Behavior of Zirconium on Passivated U-Zr Based Fuel Alloys

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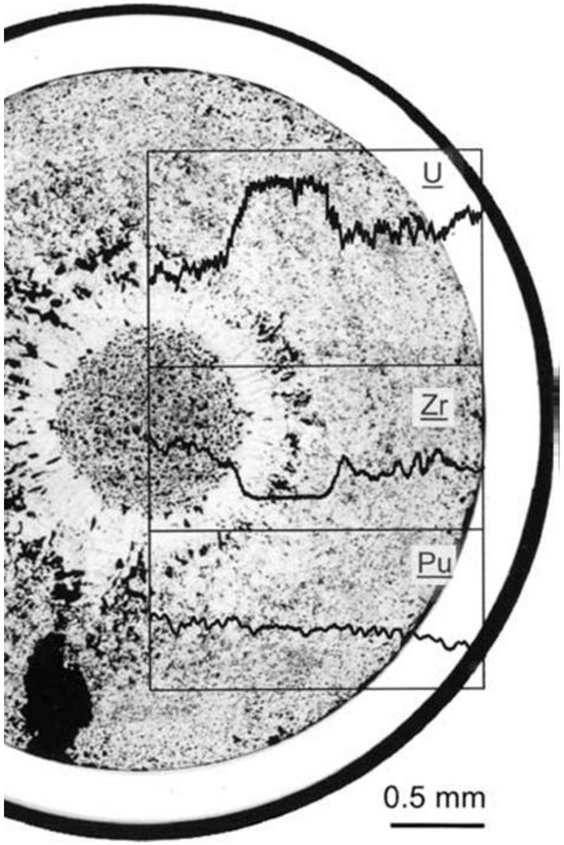
Robert Mariani, Leah Squires, Thomas Hartmann,*
Dawn Janney, Spencer Taylor, Thomas O'Holleran,
J. Rory Kennedy
INL, Nuclear Fuels and Materials Division
*University of Nevada, Las Vegas, NV

Presentation Overview

- Range of zirconium behaviors
- Potential problem for Zr-alloy fuels
- Ternary U-Pu-Zr alloys cast and examined
- Sample preparation methods
- XRD Rietveld refinement results
- Preliminary SEM results
- Conclusion and planned further studies

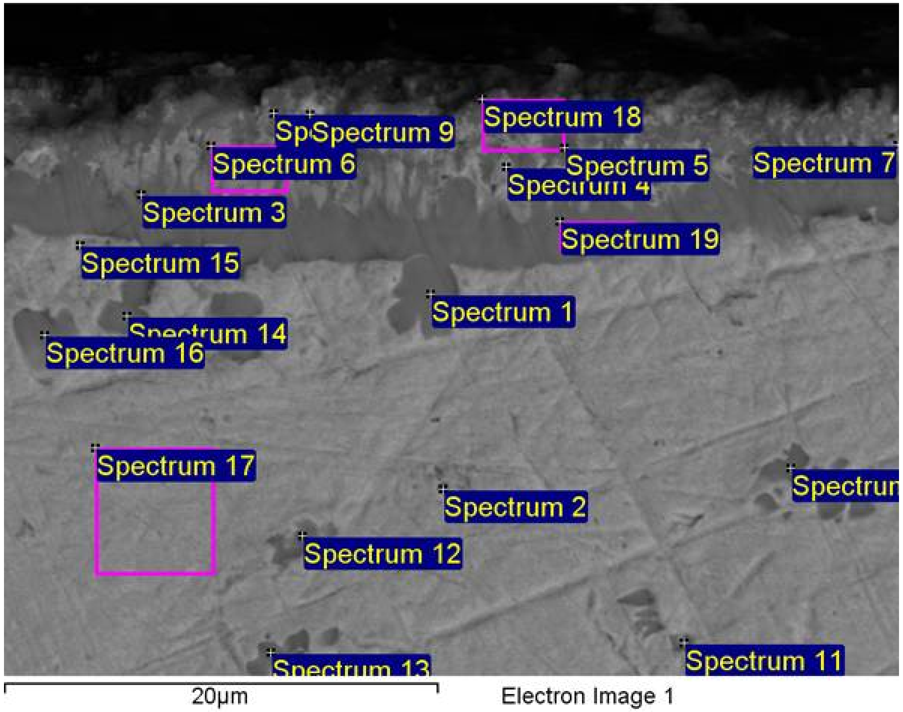
Zirconium Behavior in Metallic Fuels

- **Constituent Redistribution**
  - **In-pile, thermal and chemical potential gradients drive Zr redistribution in three fuel zones**
- **Zr rind formation on fuel surfaces**
  - **Observed for fresh fuels**
  - **Apparently accentuated for annealed and irradiated fuels**
- **Four distinct zones in irradiated fuel: rind plus three zones for constituent redistribution**



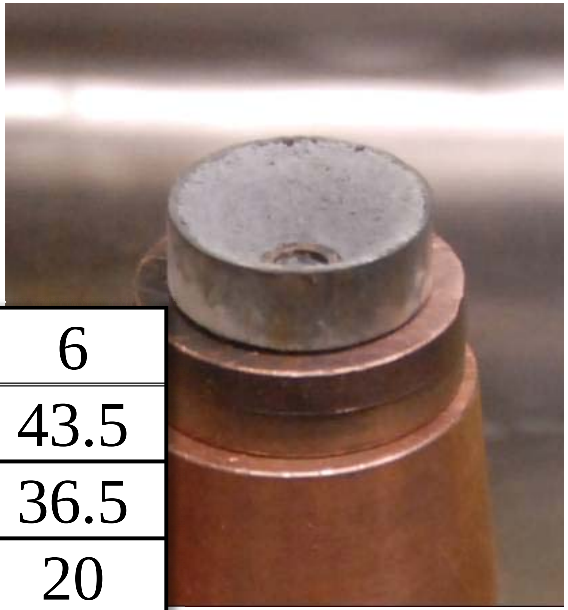
Y.S. Kim, G.L. Hofman, S.L. Hayes, Y.H. Sohn, *J. Nuc. Mat.* 327 (2004) 27-36.

Rind Microstructure, Alloy 1 As-Cast
50U-30Pu-20Zr wt%



Marker	Si	U	Pu	Zr
1	0.42	2.92	4.79	91.87
2	12.48	47.34	28.24	11.94
3	0.80	3.99	9.40	85.82
4	3.56	22.68	43.76	30.01
5	10.24	4.24	13.64	71.87
6	2.30	17.26	40.71	39.73
7	1.68	58.09	27.04	13.19
8	10.07	12.56	28.79	48.59
9	2.30	37.78	49.66	10.27
10	0.31	10.61	7.02	82.06
11	0.48	23.69	12.84	62.99
12	0.37	20.67	10.09	68.86
13	0.70	22.74	11.59	64.98
14	0.84	4.00	5.08	90.09
15	7.53	52.92	26.66	12.88
16	0.44	9.30	7.12	83.13
17	0.12	53.01	28.52	18.36
18	8.00	17.72	32.95	41.33
19	0.41	4.34	4.57	90.67

Alloys Cast and Tested
(wt%)

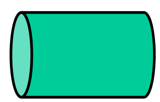


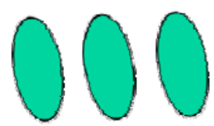
Alloy	1	2	3	4	5	6
U	50	25	35	63.5	61	43.5
Pu	30	55	20	20	24	36.5
Zr	20	20	45	15	15	20
Ln	-	-	-	1.5	-	-
Measured (ICP-MS, ± 5% at two-sigma)						
U	49.6	23.4	33.9	63.4	59.3	41.8
Pu	28.5	52.7	19.1	19.7	23.1	34.9
Zr	19.8	20.1	43.3	15.0	14.4	20.3
Ln	-	-	-	1.45	-	-

Zr Behavior Studies
XRD, SEM and Annealing Methods

- **As- Cast Alloy Samples**
- **Pellet Anneal Samples**
  - **Section**
  - **Then Anneal**

- **Slug Anneal Samples**
  - **Anneal**
  - **Then Section**



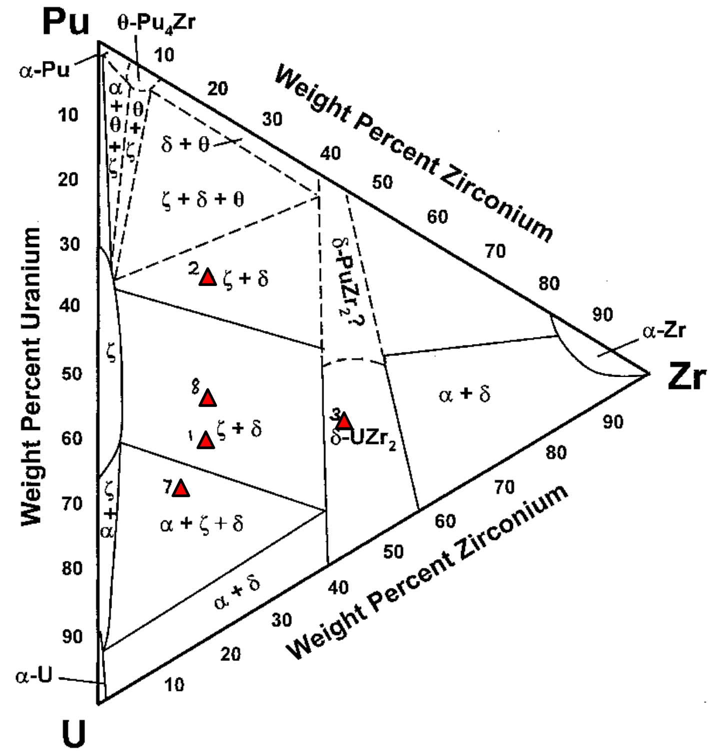


- **Samples should give the room temperature equilibrium phases if cooled slowly enough.**
- **The two annealing methods would give a different surface phase if the oxidation layer on pellet anneal samples does indeed influence α-Zr formation.**

(In each XRD room temp. measurement, three sample disks are sectioned to increase signal.)

XRD Rietveld Refinement of Alloys

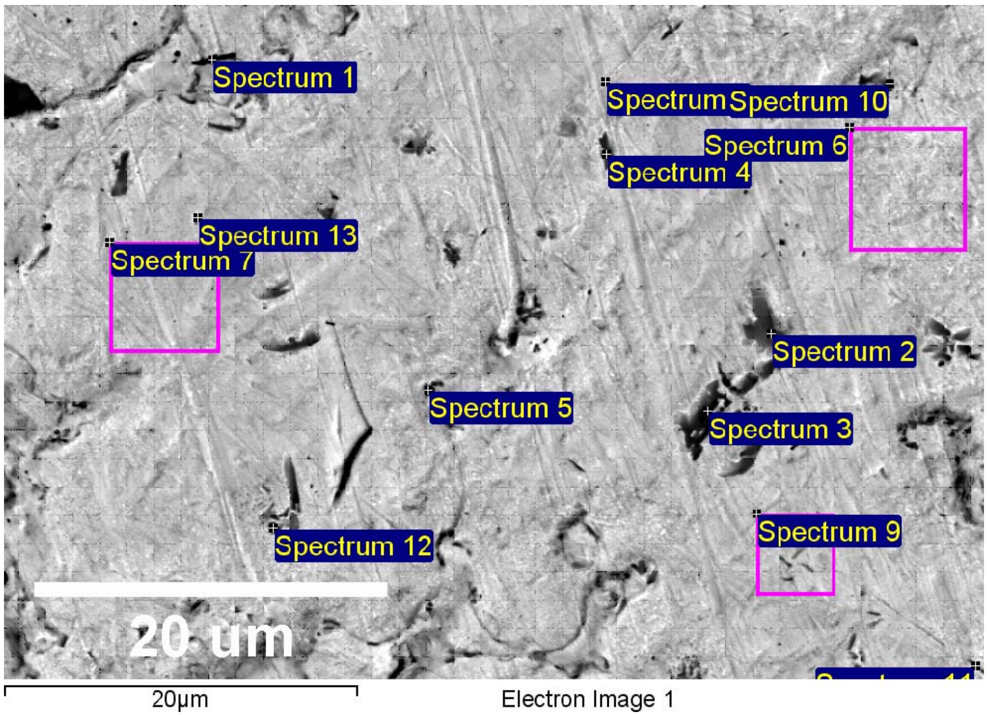
- As-cast fuels (i.e., prior to annealing) show no α-Zr.
- Pellet anneal samples show α-Zr
- Slug anneal samples show little or no α-Zr
- The ingress of α-Zr is attributed to heat treatment of a passivated, oxygen-bearing surface.



Rietveld Refinement Phase Analysis, wt%

		α U	γ U	ζ Pu-U	δ Pu	δ UZr2	β Zr	α Zr
Alloy 1: U-30Pu-20Zr	As-Cast		10	39		51		
	As-Cast, 2nd trial		13.21	16.23		70.6		
	Pellet Anneal	4.6		82.8		0.3		12.3
	Slug Anneal	31	69					
Alloy 2: U-55Pu-20Zr	As-Cast	2.8	0.4	20		77		
	As-Cast @ 650 C	0.6	4.8	29.6		57.9		7.2
	Pellet Anneal	11		24.6	4.1	1.2		59.1
	Slug Anneal	1.7	0.2	33.2		56.3	5.8	2.8
Alloy 3: U-20Pu-45Zr	As-Cast					100		
	Pellet Anneal	13.8	0.2	18.4	4	6.6		57
	Slug Anneal	1.4	0.1	1.8		96.8		
Alloy 4: U-20Pu-15Zr-1.5Ln	As-Cast		13.6	69.7		16.6		0.4
	Pellet Anneal- nm							
	Slug Anneal	3.2	8.4	65.2		23.2		
Alloy 5: U-24Pu-15Zr	As-Cast			75		25		
	Pellet Anneal	1.7		56.3		1.1		40.9
	Slug Anneal	15.4	19.6	0.1		55.6		9.2
Alloy 6: U-36.5Pu-20Zr	As-Cast			56.9		43.1		
	Pellet Anneal	5.5		40.8	4			49.7
	Slug Anneal		0	67.1		21.9	4.3	6.7

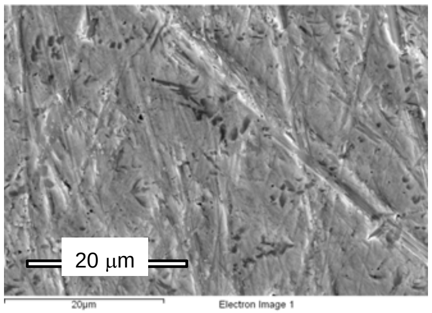
SEM Results for Alloy 1 As-cast
50U-30Pu-20Zr (wt%)

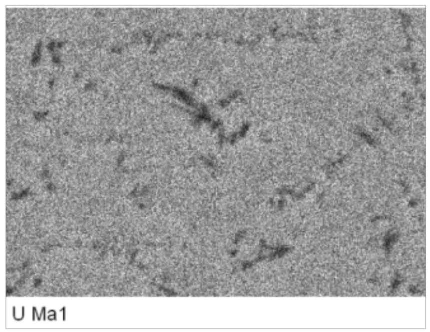


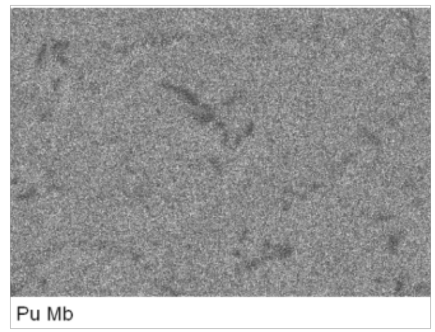
Spectrum	Si	Zr	U	Pu
1	13.8	15.8	44.6	25.8
2	0.4	77.1	13.3	9.2
3	0.4	79.2	13.5	6.9
4	0.4	68.7	20.0	11.0
5	0.4	48.7	35.3	15.6
6	0.3	19.0	51.4	29.3
7	0.4	25.3	44.8	29.5
8	0.3	25.7	48.3	25.8
9	0.3	28.4	42.9	28.4
10	0.4	50.0	32.4	17.3
11	0.2	19.1	50.3	30.4
12	0.4	46.6	35.6	17.4
13	6.7	23.1	41.7	28.5

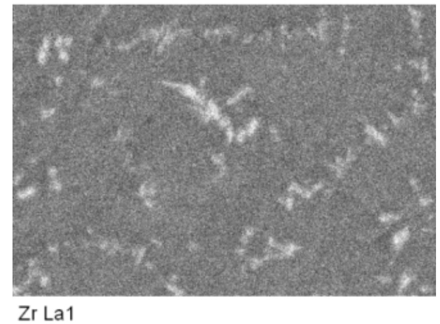
X-ray Maps For Alloy 3
35U-20Pu-45Zr (wt%)

As-Cast



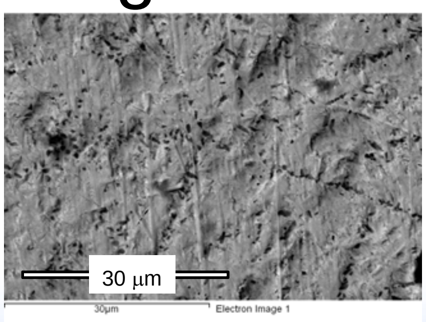


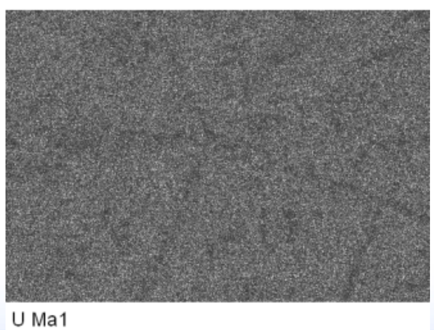


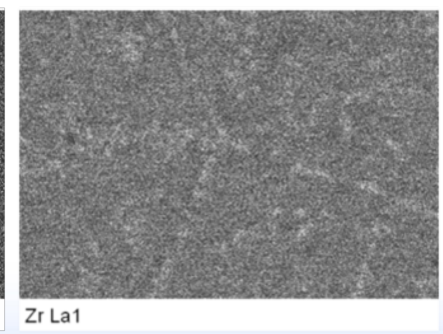


U Pu Zr

Slug Anneal







Improvements for Metallic Transmutation Fuels

- For metallic fuels that are never exposed to ambient air, an oxide passivation layer never forms.
- If a new casting method is employed, without oxide wash coatings on silica glass molds (such as ZrO_2 or Y_2O_3), even less oxide passivation is available.
- Under the above conditions, there is no driving force for an α-Zr layer to form.
- In the absence of the α-Zr, the extent of FCCI is greater and the fuel lifetime/safety may be impacted.
- Fuel slugs may possibly be passivated to some optimal level using a heat-treatment procedure.

Conclusions / Acknowledgements

Conclusions

- SEM/X-ray mapping of 'pellet anneal' samples may help assess extent of α-Zr formation, especially cross-sections that reveal depth-profiling.
- Casting of U-Pu-Zr alloys in argon-filled hot cell may need to undergo or may benefit from a separate passivation step.

Acknowledgments

- Messrs. Tim Hyde, Paul Hansen, Mark Lounsbury and Ms. Ginger Dexter prepared the alloys.
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