Minor Actinide Transmutation Performance in Fast Reactor Metal Fuel

Isotope Ratio Change in Actinide Elements upon Low-Burnup Irradiation

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Background
– MA-Containing Metal Fuel Development –

- MA burnup technology using FR metal fuel cycle is being developed in CRIEPI.
  - Metal fuel FRs have some advantages for MA burnup.
    High heavy metal density, Hard neutron spectrum
  - MA content in recycled fuel < 1wt% : Metal fuel FR cycle.
    ~ 2wt% : MA from LWR spent fuels is added.
    > 2wt% : MA from HLLW is added.
    The most efficient burnup of MA ~5wt%

- In pyrometallurgical processing, MA are recovered with some amounts of RE fission products.
  - *MA : RE ~ 1 : 1*, depending on the separation process specifications.

- Experimental studies on *U-Pu-Zr-MA-RE* alloys have been conducted in CRIEPI and ITU.
  In the case of U-Pu-Zr alloys containing ≤ *5wt%* MA and ≤ *5wt%* RE,
  - homogeneous alloys can be fabricated,
  - no significant change in mechanical and thermal properties is found.

Irradiation Experiment
– METAPHIX –

Purposes: To investigate the effects of MA and RE addition on the irradiation behavior,
To confirm MA transmutation performance in FR metal fuels.

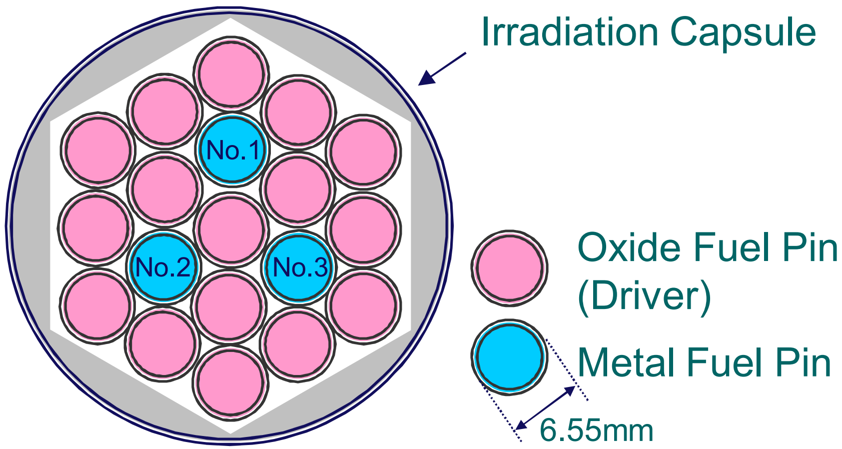
3 metal fuel pins were arranged in an irradiation capsule

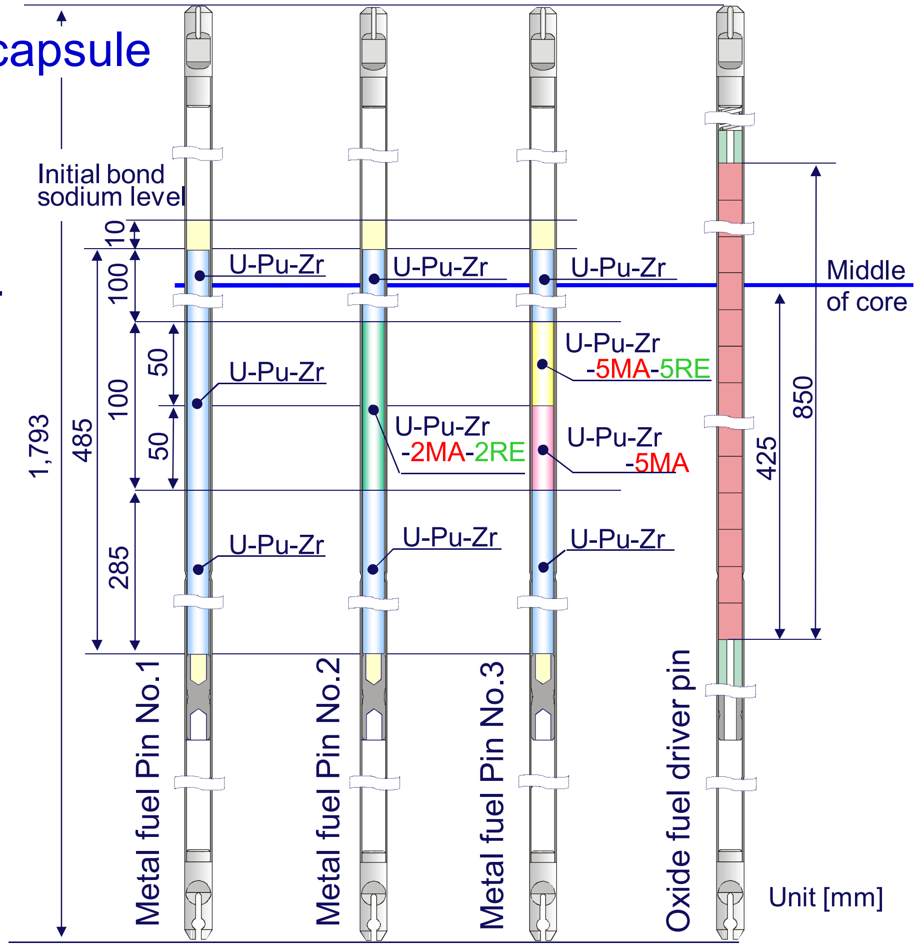
Pin No.1 : U-19Pu-10Zr (without MA and RE)

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

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

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





Target burnups: ~2.5at.% (METAPHIX-1),
~ 7at.% (METAPHIX-2),
~ 10at.% (METAPHIX-3).

Burnup Evaluation

- Postirradiation compositions of METAPHIX-1 fuel alloys were analyses by ICP-MS.
- Discharged burnups of U-19Pu-10Zr, U-19Pu-10Zr-2MA-2RE, U-19Pu-10Zr-5MA were evaluated.

$$\text{Burnup [FIMA, at.\%]} = \frac{N_{BI}/Y_{BI}}{\sum N_i + N_{BI}/Y_{BI}} \times 100$$

N_i: *Atom number density of actinide nuclide, i,*
N_{BI}: *Atom number density of burnup indicator,* Y_{BI}: *Fission yield of burnup indicator*

- For RE-containing alloy, a conventional ^{148}Nd method is not applicable.
→ ^{139}La, ^{102}Ru and ^{104}Ru were selected as alternative burnup indicators.

Sample		U-19Pu-10Zr	U-19Pu-10Zr-2MA-2RE	U-19Pu-10Zr-5MA
Irradiation position †1		~360mm	~350mm	~330mm
Burnup indicator	^{148}Nd	2.11	-	2.30
	^{139}La	2.18	2.52	2.50
	^{102}Ru	2.19	2.40	2.32
	^{104}Ru	2.14	2.51	2.27
Prediction †2		2.10-2.52	2.15-2.58	2.12-2.55

†1: Axial position from the bottom of the fuel stack. †2: Based on the irradiation condition.

Burnup evaluations using ^{148}Nd, ^{139}La, ^{102}Ru and ^{104}Ru correspond well with each other, and are within the range of predictions based on the irradiation condition.

Initial Composition of Irradiation Fuels

	U-19Pu-10Zr	U-19Pu-10Zr-2MA-2RE	U-19Pu-10Zr-5MA	U-19Pu-10Zr-5MA-5RE
	Average 14 samples*	Average 2 samples**	Average 2 samples**	Average 2 samples**
U	70.43 $^{+2.60}_{-5.46}$	66.85±1.55	66.30±1.30	63.50±2.50
Pu	18.85 $^{+1.02}_{-0.74}$	19.80±0.20	19.35±0.05	19.75±0.55
Np	-	1.29±0.09	2.97±0.02	3.04±0.16
Am	0.03 $^{+0.11}_{-0.02}$	0.67±0.02	1.45±0.05	1.52±0.20
Cm	-	0.18±0.02	0.32±0.02	0.31±0.14
Zr	10.73 $^{+5.07}_{-4.30}$	9.46±1.16	8.97±1.17	8.19±1.69
Y	-	0.12±0.07	-	0.31±0.05
Ce	-	0.20±0.00	-	0.45±0.11
Nd	-	1.25±0.12	-	2.30±1.11
Gd	-	0.16±0.03	-	0.32±0.15

*: 14 samples were prepared from many fuel rods fabricated by several separate batches.
**: 2 samples were prepared from a fuel rod fabricated by a single batch.

Large uncertainties in element concentration (>5%) are found by fuel fabrication batches.
Fuel stack was formed with several fuel rods. → Fuel composition varies by stack position.

MA Transmutation Performance
– METAPHIX-1 –

Problems:

Low-burnup experiment, METAPHIX-1

No more than ~2.1-2.6at.% of HM fissioned.

Large uncertainties in the initial fuel composition, a point of reference in MA transmutation

More than 5% in element concentration can be varied by fuel alloys.

→ To evaluate MA transmutation performance through the elemental composition change is difficult.

Isotope Ratio

Isotope ratio (IR) is uniform in the initial fuel stack.

Isotope ratio in U, Pu, Am and Cm at the time of fuel fabrication

U	Ratio [%]
^{234}U	0.001
^{235}U	0.400
^{236}U	0.002
^{238}U	99.597

Pu	Ratio [%]
^{238}Pu	0.007
^{239}Pu	95.285
^{240}Pu	4.620
^{241}Pu	0.084
^{242}Pu	0.007

Am	Ratio [%]
^{241}Am	40.92
^{242m}Am	0.29
^{243}Am	58.79

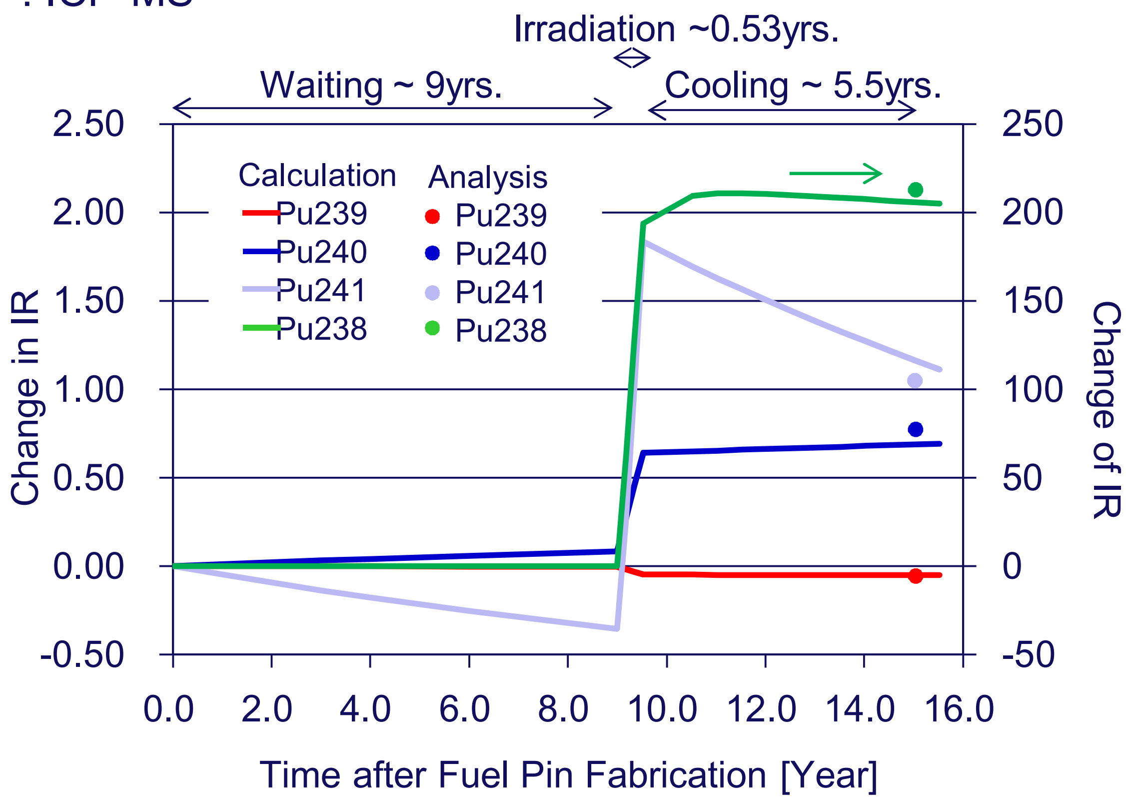
Cm	Ratio [%]
^{244}Cm	85.66
^{245}Cm	7.78
^{246}Cm	6.27
^{247}Cm	0.29

→ Change of IR in each actinide element is investigated.

$$Change\ of\ IR = \frac{(IR\ after\ fuel\ fabrication) - (IR\ at\ the\ time\ of\ fuel\ fabrication)}{IR\ at\ the\ time\ of\ fuel\ fabrication}$$

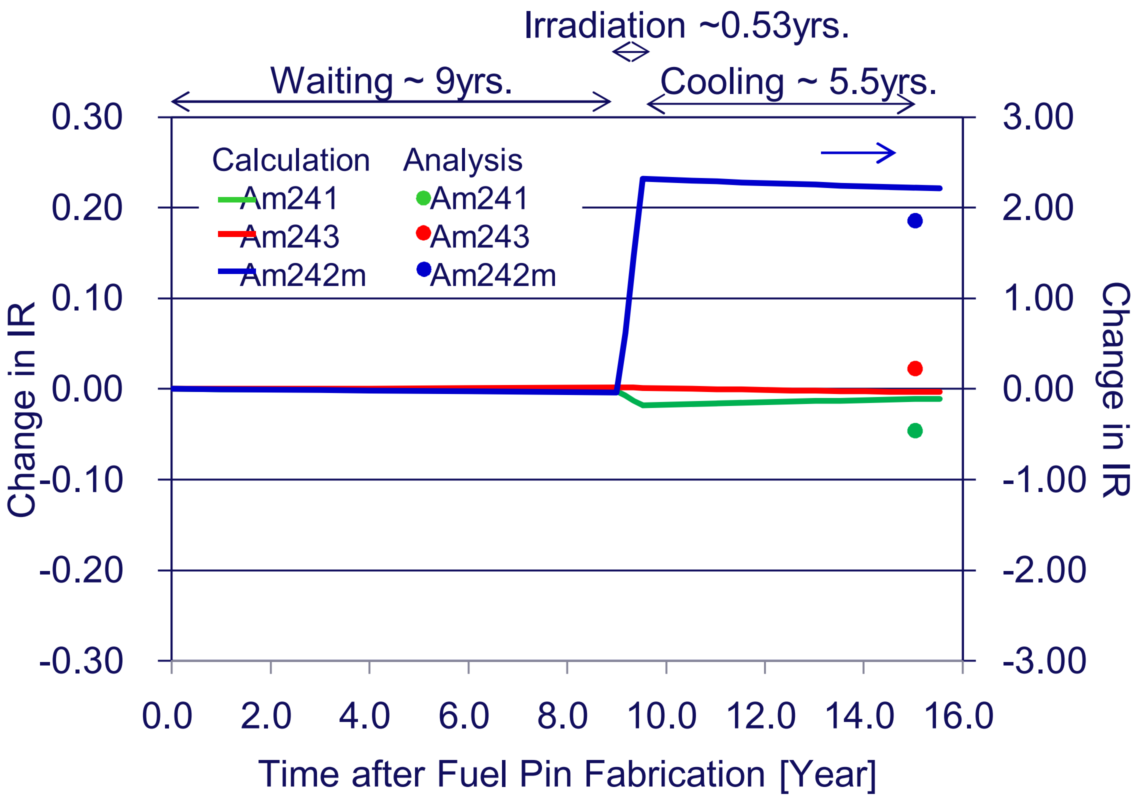
Isotope Ratio Change in Pu
– U-19Pu-10Zr-5MA –

Calculation : ORIGEN2 (ORLIBJ40 Library)
Analysis : ICP-MS



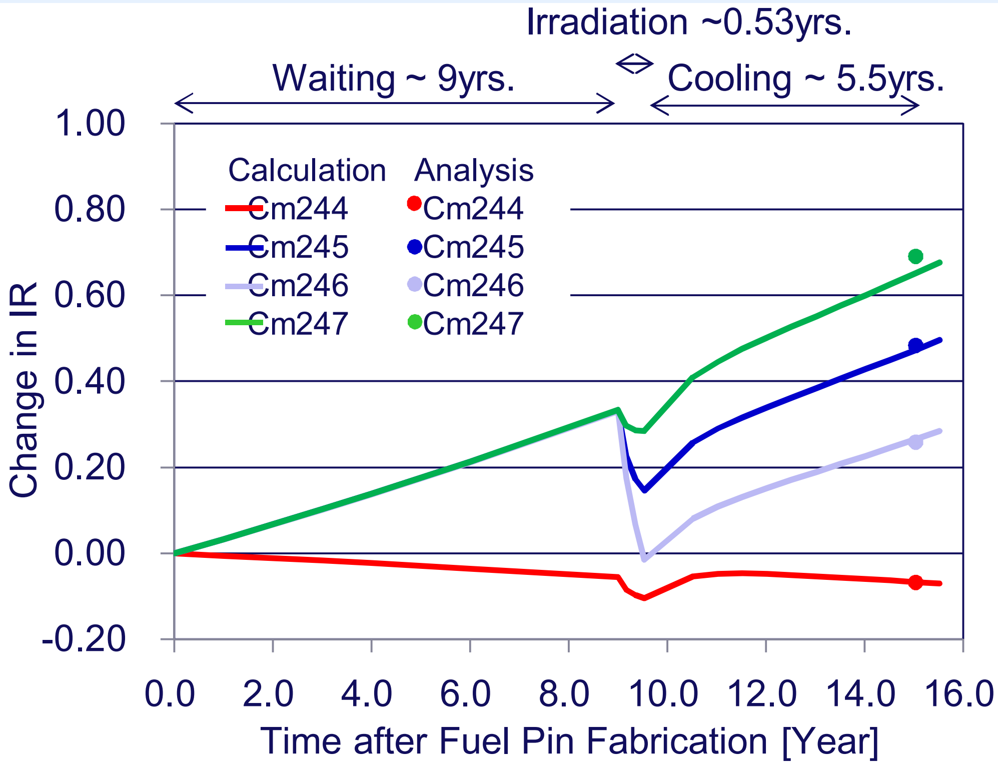
$$\text{Change of IR} = \frac{(\text{IR after fuel fabrication}) - (\text{IR at the time of fuel fabrication})}{\text{IR at the time of fuel fabrication}}$$

Isotope Ratio Change in Am
– U-19Pu-10Zr-5MA –



$$\mathit{Change\ of\ IR} = \frac{(\mathit{IR\ after\ fuel\ fabrication}) - (\mathit{IR\ at\ the\ time\ of\ fuel\ fabrication})}{\mathit{IR\ at\ the\ time\ of\ fuel\ fabrication}}$$

Isotope Ratio Change in Cm
− U-19Pu-10Zr-5MA −



$$\textit{Change of IR} = \frac{(\textit{IR after fuel fabrication}) - (\textit{IR at the time of fuel fabrication})}{\textit{IR at the time of fuel fabrication}}$$

- Isotope composition was analysed with high accuracy.
- MA burnups and decay reactions can be simulated by ORIGEN2 code.

Pu, Am and Cm nuclides are transmuted as expected.

Discussion
– Estimate of Initial Fuel Composition –

Initial fuel compositions were estimated from the postirradiation composition analyses and the burnup calculations.

	U-19Pu-10Zr	U-19Pu-10Zr-2MA-2RE	U-19Pu-10Zr-5MA
U	78.73 (78.86)	73.62 (75.30)	72.77 (73.35)
Pu	21.24 (21.11)	23.78 (22.30)	22.03 (21.41)
Np	0.00 (0.00)	1.48 (1.45)	3.13 (3.29)
Am	0.01 (0.03)	0.81 (0.75)	1.68 (1.60)
Cm	0.00 (0.00)	0.21 (0.20)	0.39 (0.35)

Within parentheses are average values in initial analysis results.

The estimated compositions are approximately within the uncertainties of initial analysis data.

Summary

Burnups of METAPHIX-1 fuels, U-19Pu-10Zr, U-19Pu-10Zr-2MA-2RE and U-19Pu-10Zr-5MA were determined quantitatively on the basis of the composition analyses by ICP-MS.

- ^{102}Ru and ^{104}Ru are applicable as alternative burnup indicators for recycled low-decontamination fuel.
- The evaluated burnups were within the range of predictions.

Actinide isotope ratio before and after METAPHIX-1 irradiation experiment was analysed and compared with ORIGEN2 calculations.

- Isotope composition was analysed with high accuracy.
- MA burnups and radioactive decay reactions after fuel fabrication can be simulated by ORIGEN2 code.
- Pu, Am and Cm nuclides in U-Pu-Zr alloy are transmuted properly as expected.

END

Thank you for your attention!!

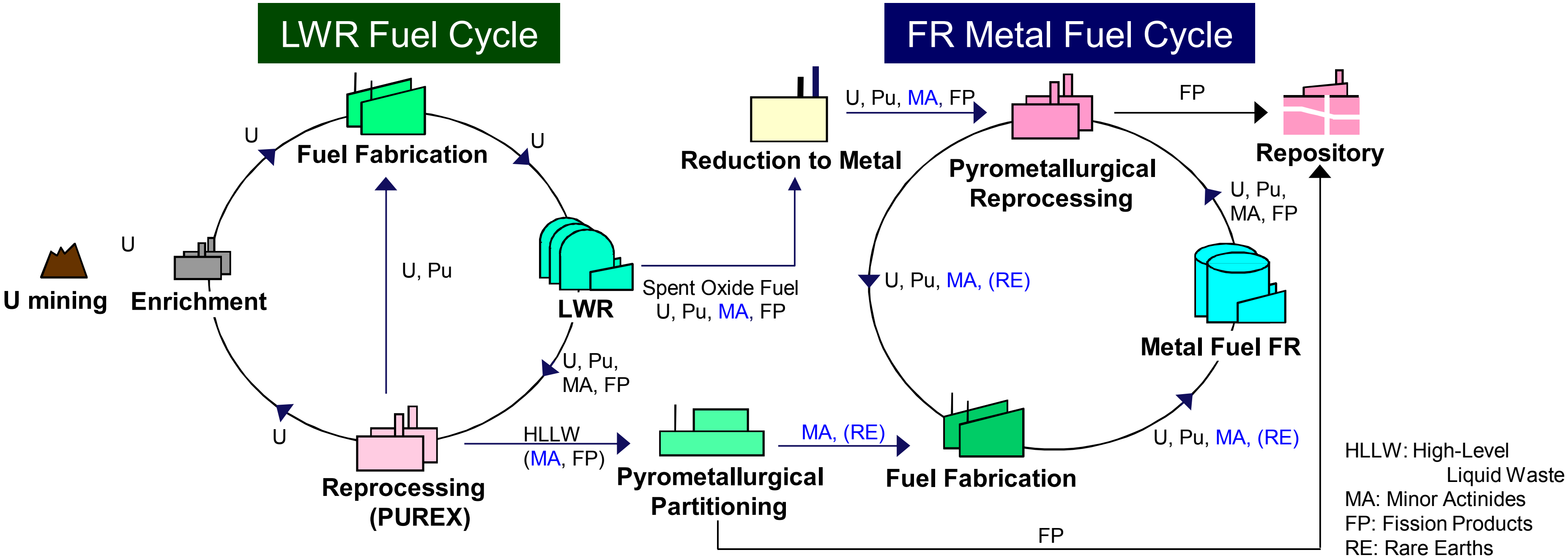
Metal Fuel & Pyrometallurgical Processes

Goals:

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

⇨ Innovative fuel cycle system is under development in CRIEPI

U-Pu-Zr Metal Fuel : Excellent nuclear performances and safety features are achievable.
Pyrometallurgical Reprocessing : MA can be recovered simultaneously with U and Pu.



Initial Composition of Irradiated Fuels

	U-19Pu-10Zr		U-19Pu-10Zr-2MA-2RE		U-19Pu-10Zr-5MA		U-19Pu-10Zr-5MA-5RE	
	Target	Average 14samples*	Target	Average 2samples**	Target	Average 2samples**	Target	Average 2samples**
U	71	$70.43^{+2.60}_{-5.46}$	67	66.85±1.55	66	66.30±1.30	61	63.50±2.50
Pu	19	$18.85^{+1.02}_{-0.74}$	19	19.80±0.20	19	19.35±0.05	19	19.75±0.55
Np	-	-	1.2	1.29±0.09	3.0	2.97±0.02	3.0	3.04±0.16
Am	-	$0.03^{+0.11}_{-0.02}$	0.6	0.67±0.02	1.6	1.45±0.05	1.6	1.52±0.20
Cm	-	-	0.2	0.18±0.02	0.4	0.32±0.02	0.4	0.31±0.14
Zr	10	$10.73^{+5.07}_{-4.30}$	10	9.46±1.16	10	8.97±1.17	10	8.19±1.69
Y	-	-	0.2	0.12±0.07	-	-	0.5	0.31±0.05
Ce	-	-	0.2	0.20±0.00	-	-	0.5	0.45±0.11
Nd	-	-	1.4	1.25±0.12	-	-	3.5	2.30±1.11
Gd	-	-	0.2	0.16±0.03	-	-	0.5	0.32±0.15

*: 14 samples were prepared from fuel alloys fabricated by several different batches.

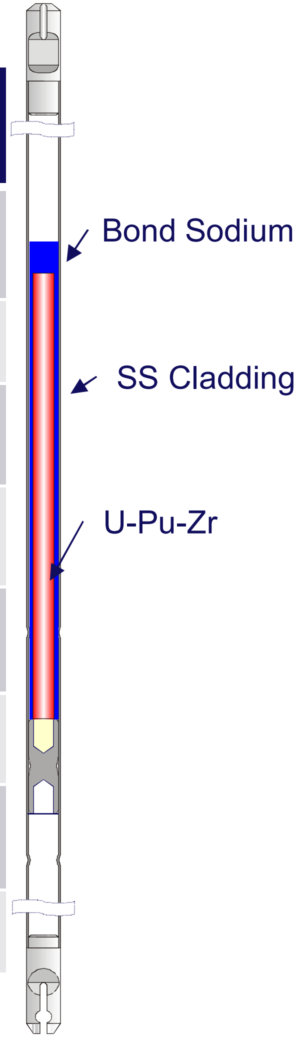
**: 2 samples were prepared from a fuel alloy fabricated by a single batch.

Average composition agrees well with target.

Large uncertainties in element concentration (>5%) are found by fuel fabrication batches.

Characteristics of FP

Mass Number	Stable FP	Characteristics	Applicability as Burnup Indicator
86-88	Sr, Rb	High mobility Soluble in Bond-sodium	Impossible
89	Y	Contamination FP in fresh fuel	Difficult
90-100	Zr, Mo, etc.	Fuel matrix Cladding material	Difficult
101-105	Ru, Rh, Pd	Low mobility	Possible
106-108	Pd	Significant Interference by isober（ZrO^+）	Difficult
109-126	Cd, Sn, etc.	Low fission yield	Difficult
127-138	I, Cs, etc.	High mobility Soluble in Bond-sodium	Impossible
139-170	Ce, Nd, etc.	Contamination FP in fresh fuel	Difficult



Fission Yield

Fast Fission Yield * of Stable Fission Products [%]

	^{101}Ru	^{102}Ru	^{103}Rh	^{104}Ru	^{105}Pd	^{139}La	^{148}Nd	^{148}Nd(T)
^{238}U	6.21	6.45	6.28	5.04	4.05	5.67	2.11	-
^{239}Pu	6.67	6.73	6.83	6.58	5.36	5.61	1.66	1.64
^{240}Pu	6.48	6.67	6.61	6.84	5.58	5.98	1.77	1.77
^{241}Pu	6.49	6.87	6.51	7.31	6.58	6.31	1.96	1.93
^{242}Pu	5.99	6.42	6.54	6.49	6.97	6.02	2.00	1.99
^{237}Np	6.12	5.77	5.56	4.13	3.10	5.63	1.72	1.66
^{241}Am	6.10	6.06	6.45	6.02	5.34	6.75	1.90	1.83
^{243}Am	5.92	5.99	5.91	5.99	5.87	5.95	1.87	-
^{244}Cm	6.20	6.50	6.70	6.70	6.40	6.57	2.48	-
平均	**6.42**	**6.38**	**6.38**	**6.12**	**5.47**	**6.05**	**1.94**	**1.80**
標準偏差	**0.24**	**0.35**	**0.38**	**0.93**	**1.16**	**0.39**	**0.23**	**0.13**

T.R. England, et al., ENDF-349 (1992) *: Incident Neutron Energy= 500keV ,
^{148}Nd(T) : Thermal Fission Yield

Neutron Capture Cross-section

Neutron capture cross-section [barn]

	^{101}Ru	^{102}Ru	^{103}Rh	^{104}Ru	^{105}Pd
Fast Reactors[1]	0.692	0.157	0.530	0.157	0.899
Thermal Reactor[2]	2.942	0.235	34.40	0.251	4.151

	^{139}La	^{148}Nd
Fast Reactors[1]	0.032	0.129
Thermal Reactor[2]	0.878	0.630

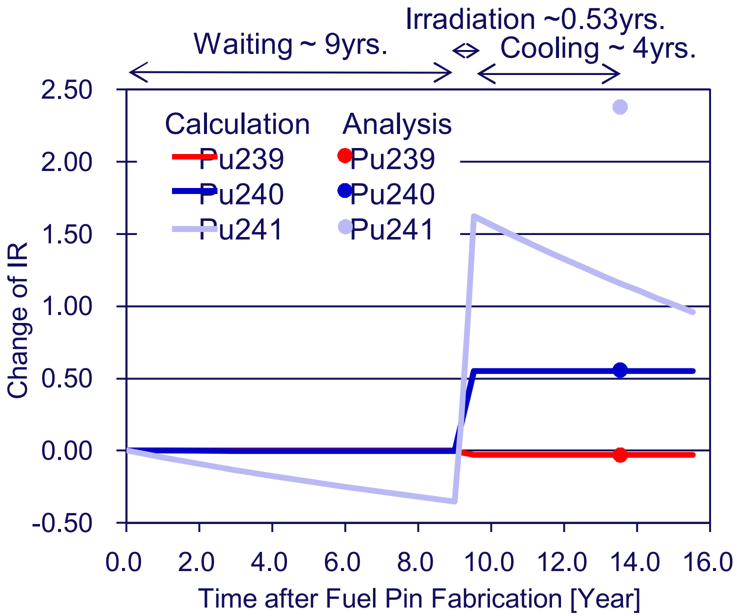
K. Suyama et al., JAERI-Data/Code 99-0003 (1999)

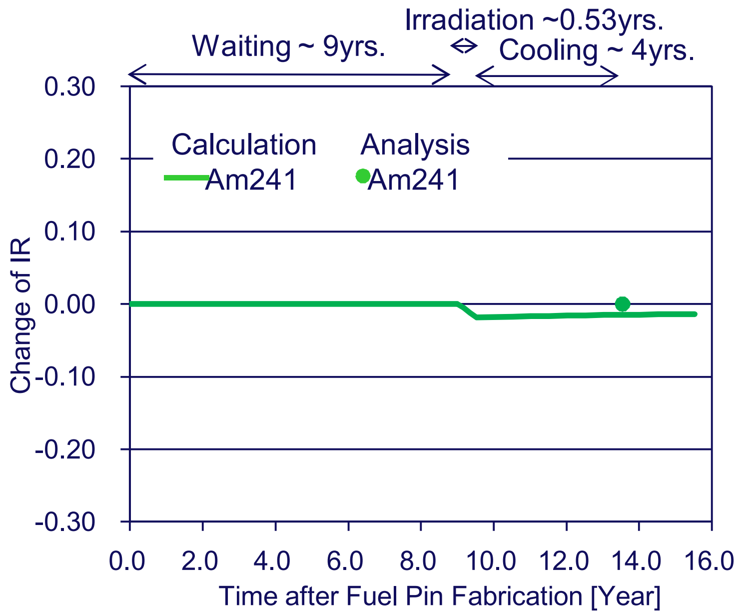
1: Prototype FBR（Inner core）: MONJMXIC,

2: U concentration=4.1% PWR: PWR41J32

Isotope Ratio Change in Actinide Elements (1)
– U-19Pu-10Zr –

Calculation : ORIGEN2 (ORLIBJ40 Library)
Analysis : ICP-MS

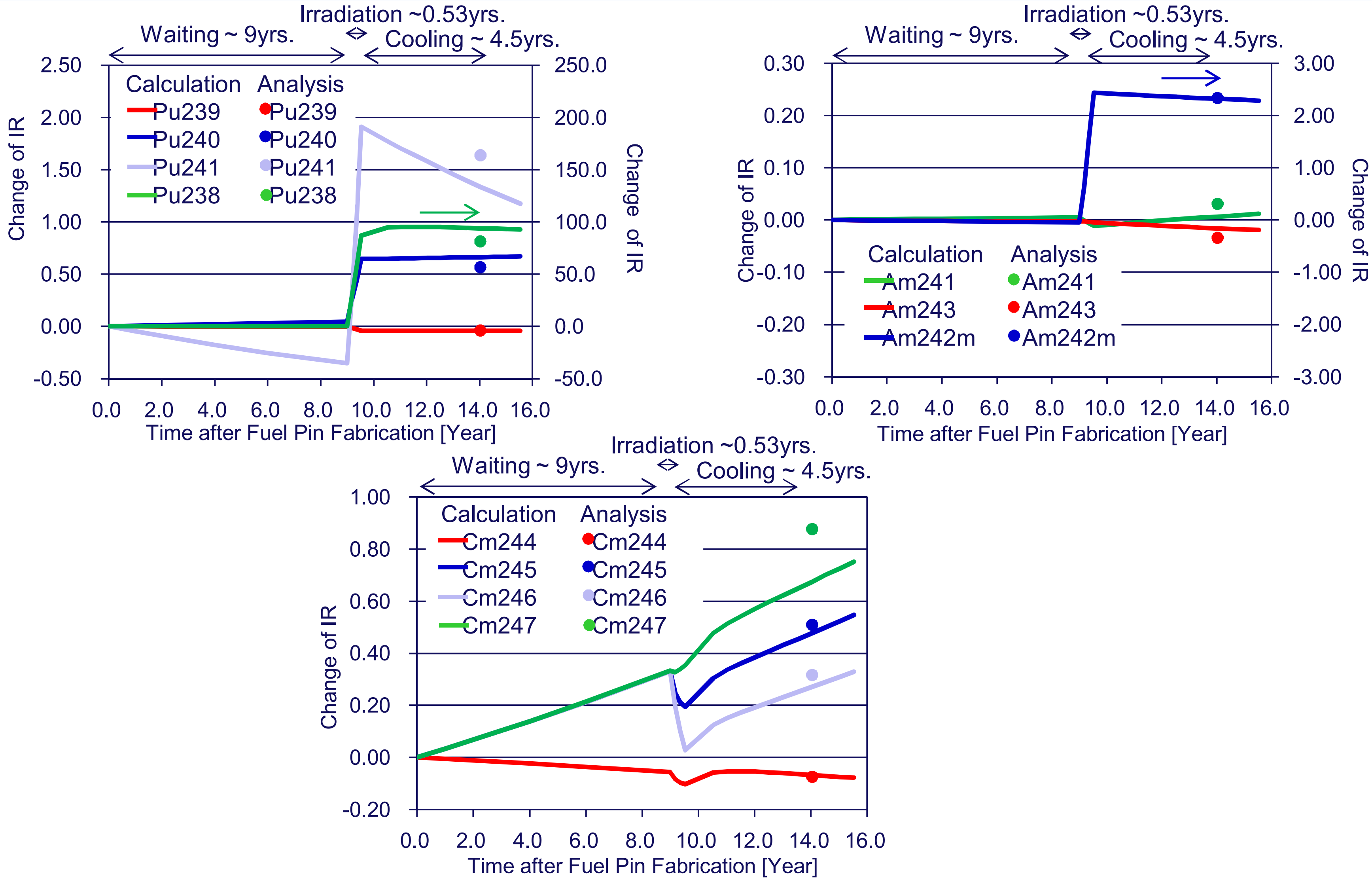




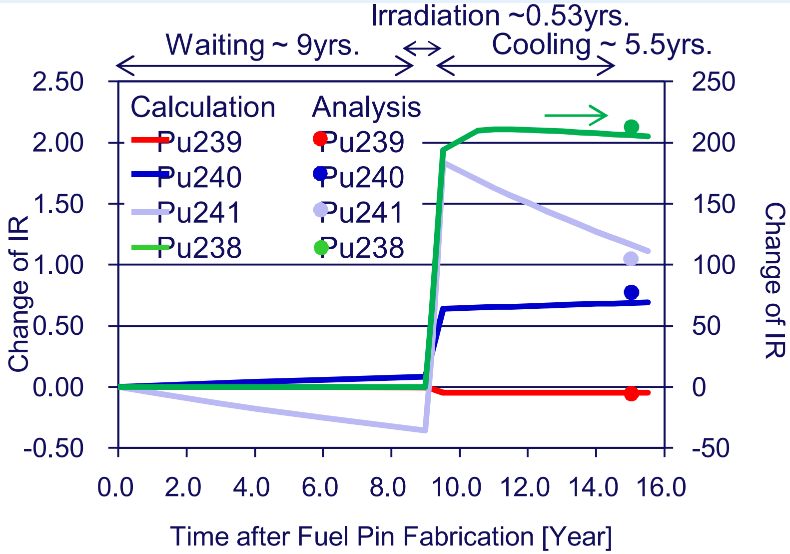
$$\textit{Change of IR} = \frac{(\textit{IR after fuel fabrication}) - (\textit{IR at the time of fuel fabrication})}{\textit{IR at the time of fuel fabrication}}$$

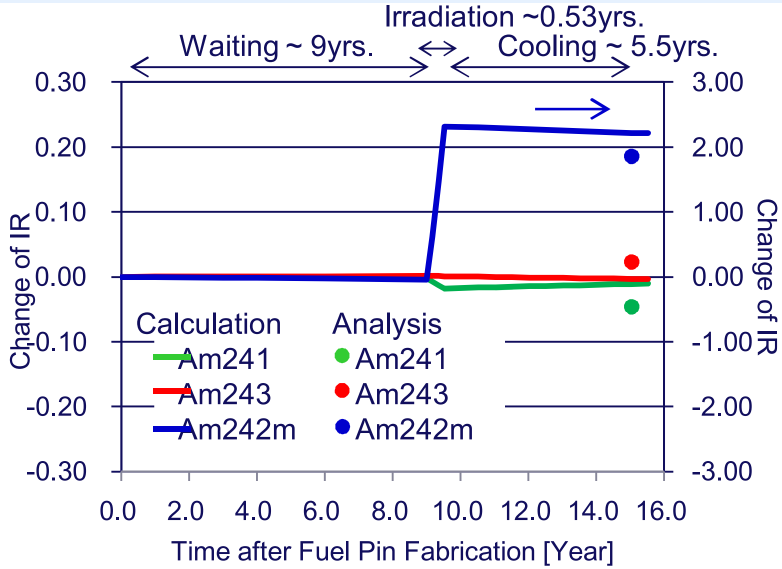
Accuracy of ICP-MS analysis for ^{241}Pu is low because of the presence of ^{241}Am.

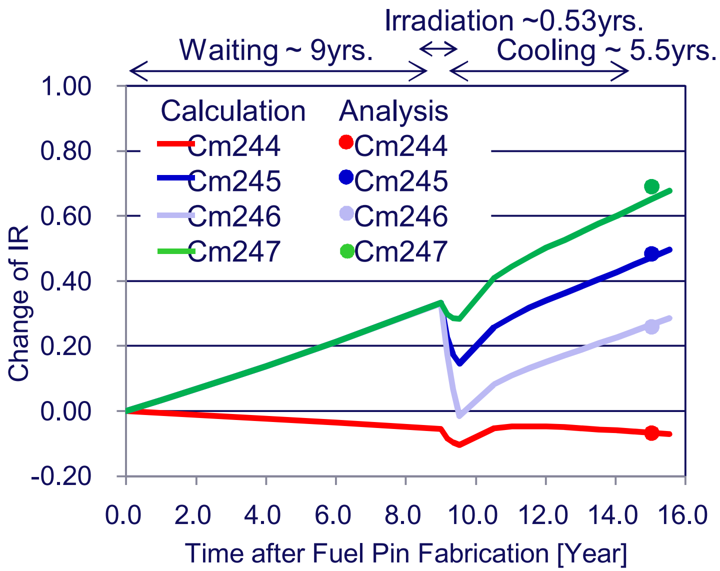
Isotope Ratio Change in Actinide Elements (2)
– U-19Pu-10Zr-2MA-2RE –



Isotope Ratio Change in Actinide Elements (3)
− U-19Pu-10Zr-5MA −







- Actinide isotope composition was analysed with high accuracy.
- MA burnups and radioactive decay reactions can be simulated by ORIGEN2 code (ORLIBJ40).

Pu, Am and Cm nuclides are transmuted as expected.