

Overview of Post-Irradiation Examination results on Minor Actinide bearing fuels within the FUTURIX-FTA international collaboration

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14th International Exchange Meeting on Partitioning & Transmutation

Overview of Post-Irradiation Examination results on Minor Actinide bearing fuels within the FUTURIX-FTA international collaboration

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Overview

- 1. The Joint Research Centre Directorate G
- 2. The FUTURIX-FTA Irradiation Experiment
- 3. Fuel Preparation (FX 5 & 6) / Fuel Characteristics
- 4. Irradiation Conditions
- 5. Post Irradiation Examination of CerCer & CerMet Fuels
- 6. Summary



The Joint Research Centre (JRC)



The JRC is the European Commission's science and knowledge service.

JRC has 10 Directorates, is located across 5 EU Member States and employs over 3000 people.

JRC's nuclear work programme, funded by EURATOM is carried out by Directorate G – Nuclear Safety and Security, which employs more than 500 people at sites in Karlsruhe (former ITU), Petten, Geel and Ispra (red circles).

Directorate G contributes to the scientific foundation for the protection of the European citizen against risks associated with the handling and storage of highly radioactive material.



The FUTURIX-FTA Irradiation Experiment

- **Title:** FUels for Transmutation of transURanium elements in phenIX / Fortes Teneurs en Actinides [high actinide content]
- Objectives: Comparison of irradiation behavior of oxides, nitrides & metallic MA-bearing fuels (U-free fuels for ADS and metallic low fertile fuels for SFR burner) in representative conditions -> 8 pins irradiated in PHÉNIX
- Participants: CEA, DOE, JAEA & JRC-Karlsruhe through bilateral agreements, FP-6 EUROTRANS (2005-2010) & FP-7 FAIRFUELS (2009-2015)
- Key dates:

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- Experimental grid: 2003
- Transportations of fresh fuels from JRC-Karlsruhe and INL to ATALANTE facility: 2006
- Irradiation in PHÉNIX: May 2007 till March 2009 for 235 EFPD (except nitrides)
- Pins transportations from PHÉNIX to Cadarache (June 2013), INL (summer 2014), JRC-Karlsruhe (February 2015)
- PIE for JRC-Karlsruhe & CEA pins firstly in FP-7 FAIRFUELS and in national programs since mid.
 2015



FUTURIX-FTA Fuels

Pin	Fuel type	Composition ^[1]	Am content
DOE 1	Metal / low-fertile	35U-29Pu-4Am-2Np-30Zr (wt %)	0.5 g/cm ³
DOE 2	Metal / U-free	U 48Pu-12Am-40Zr (wt %)	1.2 g/cm³
DOE 3	Nitride / low-fertile	(U _{0.5} ,Pu _{0.25} ,Am _{0.15} ,Np _{0.10})N	1.7 g/cm³
DOE 4	Nitride / U-free	(Pu _{0.21} ,Am _{0.21} ,Zr _{0.58})N	2.7 g/cm ³
ITU 5	Cer-Met / U-free	(Pu _{0.8} ,Am _{0.2})O ₂ + 86 vol% Mo	0.3 g/cm³
ITU 6	Cer-Met / U-free	(Pu _{0.23} ,Am _{0.25} ,Zr _{0.52})O ₂ + 60 vol% Mo	1.0 g/cm³
CEA 7	Cer-Cer / U-free	(Pu _{0.5} ,Am _{0.5})O ₂ + 80 vol% MgO	1.0 g/cm³
CEA 8	Cer-Cer / U-free	(Pu _{0.2} ,Am _{0.8})O ₂ + 75 vol% MgO	2.0 g/cm ³

PIE on CEA 7 & 8 was already presented at Global 2015^[2]. PIE results on pins ITU 5 & 6 are newly presented^[3].



Fuel Preparation – FX 5 & 6



- Particle synthesis by external gelation
- Infiltration of Am into porous (Pu,Zr)O_{2-x} & PuO_{2-x} particles
- Mixing of fuel particles and Molybdenum matrix by conventional powder blending
- Bi-axial pressing (500 MPa)
- Sintering (1600°C, 8h)



Visual Aspect of ITU 5 & 6



ITU 5 86% Mo



ITU 6 60% Mo



Ceramographs



ITU 5 86% Mo



ITU 6 60% Mo



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Fuel Preparation – FX 7 & 8





- Preparation in shielded cells (ATALANTE facility)
- Synthesis of (Pu,Am)O_{2-x} particles by oxalic coconversion
- Particles dispersion into inert magnesia matrix by conventional powder metallurgy process
- Sintering



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Fuel Characteristics

Fuel ID	ITU-5	ITU-6	CEA 7	CEA 8
Pu content (wt%)	10.17	9.40	19.9	9.1
Am content (wt%)	2.68	10.33	19.6	35.6
Mean diameter (mm)	5.18	5.19	5.19	~5.14
Average Height (mm)	6.00	5.80	6.66	6.49
Density (g.cm ⁻³)	10.37	9.50	4.88	5.12
Total porosity (%)	4.1	4.8	4.5	7.5
Open porosity (%)	0.5-0.6	0.7-0.8	0.3*	0.4*
O/M (particle)	1.896	1.786	1.88	1.73
Particle size (µm)	< 225	< 225	< 50	< 50
Particle fraction	14 (volume %)	40 (volume %)	20 (volume %)	25 (volume %)



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*estimated

FUTURIX CerMet & CerCer Fuel Pins





CerMet ITU 5&6

CerCer CEA 7&8



 UO_2 with 7% enr.

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FUTURIX-FTA Position in Phénix Core



FUTURIX-FTA Irradiation Conditions

Pin	Burn-up (at%)	Transmu- tation rate (at%)	Linear Heat Rate (W/cm)	Neutron fluence	Dose (dpa)
ITU 5	17.81	27.0	122 - 133	14.59	20 50
ITU 6	12.88	27.1	124 - 135	x 10 ²⁶ n/m ²	30.30
CEA 7	8.6	17.6	82 - 96	9.33 x 10 ²⁶ n/m ²	
CEA 8	5.75	19.8	82 - 87		28.69



Power Evolution ITU 5 & 6





Power Evolution CEA 7 & 8





Post Irradiation Examinations

- CerMet Fuel (ITU 5 & 6): JRC-ITU hot cells
 - Pin length & profilometry ✓
 - Gamma spectrometry ✓
 - Gas puncturing ✓
 - Ceramography ✓
 - Pellet Metrology and Hydrostatic Density (ongoing)
 - SEM, EPMA (to be done)
 - LaserFlash (to be done)
- CerCer Fuel (CEA 7 & 8): LECA facility (CEA-Cadarache)
 - Pin length & profilometry ✓
 - Gamma spectrometry ✓
 - Gas puncturing ✓
 - Pellet Metrology and Hydrostatic Density \checkmark
 - Ceramography ✓

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• SEM, EPMA (to be done)

Pin Length Measurement & Profilometry showing no significant changes

FUTURIX-FTA Pins:	ITU-5	ITU-6	CEA-7	CEA-8
Length before irradiation (mm):	1793	1793	1793	1793
Length after irradiation (mm):	1793.0	1793.1	1792.2	1792.0

FUTURIX-FTA Pins:	ITU-5	ITU-6	CEA-7	CEA-8
Maximum diameter at experimental column after irradiation (mm)	6.571	6.558	6.562	6.557
Change in % (relative to 6.55 mm)	0.3	0.1	0.2	0.1



Profilometry ITU-5



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Gamma spectrometry



Gamma Spectrometry of ITU-5 (137Cs)



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Gamma Spectrometry of ITU 5&6 (137Cs)





Gamma Spectrometry of ITU-5 (⁵⁴Mn)



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Gamma Spectrometry of ITU 5&6 (241Am)



Gamma Spectrometry of CEA 7 (¹³⁷Cs (red), ⁵⁴Mn (green) and ⁶⁰Co (blue))





Gamma spectrometry of CEA 8 (¹³⁷Cs (red), ⁵⁴Mn (green) and ⁶⁰Co (blue))





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Comparison of the ¹³⁷Cs distribution between pins CEA 7 (blue) and CEA 8 (red)





Determination of Pellet Positions ITU-5



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Estimation of Pellet Elongation from Gamma Spectrometry for ITU-5

Pellet No.	Measured length (mm)	Original length (mm)	Change (%)
1	6.28	6.15	2.05
2	6.28	5.89	6.55
3	6.21	6.04	2.80
4	6.41	6.07	5.59
5	6.41	6.16	4.05
6	6.48	6.04	7.22
7	6.41	6.14	4.39
8	6.08	5.7	6.59
9	5.67	5.5	3.18
10	5.94	5.62	5.73
11	6.54	6.2	5.53
12	6.68	6.24	6.99
13	6.28	6.04	3.91
14	6.41	6.17	3.88
15	6.28	6.01	4.42
16	6.34	6.04	5.01
17	6.34	6.06	4.66
Total column:	107.02	102.07	4.85



Estimation of Pellet Swelling from Gamma Spectrometry

Pin ID:	ITU 5	ITU 6	CEA 7	CEA 8
Fuel column length before irradiation (mm):	102.07	98.68	99.21	97.35
Fuel column length after irradiation (mm):	107.02	101.70	100.1	98.0
Relative elongation (%):	4.85	3.06	0.9	0.7
Equivalent to volume swelling of (%)*:	15.3	9.5	2.7	2.1

Results indicate that there might have been no gap closure (together with profilometry).



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* isotropic swelling assumed

Gas Puncturing Results Low Fractional Releases in Both Pins

		He(mol)	Kr(mol)	Xe(mol)	Total (mol)
TTU 6	produced	4.68x10 ⁻⁴	7.00x10 ⁻⁵	7.32x10 ⁻⁴	1.27x10 ⁻³
110-5	FR (%)	43.23	33.40	22.44	31.06
	produced	1.55x10 ⁻³	6.95x10 ⁻⁵	7.20x10 ⁻⁴	2.34x10 ⁻³
110-6	FR (%)	35.87	43.17	21.67	30.10
CEA_7	produced	1.20x10 ⁻³	4.93x10 ⁻⁵	5.04x10 ⁻⁴	1.75x10 ⁻³
CEA-7	FR (%)	~0	0.75		<1
CEV-8	produced	2.32x10 ⁻³	4.42x10 ⁻⁵	4.25x10 ⁻⁴	2.79x10 ⁻³
CEA-0	FR (%)	~0	0.	22	<1



Recovery of CerMet Pellets

- All pellets were found loose inside pin, no indication of gap closure
- All pellets were completely intact and could be extracted
- Confirmation of gamma spectrometry





Ceramography of ITU 5 (Pu_{0.8}Am_{0.2}O_{1.9} + 86 vol% Mo)

Fresh fuel

Irradiated fuel





No cracks in Mo matrix

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Particle at pellet boundary formed open porosity -> gas release



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Ceramography of ITU 5 (Pu_{0.8}Am_{0.2}O_{1.9} + 86 vol% Mo)

Fresh fuel



- No cracks in Mo matrix
- Metallic precipitates in particles
- Gas bubbles mainly located in particles
- "Finger"-like particle-matrix transition
- Halo around particles

Irradiated fuel





Ceramography of ITU 6 (Pu_{0.23}Am_{0.2}Zr_{0.52}O_{1.8} + 60 vol% Mo)

Fresh fuel

Irradiated fuel





• No cracks in Mo matrix

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Particle at pellet boundary formed open porosity -> gas release



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Ceramography of ITU 6

Fresh fuel

Irradiated fuel



- No cracks in Mo matrix
- Metallic precipitates in particles
- Gas bubbles mainly located in particles
- "Finger"-like particle-matrix transition
- Halo around particles





Recovery of CerCer Pellets

- Pellets were found loose inside pin, no indication of gap closure
- CEA 7
 - 8 of 15 pellets recovered in one piece

MgO pellet being extracted (a) and F17 spacer (b), from pin CEA-8 $\,$





- CEA 8
 - 5 of 15 recovered in one piece
- Other pellets were extracted in relatively large fragments

extracted Am-bearing CERCER fuels from CEA-7







Pellet Metrology of CEA 7 & 8

Target #	∆ Height (%)	Δ Diameter (%)	∆ Geometrical density (%)				
	Futurix FTA 7						
5114	0,7	1,5	-3,6				
5119	0,6	1,3	-3,1				
5120	0,6	1,5	-3,4				
5121	1,2	1,2	-3,6				
5126	0,8	1,3	-3,3				
5127	0,7	0,9	-2,5				
5128	0,9	1,3	-3,4				
5129	0,7	1,1	-2,8				
	Futuri	c FTA 8					
2112	1,1	1,1	-3,2				
2113	0,9	1,2	-3,2				
2117	0,7	1,4	-3,5				
2124	0,8	0,7	-2,2				
2120	0,3	1,5	-3,2				

Consistent with results from gamma spectrometry, indicating circa 3% volume swelling.



Ceramography of CEA 7 (Pu_{0.5}Am_{0.5}O_{1.88} + 80vol% MgO)

No significant evolution of the diameter of the pellets along the cuts; rounded shaped bubbles inside $(Pu,Am)O_{2-x}$ particles that probably host gases.



- -> some cracks (consistent with broken pellets shapes)
- -> metallic precipitates

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- -> gas bubbles mainly located in particles (consistent with low gas release rate)
- -> secondary phase at MgO grain boudaries



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Ceramography of CEA 8 (Pu_{0.2}Am_{0.8}O_{1.73} + 75 vol% MgO)



few cracks

-> gas bubbles mainly located in particles (but features different from CEA-7)

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-> secondary phase at MgO grain boudaries ?

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Summary 1/2

- Within the frame of the international collaboration FUTURIX-FTA as well as the European research programmes EUROTRANS (FP6) and FAIRFUELS (FP7) CEA and JRC have conducted together preparation, irradiation and PIE of 2 CerCer and 2 CerMet fuels for transmutation of Am in the Phénix fast reactor.
- CEA has achieved promising PIE results regarding CerCer fuels based on MgO matrix (CEA 7 & CEA 8):
 - Non-significant elongations and deformation of the pins
 - No ¹³⁷Cs migration in the fuel stacks
 - Low Helium and Fission Gas release rates
 - A significant fraction of fuel pellets recovered as one piece
 - Moderate and similar pellet swelling (~3%) for both fuels (0.3-0.5 vol.%/at%)
 - Low hydrostatic density changes (~0,4%)



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Summary 2/2

- ITU has achieved promising PIE results regarding CerMet fuels based on Mo matrix (ITU 5 & ITU 6):
 - No significant pin length elongation
 - No significant pin diameter change at position of the CerMet columns
 - No or very low fission product migration
 - No or very low Cs release from CerMet pellets
 - Pellet length changes ~5% & ~3% for ITU 5 & 6, respectively
 - Volume swelling circa 15% & 9% (0.7-0.9 vol.%/at%)
 - A fractional gas release of circa 30 % was measured for both pins
 - All pellets were recovered completely intact
 - No cracks inside the Mo matrix, Bubbles located inside particles
 - Outer particles formed open porosity during irradiation (improve synthesis in the future)



Thank you for your attention!



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Back-up slides



Material Data

Mo

- Molar mass
- Density
- Thermal expansion
- Thermal conductivity
- Specific heat
- Melting point

MgO

- Molar mass
- Density
- Thermal expansion
- Thermal conductivity
- Specific heat
- Melting point

95.95 g/mol 10.28 g/cm3 4.8 µm/(m·K) (at 25 °C) 138 W/(m·K) 250 J/kg.K

2896 K (2623 °C, 4753 °F)

40.30 g/mol 3.58 g/cm3 9-12 µm/(m·K) 45 - 60 W/(m·K) 880 - 1030 J/kg.K 3125 K (2852 °C, 5166 °F)



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Table 1.- Impurity analysis of molybdenum metal

Element	Concentration	Eleme	nt Concentration µg/g
µg∕g			
Na	22.99 ± 2.76	Ce	3.91 ± 0.47
Mg	9.14 ± 1.10	Re	2.83 ± 0.34
AI	19.29 ± 2.32	W	80.80 ± 9.70
Rb	0.72 ± 0.09	Pb	20.22 ± 2.43
Y	0.12 ± 0.01	Bi	0.18 ± 0.02
Ru	2.78 ± 0.33	V	0.47 ± 0.06
Cd	1.19 ± 0.14	Cr	8.03 ± 0.96
Ag	0.17 ± 0.02	Mn	1.16 ± 0.14
Sb	0.67 ± 0.08	Fe	93.21 ± 11.19
Cs	0.89 ± 0.11	Со	0.43 ± 0.05
Ba	1.00 ± 0.12	Cu	3.90 ± 0.47
La	0.05 ± 0.01	к	30.03 ± 3.60

Table 2.- Isotopic composition of plutonium (analysis 08.08.2005)

TIMS	
²³⁸ Pu	0.01535
²³⁹ Pu	90.6340
²⁴⁰ Pu	9.11570
²⁴¹ Pu	0.16188
²⁴² Pu	0.07263

Table 3.- Particle size distribution of plutonium compounds following calcination.

Range µm	FX5	FX6
	(%wt)	(%wt)
>225	0.02	0.03
125-225	28.24	38.51
90-125	47.21	38.07
63-90	21.82	14.24
32-63	2.33	6.03
<32	0.38	3.11

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Table 1.-Am and Pu content in the beads

	FX5	FX6	
Beads	PuO ₂	(Zr,Pu)O ₂	
Analysis number	3458	3459	
Pu (wt%)	68.65 ± 0.89	28.42 ± 0.37	
Am (wt%)	17.59 ± 0.14	30.05 ± 0.24	
Zr (wt% ¹)		25.31	
Am/ΣM	20.4	35.9	



Profilometry ITU-6



Gamma Spectrometry of ITU-5 (⁶⁰CO)



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Gamma Spectrometry of ITU-5 (106Ru)



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Gamma Spectrometry of ITU-5 (125Sb)



Gamma Spectrometry of ITU-5 (154Eu)



Gamma Spectrometry of ITU-6 (137Cs)



Gamma Spectrometry of ITU-6 (¹³⁴Cs)



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Gamma Spectrometry of ITU-6 (⁶⁰Co)



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Gamma Spectrometry of ITU-6 (⁵⁴Mn)



Determination of Pellet Positions ITU-6



Gas Puncturing Results

Pin ID	ITU-5	ITU-6	CEA-7	CEA-8	
Date of puncturing	June 2015		February 2014		
Pressure at puncturing stage (MPa)	0.148	0.195	0.106	0.118	
Free volume in the pin(cm ³)	19.18	18.93	18.9±3.1	18.9±3.2	

Initial free volume: 18.6 ml

Pin	ITU-5	ITU-6	CEA-7	CEA-8
%H ₂	-	-	0.002	0.002
% He	83.23	87.18	98.942	99.336
% Xe	14.34	10.3	0.466	0.100
% Kr	2.04	2.02	0.033	0.008
% O ₂	0.03	0.04	0.102	0.065
% N ₂	0.11	0.12	0.425	0.330
% Ar	0.05	0.14	0.024	0.154
% CO ₂	0.19	0.21	0.006	0.005



The Joint Research Centre – Directorate G



Facilities and Equipment at JRC Karlsruhe

24 hot cells with capacities up to **1 Mio Curies** and some **400 glove boxes** in **30 alpha-laboratories.**

Minor Actinide Lab for preparing *irradiation targets* containing amounts of *actinides for transmutation* experiments

Clean Lab category 10-100-Large Geometry SIMS for safeguards and nuclear forensic applications

Destructive and non-destructive analyses

Basic materials science investigations

- Materials studies down to 1 K and up to 8000 K and 60 GPa
- Photo-electron spectroscopy, X-ray diffraction, (electrical resistance and optical properties) on highly radioactive samples, Mössbauer spectroscopy at normal and elevated pressures
- High-resolution electron microscopes,
- Shielded microprobes,



