Progress in Understanding of Fuel-Cladding Chemical interaction in Metal Fuel

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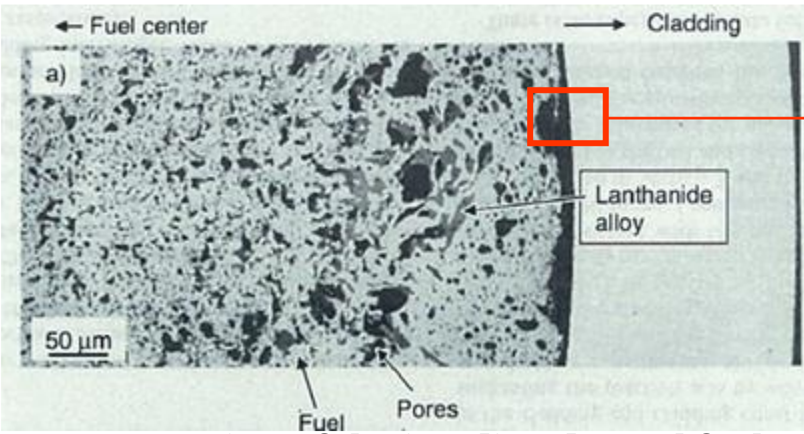
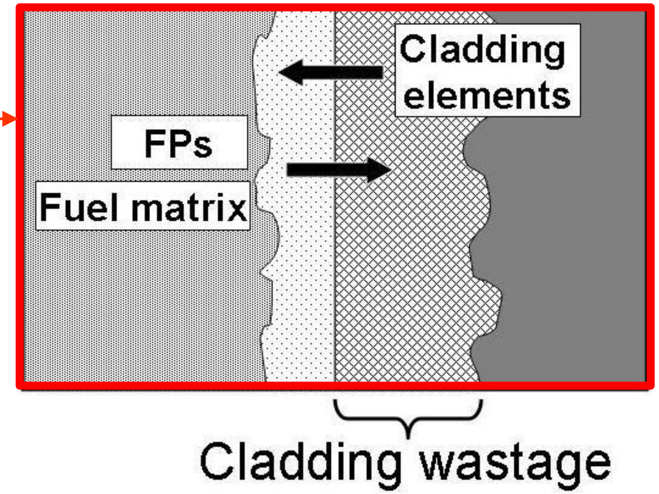
U-Pu-Zr alloy is a candidate for the metallic fuel for Fast Reactor.



*Cross section of the irradiated metal fuel**



FCCI(Fuel-Cladding Chemical Interaction)
Reaction occurs between cladding and FPs or fuel matrix.

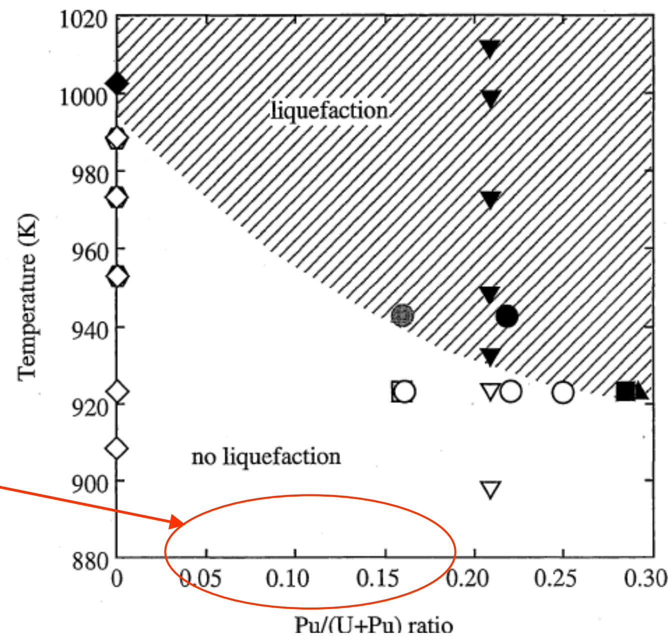
- FPs and cladding
  - The reaction of lanthanide elements is dominant among other FPs.
  - During normal operation temperature lower than 923K.
- Fuel alloy (U-Pu-Zr) and cladding
  - Liquefaction can occur at transient period.
  - Its threshold temperature depends on the Pu concentration.

*Y.S.Kim et al., Journal of Nucl. Mat. 392 (2009) 164

Purpose

Out-of-pile diffusion couple tests were conducted to investigate the characteristics of the reactions.

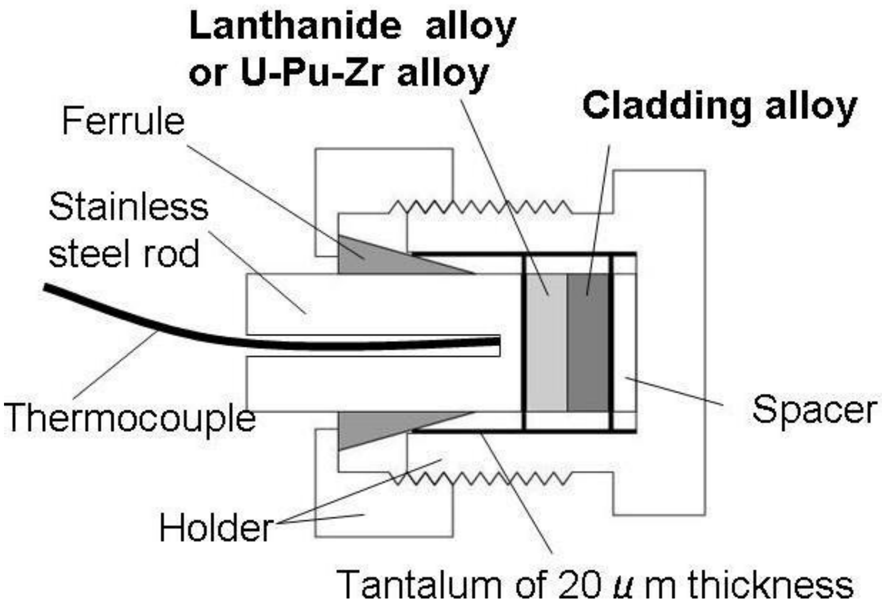
- Lanthanide alloy / several cladding materials
  - Identify the phases formed in the reaction
  - Investigate the influence of the alloying elements in the cladding materials
  - Formulate the growth rate of the wastage layer formed in the cladding side
- U-Pu-Zr / Fe
  - Identify the phases formed in the reaction
  - Estimate the threshold temperature for the liquefaction in lower Pu region



Summary of liquefaction data in the reaction between U-Pu(-Zr) alloy with Fe-based alloys*

**K.NAKAMURA et al., Journal of Nucl. Sci. Tech.. 38,2 (2001) 112*

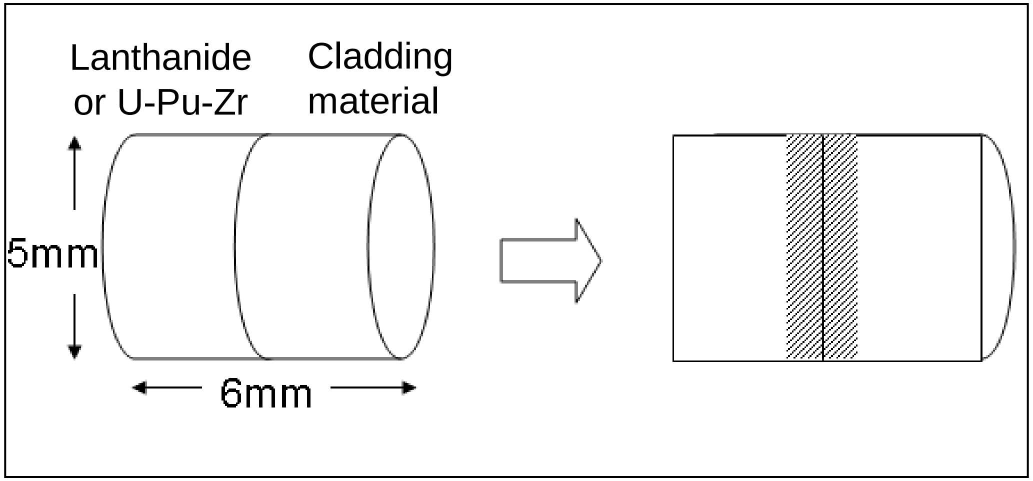
Experimental procedure



Configuration of the diffusion couple

- Diffusion couples
  ① Lanthanide alloy / cladding
  ② U-Pu-Zr alloy / Fe
- The reaction layer was examined by X-ray spectrometry.

After the isothermal annealing



Examination of the reaction layer in the specimens

Reaction between Lanthanide alloy (La-Ce-Nd-Pr-Sm) and cladding

Tests of the lanthanide / cladding couples

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Summary of test conditions for lanthanide alloy with cladding materials

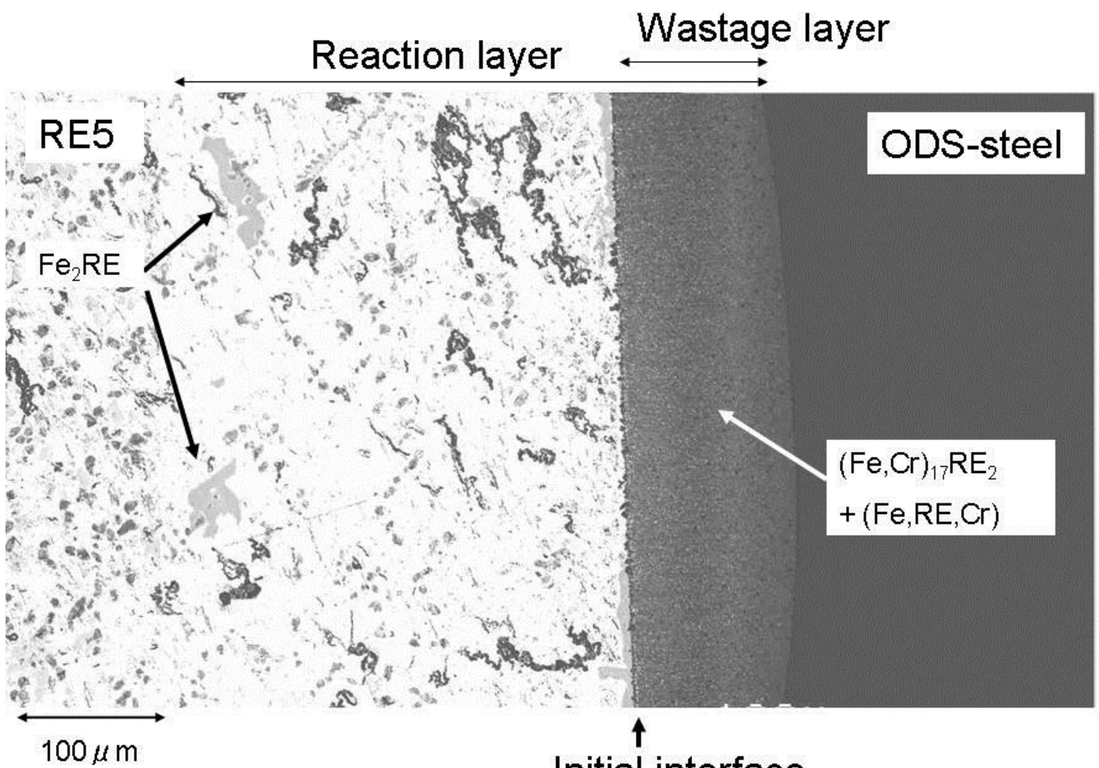
Combination of the diffusion couple	Temperature [K]	Time [hours]	Remarks of cladding
RE5 / ODS-steel	853, 923	12-170	Candidate cladding material in Japan
RE5 / PNC-FMS	853, 892, 923	12-170	Candidate cladding material in Japan
RE5 / HT9	853	162	Candidate cladding material in U.S.
RE5 / Fe-12wt%Cr	923	12	Simulating alloy for cladding materials

RE5 : 13La –24Ce –12Pr –39Nd –12Sm wt.%

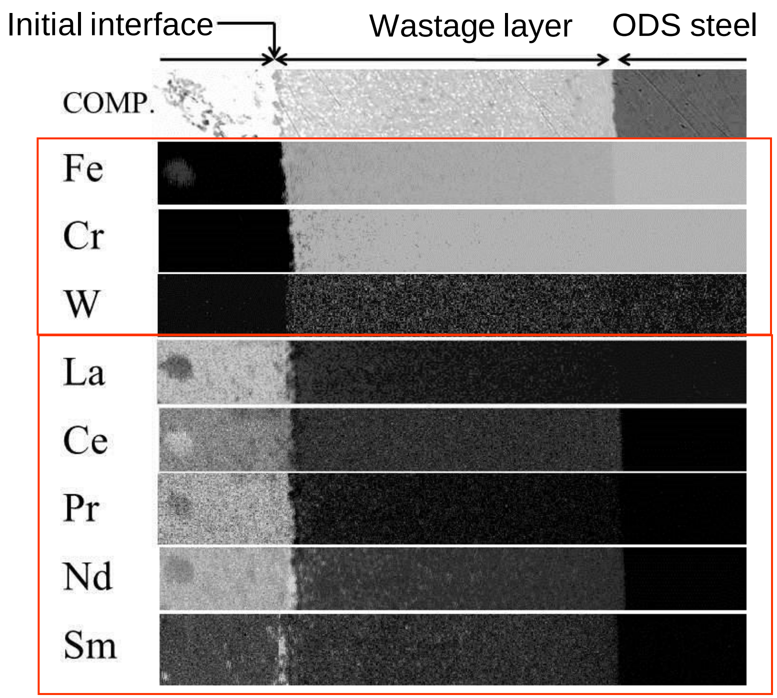
- Tests were conducted between 853K and 923K.
  - 923K is the maximum inner cladding temperature during the normal operation of metal fuel FBR in the case of Pu/(U+Pu)≦0.25.
  - reaction between metal fuel components (U,Pu,Zr) and Fe becomes dominant above 923K

  -the reaction is not significant below 853K.

Reaction layer in RE5/ODS-steel couple



RE5/ODS-steel annealed at 923 K for 50 hours.



X-ray intensity map in the cladding wastage layer

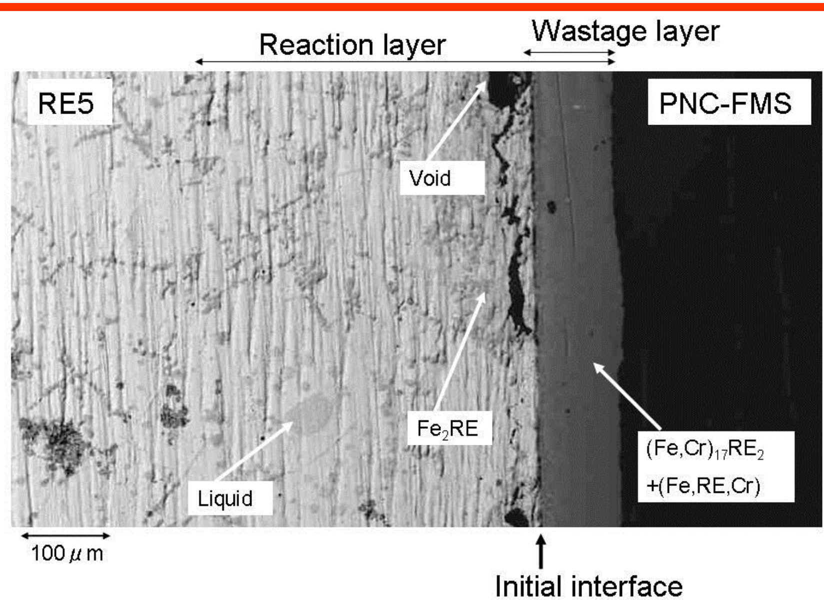
RE5 side

- Fe diffused and precipitated phase of Fe_2RE was formed.
- Diffusion of other cladding elements (Cr, W, etc) was not obvious.

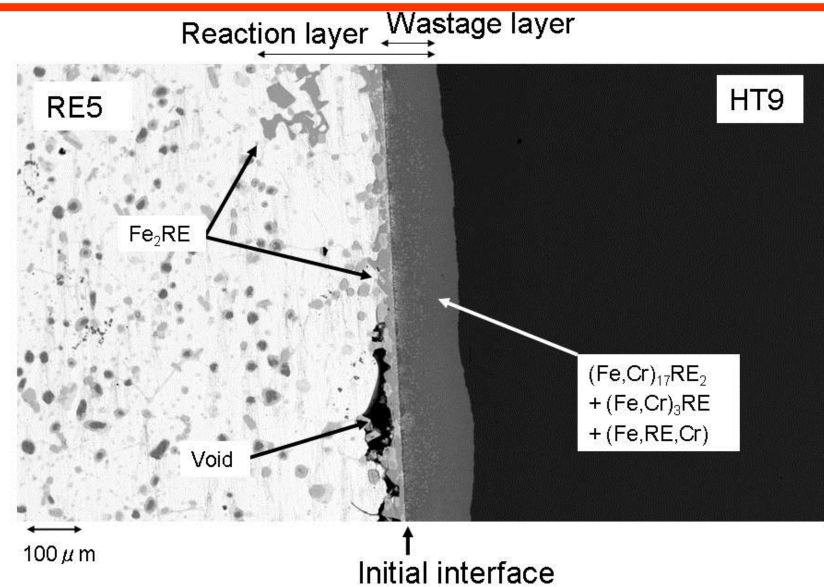
cladding side

- Wastage layer consisted of $(Fe,Cr)_{17}RE_2$ and RE-rich precipitates.

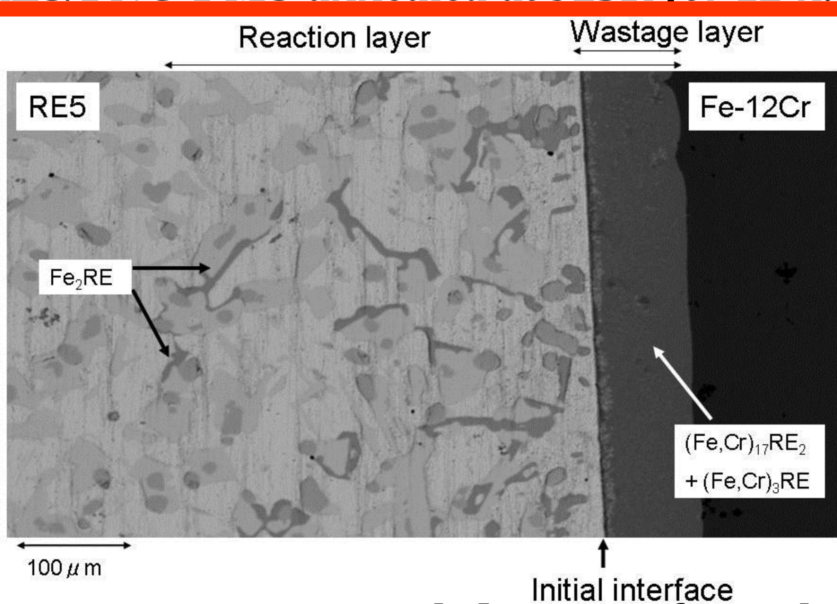
Reaction layers in other couples



RE5/PNC-FMS annealed at 923K for 12 h.



RE5/HT9 annealed at 853K for 162 h.



RE5/Fe-12Cr annealed at 923K for 12 h.

- The liquid phase and void were formed on RE5 side in RE5/PNC-FMS

- Void were formed on RE5 side in RE5/HT9
- Ni depleted from HT9 near interface.

- Reaction in RE5/Fe-12Cr was similar to those in ODS-steel, PNC-FMS and HT9.

Summary of the reaction layers (RE5/cladding)

Temp.	RE5 side	Cladding side
		ODS-steel
923	Fe_2RE	$(Fe,Cr)_{17}RE_2$ + RE-rich
853	Fe_2RE	$(Fe,Cr)_{17}RE_2$ + RE-rich
		PNC-FMS
923	Fe_2RE + Liquid+ Void	$(Fe,Cr)_{17}RE_2$ + RE-rich
892	Fe_2RE + Void	$(Fe,Cr)_{17}RE_2$ + RE-rich
853	Fe_2RE	$(Fe,Cr)_{17}RE_2$ + RE-rich
		HT9
853	Fe_2RE + Void	$(Fe,Cr)_{17}RE_2$ + $(Fe,Cr)_3RE$ + RE-rich
		HT9
923	Fe_2RE	$(Fe,Cr)_{17}RE_2$ + $(Fe,Cr)_3RE$

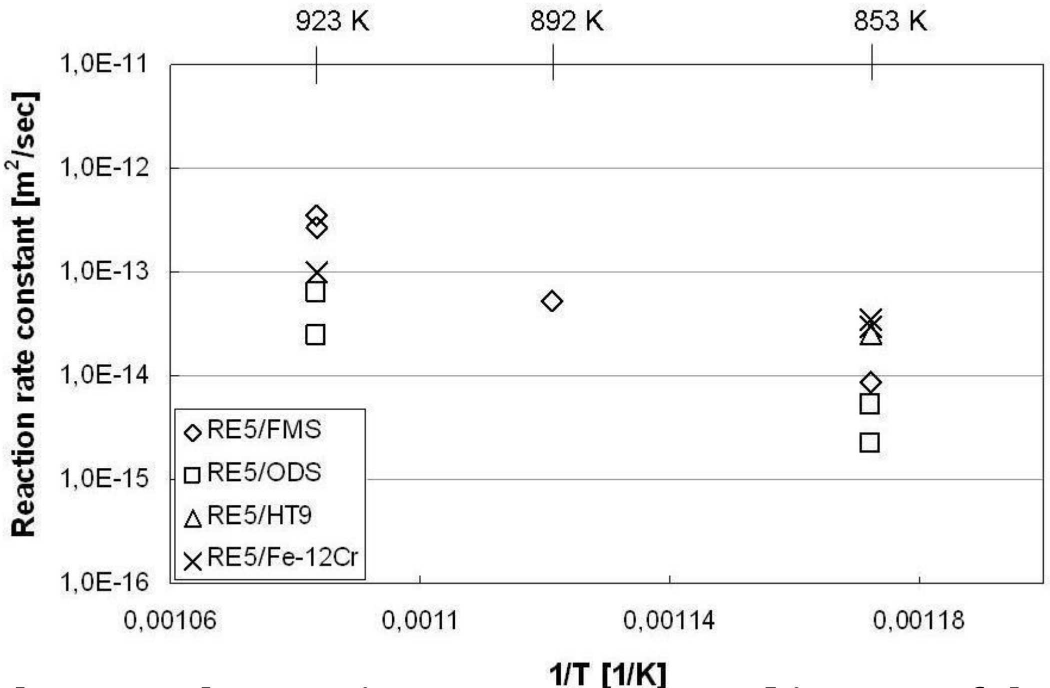
- Although several differences were observed in the reaction layer in RE5 side, the wastage layer consisted of the matrix of $(Fe,Cr)_{17}RE_2$ and the lanthanide-rich precipitated phases in common.

- The influence of the alloying elements to the cladding wastage layer formation was not significant.

Characteristics of the growth of wastage layer

- The growth of the wastage layer was diffusion controlled.

$$(\text{Reaction rate constant}) = \frac{\delta^2}{t}$$



Relation between the reaction rate constant and inverse of the temperature

- This result can be the basis for quantification of FCCI.
- ODS-steel provided better resistance characteristics compared to the PNC-FMS.

Reaction between U-Pu-Zr alloy and Fe

Tests of the U-Pu-Zr / Fe couples

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- These tests were conducted in the globe box in Plutonium Fuel Research Facility of JAEA-Oarai in Japan

Casted alloy of U-16Pu-10Zr

- U-9Pu-10Zr alloy fabricated in the previous study* was used.

**NAKAMURA, K., et al., "U-Pu-Zr metallic fuel fabrication for irradiation test at Joyo", in: Proc. FR09, Kyoto, Japan, Dec. 7-11, (2009)*

- U-16Pu-10Zr alloy was manufactured by mixing existing U-20Pu-10Zr alloy rod with pure U and pure Zr by arc melting and casted by injection casting technique .

Summary of test conditions for U-Pu-Zr with Fe.

Combination of the diffusion couple	Temperature [K]	Time [hours]
U-16Pu-10Zr/Fe	943, 963	25
U-9Pu-10Zr/Fe	963, 983	30

- The temperature of the tests were selected to estimate the threshold temperature for the liquefaction

Reaction layer in U-16Pu-10Zr/Fe at 963K

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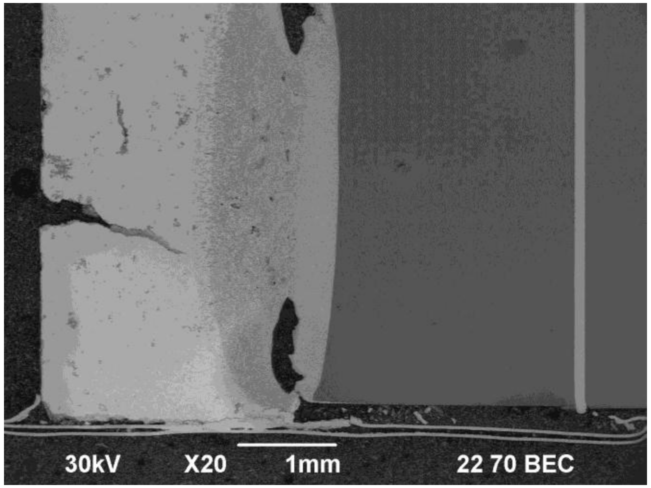
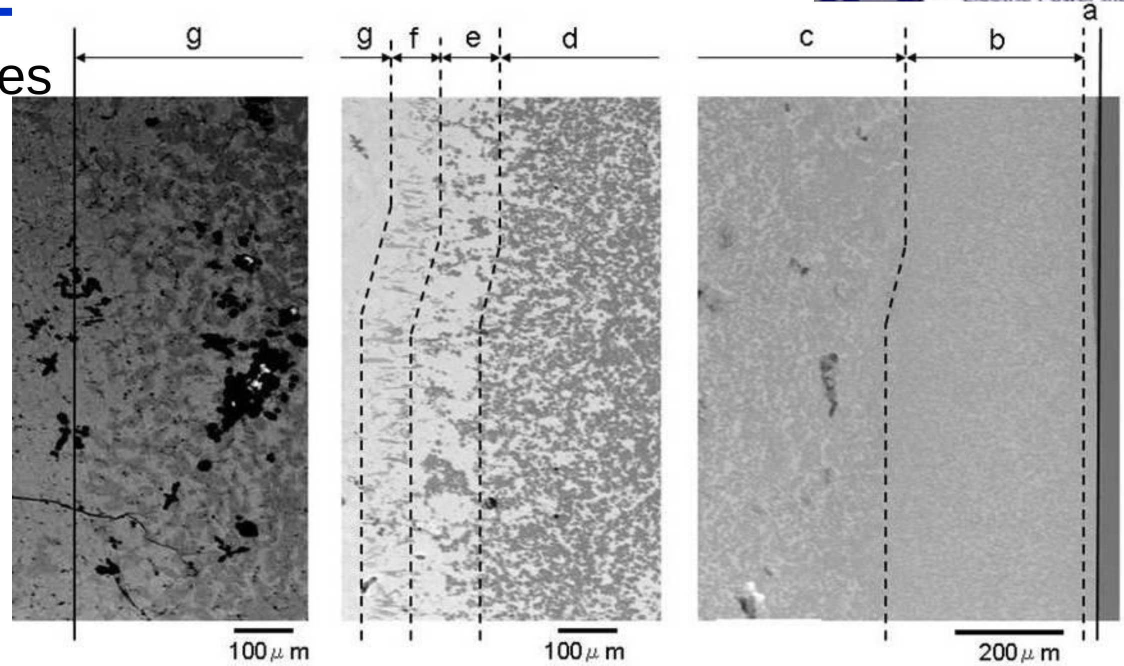
Back-scattered electron images



Whole view

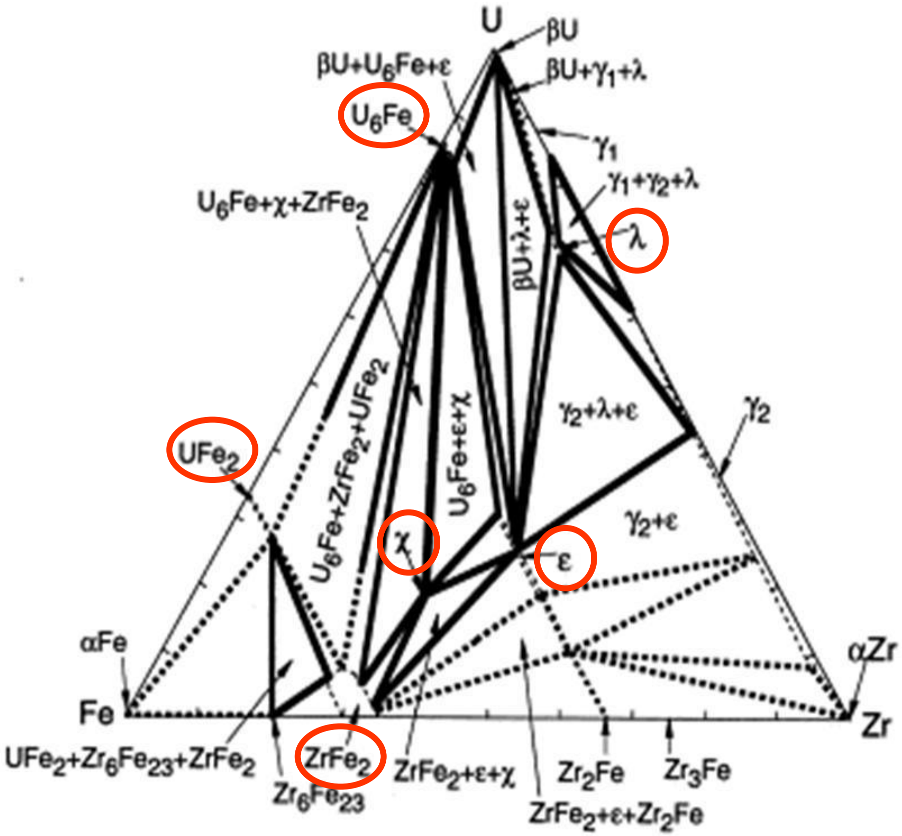


Enlarged image

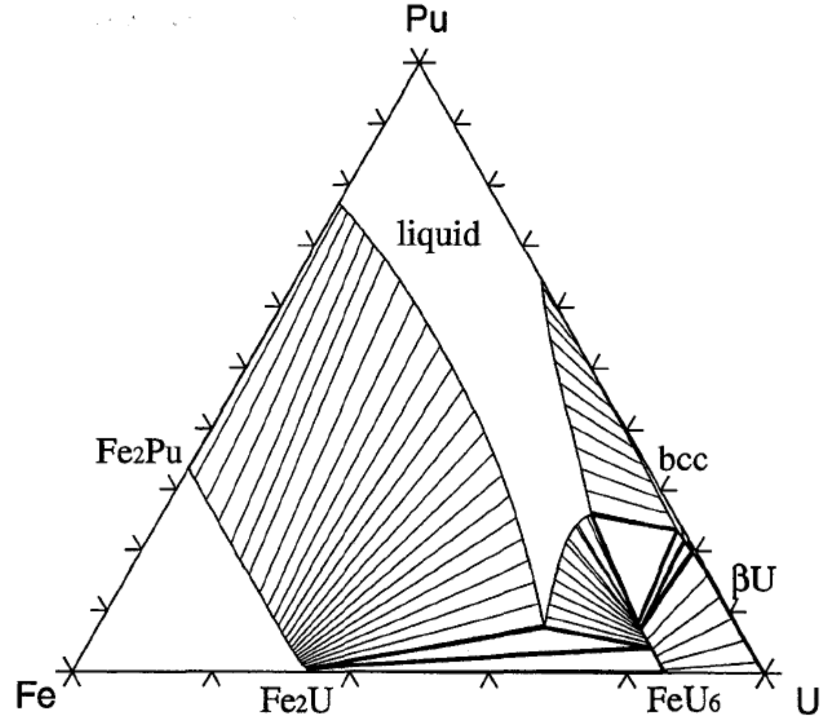
Identified phases

g	f	e	d	c	b	a
bcc + λ	$(U,Pu,Zr)_6Fe$ + χ + ε	$(Zr,U,Pu)Fe_2$ + $(U,Pu,Zr)_6Fe$ + Liquid	$(Zr,U,Pu)Fe_2$ + $(U,Pu,Zr)_6Fe$ + Liquid	$(U,Zr,Pu)Fe_2$ + $(Zr,U,Pu)Fe_2$ + $(U,Zr,Pu)_6Fe$ + Liquid	$(U,Zr,Pu)Fe_2$ + $(U,Zr,Pu)_6Fe$	$(U,Zr,Pu)Fe_2$

Ternary phase diagrams (U-Zr-Fe, U-Pu-Fe)



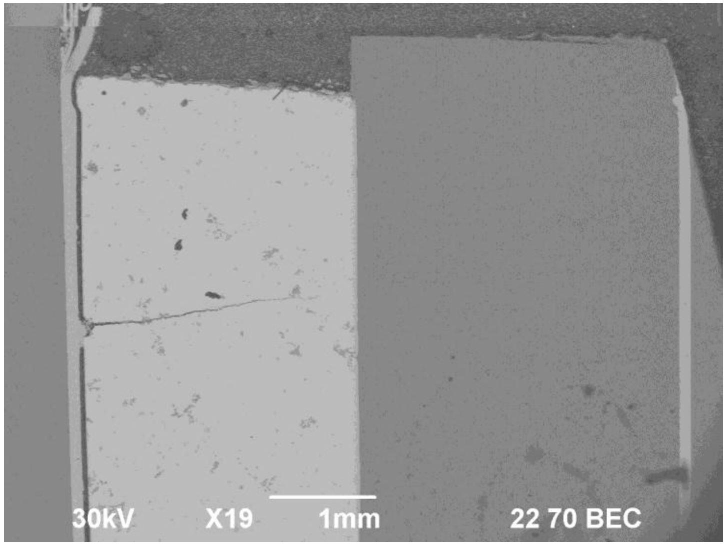
U-Zr-Fe ternary phase diagram at 973 K



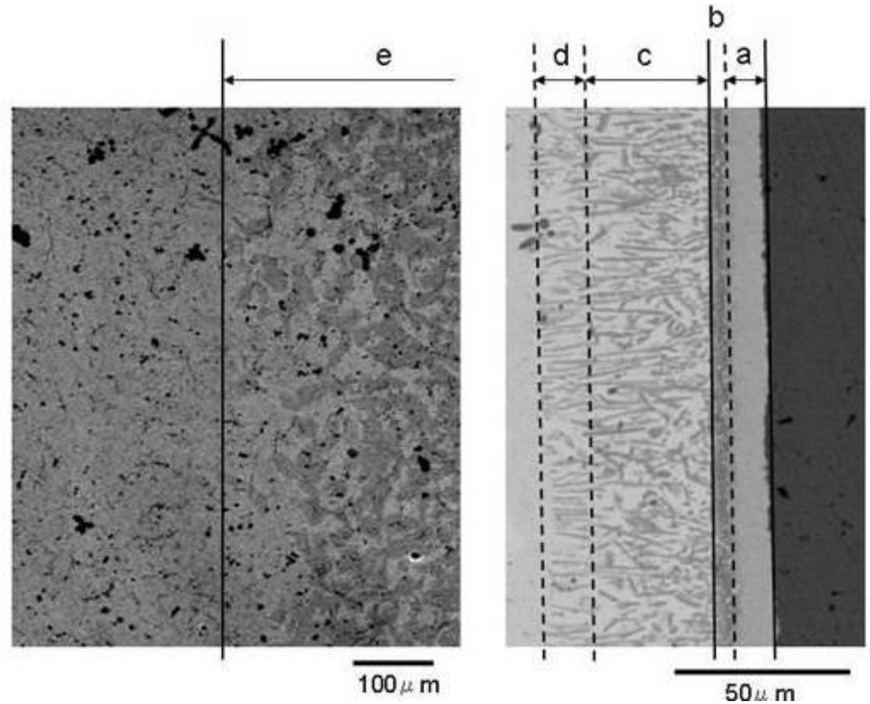
U-Pu-Fe ternary phase diagram at 983 K

Reaction layer in U-16Pu-10Zr/Fe at 943K

Back-scattered electron images



Whole view



Enlarged image

Identified phases

e	d	c	b	a
bcc + λ	$(U,Pu,Zr)_6Fe$ + χ	$(Zr,U,Pu)Fe_2$ + $(U,Zr,Pu)_6Fe$	$(Zr,U,Pu)Fe_2$	$(U,Zr,Pu)Fe_2$

Reaction layer in U-9Pu-10Zr/Fe at 983K

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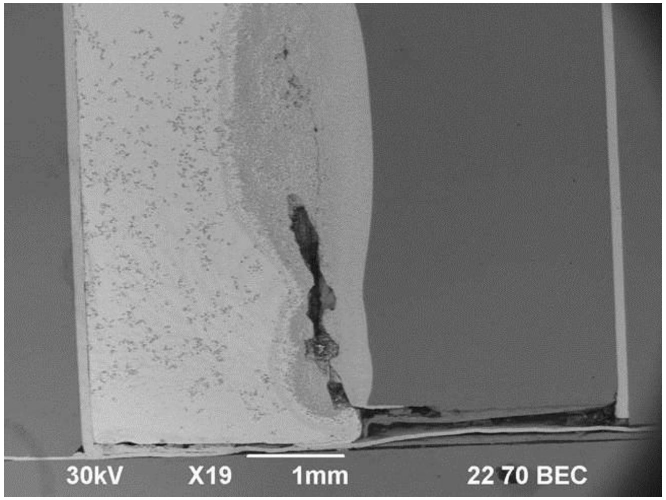
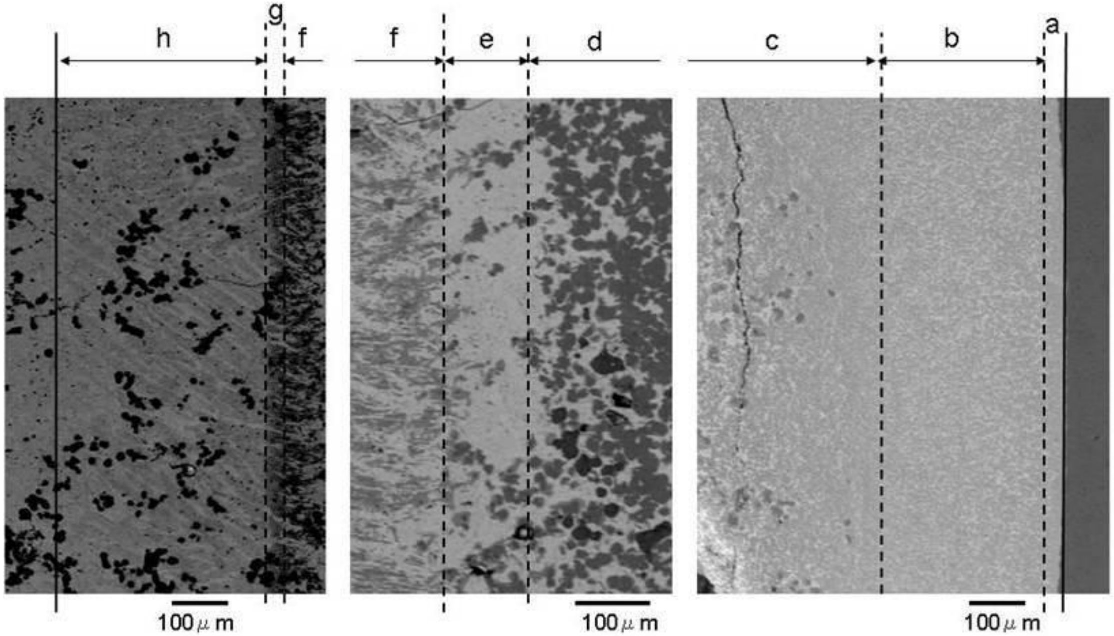
Back-scattered electron images



Whole view



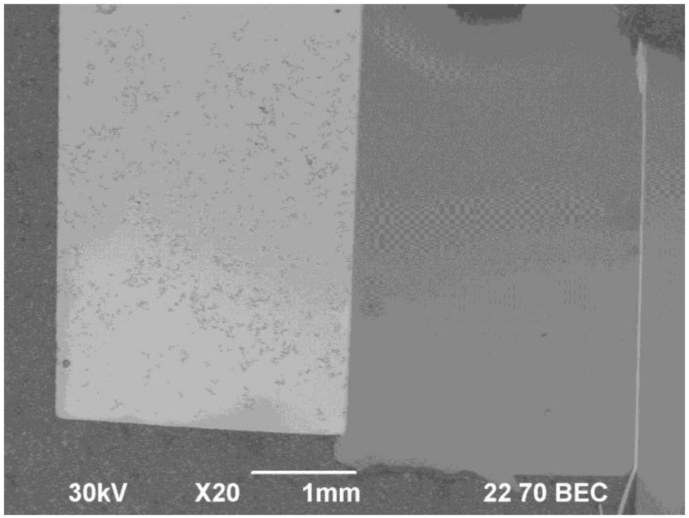
Enlarged image

Identified phases

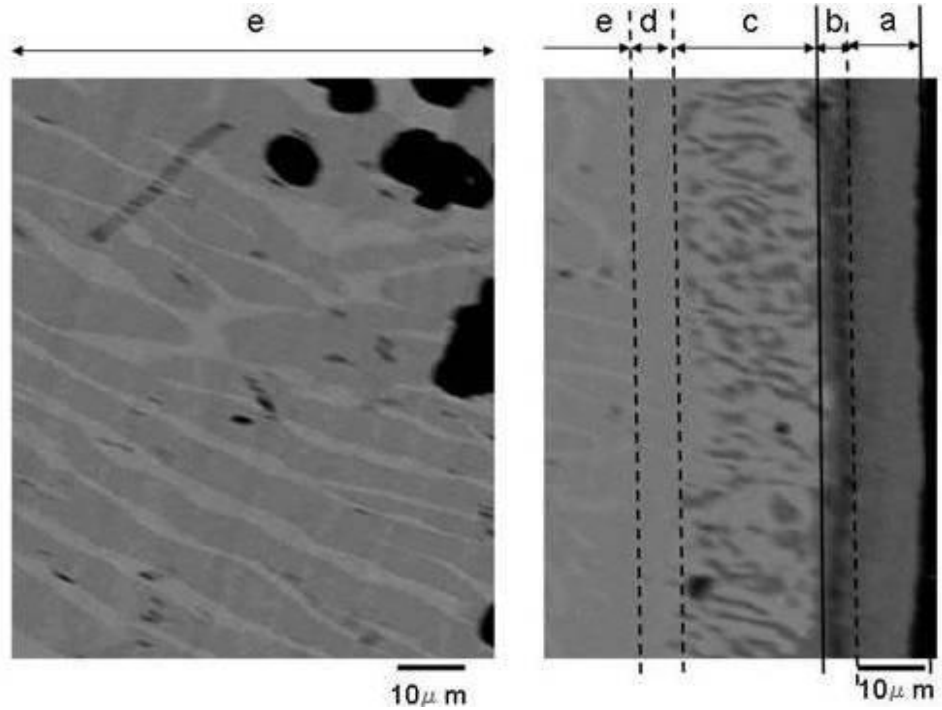
h	g	f	e	d	c	b	a
bcc + λ	(U,Zr,Pu)$_6$Fe + χ + ε	(U,Zr,Pu)$_6$Fe + (Zr,U,Pu)Fe$_2$	(U,Zr,Pu)$_6$Fe + Liquid	(Zr,U,Pu)Fe$_2$ + (U,Zr,Pu)$_6$Fe + Liquid	(U,Zr,Pu)Fe$_2$ + (U,Zr,Pu)$_6$Fe + χ + Liquid	(U,Zr,Pu)Fe$_2$ + (U,Zr,Pu)$_6$Fe	(U,Zr,Pu)Fe$_2$

Reaction layer in U-9Pu-10Zr/Fe at 963K

Back-scattered electron images



Whole view



Enlarged image

Identified phases

e	d	c	b	a
bcc + λ	λ	$(U,Zr,Pu)_6Fe$ + χ	$(Zr,U,Pu)Fe_2$	$(U,Zr,Pu)Fe_2$

Summary of the reaction (U-Pu-Zr/Fe)

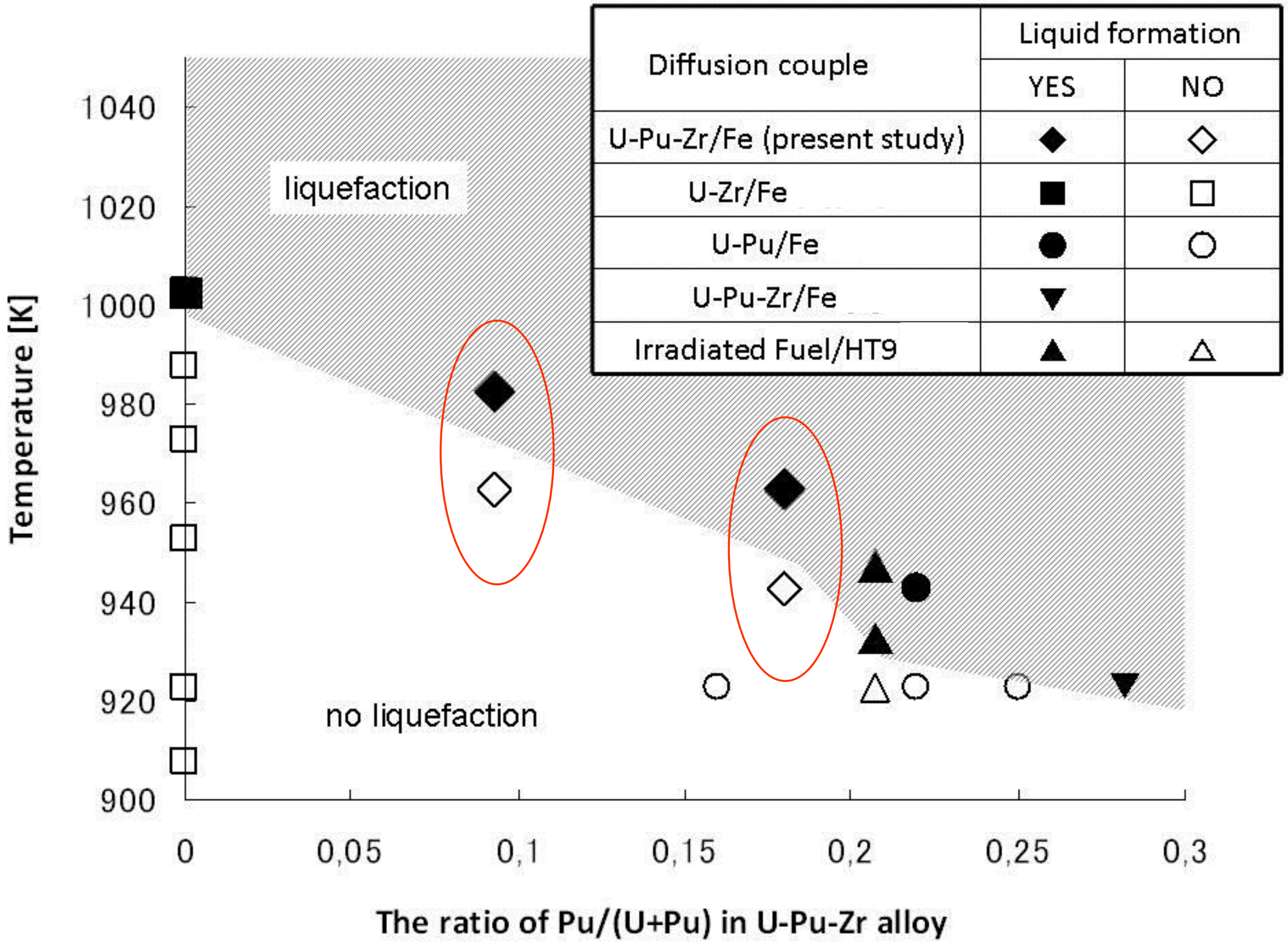
It was confirmed that

- Stoichiometry of all the phases formed in the U-Pu-Zr/Fe couples were found in the ternary phase diagram of U-Zr-Fe system.

- In the U-16Pu-10Zr/Fe couple, the liquefaction threshold temperature is between 943 and 963K.
- In the U-9Pu-10Zr/Fe couple, the liquefaction threshold temperature is between 963 and 983K.

- The growth of wastage layer is accelerated due to the formation of liquid phase.

Liquid phase formation criteria



The threshold temperature for the lower Pu content was clarified.

Representative phases formed in FCCI were identified.

- The reaction between lanthanide elements and cladding
- The reaction between U-PU-Zr and cladding (Fe)

Characteristics of the wastage layer were clarified.

- Time and temperature dependency of the growth ratio of the wastage layer formed by lanthanide elements.
- Threshold temperature of the liquid phase formation in the reaction between U-Pu-Zr and Fe.

These results are used
-as a basis for the FCCI modeling.
-as a reference data in post-irradiation examination of irradiated metallic fuels.

Thank you for your attention.

Preparation of the lanthanide alloy (RE5)

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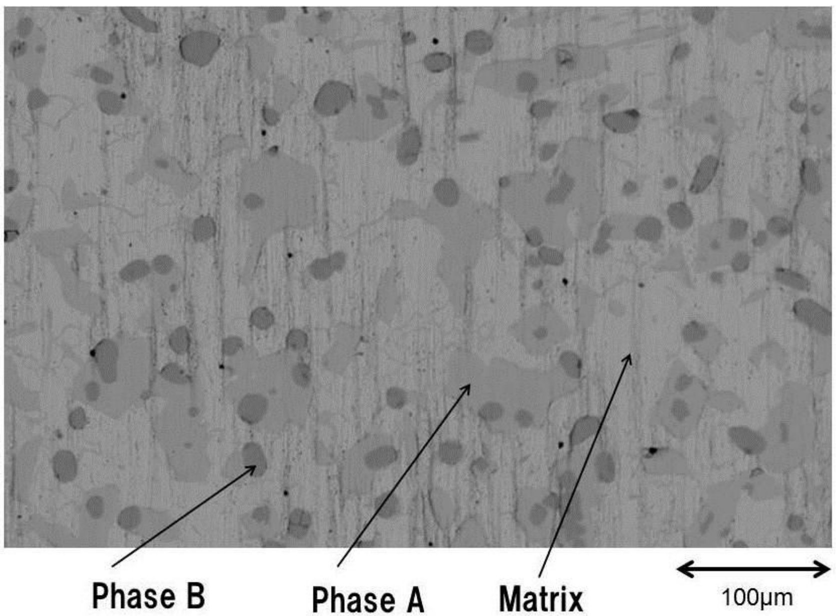
13wt.%La-24wt.%Ce-12wt.%Pr-39wt.%Nd-12wt.%Sm *

*The composition of the determined based on the fission yield of the lanthanide elements.

- After the annealing for homogenization, RE5 alloy had three-phase structure.



Back-scattered electron image of RE5 alloy after homogenization annealing.

The composition of the phases formed in RE5 [wt. %]

	La	Ce	Pr	Nd	Sm
Average	**13.0**	**24.0**	**12.0**	**39.0**	**12.0**
Matrix	16.7	23.2	13.3	36.7	10.0
Phase A	8.9	13.9	12.0	48.8	16.3
Phase B	2.5	10.9	9.4	48.0	29.1

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Cladding materials

- ODS-steel
  - Ferritic steel
  - Candidate material for FR cladding developed in Japan
- HT9
  - Ferritic/martensitic stainless alloy
  - Irradiation experience in EBR-2 In U.S.
- PNC-FMS
  - Ferritic/martensitic stainless alloy
  - Candidate material for FR cladding developed in Japan
- Fe-12wt%Cr
  - Simulating alloy for the cladding materials

Table : Compositions of the cladding materials

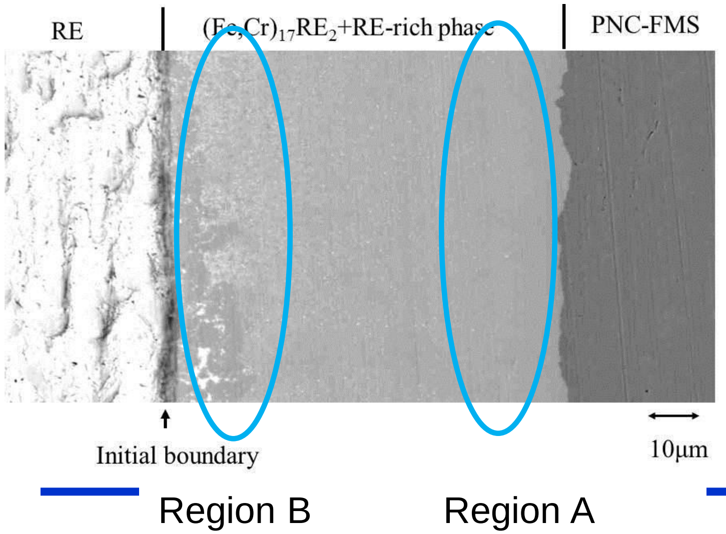
	C	Si	Mn	P	S	Ni	Cr	Mo	W	Ti	Nb	V	Y_2O_3	Fe
ODS-steel	0.13	<0.1	<0.1	<0.1	<0.1	<0.1	9.0	-	1.95	0.21	-	-	0.36	Bal.
PNC-FMS	0.1	<0.1	0.5	<0.1	<0.1	<0.1	11.1	0.5	2.1	-	<0.1	0.19	-	Bal.
HT9	0.2	0.2	0.2	-	-	0.6	12.0	1.0	0.5	-	-	0.3	-	Bal.
Fe-12wt%Cr	-	-	-	-	-	-	12.0	-	-	-	-	-	-	Bal.

Difference of the behavior of each rare-earth element

Weight ratio of rare-earth elements contained in the wastage layer. [wt.%]

	Region	Phase	La	Ce	Pr	Nd	Sm
RE5/ODS-steel	A	$(Fe,Cr)_{17}RE_2$	1.4	40.2	4.4	41.0	13.0
	B	$(Fe,Cr)_{17}RE_2$ + RE-rich	3.4	25.7	5.0	51.2	14.7
RE5/PNC-FMS	A	$(Fe,Cr)_{17}RE_2$	2.5	35.1	5.6	32.6	24.2
	B	$(Fe,Cr)_{17}RE_2$+ RE-rich	4.0	30.3	6.6	34.5	24.6
RE5 (Initial composition)			13.0	24.0	12.0	39.0	12.0

*Region A: Near the unreacted ODS-steel or PNC-FMS in the wastage layer
Region B: Near the initial interface in the wastage layer



Relative reactivity of the rare earth elements with Fe:

Ce > Nd, Sm > La, Pr

16Pu 963K

Area	Colour of the phase	Composition ratio [at.%]				Identified phase
		Fe	Zr	U	Pu	
a	(single phase)	71.5	6.5	20.2	1.8	$(U,Zr,Pu)Fe_2$
b	(multi-phase)*	63.7	7.8	24.7	3.8	$(U,Zr,Pu)_6Fe+(U,Zr,Pu)Fe_2$
c	bright*	51.8	11.0	31.6	5.6	$(U,Zr,Pu)_6Fe$+Liquid
c	dark	67.9	13.6	16.4	2.1	$(Zr,U,Pu)Fe_2+(U,Zr,Pu)Fe_2$
d	bright	19.3	4.2	64.1	12.4	$(U,Pu,Zr)_6Fe$+Liquid
d	dark	65.9	19.1	13.0	2.0	$(Zr,U,Pu)Fe_2$
e	bright	19.8	3.5	69.1	7.6	$(U,Pu,Zr)_6Fe$+Liquid
e	dark	72.4	16.9	9.8	0.9	$(Zr,U,Pu)Fe_2$
f	bright	23.4	4.0	65.6	7.6	$(U,Pu,Zr)_6Fe$
f	dark-1*	29.9	15.7	46.6	7.9	χ
f	dark-2	34.9	35.9	25.4	3.8	ε
g	bright	0.9	14.9	71.9	12.2	bcc
g	dark	8.9	22.1	57.1	11.9	λ

16Pu 943K

Area	Colour of the phase	Composition ratio [at.%]				Identified phase
		Fe	Zr	U	Pu	
a	(single phase)	73.1	3.5	21.9	1.5	$(U,Zr,Pu)Fe_2$
b	(single phase)	67.5	20.2	11.2	1.1	$(Zr,U,Pu)Fe_2$
c	(multi-phase)*	39.1	13.9	43.0	4.0	$(U,Zr,Pu)_6Fe+(Zr,U,Pu)Fe_2$
d	(multi-phase)*	25.6	11.6	57.6	5.3	$(U,Zr,Pu)_6Fe+\chi$
e	bright	0.6	12.5	74.0	12.9	bcc
e	dark	7.2	22.0	58.2	12.6	λ

9Pu 983K

Area	Colour of the phase	Composition ratio [at.%]				Identified phase
		Fe	Zr	U	Pu	
a	(single phase)	71.5	6.4	21.1	1.0	$(U,Zr,Pu)Fe_2$
b	bright*	49.3	6.8	39.4	4.5	$(U,Zr,Pu)_6Fe$
b	dark	66.5	9.6	22.1	1.8	$(U,Zr,Pu)Fe_2$
c	bright*	46.4	8.5	40.1	5.0	$(U,Zr,Pu)_6Fe$+Liquid
c	dark-1	65.9	13.7	18.0	2.4	$(U,Zr,Pu)Fe_2$
c	dark-2(granular)	44.2	27.3	24.8	3.7	χ
d	bright	34.4	6.1	53.3	6.2	$(U,Zr,Pu)_6Fe$+Liquid
d	dark	68.3	20.6	10.1	1.0	$(Zr,U,Pu)Fe_2$
e	bright	21.3	4.3	70.0	4.4	$(U,Zr,Pu)_6Fe$+Liquid
f	(multi-phase)*	59.0	16.6	22.1	2.2	$(U,Zr,Pu)_6Fe$ + $(Zr,U,Pu)Fe_2$
g	bright	-	-	-	-	$(U,Zr,Pu)_6Fe$
g	dark-1*	34.7	19.2	43.0	3.0	χ
g	dark-2*	26.7	21.0	48.0	4.3	ε
h	bright	0.7	15.1	74.2	10.0	bcc
h	dark	8.8	22.5	60.1	8.6	λ

9Pu 963K

Area	Colour of the phase	Composition ratio [at.%]				Identified phase
		Fe	Zr	U	Pu	
a	(single phase)	70.4	5.2	23.4	0.9	$(U,Zr,Pu)Fe_2$
b	(single phase)*	68.3	15.4	15.8	0.4	$(Zr,U,Pu)Fe_2$
c	(multi-phase)*	31.2	13.9	52.6	2.3	$(U,Zr,Pu)_6Fe+\chi$
d	(single phase)*	10.9	19.7	64.2	5.1	λ
e	bright	0.7	16.2	76.9	6.2	bcc
e	dark	4.6	18.2	70.6	6.6	λ

Assessed isothermal sections at 923K