted from  $^{106}\text{Ru}$  and axial power profile of Pin No. 1

# Discussion - Irradiation Temperature -



Due to the uncertainty of linear power,

-Temperature fluctuation for each fuel rod reaches  $\sim 20^{\circ}\text{C}$  at the fuel center,  
→Irradiation temperature at higher axial level

can be lower than that at lower level,

-The highest temperature can be  $\sim 660^{\circ}\text{C}$ .  
→High-temperature  $\gamma$ -phase appears at only limited fuel rods.

Fig. Evaluated irradiation temperature for METAPHIX-1 fuel pins, taking account of the uncertainties of linear power.