

# *Advanced Fuel Fabrication Processes for Transmutation*



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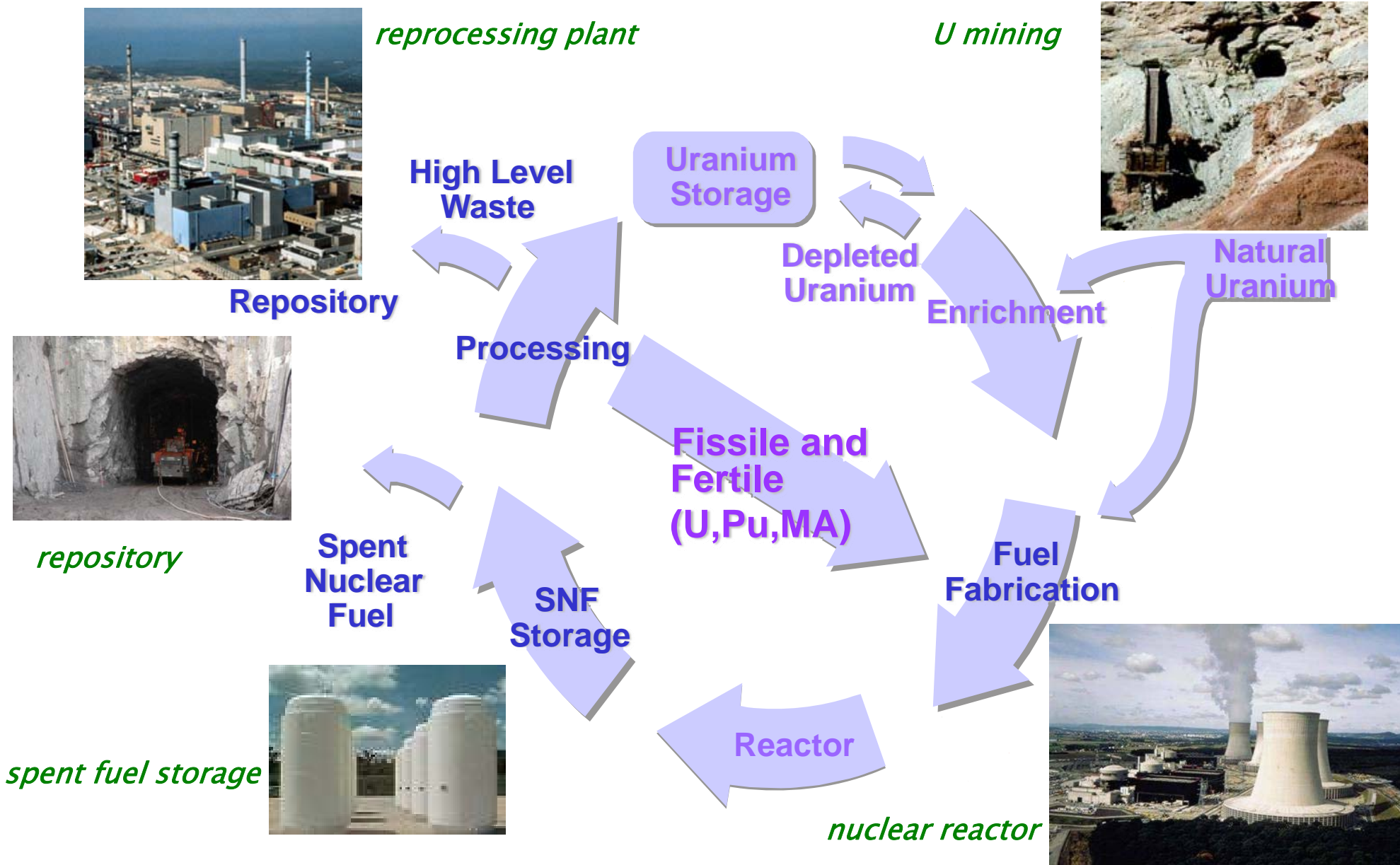
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<http://www.jrc.org/>

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

# The Nuclear Fuel Cycle



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
$^{239}\text{Pu}$	$2.29 \cdot 10^9$	5.156	0.07	
$^{237}\text{Np}$	$2.610 \cdot 10^7$	4.79	29.4	
$^{241}\text{Am}$	$1.271 \cdot 10^{11}$	5.49	59.5	•
$^{242\text{m}}\text{Am}$	$3.598 \cdot 10^{11}$	5.20	49.4	
$^{243}\text{Am}$	$7.391 \cdot 10^9$	5.28	74.7	•
$^{243}\text{Cm}$	$1.911 \cdot 10^{12}$	5.79	277.6	
$^{244}\text{Cm}$	$2.997 \cdot 10^{12}$	5.80	42.8	•

# Transmutation Fuels

Fabrication facility: MA LAB

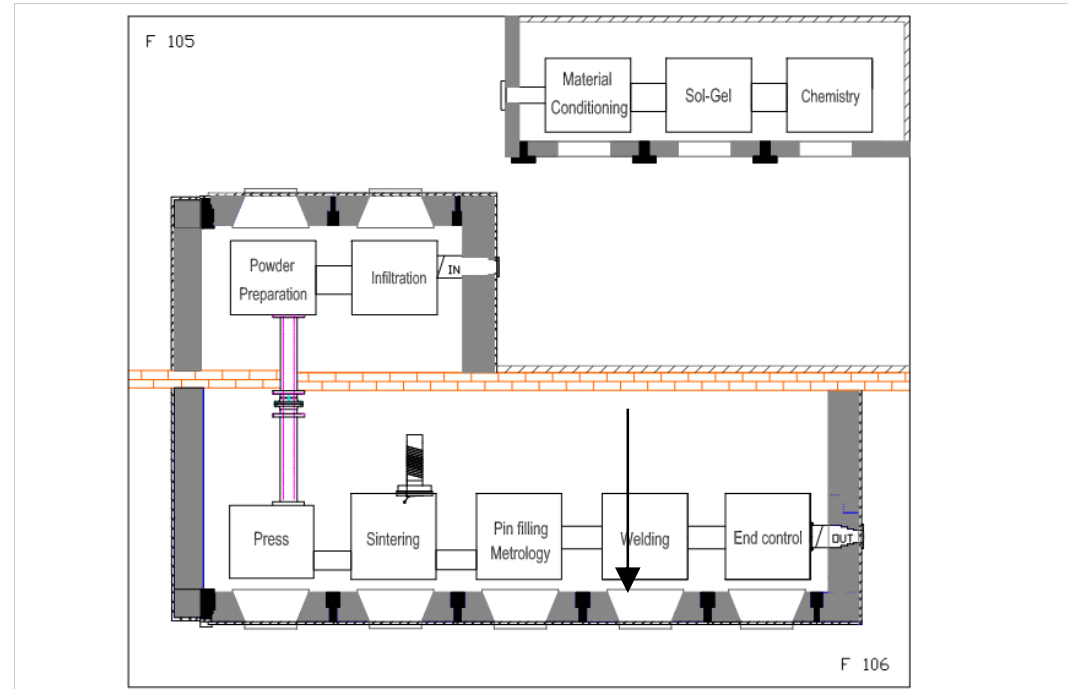


Isotope	Limiting mass (g) <sup>a</sup>	Criterion
<sup>231</sup> Pa	10	Shielding
<sup>237</sup> Np	- <sup>b</sup>	
<sup>241</sup> Am	50	License <sup>c</sup>
<sup>242m</sup> Am	0.1	Shielding
<sup>243</sup> Am	65	License <sup>c</sup>
<sup>244</sup> Cm	5	Shielding & licence <sup>c</sup>

<sup>a</sup> to yield max 2 μSv/h at 1 metre

<sup>b</sup> no practical limit

<sup>c</sup> 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

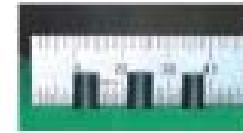
MA bearing metal fuels

CRIEPI/ITU; INL

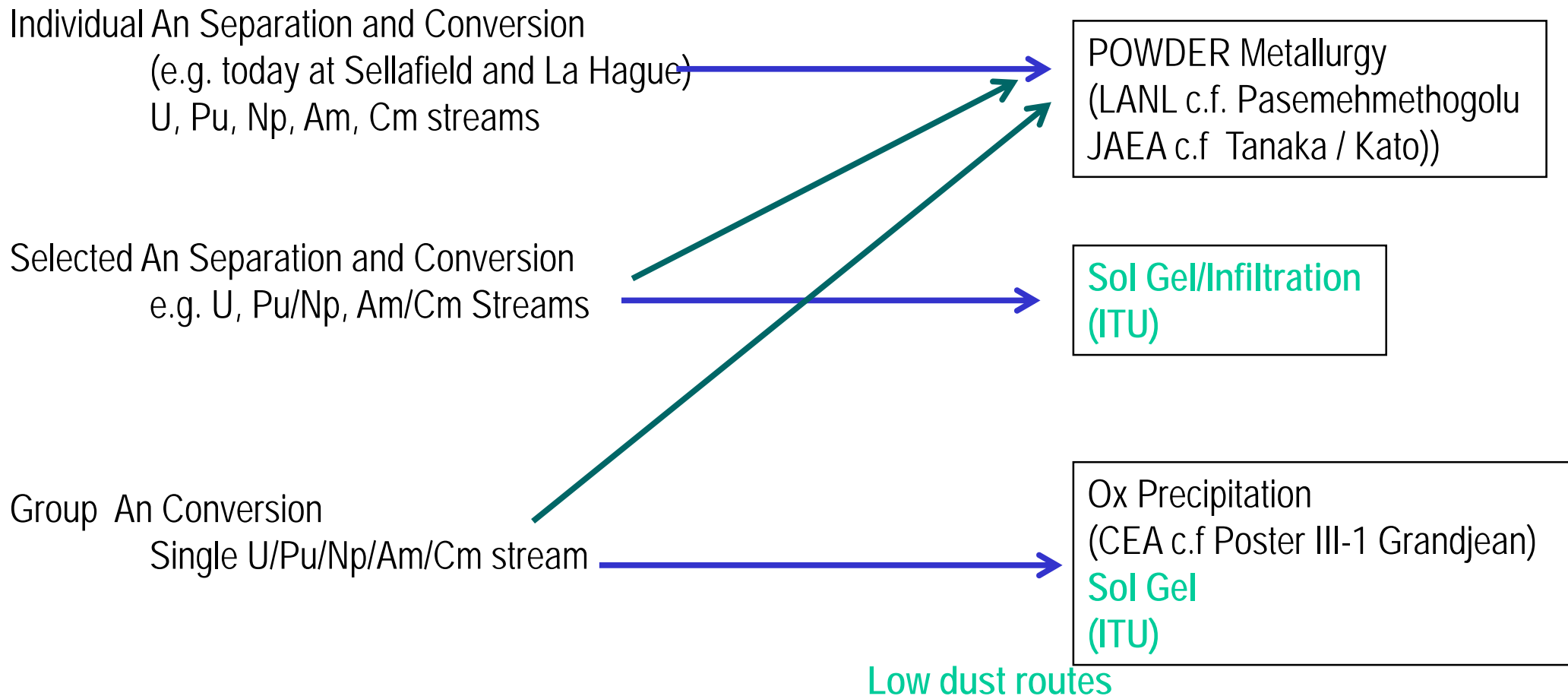
MA bearing nitride fuels

JAEA; LANL

# Transmutation Fuel Fabrication: Strategic choice at conversion step



$(U_{0.65}Pu_{0.30}Am_{0.05})O_2$  fuel  
(Tanaka Global 2007)



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**Sol Gel Routes - SUPERFACT**

Infiltration

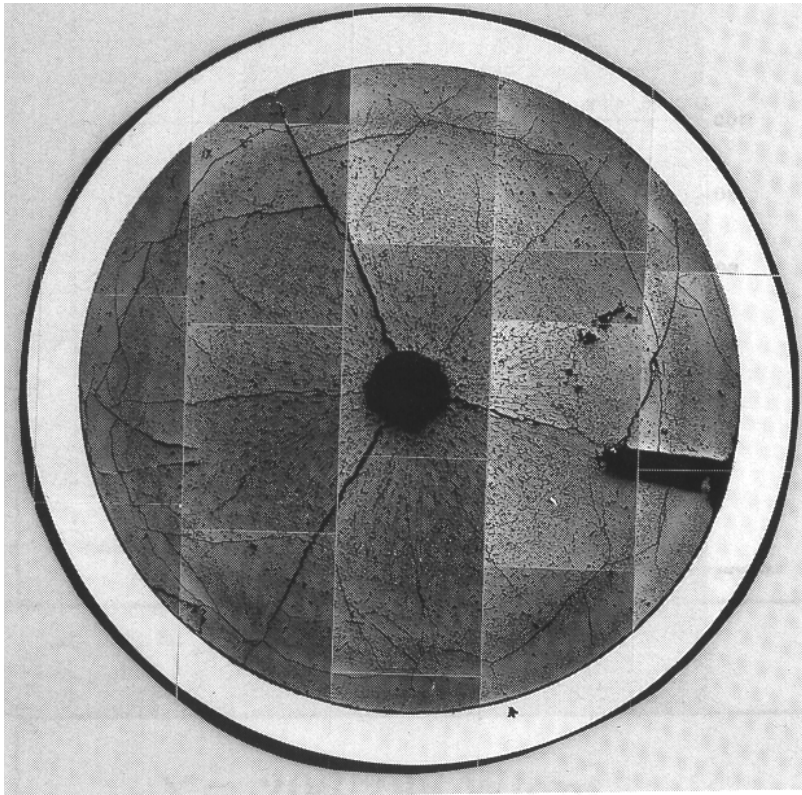
CERMETS/CERCERS

Gen IV Oxides and Nitrides

Conclusions

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

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



Typical observations for  
(U<sub>0.74</sub>Pu<sub>0.24</sub>Am<sub>0.02</sub>)O<sub>2</sub> 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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**INFILTRATION**

CERMETS

Gen IV Oxides and Nitrides

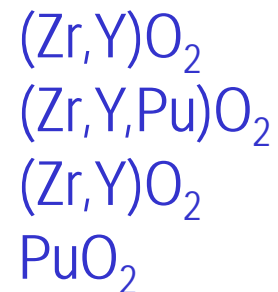
Conclusions

## HELIOS FUELS FABRICATED USING Sol Gel /Infiltration processes

Fuel	Compound	Am content*	Pu content*	Particle size	Density
		$g \cdot cm^3$	$g \cdot cm^3$	$\mu m$	%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	$ZrYAmO_2 + Mo$	0.76		80-100	
HELIOS 5	$PuAmO_2 + Mo$	0.32	1.28	20-150	

CEA

ITU

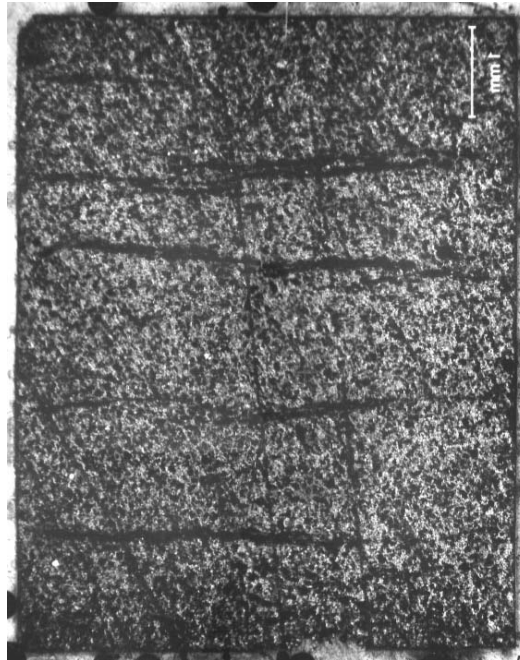


Sol Gel precursors  
(prepared in inactive  
or Pu facilities)

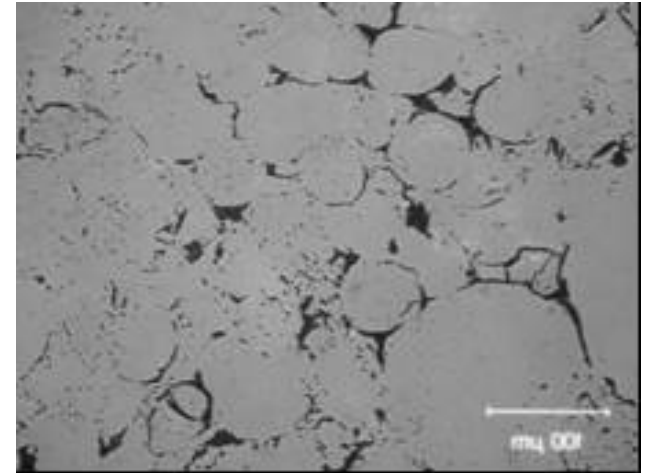
$\text{YZrAmO}_{2-x}$



Good visual aspect



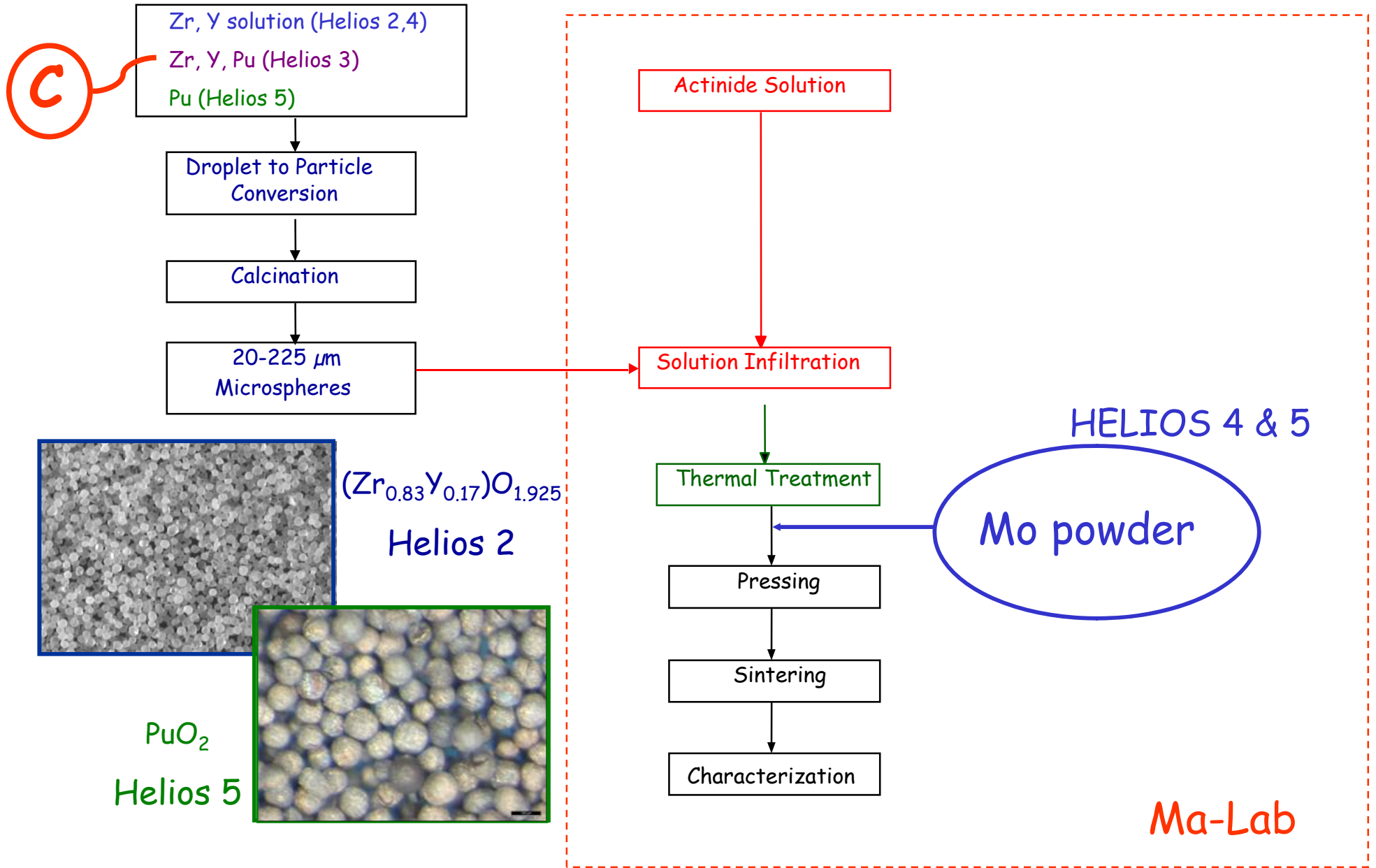
But microstructure → cracks and large localised porosity



Carbon addition



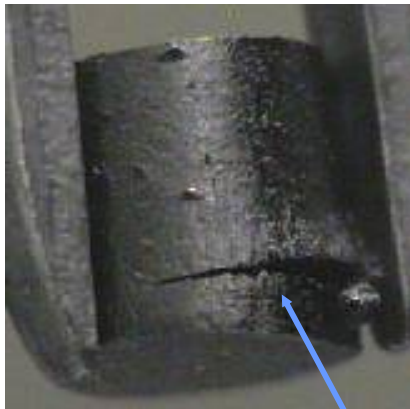
improve microstructure  
improve infiltration behaviour



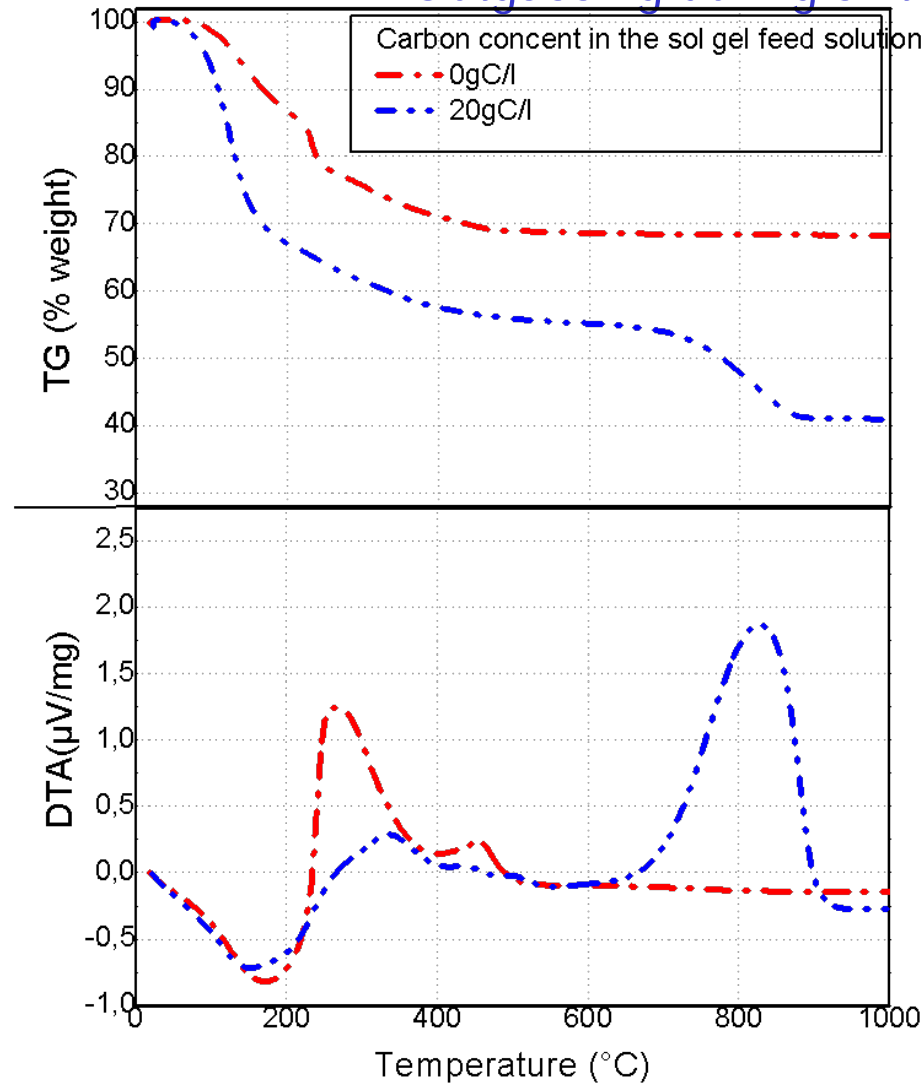
CARBON ADDITION: Surface spalling, higher than normal mass loss  
 → Outgassing during sintering (Ar/H<sub>2</sub>)



Non uniform shrinkage



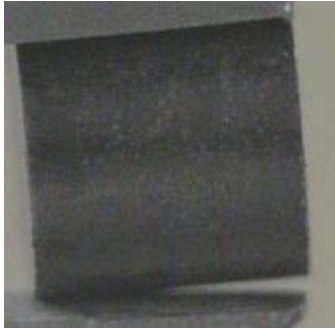
cracks



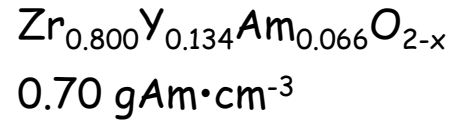
CH<sub>4</sub> or ???

Process modification

- Calcine 800 C (air)
- Infiltration
- Calcine 800C (air)
- Pellet compaction
- Heat 1000 C (air)
- Sinter Ar/H<sub>2</sub>



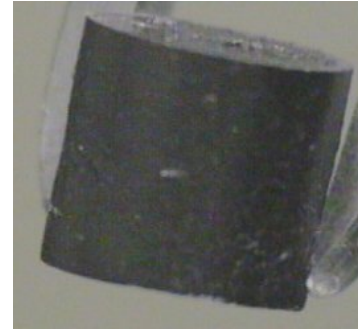
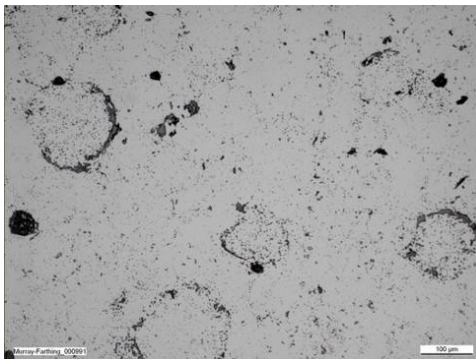
## HELIOS 2



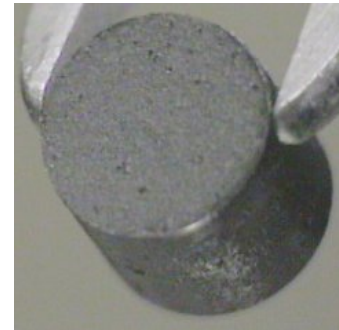
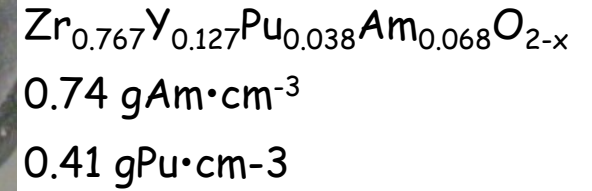
92.6 ± 1.2 %TD



90.9 ± 0.3 %TD



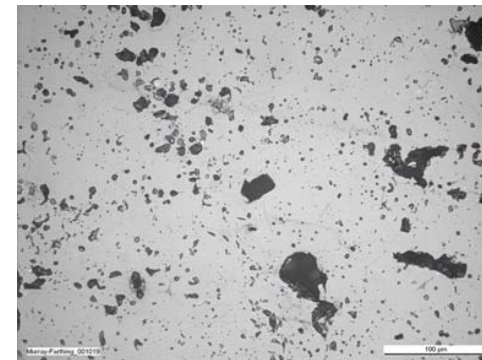
## HELIOS 3

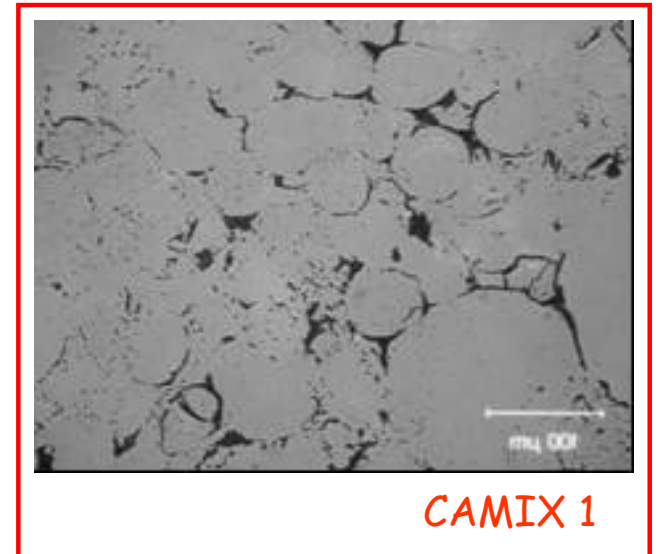
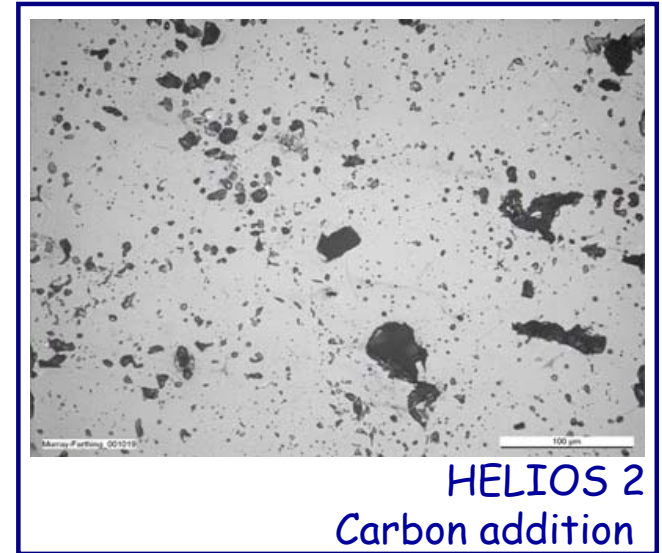
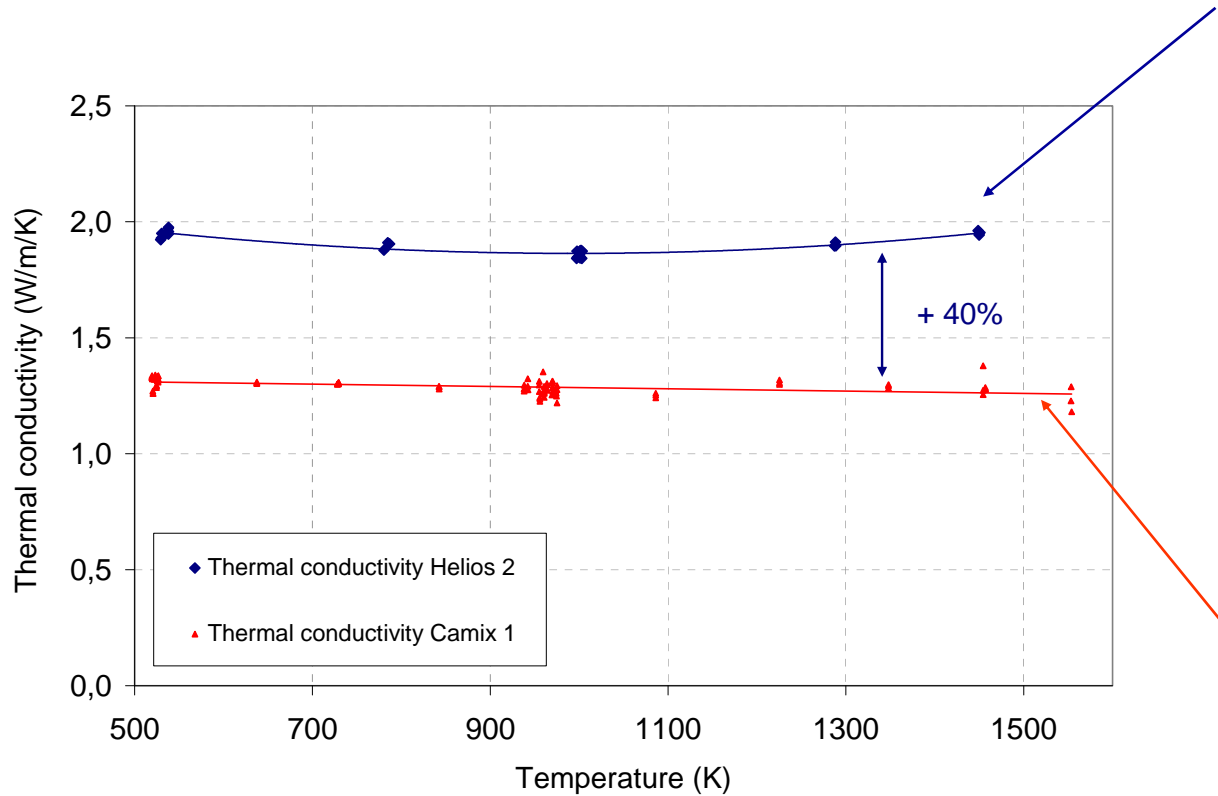


89.7 ± 0.4 %TD



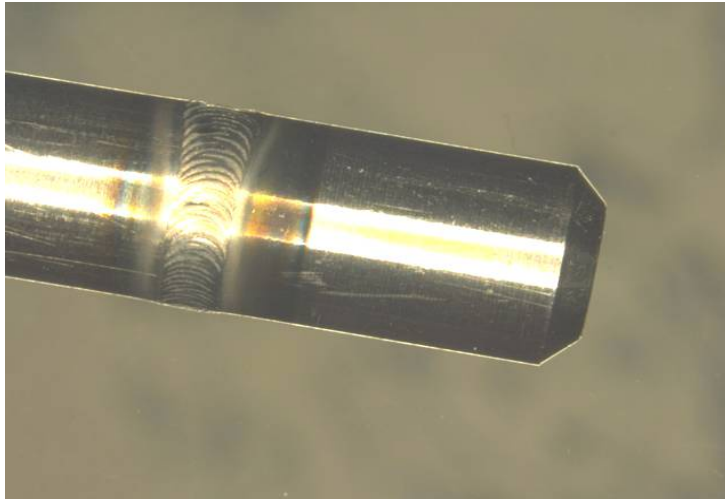
89.1 ± 1.1 %TD





Carbon addition → improve microstructure → **Improve Thermal Conductivity<sup>1</sup>**

<sup>1</sup> measured by Dragos Staicu (MR)



Measured dose rate

	Contact (mSv/h)	1m ( $\mu$ Sv/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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Difficulties in Fabricating MA fuels

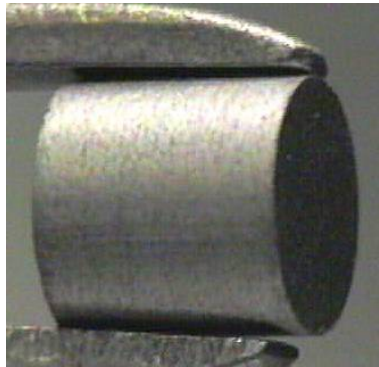
Sol Gel Routes - SUPERFACT

INFILTRATION

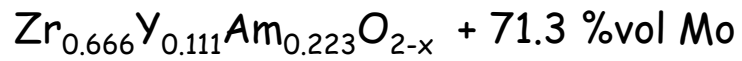
CERMETS

Gen IV Oxides and Nitrides

Conclusions

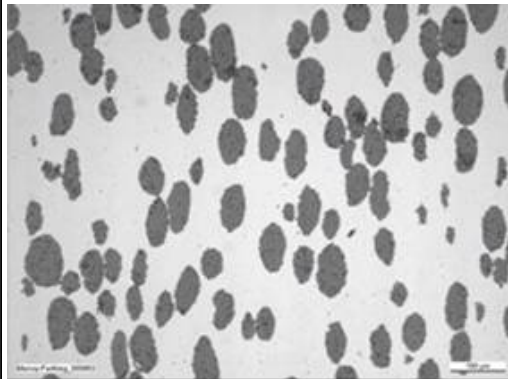
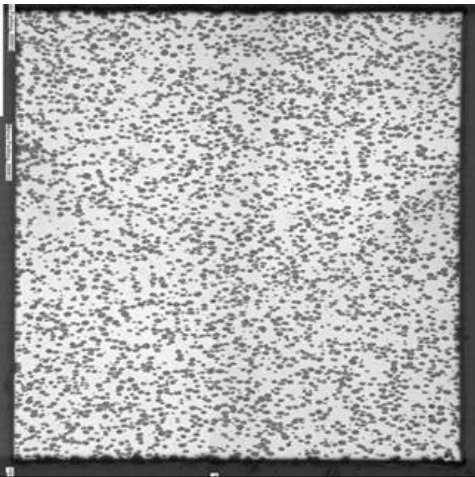


HELIOS 4

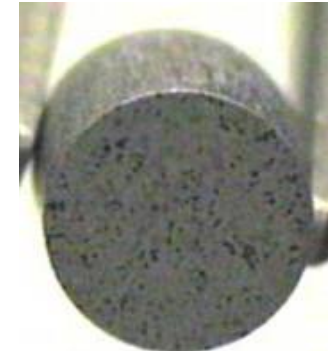


$0.69 \text{ gAm}\cdot\text{cm}^{-3}$

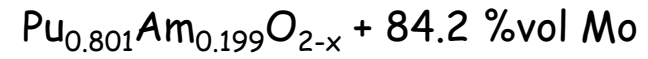
$94.2 \pm 0.4 \text{ \%TD}$



HR = 4.83

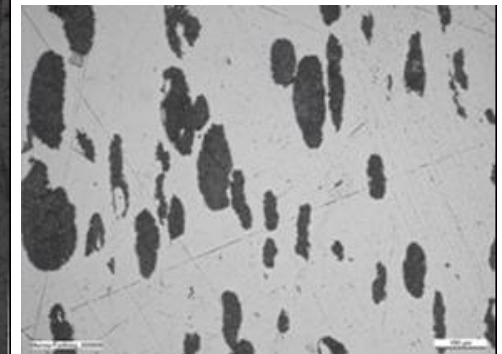


HELIOS 5



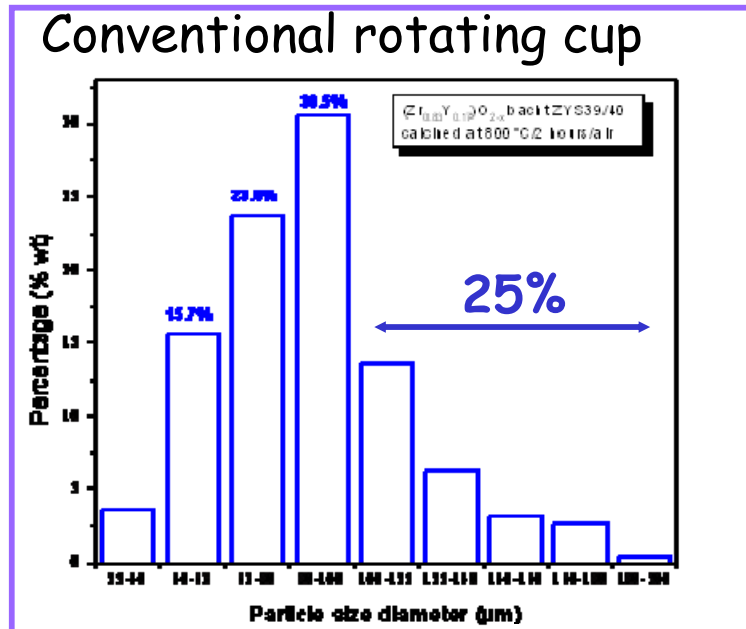
$0.295 \text{ gAm}\cdot\text{cm}^{-3}$ ,  $1.24 \text{ gPu}\cdot\text{cm}^{-3}$

$95.9 \pm 0.4 \text{ \%TD}$



HR = 12.01

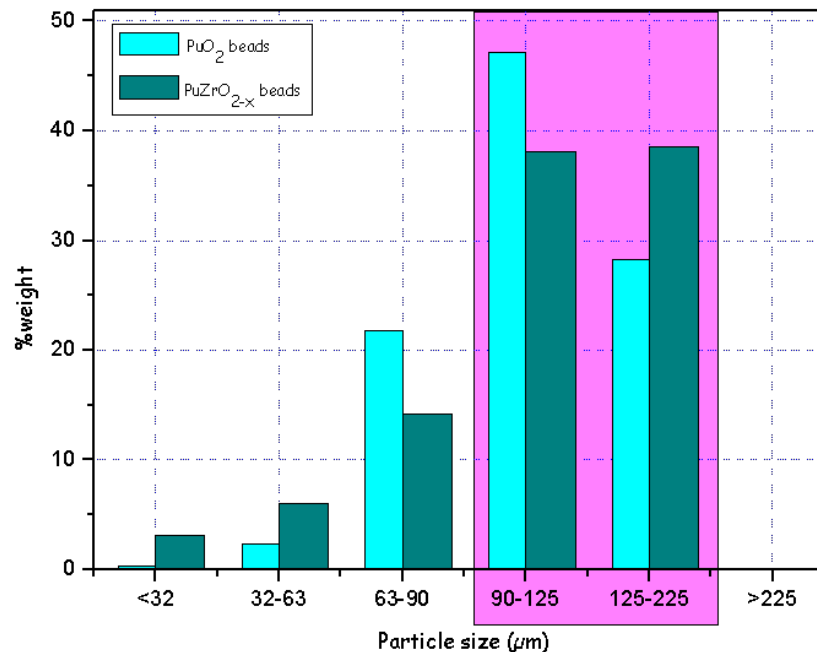
# Fabrication of porous $\text{PuO}_2$ and $(\text{Zr}_{0.705}\text{Pu}_{0.295})\text{O}_2$ beads (100-200 $\mu\text{m}$ )



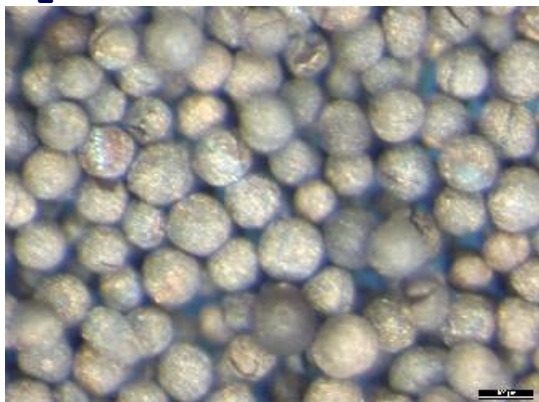
- Pu concentration
- Denitration
- Viscosity
- Rotating cup (rpm, height, etc)
- Polydisperse size distribution



## FUTURIX fuels

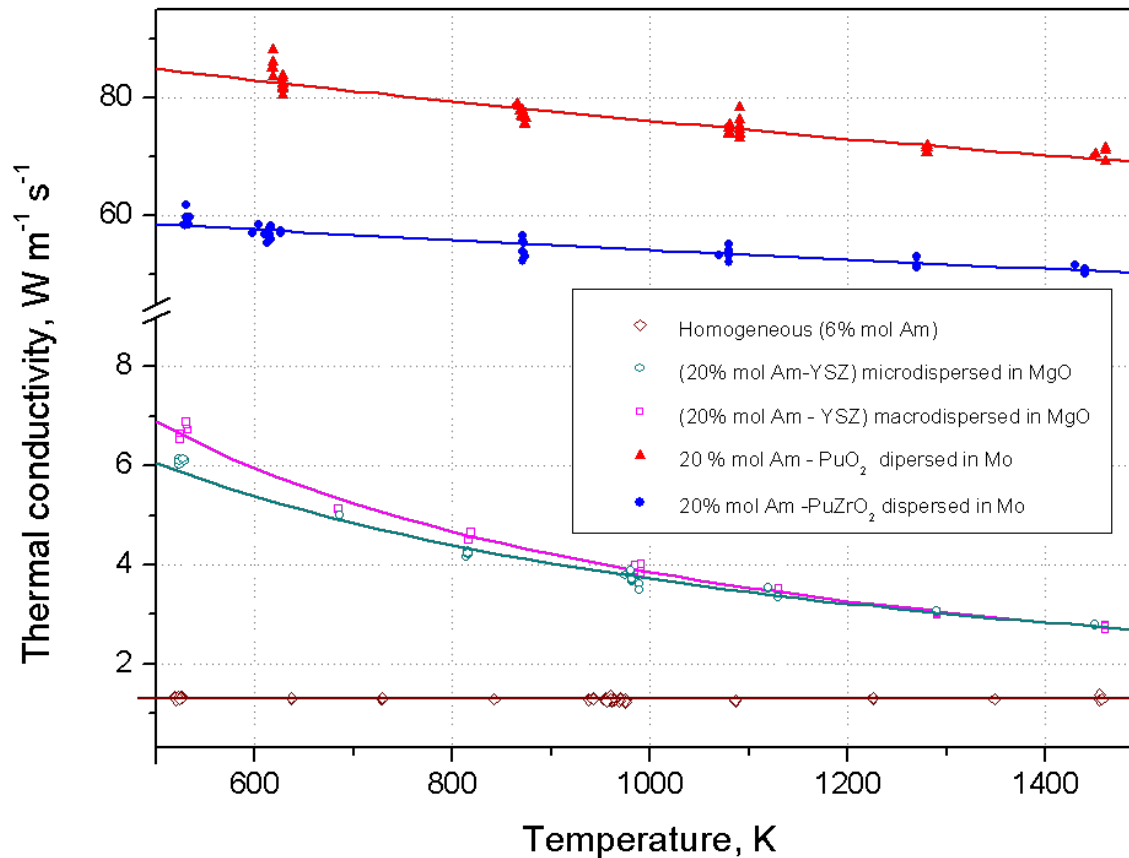


### $\text{PuO}_2$ beads



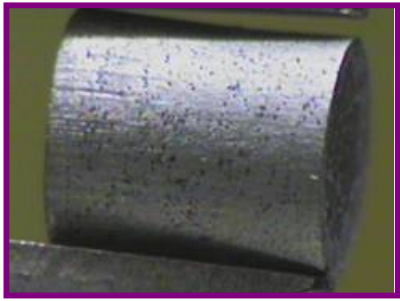
Calculated from measured thermal diffusivity and specific heat.

CERCER compared to CERMET

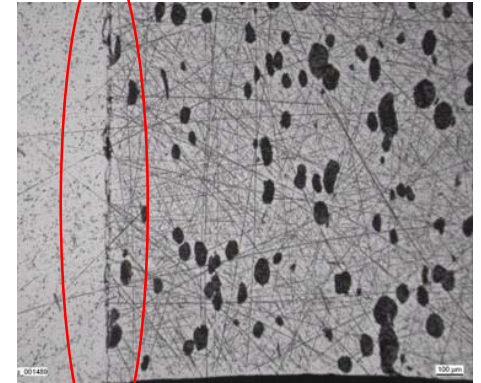
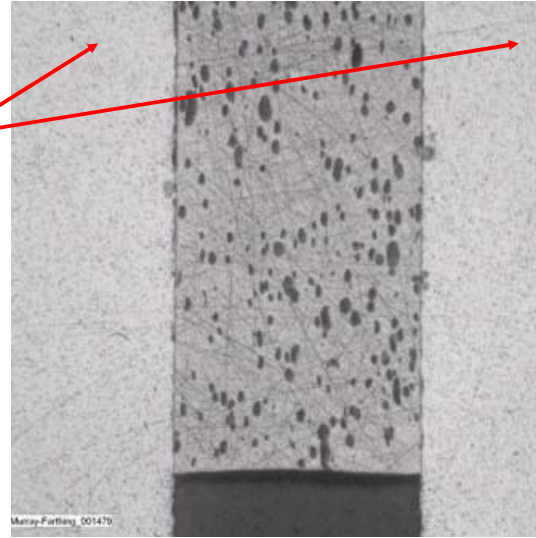


## Compatibility test- T91

Mo- (Pu,Am)O<sub>2-x</sub>  
(FX 5)



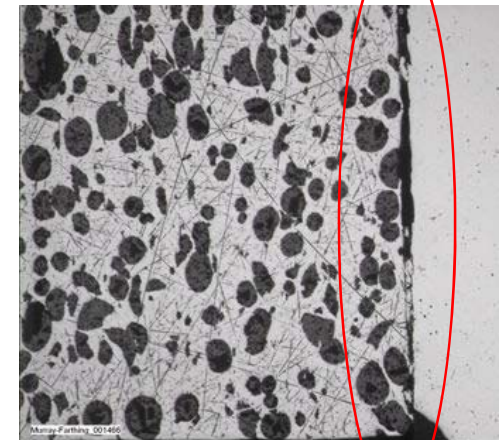
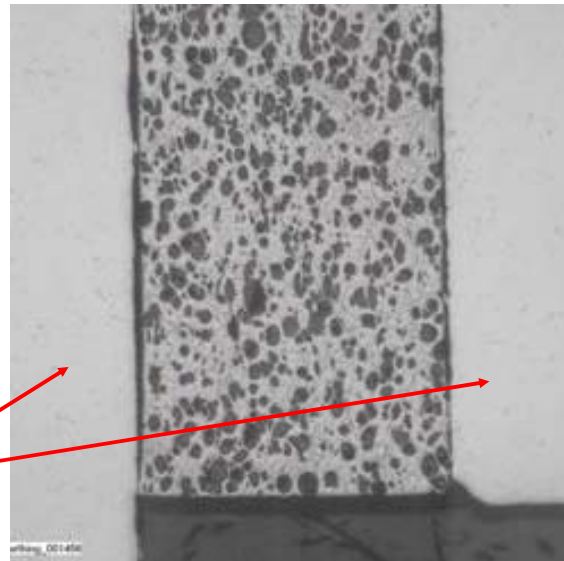
**Cladding**



Mo- (Zr,Pu,Am)O<sub>2-x</sub>  
(FX 6)



**Cladding**



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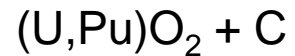
INFILTRATION

CERMETS

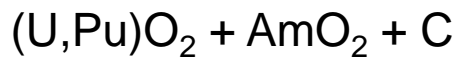
Gen IV Oxides and Nitrides

Conclusions

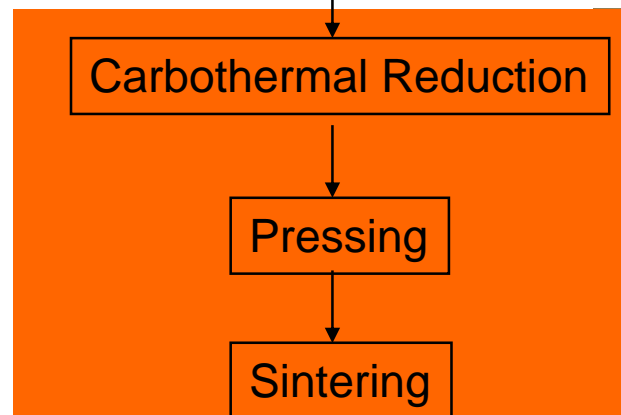
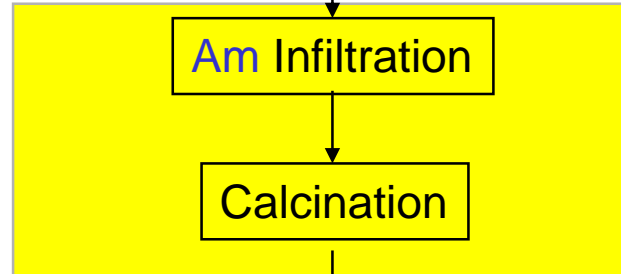
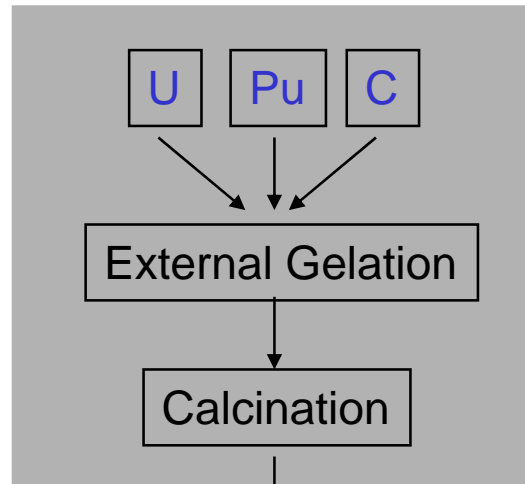
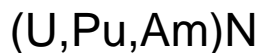
Conventional  
Gloveboxes



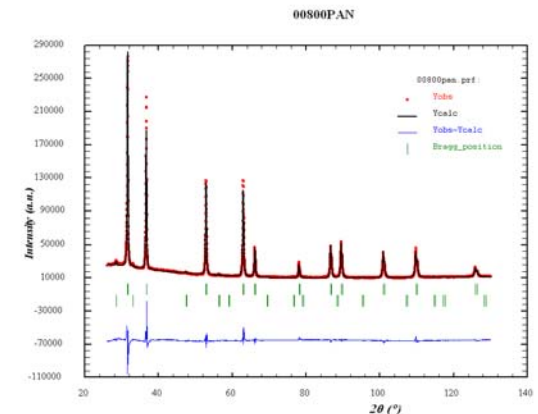
MALAB



Conventional  
Gloveboxes with  
Purified  
Atmospheres



FR Nitride (Carbide)  
Fuel Production

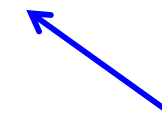


MA Production Losses : Particularly for carbides

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	seems ok
Carbide 2 pin 4	74,7	2,58	0,112	



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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## 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

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.....

## 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

