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Kernels for the Disposition of Pu in High Temperature Reactors

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Outline

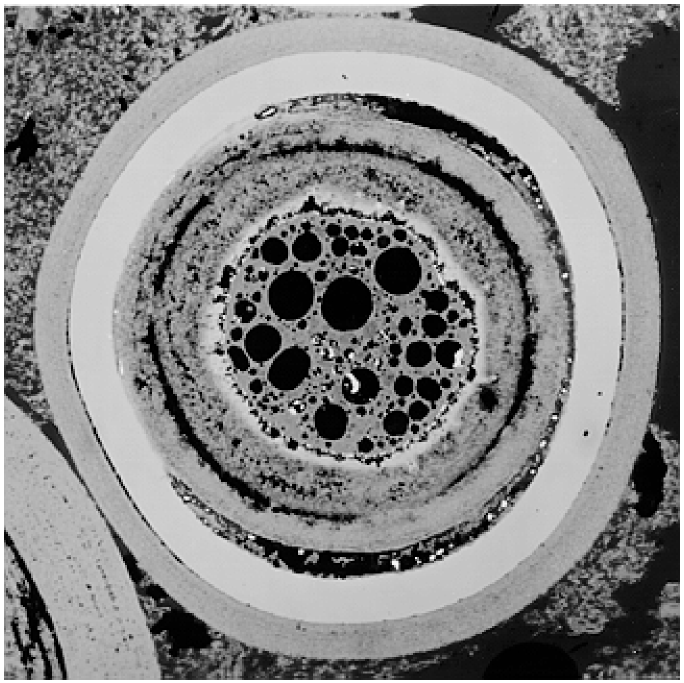
- Kernel Compositions for Pu and MA Transmutation
- Irradiation Performance: Prospects for such kernels
- Fabrication Routes for Pu (or MA) based kernels
- Conclusions

Kernels for Pu and MA Transmutation

FTE 13 Test in Peach Bottom (GA)

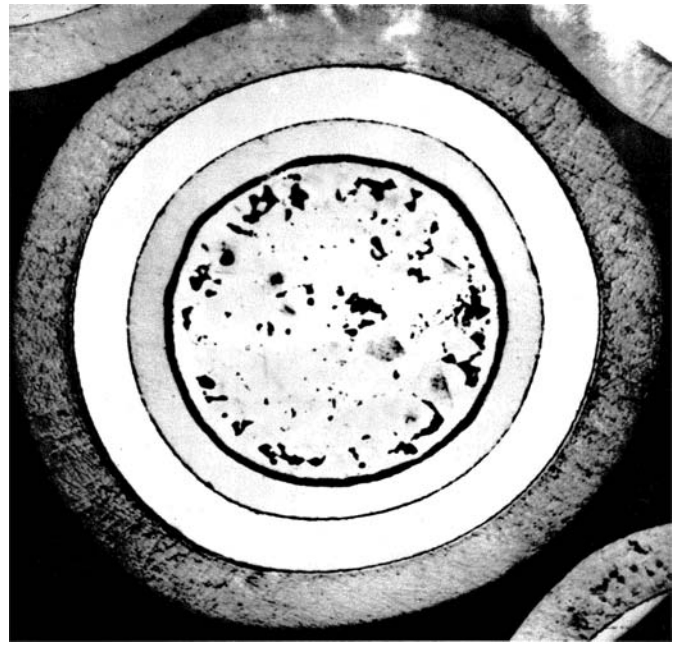
PuO_2 110 and 200 µm

$(Th,Pu)O_2$ 350 µm



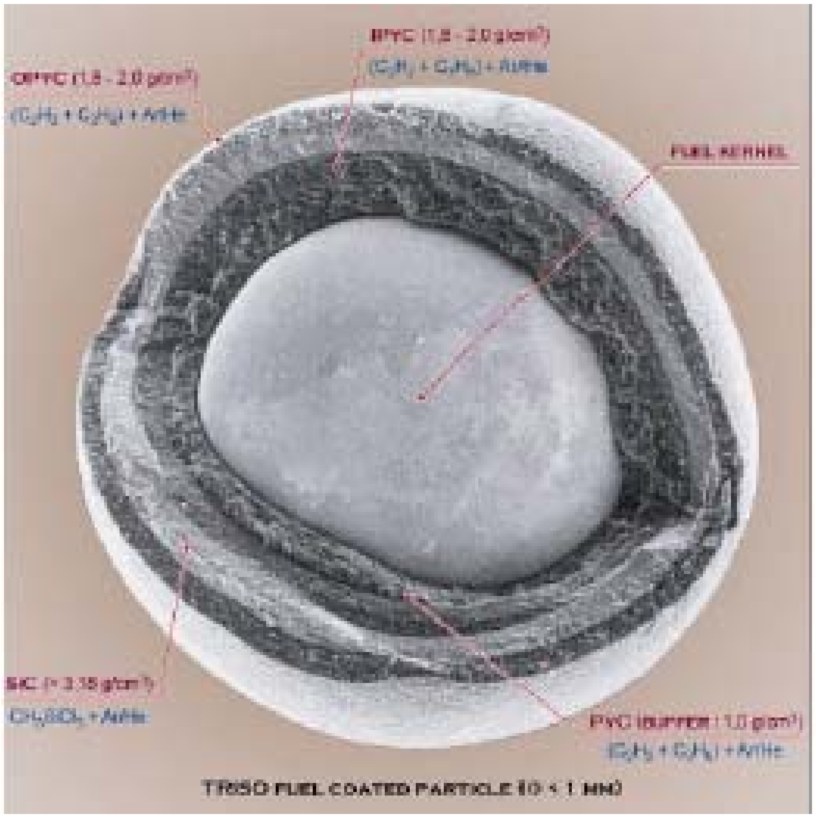
TRISO-Coated $PuO_{1.68}$
~75% Burnup,
2.3×10^{25} n/m^2
T up to 1450 ºC

Tests in Europe (Belgonucleaire)



PuO_2
600 MWd/Tonne HM
(Gaube et al 1972)

Coated Particle Layers



Fuel Kernel:

- Provides fission energy (heat)
- Retains a great part of fission products (FP)

Buffer:

- Protects main layers against FP recoil
- Free volume for released fission gas (FG)
- Accommodates Kernel swelling

Inner PyC:

- Prevents Kernel corrosion by chlorine during manufacture
- Provides mechanical support for SiC
- Retains FG

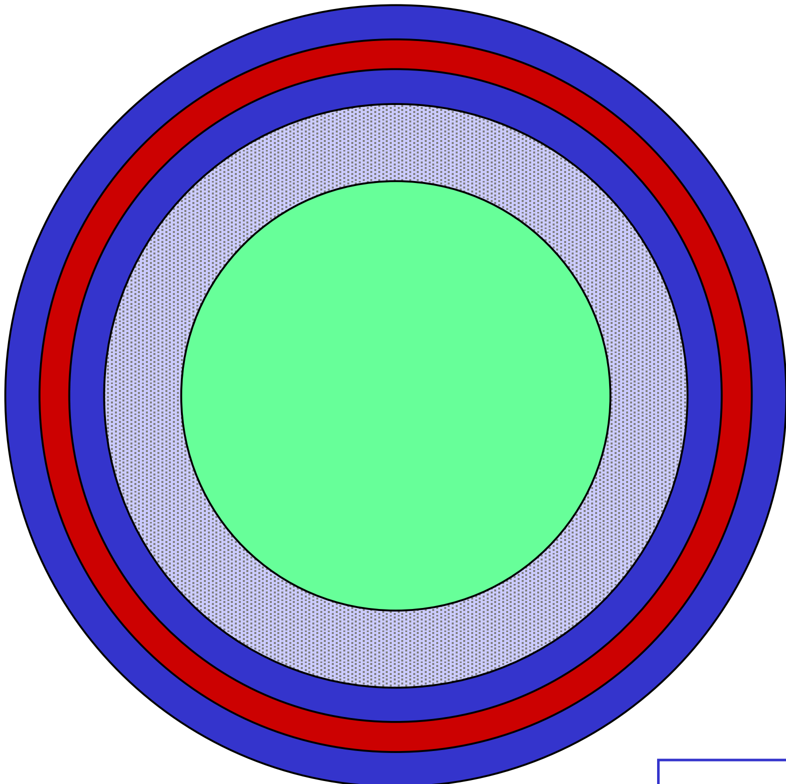
SiC:

- Main load bearing member
- Retains FG, volatile and metallic FP

Outer PyC:

- Provides mechanical support for SiC
- Provides FP barrier for particles with defective SiC
- Provides bonding surface for compacting

Irradiation Performance: Prospects for such kernels



Coated particles

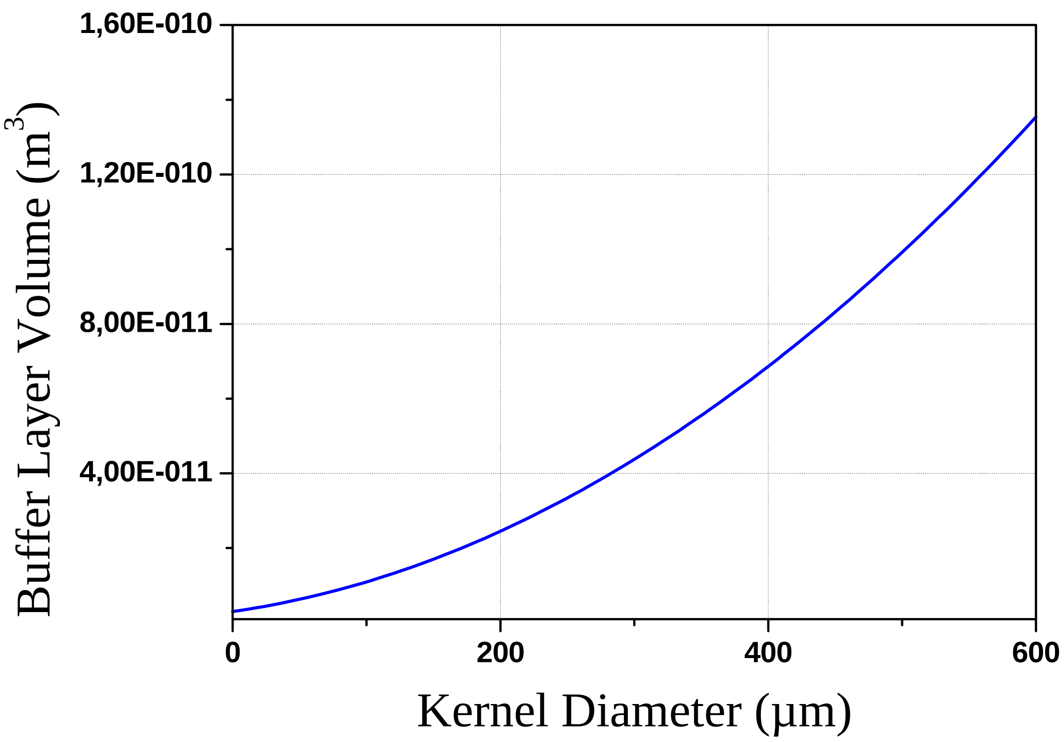
Buffer layer (90 µm)

Damage resistance

Free volume (porosity 50%)
(Depends on layer thickness and kernel diameter)

90 µm Buffer is a consensus
➔Bufer volume depends on kernel diameter

Irradiation Performance: Prospects for such kernels (ii)



➔Factor 4 (500µm) for FP and CO accomodation

➔Higher Burnup achievable

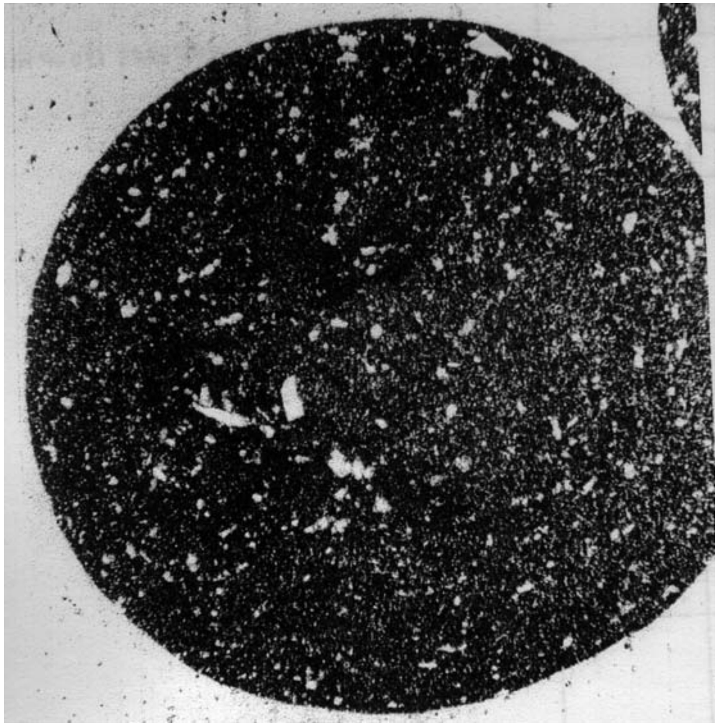
Irradiation Performance: Prospects for such kernels (iii)

Diluted kernels: Who has thought of it before?

Belgonucleaire. Bairiot et al
Irradiation Test in FRJ2

PuO2 / Graphite

1: 20 in mass



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Irradiation Performance: Prospects diluted kernels

Diluted kernels: Pu (Am) dilution in Yttria Stabilised Zirconia: (Zr,Y,Pu)O2

- Cubic structure
- High Melting point
- IMF (neutonically Inert Matrix Fuel)
- Current Concept for LWR and FR

	PuO2	**(Zr,Y,Pu)O2**	**(Zr,Y,Pu)O2**
Diameter (µm)	200	500	600
Mass Pu (mg)	0,041	0,041	0,041
Mass YSZ (mg)	0	0,365	0,649
Pu/total oxide (wt%)	**88,2**	**9,9**	**5,8**

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Irradiation Performance: Prospects for YSZ kernels

(a) Temperature Difference between centre and edge of kernel

Material	***λ (W.m^{-1}.K^{-1})***	***ΔT (°C)***
UO_2	***2.5***	***16***
$(Zr,Y,Pu)O_2$	***1.5***	***26***
Graphite	***41***	***1***

Negligible: so no drastic effect expected on fission gas release

(b) Core neutronics: Negative temperature coefficient

➔ Burnable poison in fuel kernels - Erbium

Kernels based on $(Zr,Y,Er)O_2$

Fabrication Routes for YSZ based Pu (or MA) kernels

Sol Gel External or Internal Gelation

Solution Preparation
↓
Atomisation
↓
Gelation in Ammonia / Hot oil
↓
Washing and Drying
↓
Calcination
↓
Sintering

Active

Gloveboxes

Active Wastes

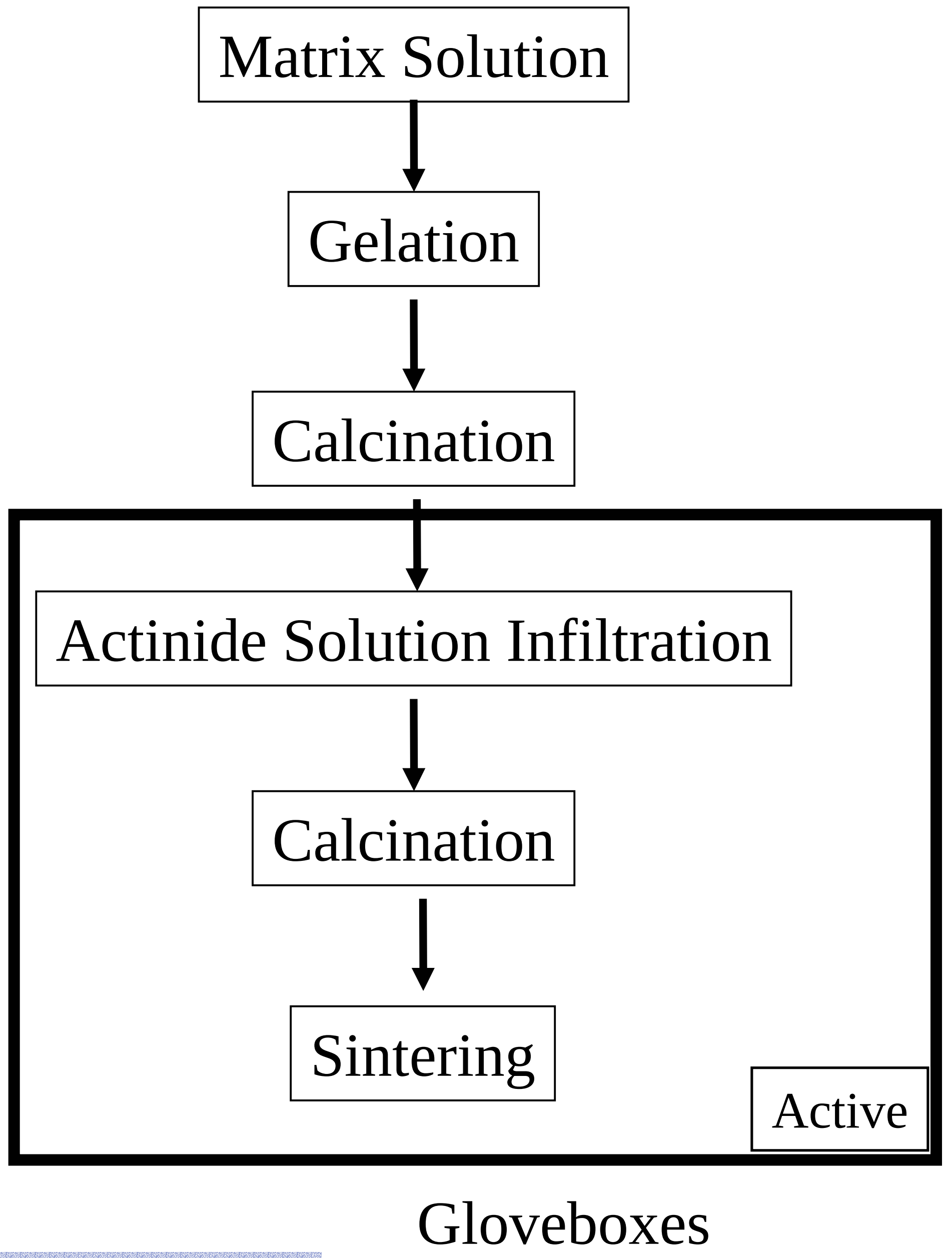
Support Polymer
Water
Ammonia solution
Ammonium nitrate solution
Alcohol

Silicone oil
Organic solvent (Actrel)

Non conforming kernels

Fabrication Routes for Novel Pu or MA based kernels

INFILTRATION Process



Fabrication Routes for Novel Pu or MA based kernels

INFILTRATION Process
Step 1: Production of Porous YSZ Kernels

Matrix Solution

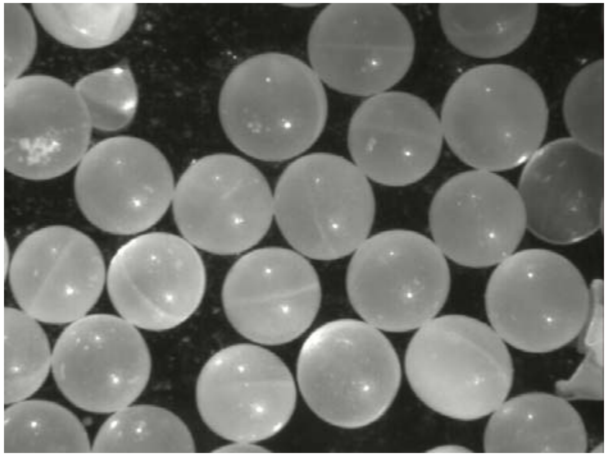
↓

Gelation

↓

Calcination

YSZ Porous beads



External Gelation
Y/(Y+Zr) = 16.0%

Fabrication Routes for Novel Pu or MA based kernels

Matrix Solution

↓

Gelation

↓

Calcination

↓

Actinide Solution* Infiltration

↓

Calcination (NOx)

Active

INFILTRATION Process

Step 2: Put porous kernels in glovebox

Step 3: Infiltrate porous kernels (An nitrate)

Step 4: Calcination

Only Effluent in Active cells

* 50-400 g/l An - Here 155 g/l Pu

Fabrication Routes for Novel Pu or MA based kernels

INFILTRATION Process
Step 5 Sintering of the kernels
Densification and Homogenisation

Matrix Solution

↓

Gelation

↓

Calcination

↓

Actinide Solution Infiltration

↓

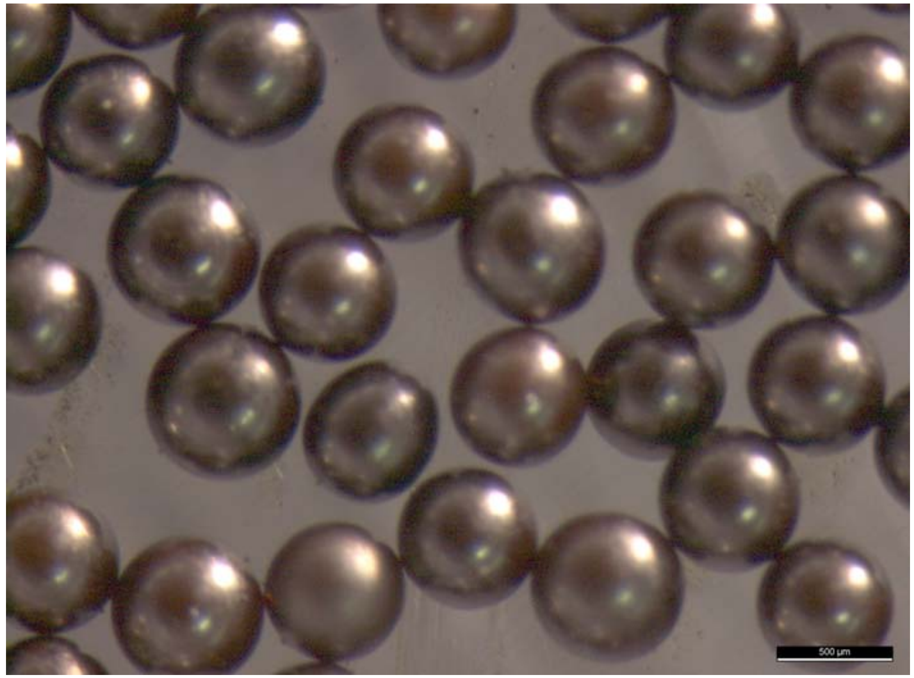
Calcination

↓

Sintering

Active

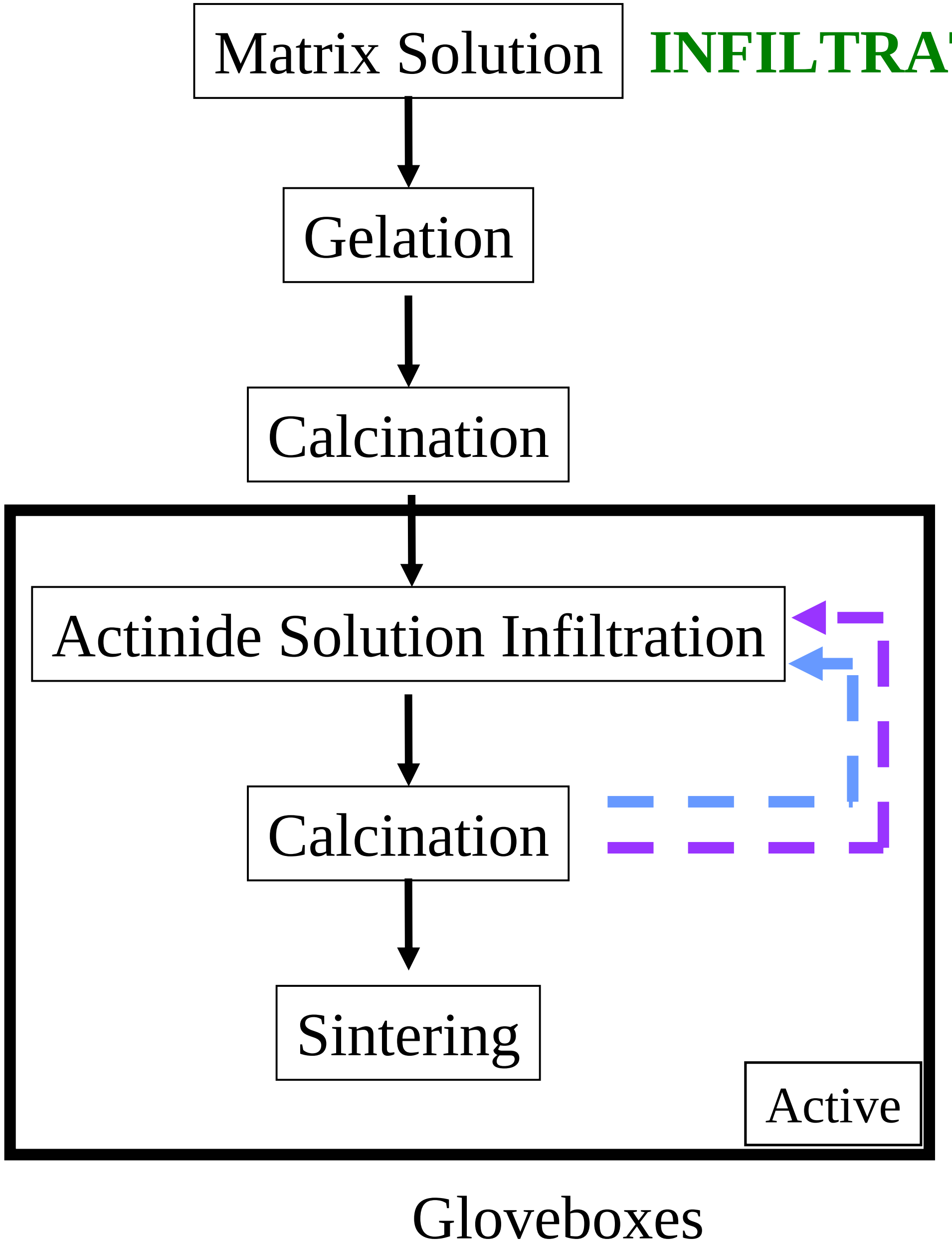
Gloveboxes



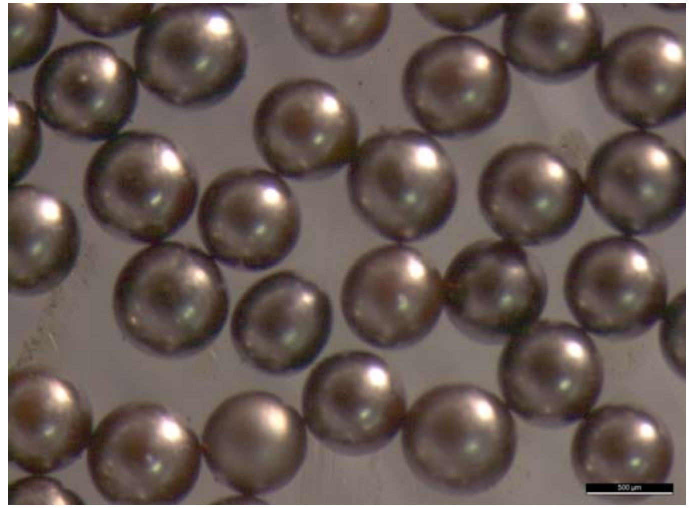
$(Zr,Y,Pu)O_2$ Kernels ϕ = 670 µm

Fabrication Routes for Novel Pu or MA based kernels (xi)

INFILTRATION Process: Increasing An Content

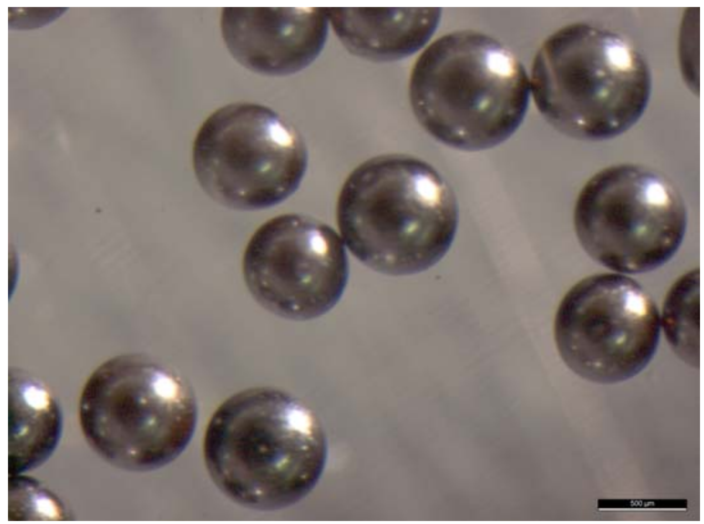


$(Zr,Y,Pu)O_2$



6.1 wt%Pu

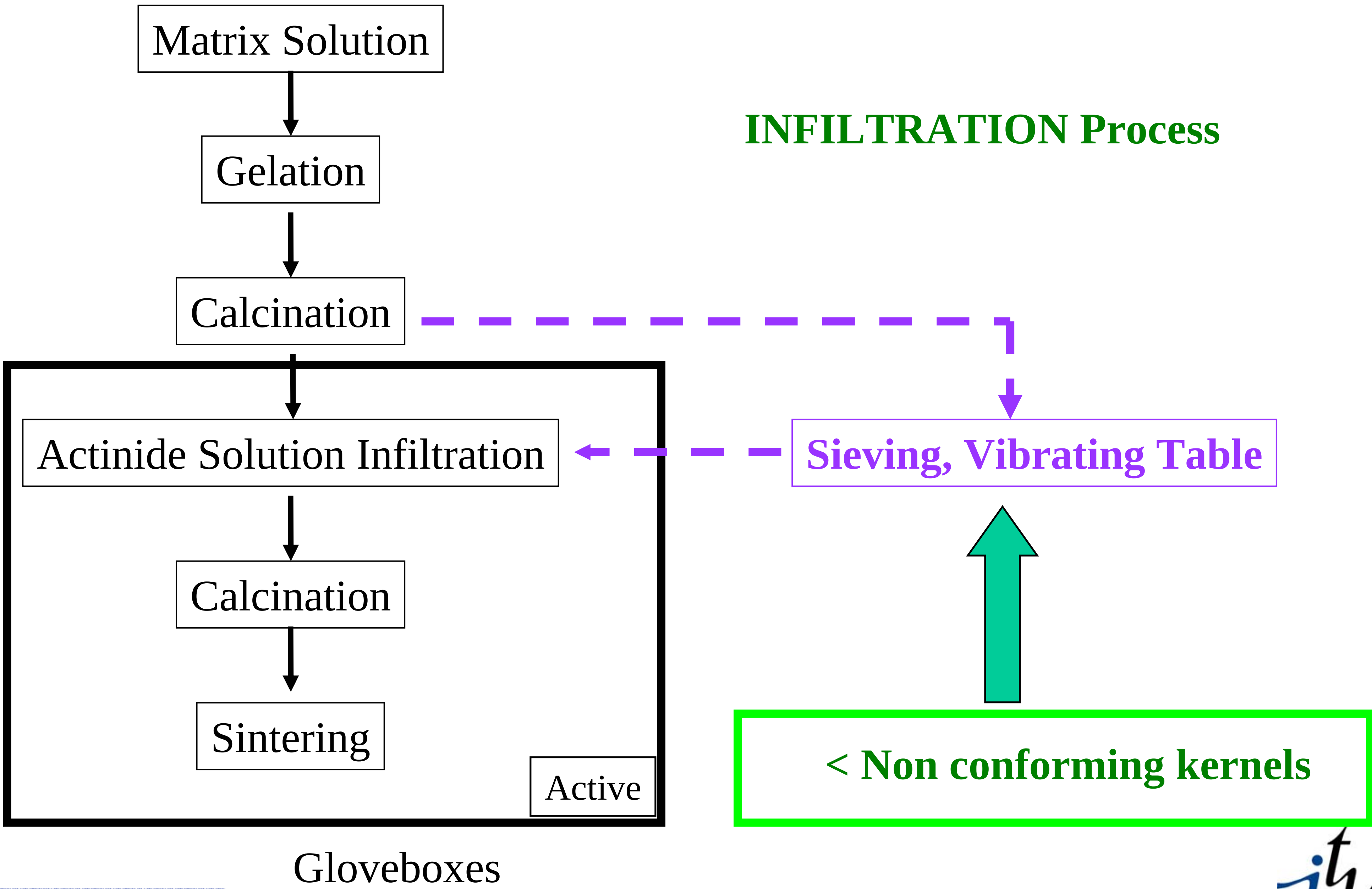
Single



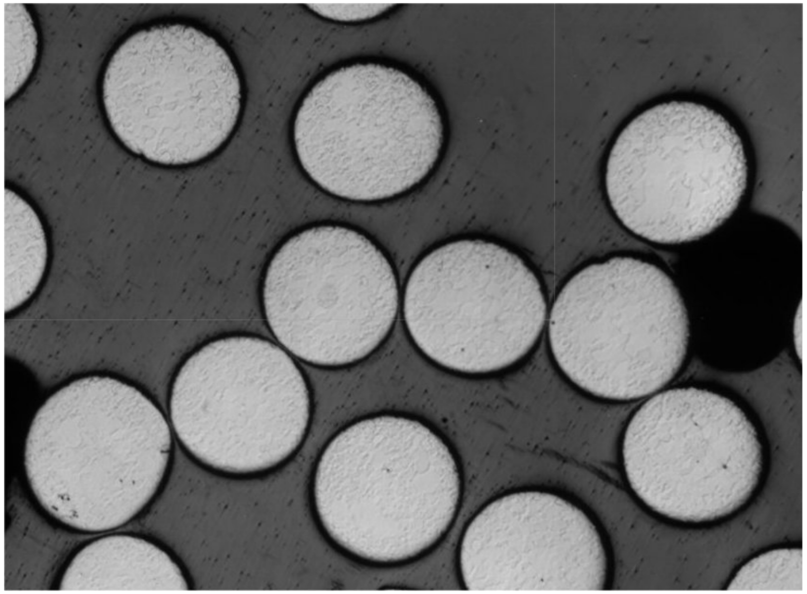
Double

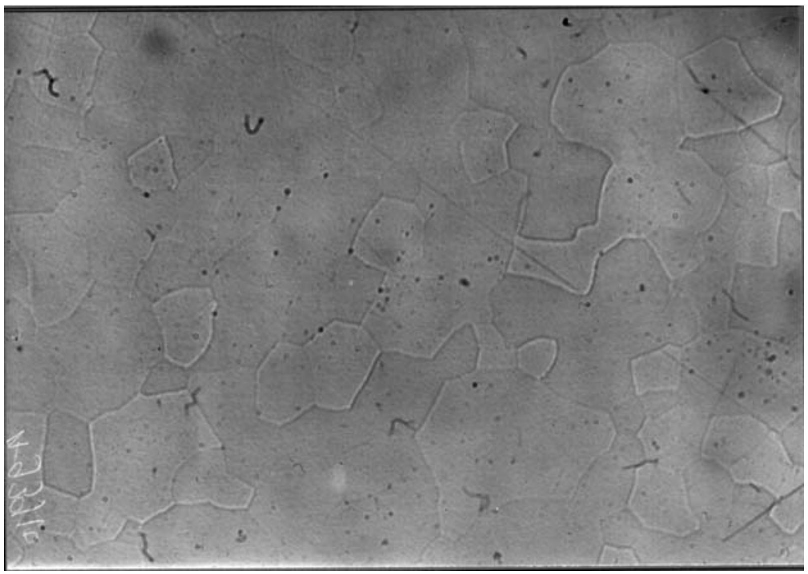
11.0 wt%Pu

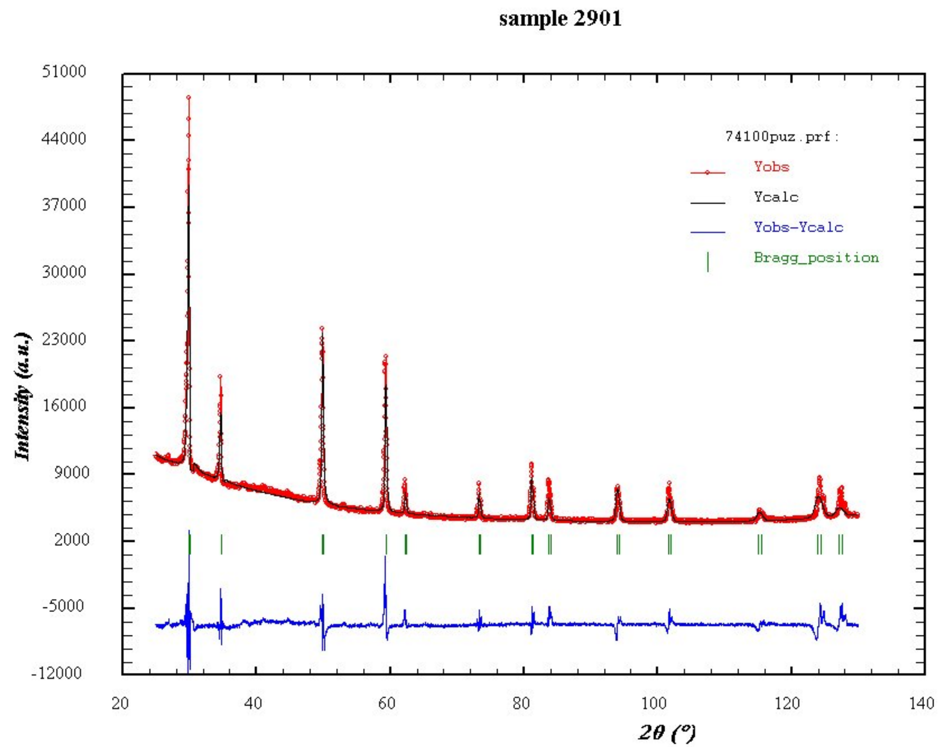
Fabrication Routes for Novel Pu or MA based kernels (xii)



Kernel cross sections (1st Infiltration)

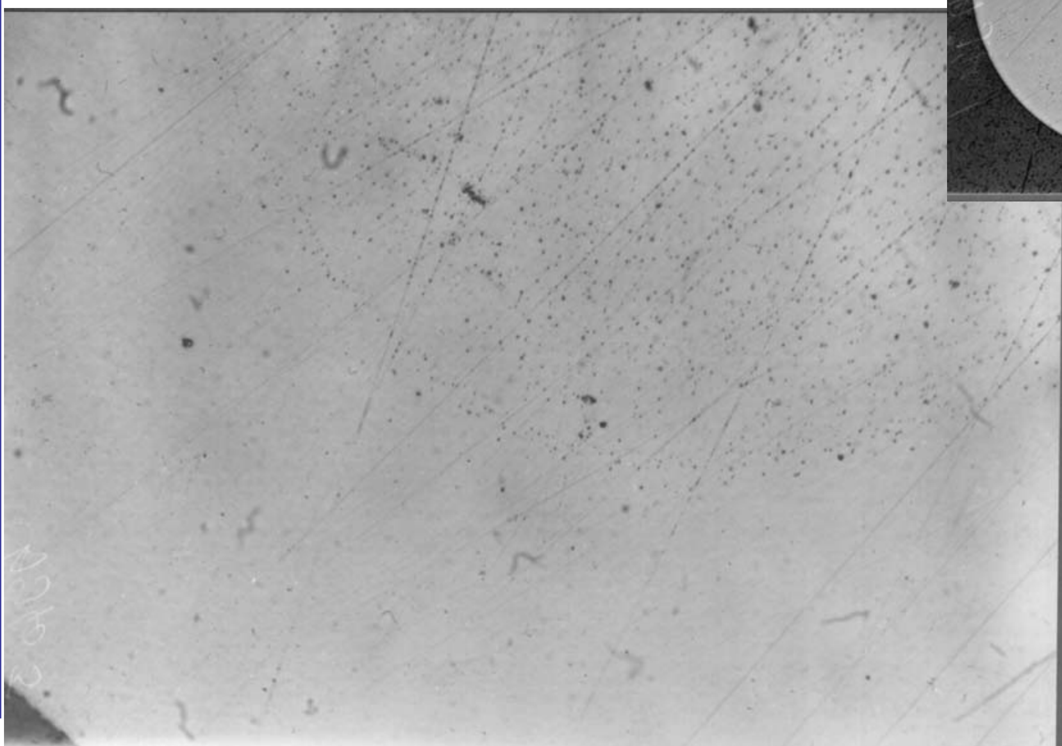


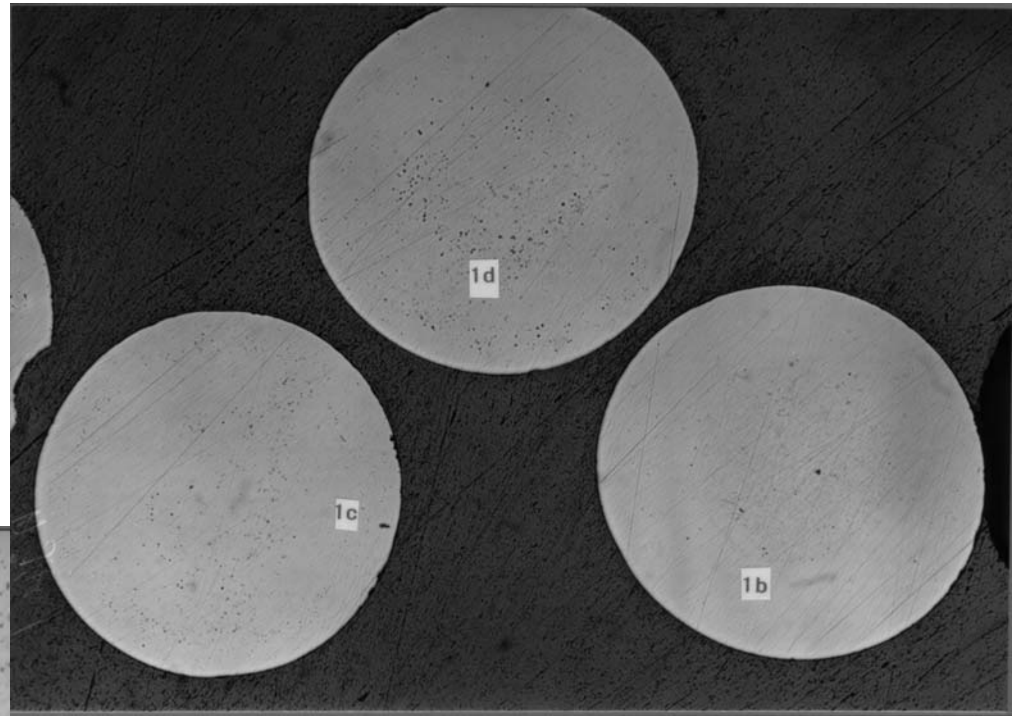




Grain Size ca. 20 μm

Kernel cross sections 2nd Infiltration

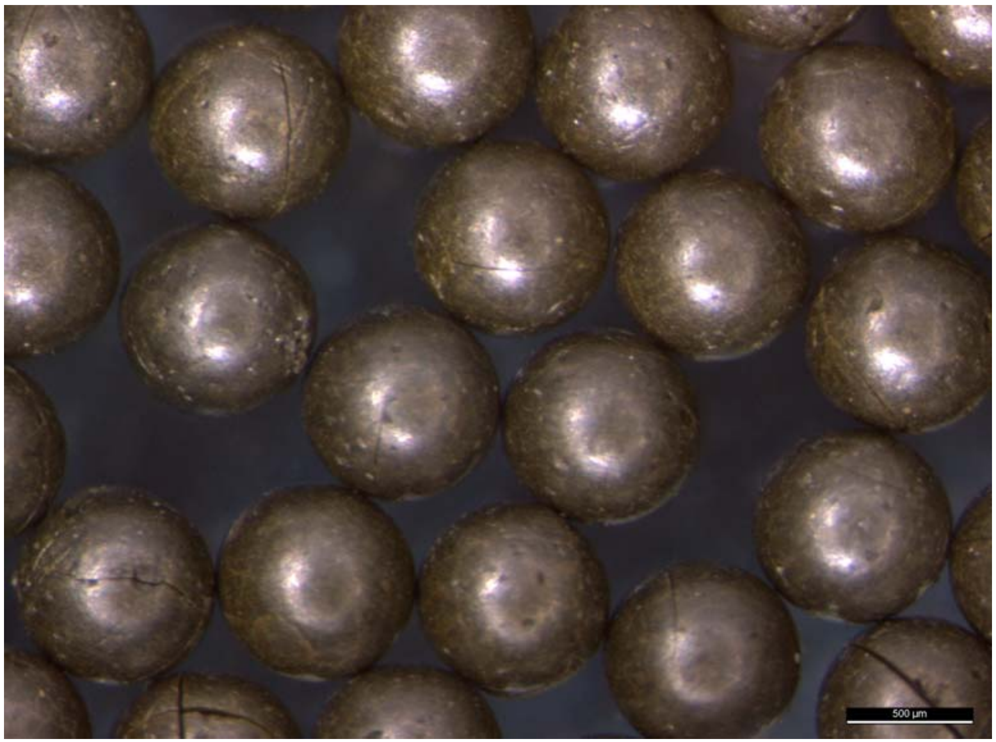




More porous in central region

Porous Beads by Internal gelation

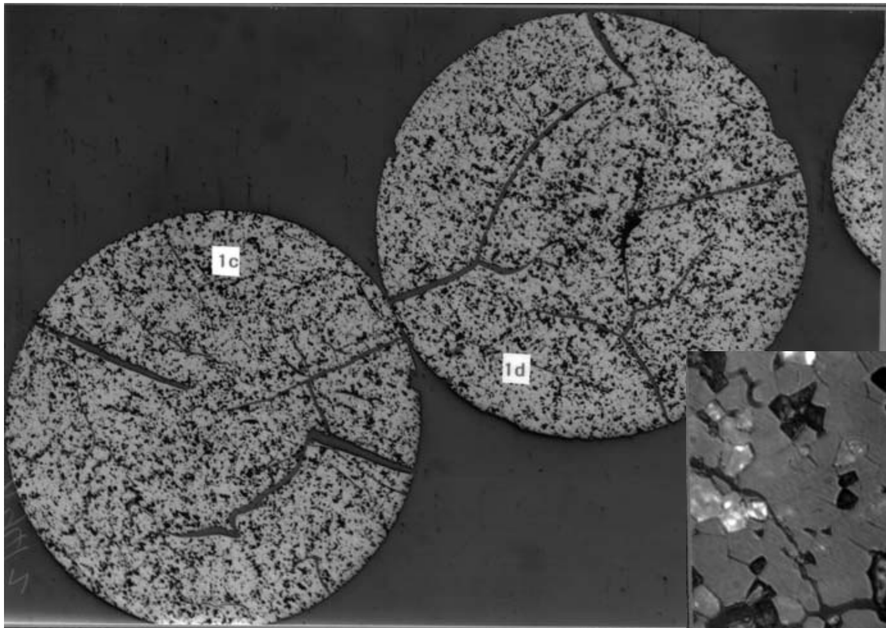
Internal Gelation
Y/(Y+Zr) = 15.6%



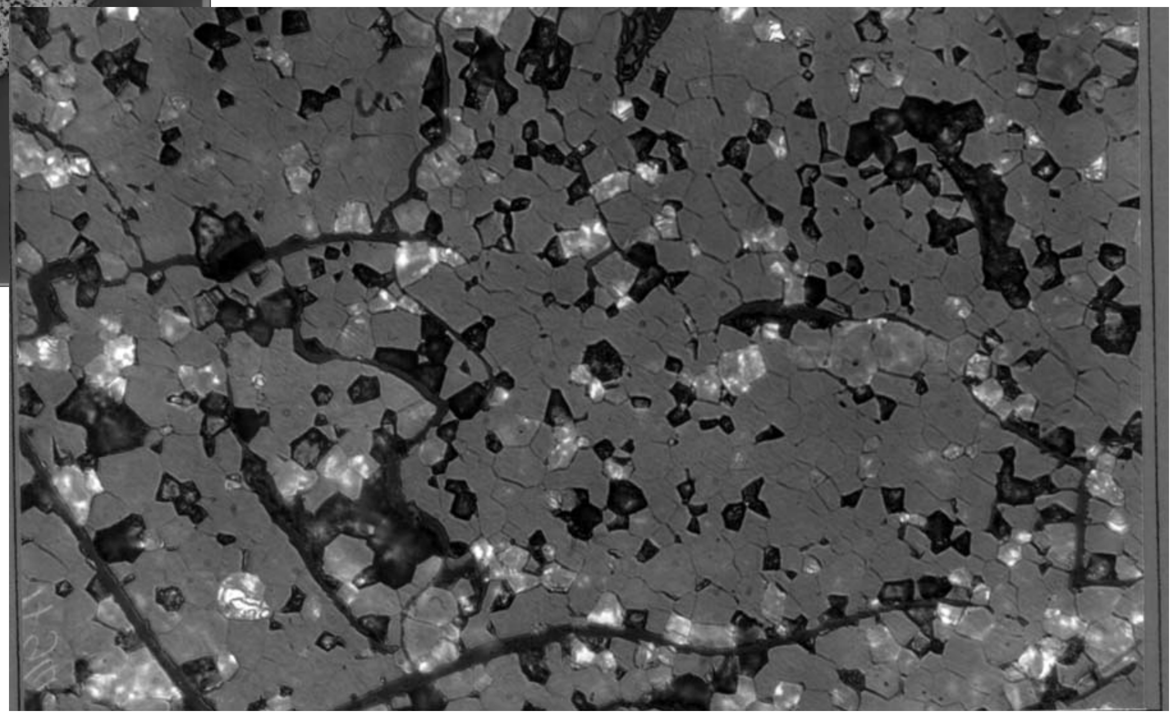
($Zr,Y,U)O_2$ Kernels
ϕ= 770 µm

U Soln 200 g/l

Porous Beads by Internal gelation



(Zr,Y,U)O2 Kernels
ϕ= 770 µm



2 phases present – Why ?

Fabrication Routes for Novel Pu or MA based kernels (xiii)

INFILTRATION Process

Limitations

An content determined by (a) porosity
(b) An Solution concentration

Porous kernels must be insoluble in acidic solutions

Fabrication Routes for Novel Pu or MA based kernels (xiv)

INFILTRATION Process

Improvements

Sphericity of porous beads by external gelation
Cracks in products from kernels produced by internal gelation

Maximising the actinide content per infiltration cycle
- Pu 200 g/l limit (155 g/l used so far)
- Am ca 400 g/l limit
- Complete kernel infiltration (not just periphery)

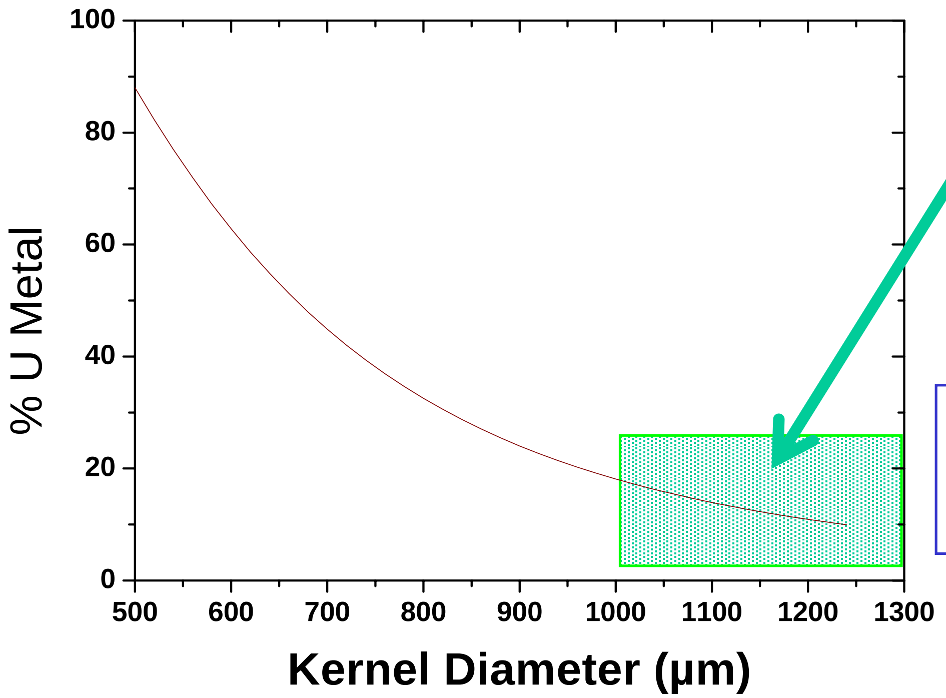
Pareto 20: 80 Rule (even over optimistic here)
- Much to do before process is ready to meet HTR specs.

Backend of the Fuel Cycle

- Geological Disposal
  - YSZ corrosion resistant
- Reprocessing
  - YSZ difficult to dissolve - Cost

Conclusions

- Dilution of Pu and MA in Stabilised zirconia can give performance advantages (burnup)
- Infiltration is a very efffective means to produce $(Zr,Y,Pu)O_2$
  - No wastes, Flexibility
  - Incorporation of Er in $(Zr,Y,Er)O_2$ precursor porous beads
  - Can process be applied for LEU (<20% ^{235}U)? Yes, but.......



Very big kernels