Simulated UK Advanced Gas Reactor Fuel by Hot Isostatic Pressing (HIP)

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Introduction

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AGR SIMFuel

Compounds	25GWd/t U		43GWd/t U	
Precursors	at.%	wt%	at.%	wt%
UO_2	97.587	98.193	95.870	97.586
Nd_2O_3	0.483	0.605	0. 827	0.364
ZrO_2	0.369	0.169	0.602	0.279
MoO_3	0.334	0.179	0.566	0.307
RuO_2	0.257	0.127	0.454	0.228
$BaCO_3$	0.244	0.179	0.435	0.323
CeO_2	0.193	0.124	0.329	0.213
La_2O_3	0.096	0.117	0.160	0.196
SrO	0.032	0.012	0.050	0.019
Y_2O_3	0.041	0.034	0.064	0.054
Cs_2CO_3	0.191	0.231	0.309	0.379
TeO_2	0.044	0.026	0.079	0.047

The compositions are calculated by FISPIN in at% for 25GWd/t and 43GWd/t .
(Z. Hiezl et al./Journal of Nuclear Materials 456 (2015))

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Bulk Property

Density and grain size of UO_2 pellet (Z. Hiezl et al./Journal of Nuclear Materials 456 (2015))

Parameters	AGR fuel pellet	Low doped pellet	High-doped pellet
Relative density(%)	97.2 (98.5)	91.2	91.7
Mean fine pore area(%)		15.4	14.2
Grain size(μm)	3.0~30.0 (SNF)	3.6~4.8	1.6~4.2

Microstructure Characterization

Fission products behaviours

- Dissolved in the UO_2 matrix √
- Oxide precipitates √
- Metallic precipitates √
- Volatile elements ×

Sintered at reduced atmosphere of 99.5 vol.% H_2 and 0.5 vol.% CO_2

SPS for incorporation of volatile fission product into simulant fuel pellet but not densified.
(T. Wangle et al. / Journal of Nuclear Materials 466 (2015))

Hot Isostatic Pressing

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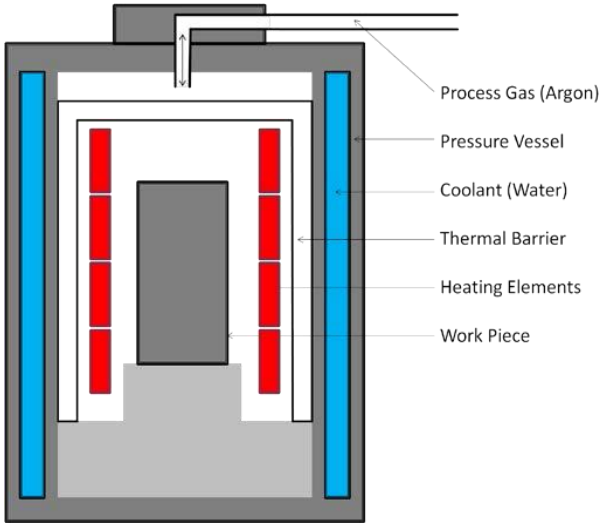
Characteristics

- Independent and simultaneous pressure and temperature cycle
- Isostatic pressure application



Advantages compared with normal sintering method

- Porosity elimination and higher density
- Relatively homogenous material
- Closed system without volatile gas release

Experimental

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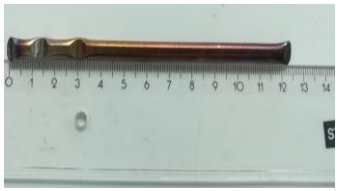
Preparation
- Batch Weighed
- Planetary milled for 10 mins
- Dried at 80 °C

HIP
- Baked out at 500 °C, 5h
- 10 °C/min ramp, 1250 °C, 5h
- 200 MPa, Argon

Characterization
- XRD
- SEM
- Raman

HIP preparation and operation

Packing into the tube → Baking out → Evacuating → Welding → HIP cycle



- Reduced atmosphere
- Hermetically sealed for closed system
- Water evaporation below the Cs_2CO_3 decomposition

Result-XRD

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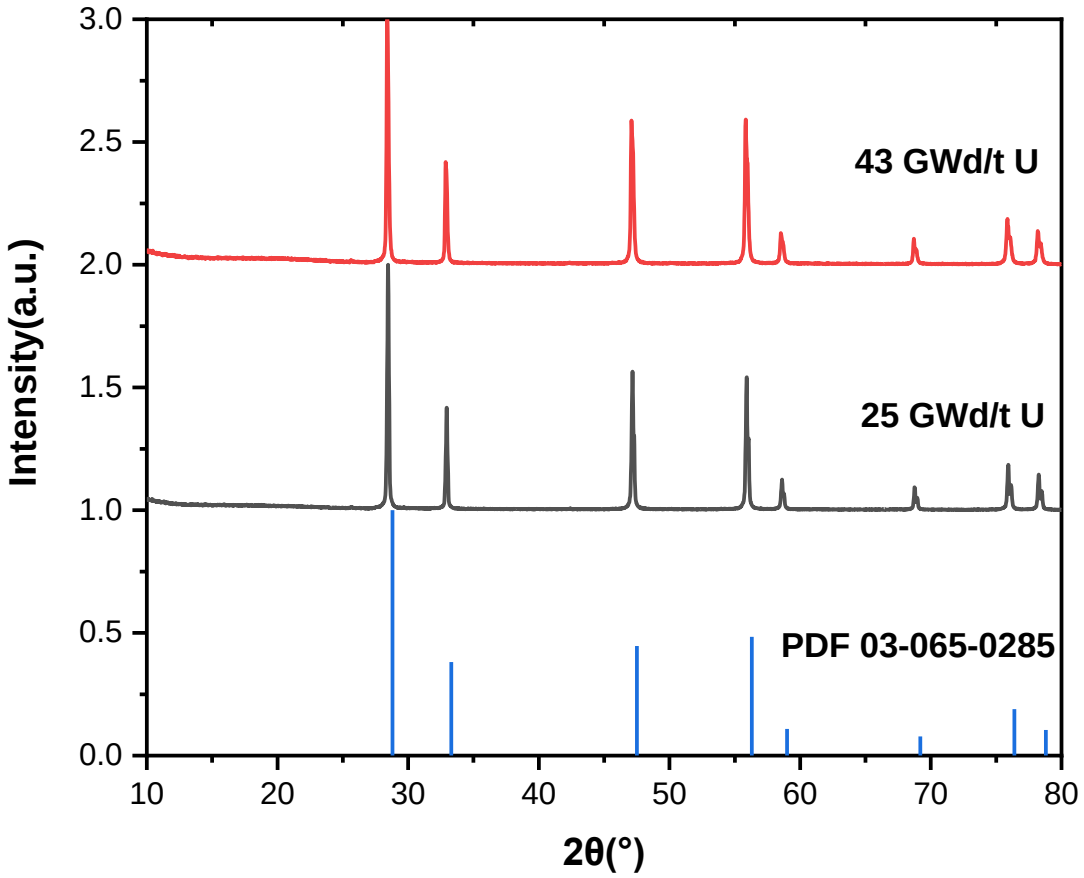
Phase identification

Peaks of two burnup SIMFuels are corresponding to UO_2.

- No obvious interaction between tube and pellet
- Amount of fission products are tiny

Refinement

Lattice parameters increase with the extent of burnup.



	25 GWd/t U	43 GWd/t U
Lattice Parameter a(Å)	5.472	5.475
Errors	0.00006	0.00008
Rwp	9.585	9.616

Result-XRD

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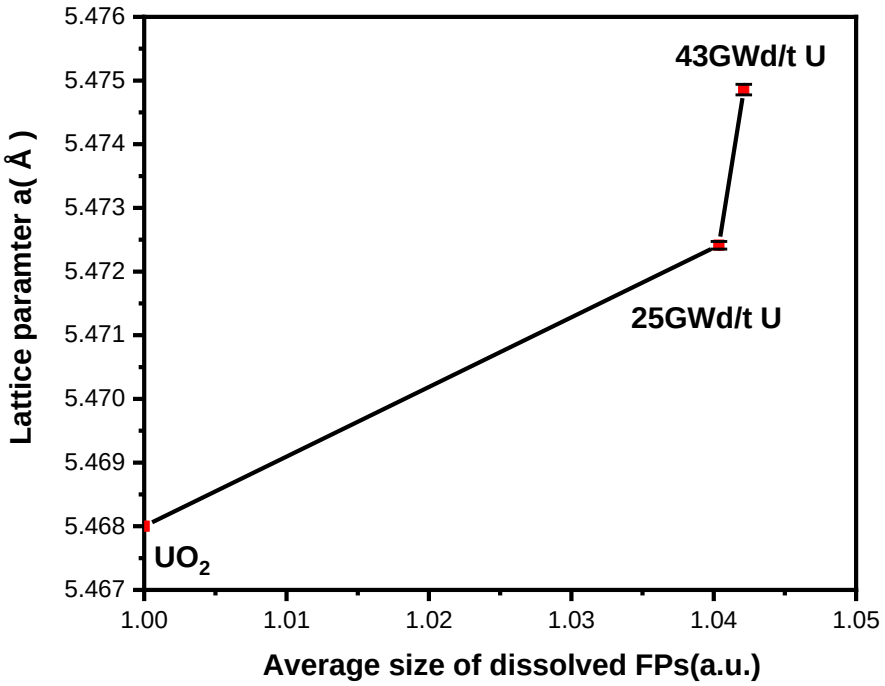
Lattice parameter

Fission products (FPs) dissolved in the UO_2 matrix enlarge the unit cell and the lattice parameter with larger ionic radius.

Element	Coordination number	Ionic radius
U^{4+}	VIII	1.000
Nd^{3+}	VIII	1.109
La^{3+}	VIII	1.160
Ce^{4+}	VIII	0.970
Zr^{4+}	VIII	0.840
Y^{3+}	VIII	1.019

	25GWd/t U	43GWd/t U
Average Ionic Size	1.040	1.042



Not strict linear relationship due to the different solubility of dissolved FPs.

Result-Raman

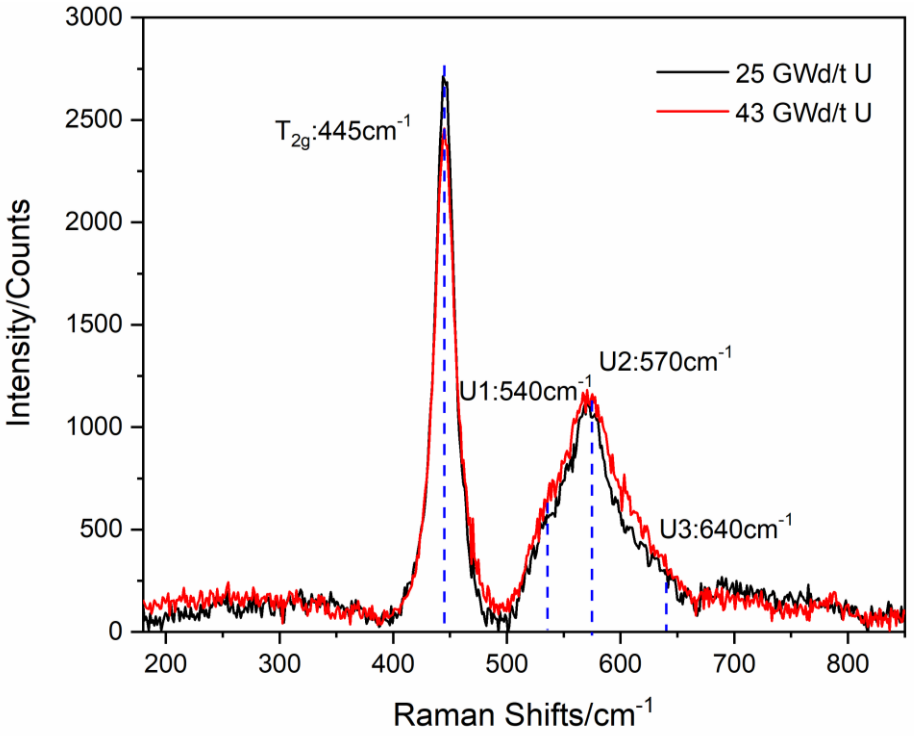
Bands identification

T_{2g}: U-O stretching

U1: Oxygen defects

U2: Longitudinal optical (LO)

U3: Oxygen Interstitials



- T_{2g} intensity decrease shows weak U-O stretching indicating the more dopants in UO_2 matrix.
- U1 peak shows larger oxygen defects
- U2 peak increase reflect a more disordered lattice crystal
- U3 peak shows no obvious oxygen interstitials

Element	Valence
Nd	3
La	3
Ce	4
Zr	4
Y	3

Result-Raman

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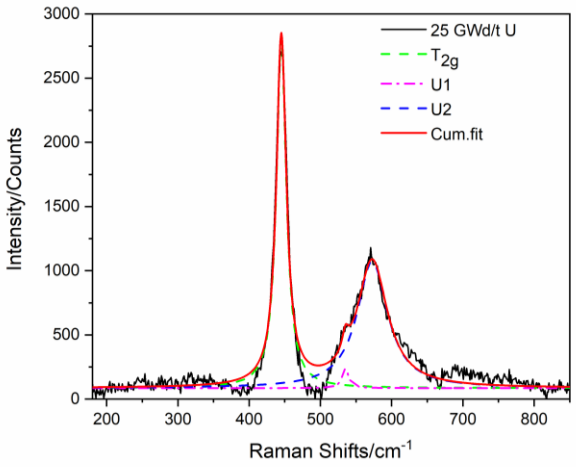
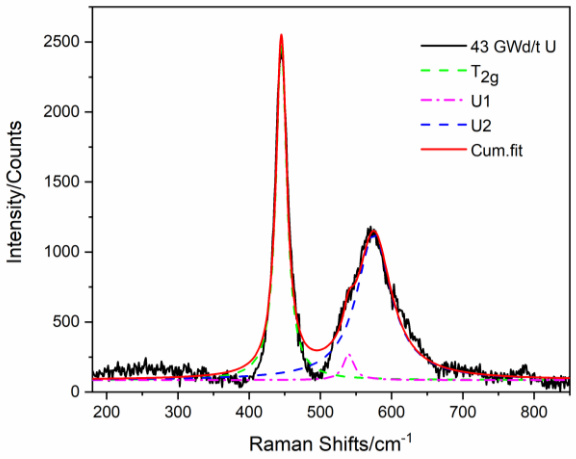
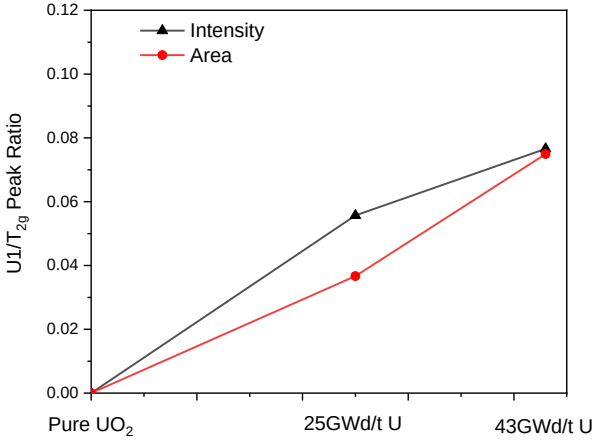
Deconvolution

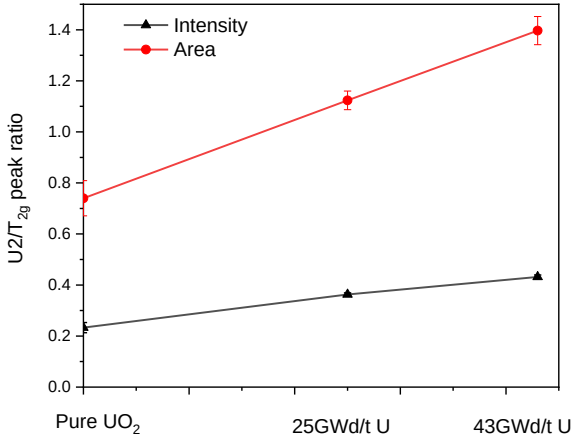
- There is no peak at U3 position for both spectra showing no oxygen interstitial.





- The ratio of U1/T_{2g} and U2/T_{2g} peak and intensity increase with burnup matching the more FPs dissolved in UO_2 matrix.

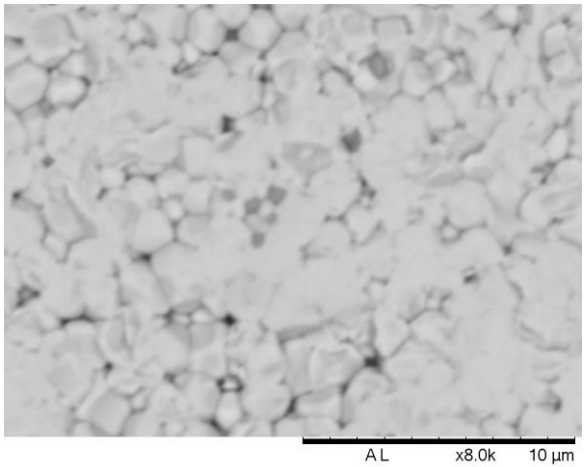




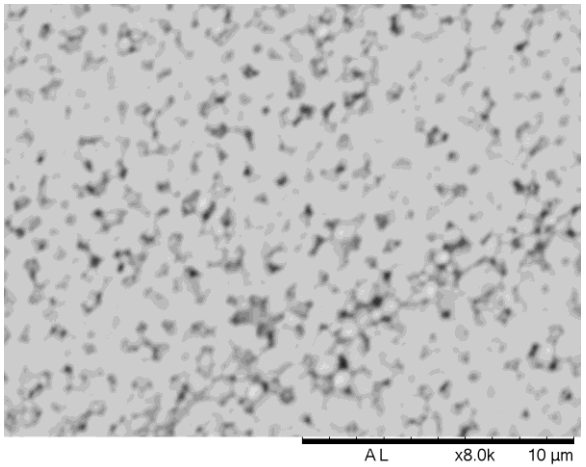
Result-SEM

Porosity calculation

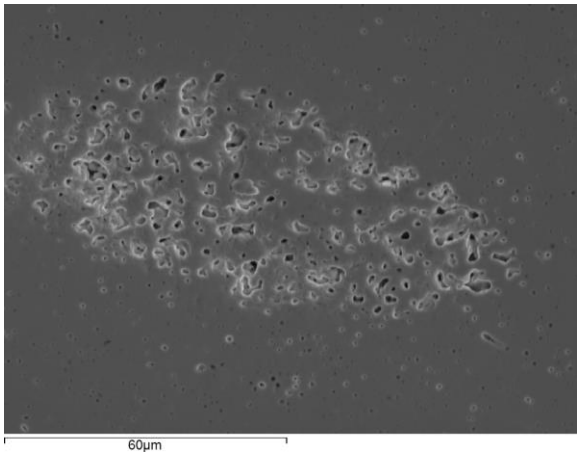
(Z. Hiezl et al./Journal of Nuclear Materials 456 (2015))



25 GWd/t U



43 GWd/t U



Normal sintering method

	HIP		Normal Sintering	
	25GWd/t U	43GWd/t U	25GWd/t U	43GWd/t U
Porosity	2.173%	2.849%	5.45%	4.83%
Standard Deviation	0.327%	0.326%	2.94%	0.94%

- The porosity of HIP is lower than that of normal sintering method
- The porosity increase and grain size decrease due to the more precipitates formation

Result-SEM&EDX

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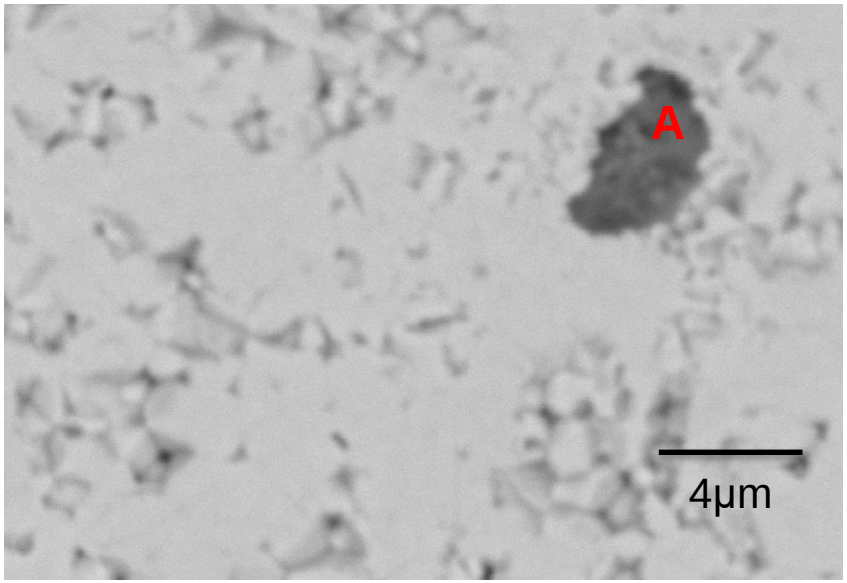
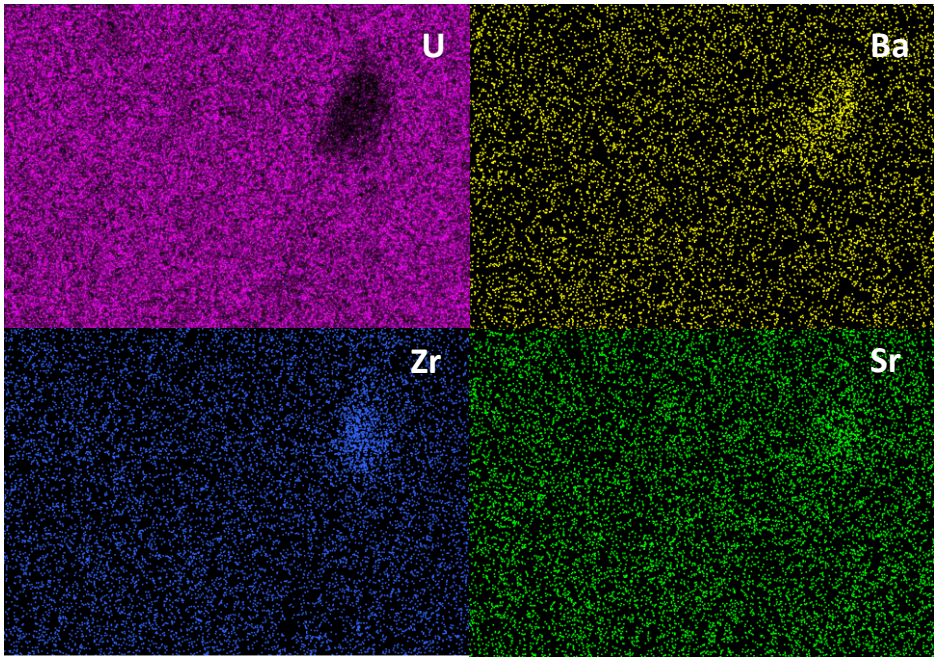
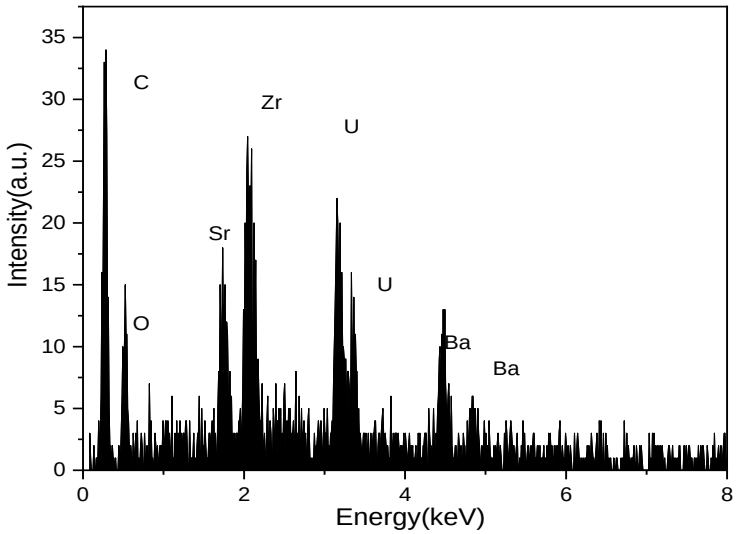
Oxide precipitates





$BaZrO_3$-containing oxide precipitate with significant amounts of $SrZrO_3$



Result-SEM&EDX

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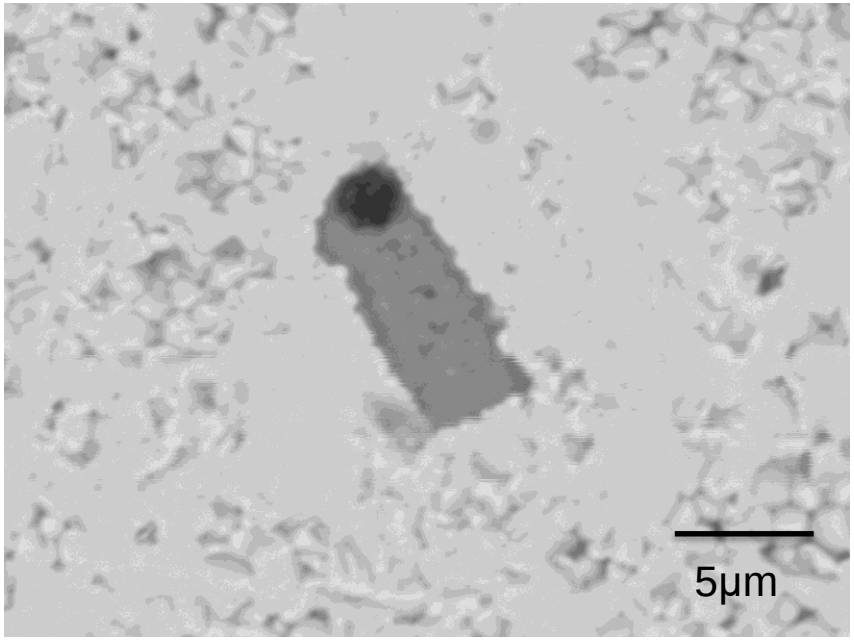
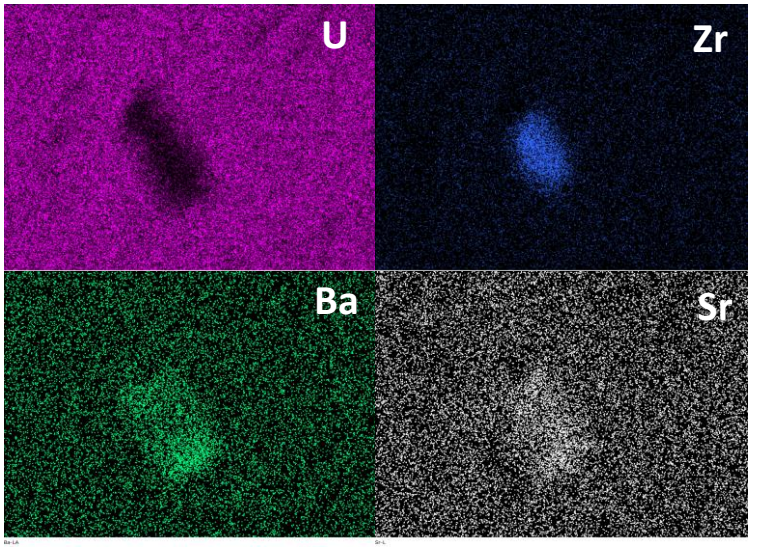
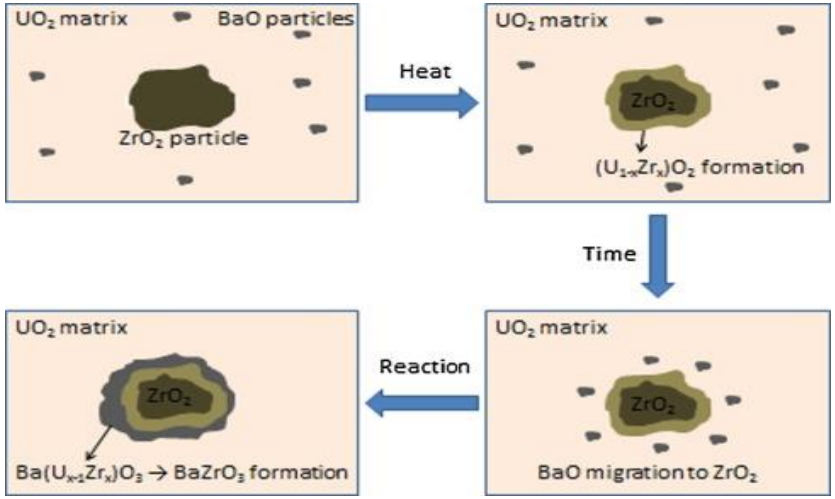
Oxide precipitates







Edge of ZrO_2 was dissolved in UO_2 matrix and then enveloped by BaO and SrO formed a growth zone dominated by ZrO_2 , $(U,Zr)O_2$ and $BaZrO_3$ or $SrZrO_3$ from the center to the edge.

(Z. Hiezl et al./Journal of Nuclear Materials 456 (2015))

Result-SEM&EDX

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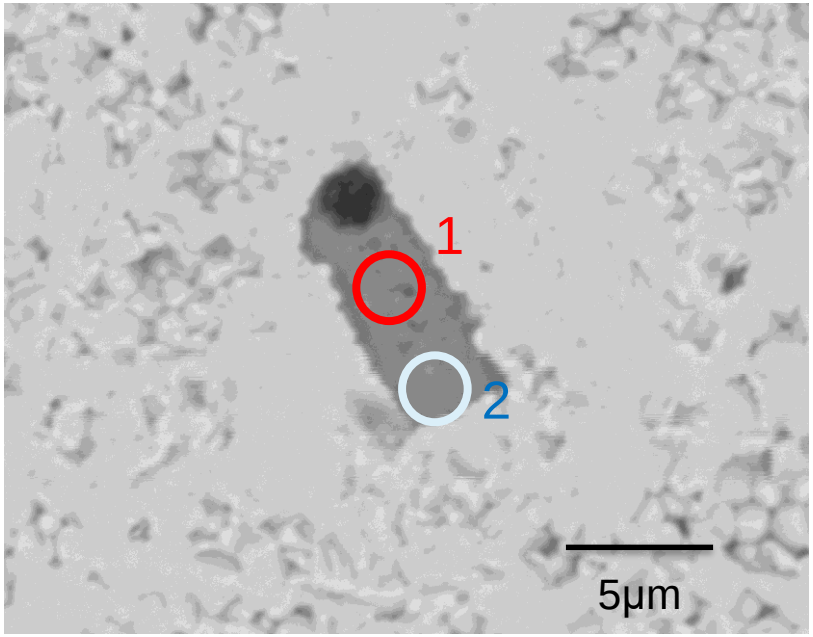
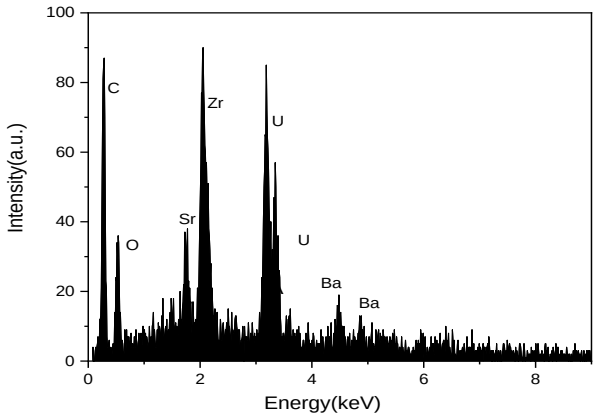
Oxide precipitates



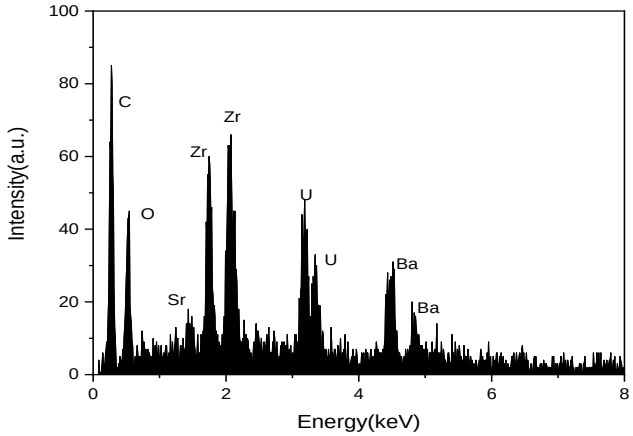
Point 1

Element	Ba	Zr	U	Sr	O
Ratio(%)	8.71	20.53	39.51	2.03	13.02



Point 2

Element	Ba	Zr	U	Sr	O
Ratio(%)	20.03	15.74	24.79	20.01	14.50



- U and Zr decrease from the center to the edge.
- Ba and Sr follow the inverse trend.

Result-SEM&EDX

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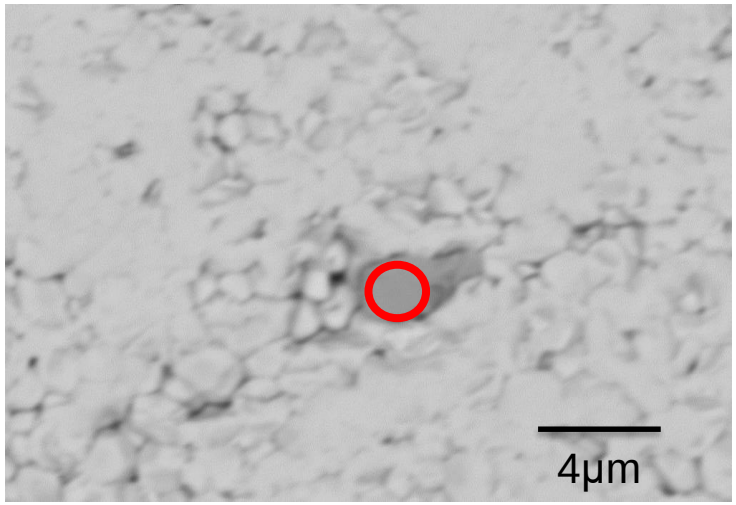
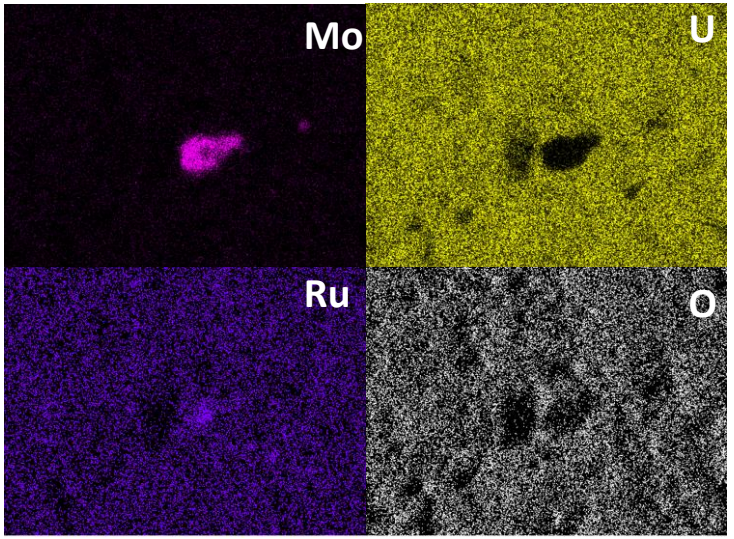
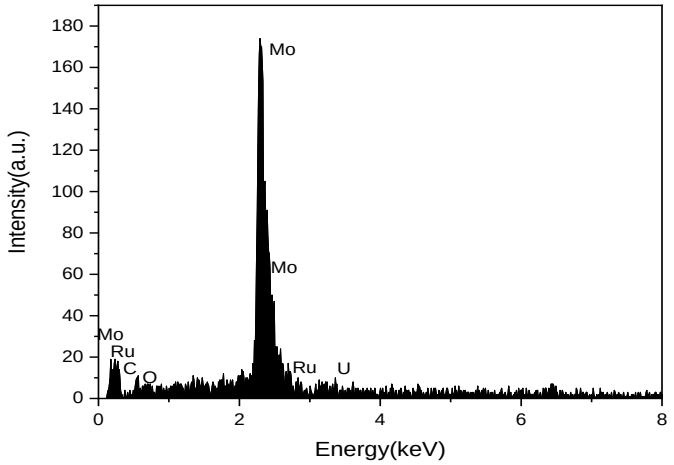
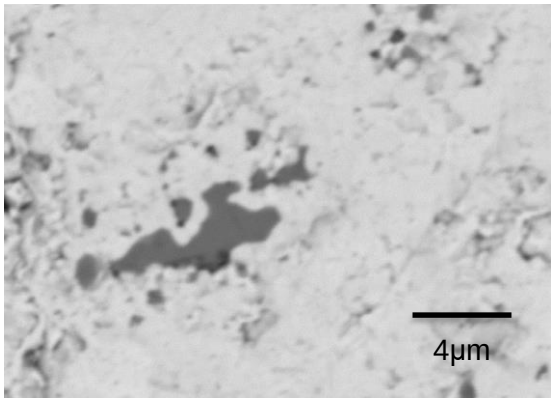
Metallic precipitates





Element	Mo	Ru	U	O
Ratio(%)	63.02	8.66	3.03	7.53





- Mo-Ru Metallic precipitates
- Varity of morphologies

Result-SEM&EDX

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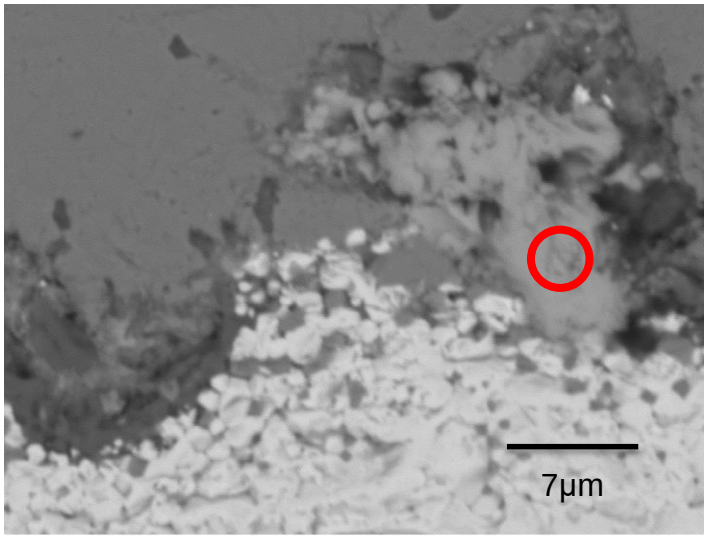
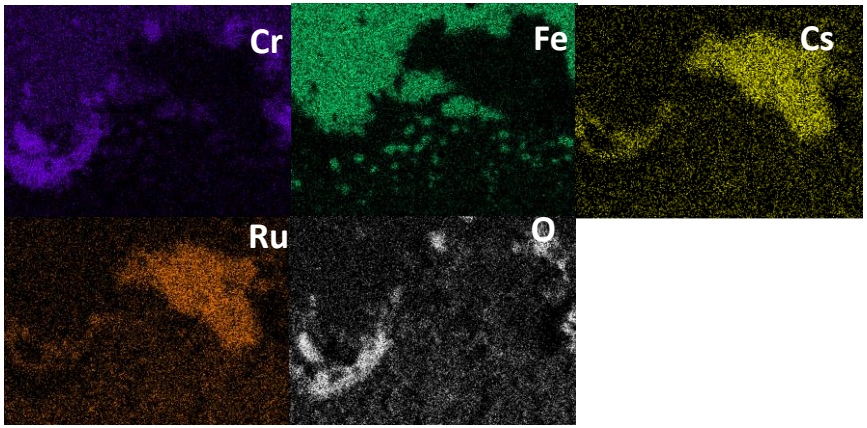
Volatile fission product: Cs



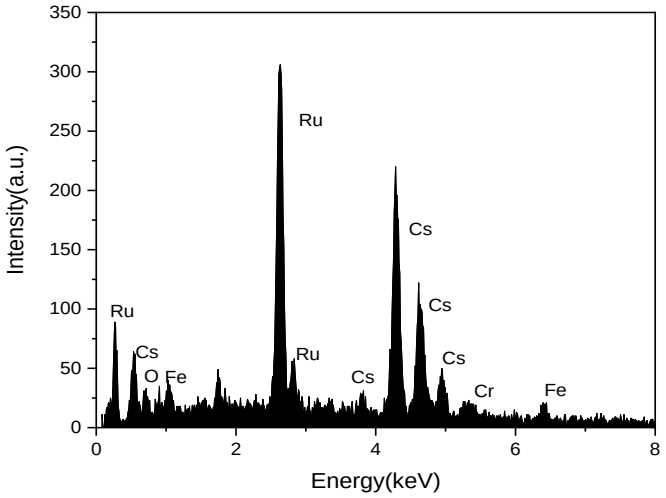
Tube

Pellet



Element	Fe	Cr	Cs	Ru	Ni	O
Ratio(%)	3.35	0.84	49.11	20.01	0.88	7.64

- Tube is made of stainless steel 316 (16Cr/10Ni) similar to the AGR stainless steel cladding (20Cr/25Ni)
- Cs was captured by Ru forming the metallic phase along the gap between tube and pellet.



Result-SEM&EDX

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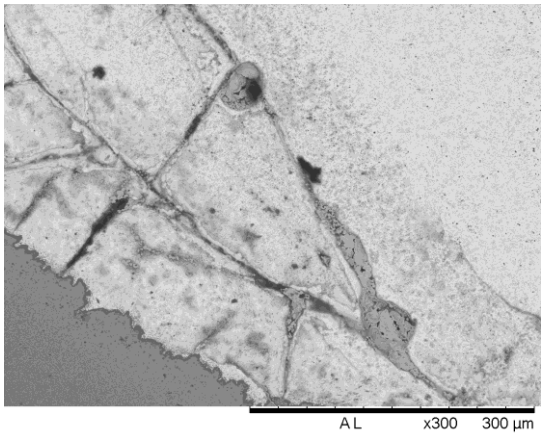
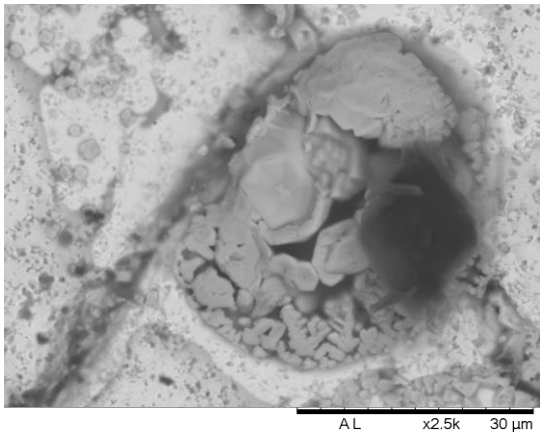
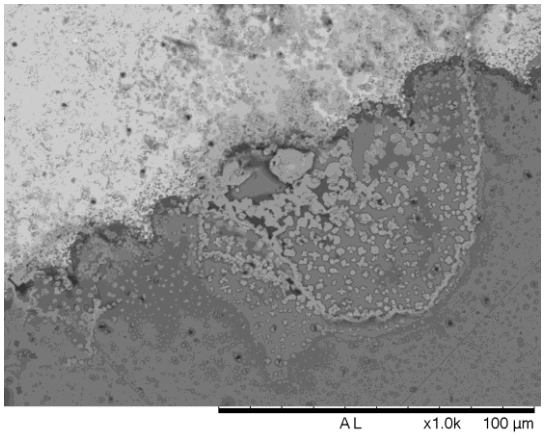
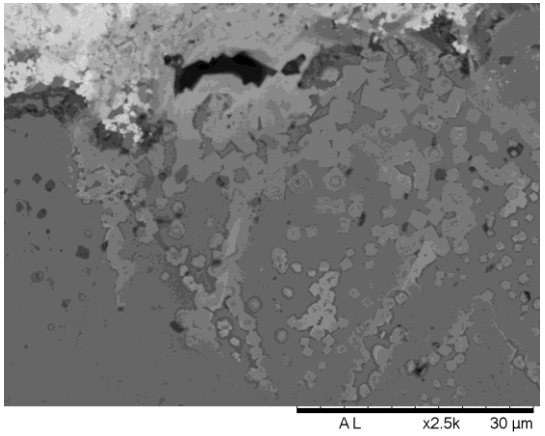
Volatile fission product: Cs









- The channel for Cs release was observed.
- Cs diffused into stainless steel tube with Ru.

Conclusion & Future work

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- Bulk property
  - Higher density & lower porosity
  - Larger grain size
- Fission product behaviour
  - Dissolved in UO_2 matrix
  - Oxide precipitates
  - Metallic precipitates
  - Volatile elements
- Large batch SIMFuels fabrication
  - Small precursors grain size
  - Sufficient mixing
  - Detailed characterization
- Dissolution experiment
  - Cs behaviour
  - Geological disposal
  - Reprocessing potential

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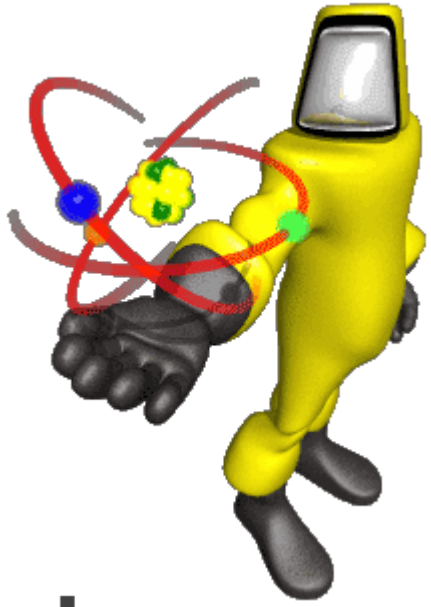
Thank you for listening!

Acknowledgment

Supervisors: Prof. Neil Hyatt and Dr. Claire Corkhill

ISL Group members



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