
Development of Minor Actinide- Containing Metal Fuels

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Background

MA-Containing Metal Fuel Development

- Minor actinides (MA) burnup technology using FR metal fuel cycle is being developed in CRIEPI.
 - Metal fuel FR have some advantages for MA burnup.
 - MA content in recycled fuel is < 1wt% : Metal fuel FR cycle.
 - ~ 2wt% : MA from LWR spent fuels are recycled.
 - > 2wt% : MA from HLLW are recycled.
 - In pyro-processing, MA is recovered with rare-earth (RE) fission products.
MA : RE ~ 1 : 1, depending on decontamination specifications.
- Experimental studies on U-Pu-Zr-MA(-RE) alloys have been conducted in cooperation with JRC-ITU.
 - Characterization of U-Pu-Zr-MA-RE alloys,
 - Fuel fabrication for irradiation experiment,
 - Irradiation in fast reactor Phénix,
 - Non-destructive and destructive postirradiation examinations.

Miscibilities among U-Pu-Zr-MA-RE

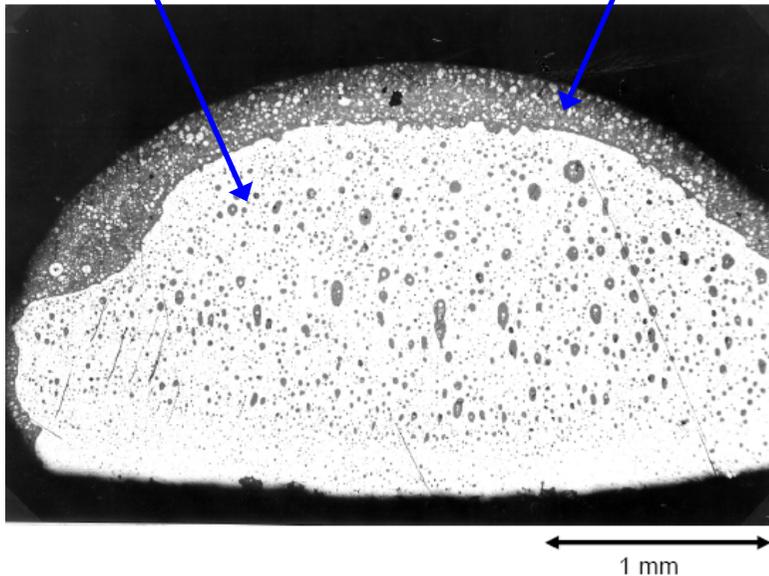
U-Pu-Zr-MA-RE alloys of different compositions were mixed by arc-melting.

Metallography of CR10 alloy:

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

U-Pu-Zr-Np phase

Pu-Am-RE phase



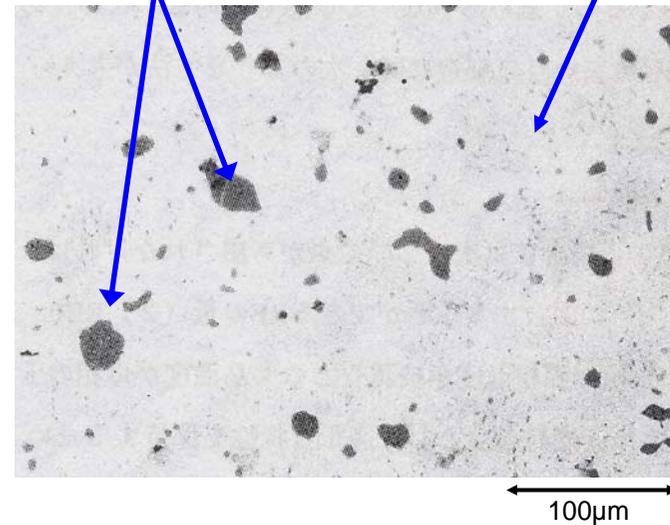
In the alloys of high RE content,
→ Matrix segregates into upper
and lower parts.

Metallography of CR101/3 alloy:

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

Pu-Am-RE precipitates

U-Pu-Zr-Np phase



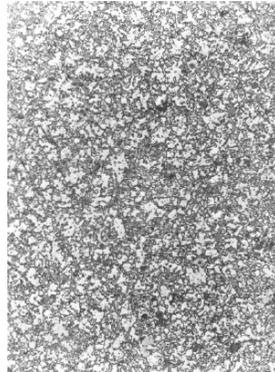
In the alloys of low RE content ($\leq 5\%$),
→ RE-rich precipitates ($\leq 30\mu\text{m}$) were
uniformly dispersed.

RE content should be reduced to $\leq 5\%$.

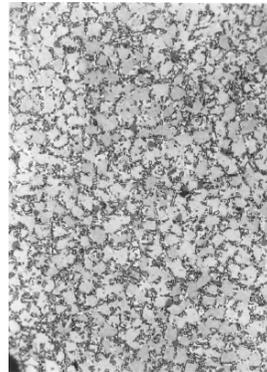
Phase Structures of annealed U-Pu-Zr-MA-RE

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

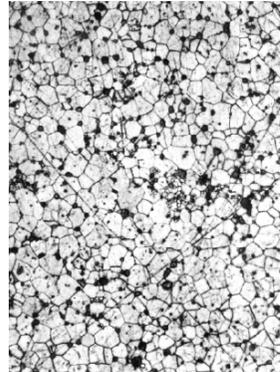
Metallography of CR11: U-Pu-Zr-2MA-2RE.



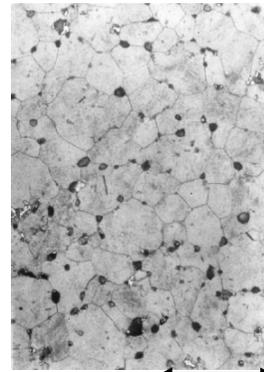
500°C



600°C



700°C

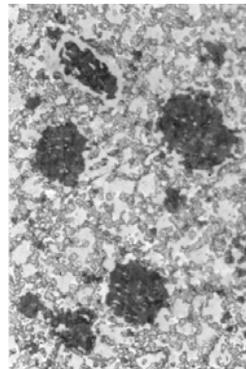


850°C

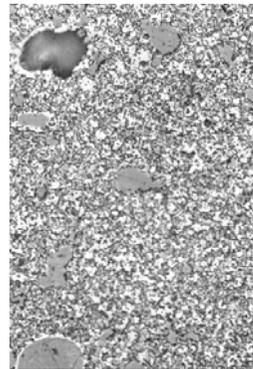
Matrix phase

- $\leq 600^\circ\text{C}$: Two phase structures
 $\zeta+\delta$ at 500°C
 $\gamma+\delta$ (or $\zeta+\delta$) at 600°C
- $\geq 700^\circ\text{C}$: Single γ -phase

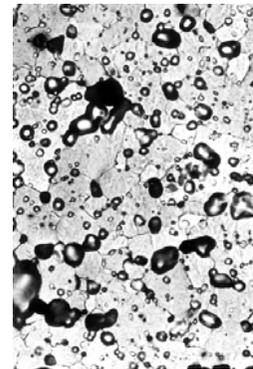
Metallography of CR12: U-Pu-Zr-5MA-5RE.



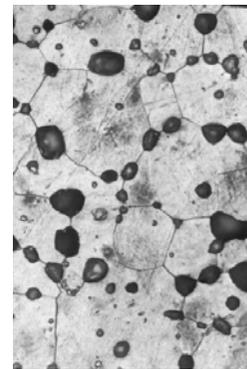
500°C



600°C



700°C



850°C

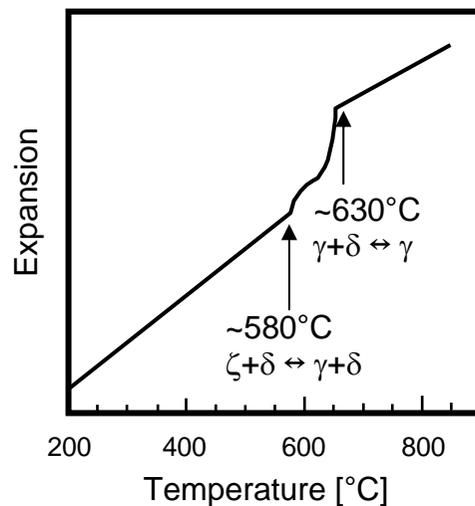
Am & RE-rich precipitations

- Uniformly dispersing
- Cohesion at grain boundary ($\geq 700^\circ\text{C}$)
- $\sim 3\mu\text{m}$ (CR11), $\geq 10\mu\text{m}$ (CR12)

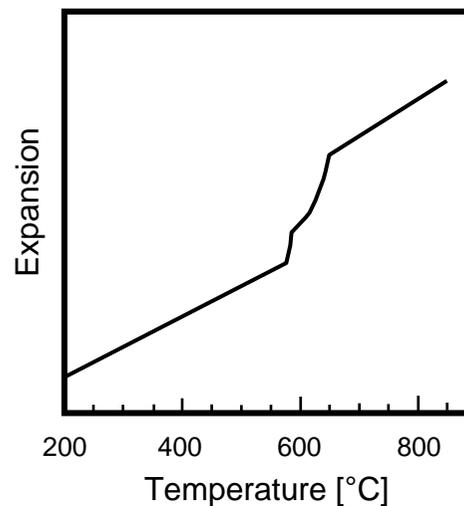
Phase transition temperature

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

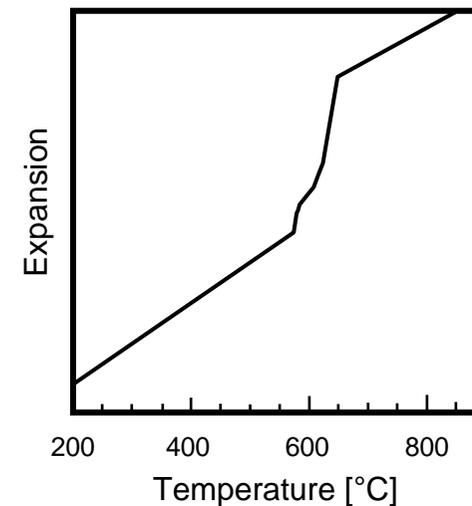
Dilatometric curves



(a) CR11: U-Pu-Zr-2MA-2RE



(b) CR12: U-Pu-Zr-5MA-5RE



(c) CR13: U-Pu-Zr

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

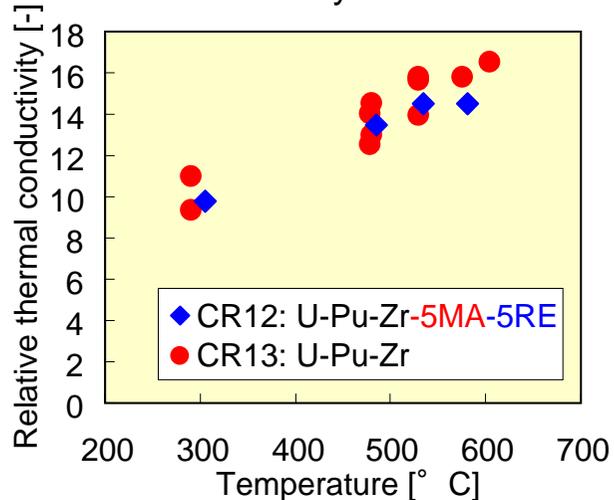
→ Insignificant influence of MA and RE addition up to 5wt%

Other properties

	U-19Pu-10Zr -2MA-2RE	U-19Pu-10Zr -5MA	U-19Pu-10Zr -5MA-5RE	U-19Pu-10Zr	Reported U-19Pu-10Zr
Density [g/cm ³]	14.73	15.31	14.66	15.77	15.8 [2]
Melting point [°C]			1207±10	1217±10	1214±75 [3]
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.

Thermal conductivity



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

[1] C. Sari, et al, J. Nucl. Mater., 208 (1994) 201.

[2] J. H. Kittel, et al., *Nuclear Engineering and Design* 15, 373-440 (1971).

[3] M.C. Billon, et al., Int. Conf. on Reliable Fuels for Liquid Metal Reactors, Tucson, Arizona, Sep. 7-11 (1986).

Irradiation Experiment

MA-containing alloys were irradiated in Phénix.

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

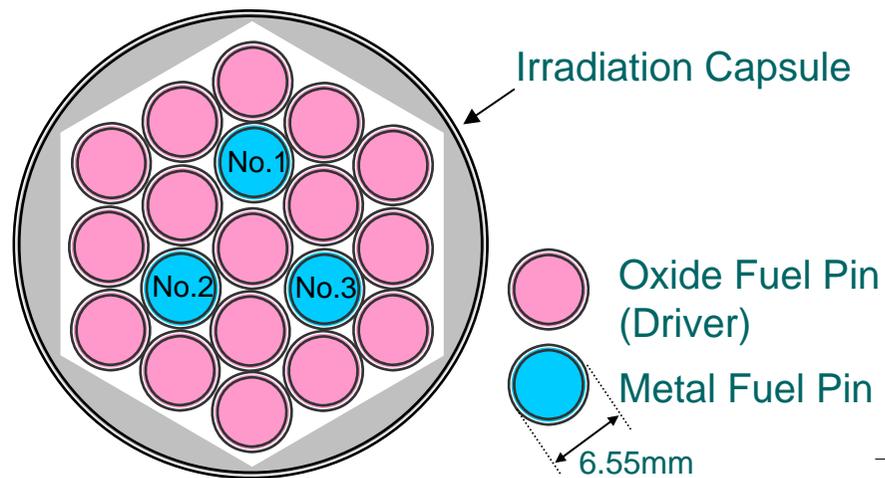
Pin No.1 : U-19Pu-10Zr

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

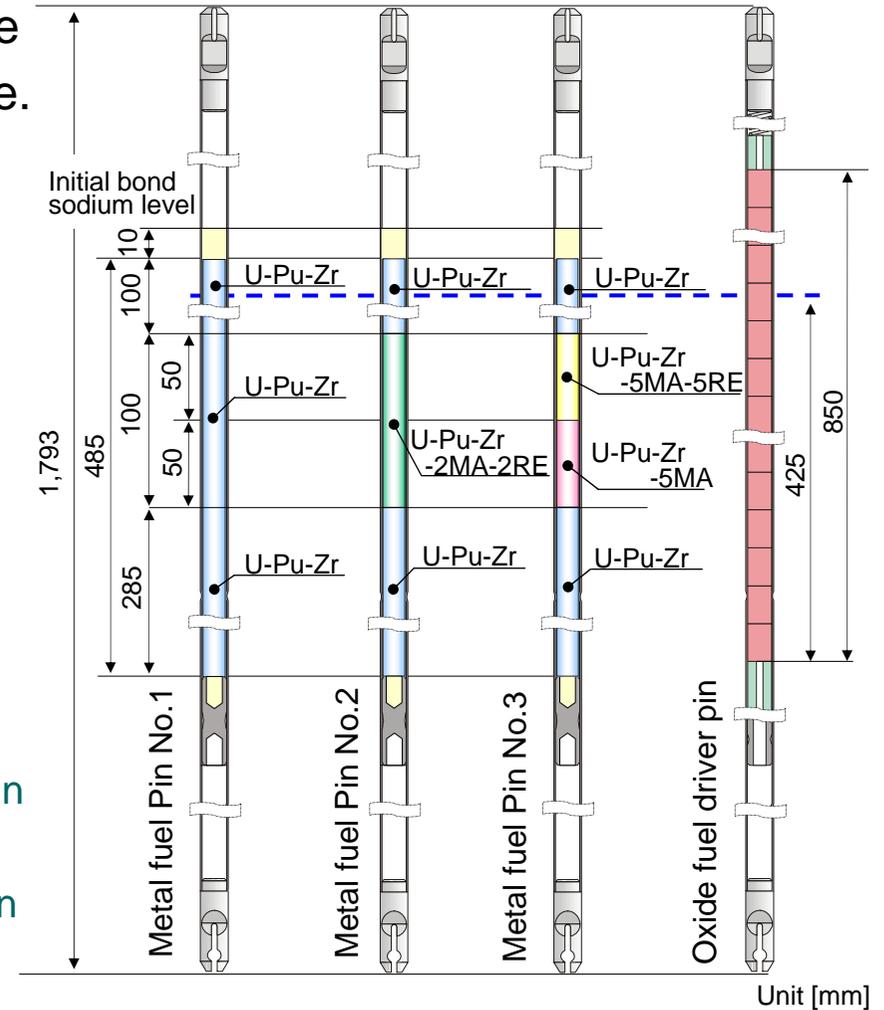
Pin No.3 : U-19Pu-10Zr-5MA / -5MA-5RE

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

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



Fuel pin arrangement in irradiation capsule.



Schematic views of irradiated fuel pins.

Irradiation Conditions

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

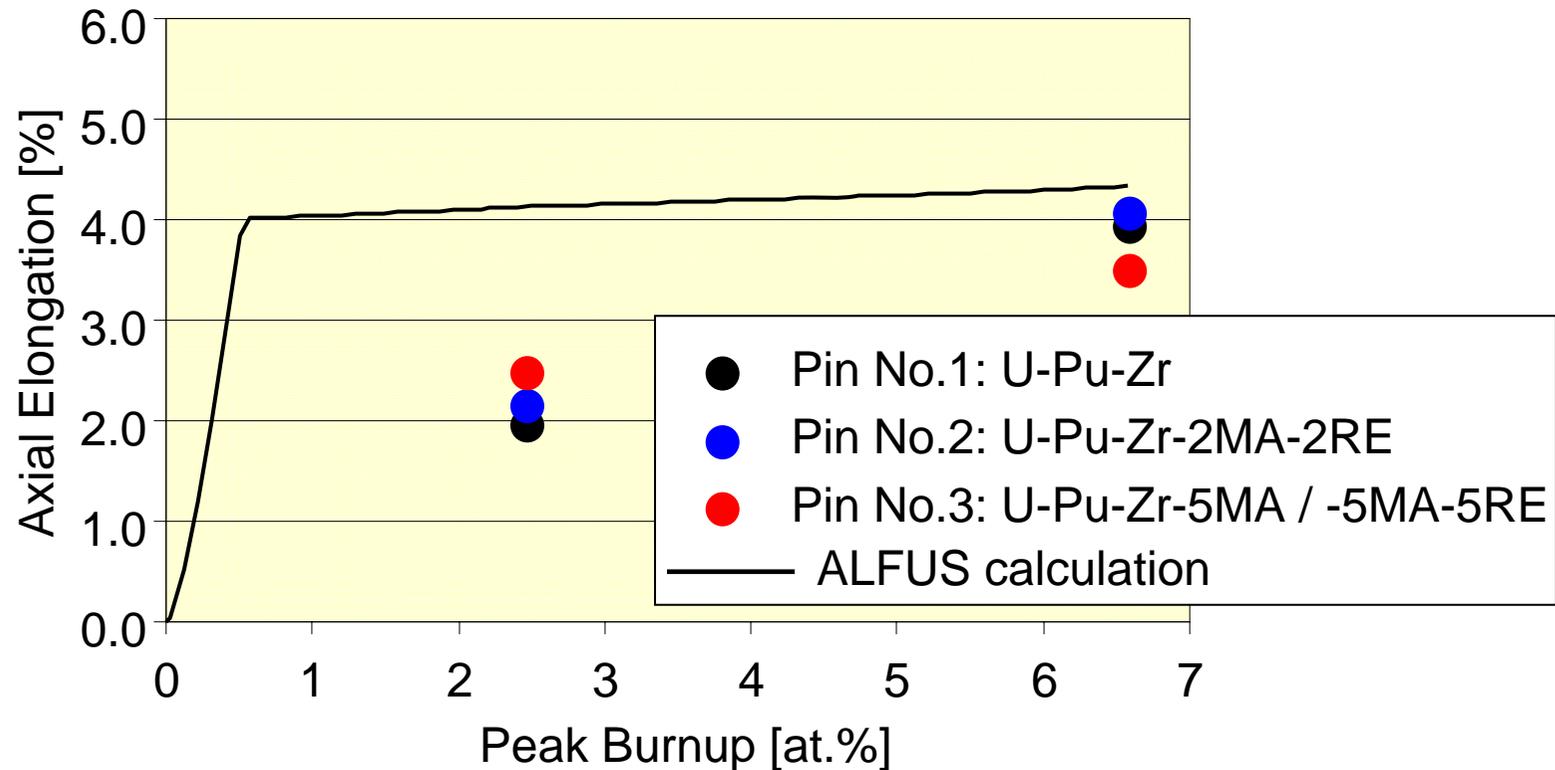
Projected Irradiation Conditions for METAPHIX Experiment

	<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>
Begin of Irradiation <i>Max. Linear Power</i> ¹ [W/cm] <i>Max. Cladding Temp.</i> ² [°C]	350 581	327 581	343 581	332 ←
End of METAPHIX-1 (120EFPD ³) <i>Max. Linear Power</i> ¹ [W/cm] <i>Max. Cladding Temp.</i> ² [°C] <i>Max. Burnup</i> [at.%]	330 572 2.4	308 572 2.5	325 572 2.4	313 ← 2.6
End of METAPHIX-2 (360EFPD ³) <i>Max. Linear Power</i> ¹ [W/cm] <i>Max. Cladding Temp.</i> ² [°C] <i>Max. Burnup</i> [at.%]	295 556 6.9	276 556 7.1	294 556 7.0	282 ← 7.5
End of METAPHIX-3 (900EFPD ³) <i>Max. Linear Power</i> ¹ [W/cm] <i>Max. Cladding Temp.</i> ² [°C] <i>Max. Burnup</i> [at.%]	268 543 10.9	251 543 11.2	269 543 11.2	256 ← 11.9

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

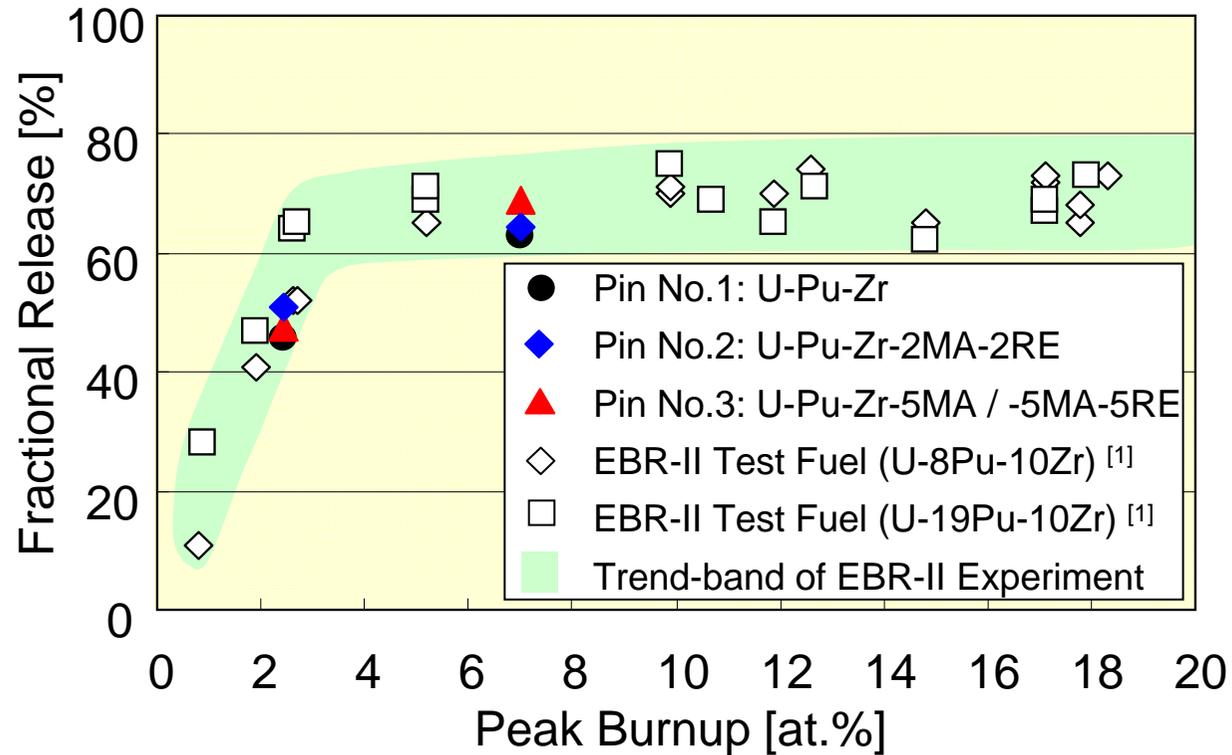
Axial Swelling of Fuel alloy

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



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

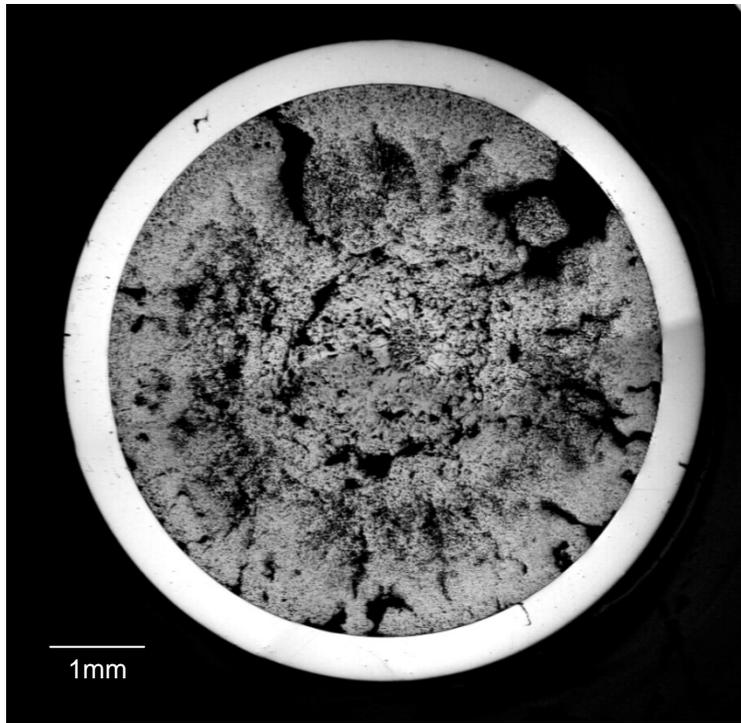
Fission Gas Release



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

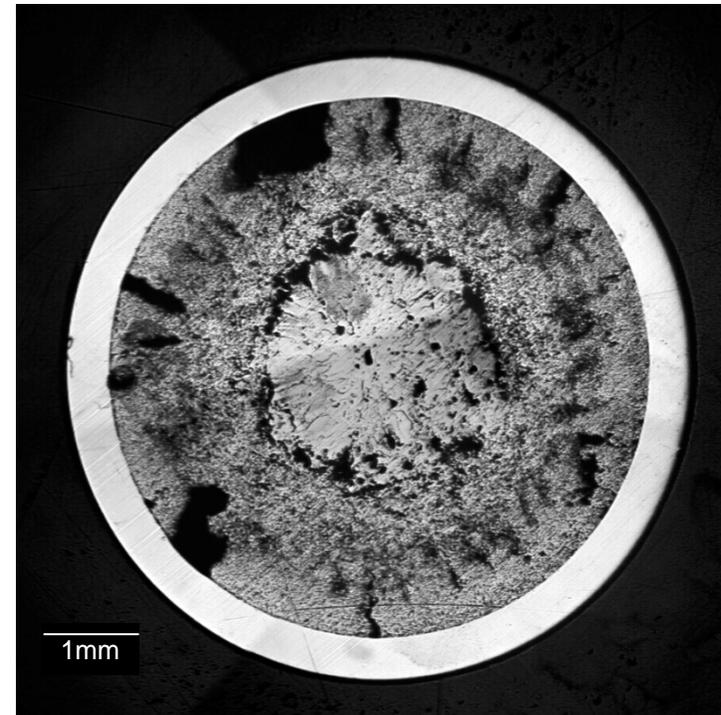
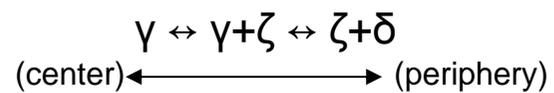
[1] : R. G. Pahl, et al., *Proc. Int. Fast Reactor Safety Meeting*, Session 2 Vol. IV, 129 (1990).

Metallography (1) Sample #1 & #2: U-19Pu-10Zr



(a) Sample #1

Three concentric regions are formed.



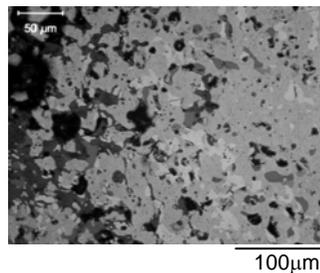
(b) Sample #2

Two concentric regions are formed.

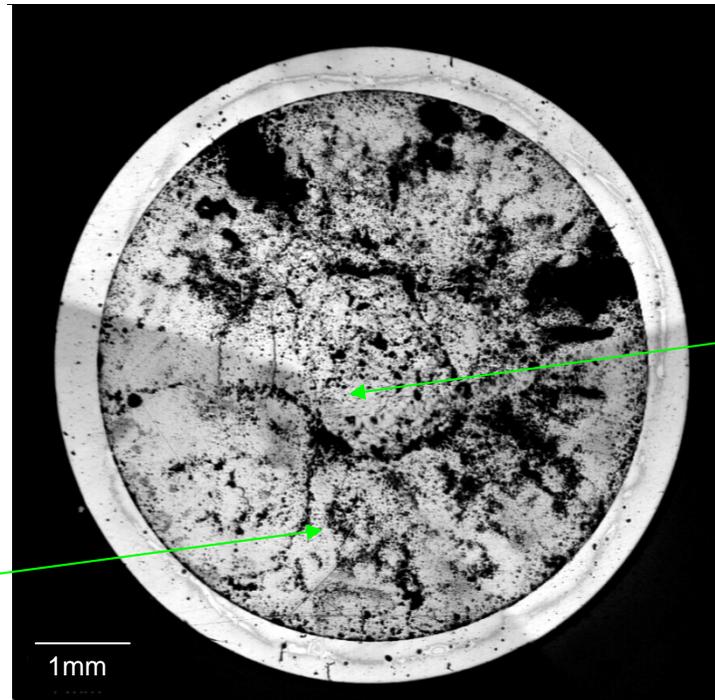
(γ -phase is not observed.)

→ Irradiation temperature < 650°C

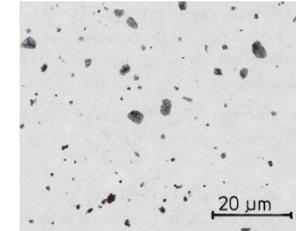
Metallography (2) Sample #3: U-19Pu-10Zr-2MA-2RE



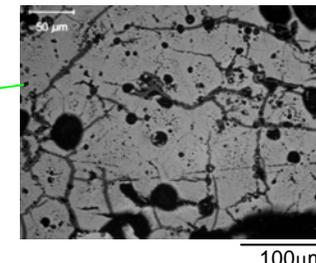
Intermediate Zone



Cross-Sectional Overview



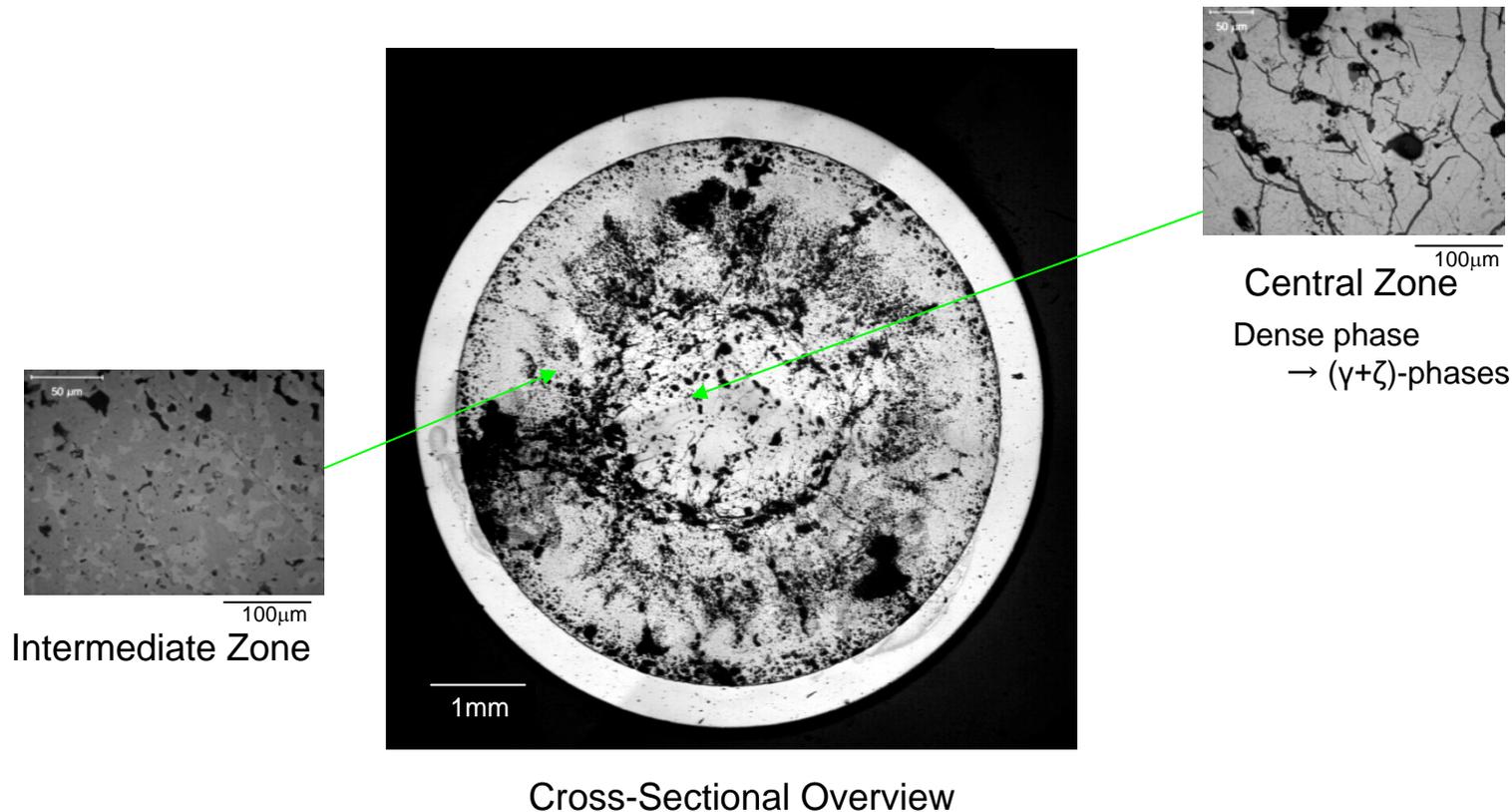
Unirradiated U-Pu-Zr-2MA-2RE



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

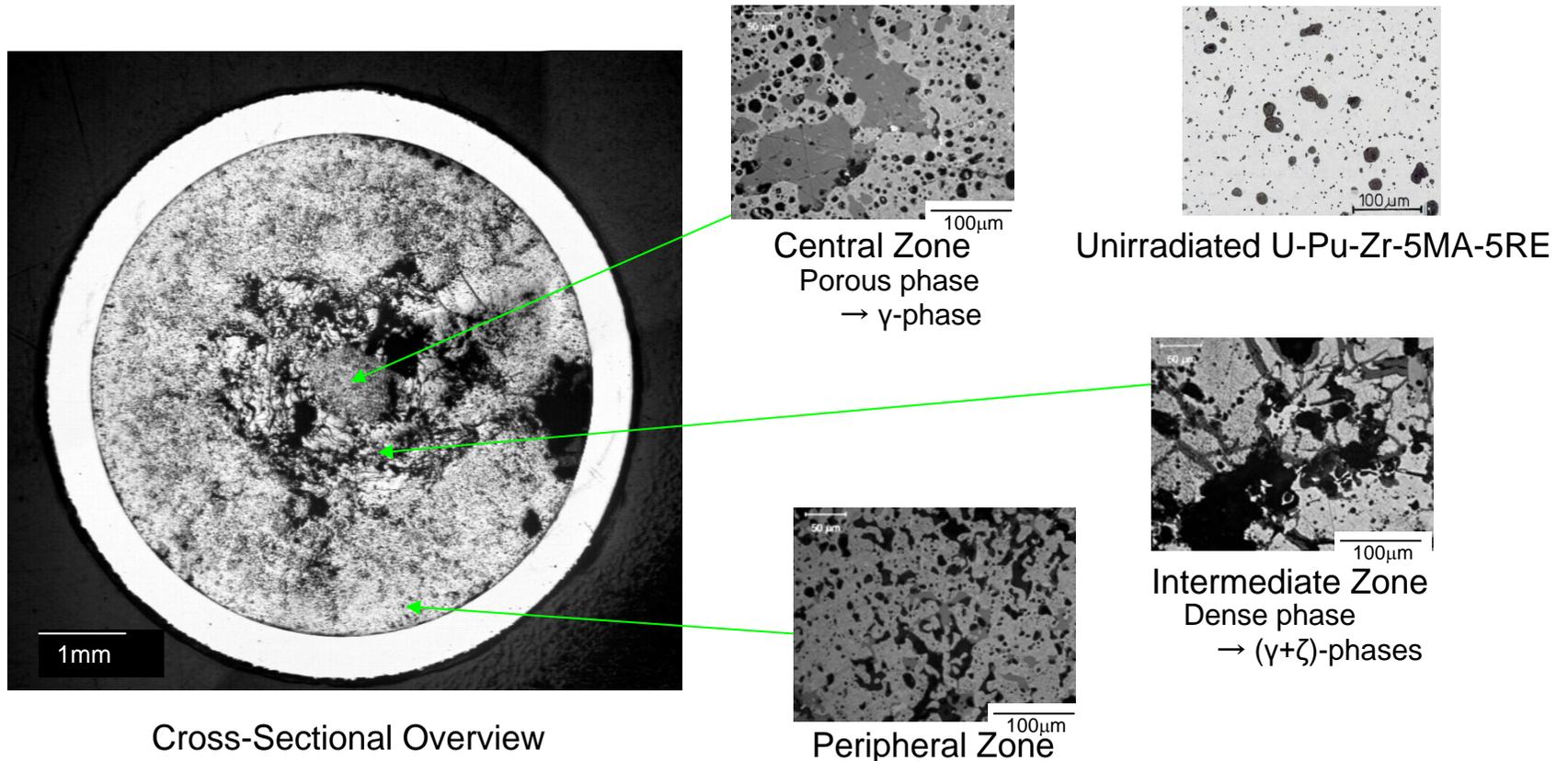
Matrix morphology is similar to that of U-Pu-Zr fuel (Sample #2).
Some narrow layered phases (MA-RE inclusions) spread along grain boundaries in γ+ζ zone.
In low-temperature regions, some dark spots (MA and RE inclusions) are visible.

Metallography (3) Sample #4: U-19Pu-10Zr-5MA



Matrix morphology is similar to that of U-Pu-Zr fuel (Sample #2).
Some narrow layered phases (MA-RE inclusions) spread along grain boundaries in $\gamma+\zeta$ zone.
In low-temperature regions, small dark spots (MA and RE inclusions) are observed.

Metallography (4) Sample #5: U-19Pu-10Zr-5MA-5RE



Matrix morphology is similar to that of U-Pu-Zr fuel (Sample #1).

Large precipitations (MA and RE inclusions) appear in γ phase zone.

Some narrow layered phases (MA-RE inclusions) spread along grain boundaries in $\gamma+\zeta$ zone.

In low-temperature regions, 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 precipitations (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 regions, some dark spots (MA and RE inclusions) are visible.

Summary

Relevant characteristics of U-Pu-Zr alloys containing MA (and 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 (and RE)-containing U-Pu-Zr alloys were irradiated in Phénix.

- Fuel compositions: U-19Pu-10Zr, U-19Pu-10Zr-2MA-2RE, U-19Pu-10Zr-5MA, U-19Pu-10Zr-5MA-5RE.
- Peak burnups: $\sim 2.5\text{at.}\%$ (METAPHIX-1), $\sim 7\text{at.}\%$ (METAPHIX-2), $\sim 10\text{at.}\%$ (METAPHIX-3).

PIE on METAPHIX-1 & -2 started.

- NDT of the pins
 - No damage had occurred during irradiation.
- Gamma-ray spectrometry & Plenum gas analysis
 - Axial swelling and FP gas release are essentially the same as those of U-Pu-Zr fuels.
- Metallography
 - Radial distribution of matrix morphology is similar to that of U-Pu-Zr fuels.
 - Some large precipitates (MA and RE inclusions) appear in γ -phase zone.
 - In $\gamma+\zeta$ phase region, some layered phase (MA and RE inclusions) spread along grain boundaries.
 - In low-temperature region, some dark spots (MA and RE inclusions) are dispersed.

Quantitative analyses are being carried out.

Thank you for your attention!!

Compositions of Metal Fuel Alloys

4 types of metal fuel alloy were prepared.

TABLE. 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%

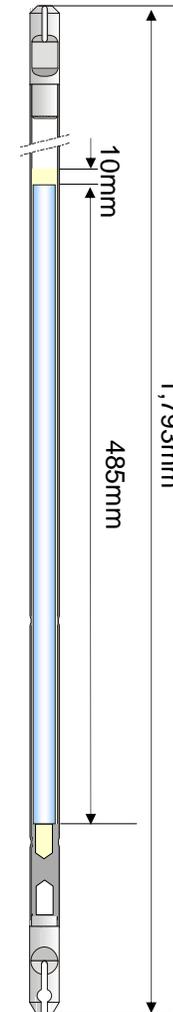
Specifications of Metal Fuel Pins

Fuel pins were manufactured according to Phénix geometry.

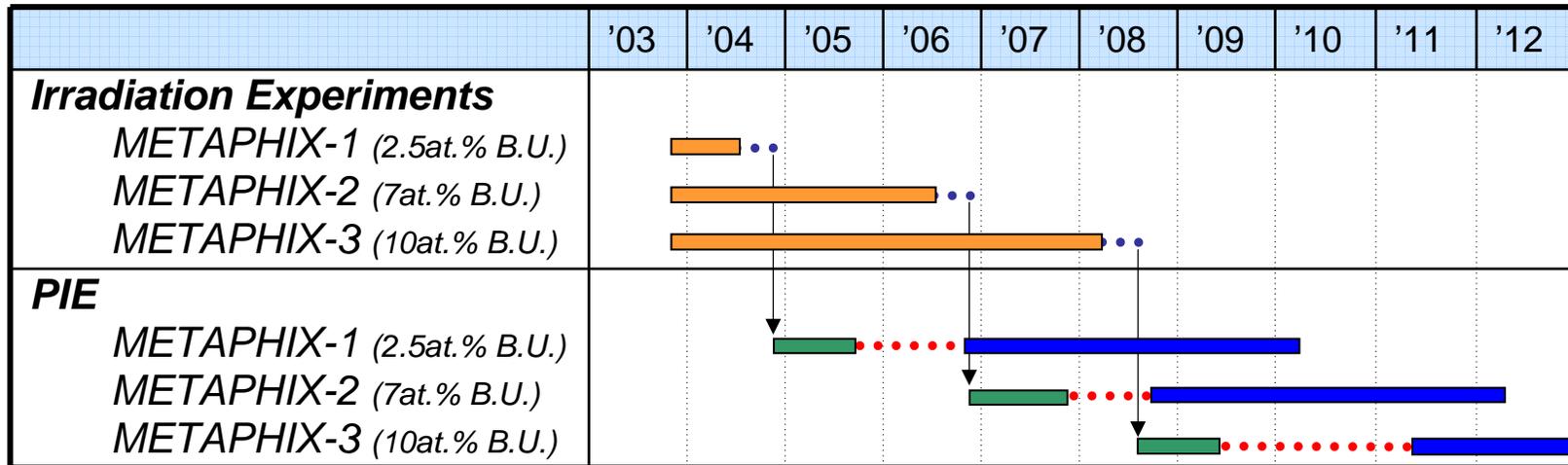
TABLE. Fuel Pin Specifications in this Irradiation Experiments

<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.



METAPHIX Program



●●● Cooling time ●●● Transport from PHÉNIX to ITU
 ■ Nondestructive tests ■ Nondestructive & Destructive PIEs

- Irradiation experiments were conducted from Dec. 2003 to May 2008 at Phénix.
- After cooling, NDT was 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, pyrometallurgical reprocessing experiment is planned.