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Comparison of neutron, proton, and self-ion irradiation of Fe-9%Cr ODS at 3 dpa, 500°C

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Objective

Develop a simple model to describe oxide nanocluster evolution in ODS alloys upon various irradiation conditions.

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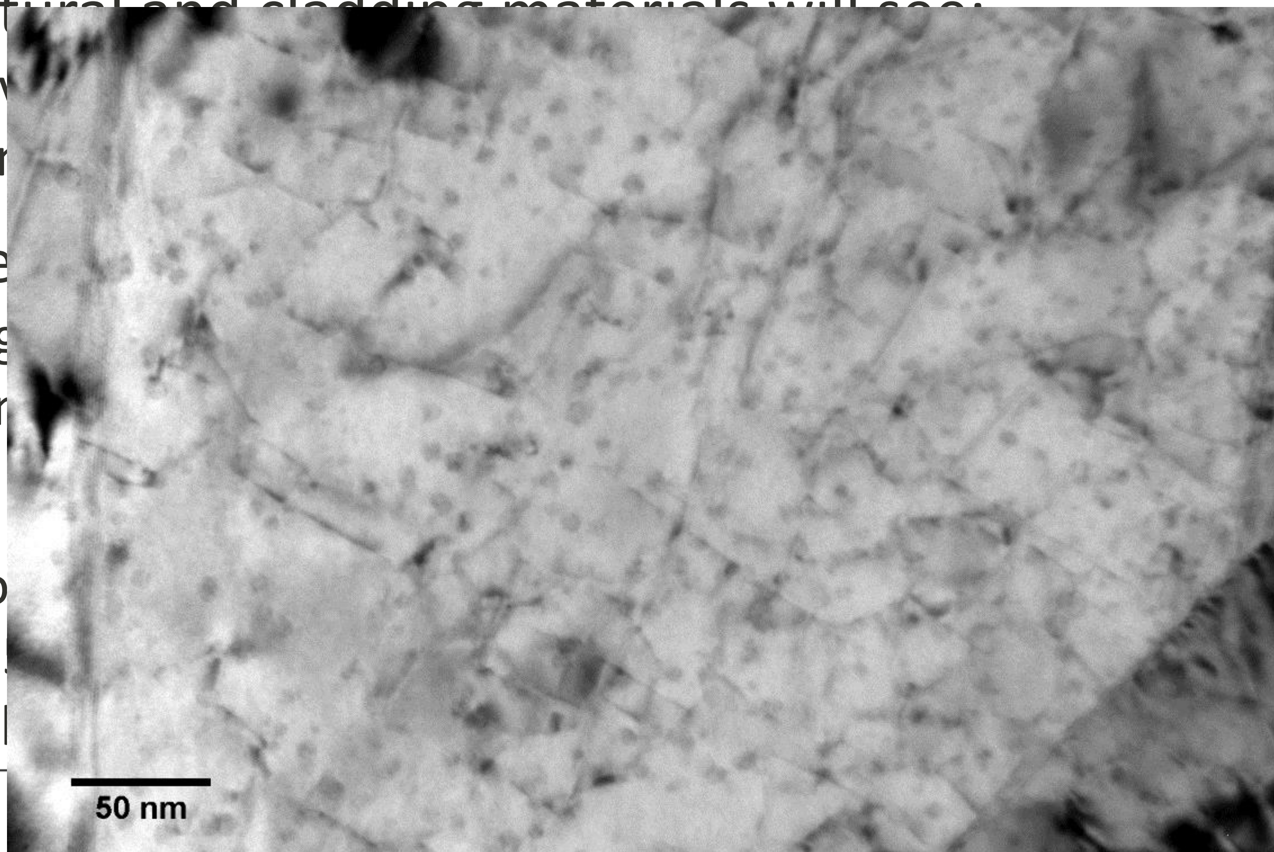
Materials Challenges for Gen IV Reactors

- Structural and cladding materials will see:
  - Se
  - Te
- Oxide
  - Hig
  - Dir
- JAEA
  - Ho
  - 10
  - Ful



C	Si	Mn	P	S	Ni	Cr	W	Ti	Y	O	N	Ar	Y_2O_3	Ex. O
0.14	0.048	0.05	<0.005	0.004	0.06	8.67	1.96	0.23	0.27	0.14	0.017	0.004	0.34	0.07
$[Y_2O_3] = 1.27 \times [Y]$														
$[Ex.O] = [Total\ O] - [O\ in\ Y_2O_3\ powder] = [O] - 0.27 \times [Y]$														

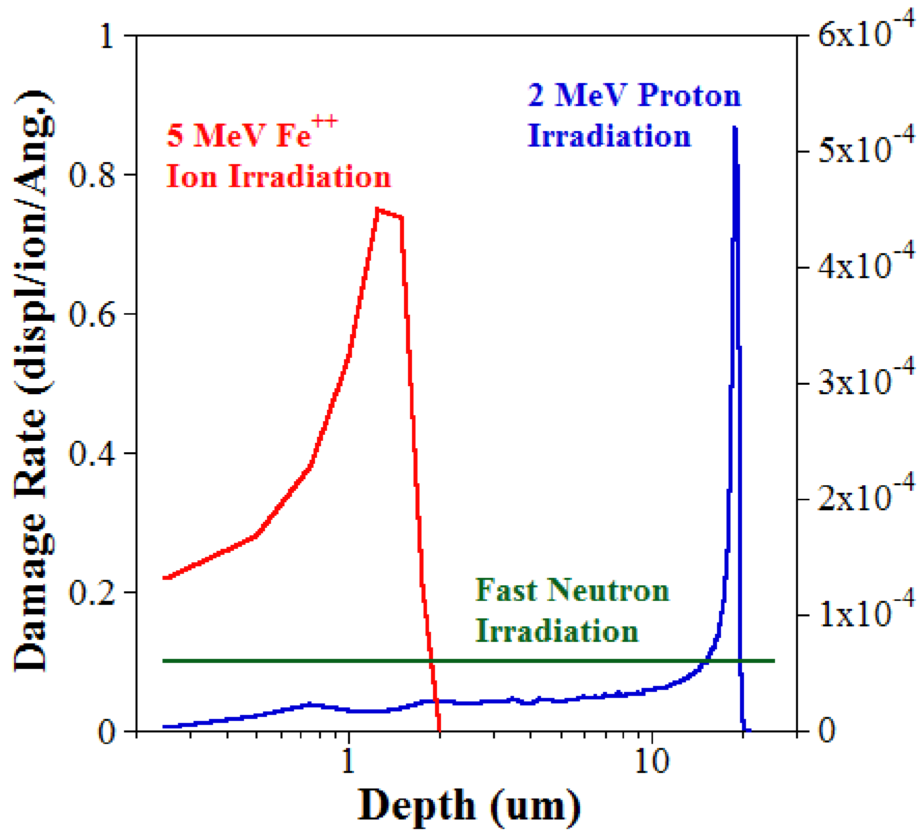
Irradiations

Charged particle Irradiation as surrogates for neutron irradiation:

- Different Dose rate
- Different Cascade morphology
  - Volume and efficiency
- Different Depth profiles

Increasing Dose Rate →

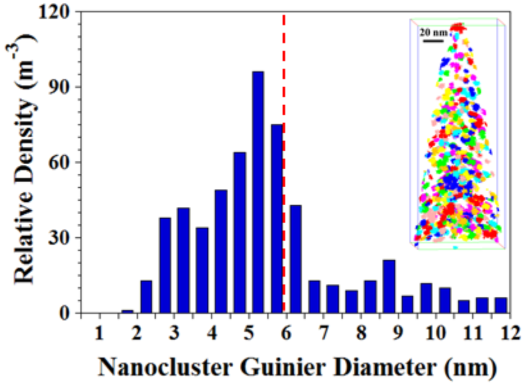
Temp.	Fast Neutron Irradiation (10^{-7} dpa/s)	2 MeV Proton Irradiation (10^{-5} dpa/s)	5 MeV Fe^{++} Ion Irradiation (10^{-4} dpa/s)	
500°C	3 dpa	3 dpa	3 dpa	100 dpa
400°C				100 dpa



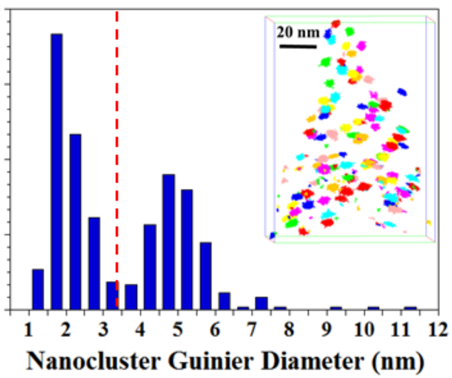
Neutron irradiation in Advanced Test Reactor (ATR)
Ion irradiations at Michigan Ion Beam Laboratory (MIBL)

Results – Particle Size Distributions

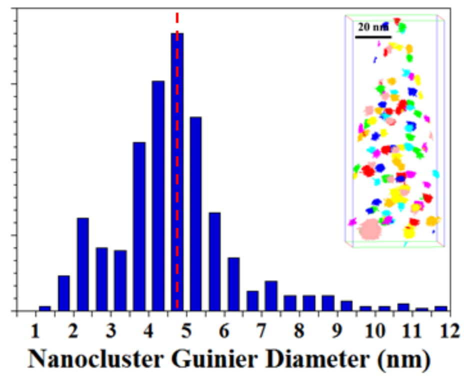
As-received



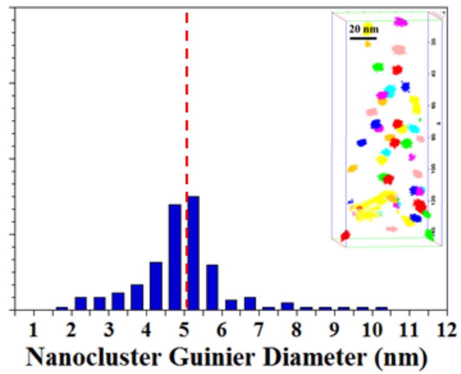
Neutron-Irradiated (3 dpa, 500°C)



Proton-Irradiated (3 dpa, 500°C)

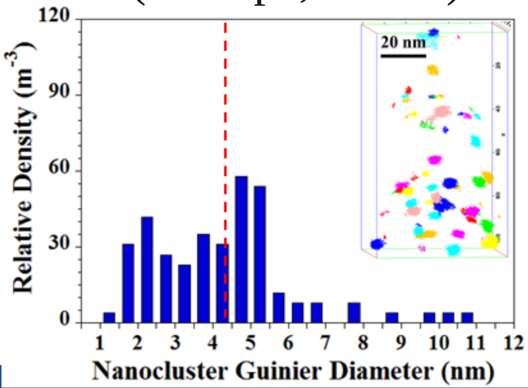


Fe^{++} Ion-Irradiated (3 dpa, 500°C)

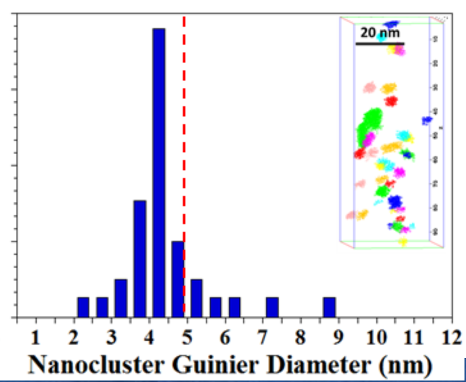


Increasing Dose Rate

Fe^{++} Ion-Irradiated (100 dpa, 400°C)

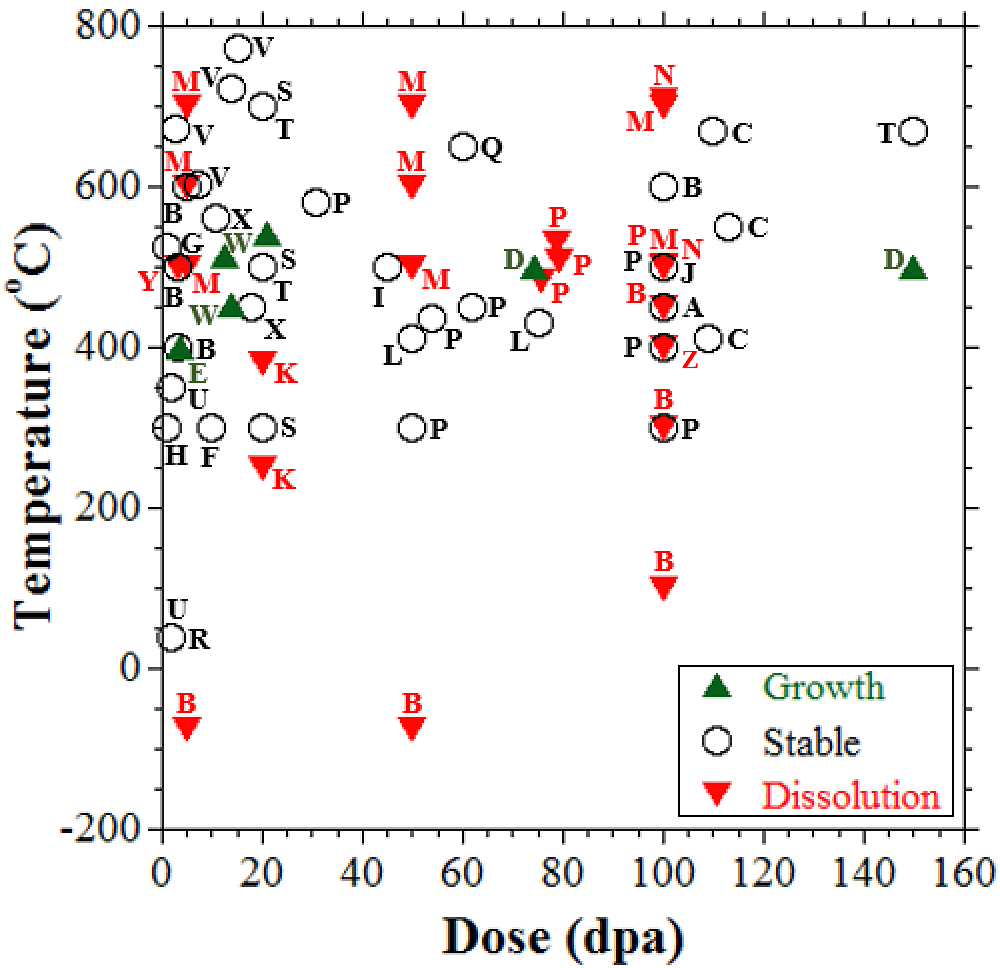


Fe^{++} Ion-Irradiated (100 dpa, 500°C)



Increasing Temperature

Literature Survey



A 14YWT - He, *JNM* 455 (2014)
B 14YWT, 9Cr ODS – Certain, *JNM* 434 (2013)
C MA957 – Bailey, *JNM* 459 (2015)
D Fe-18Cr-Y_2O_3 – Lescoat, *AM* 78 (2014)
E 9Cr ODS – He, *JNM* 452 (2014)
F EUROFER97 ODS – Rogozhkin, *AE* (2013)
G 9Cr ODS – Certain, *JNM* 407 (2010)
H 12YWT – Pareige, *JNM* 360 (2007)
I Fe-18Cr-Y_2O_3 – Lescoat, *JNM* 428 (2012)
J Model ODS – Robertson, *JNM* 426 (2012)
K F82H – Liu, *JNM* 417 (2011)
L MA957 – Ribis, *JNM* 444 (2014)
M 9Cr ODS – Allen, *JNM* 375 (2008)
N MA957 – Yamashita, *JNM* 367 (2007)
P DY & EM10+Y_2O_3 – Monnet, *JNM* 335 (2004)
Q 16Cr0.1Ti – Kishimoto, *JNM* 386 (2009)
R EUROFER97 ODS – Ramar, *JNM* 367 (2007)
S 19Cr-4.5Al ODS – Kimura, JNST 44 (2007)
T K1 & K4 – Kishimoto, *JNM* 367 (2007)
U EUROFER97 ODS – Schaublin, *JNM* 351 (2006)
V 9Cr and 12Cr ODS – Akasaka, *JNM* 329 (2004)
W F95, M93 – Yamashita, *JNM* 329 (2004)
X 1DS, 1DK – Yamashita, *JNM* 307 (2002)
Y Fe-9%Cr ODS – Swenson, *JNM* 467 (2015)
Z Fe-9%Cr-ODS – Swenson (this study)

Cluster Evolution – Simple Model

$$\frac{dr}{dt} = \underbrace{-\frac{\phi}{N}}_{\text{Recoil}} \underbrace{-\varphi K}_{\text{Disorder}} + \underbrace{\frac{3D^{irr}C}{4\pi pr} - D^{irr}r^2 n}_{\text{Growth}} \qquad [1]$$

Change in cluster radius upon irradiation

[1] R.S. Nelson, J.A. Hudson, and D.J. Mazey, "The stability of Precipitates in an Irradiation Environment," *J. Nucl. Mater.*, 44 (1972).

Cluster Dissolution – Recoil

$$\frac{dr}{dt} = \underbrace{-\frac{\phi}{N}}_{\text{Recoil}} - \varphi K + \frac{3D^{irr}C}{4\pi pr} - D^{irr}r^2 n$$

flux of atoms $\phi \sim 10^{14} \bullet K\,(\mathrm{cm}^2/\mathrm{s})$

$K =$ dose rate (dpa/s)

$N =$ atomic density of target $= 84.6$ atoms/nm^3 (b.c.c. Fe)

Depends on dose rate (i.e. irradiating particle)

Cluster Dissolution – Disorder

$$\frac{dr}{dt} = -\frac{\phi}{N} \underbrace{- \varphi K}_{\text{Disorder}} + \frac{3D^{irr}C}{4\pi p r} - D^{irr} r^2 n$$

$\varphi = l \bullet f$

[2]

- $l \sim$ size of damage cascade

  $l \sim$ 2.3 nm (2 MeV proton irradiation)

  $l \sim$ 6.8 nm (5 MeV Fe^{++} ion irradiation)

  $l \sim$ 10.4 nm (Fast neutron irradiation)

$l = f(\bar{T}, E_D, Z_1, Z_2, A_1, A_1, a, \varepsilon)$

$a =$ screening radius

$\varepsilon =$ unit electronic charge

(empirical functions)

- $f =$ fraction of dissolved solutes – per cascade (empirically fit)

Depends on dose rate and cascade morphology (i.e. irradiating particle)

[2] G.S. Was, *Fundamentals of Radiation Materials Science: Metals and Alloys*, Springer, (2007).

Cluster Evolution – Growth

$$\frac{dr}{dt} = -\frac{\phi}{N} - \varphi K + \underbrace{\frac{3D^{irr}C}{4\pi pr} - D^{irr}r^2 n}_{\text{Growth}}$$

$C =$ Total concentration of solutes (Y, Ti)

$p =$ fraction of solutes (Y, Ti) in clusters

$r =$ average radius of clusters

$n =$ number density of clusters

From Atom Probe Tomography Cluster Analysis (IVAS)

$D^{irr} =$ Radiation-enhanced diffusion of Y, Ti solutes - based on matrix content of Y, Ti

Depends on dose rate (irradiating particle), temperature and the target alloy (solute and cluster morphology)

Cluster Evolution – Calculation

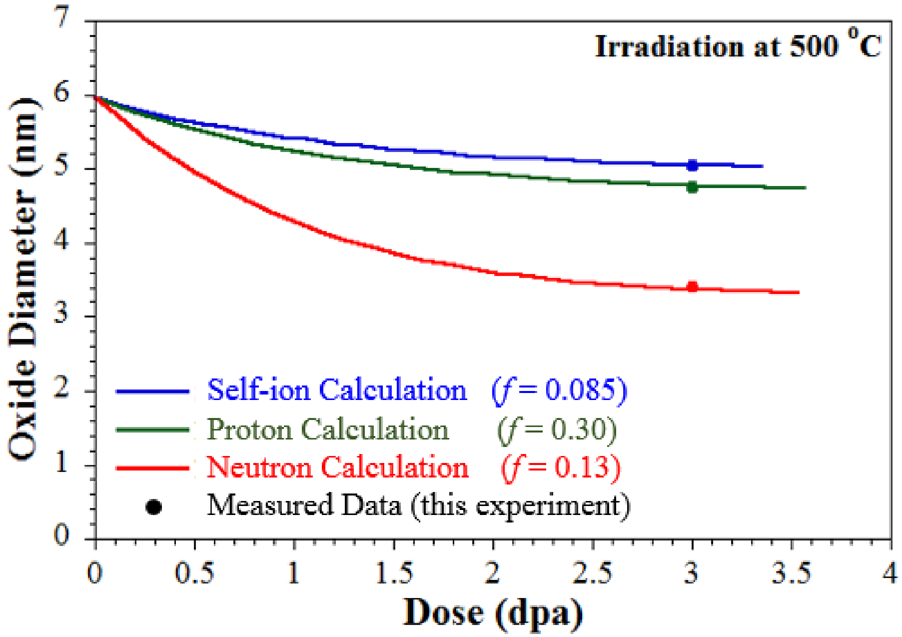
$$\frac{dr}{dt} = -\frac{\phi}{N} - \overbrace{l \cdot f}^{\varphi} \cdot K + \frac{3D^{irr}C}{4\pi pr} - D^{irr}r^2 n$$

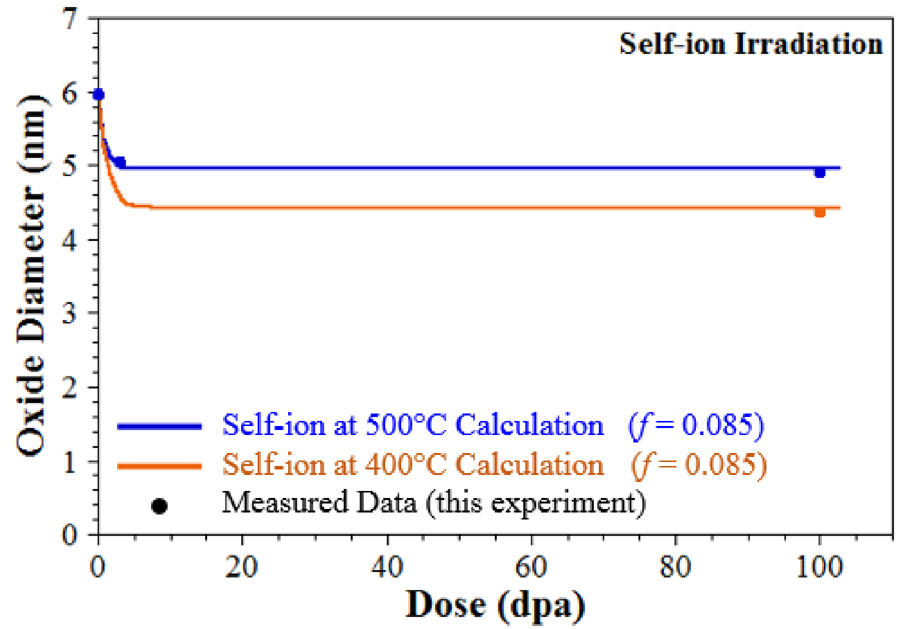
Empirically fit parameter

Cluster Evolution Iteration:

1) Use As-received parameters and estimate $\frac{dr}{dt}$
2) Apply $\frac{dr}{dt}$ over a period of time Δt (e.g. 3600 sec)
3) Calculate the cluster radius after Δt
4) Repeat calculation of $\frac{dr}{dt}$

Cluster Evolution – Simple Model





Model predicts that clusters will converge onto a stable size

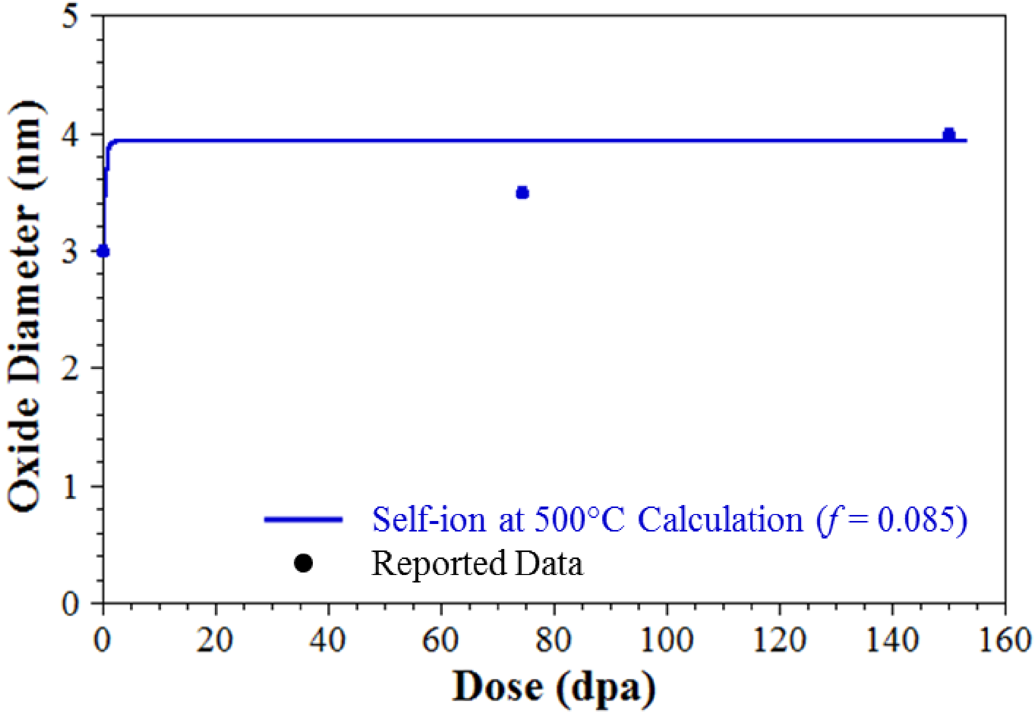
Limitations:

- Alloy system parameters are NOT updated after each Δt.
- Does NOT predict changes in cluster number density
- **Does NOT consider bimodal particle size distributions**

Model – Applied to Literature

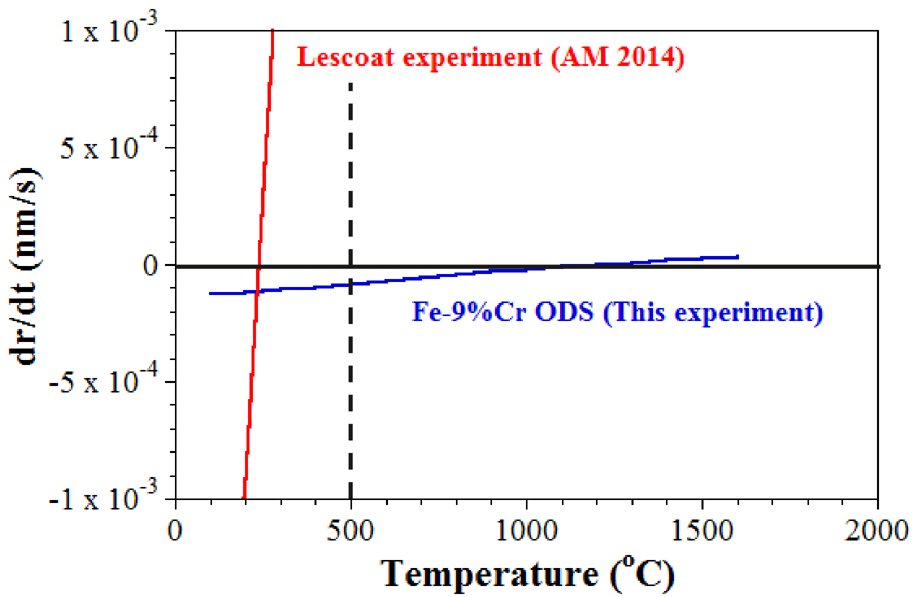
Lescoat, et al., Acta Materialia 78 (2014)[3]

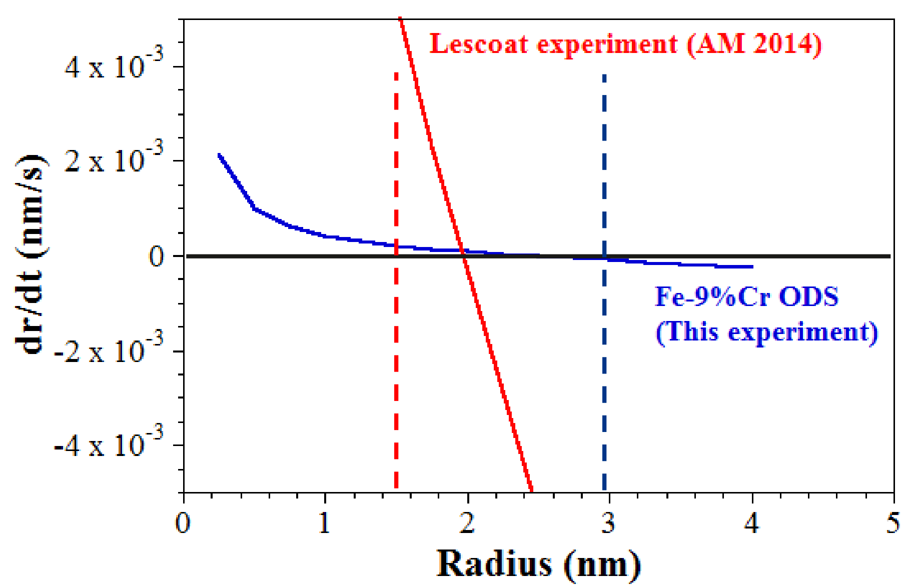
- Fe18Cr-Y_2O_3 ODS Alloy
  - $\bar{d} = 3.0\ nm$
  - $n = 2.3\ x\ 10^{23}\ m^{-3}$
  - Different Compositions
- 500 eV Fe^+ irradiation
  - at 500°C
  - $K = 6.4\ x\ 10^{-3}$ dpa/s



[3] M.-L. Lescoat, J. Ribis, Y. Chen, E.A. Marquis, E. Bordas, P. Trocellier, Y. Serruys, A. Gentils, O. Kaïtasov, Y. de Carlan, A. Legris, "Radiation-induced Ostwald ripening in oxide dispersion strengthened ferritic steels irradiated at high ion dose," *Acta Mater.*, 78 (2014).

Cluster Stability – At Onset of Irradiation



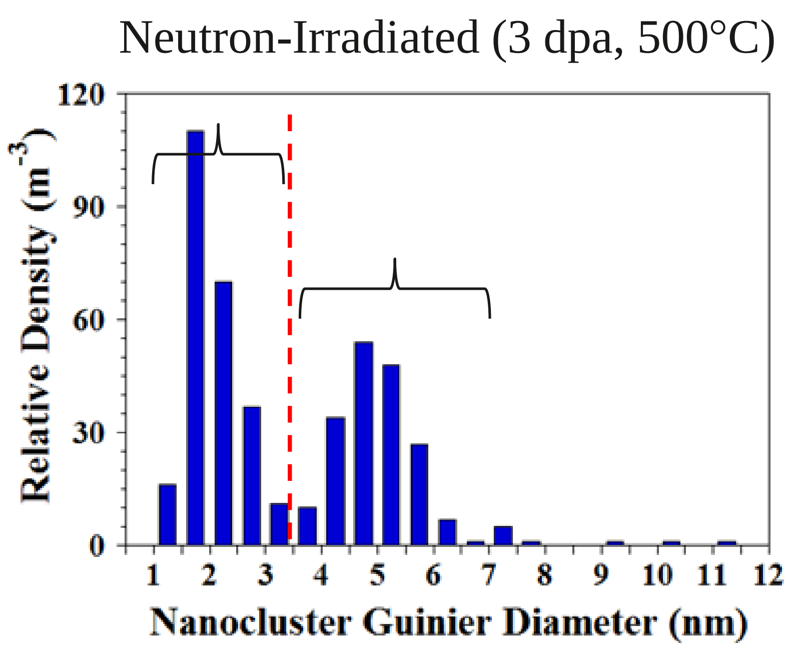


Notes:

- Lescoat experiment has higher does rate (6.4 x 10^{-3} vs. 2.2 x 10^{-4} dpa/s)
- Model correctly predicts direction of evolution (Growth or Dissolution)
- **Oxide evolution depends on Irradiation conditions AND alloy system**

Future Work

Consider bimodal Particle Size Distributions



1) Modelling the nucleation process for new clusters
2) Describing how number density will evolve

Conclusions

Oxide evolution upon irradiation depends on <u>irradiation conditions</u> AND <u>alloy system</u>

- Model predicts → stable cluster size
- Model predicts →Oxides may growth or dissolve

Consistent with variety of results reported in literature

Acknowledgements

Special Thanks:

- Jatu Burns, Dr. Yaqiao Wu

  Center for Advanced Energy Studies, MaCS

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  Michigan Ion Beam Laboratory

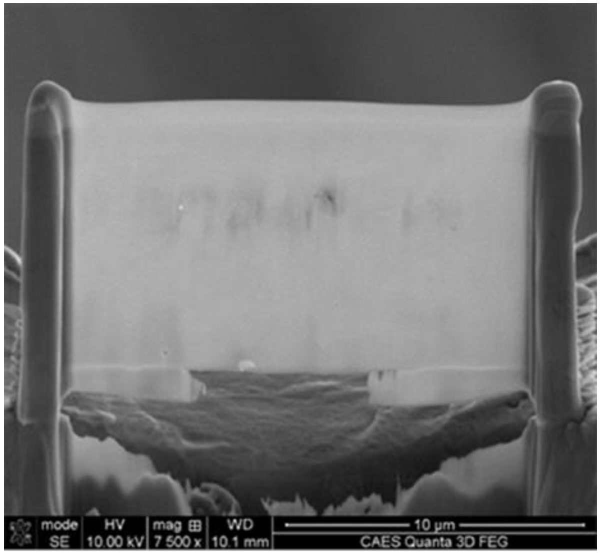
Matthew Swenson, Graduate Research Assistant
Email Matthewswenson1@u.boisestate.edu

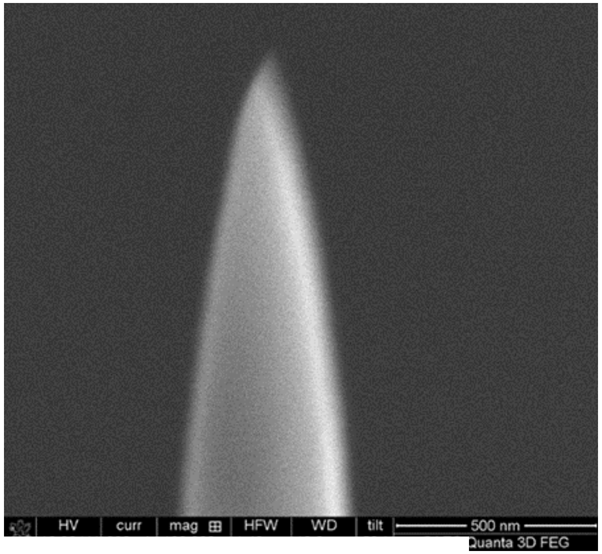
THANK YOU

Appendix

Microscopy

- Transmission electron microscopy (TEM)
  - FEI Tecnai TF30-FEG (300 kV)
  - Imaging grains/laths, dislocations, carbides, voids, dislocation loops
- Atom probe tomography (APT)
  - Cameca LEAP 4000X HR
  - Cluster analysis and solute composition measurements (IVAS)

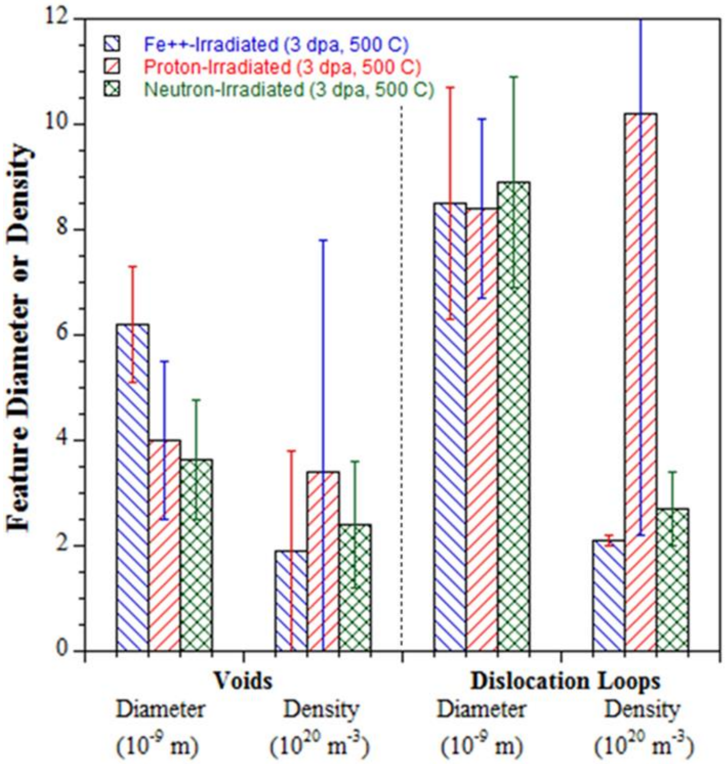


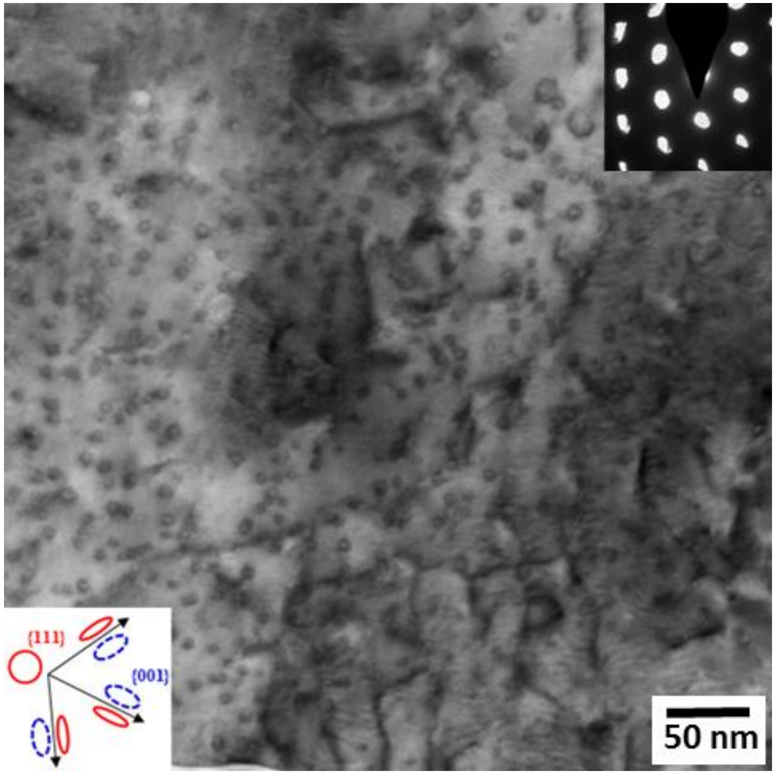


Performed at:
Center For Advanced Energy Studies (Idaho Falls, ID)
Microscopy and Characterization Suite (MaCS)

[3] M.J. Swenson and J.P. Wharry, "The comparison of microstructure and nanocluster evolution in proton and neutron irradiated Fe-9%Cr ODS steel to 3 dpa at 500°C," *J. Nucl. Mater.*, 467 (2015).

Results – Voids / Dislocation Loops





Grains/laths, dislocations and carbides were also statistically indifferent.
Charged particle irradiations replicate neutron irradiation for these features.

[3] M.J. Swenson and J.P. Wharry, "The comparison of microstructure and nanocluster evolution in proton and neutron irradiated Fe-9%Cr ODS steel to 3 dpa at 500°C," *J. Nucl. Mater.*, 467 (2015).

Cluster Dissolution – Disorder

$$\frac{dr}{dt} = -\frac{\phi}{N} - \underbrace{\varphi K}_{\text{Disorder}} + \frac{3D^{irr}C}{4\pi pr} - D^{irr}r^2 n$$

$$E_D = \frac{4}{3}\pi\left(\frac{l}{2}\right)^3 U_a N = \frac{T}{[1 + k_N g(\varepsilon_N)]}$$ [2]

φ (dissolution parameter) $= l \bullet f$

- $l \sim$ size of damage cascade

  $l \sim 2.3$ nm (2 MeV proton irradiation)

  $l \sim 6.8$ nm (5 MeV Fe^{++} ion irradiation)

  $l \sim 10.4$ nm (Fast neutron irradiation)

$$k_N = 0.1337 Z_1^{1/6}\left(\frac{Z_1}{A_1}\right)^{1/2}$$

$$g(\varepsilon_N) = 3.4008\varepsilon_N^{1/6} + 0.40244\varepsilon_N^{3/4} + \varepsilon_N$$

$$\varepsilon_N = \left(\frac{A_2 T}{A_1 + A_2}\right)\left(\frac{a}{Z_1 Z_2 \varepsilon^2}\right)$$

- $f =$ fraction of dissolved solutes – per cascade

$$a = \left(\frac{9\pi^2}{128}\right)^{1/3} a_0 \left(Z_1^{2/3} + Z_2^{2/3}\right)^{-1/2}$$

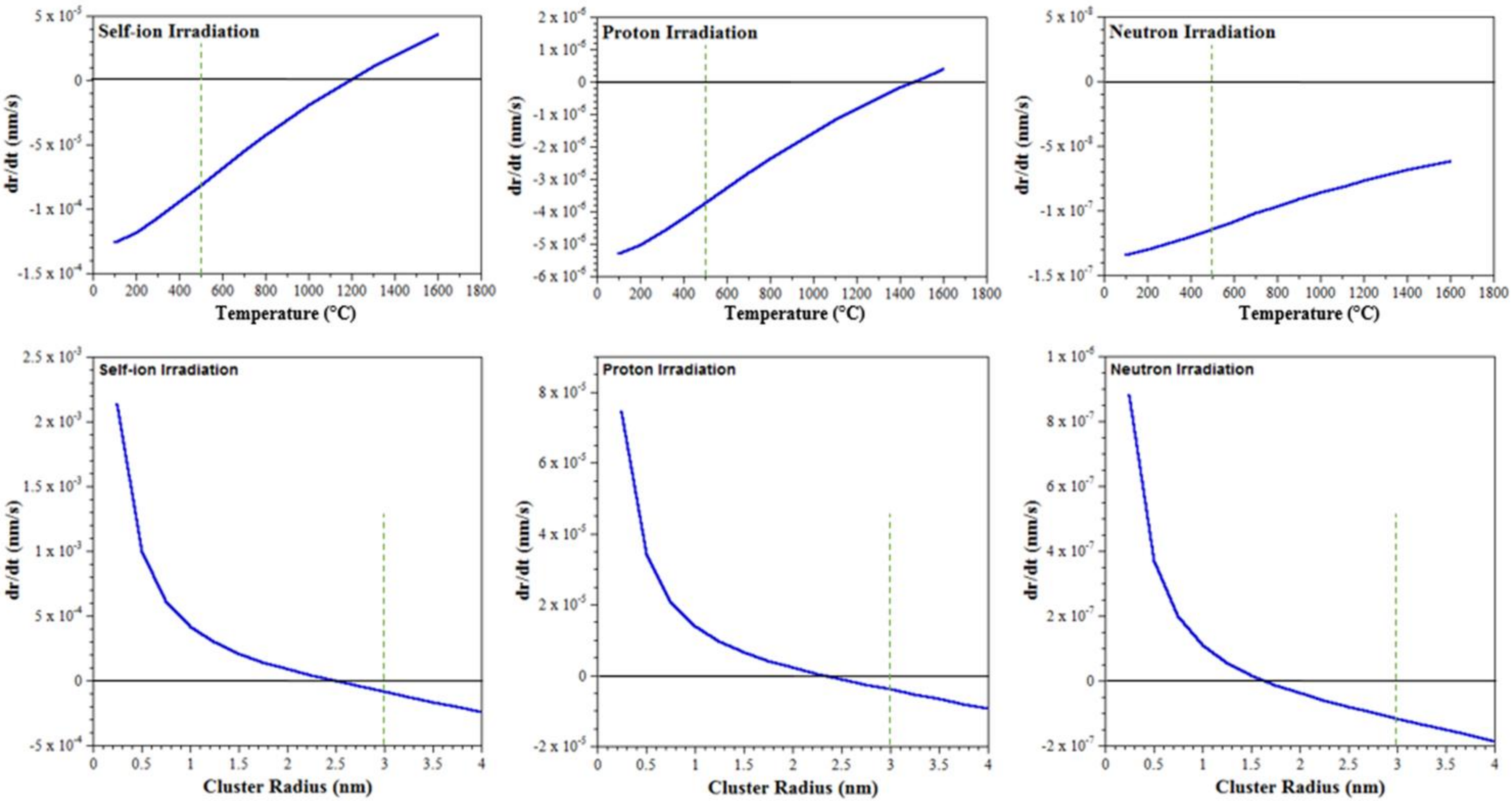
Depends on dose rate and cascade morphology (i.e. irradiating particle)

[2] G.S. Was, *Fundamentals of Radiation Materials Science: Metals and Alloys*, Springer, (2007).

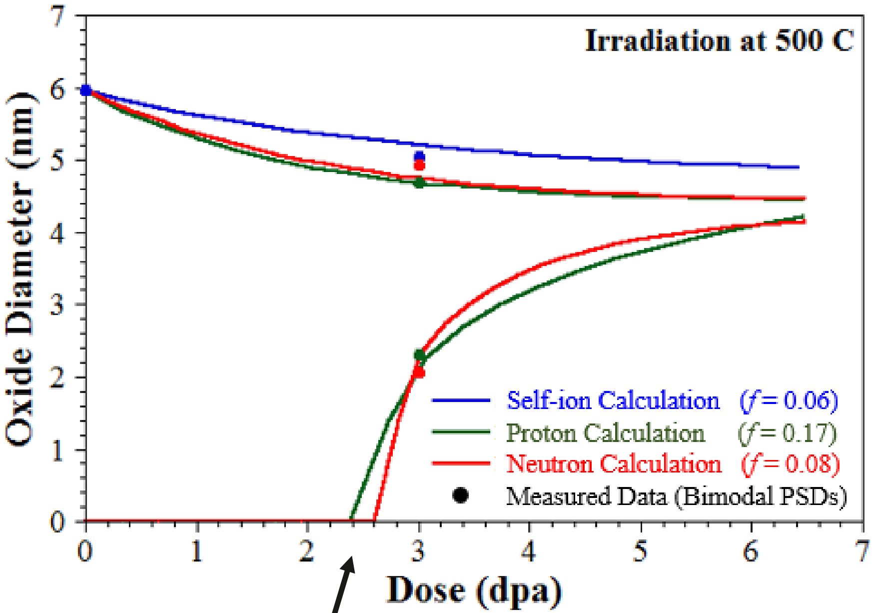
Solute Concentrations

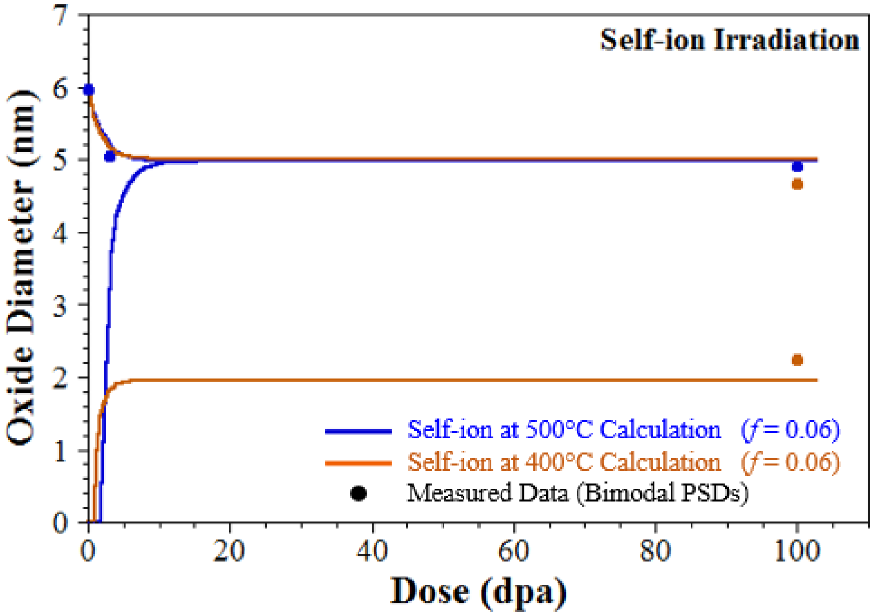
Oxide nanoclusters	As-received	Fe^{++} Irr. (100 dpa, 400°C)	Fe^{++} Irr. (100 dpa, 500°C)	Fe^{++} Irr. (3 dpa, 500°C)	Proton-Irr. (3 dpa, 500°C)	Neutron-Irr. (3 dpa, 500°C)
Cluster Composition	**at. %**	**at. %**	**at. %**	**at. %**	**at. %**	**at. %**
Y	2.41 ± 1.24%	2.57 ± 1.85%	2.44% ± 1.41%	4.24 ± 1.66%	2.43 ± 1.31%	2.93 ± 1.85%
Ti	4.77 ± 1.75%	6.46 ± 3.43%	5.98% ± 2.77%	5.35 ± 3.14%	4.64 ± 1.74%	3.75 ± 2.28%
O	6.25 ± 3.49%	7.87 ± 4.91%	7.64% ± 4.50%	8.56 ± 5.80%	6.24 ± 3.14%	5.85 ± 4.45%
Cr	9.78 ± 2.08%	11.11 ± 2.69%	6.56% ± 1.66%	11.32 ± 2.21%	11.80 ± 2.52%	10.55 ± 3.25%
Si	0.19 ± 0.17%	0.25 ± 0.30%	0.07% ± 0.09%	0.32 ± 0.19%	0.30 ± 0.23%	0.38 ± 0.49%
Mn	0.07 ± 0.10%	0.09 ± 0.23%	0.03% 0.05%	0.11 ± 0.10%	0.08 ± 0.12%	0.19 ± 0.30%
Ni	0.04 ± 0.08%	0.05 ± 0.09%	0.09% 0.10%	0.14 ± 0.10%	0.10 ± 0.10%	0.13 ± 0.23%
C	0.27 ± 0.30%	0.48 ± 0.38%	0.08% 0.09%	0.38 ± 0.30%	0.25 ± 0.25%	0.28 ± 0.45%
W	0.58 ± 0.39%	0.75 ± 0.61%	0.48% ± 0.25%	0.57 ± 0.33%	0.37 ± 0.30%	0.58 ± 0.62%
Matrix Composition	**at. %**	**at. %**	**at. %**	**at. %**	**at. %**	**at. %**
Y	0.05%	0.06%	0.02%	0.07%	0.04%	0.10%
Ti	0.11%	0.16%	0.09%	0.23%	0.15%	0.32%
O	0.17%	0.25%	0.11%	0.27%	0.19%	0.37%
Cr	8.38%	8.02%	3.72%	8.15%	8.21%	8.14%
Si	0.15%	0.12%	0.01%	0.13%	0.13%	0.12%
Mn	0.06%	0.05%	0.03%	0.08%	0.06%	0.08%
Ni	0.03%	0.03%	0.03%	0.05%	0.03%	0.05%
C	0.15%	0.21%	0.03%	0.16%	0.12%	0.16%
W	0.61%	0.61%	0.84%	0.74%	0.60%	0.67%
Trace amounts of P, S, N and H, Ga detected (balance is Fe)						

Cluster Stability – At Onset of Irradiation



Cluster Evolution – Split PSDs





Introduced nucleation

Denuded Regions for Clusters

Fe^{++} Irradiated to 3 dpa at 500°C