Advanced Fuel Fabrication Processes for Transmutation

A. Fernandez-Carretero, C. Nästren, D. Staicu, J. Somers

Nuclear Fuels Unit

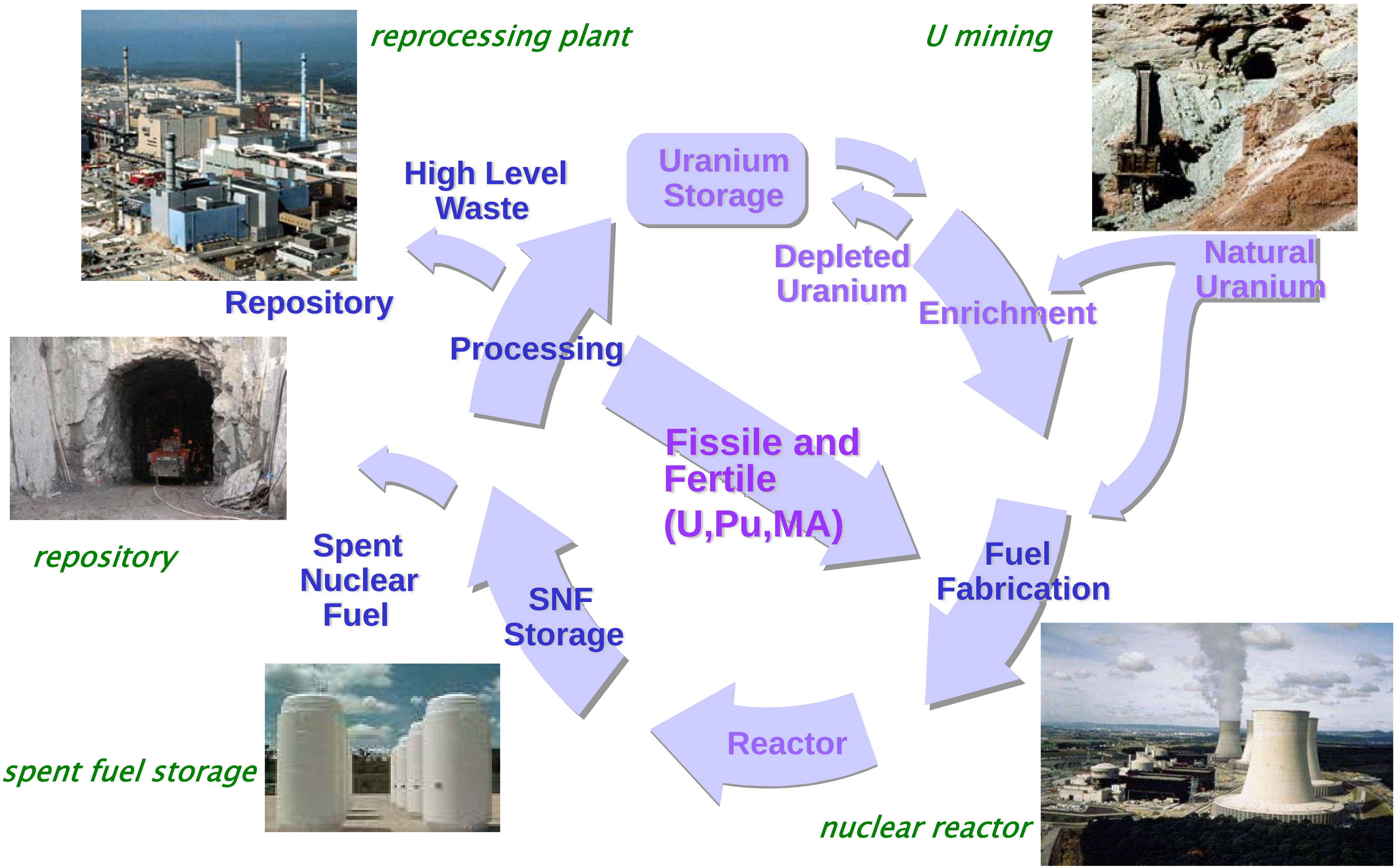
Institute for Transuranium Elements (ITU)

Karlsruhe, Germany

http://www.jrc.org/

http://itu.jrc.cec.eu.int/

The Nuclear Fuel Cycle



Outline

Introduction

Difficulties in Fabricating MA fuels

Sol Gel Routes - SUPERFACT

Infiltration

CERMETS/CERCERS

Gen IV Oxides and Nitrides

Conclusions

Implications of MA on the fabrication process

- **Shielded** installations
  → remote handling
- **Automation** → use of robots
- **Dust-free** processes → avoid the use of fine powders that produce dust that accumulates in the production cells
- Process **simplification**
  → limit the number of (active) fabrication steps (e.g. vibrocompaction instead of pressing)

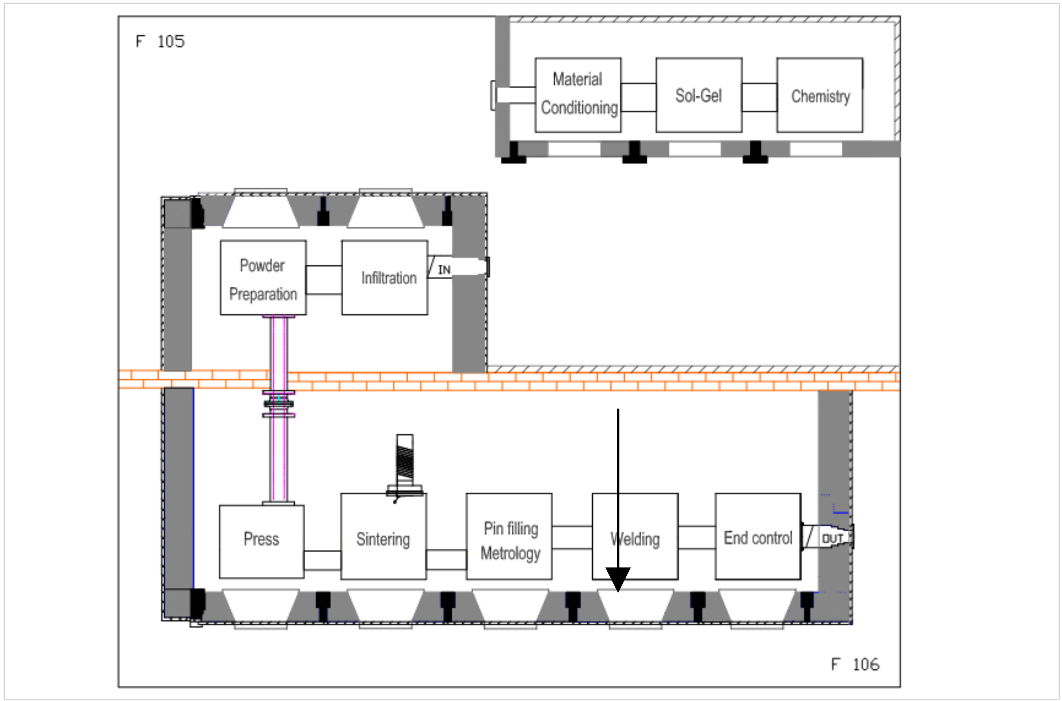
Nuclide	Specific Activity (Bq/g)	Alpha Energy (MeV)	Gamma Energy (keV)	SF
^{239}Pu	$2.29\ 10^{9}$	5.156	0.07	
^{237}Np	$2.610\ 10^{7}$	4.79	29.4	
^{241}Am	$1.271\ 10^{11}$	5.49	59.5	•
^{242m}Am	$3.598\ 10^{11}$	5.20	49.4	
^{243}Am	$7.391\ 10^{9}$	5.28	74.7	•
^{243}Cm	$1.911\ 10^{12}$	5.79	277.6	
^{244}Cm	$2.997\ 10^{12}$	5.80	42.8	•

Transmutation Fuels

Fabrication facility: MA LAB

Isotope	Limiting mass (g) [a]	Criterion
^{231}Pa	10	Shielding
^{237}Np	- [b]	
^{241}Am	50	License [c]
^{242m}Am	0.1	Shielding
^{243}Am	65	License [c]
^{244}Cm	5	Shielding & licence [c]

[a] to yield max 2 µSv/h at 1 metre
[b] no practical limit
[c] corresponding to the dosis equivalent of 200 g Pu in the form of powder (oxide)



Transmutation Fuels at the ITU MALAB

Since 2004

- 75 grams Am processed for fuel property and irradiation campaigns
  - CAMIX-COCHIX
  - FUTURE
  - FUTURIX
  - HELIOS

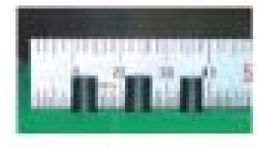
  - Am cross section targets (c.f. Poster V-4 P. Rullhusen)

Transmutation Fuels

Presentation focuses on oxides, but active programmes on

MA bearing metal fuels	CRIEPI/ITU; INL
MA bearing nitride fuels	JAEA; LANL

Transmutation Fuel Fabrication: Strategic choice at conversion step



$(U_{0.65}Pu_{0.30}Am_{00.5})O_2$ fuel

(Tanaka Global 2007)

Individual An Separation and Conversion
(e.g. today at Sellafield and La Hague)
U, Pu, Np, Am, Cm streams

Selected An Separation and Conversion
e.g. U, Pu/Np, Am/Cm Streams

Group An Conversion
Single U/Pu/Np/Am/Cm stream

POWDER Metallurgy
(LANL c.f. Pasemehmethogolu
JAEA c.f Tanaka / Kato))

Sol Gel/Infiltration
(ITU)

Ox Precipitation
(CEA c.f Poster III-1 Grandjean)
Sol Gel
(ITU)

Low dust routes

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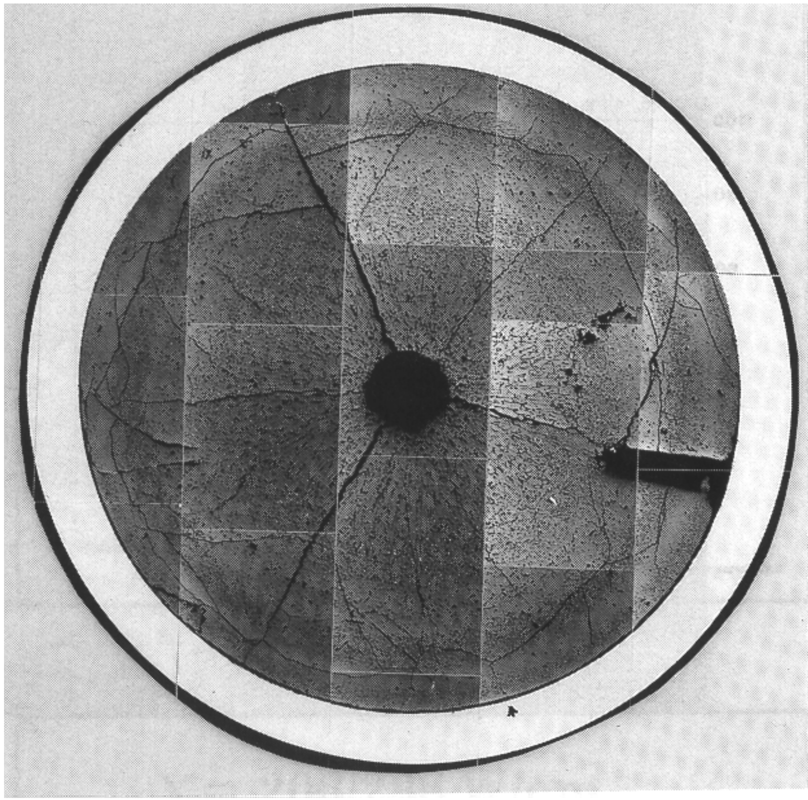
SUPERFACT – As yet unsurpassed irradiation test (CEA/ITU)

Sol Gel conversion of (U,Pu,Am,Np solutions



Typical observations for $(U_{0.74}Pu_{0.24}Am_{00.2})O_2$ fuel:

- Fuel restructuring similar to standard fuel irradiated under similar conditions
- U and Pu did not show significant radial re-distribution
- Nodular oxide layer (few tens of microns) on inner cladding
- Reprocessing demonstrated

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HELIOS FUELS FABRICATED USING Sol Gel /Infiltration processes

CEA (HELIOS 1); ITU (HELIOS 2–5)

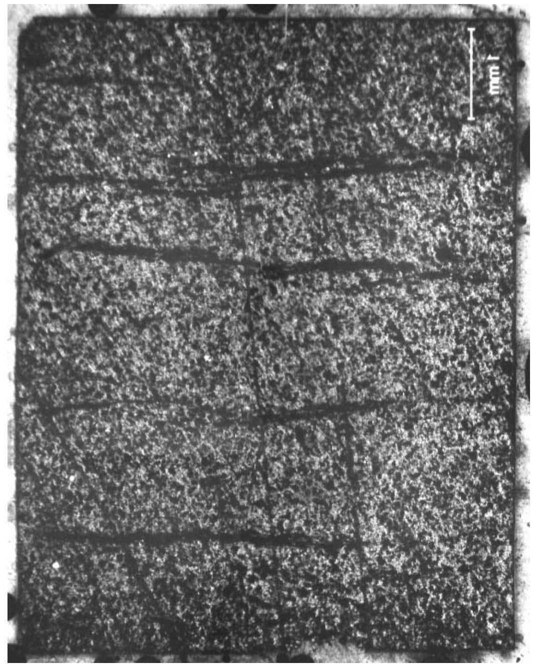
Fuel	Compound	Am content*	Pu content*	Particle size	Density
		$g \cdot cm^3$	$g \cdot cm^3$	µm	%TD
HELIOS 1	$Am_2Zr_2O_2$-MgO	0.76			90 ± 5
HELIOS 2	**$ZrYAmO_2$**	**0.76**			
HELIOS 3	**$ZrYPuAmO_2$**	**0.76**	**0.42**		
HELIOS 4	**$ZrYAmO_2$ + M**	**0.76**		**80-100**	
HELIOS 5	**$PuAmO_2$+ Mo**	**0.32**	**1.28**	**20-150**	

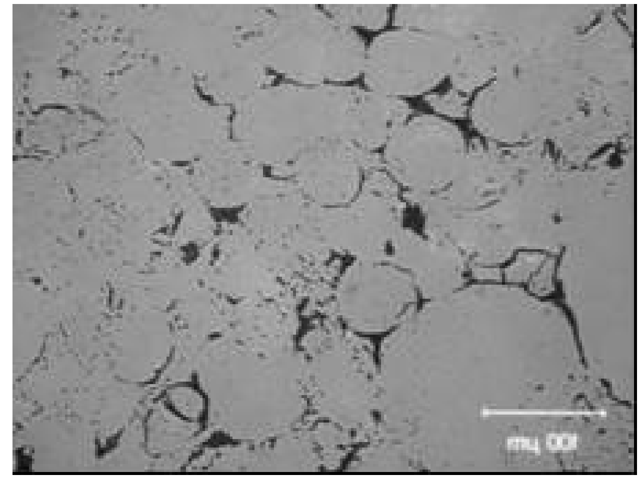
$(Zr,Y)O_2$
$(Zr,Y,Pu)O_2$
$(Zr,Y)O_2$
PuO_2

Sol Gel precursors (prepared in inactive or Pu facilities)

HELIOS FUELS INFILTRATION ROUTE OPTIMISATION

$YZrAmO_{2-x}$





Good visual aspect

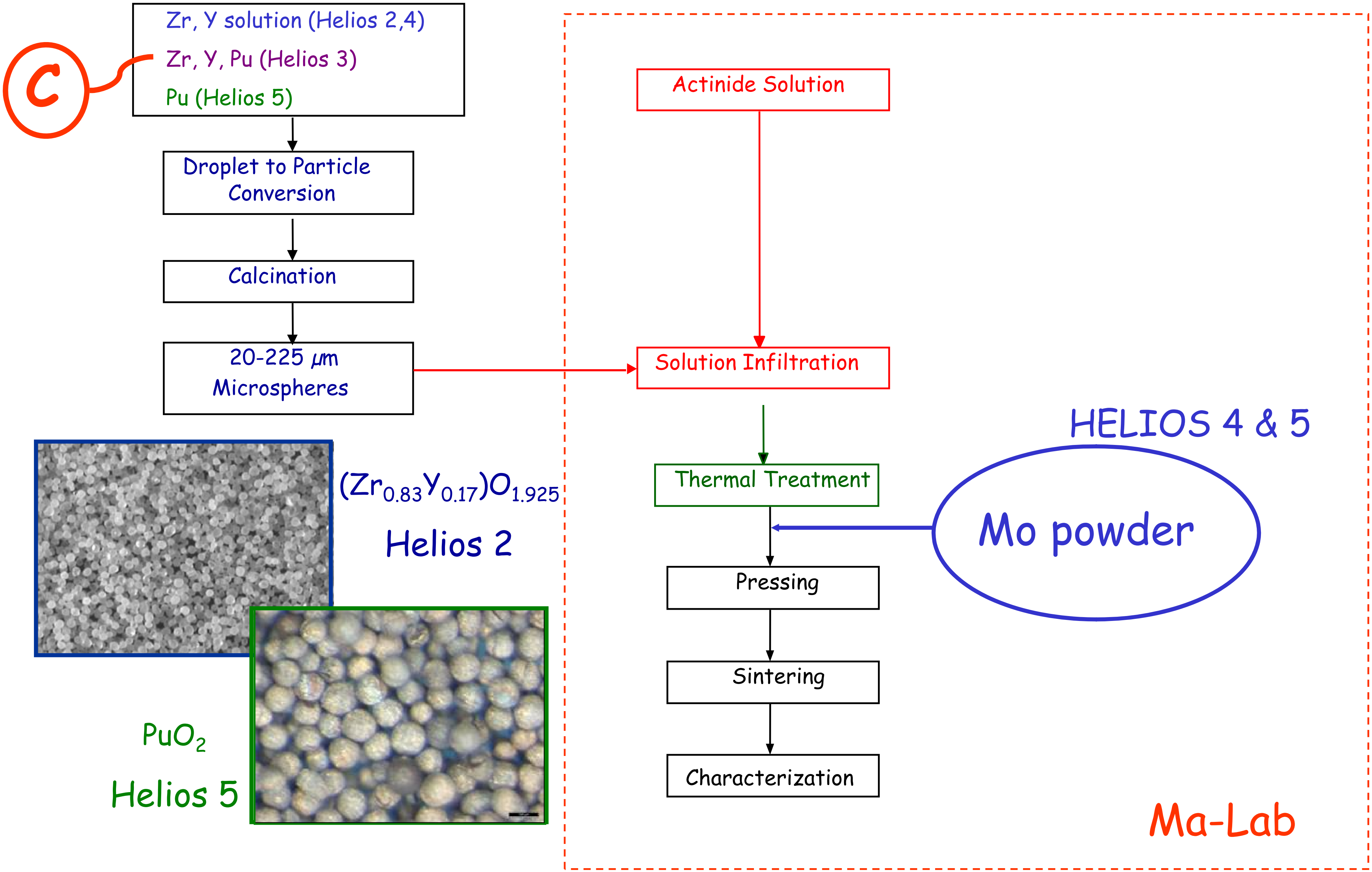
But microstructure→ cracks and large localised porosity

Carbon addition

improve microstructure

improve infiltration behaviour

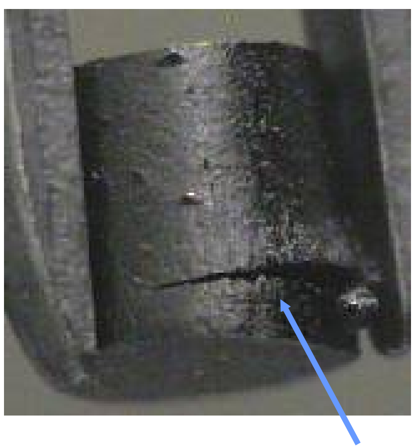
HELIOS FUELS: Fabrication Process Optimisation



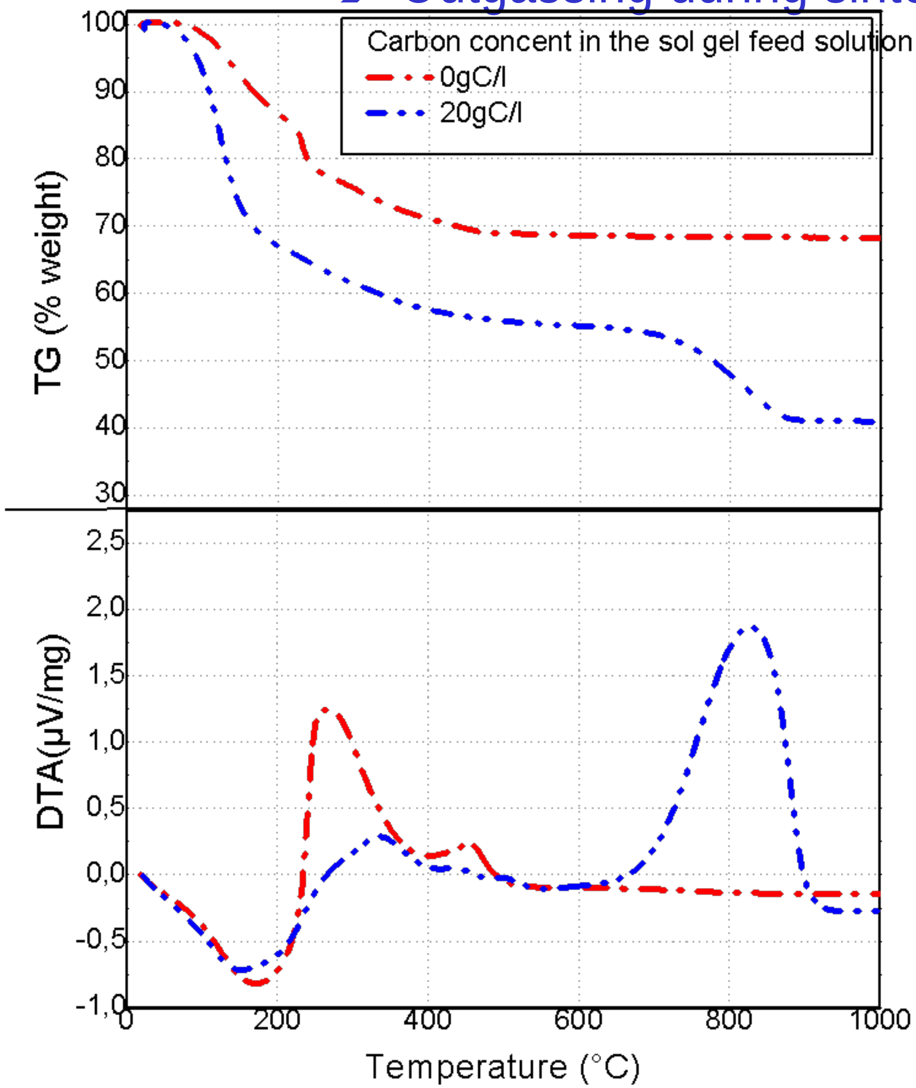
CARBON ADDITION: Surface spalling, higher than normal mass loss

➔ Outgassing during sintering (Ar/H_2)

Non uniform shrinkage



cracks

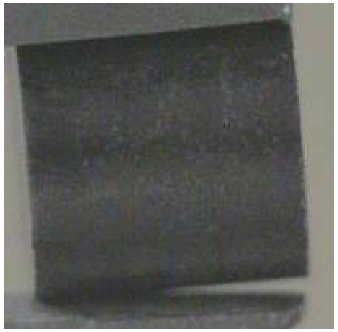


CH_4 or ???

Process modification

Calcine 800 C (air)
Infiltartion
Calcine 800C (air)
Pellet compaction
Heat 1000 C (air)
Sinter Ar/H_2

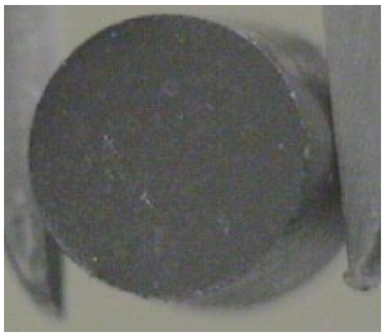
HELIOS FUELS INFILTRATION ROUTE OPTIMISATION



HELIOS 2

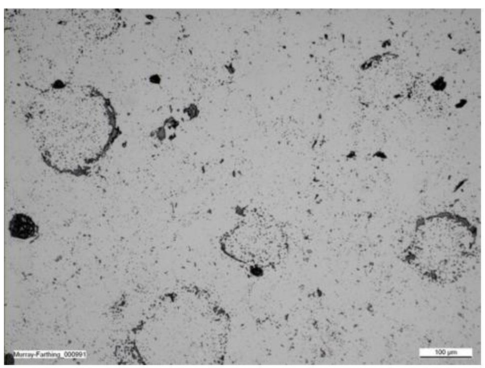
$Zr_{0.800}Y_{0.134}Am_{0.066}O_{2-x}$

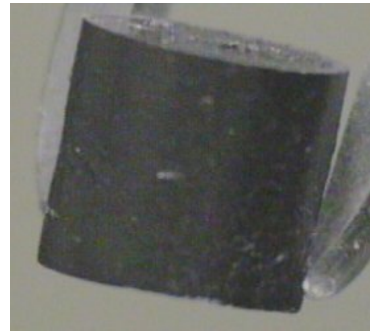
0.70 gAm•cm^{-3}



92.6 ± 1.2 %TD

90.9 ± 0.3 %TD



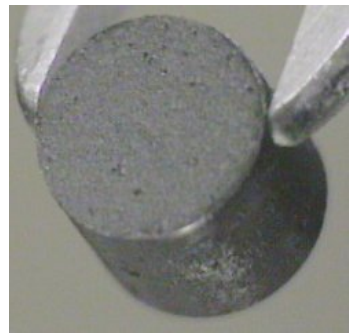


HELIOS 3

$Zr_{0.767}Y_{0.127}Pu_{0.038}Am_{0.068}O_{2-x}$

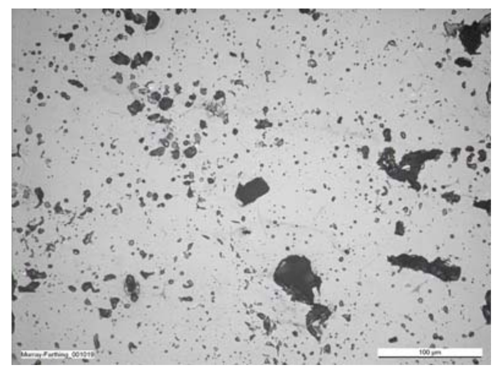
0.74 gAm•cm^{-3}

0.41 gPu•cm-3

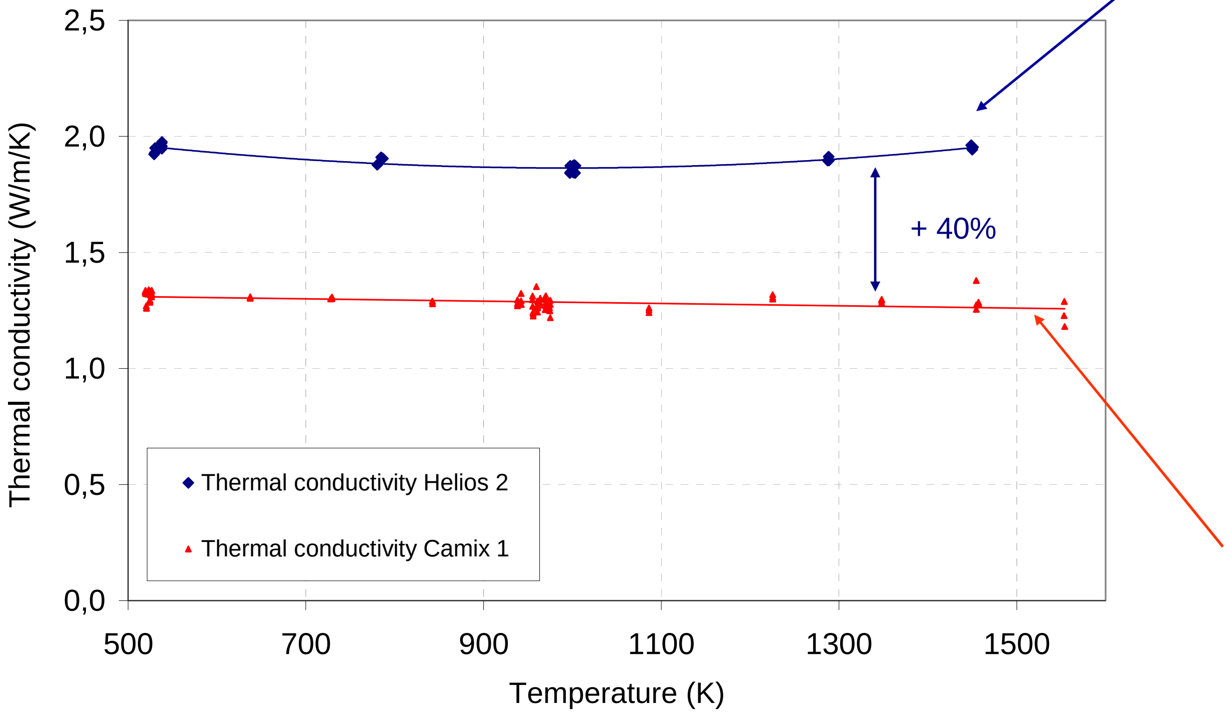


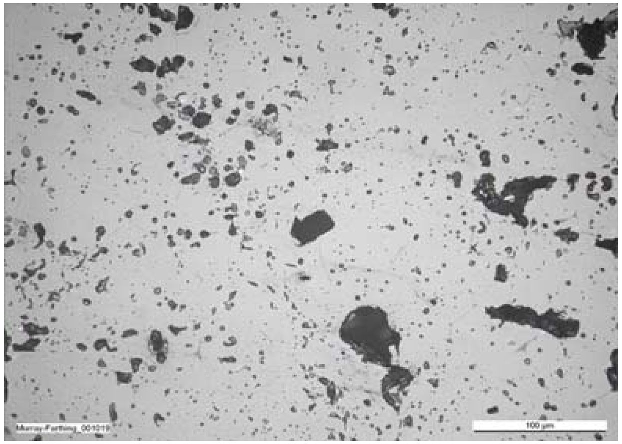
89.7± 0.4 %TD

89.1 ± 1.1 %TD

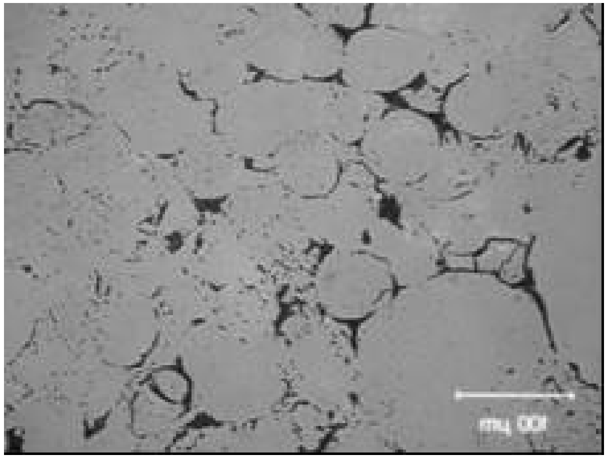


HELIOS FUELS: Characterization





HELIOS 2
Carbon addition

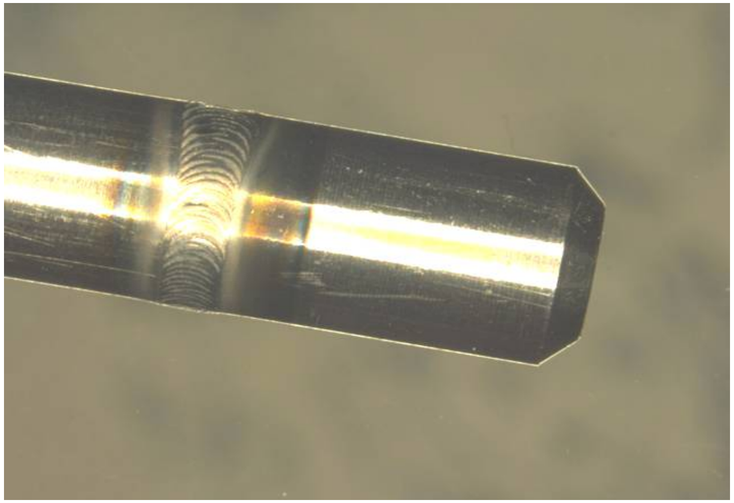


CAMIX 1

Carbon addition → improve microstructure → **Improve Thermal Conductivity**[1]

[1] measured by Dragos Staicu (MR)

HELIOS FUELS: Pin fabrication & Transport



Measured dose rate

	Contact (mSv/h)	1m (μSv/h)
1	175	105
2	66	42
3	58	43
4	36	22
5	12	6

New Design
Transport carousel
5 pins

Transport to HFR-Petten 11.10.2007

Beginning of Irradiation – October 2008

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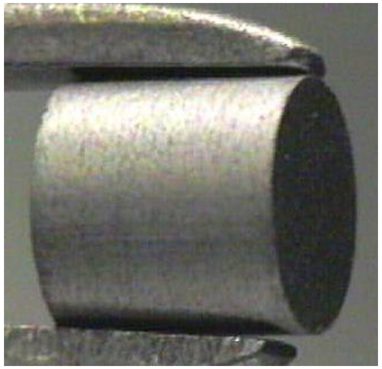
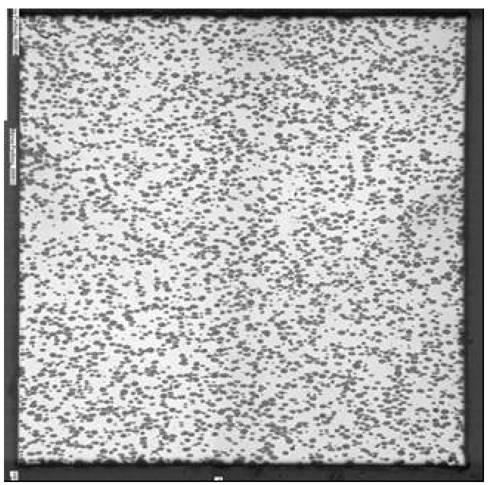
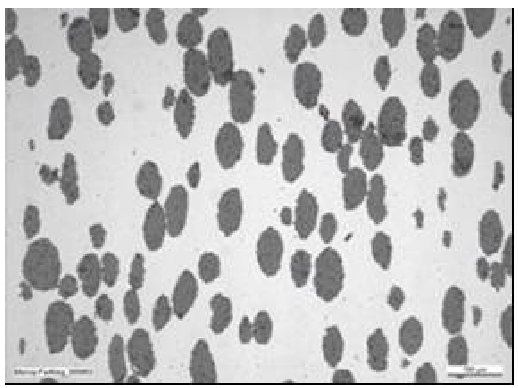



HELIOS 4

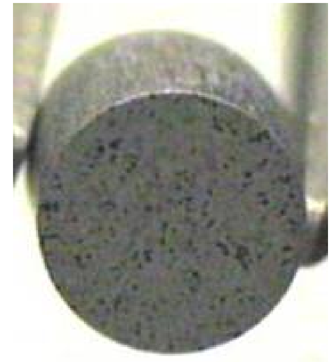
$Zr_{0.666}Y_{0.111}Am_{0.223}O_{2-x}$ + 71.3 %vol Mo

0.69 gAm•cm^{-3}

94.2 ± 0.4 %TD

HR = 4.83

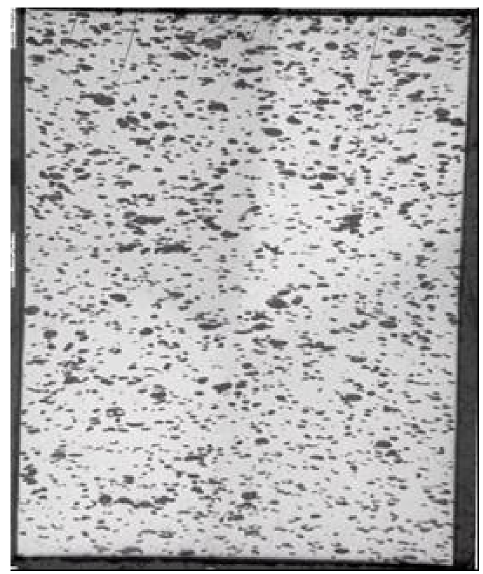
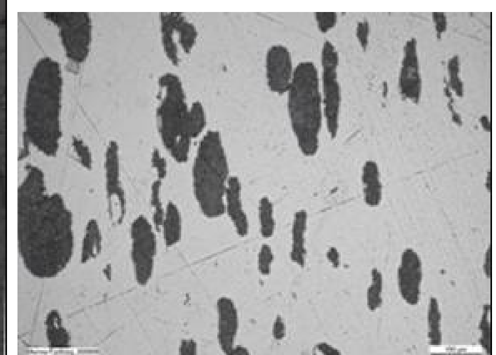



HELIOS 5

$Pu_{0.801}Am_{0.199}O_{2-x}$ + 84.2 %vol Mo

0.295 gAm•cm^{-3} , 1.24 gPu•cm^{-3}

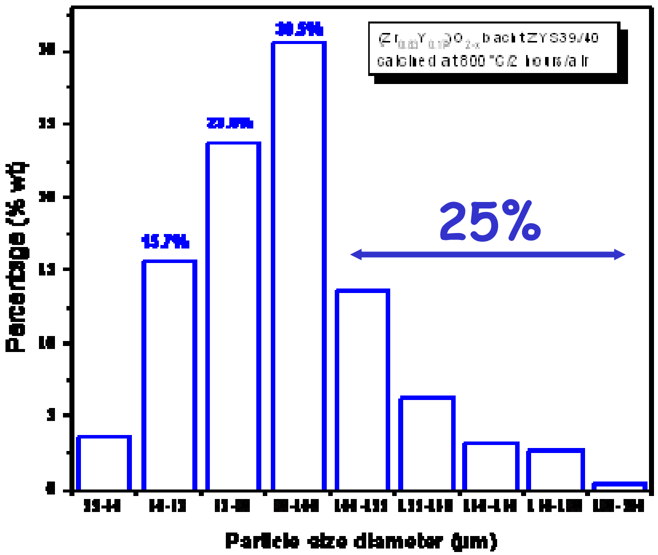
95.9 ± 0.4 %TD

HR = 12.01

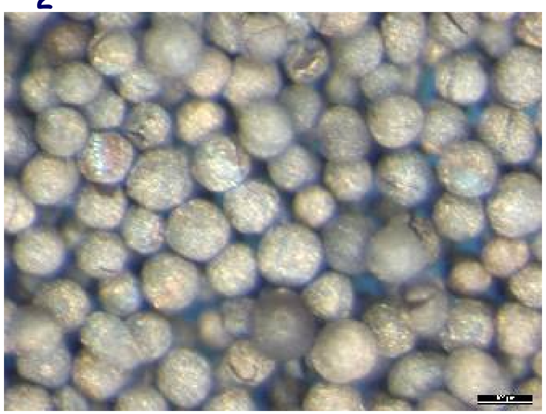
Fabrication of porous PuO_2 and $(Zr_{0,705}Pu_{0.295})O_2$ beads **(100-200µm)**

Conventional rotating cup

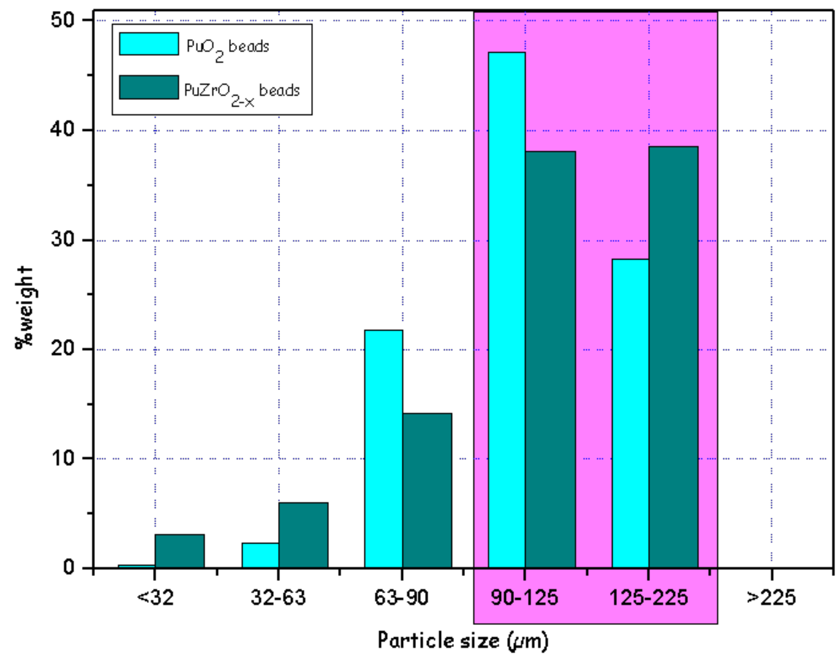


- Pu concentration
- Denitration
- Viscosity
- Rotating cup (rpm, height, etc)
- Polydisperse size distribution

PuO_2 beads



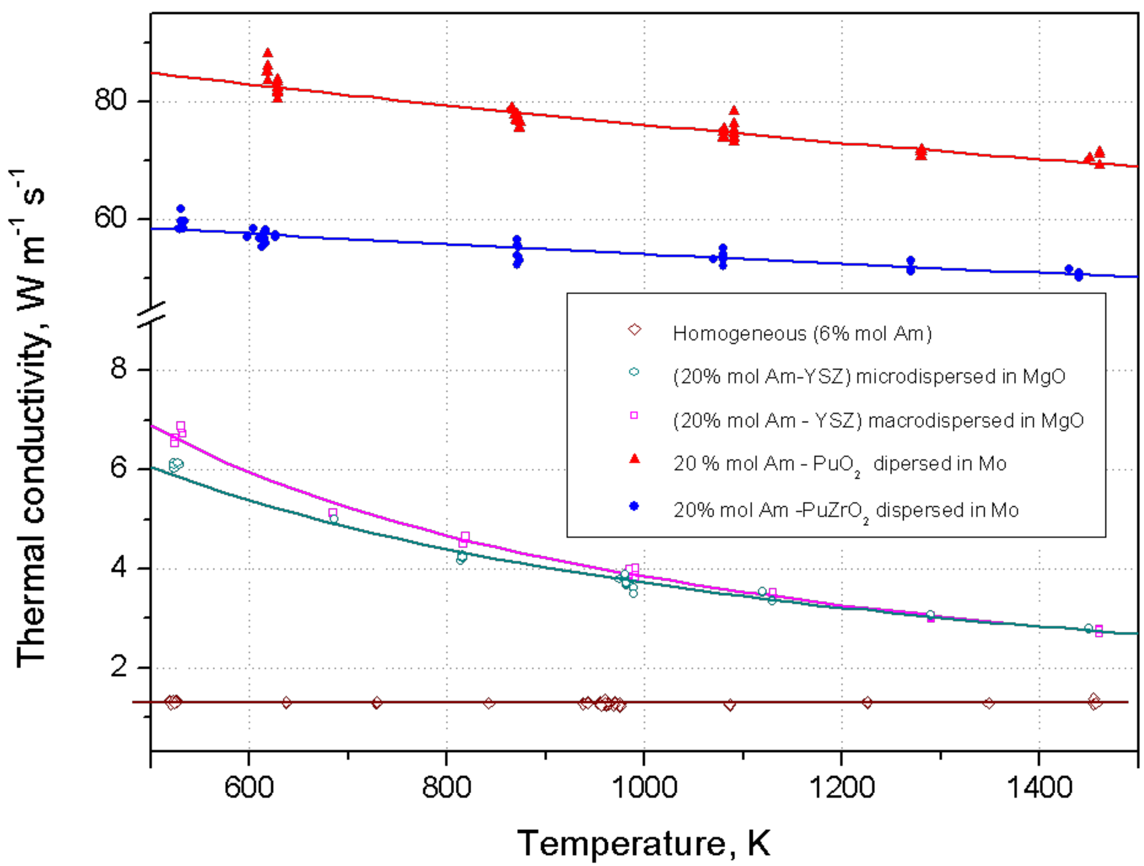
FUTURIX fuels



Thermal conductivity

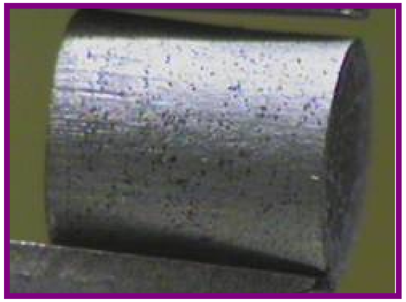
Calculated from measured thermal diffusivity and specific heat.

CERCER compared to CERMET

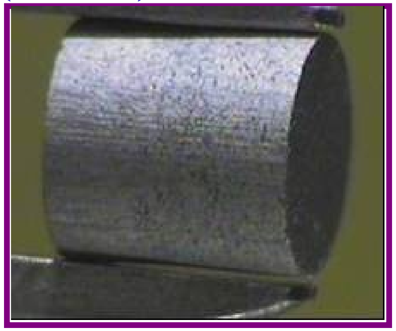


Compatibility test- T91

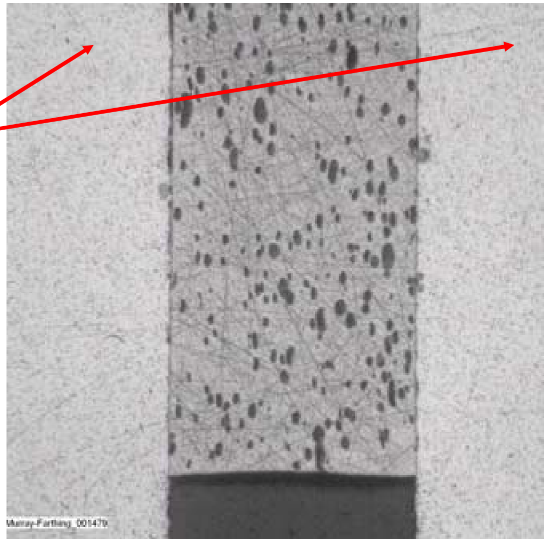
Mo- (Pu,Am)O_{2-x}
(FX 5)

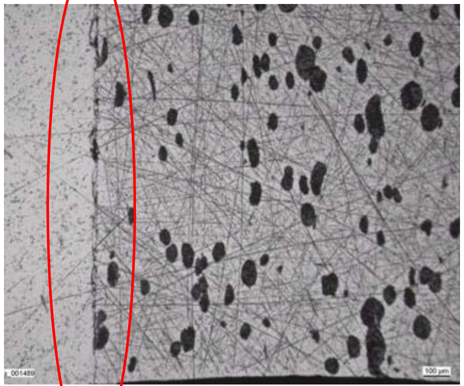


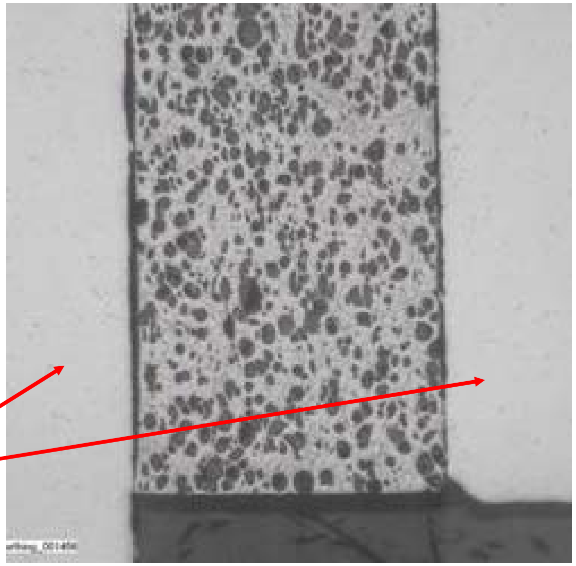
Mo- (Zr,Pu,Am)O_{2-x}
(FX 6)

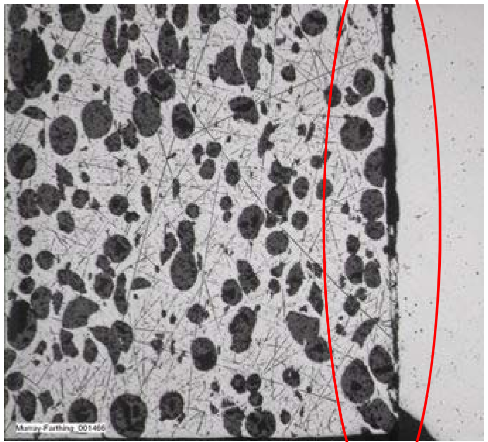


Cladding









Cladding

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Nitride/Carbide Fuel Fabrication

FR Nitride (Carbide) Fuel Production

Conventional Gloveboxes

U | Pu | C

External Gelation

Calcination

$(U,Pu)O_2 + C$

MALAB

Am Infiltration

Calcination

$(U,Pu)O_2 + AmO_2 + C$

$U_{0,805}Pu_{0,175}Am_{0,02}N$

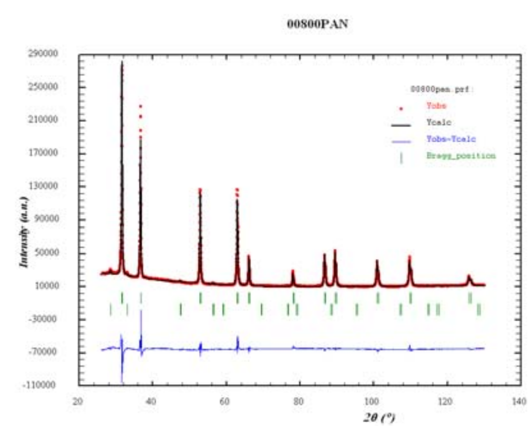
Conventional Gloveboxes with Purified Atmospheres

Carbothermal Reduction

Pressing

Sintering

(U,Pu,Am)N



Nitride/Carbide Fuel Fabrication

MA Production Losses : Particularly for carbides
but also known for nitrides (LANL)

Experience in NIMPHE2

NIMPHE Pu Isotopes	239	241	Am/(Pu+Am) % as starting material
Pu as delivered	74,6	2,62	0,365
Nitride 2 pin 3	74,7	2,573	0,436
Carbide 2 pin 4	74,7	2,58	0,112

Nitride 2 pin 3: seems ok

Carbide 2 pin 4: 75% Am Losses!!

Production Losses : Needs

Vapor Pressure determination of Am over (U,Pu,Am)C

New Lower Temperature Fabrication Routes

- Precursor to carbide or nitride directly
  - Pyrochemistry through azide precipitation from molten salt

- Alternatives to carbothermal reduction?

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Transmutation Fuel Fabrication:

Options today

Reprocessing: Aqueous or Pyro (potential for “and/or” especially of MC and MN)

Fabrication:

Separation strategy influences fabrication options

(Homogeneous vs heterogeneous recycling)

- Individual An Separation & Conversion

- Partial An Separation & Conversion

- Grouped An Separation & Conversion

Transmutation Fuel Fabrication:

Decisions must include plant concept

Dust or not
Production Scrap recycling
Primary and secondary waste issues

Powder metallurgy - Very flexible; dust

Grouped Oxalate precipitation - limited flexibility; dust

Sol Gel - flexible if partial separation; limited flexibility for grouped separation; no dust

Infiltration – Medium flexibility, partial separation needed, no dust

Microstructure attributes for Hi BU, He MGT, swelling…….

Outlook for the Future

Gen IV Fast Reactors and ADS

Demo with MOX cores

R&D for advanced MA bearing fuels

Oxide, nitride, carbide

Three Pillars for Fuel R&D recognised in the SNETP Strategic Research Agenda

1. Fabrication and basic properties of advanced fuels
2. Integral irradiation testing of fuel in appropriate advanced cladding materials
3. Separate effect studies and multiscale modelling approach

Goals to be reached via

National, European, Global research programmes

Fuels Research