Microstructural Characterization of Irradiated Fuels

Jian Gan

Nuclear Fuels and Materials Division
Idaho National Laboratory

ATR User's Week-MeV Summer School
June 18-22, 2012, Idaho Falls, ID

Idaho National Laboratory

INL

Contributors

Dennis Keiser, Jr.
Brandon Miller,
Jan-Fong Jue,
Adam Robinson,
Pavel Medvedev,
James Madden,
Dan Wachs

Acknowledgements

The TEM work for RERTR program was supported through funding provided by the U.S. Department of Energy (DOE), Office of National Nuclear Security Administration (NNSA) to the RERTR program
at Idaho National Laboratory (INL), operated by Battelle Energy Alliance (BEA), LLC, under DOE Idaho Operations Office Contract DE-AC07-05ID14517.

Idaho National Laboratory

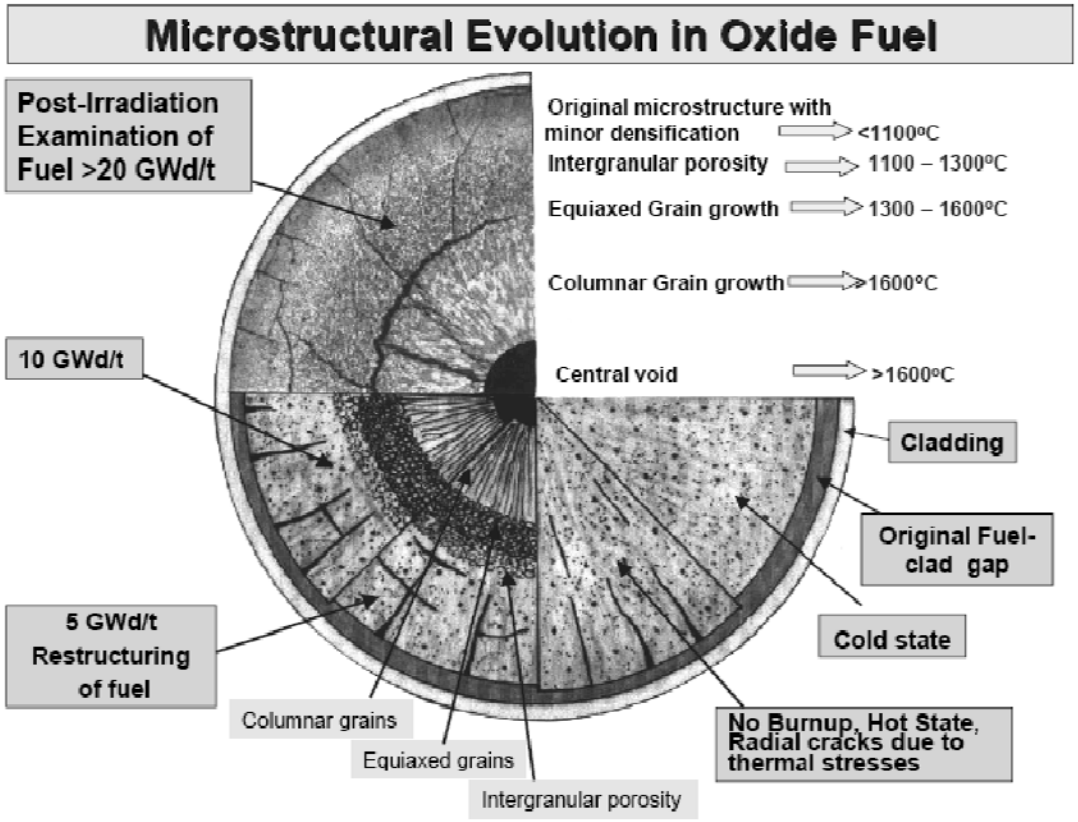
Outline

- Introduction.
- Microstructure of the irradiated fuel highlights.
- Sample preparation for SEM and TEM microstructural characterization.
- Microstructural characterization highlights.
- TEM characterization of irradiated fuels.
- Summary.

Relevant Lectures of The MeV School

Topic	Lecturer
Importance of Materials and Fuels	Steve Zinkle
Overview of Radiation Damage	Gary Was
Characterization of Irradiation Damage	Chad Parish
Characterization	Gene Ice
Electron Microscopy	Karren More

Microstructure of Fast Reactor Oxide Fuel



Graphic from S. Banerjee,
Bhaba Atomic Research Center

Estimate for UO_2 fuel Burnup:

1 at%BU of H.M. ≈ 10 GWd/tU
= 2.3×10^{20} fissions/cm^3

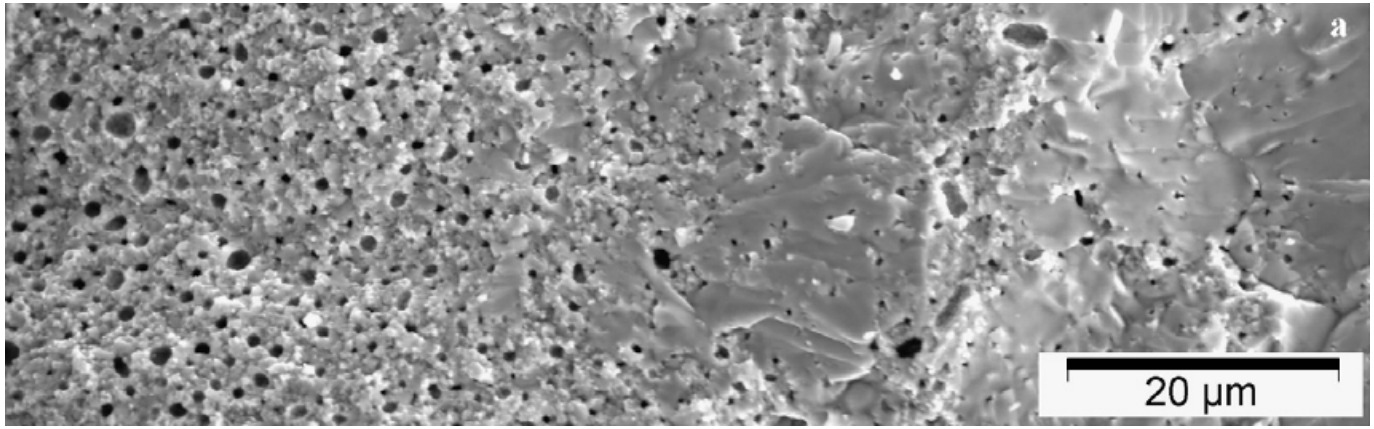
- A typical fresh LWR UO_2 fuel is enriched to ~6%U-235 and discharged from service with ~1%U-235 left.
- Therefore EOL burnup is approximately ~50 GWd/tU (for~ 7 cycles). This corresponds to atomic displacement damage of ~5,000 dpa, and introduces ~10% impurities (relative to metal sublattice)

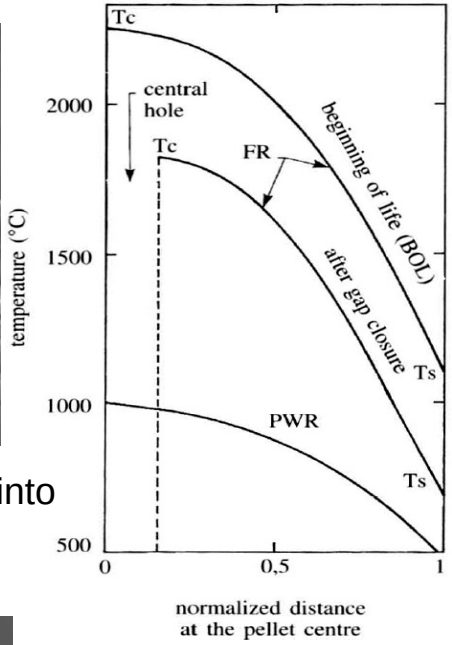
[Ref: Matzke, Nucl. Inst. and Meth. In Phys. Res. B65 (1992) 30]

Complex Microstructure in irradiated UO_2

SEM images

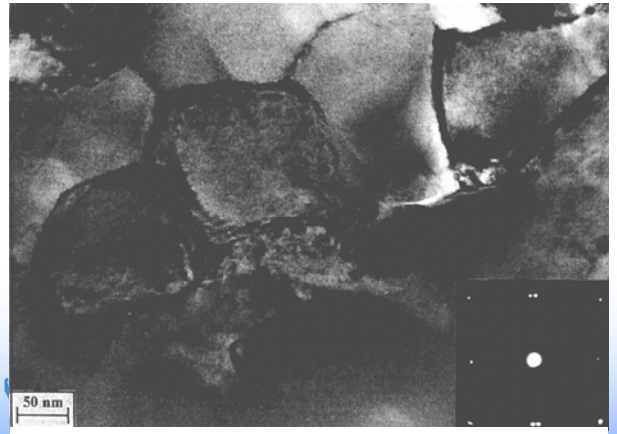
← Pellet edge Pellet center →



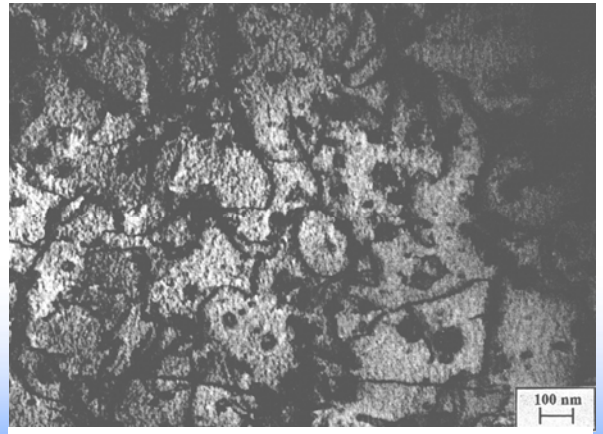


High local burn-up at rim near pellet surface. Original grain size ~ 10 µm divided into subgrains of 0.15-0.30 µm. The width of the rim may be up to ~ 1 mm at edge.

TEM images



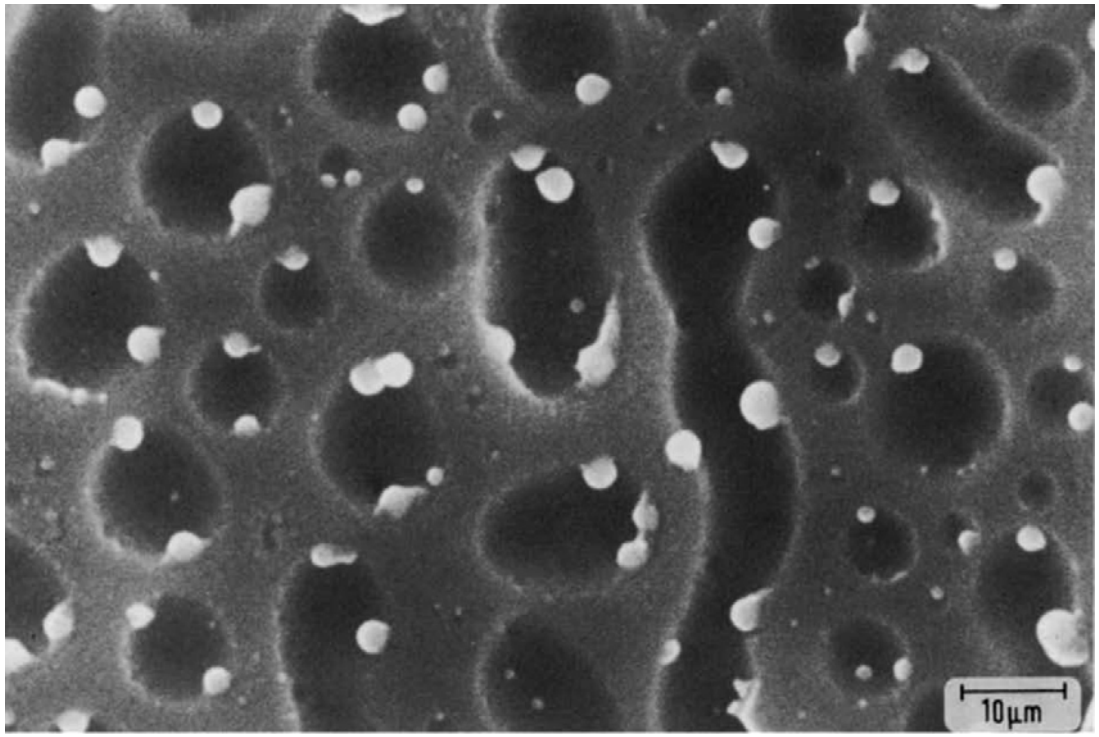
Grain subdivision in “rim” region near fuel pellet surface



Dislocations and loops near fuel pellet center

Ref:
[1] J. Noirot, L. Desgranges and J. Lamontagne, J Nucl Mater 372 (2008) 318.
[2] I.L.F. Ray, J Nucl Mater 245 (1997) 115.
[3] A. Berthet et al, “The Nuclear Fuel of Pressurized Water Reactors and Fast Reactors – Designe and Behaviour”, Edited by Henri Bailly et al

SEM of Grain Boundary Bubbles in LWR UO_2 Fuel



LWR UO_2 fuel showing grain boundary bubbles containing 5-metal particles.

The 5-metal precipitates consisting of: Pd-Rh-Ru-Tc-Mo.

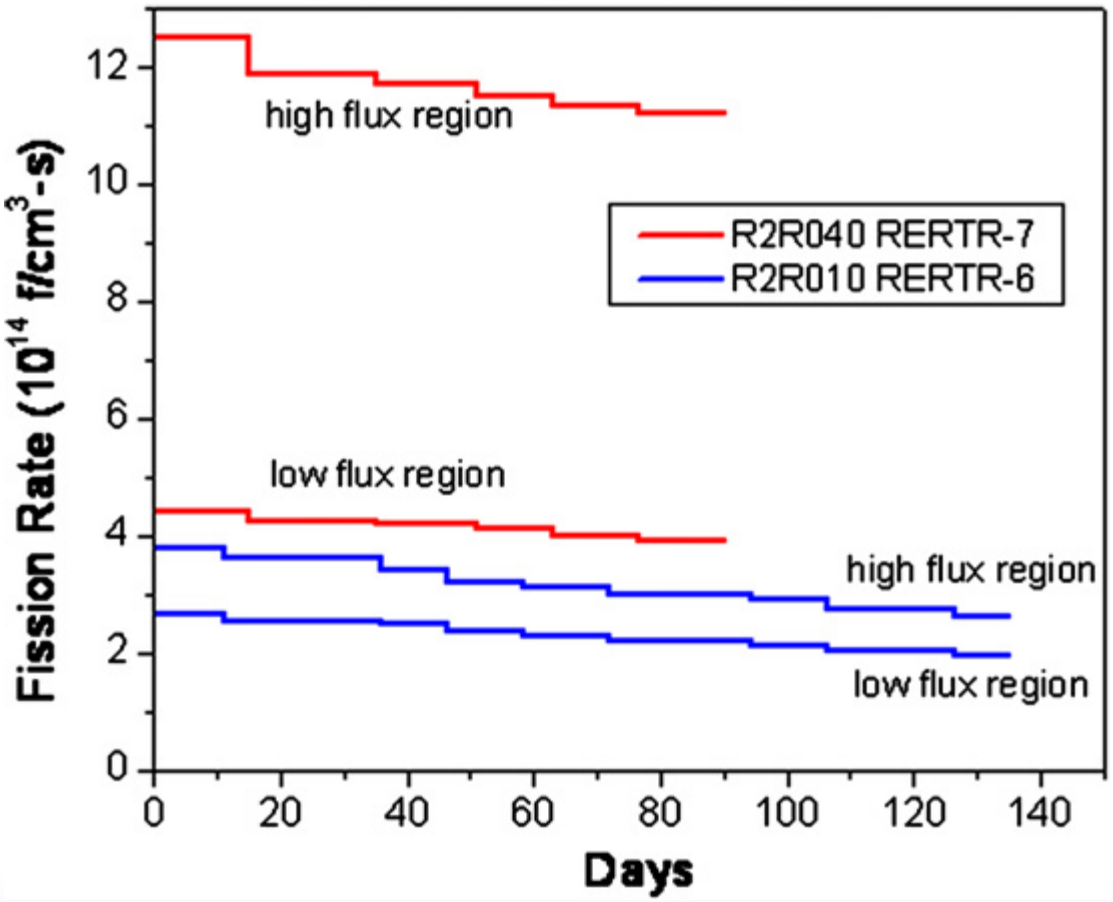
Ref: Hj. Matzke and H. Blank, J. Nucl. Mater., 166 (1989) p120

OBJECTIVES

- Develop TEM characterization capability for the irradiated fuel for RERTR fuel program.
- Investigate the irradiated microstructure as a function of fission density in U-Mo dispersion and monolithic fuel plates.
- Develop site-specific TEM characterization from an SEM sample using Focused Ion Beam (FIB) lift-out technique.
- Improve the understanding of the role of microstructure on fuel performance.

Microstructure Characterization → **Key to understand fuel performance**

Fuel Irradiation Condition in ATR

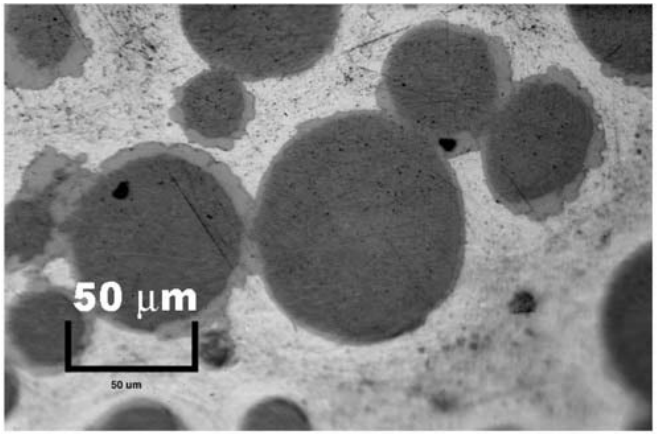


Keiser, et al.
JNM 425 (2012) 156

R2R010 (19% enrichment)
R2R040 (58% enrichment)

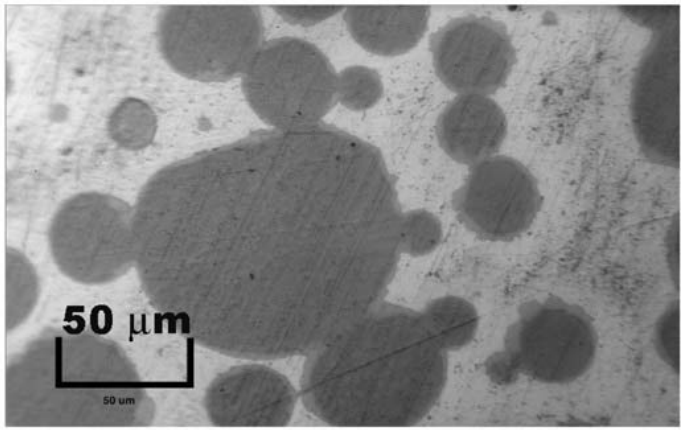
OM Images of Irradiated U-7Mo/Al-2Si

19% enrichment, 3.26×10^{21} f/cm^3 (avg)

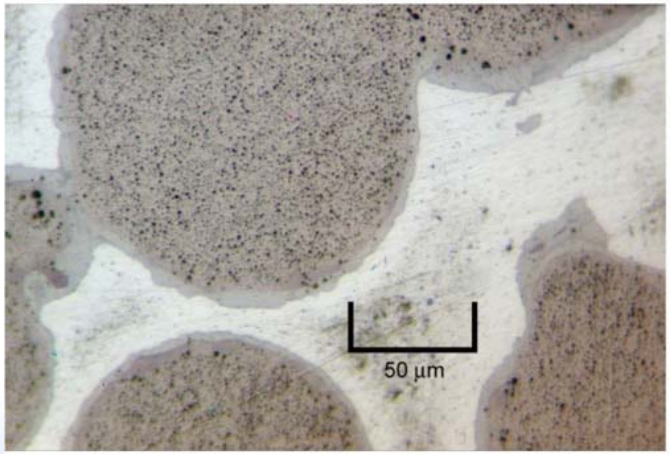


(a)

19% enrichment, 3.22×10^{21} f/cm^3 (avg)

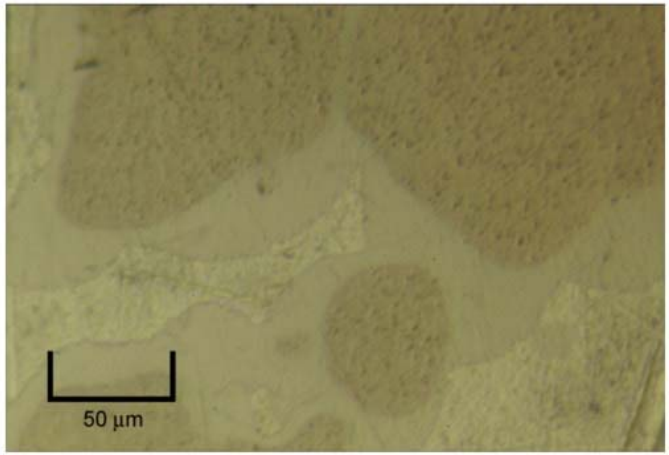


(b)



(c)

58% enrichment, 5.11×10^{21} f/cm^3 (avg)



(d)

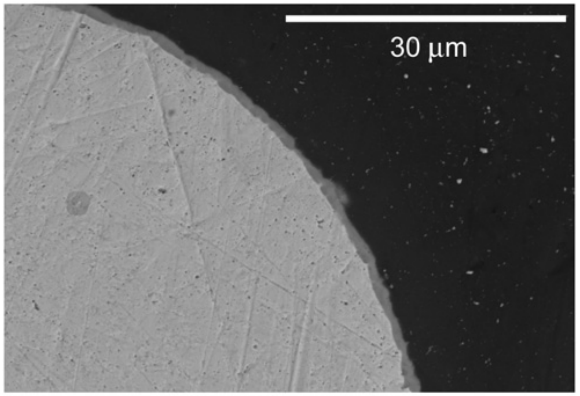
58% enrichment, 4.89×10^{21} f/cm^3 (avg)

Keiser, et al.
JNM 425 (2012)
p156

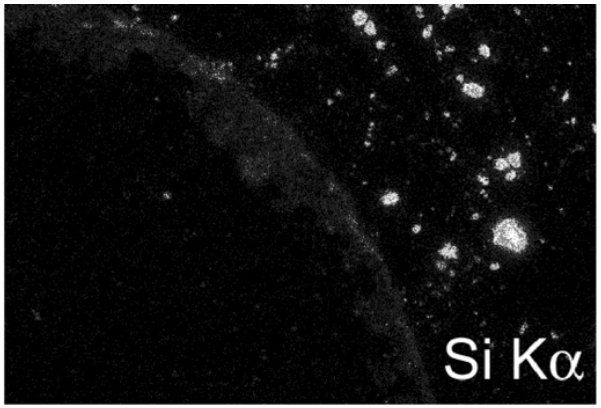
SEM Images of Irradiated U-7Mo/Al-2Si

Backscattered Electron image

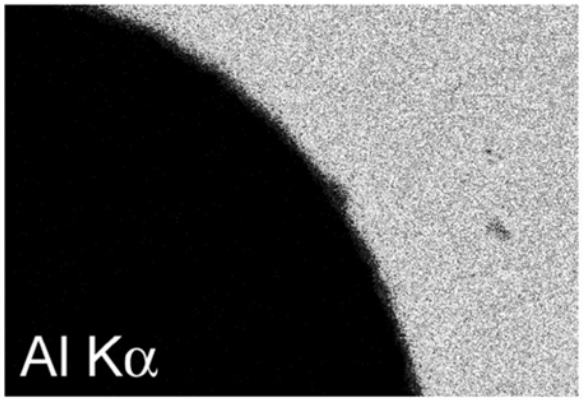
WDS X-ray Map



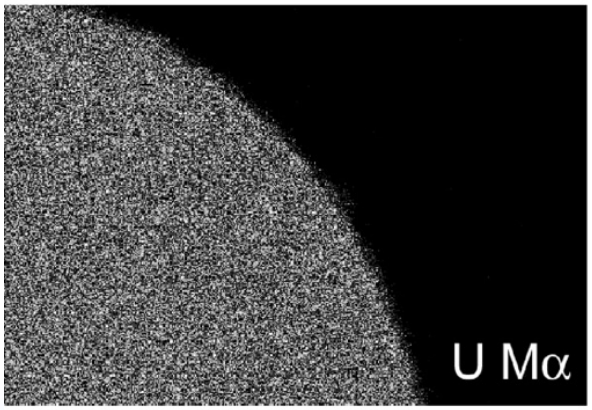
(a)



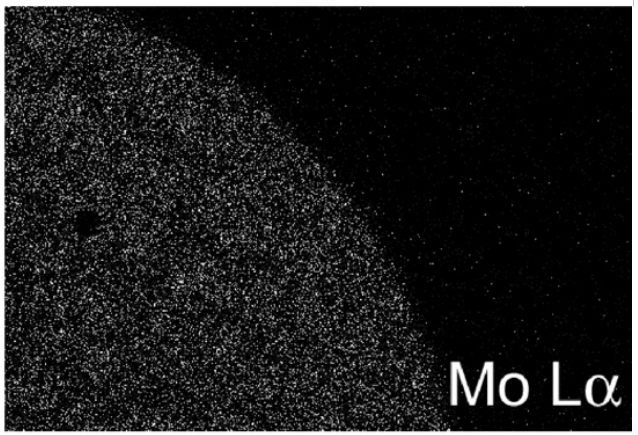
(b)



(e)



(c)



(d)

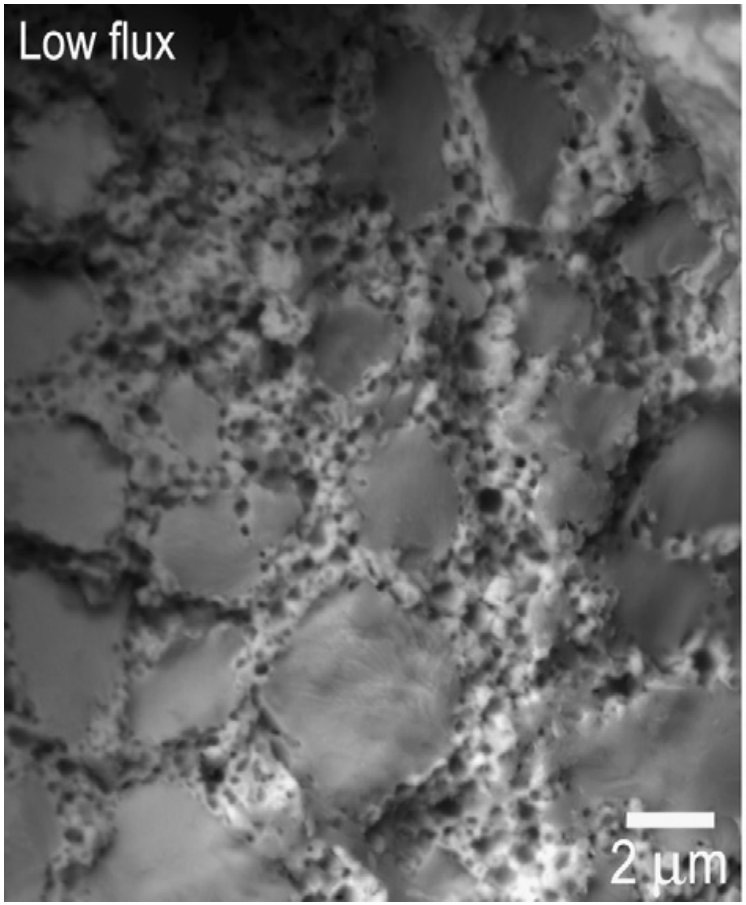
Keiser, et al.
JNM 425
(2012) p156

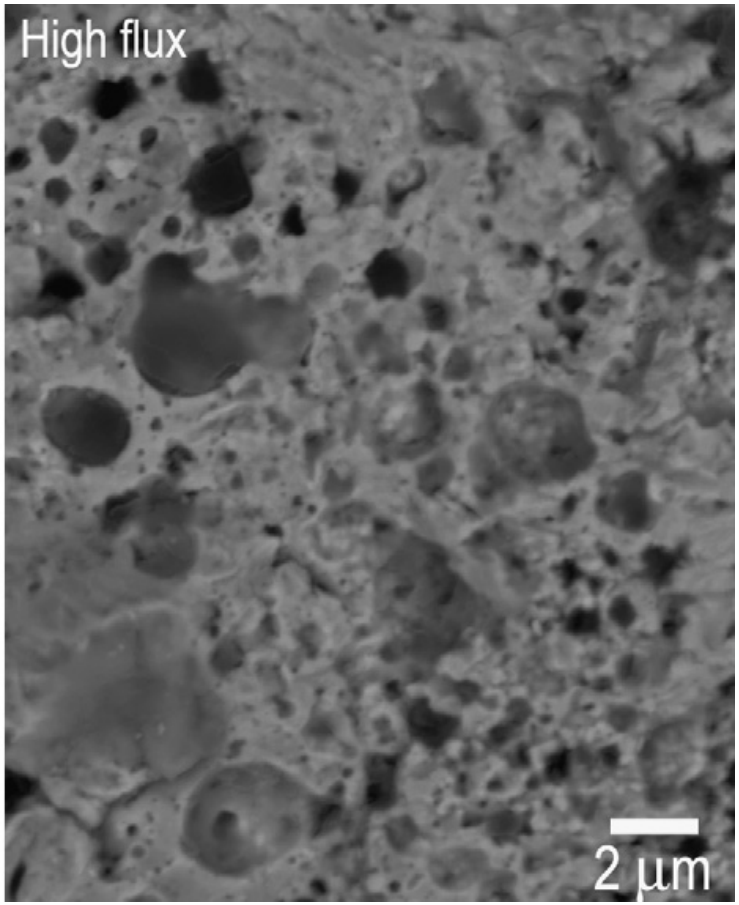
Idaho National Laboratory

58% enrichment, 3.3×10^{21} f/cm^3 (local)

SEM Images of Irradiated U-7Mo/Al-2Si

R2R040 fuel plate, 58% enrichment





Low flux side: 3.3×10^{21} f/cm^3

High flux side: 6.3×10^{21} f/cm^3

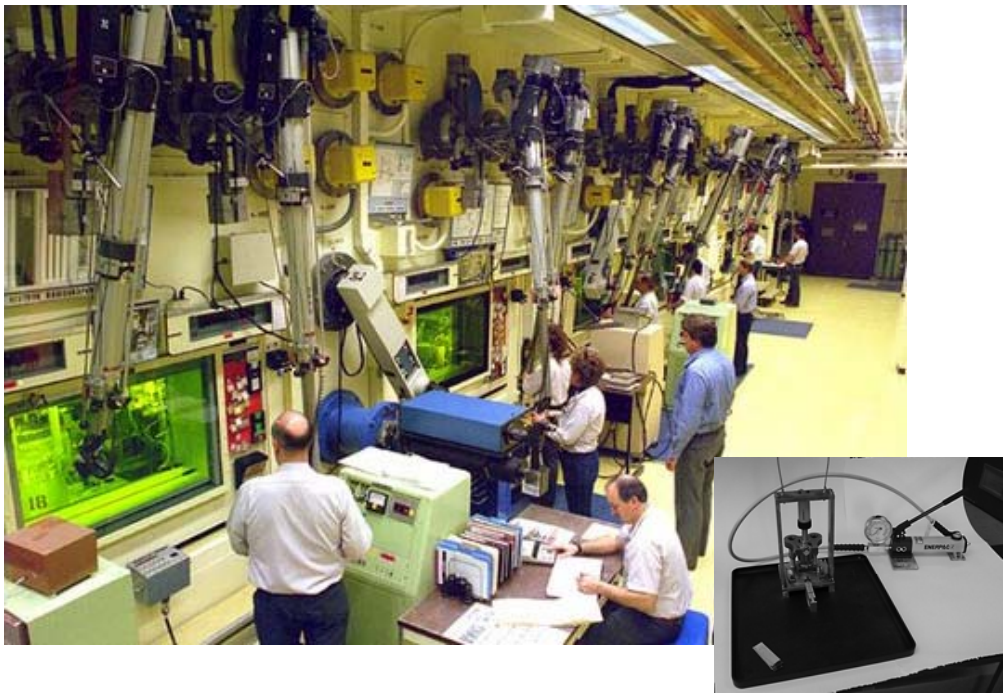
Fuel Irradiation Condition in ATR

Fuel Type Fuel Composition Matrix	Dispersion **U-7Mo** **Al-2Si**	Dispersion **U-7Mo** **Al-2Si**	Dispersion **U-7Mo** **Al-2Si**	Dispersion **U-7Mo** **Al-5Si**	Monolithic **U-10Mo** **n/a**	Dispersion **U_3Si_2** **Al**
Fuel Plate ID	R2R010	R2R040	R2R040	R3R050	L1F140	U0R040
Sample location	High Flux	Low Flux	High Flux	High Flux	Low flux	High Flux
^{235}U enrichment	**19%**	**58%**	**58%**	**58%**	**58%**	**75%**
Local fission density (10^{21} fiss./cm^3)	**4.5**	**3.3**	**6.3**	**5.2**	**3.5**	**5.4**
Time averaged fission rate (10^{14} fiss./cm^3-s)	**3.8**	**4.3**	**8.1**	**6.6**	**4.5**	**6.0**
Plate center-line temperature (°C)	**109**	**90**	**120**	**130**	**112**	**120**

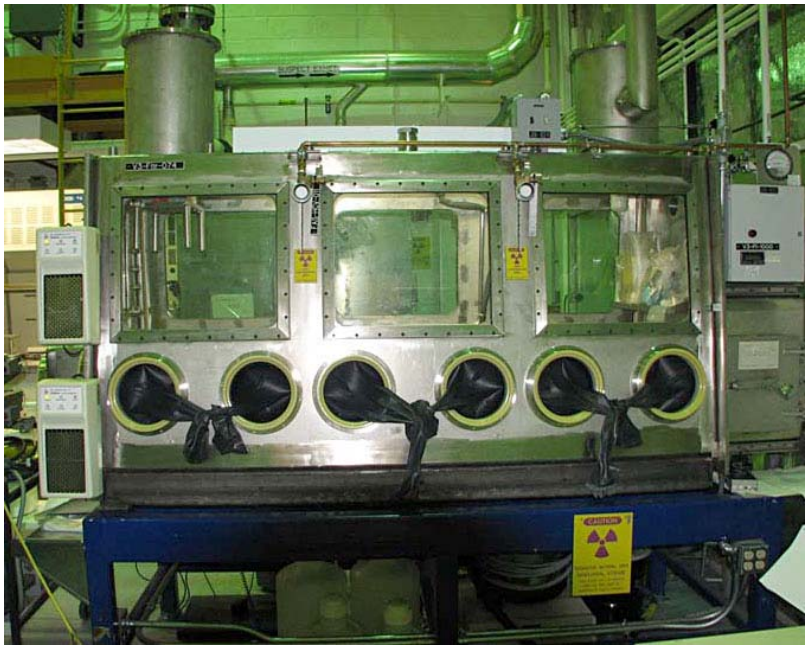
Fuel meat U loading density:
Monolithic > Dispersion
Dispersion fuel U loading:
U-7Mo > U-10Mo > U_3Si_2

Idaho National Laboratory

TEM Sample Prep. of the Irradiated Dispersion Fuel



Punch used in a hot cell for 1.0 mm punching for dispersion fuel plate

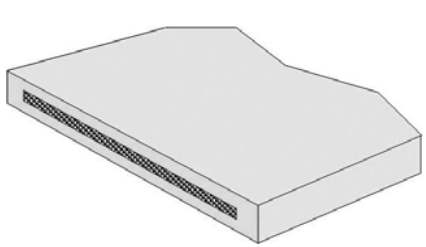


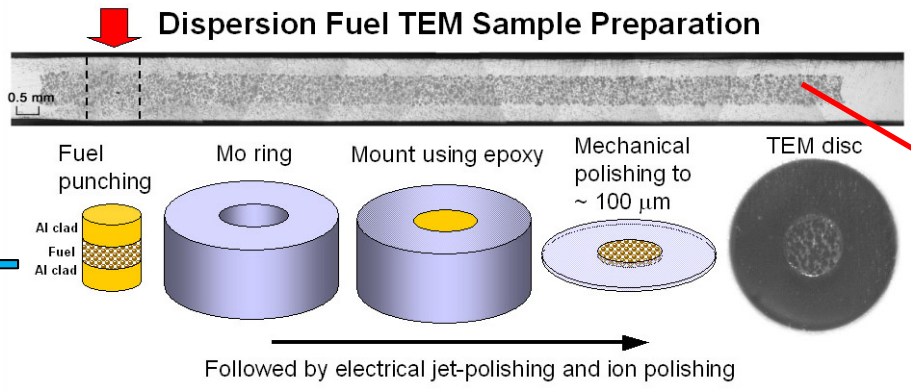
Glove-box used at EML for SEM and TEM sample preparation

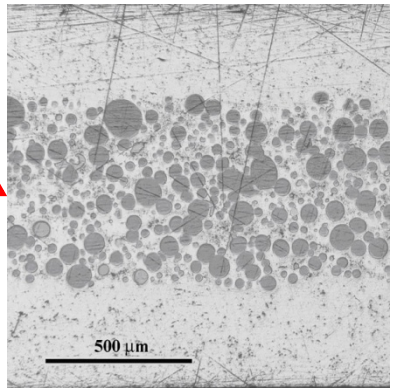
HFEF at INL, one of the largest hot cells in the world.

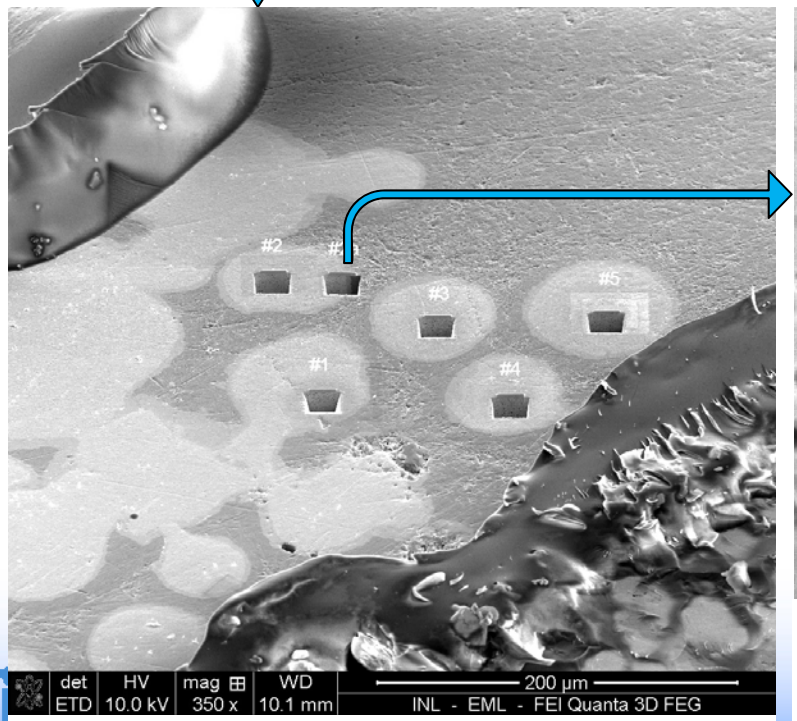
Idaho National Laboratory

Site-Specific TEM of Irrad. Disp. Fuel Using FIB

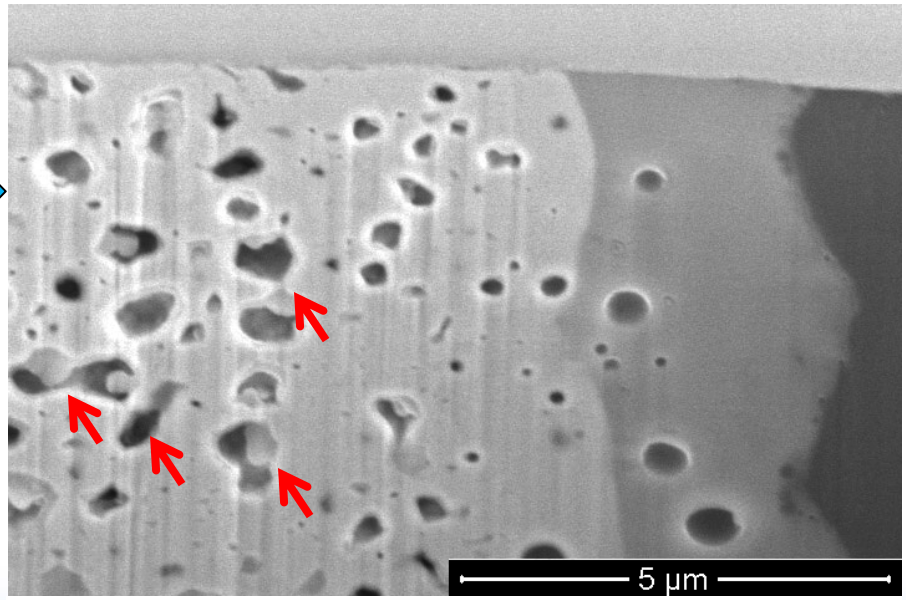






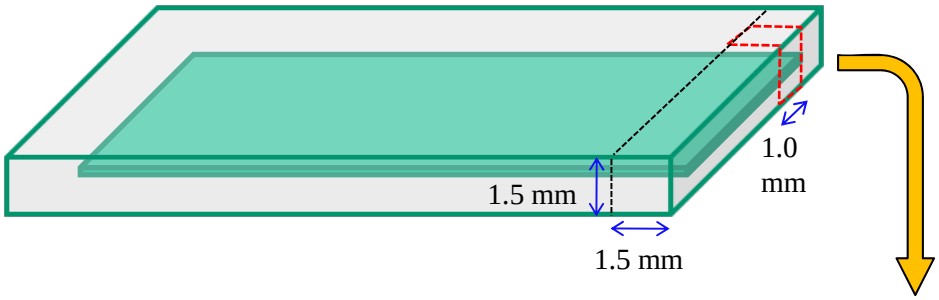


Site specific FIB lift-out from a SEM sample prepared from a 1.0 mm punch



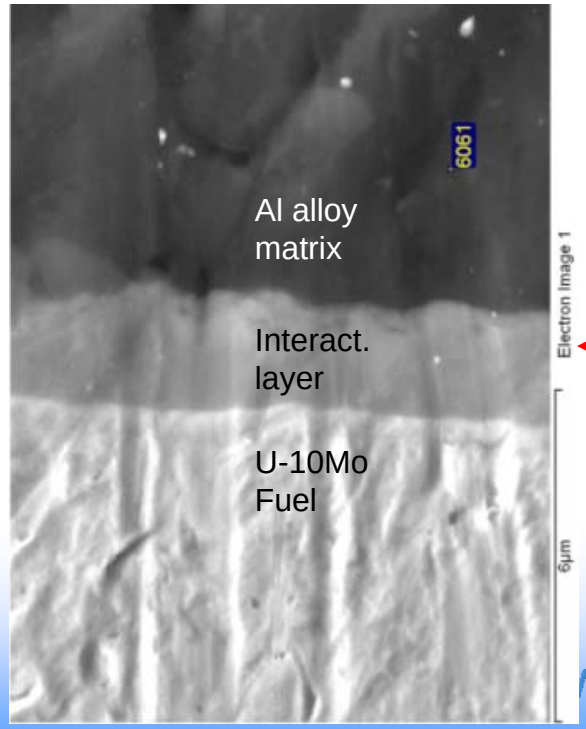
- SEM image of a FIB lift-out cut across the FCI showing circular bubbles in amorphous FCI.
- Solid fission product precipitate at gas bubble.
- More bubbles in the fuel than in FCI.
- Evidence of bubble inter-link is identified.

SEM Sample Prep. of Irrad. Monolithic Fuel

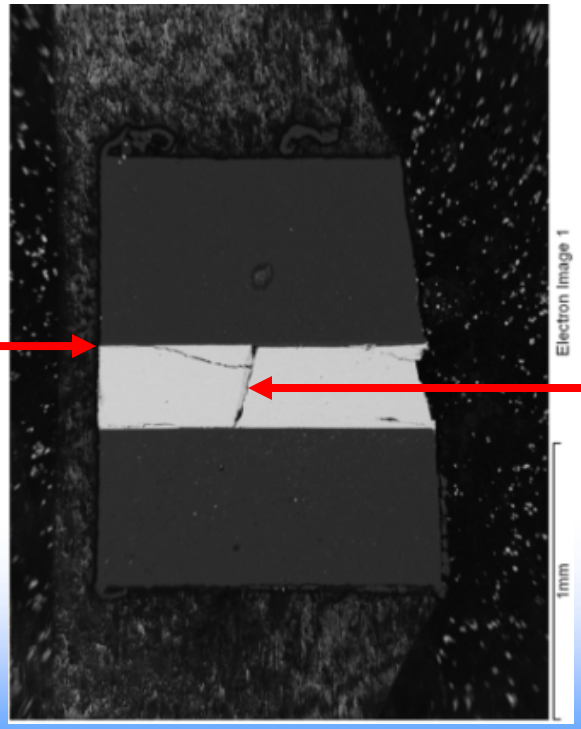


- A small sample with its total activity similar to that of a 1.0 mm diameter punch sample was sectioned in a hot cell.
- A SEM sample was prepared using a glove box and characterized.

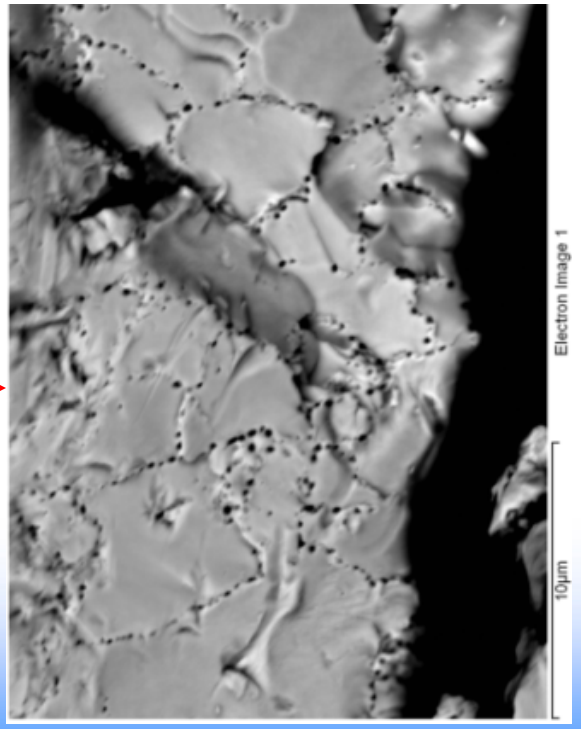
Fuel-Matrix interaction



SEM sample



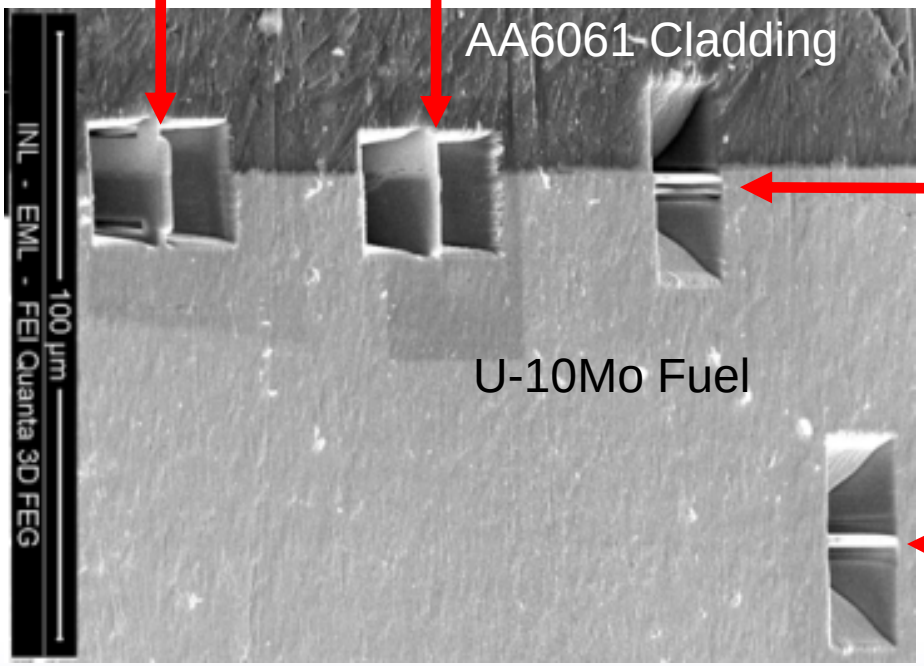
Fracture surface



Site-Specific TEM of Irrad. Monolithic Fuel

U-10Mo/AA6061 Monolithic Fuel

Fuel-Cladding-Interaction
Cross section FIB TEM

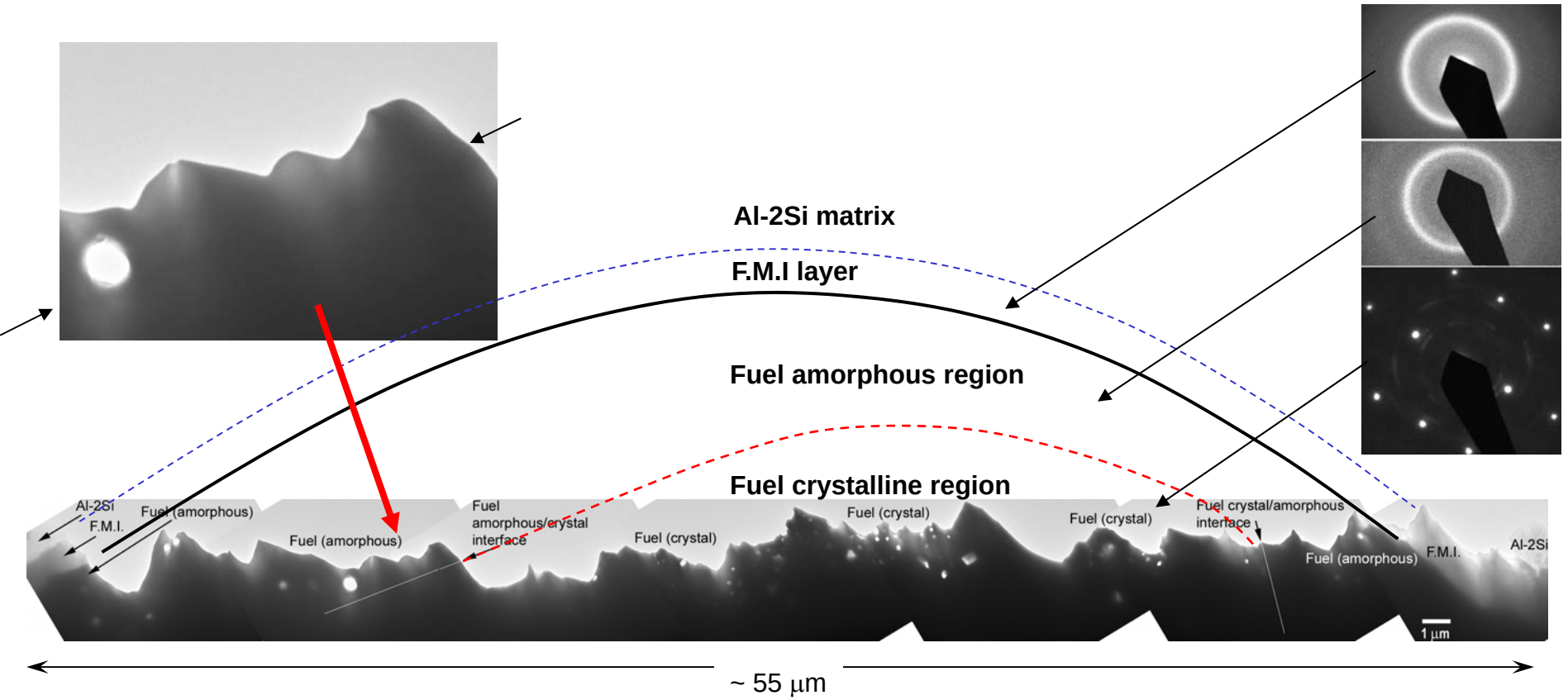


Fuel next to F.C.I.

Fuel near the plate center

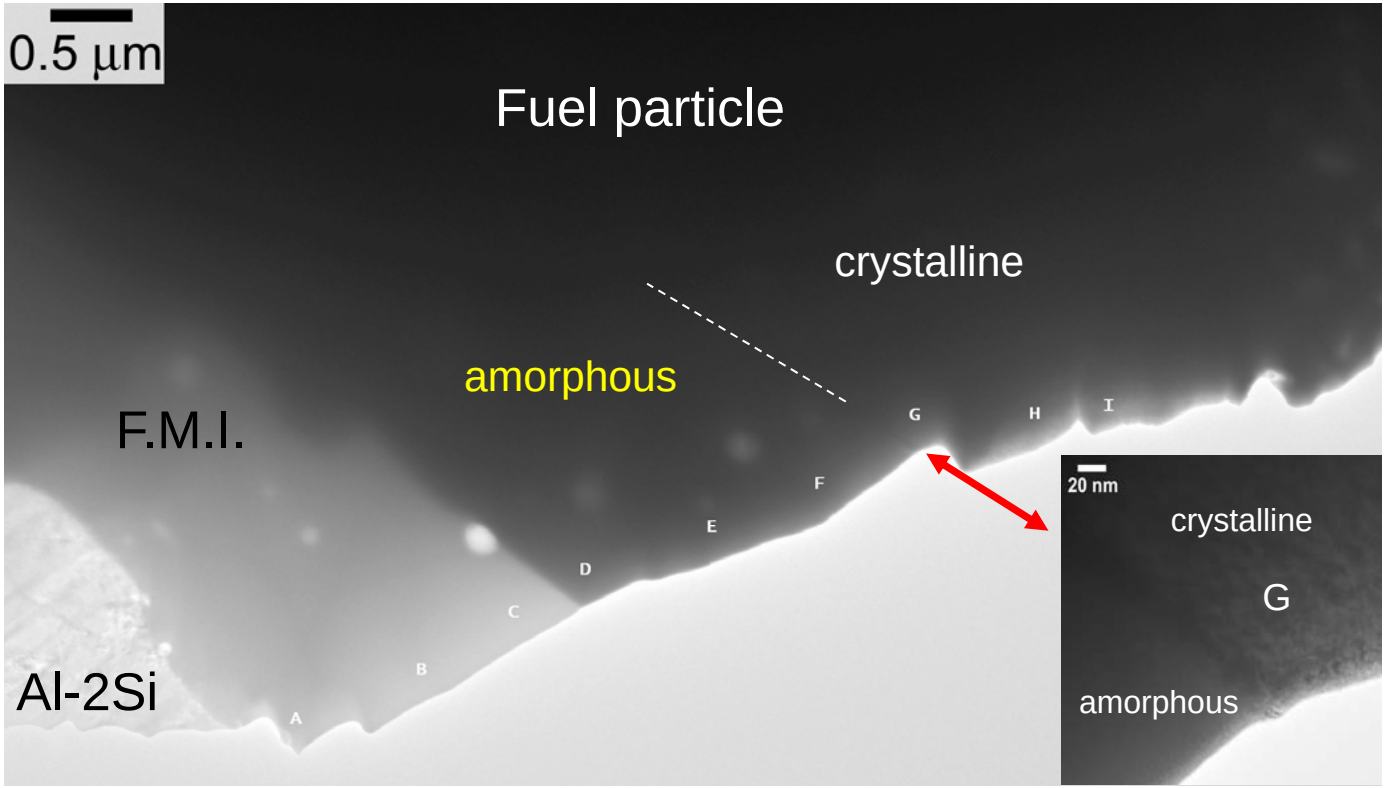
TEM Characterization of Irradiated U-Mo Dispersion Fuel Plates

Overview of RERTR-7 Low Flux Fuel Particle



- Amorphous region (roughly 2 – 6 μm) of the fuel particle (Si > 10 at%).
- Crystalline region of the fuel particle (Si < 2 at% or not detected).
- An abrupt change in Si content cross the interface of amorphous/crystalline.
- Large bubbles found in fuel particle, not the F.M.I. layer
- Small bubbles found in F.M.I. layer
- Bubble superlattice only found in crystalline region of the fuel particle.

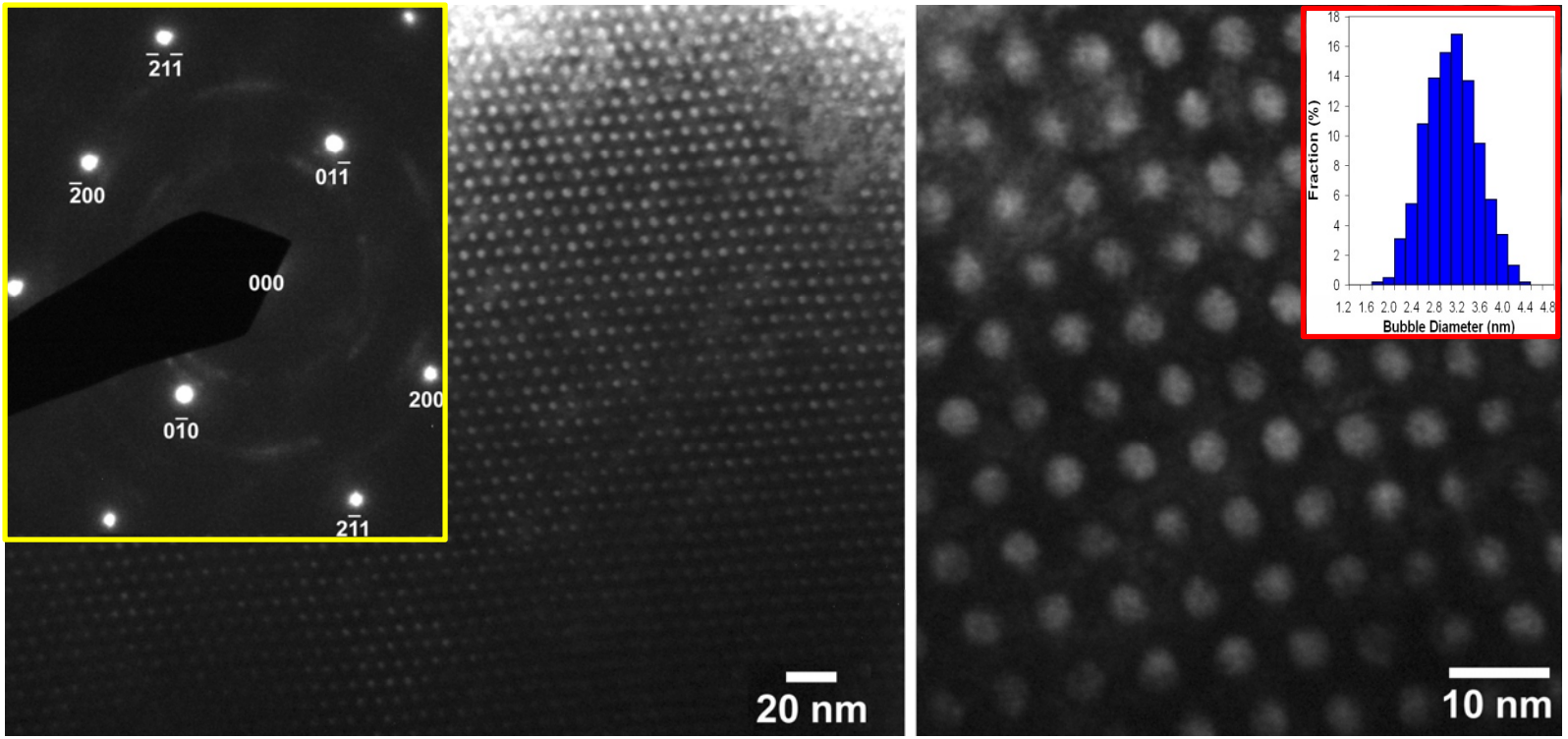
Fuel – Matrix Interfaces in RERTR-7 Low Flux



Spot	Al	Si	Mo	U	EDS Measurement Note
A	78.0	5.1	4.4	12.6	F.M.I. near Al-2Si side
B	78.7	3.5	4.9	13.0	F.M.I.
C	77.6	1.9	5.2	15.3	F.M.I. near fuel side
D	16.0	8.7	17.7	57.5	Fuel, amorphous
E	11.8	10.2	16.8	61.3	Fuel, amorphous
F	11.3	15.3	16.3	57.2	Fuel, amorphous
G	9.2	2.4*	21.0	67.5	Fuel, crystalline

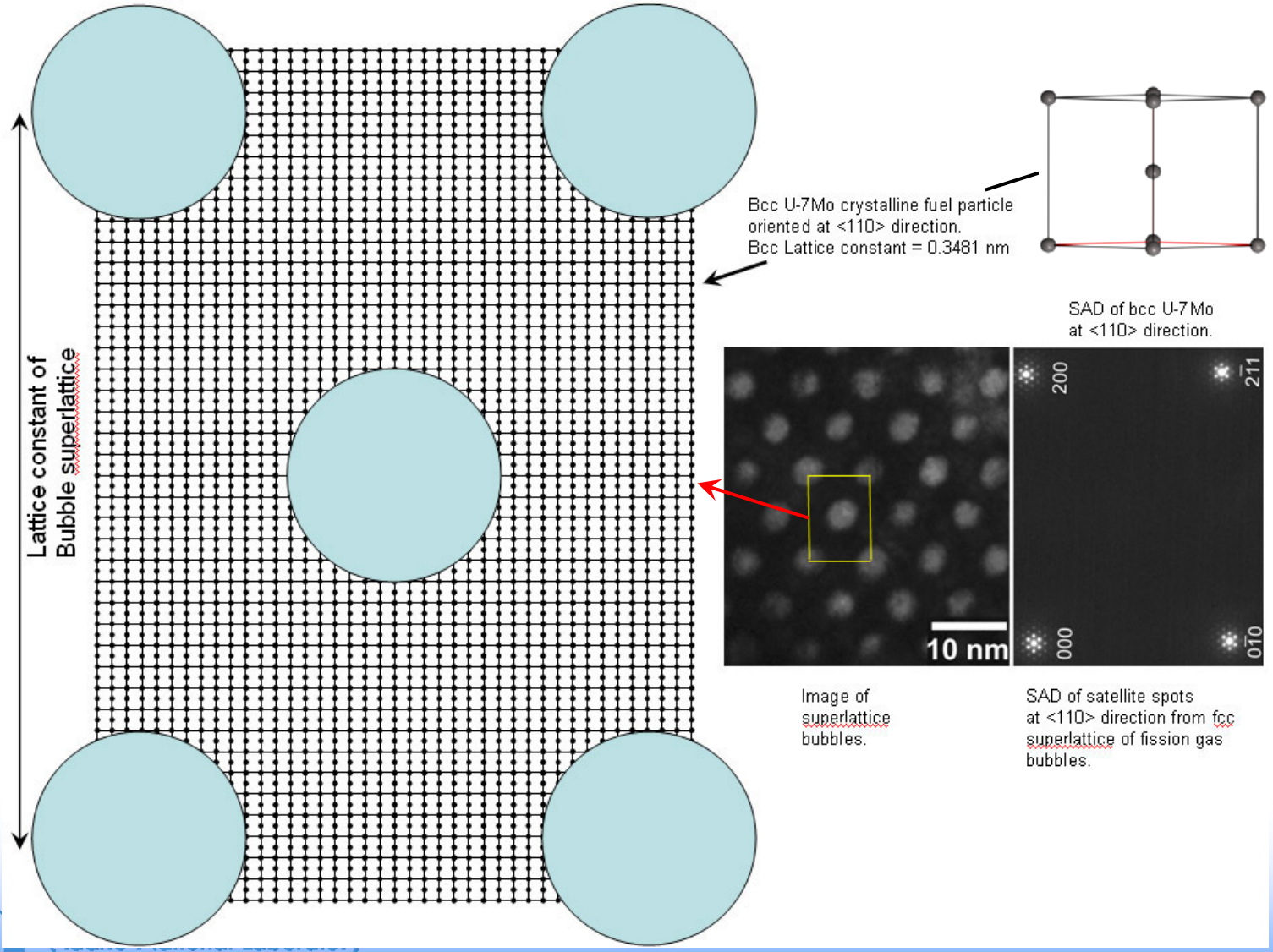
All in at.%

Fission Gas Bubble Superlattice in Irradiated U-7Mo Fuel Particles at 3.3×10^{21} fission/cm^3



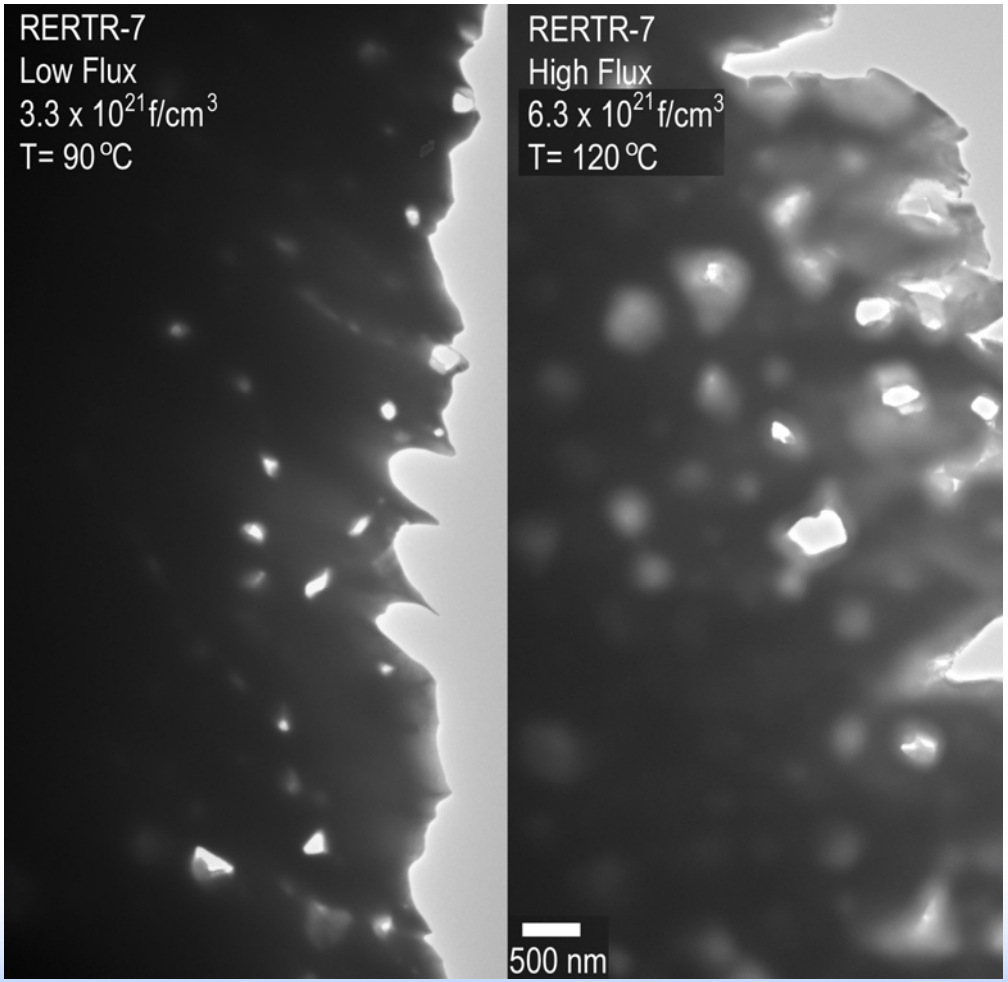
- Ordered bubbles in U-7Mo fuel (bcc) imaged at zone [011] at low and high mag.
- Coherent orientation between U-7Mo fuel (bcc) and bubble superlattice (fcc).
- The measured average bubble size = 3.1 ± 0.4 nm (1066 counts).
- The measured fcc superlattice constant = 12.07 ± 0.06 nm.

Orientation of Bubble Superlattice vs. U-7Mo Lattice



Comparison of U-7Mo Particle Microstructure between Low Flux and High Flux Conditions

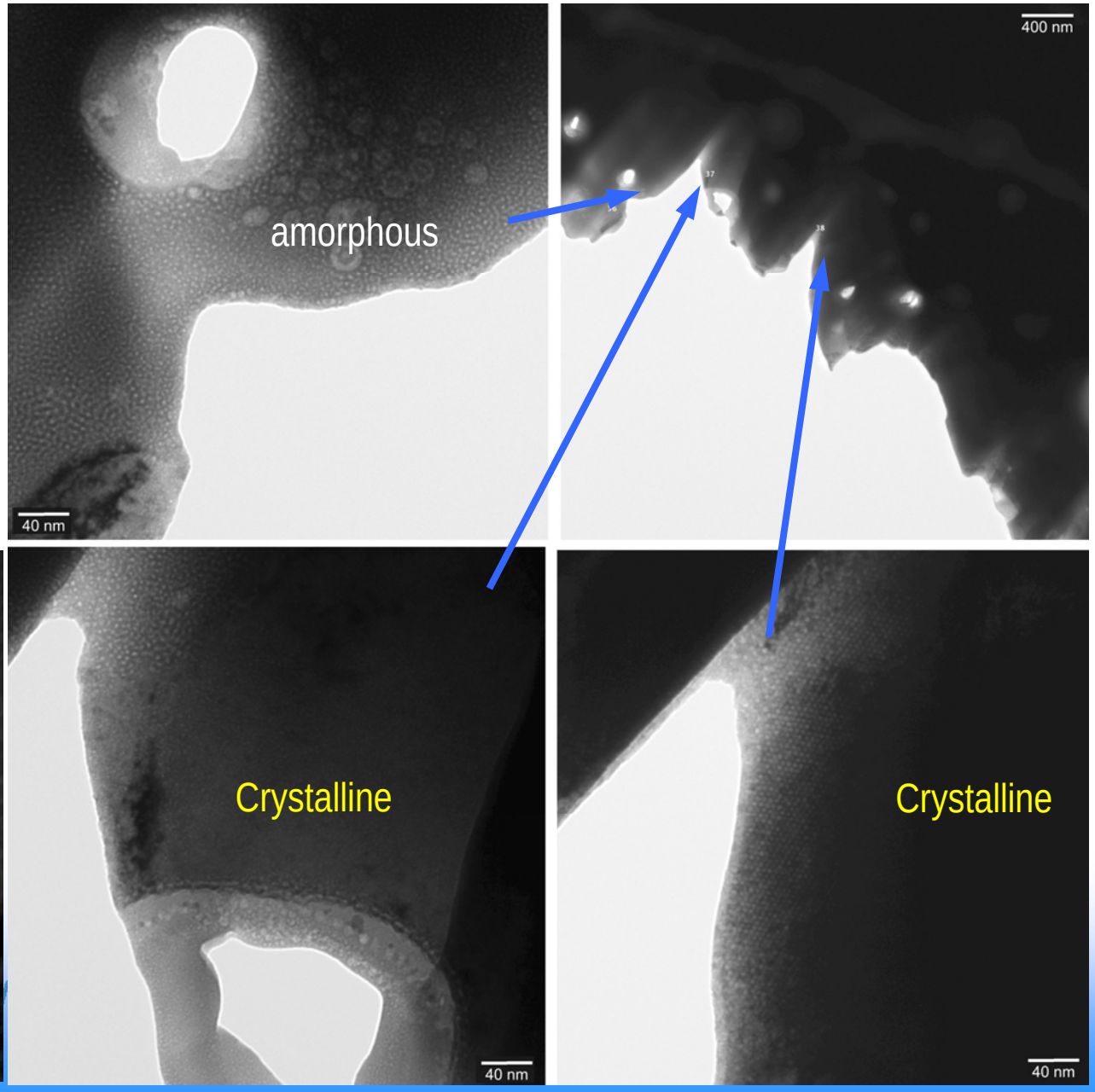
Low Flux



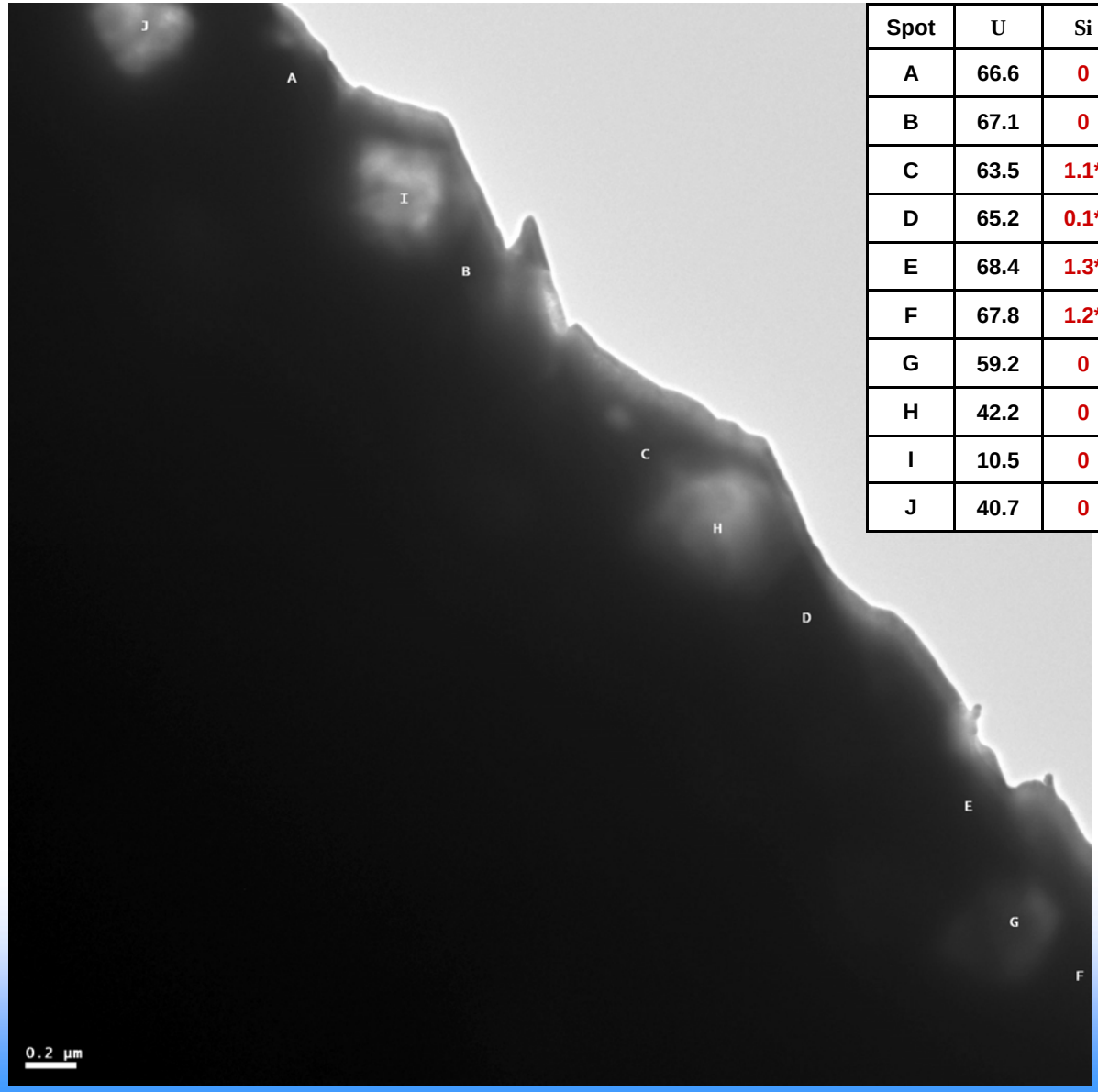
High Flux

Microstructure in U-7Mo at High Fission Density

- Low mag. and high mag. images of fuel particle showing residual ordering of fission gas bubble in the amorphous region.
- Bubble suprelattice still evident in certain part of crystalline region.



Fission Product Precipitates in the Fuel



Spot	U	Si	Al	Mo	Other
A	66.6	0	17.1	17.0	
B	67.1	0	14.4	18.6	
C	63.5	1.1*	18.6	16.6	
D	65.2	0.1*	18.1	16.5	
E	68.4	1.3*	15.2	15.1	
F	67.8	1.2*	16.6	14.4	
G	59.2	0	14.0	12.8	Sr-5.5, Ba-5.1, Nd-3.0, Y-0.4*
H	42.2	0	23.5	18.2	Sr-2.4, Ba-3.7, Nd-9.0, Y-1.3*
I	10.5	0	24.4	6.4	Sr-24.0, Ba-19.2, Nd-3.5, Y-7.3, Te-4.7
J	40.7	0	24.4	10.3	Sr-3.5, Ba-10.3, Nd-9.0, Y-1.9*

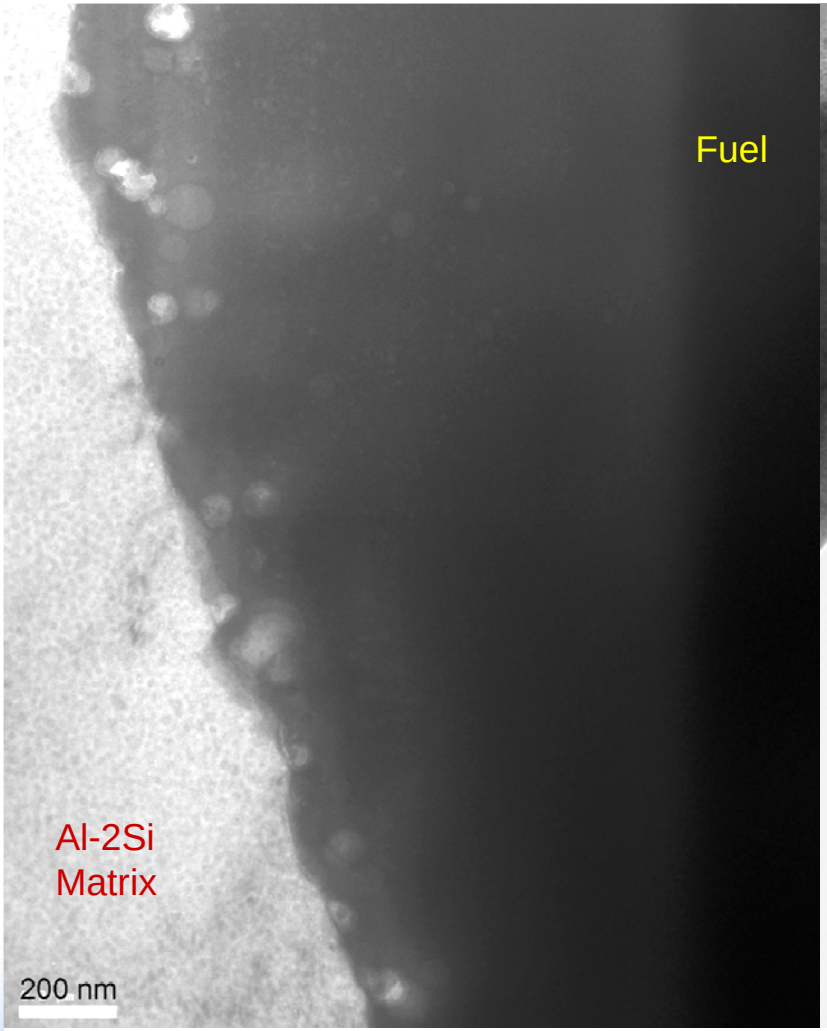
- Note that the Si content in this part of fuel is ~ 0.
- It may suggests that this area is near the center of a fuel particle where Si content is expected to be low to none.

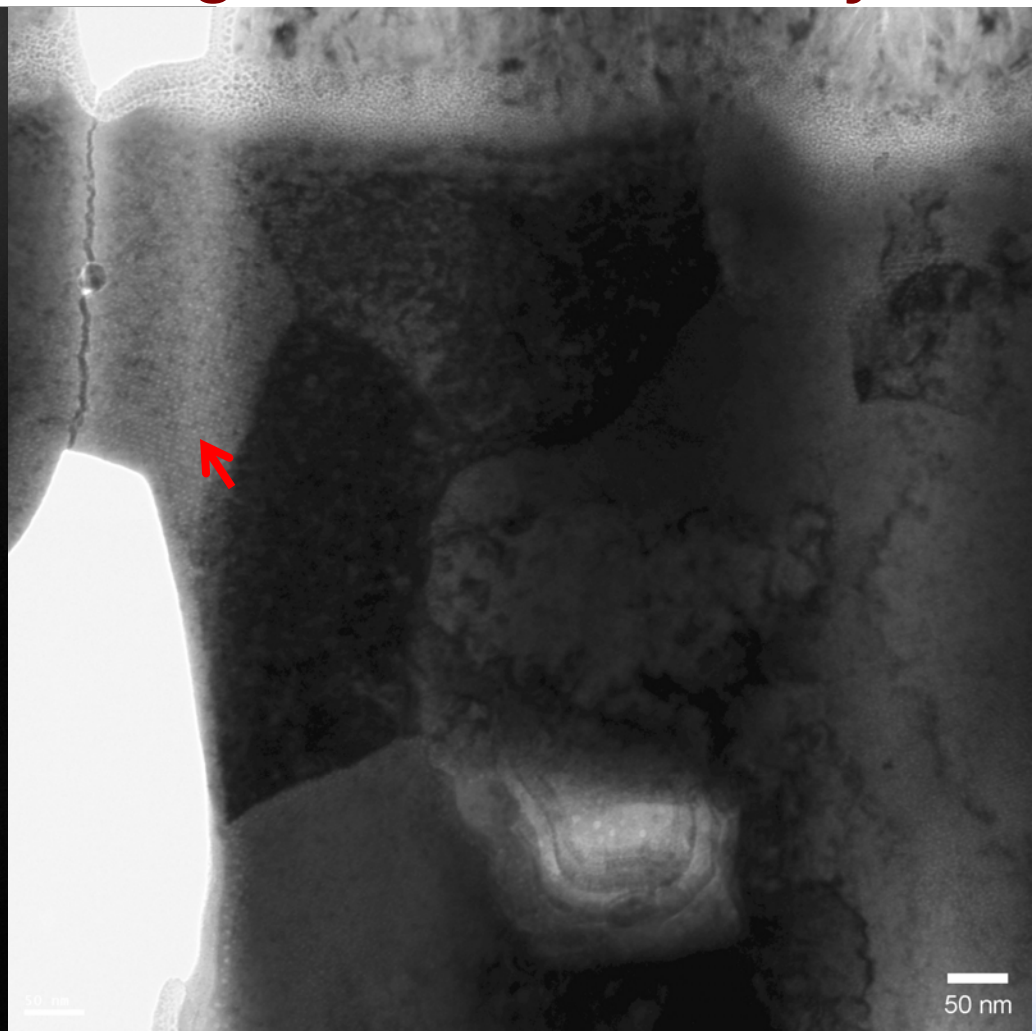
Site-Specific TEM of Irrad. Disp. Fuel Using FIB

Spot	U	Si	Al	Mo	Other (at.%)
A	0.0	0.1	99.5	0.3	Al
B	12.1	13.3	71.1	3.4	Fuel-Matrix Interaction (FMI)
C	13.7	3.5	78.8	4.0	FMI
D	16.6	1.1	77.2	1.1	FMI
E	61.1	5.3	18.2	15.4	High Si layer (amorphous)
F	67.0	10.5	5.3	17.3	High Si layer (amorphous)
G	81.9	1.5	0.0	16.8	Fuel
H	79.1	1.0	1.1	18.8	Fuel
I	75.0	2.2	3.9	18.9	Fuel
J	80.5	1.1	0.7	17.7	Fuel
K	81.3	0.0	1.4	17.5	Fuel
L1	5.2	0.1	1.3	1.7	Sr_28, Y_23, Ba_17, Zr_9
L2	2.5	0.0	1.0	0.9	Sr_37, Y_17, Ba_30, Zr_6
L3	5.2	0.0	5.9	1.8	Sr_31, Y_14, Ba_29, Zr_7
L4	13.0	0.0	1.3	5.8	Sr_30, Y_16, Ba_19, Zr_7
L5	5.2	0.0	1.2	1.4	Sr_31, Y_18, Ba_23, Zr_10
L6	67.0	0.0	1.8	23.9	Zr_4.9
L7	43.3	0.0	2.5	21.1	Sr_1, Y_3, Nd_21, Zr_3
L8	7.5	0.0	0.0	5.1	Sr_36, Y_22, Ba_14, Zr_9, Ba_14, Nd_2,
M1	13.4	0.0	60.5	2.9	Sr_2, Zr_21 (FMI)
Z1	77.0	0.7	1.5	16.8	Zr_3 (Crystalline)

Note that the area surround the large bubble is typically amorphous, which explains the previously found crystalline-amorphous alternating microstructure.

FIB TEM of Irradiated Dispersion Fuel: Grain Subdivision at High Fission Density





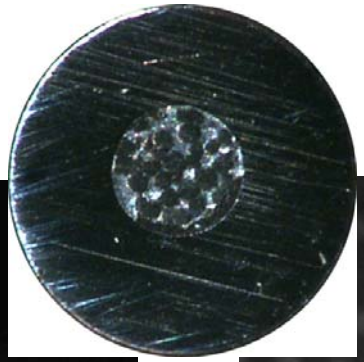
FMI bubbles

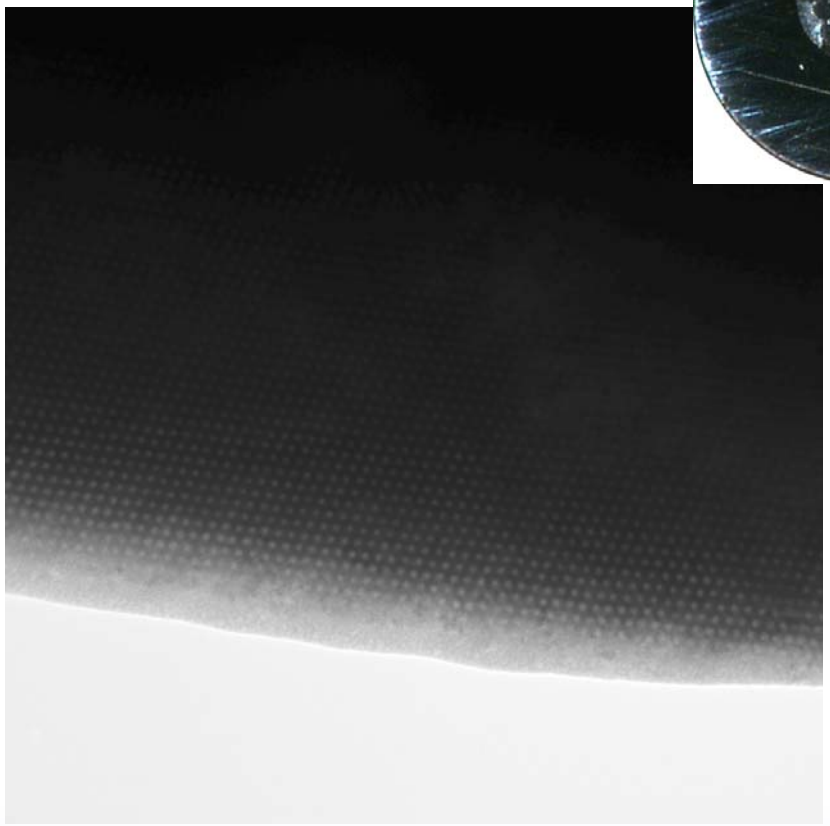
6.3×10^{21} fissions/cm^3

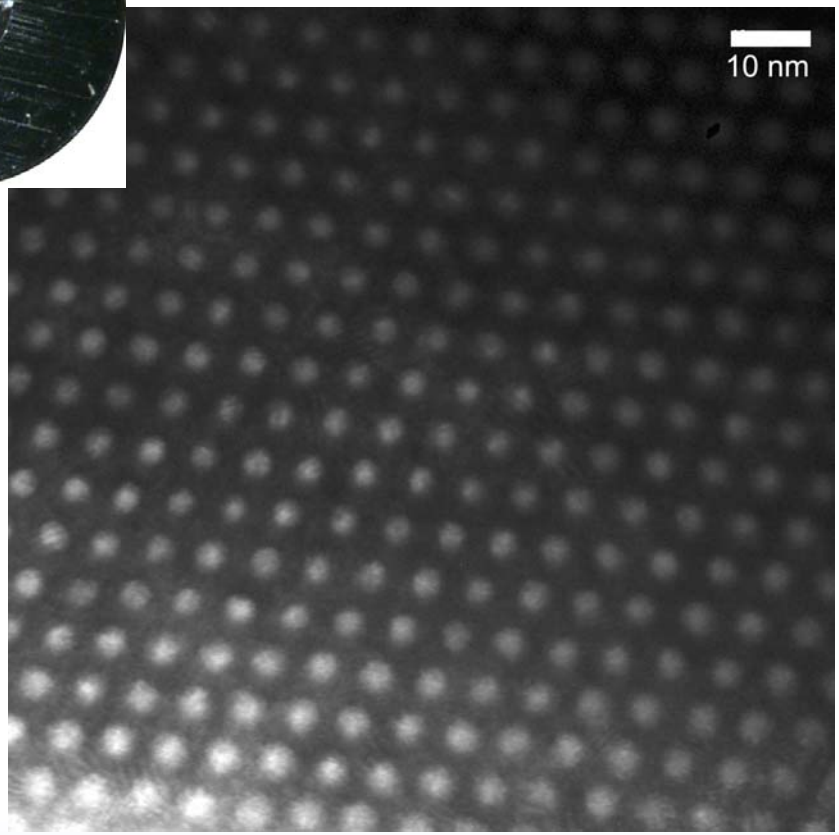
- Small fraction of bubble superlattice and grain subdivision.
- Note that there is no preferential association of bubbles with the boundaries of these small grains (200 ~ 300 nm).

Irradiated U-7Mo/Al-5Si Dispersion Fuel Plate

Fission density:
5.16×10^{21} fissions/cm^3



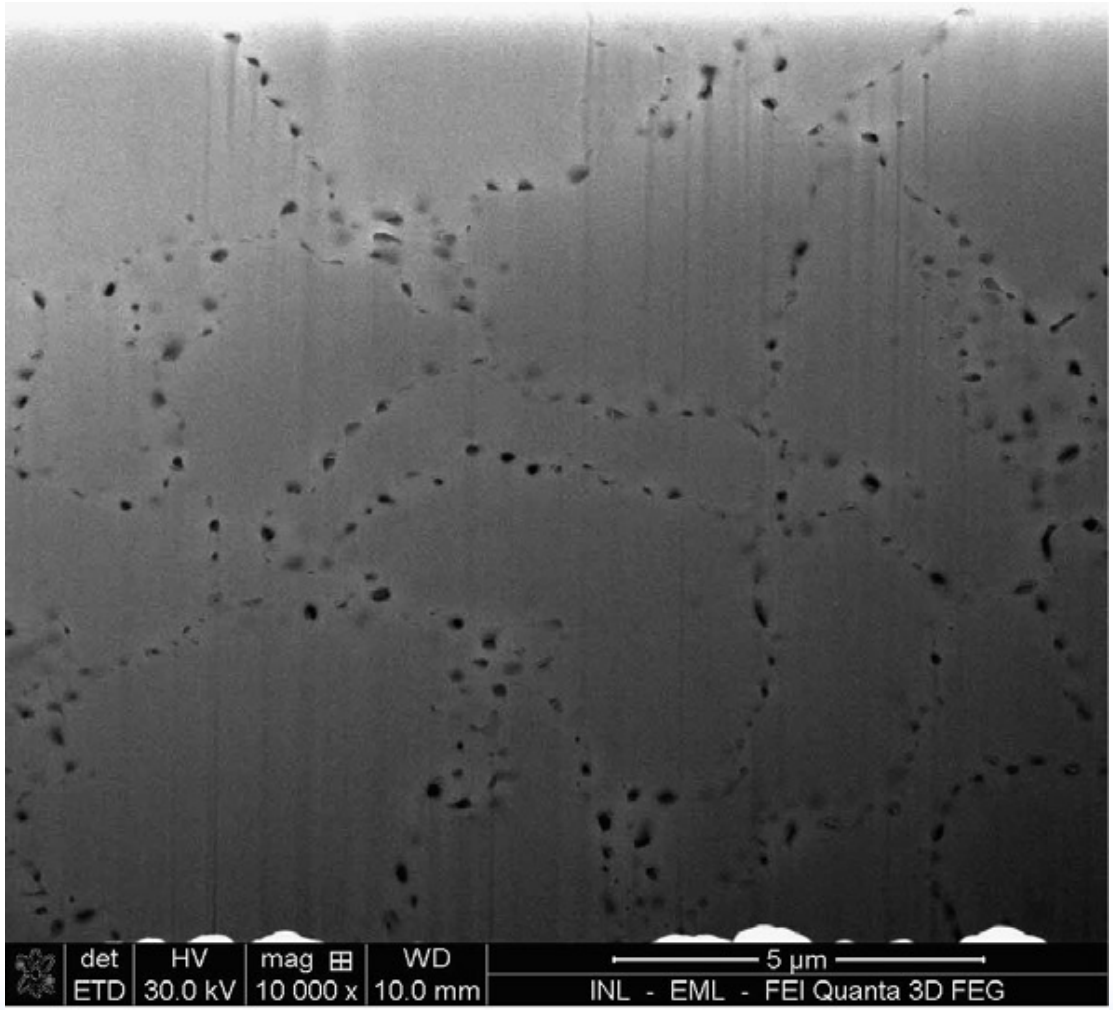




TEM image showing bubble superlattice imaged with U-7Mo oriented at [110]

TEM Characterization of Monolithic U-Mo/AA Fuel Plate

SEM of FIB Lift-out of Irrad. Monolithic Fuel



U-10Mo/AA6061
Monolithic Fuel

3.5×10^{21} f/cm^3 (local)

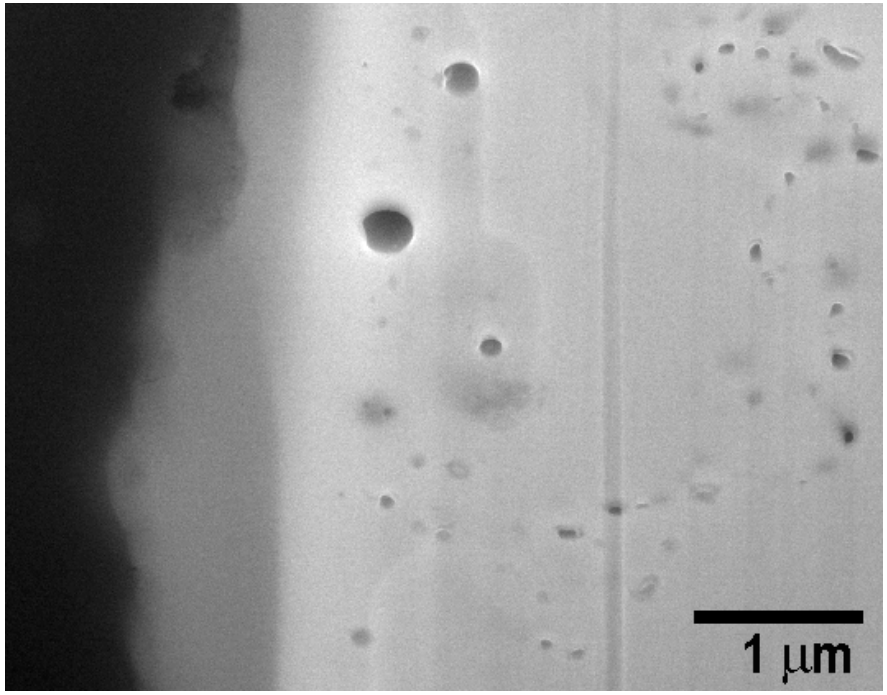
Miller, et al.
JNM 424 (2012)
p38

- Valuable SEM image of FIB Lift-out, before final thinning for TEM analysis.
- Excellent surface finish revealing the submicron size bubbles accumulated at grain boundaries.

Site-Specific TEM of Irrad. Monolithic Fuel Plate

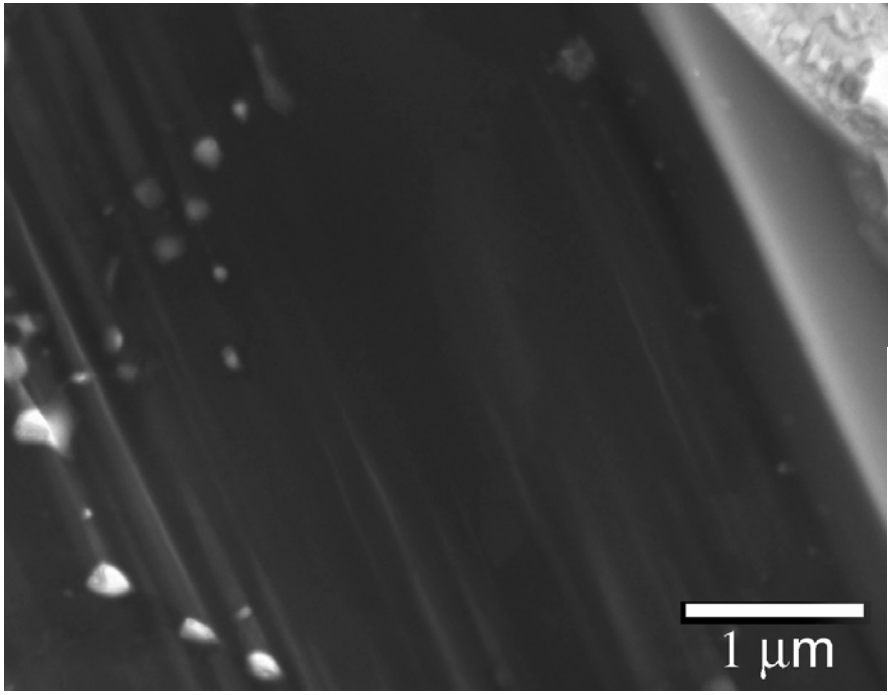
(U-10Mo/AA6061Monolithic Fuel)

SEM image of Fuel-Cladding Interface
From FIB Lift-out Sample



- FIB Lift-out, before final thinning for TEM.
- Its surface quality is excellent for SEM.
- The disadvantage is the very small sample area (10x15 μm^2).
- Few large circular bubbles found in fuel next to FCI.

Low Magnification TEM image
of a FIB Sample



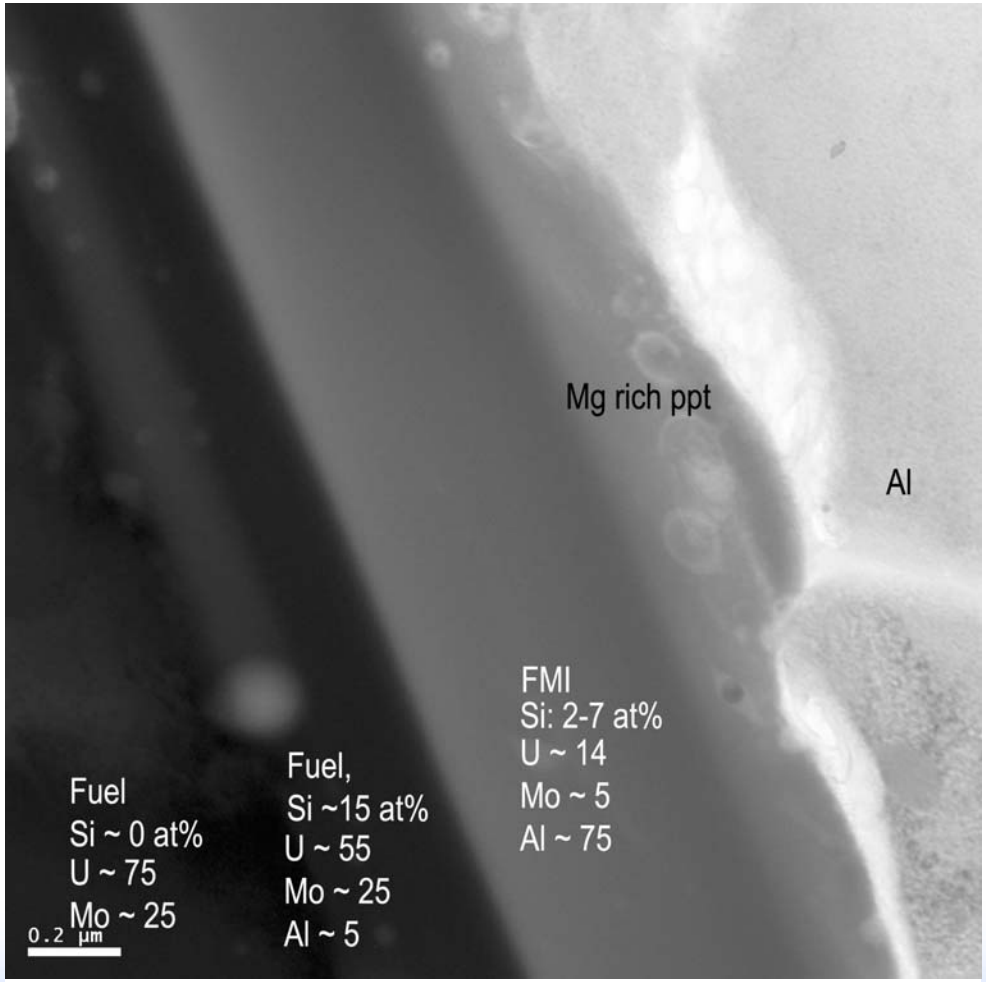
- FIB-TEM sample after final thinning.
- Note the artifact of streaks from FIB ion thinning due to fixed beam orientation and porosity.
- Large bubbles are mostly located at the grain boundary.

TEM Study of Irradiated Monolithic Fuel Plate

U-10Mo/AA6061
Monolithic Fuel Plate

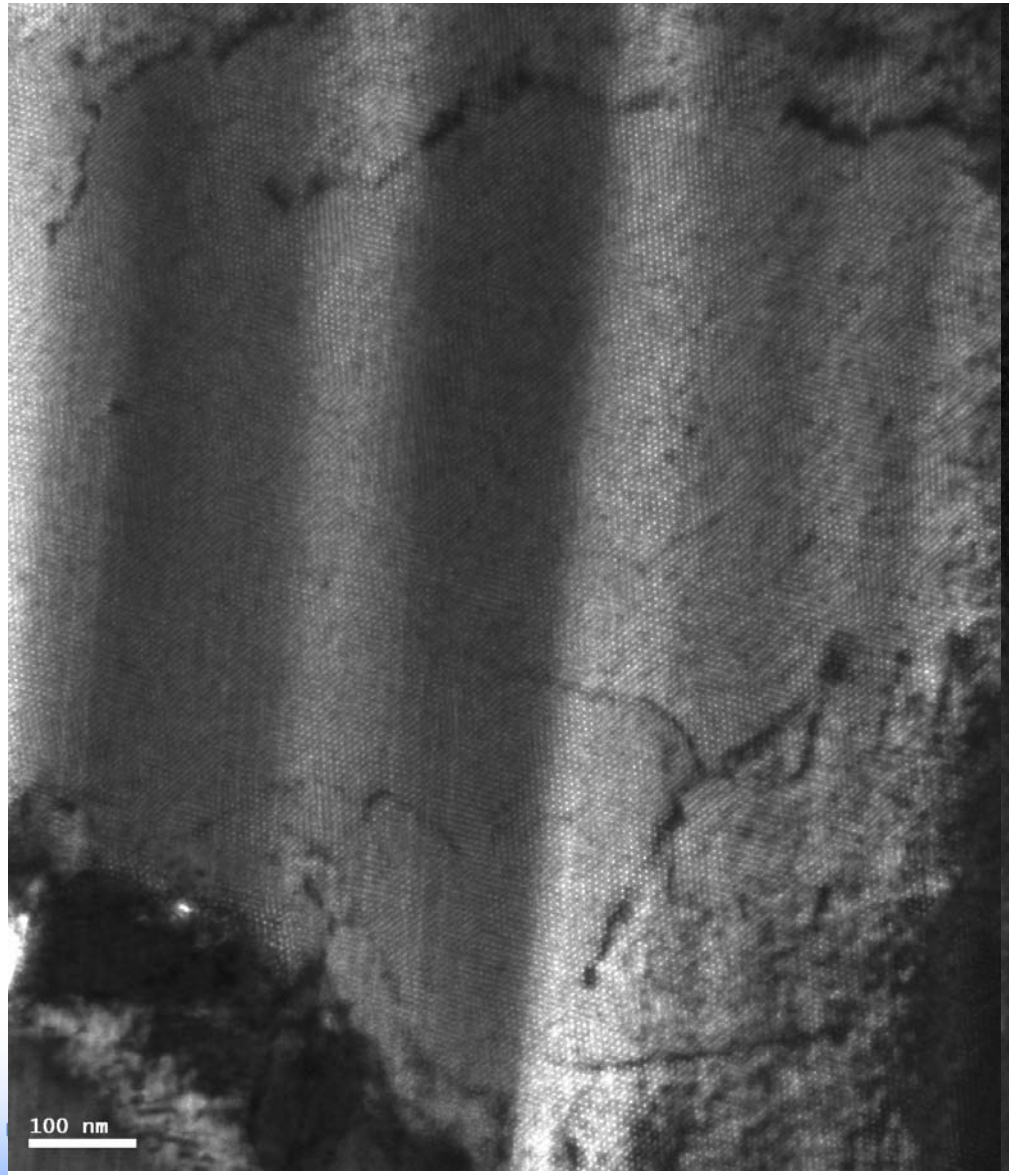
Fission density:
3.46×10^{21} fissions/cm^3

TEM image of Fuel-Cladding Interface and EDS Results



Similar to the dispersion fuel, a Si-rich layer was found in the fuel meat near the interface

Dislocations and Bubble Superlattice

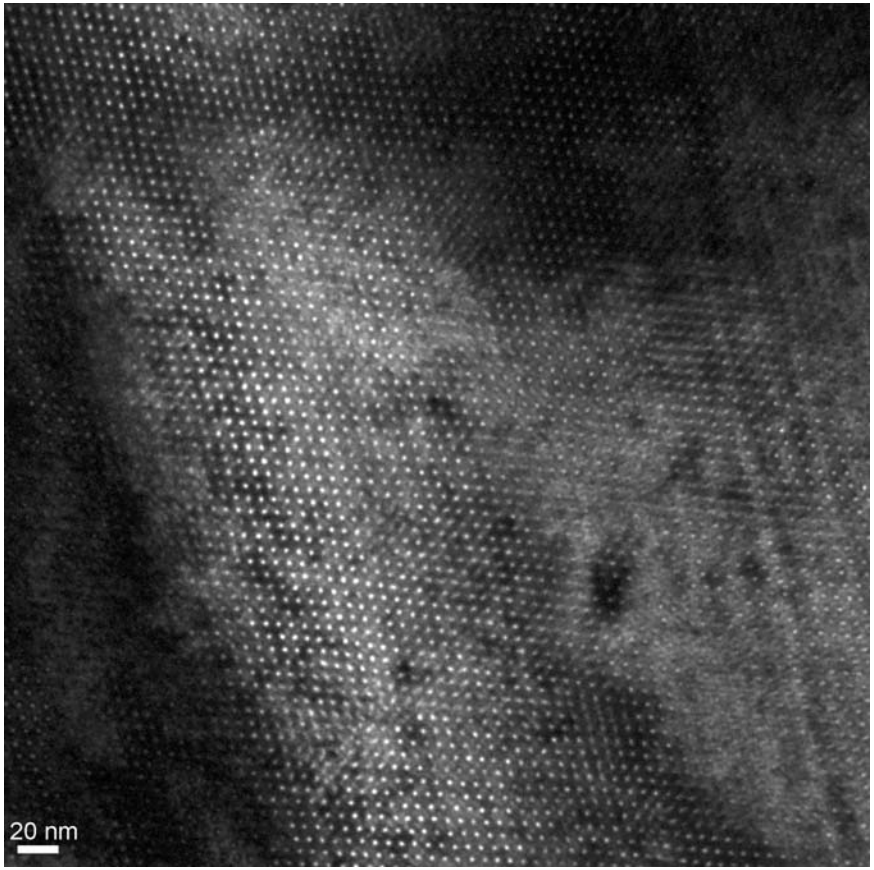


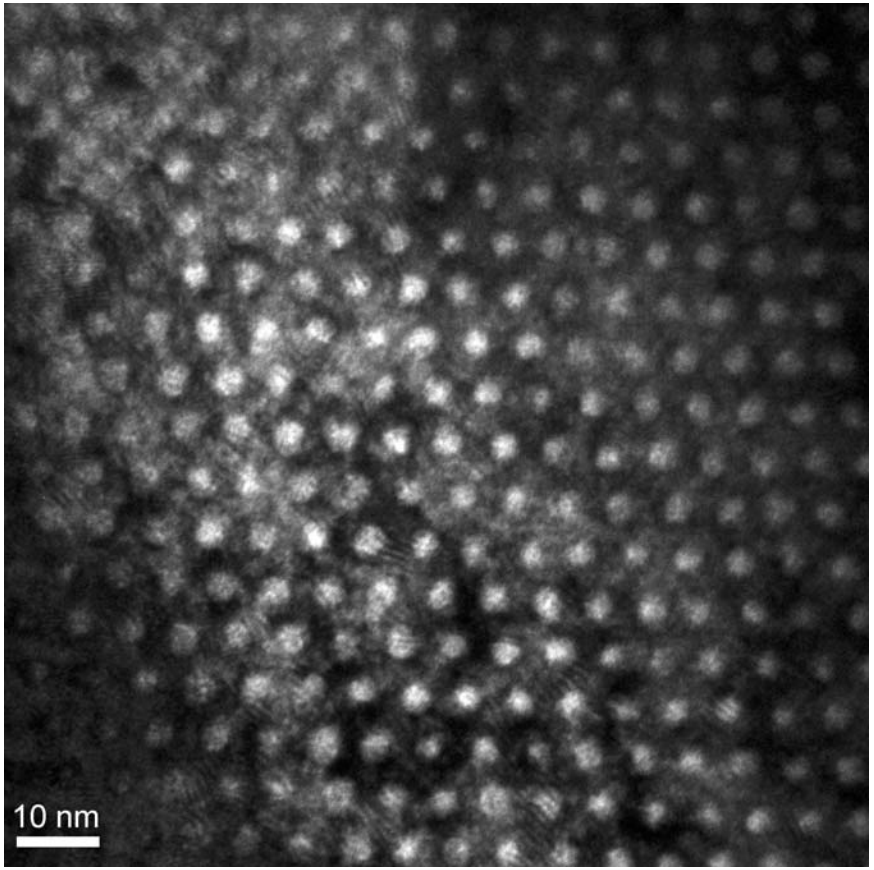
- Bubble superlattice and dislocations near zone [110].
- High internal stress partially due to 90% CW in fresh fuel

Super lattice bubbles [001] near a large grain boundary bubble located at low-right corner

TEM Study of Irradiated Monolithic Fuel Plate

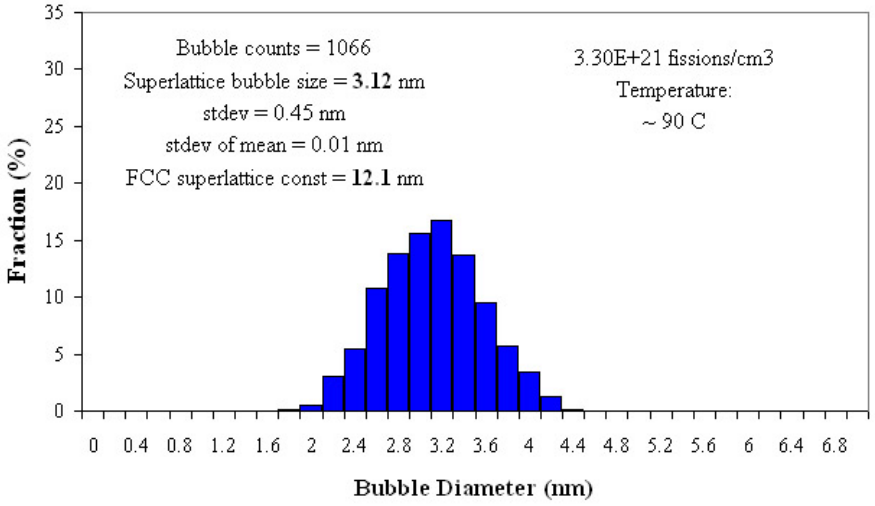
(U-10Mo/AA6061Monolithic Fuel)



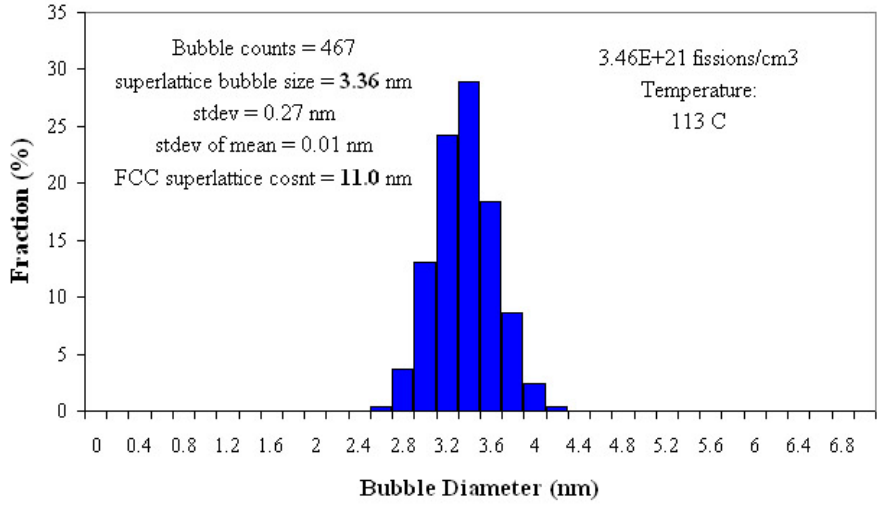


TEM Image of the fission gas bubble superlattice in U-10Mo fuel orientated at [110]

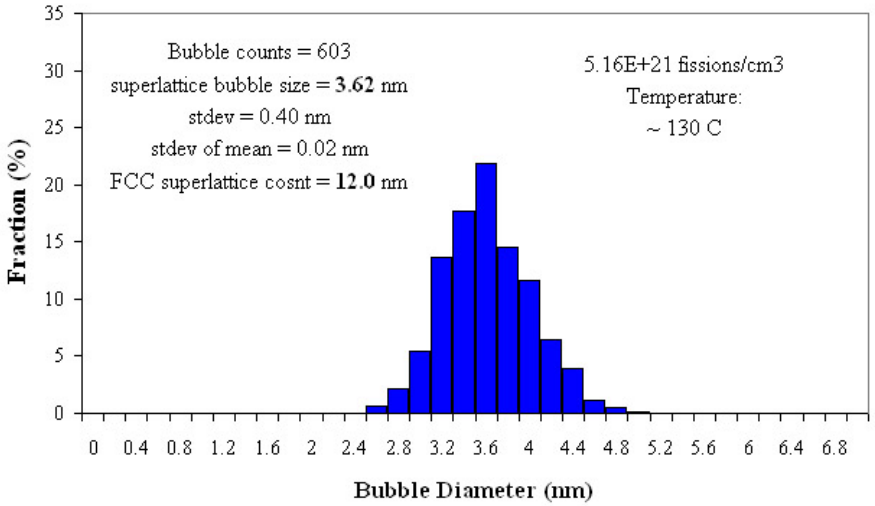
RERTR-7 (U-7Mo/Al-2Si) Irradiated Dispersion Superlattice Bubbles



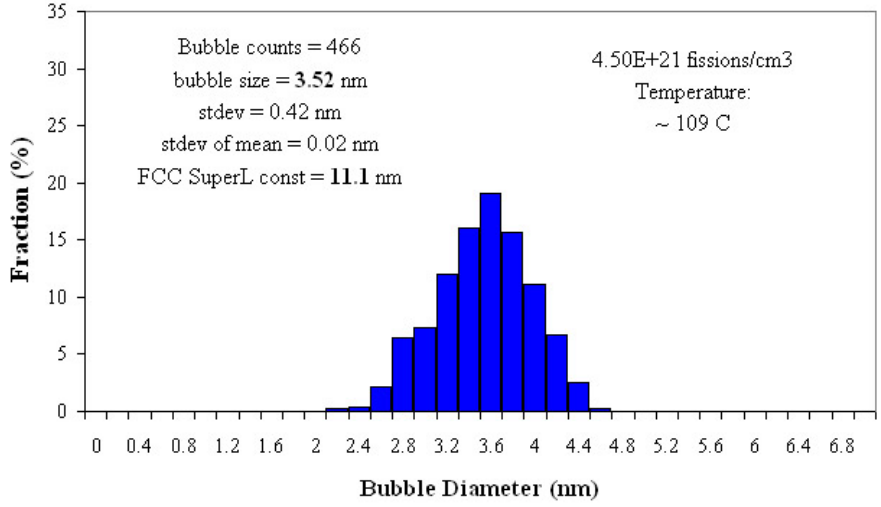
L1F140 (U-10Mo/Al) Irradiated Monolithic Plate Superlattice Bubbles



RERTR-7 (U-7Mo/Al-5Si) Irradiated Dispersion Superlattice Bubbles

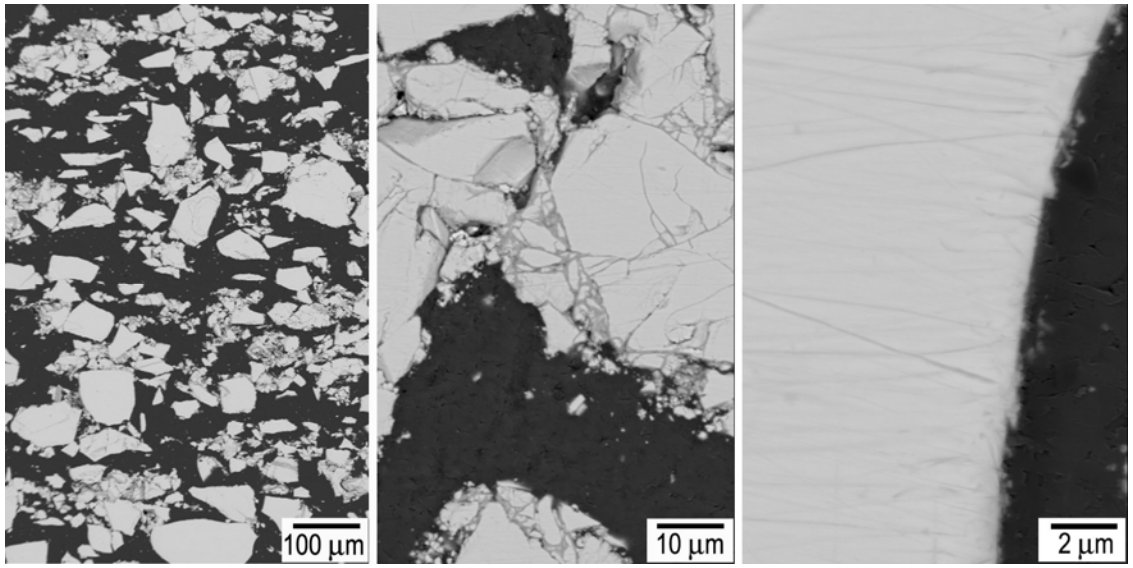


RERTR-6 Irradiated Dispersion Plate Superlattice Bubbles



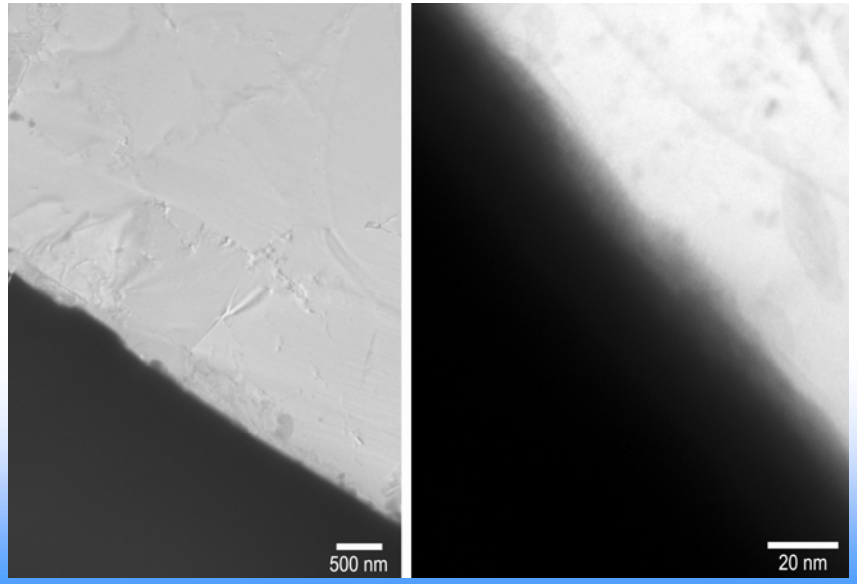
TEM Characterization of U_3Si_2/Al Dispersion Fuel Plate

General Microstructure of U_3Si_2 Fuel

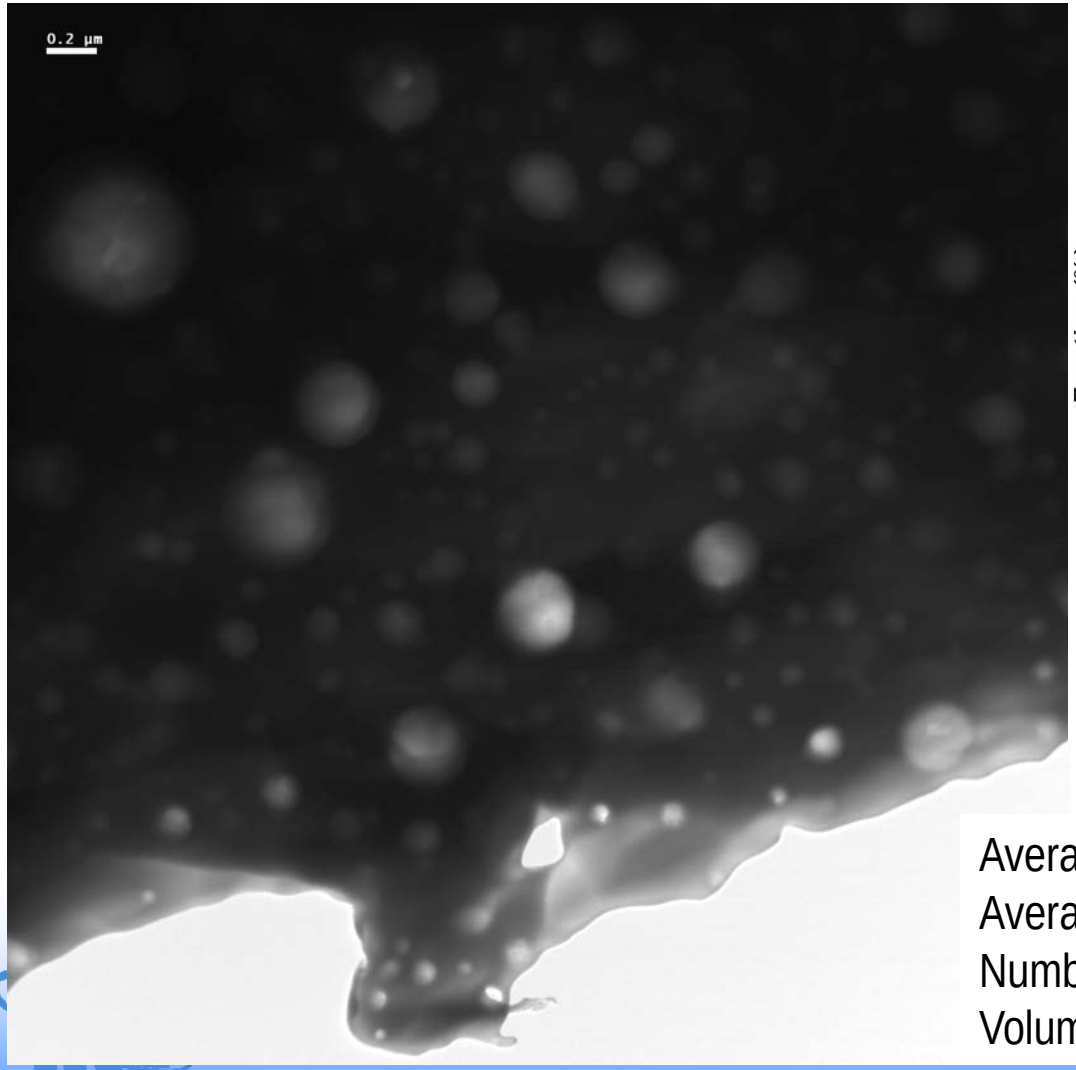


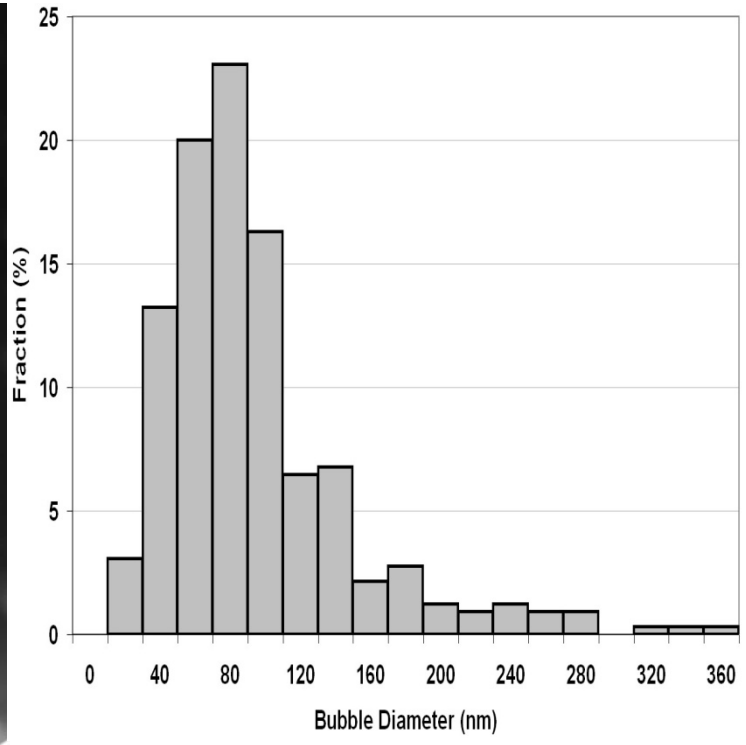
SEM images

TEM images



General Microstructure of U_3Si_2 Fuel Particle

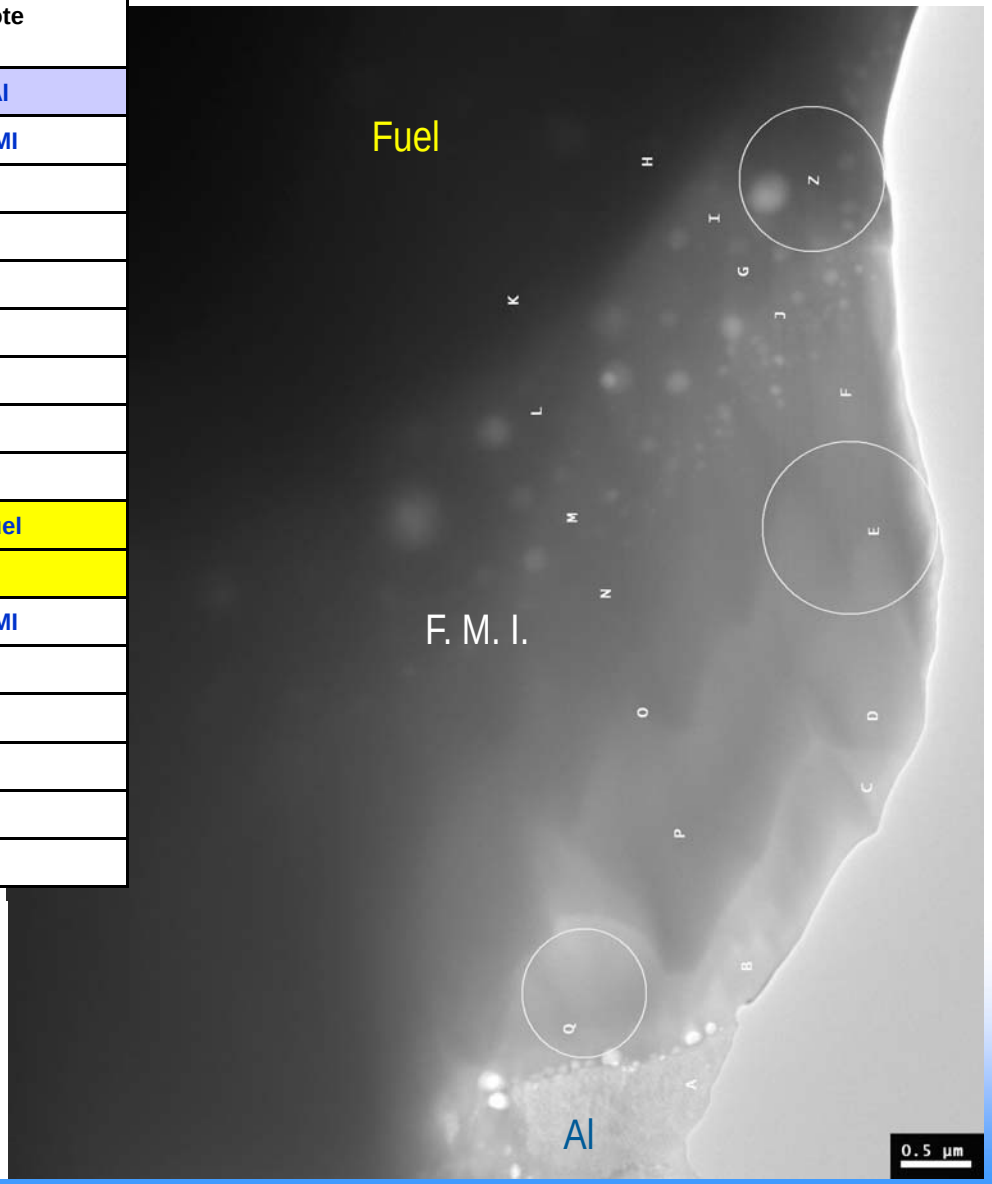




Average bubble size ~ 94 nm. (for size < 1 µm)
Average bubble vol. ~ 1.1×10^{6} nm^3.
Number density ~ 1.05×10^{20} m^{-3}.
Volume fraction ~ 11%

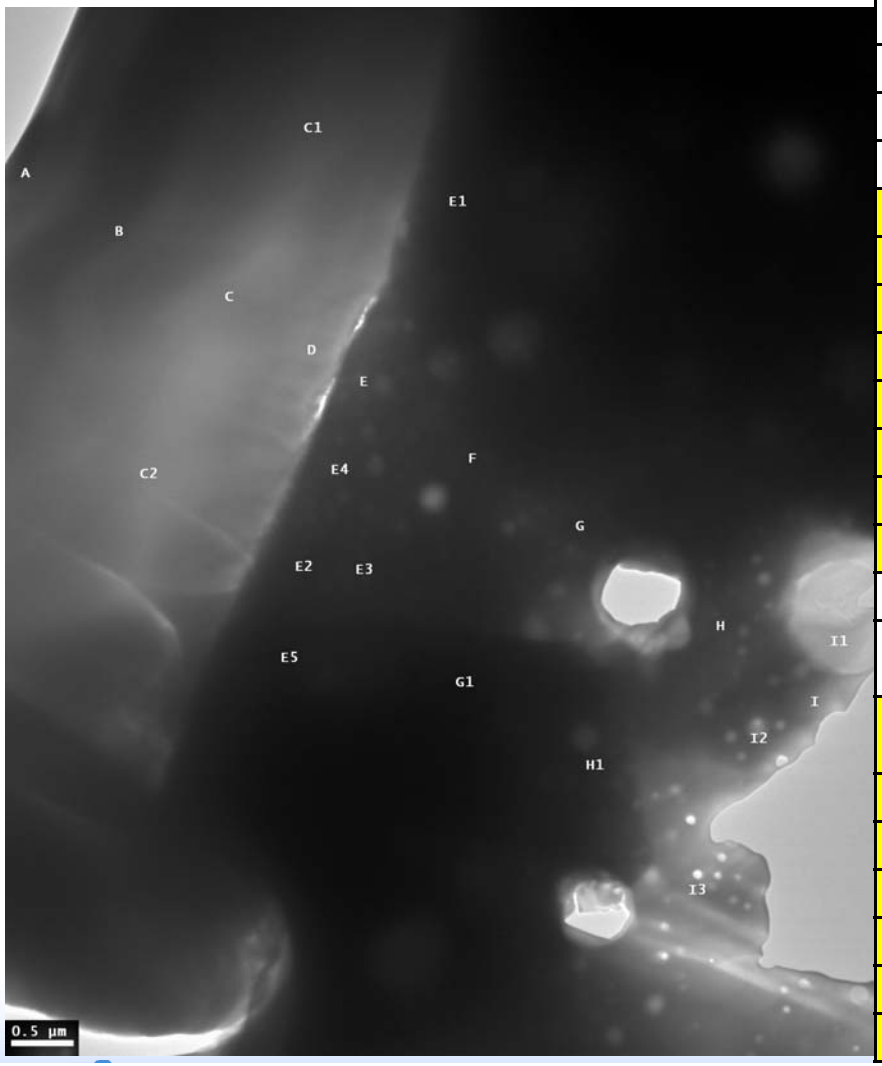
Fuel, FMI and Al Matrix in U_3Si_2/Al Fuel

Spot	U	Si	Al	Mo	(Si+Al)/(U+Mo)	U/Si	Note
A	0	0.9*	98.3	0.9	--	--	Al
B	9.1	10.8	77.5	2.5*	7.6	--	FMI
C	13.2	15.4	69.7	1.6	5.8	--	
D	13.9	14.1	71.4	0.9*	5.8	--	
E	13.4	14.7	70.7	1.3	5.8		
F	14.7	15.7	68.6	1.0	5.4	--	
J	16.4	15.5	67.8	0.3*	5.0		
G	18.3	16.4	64.1	1.2	4.1	--	
I	19.3	17.6	62.3	0.9*	4.0		
H	61.1	31.4	4.6	2.9	--	1.9	Fuel
K	48.5	34.4	17.4	0	--	1.4	
L	20.0	16.5	62.7	0.7*	3.8		FMI
M	17.4	14.5	67.3	0.8*	4.5		
N	16.6	14.7	68.2	0.6*	4.8		
O	15.6	14.5	69.5	0.5*	5.2		
P	14.0	14.0	71.0	0.9	5.7		
Q	11.2	10.5	77.4	1.0	7.2		



- Bubbles in I.L. distribute near Fuel/I.L interface where high U (> 16 at%) and low (Si+Al)/(U+Mo) (≤ 5 at%) are noticed.
- Large fraction of interaction layer shows very few visible bubble.

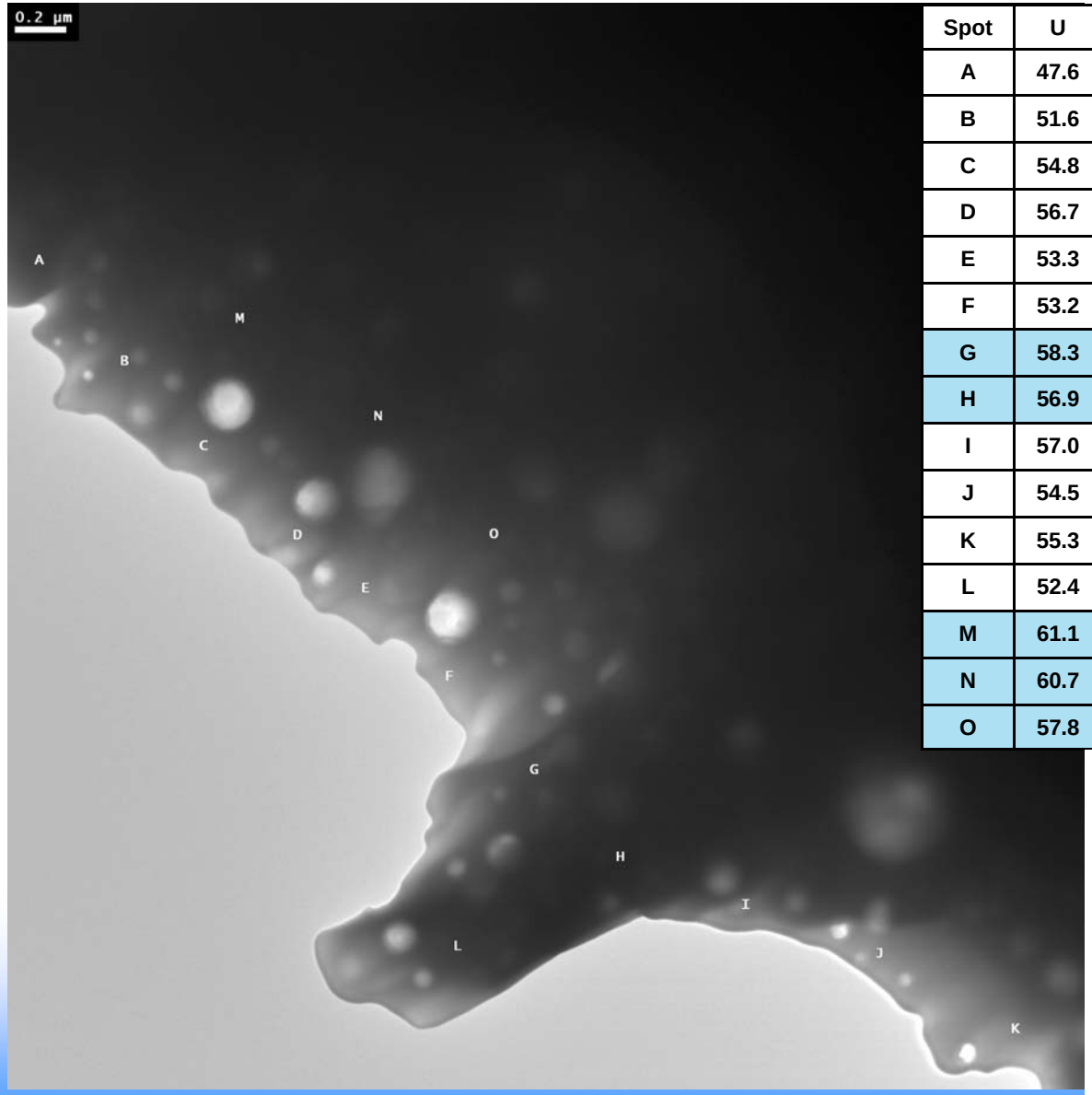
Fuel and FMI



Spot	U	Si	Al	Mo	Other	U/Si	Note
A	17.4	33.5	37.7	11.2	Y-2.1, Zr-2.8	--	I. L.
B	17.4	33.5	37.7	4.1	Y-2.6, Zr-3.4, Nd-1.2	--	I. L.
C	21.0	47.8	21.5	4.7	Y-1.7, Zr-4.3	--	I. L.
D	21.9	55.1	11.3	6.3	Y-2.0*, Zr-5.1	--	interface
E	41.0	35.3	21.6	2.1	--	1.2	interface
F	58.1	36.1	4.8	1.1*	--	1.6	Fuel
G	54.8	40.7	1.5*	3.0*	--	1.3	
H	50.1	41.5	5.4	3.1	--	1.2	
I	46.4	44.4	5.7*	3.5*	--	1.0	
I1	21.8	20.1	38.8	19.4	--	1.1	Large bb
I2	49.3	46.6	3.2	0.9*	--	1.1	Small bb
I3	48.3	46.2	4.1	1.5*	--	1.0	Small bb
C1	17.4	40.1	28.4	4.6	Y-1.7, Zr-4.3, Cs-1.9	--	I. L.
C2	12.2	44.9	29.4	6.1	Y-1.1, Zr-2.8, Cs-2.1 Nd-1.1	--	I. L.
E1	37.3	37.5	11.9	6.1	Y-1.4, Zr-1.9, Nd-2.8 Ba-1.1*,	1.0	
E2	61.1	18.3	8.5	6.0	Y-1.0, Zr-2.3, Nd-3.4	3.3	
E3	57.1	33.5	6.8	2.5	--	1.7	
E4	44.7	36.9	15.6	2.7	--	1.2	
E5	62.3	23.5	9.7	4.6	--	2.7	
G1	63.8	28.7	3.2*	4.3	--	2.2	
H1	52.5	40.8	3.6	3.2	--	1.3	

- Bubbles are only found in fuel zone, no bubbles are found in interaction layer at this magnification.
- Fission product are found in both fuel and interaction layer.
- High Al (~39 at%) and Mo (19 at%) are detected at one large bubble (spot I1).

EDS Measurement in U_3Si_2 Fuel Particle

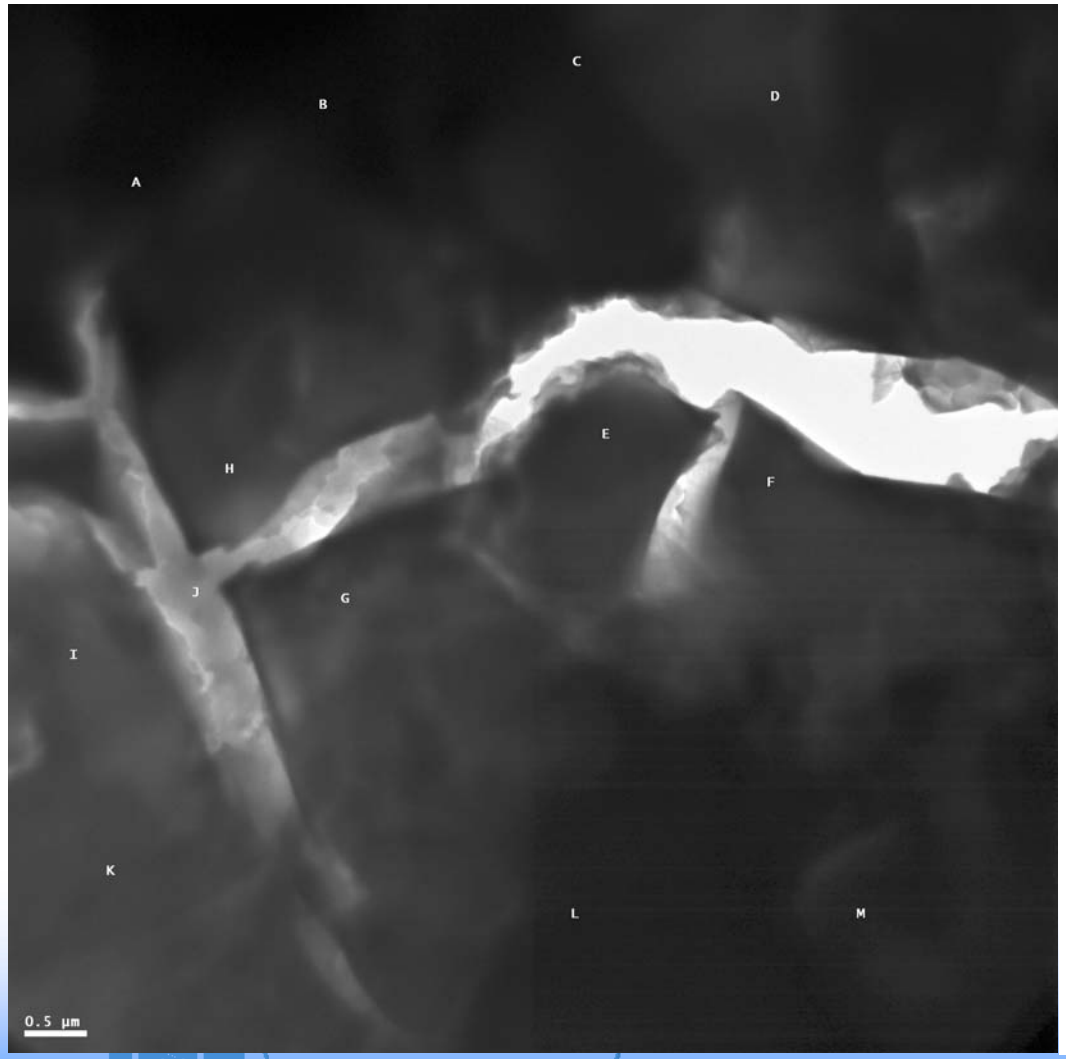


Spot	U	Si	Al	Mo	U/Si	Note
A	47.6	42.3	5.94	4.2	1.1	Thin area
B	51.6	44.0	2.6*	1.8*	1.2	
C	54.8	45.3	--	--	1.2	
D	56.7	43.3	--	--	1.3	
E	53.3	46.8	--	--	1.1	
F	53.2	46.8	--	--	1.1	
G	58.3	41.7	--	--	1.4	Thick area
H	56.9	43.1	--	--	1.3	Thick area
I	57.0	43.0	--	--	1.3	
J	54.5	45.5	--	--	1.2	
K	55.3	44.7	--	--	1.2	
L	52.4	47.6	--	--	1.1	
M	61.1	38.9	--	--	1.6	Thick area
N	60.7	39.3	--	--	1.5	Thick area
O	57.8	42.2	--	--	1.4	Thick area

- U/Si ratio in the thin area is close to USi while in the thick area it is close to U_3Si_2. The latter is likely a "local x-ray shielding" effect resulting an overestimate of the heavy element.

- With a fission density of 5.4×10^{21} fision/cm^3, the initial U/Si of 1.5 is expected to drop to approximately 1.2.

Solid Fission Product Precipitates



Spot	U	Si	Al	Mo	Other	Note
A	0	1.5	98.0	0	Ru-0.5	Al
B	0	1.4	98.5	0	--	Al
C	0	1.1	71.9	2.4	Ru-25	
D	0	1.2	67.8	3.0	Ru-28	
E	0	0.7	68.6	2.4	Ru-28	
F	0	2.9	79.5	3.2	Ru-15	
G	0	0.4*	97.0	0.5	Ru-2.1	
H	0	1.5	89.0	1.3	Ru-8.4	
I	0.1*	1.8	88.5	1.5	Ru-8.2	
J	0.2	3.4	94.5	1.3	Ru-0.5*	
K	0	1.4	98.5	0	--	Al
L	0	1.5	98.5	0	--	Al
M	0	1.4	88.5	1.0	Ru-9.2	

- Large Ru precipitates are found near light contrast strip feature consisting of mostly pure aluminum.

- This strip feature is also identified in the fresh U_3Si_2/Al fuel, likely introduced from fuel fabrication.

Summary

•TEM characterization capability for highly radioactive irradiated dispersion and monolithic fuel plates are successfully developed at INL.

•Site-specific TEM characterization using FIB lift-out from an SEM sample has great advantage over conventional TEM sample preparation:

- Easy access to areas of interest for analysis,
- Less personal radiation exposure from sample preparation,
- Low radiation background, better for EDS chemical analysis.

•General microstructural features between U-10Mo/Al monolithic fuel and U-7Mo/Al-2Si are quite similar on FMI and bubble superlattice.

•Stable fission gas bubble superlattice in U-Mo fuel is responsible for its excellent fuel performance.

•At high fission density (> 6×10^{21} fission/cm^3), significant fraction of U-Mo fuel turns to amorphous as a result of large bubble development and fission product accumulation.