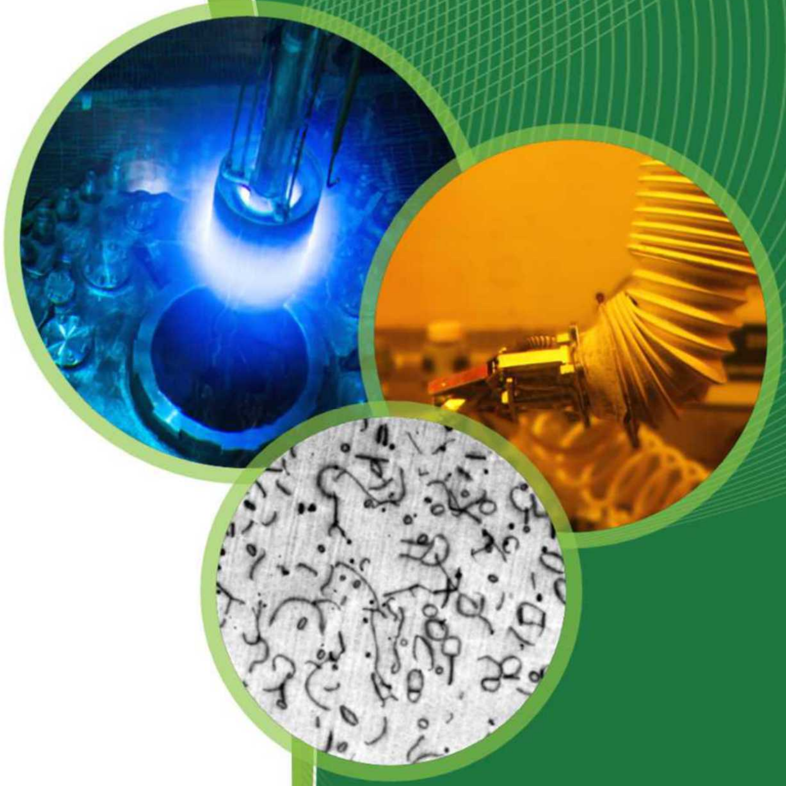
Ex-situ and In-situ Determination of α' Phase Formation/Dissolution in High-Cr Ferritic Alloys Using Small-Angle Neutron Scattering

SAND2018-2786C



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Advanced Fuels Campaign

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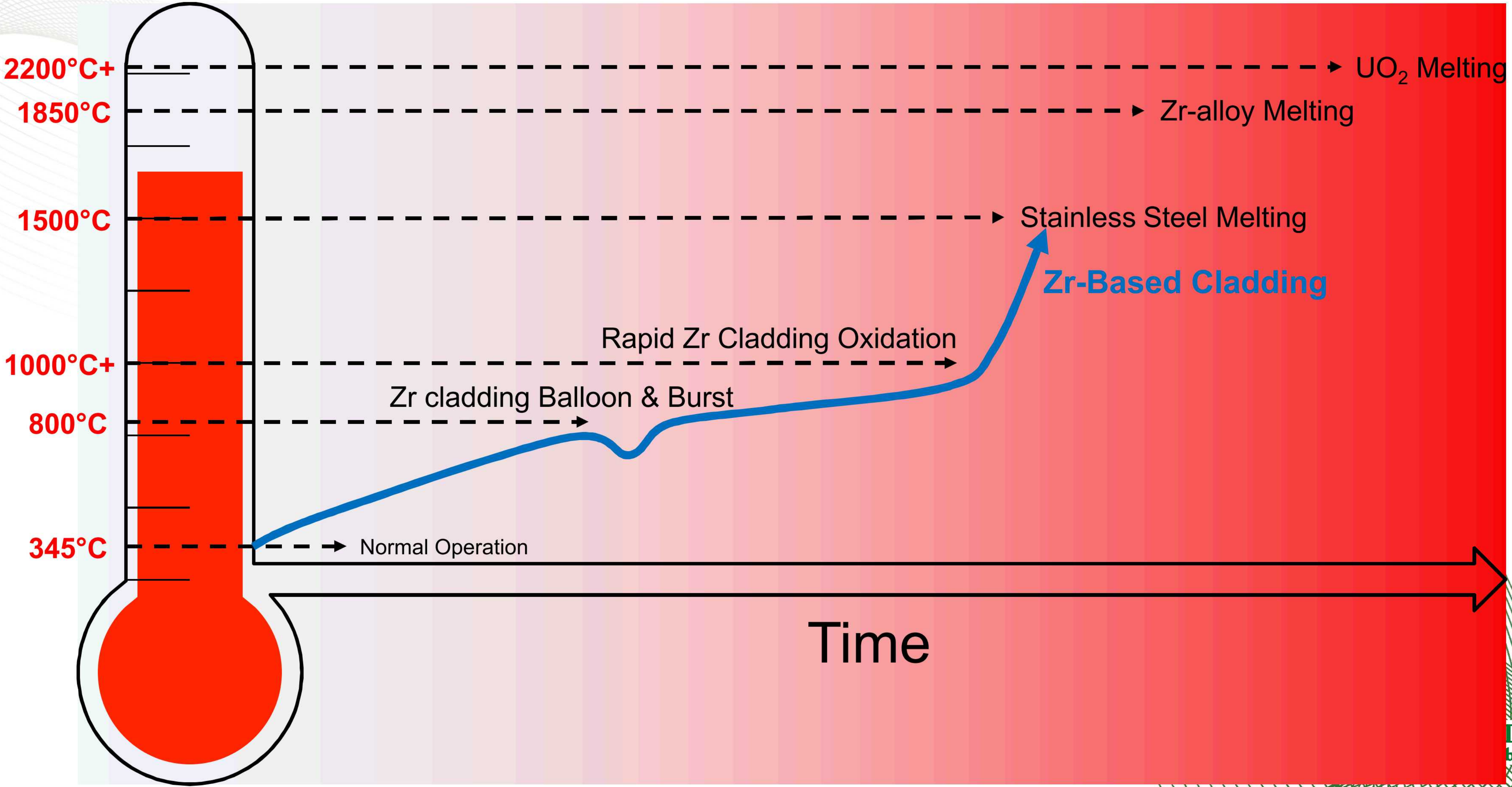
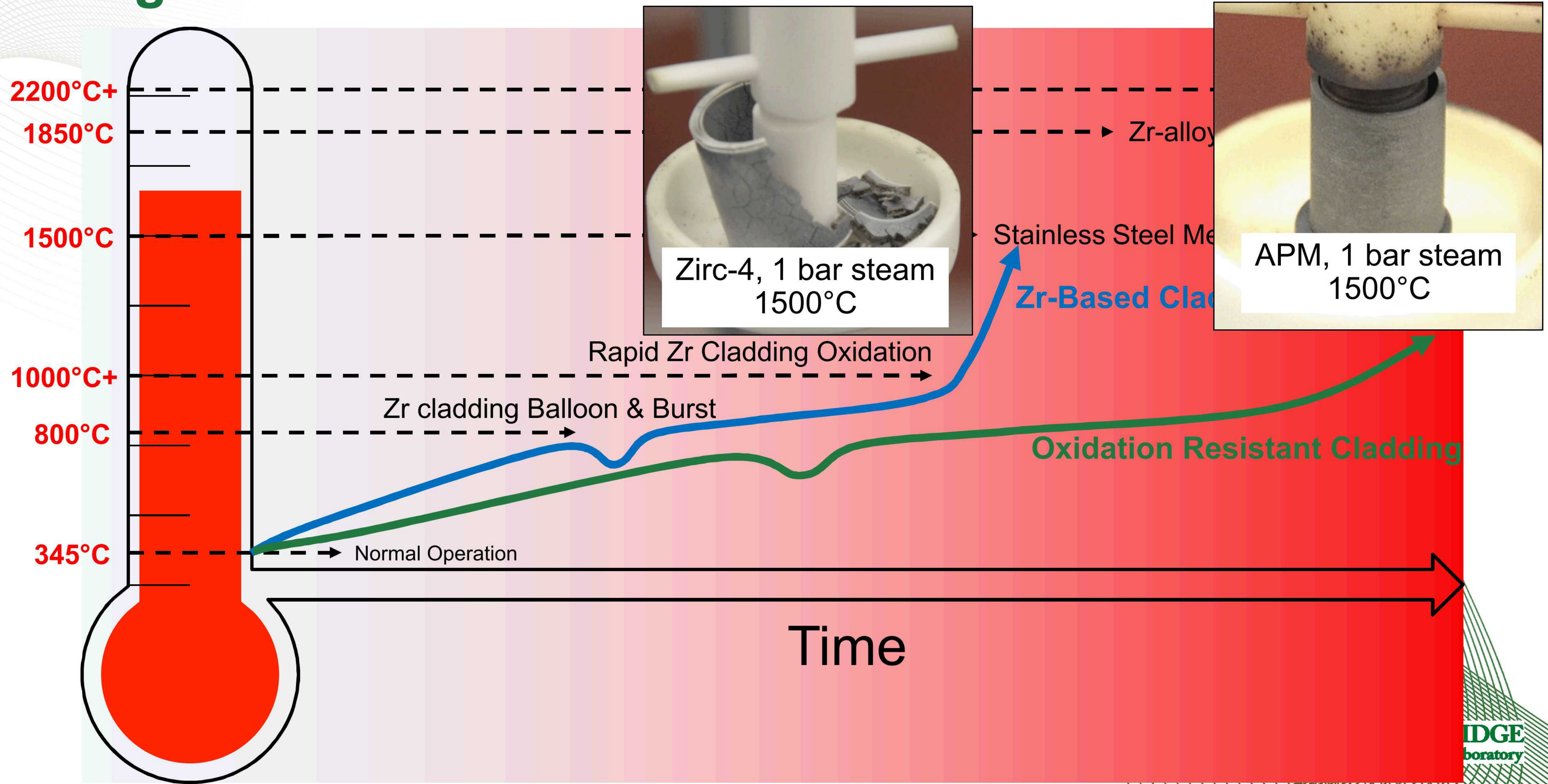
Oxidation of cladding is key towards core degradation during a coolant-limited accident scenario



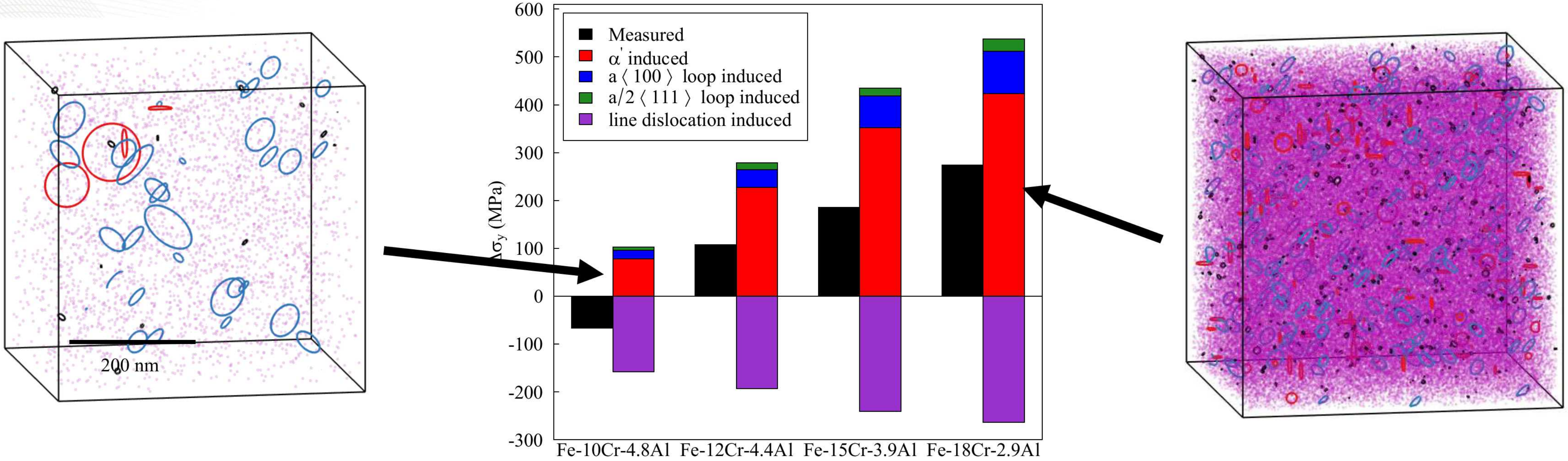
Oxidation of cladding is key towards core degradation during a coolant-limited accident scenario



A large amount of FeCrAl samples have been irradiated and/or tested over past 5 years…

…how do we characterize α' from these efficiently?

Mechanical properties can be linked to the microstructure



- Dispersed barrier hardening model can be used to develop a simplified structure-property relationship:

$$\Delta\sigma_y = M\mu b(\alpha_{\alpha'}\sqrt{\rho_{\alpha'}\cdot d_{\alpha'}} + \alpha_{a/2\langle 111\rangle}\sqrt{\rho_{a/2\langle 111\rangle}\cdot d_{a/2\langle 111\rangle}} + \alpha_{a\langle 100\rangle}\sqrt{\rho_{a\langle 100\rangle}\cdot d_{a\langle 100\rangle}} + \alpha_d(\sqrt{\rho_d}-\sqrt{\rho_o}))$$

0.04 0.05 0.31 0.68

- Even with an over-simplified model, the dominant hardening response is FeCrAl composition dependent due to the α’ precipitation

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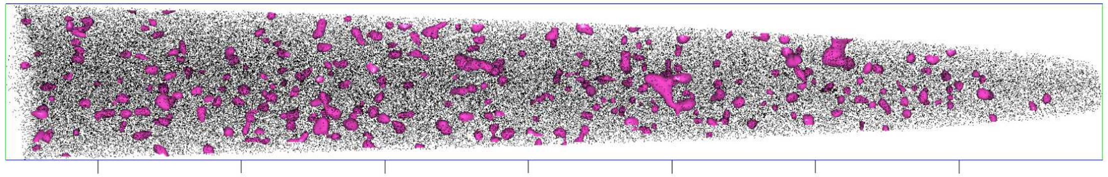
Nanoscale characterization techniques for α' precipitation

α': 2-10 nm spherical precipitates with ~80-90 wt.% Cr



Small-Angle Neutron Scattering

- \+ Non-destructive
- \+ In-situ/time resolved
- \+ Magnetic information
- \+ Large volume/good statistics
- \+ Limited sample preparation
- \- Flux limited
- \- No spatial information
- \- Indirect chemical/morphology information

TEM/STEM/EFTEM

- \+ High spatial resolution
- \+ Can provide microstructure/ chemical relationships
- \+ Fast data collection
- \- Poor signal to noise (EFTEM/EDS)
- \- Limited sample volume
- \- Destructive/time consuming
- \- Sample quality limited

Atom Probe Tomography

- \+ High spatial resolution
- \+ Direct determination of chemical information
- \- Slow data collection
- \- Limited sample volume
- \- Destructive/time consuming
- \- Prone to aberration related errors
- \- Sample quality limited

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SANS Scattered Intensity: Local Monodispersed Approximation[1]

$$\frac{d\sigma}{d\Omega}(q) = \Delta\rho^2 \int_0^\infty \phi(q,R)^2 S[q, R_{HS}(R)] N(R)\, dR + Aq^{-B} + C$$

- Contrast: $\Delta\rho^2 = (\rho_{particle} - \rho_{matrix})^2$
- Form factor (for spheres): $\phi(q,R) = 3V_o[\sin(qR) - qR\cos(qR)]/(qR)^3$
- Structure factor: $S[q, R_{HS}(R)] = [1 + 24\eta_{HS} G(R_{HS}q)/(R_{HS}q]^{-1}$
- Size distribution (Weibull density function): $N(R) = (R/\bar{R})^{b-1} exp[-(R/\bar{R})^b]$
- Low-q power-law: Aq^{-B}
- Background: C

SANS Scattered Intensity:
Contrast from Neutrons

1. ***Nuclear contrast:***

$$\Delta\rho_{nucl}^2 = \left(\rho_{nucl,particle} - \rho_{nucl,matrix}\right)^2$$

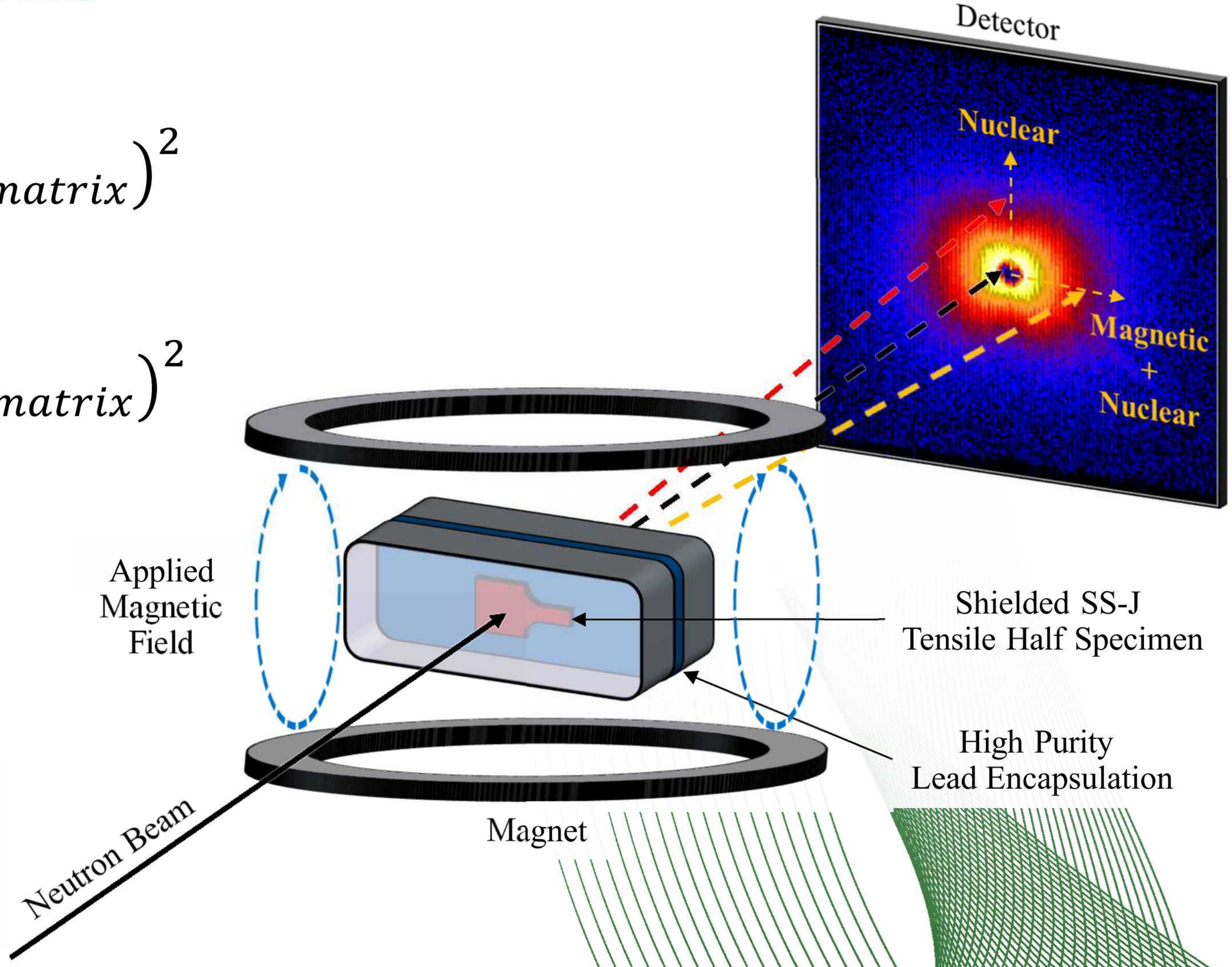
2. ***Magnetic contrast:***

$$\Delta\rho_{mag}^2 = \left(\rho_{mag,particle} - \rho_{mag,matrix}\right)^2$$

$$\frac{d\sigma}{d\Omega}(q) = \Delta\rho^2 \int_0^\infty \phi(q,R)^2 \ldots []$$

$$\left[\Delta\rho_{nucl}^2 + \Delta\rho_{mag}^2 sin^2\alpha\right]$$

Can exploit magnetization to extract composition



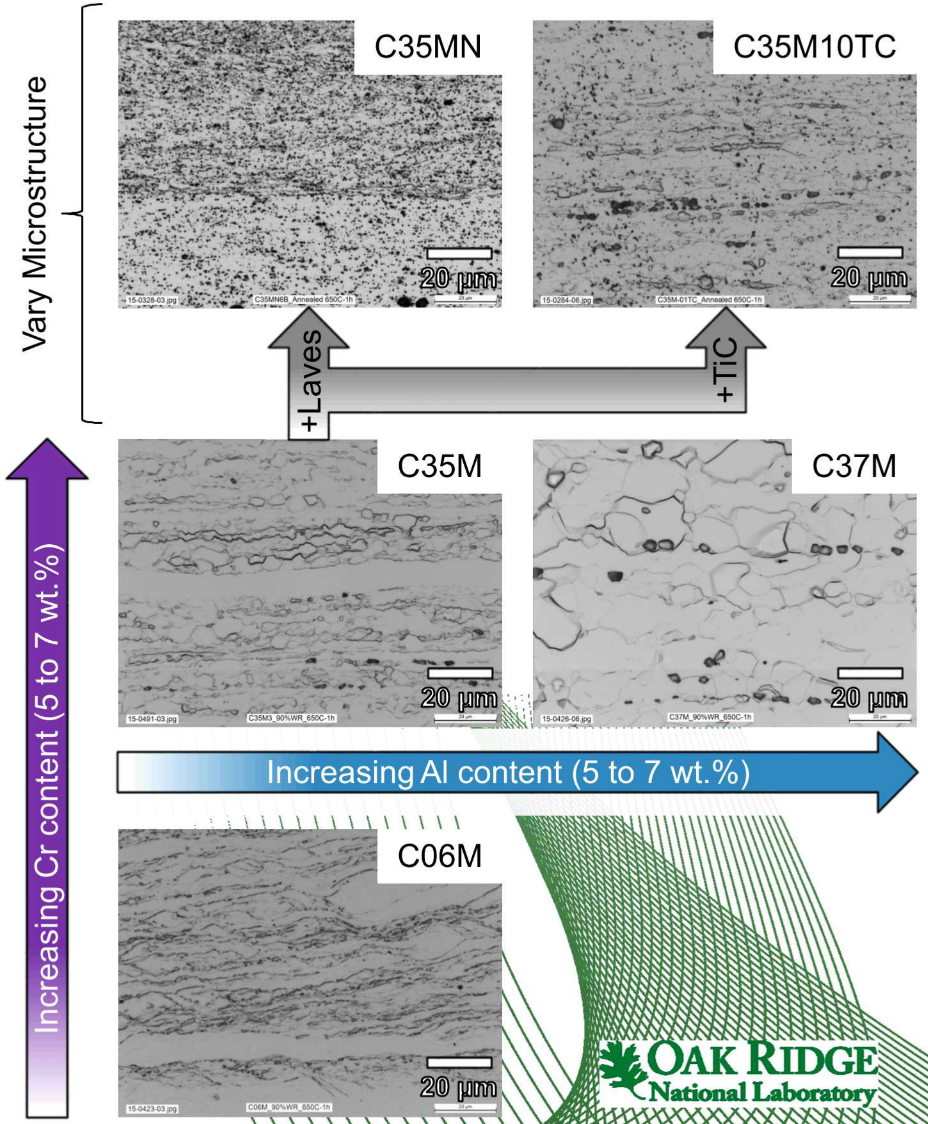
Magnetic Shielded-SANS measurements: Role of temperature and composition on α'

- Selected FeCrAl alloys irradiated in HFIR to determine temperature, composition, and microstructure trends

Capsule ID	Number Samples	Neutron Flux (n/cm^2s) E > 0.1 MeV	Neutron Fluence (n/cm^2) E > 0.1 MeV	Dose Rate (dpa/s)	Dose (dpa)	Irradiation Temperature (°C)
FCAT-01	45	1.10×10^{15}	2.17×10^{21}	9.6×10^{-7}	1.9	194.5 ± 37.9
FCAT-02	45	1.04×10^{15}	2.05×10^{21}	9.1×10^{-7}	1.8	362.7 ± 21.2
FCAT-03	45	1.10×10^{15}	2.17×10^{21}	9.6×10^{-7}	1.8	559.4 ± 28.1

Increasing Temp.

- Measurements performed at CG-2 general purpose SANS beamline at HFIR
- Data collected on broken tensile heads in magnetic shielded SANS configuration



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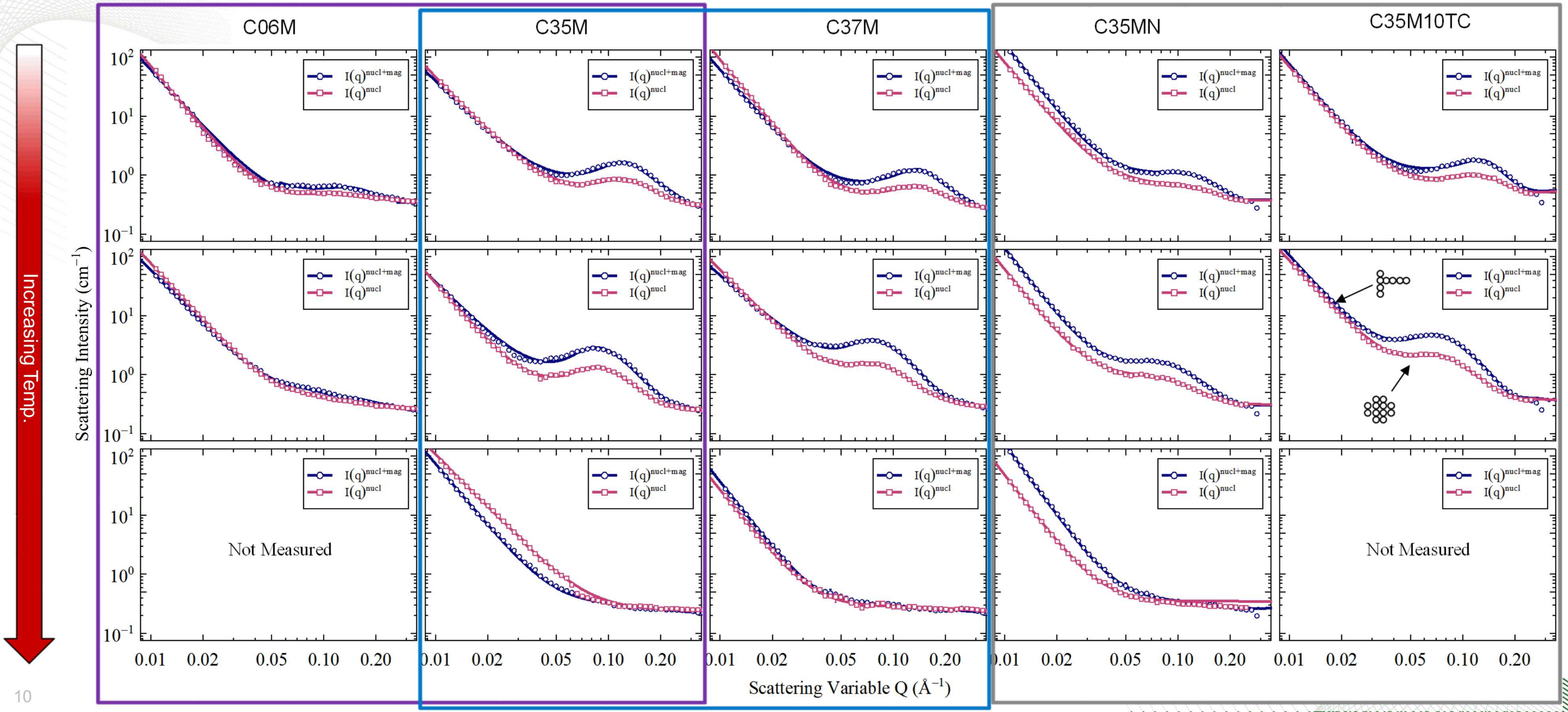
Magnetic Shielded-SANS measurements: Role of temperature and composition on α'



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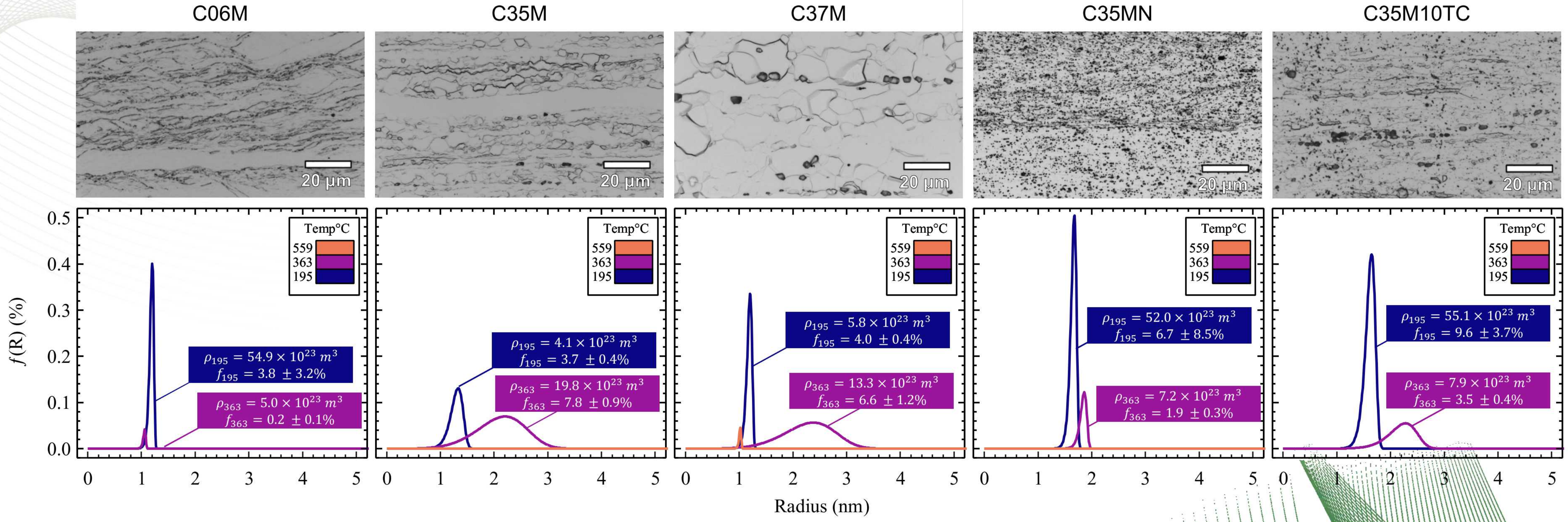

- Increasing temp. increases mean radius and distribution width
- Cr stronger than Al in control
- Microstructure (precipitates/grain size) play a role

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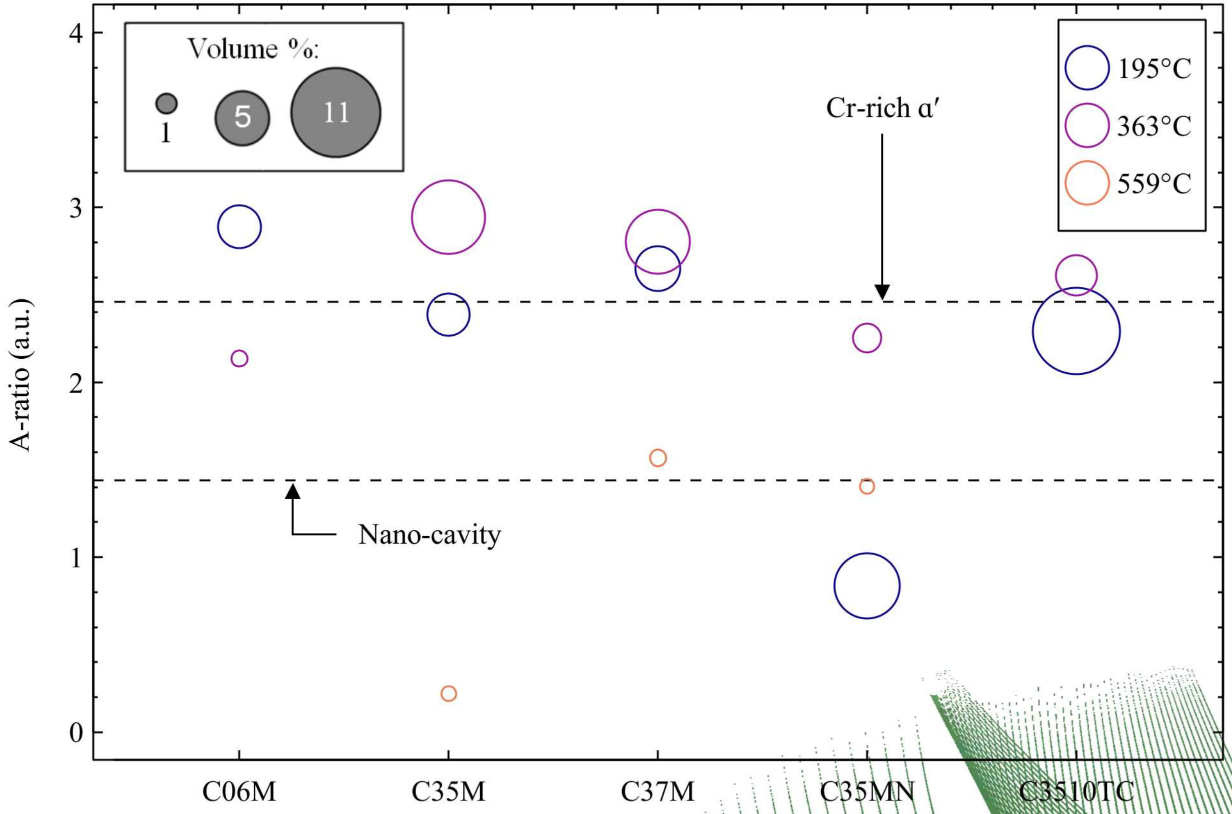
Magnetic Shielded-SANS measurements: Role of temperature and composition on α'

$$\frac{d\sigma}{d\Omega}(q) = \Delta\rho^2 \int_0^\infty \phi(q,R)^2 \ldots []$$

$$\left[\Delta\rho_{nucl}^2 + \Delta\rho_{mag}^2 sin^2\alpha\right]$$

$$A - ratio = \frac{\Delta\rho_{mag+nucl}^2}{\Delta\rho_{nucl}^2}$$



- Magnetic shielded SANS reveals that irradiations below 500°C form primarily α'
  - Lower Cr content in 195°C irradiations
- Above 500°C, scattering could be from nano-cavity formation

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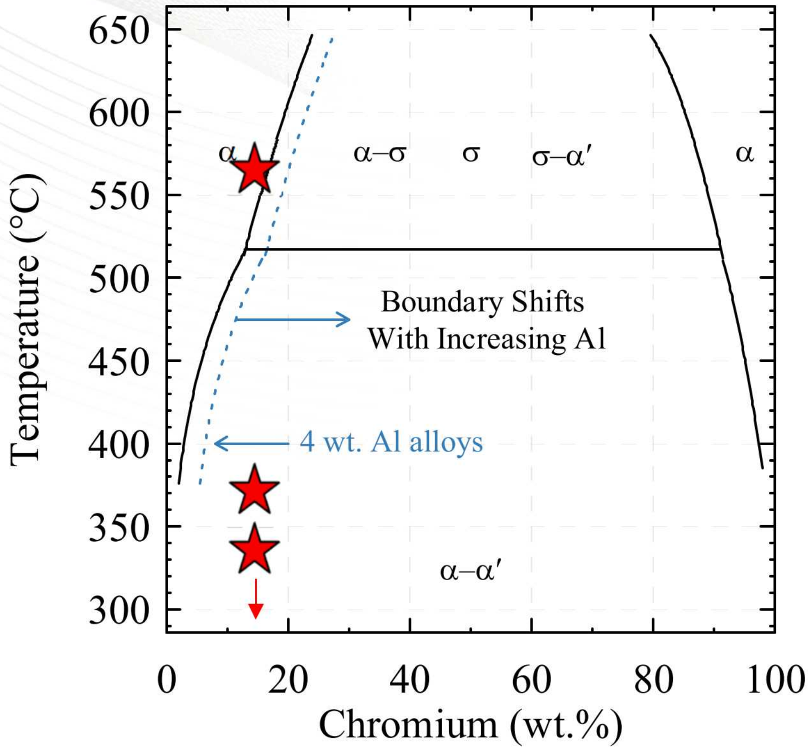
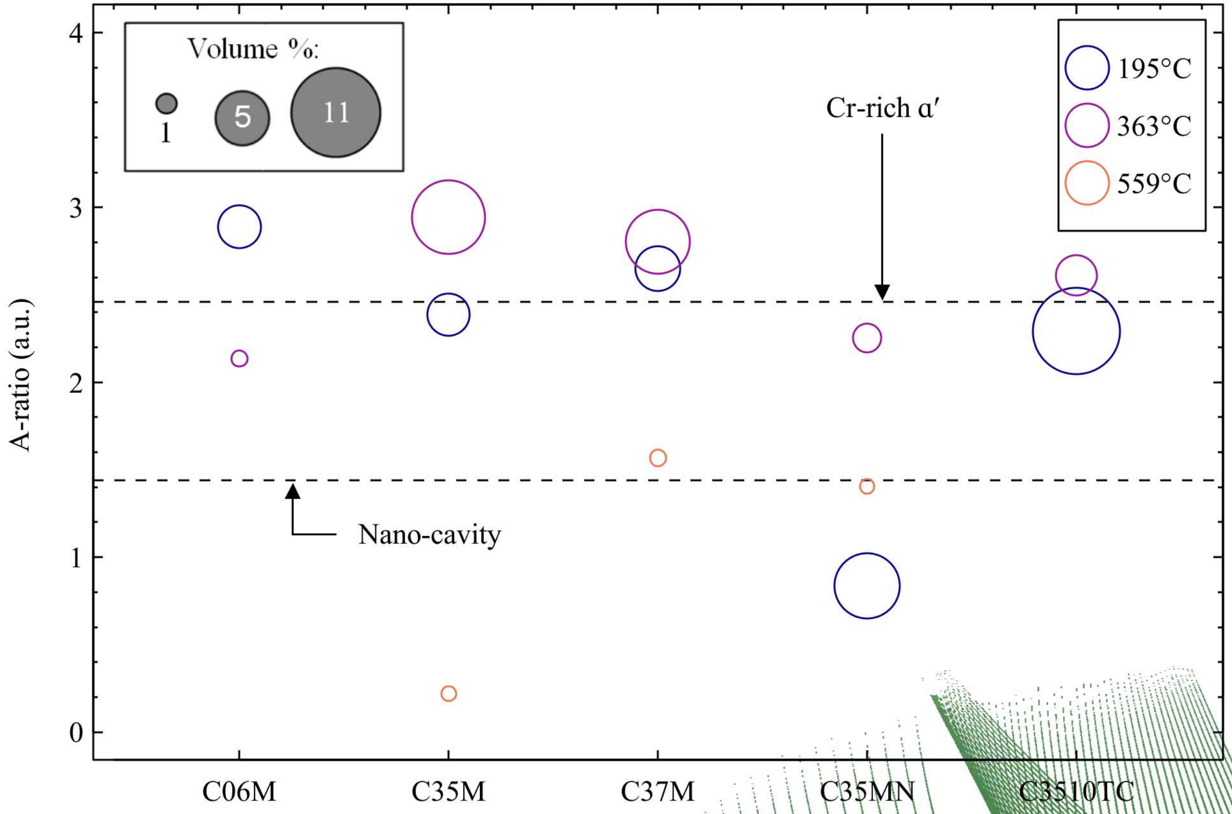
Magnetic Shielded-SANS measurements: Role of temperature and composition on α'





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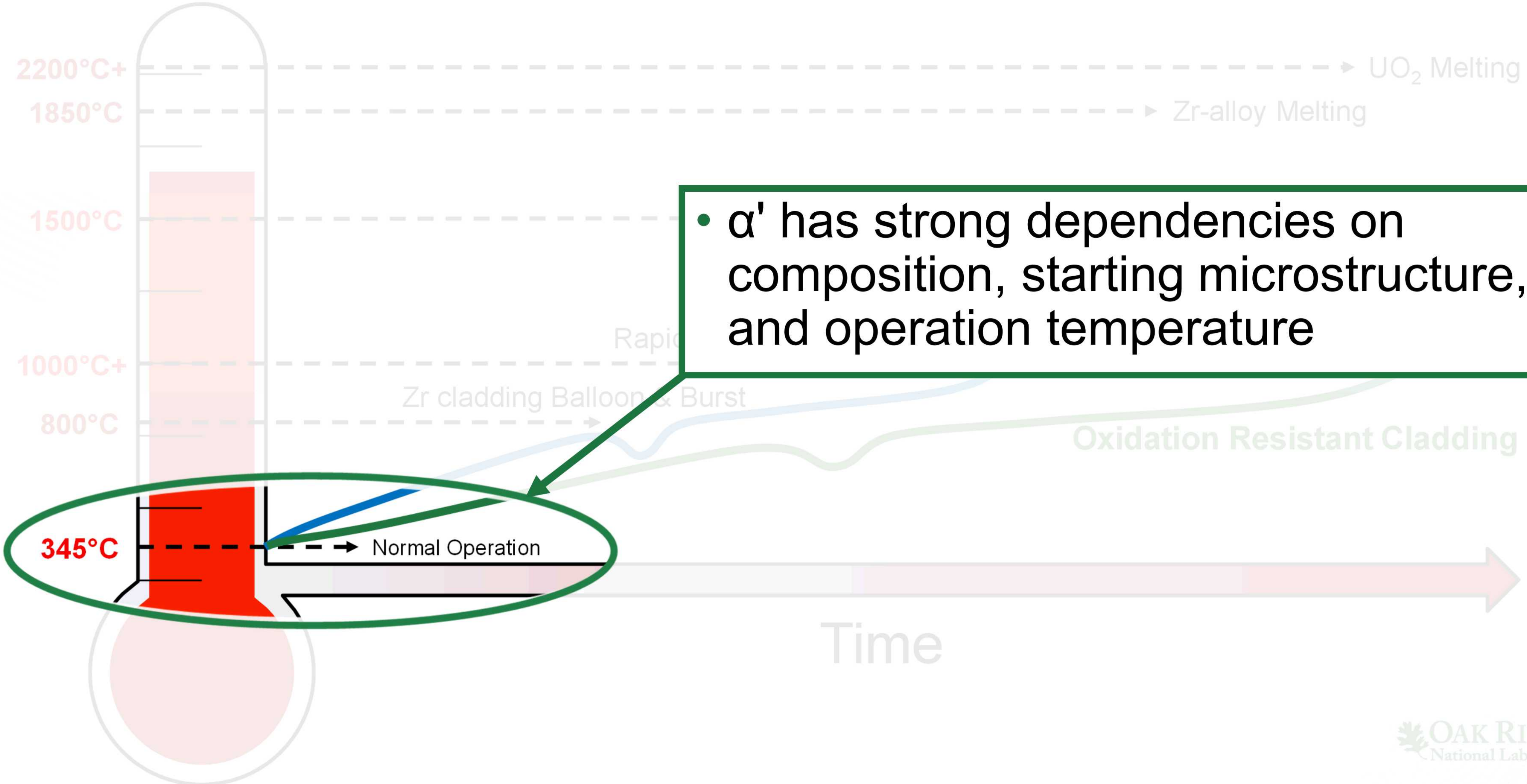
Practical Application of Post-Irradiation Annealing: α' not a concern for elevated temperature accidents



- α' has strong dependencies on composition, starting microstructure, and operation temperature

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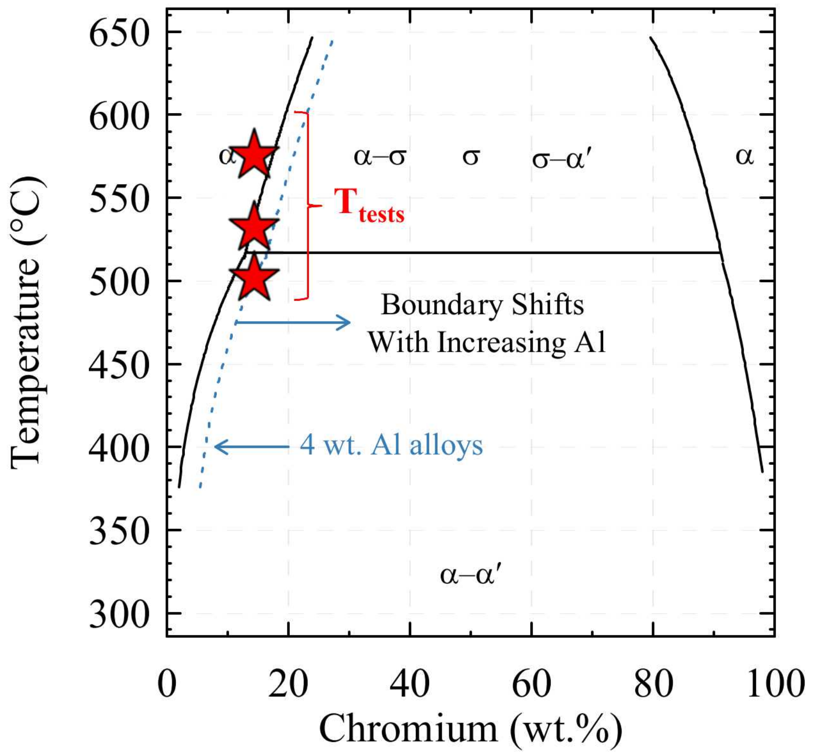
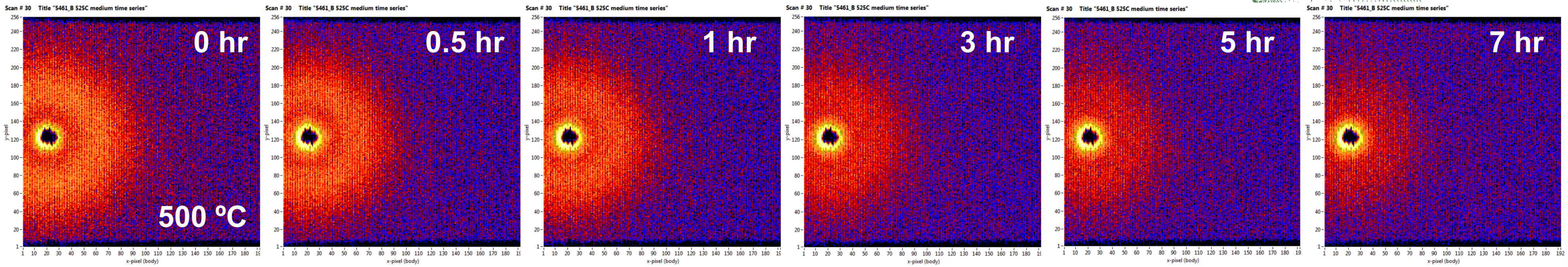
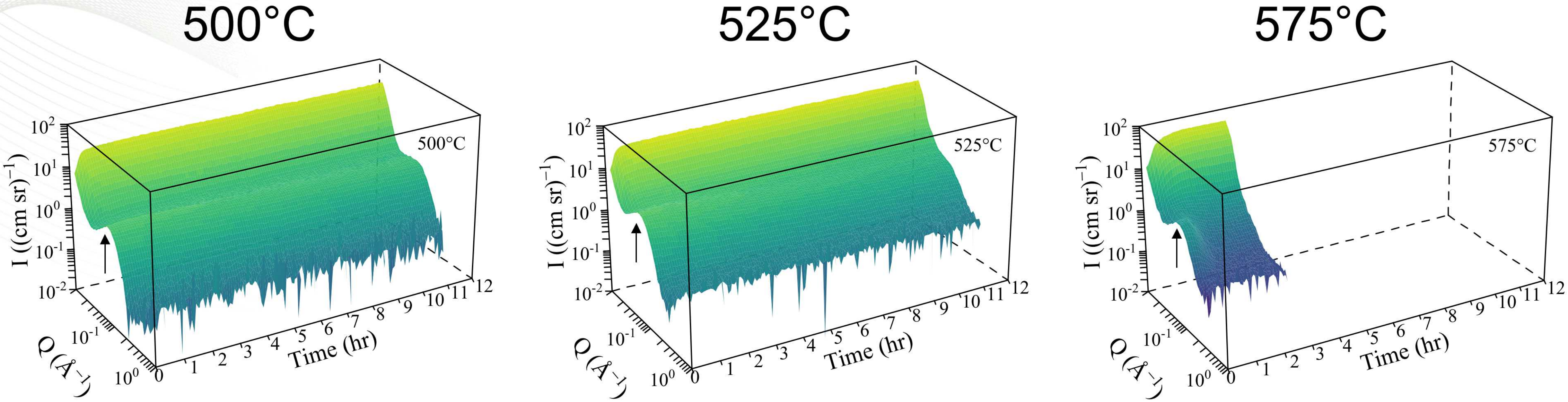
Time resolved SANS measurements: Post Irradiation Annealing of α'

- Measurements performed at CG-2 general purpose SANS beamline at HFIR
- Data collected at 5 min. intervals on broken tensile heads of Fe-15Cr-3.9Al (wt.%) irradiated to 1.8 dpa at 382°C
- Annealing performed at 500°C, 525°C, and 575°C





Time resolved SANS measurements: Post Irradiation Annealing of α'



Increasing Temperature

- Collection of SANS curves show clear time-temperature dependence on the "hump" which is indicative of α' precipitation

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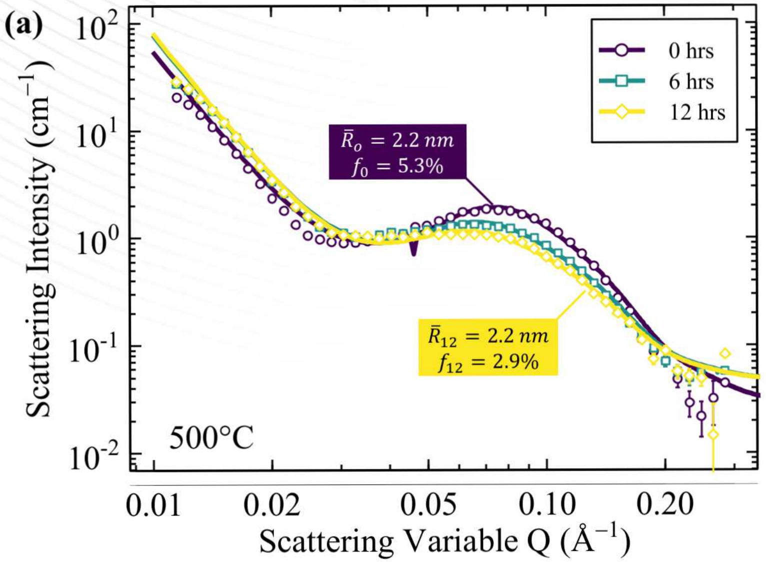
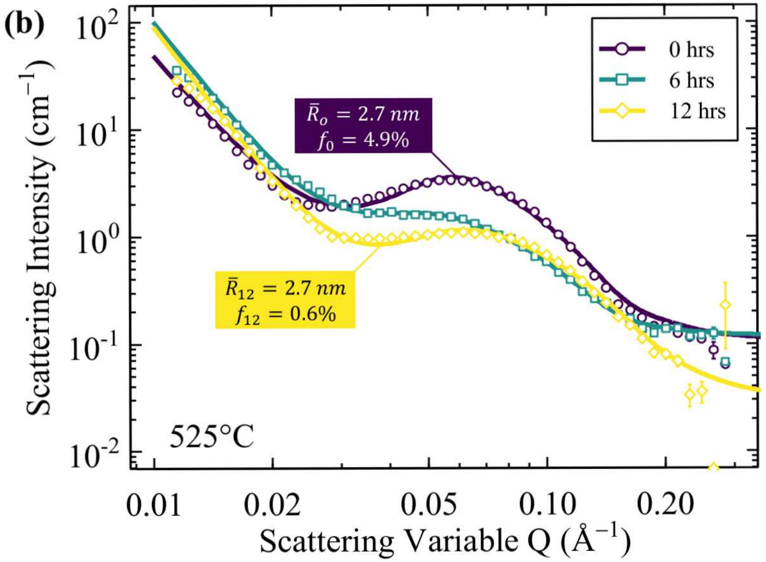
Time resolved SANS measurements: Post Irradiation Annealing of α'

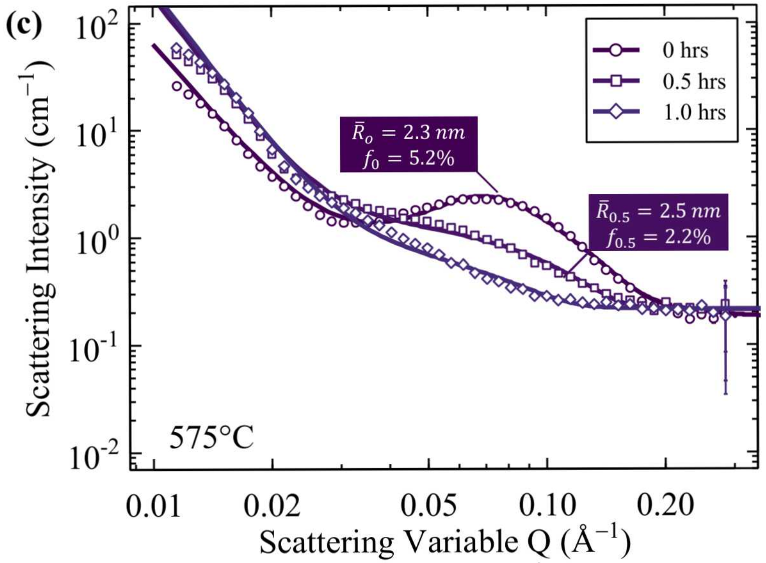
500°C



525°C



575°C



Increasing Temperature

- Local monodispersed approximation (LMA) shows good curve following with experiment over varying time-temperatures

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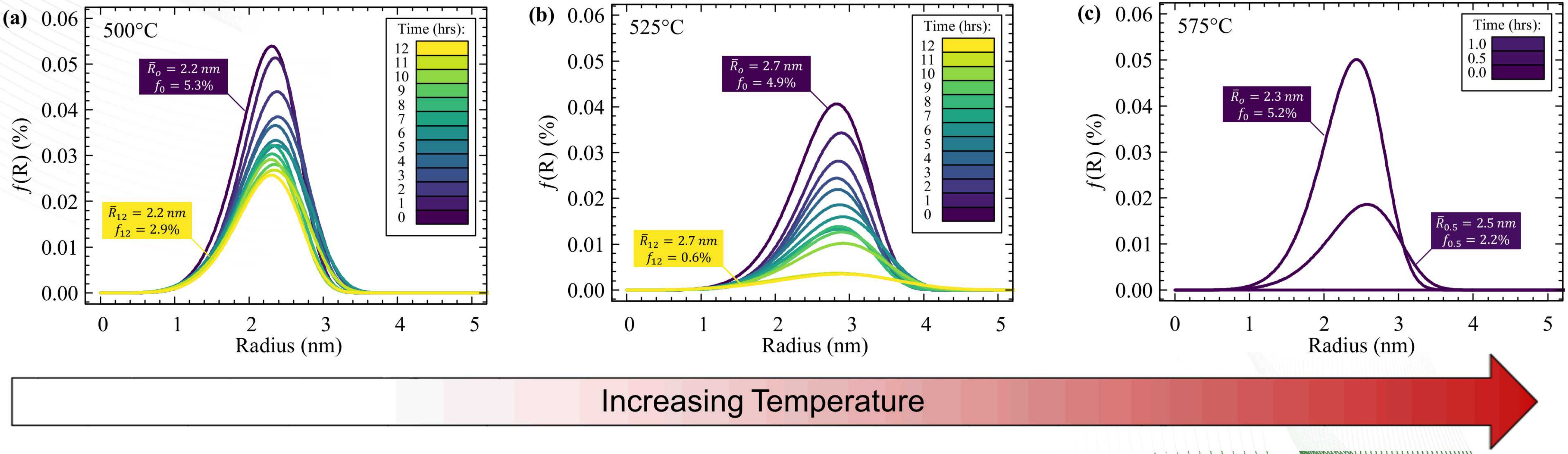
Time resolved SANS measurements: Post Irradiation Annealing of α'

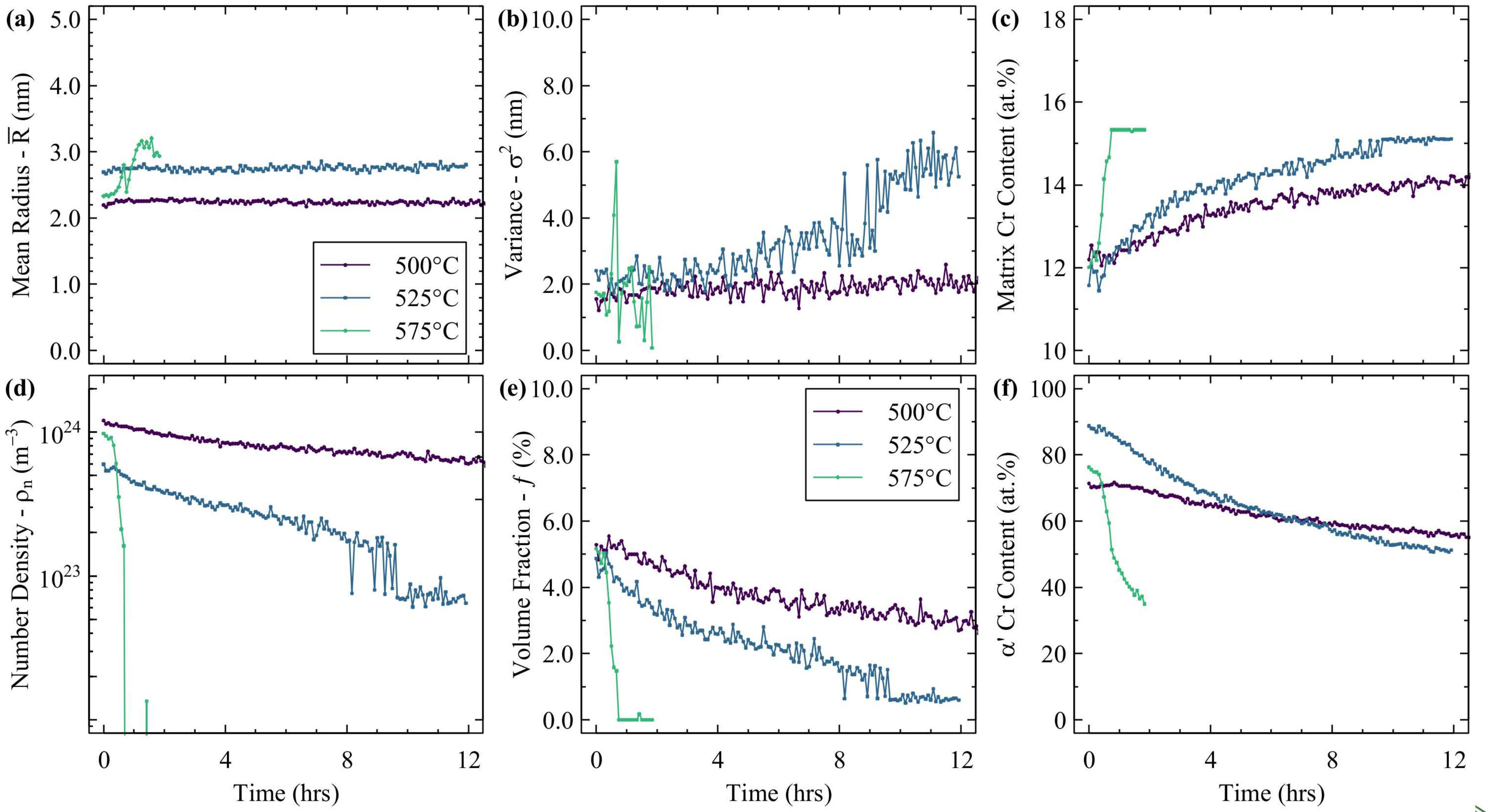
500°C 525°C 575°C



- Mean size varies little during post-irradiation annealing; volume fraction and width of size distribution strongly altered

Time resolved SANS measurements: Post Irradiation Annealing of α'

- **500°C:**
  - Minor dissolution after 12 hrs
- **525°C:**
  - Nearly complete dissolution after 12 hrs
- **575°C:**
  - Complete dissolution after 1 hrs



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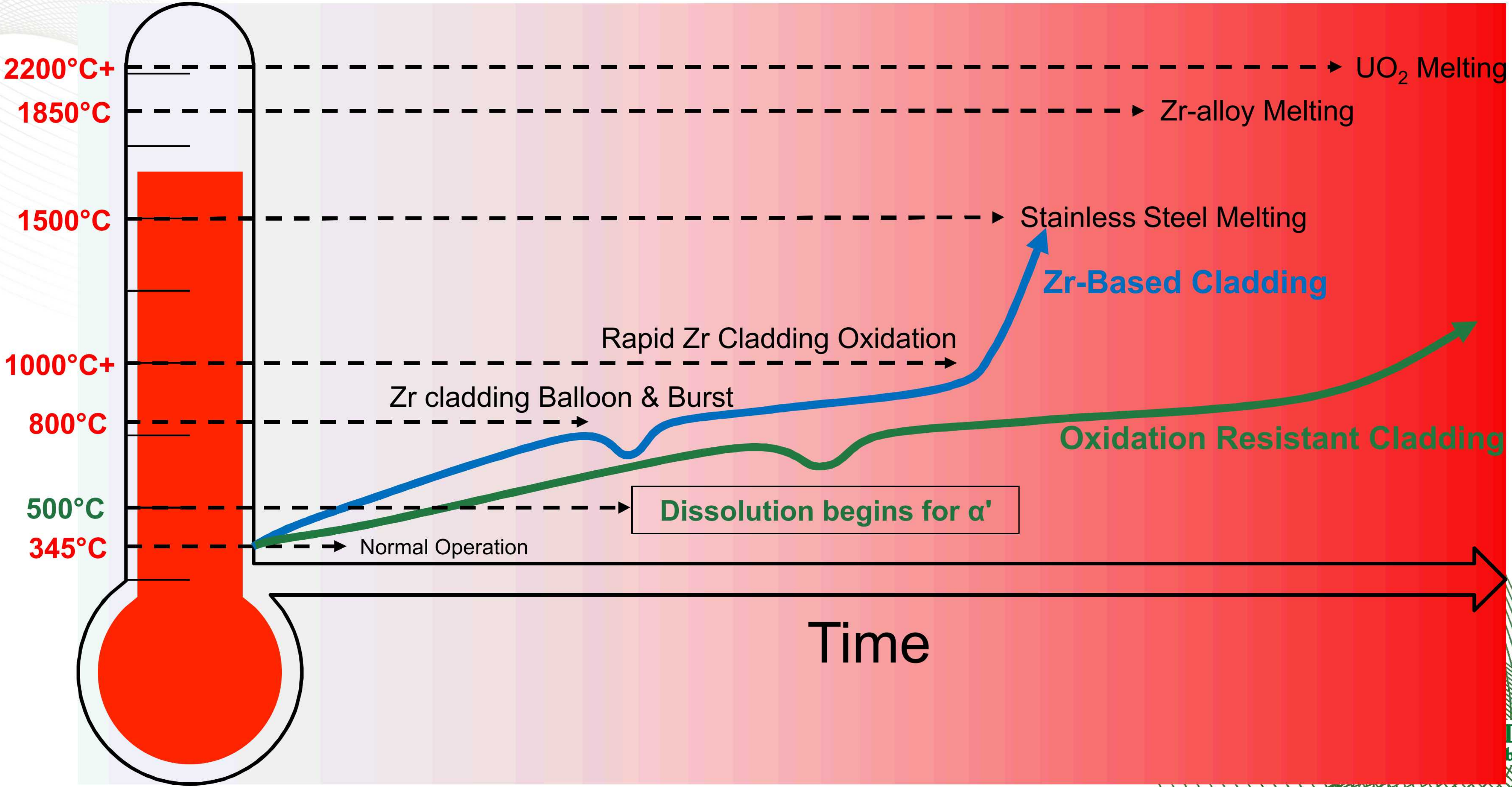
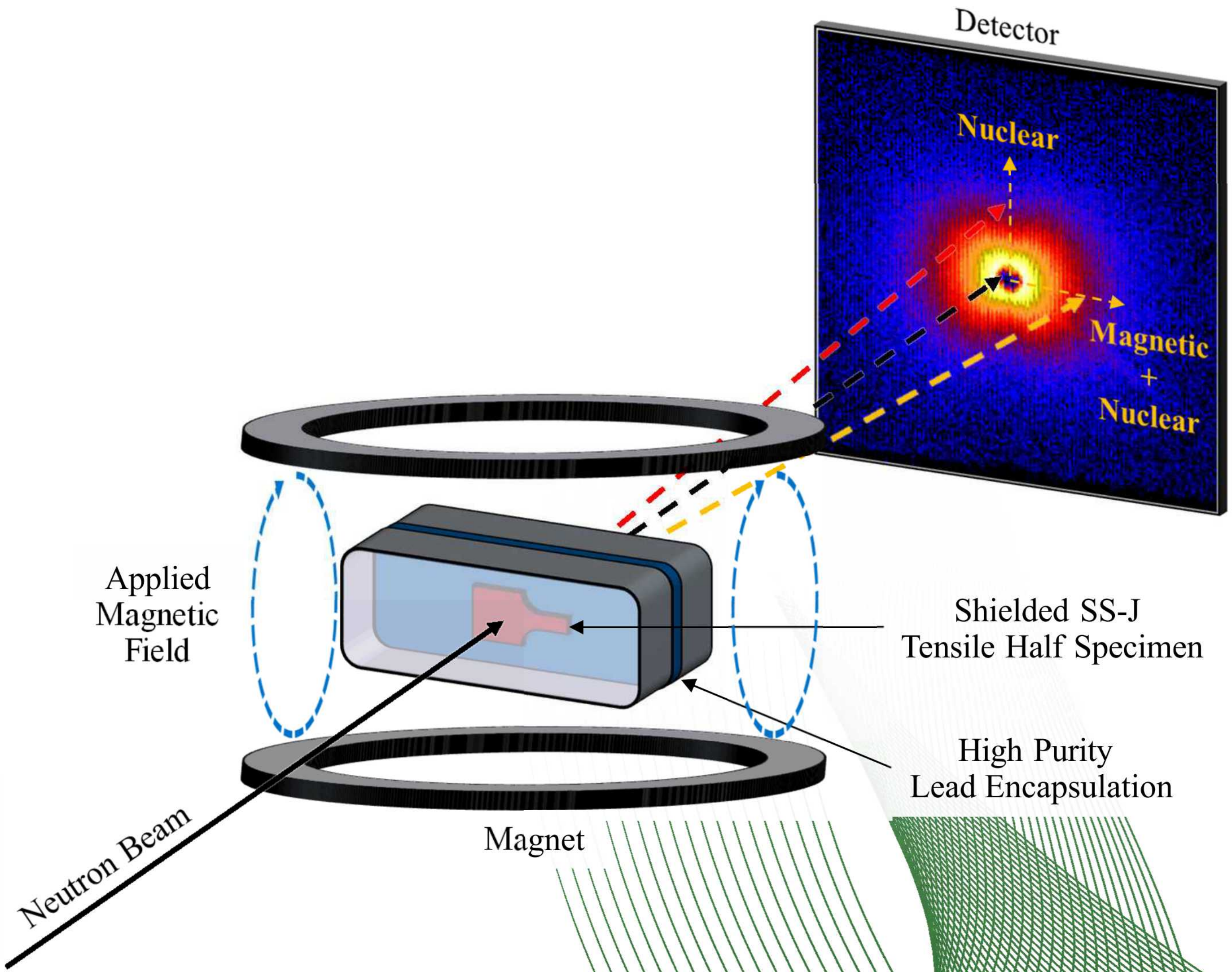


Summary

- FeCrAl alloys are considered for accident tolerance
- SANS is efficient means to detect and analyze the detrimental α' in FeCrAl alloys after irradiation
- Ex-situ SANS showed strong dependencies on a large number of factors
- In-situ SANS showed rapid dissolution of α' during accident scenarios



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Questions?

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Photo Courtesy of John Williams (GNF)