Effect of Irradiation Damage on Fission Product Transport: FY2017 Progress

AGR TRISO Fuels Program Review
July 18-19, 2017

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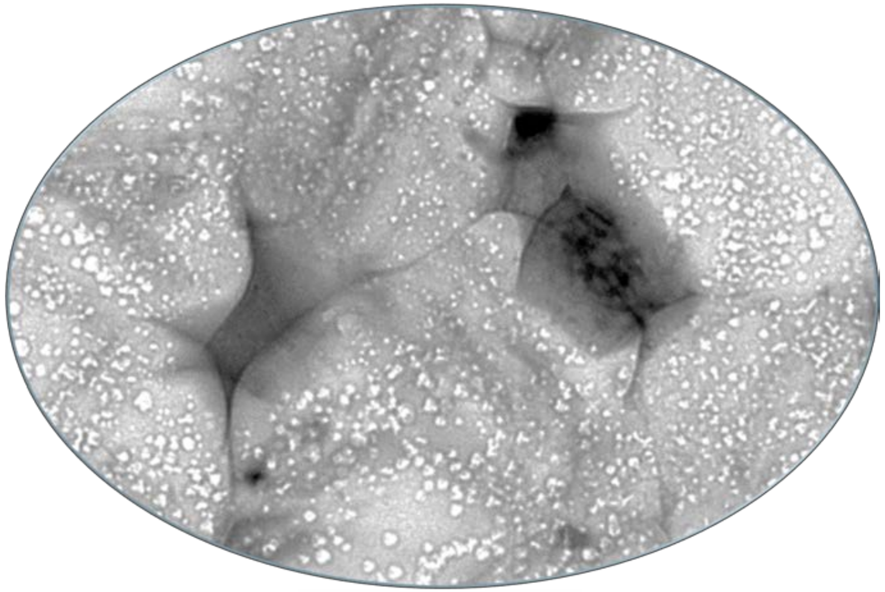
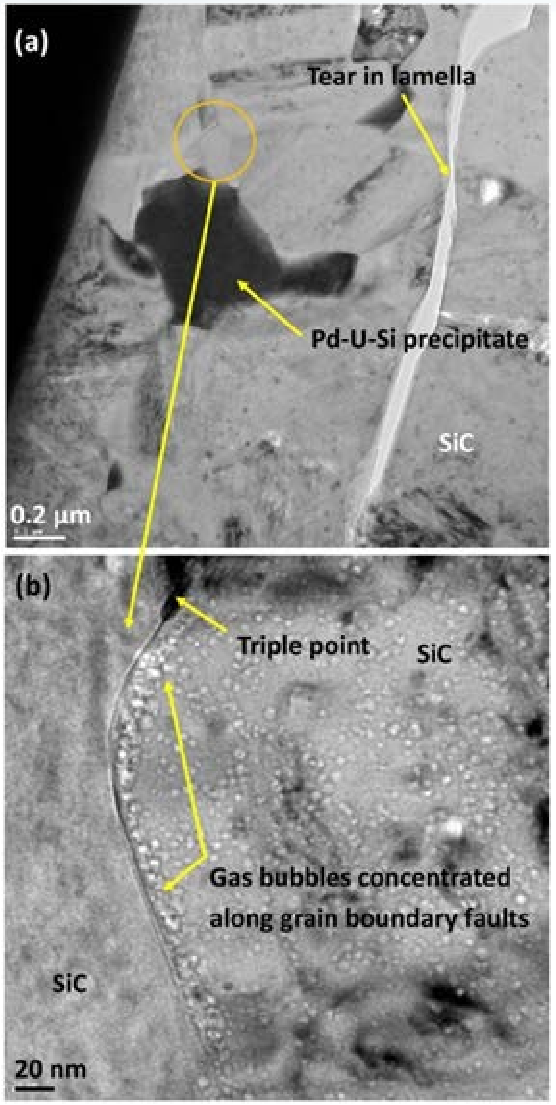


INL/MIS-17-42685

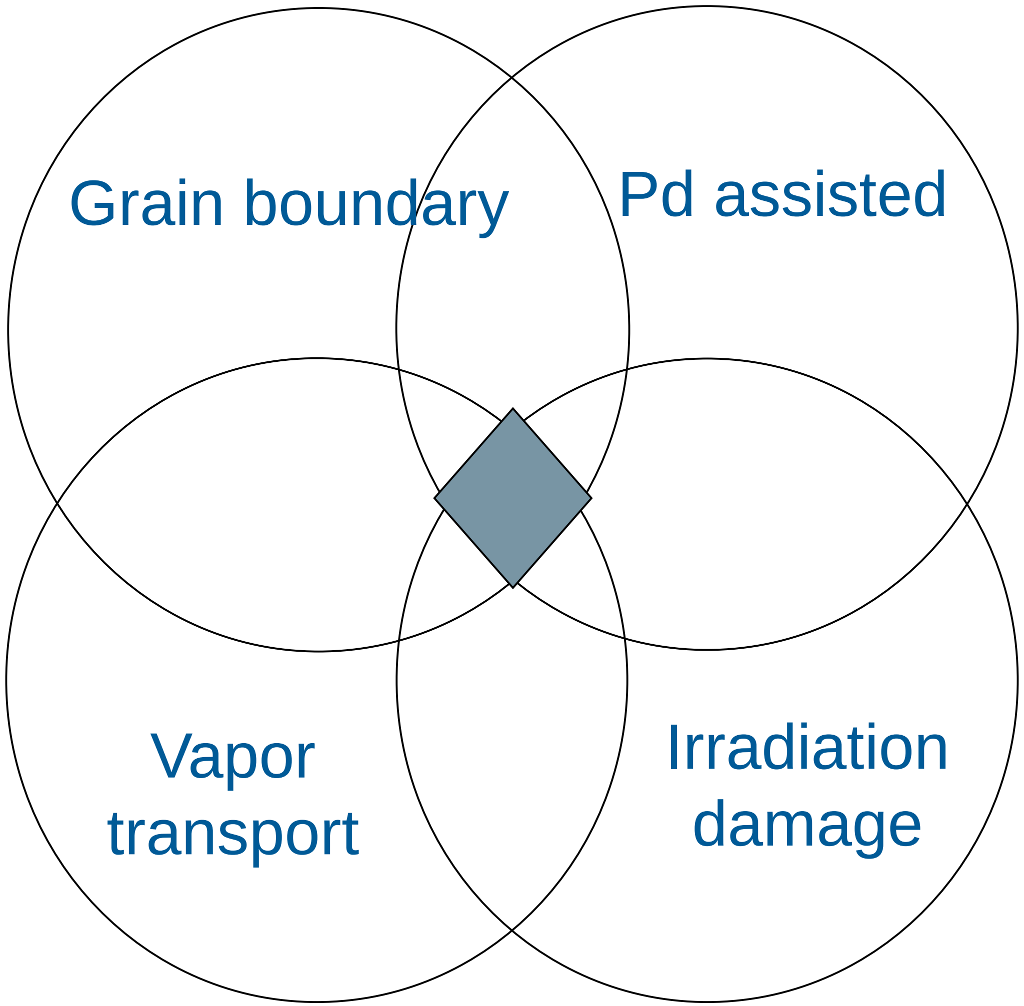
Outline

- Background and Objectives
- FY2017 work scope
- Neutron Damage: Defect density and irradiation temperature
- Neutron Induced Phase Transformations
- HRTEM structures of Particle AGR1-411-030
- Conclusions
- Recommendations: Future Work
- Acknowledgements



Background: Transport Mechanisms

- Complex interactive mechanism likely:
  - neutron damage,
  - grain boundary characteristics,
  - chemical interaction with Pd, and
  - vapor transport



[I. J. van Rooyen, H. Nabielek, J. H Neethling, M. Kania and D.A. Petti, PROGRESS IN SOLVING THE ELUSIVE AG TRANSPORT MECHANISM IN TRISO COATED PARTICLES: "WHAT IS NEW?" Paper 31261, Proceedings of the 2014 International HTR-2014 Conference of High Temperature Reactors, Weihai, China, 2014]

Neutron Damage (FY2017)

Work focus on neutron damage and its effects on:

- Fuel performance
- Fission product distribution
- Bulk and grain boundary fission product transport mechanisms.
- This work will narrow the gap in understanding the effect of neutron irradiation on fission product transport in the intact SiC layer of TRISO-coated particles.

Neutron Damage: Scope and Matrix

- Correlate neutron-induced microstructural
  - defect density,
  - volume fraction, and
  - morphology with neutron irradiation parameters (i.e., neutron fluence and temperature)
- Analyze the defect density and distribution in the vicinity of fission product precipitates

Particle	Ag retention	Fuel Type	Burnup (%FIMA)
AGR1-632-034	65%	Baseline	11.4
AGR1-523-SP01	16%	Variant 1	17.4
AGR1-131-066	39%	Variant 3	15.3
AGR1-433-001 Safety tested	66%	Variant 3	18.6
AGR2-223-R06	8%		10.8
AGR2-222-RS036 Safety tested	80%		
AGR2-633-TBD ★			7.5
AGR1-411-030 ★	90%	Variant 3	19.4

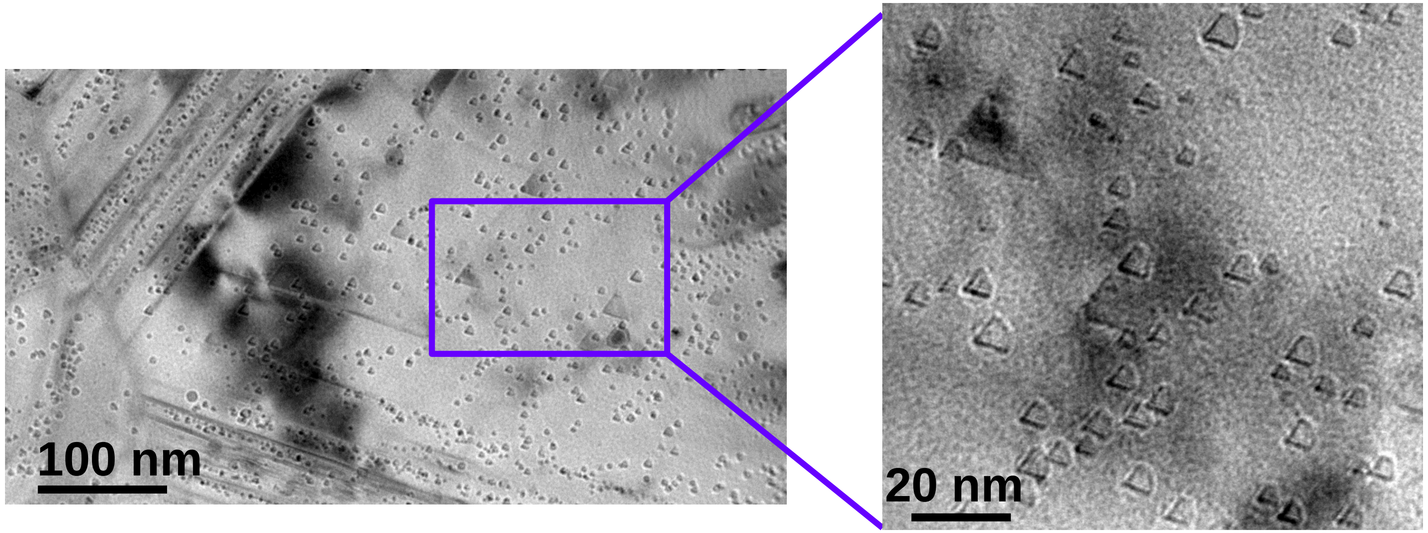
★ Future work

1 2 3 4

Neutron Damage: Defect Density and Irradiation Temperature

Neutron Damage: Defect density and irradiation temperature

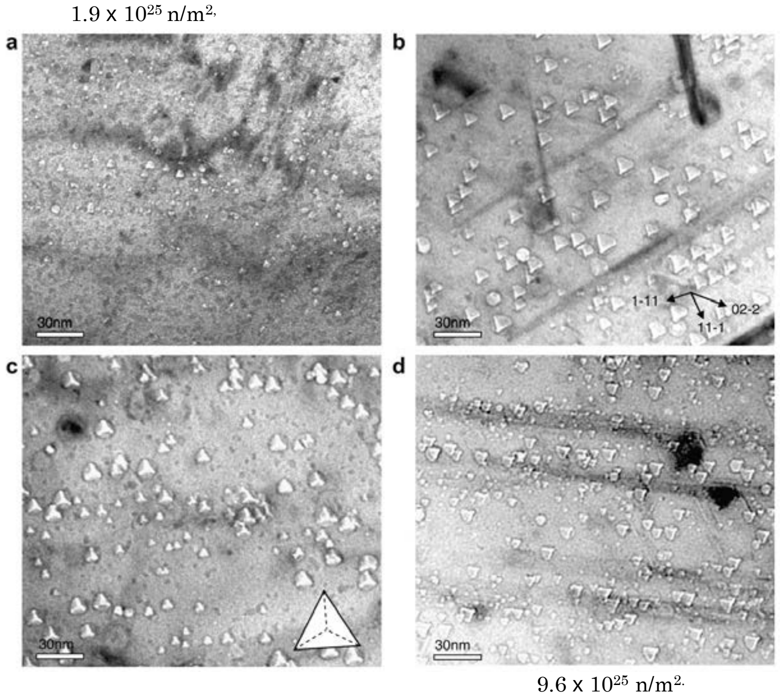
** Volume fraction and densities of **voids** can give insights to the irradiation temperature*



Neutron irradiation induced voids are non-uniformly distributed. Voids are aligned along stacking faults.

Voids have tetragonal shape

Effect of fluence on size and density



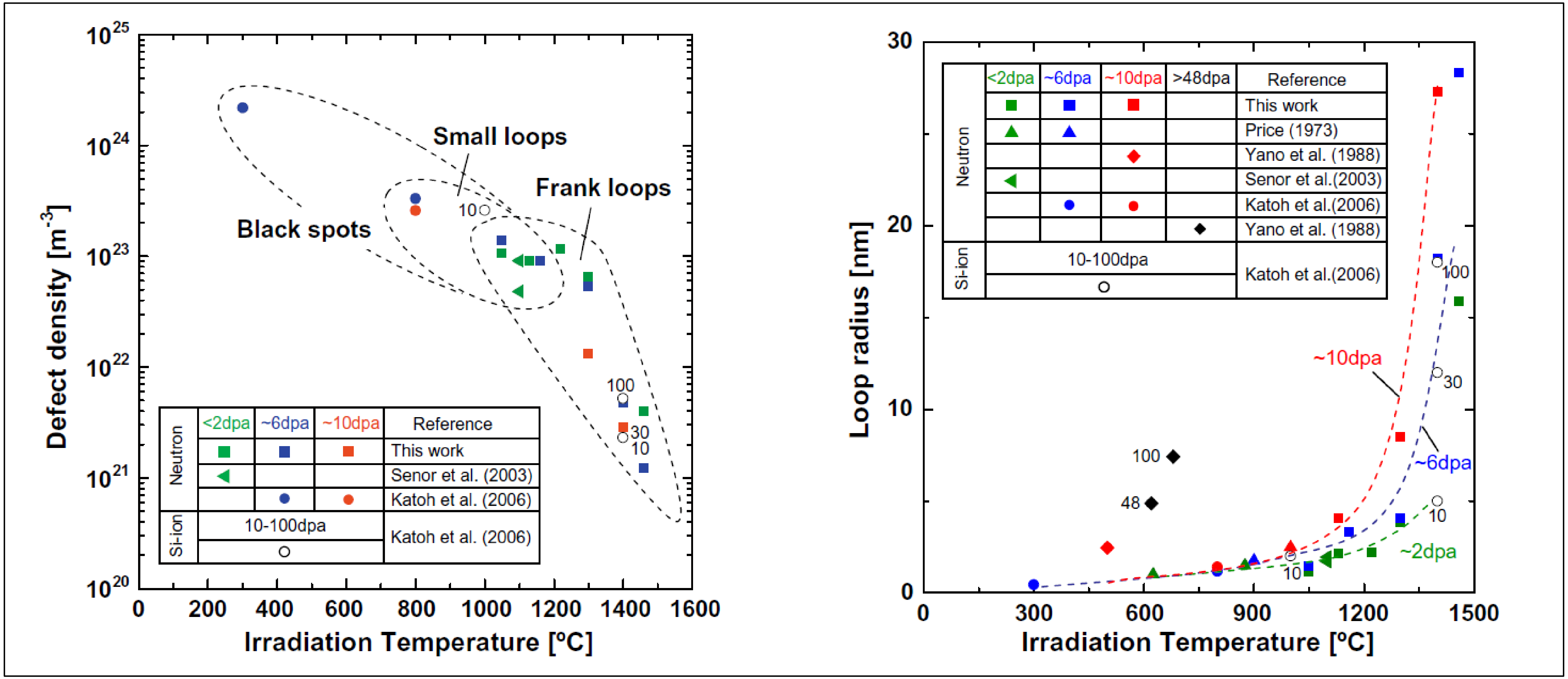
[S. Kondo, Y. Katoh, and L. L. Snead, Unidirectional formation of tetrahedral voids in irradiated silicon carbide, Appl. Phys. Lett. 93, 163110 (2008)]

S Kondo et al, JNM 382 (208) 160 -169]

AGR2-223-RS06, Lamella 9 (SiC/OPyC)

Estimated Temperature > 1400 °C

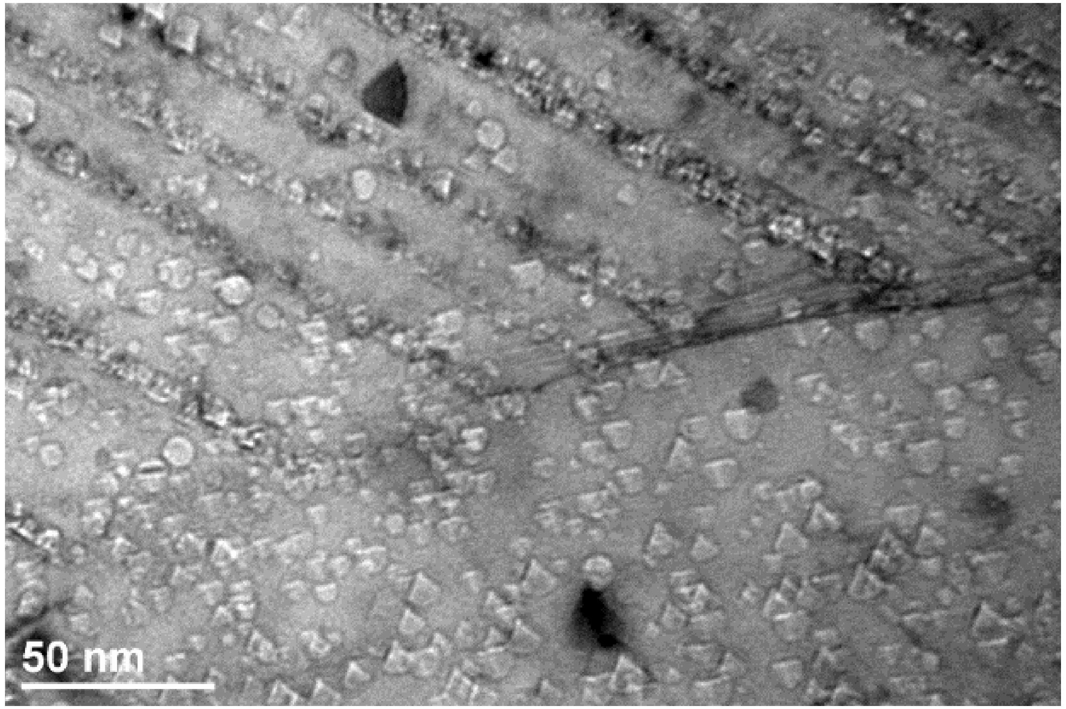
Neutron Damage: Defect density and irradiation temperature



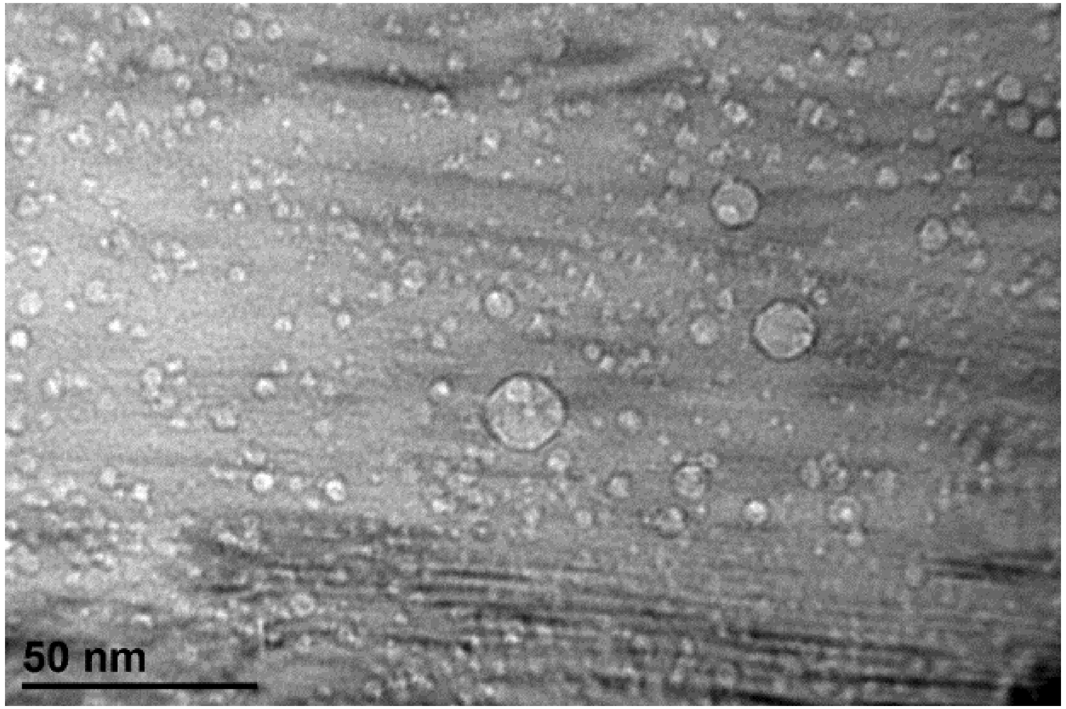
Below 800 °C: Black spots dominant defects
800 -1150 °C : some black spots, small loops, Frank Loops
1300-1460 °C : irradiation fluence affects defect density and size, larger Frank loops (>20 nm in radius).

[S. Kondo, Y. Katoh, and L. L. Snead, *J. Nuc. Matls.* (2008) 382: 160-169].

Defect Size, Morphology and Concentration differences??



Inner SiC



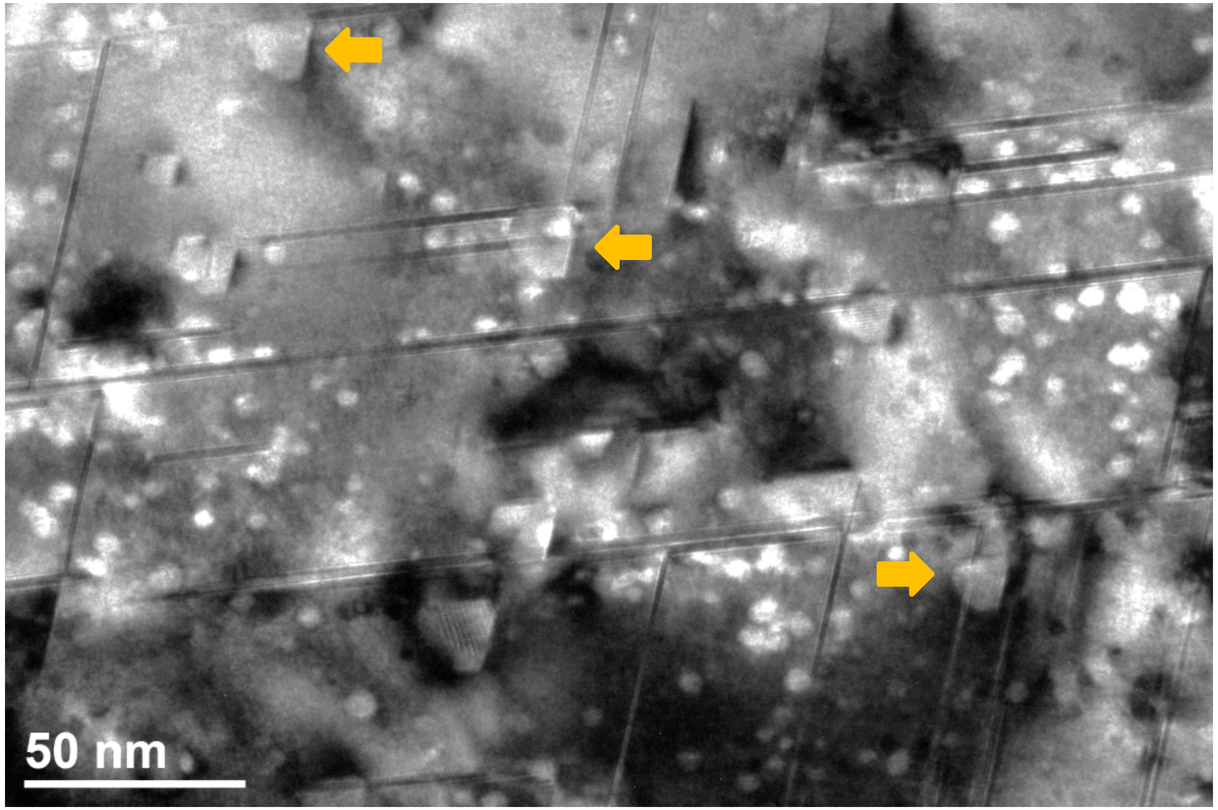
Outer SiC

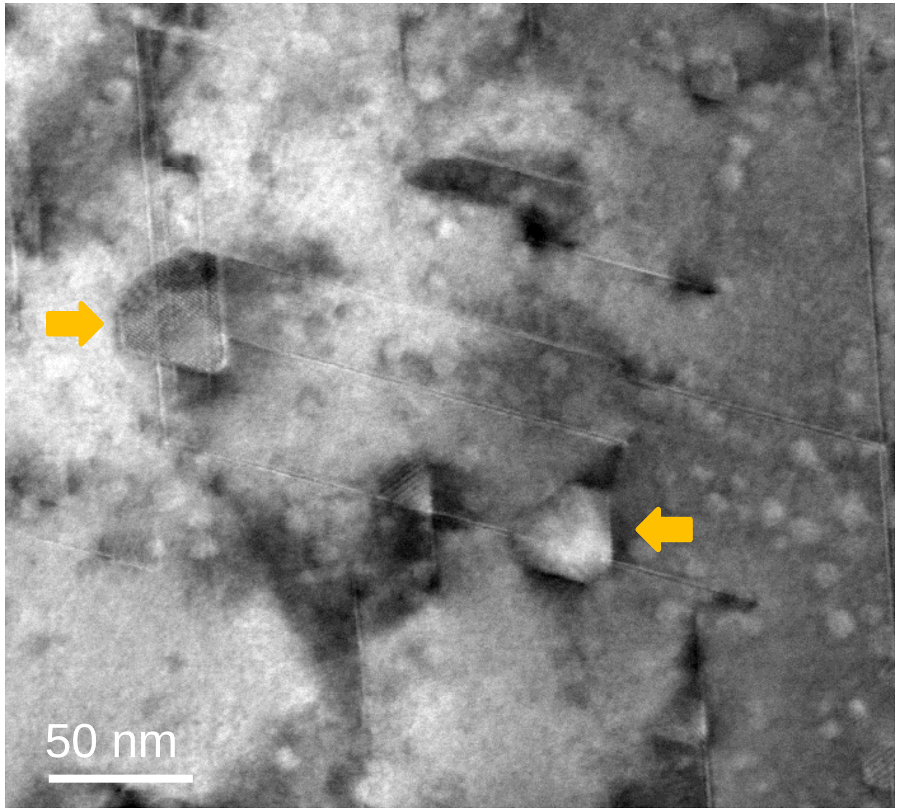
Particle AGR2-223-RS06

- Voids spherical: < 1300 °C
- Voids tetrahedral: > 1400 °C
- Preferential formations of cavities at stacking faults were confirmed above 1300 °C
- Small cavities were dispersed with low number density at 1130 °C

[S Kondo et al, JNM 382 (208) 160 -169]

Distribution of voids around nanoscale precipitates

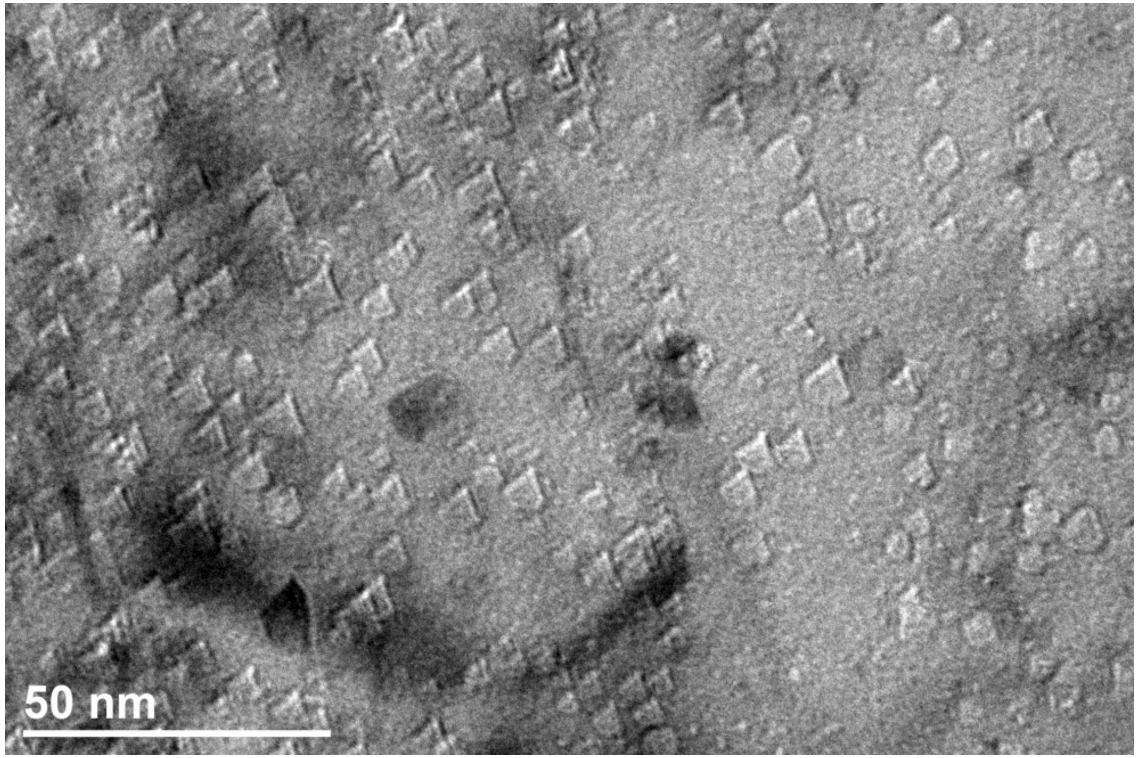


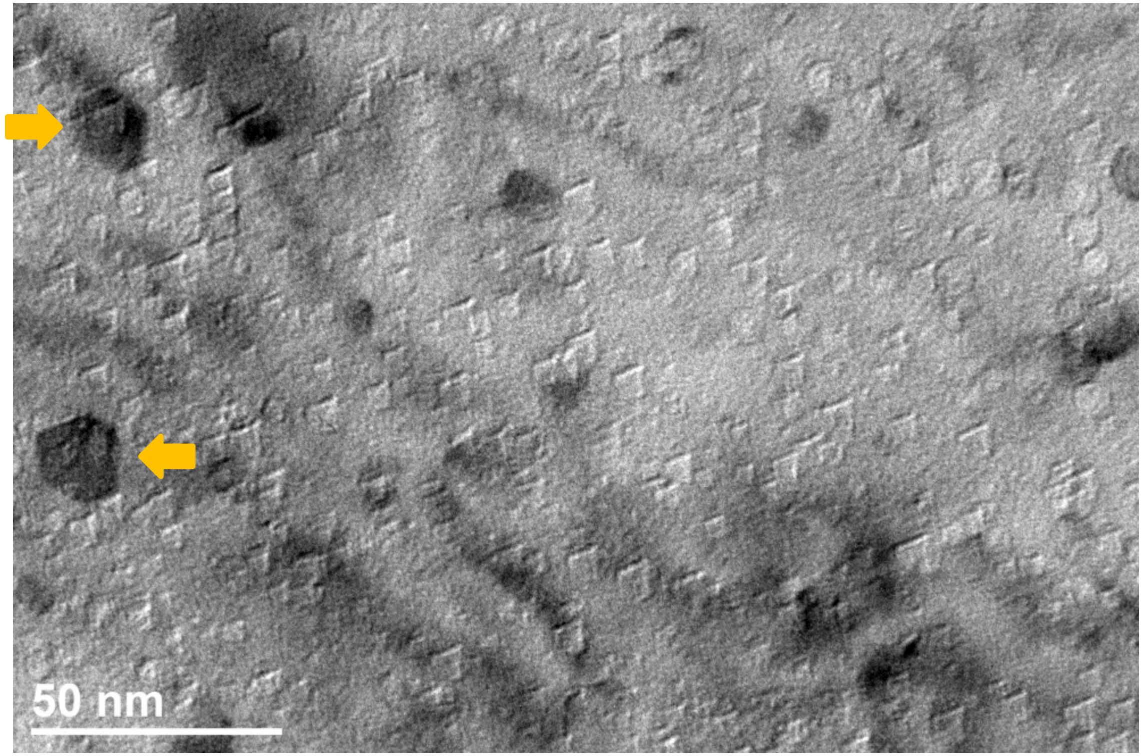


Yellow arrows indicates α-SiC or Pd precipitates

AGR1-632-034

Tetragonal shaped voids around nanoscale precipitates



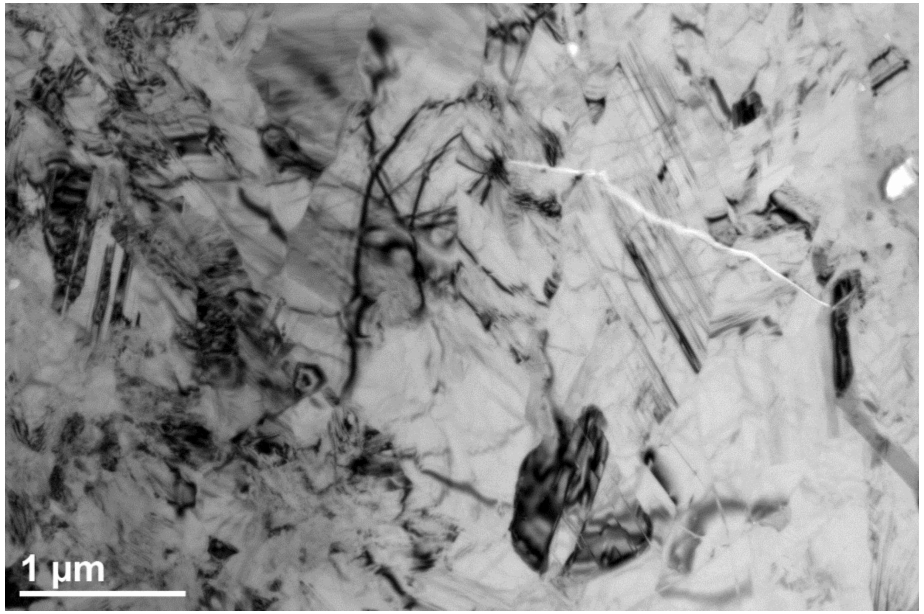


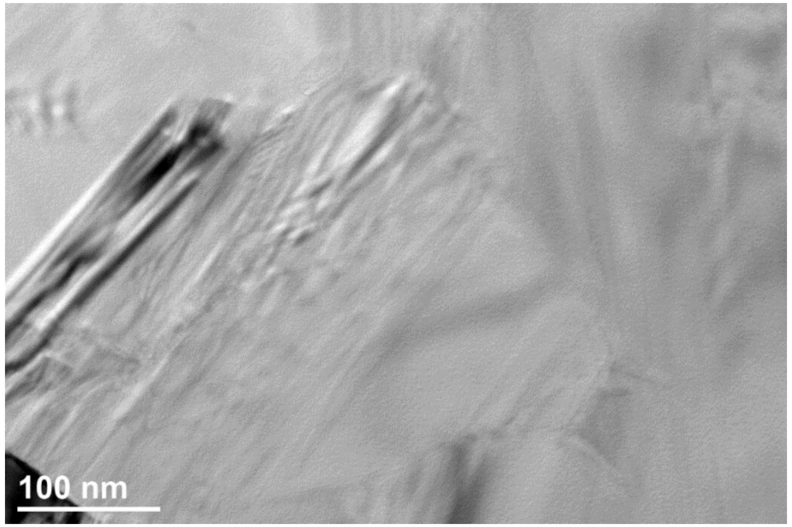
Yellow arrows indicates α-SiC or Pd precipitates

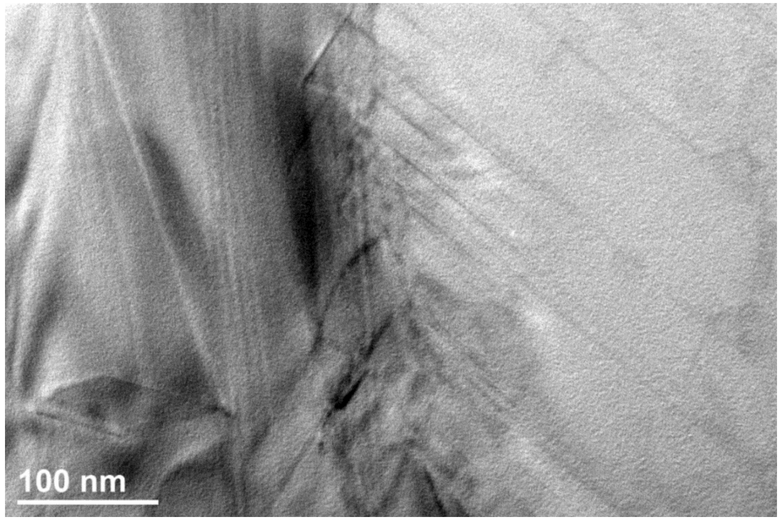
AGR1-523-SP01

Neutron Induced Phase Transformations

Unirradiated SiC (Variant 3 Fuel Compact T0650)

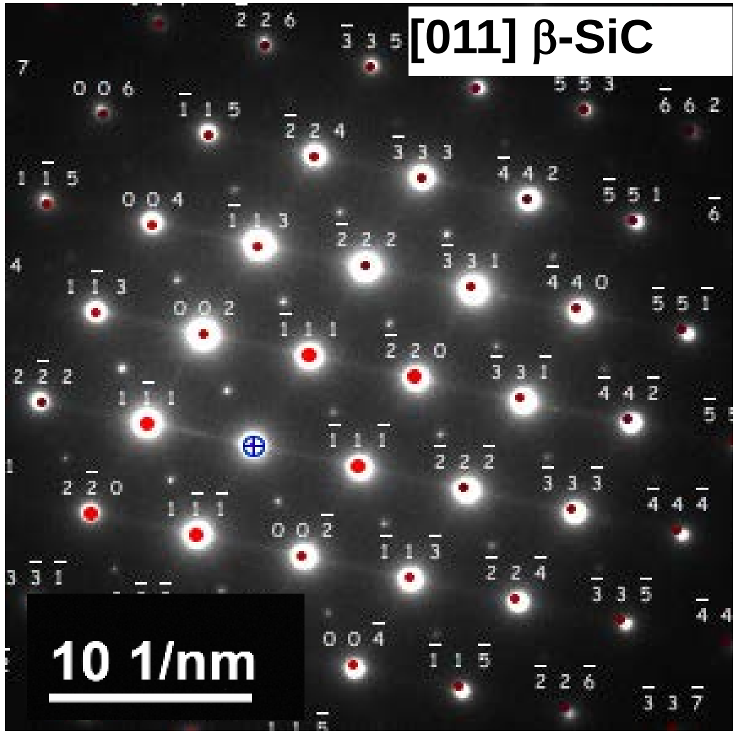




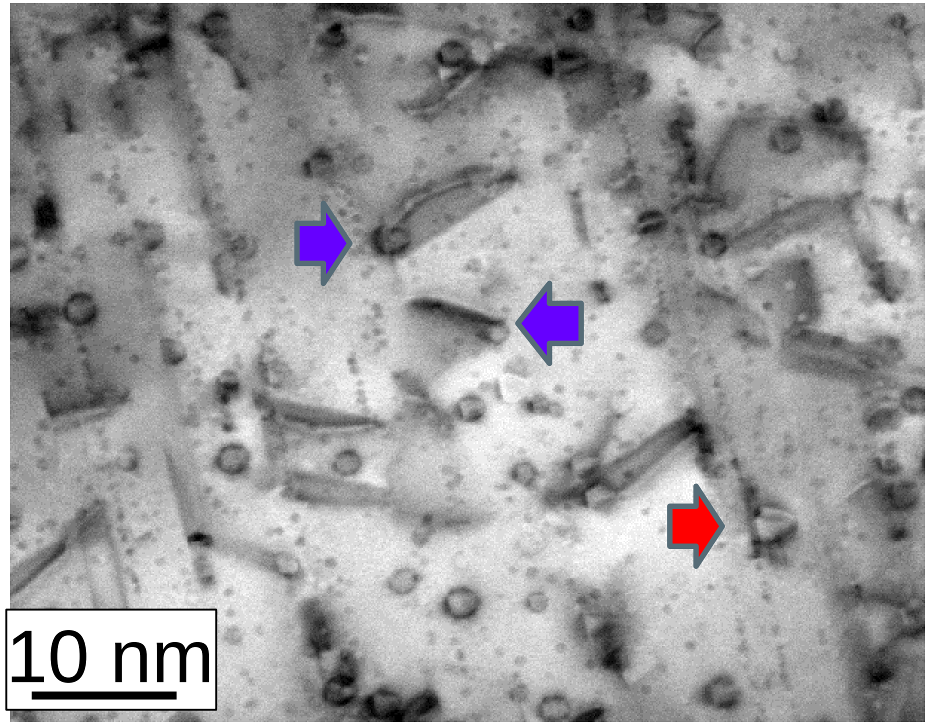


- β-SiC
- Stacking Faults
- No α-SiC region
- No apparent frank loops

Neutron Induced Phase Transformation of SiC



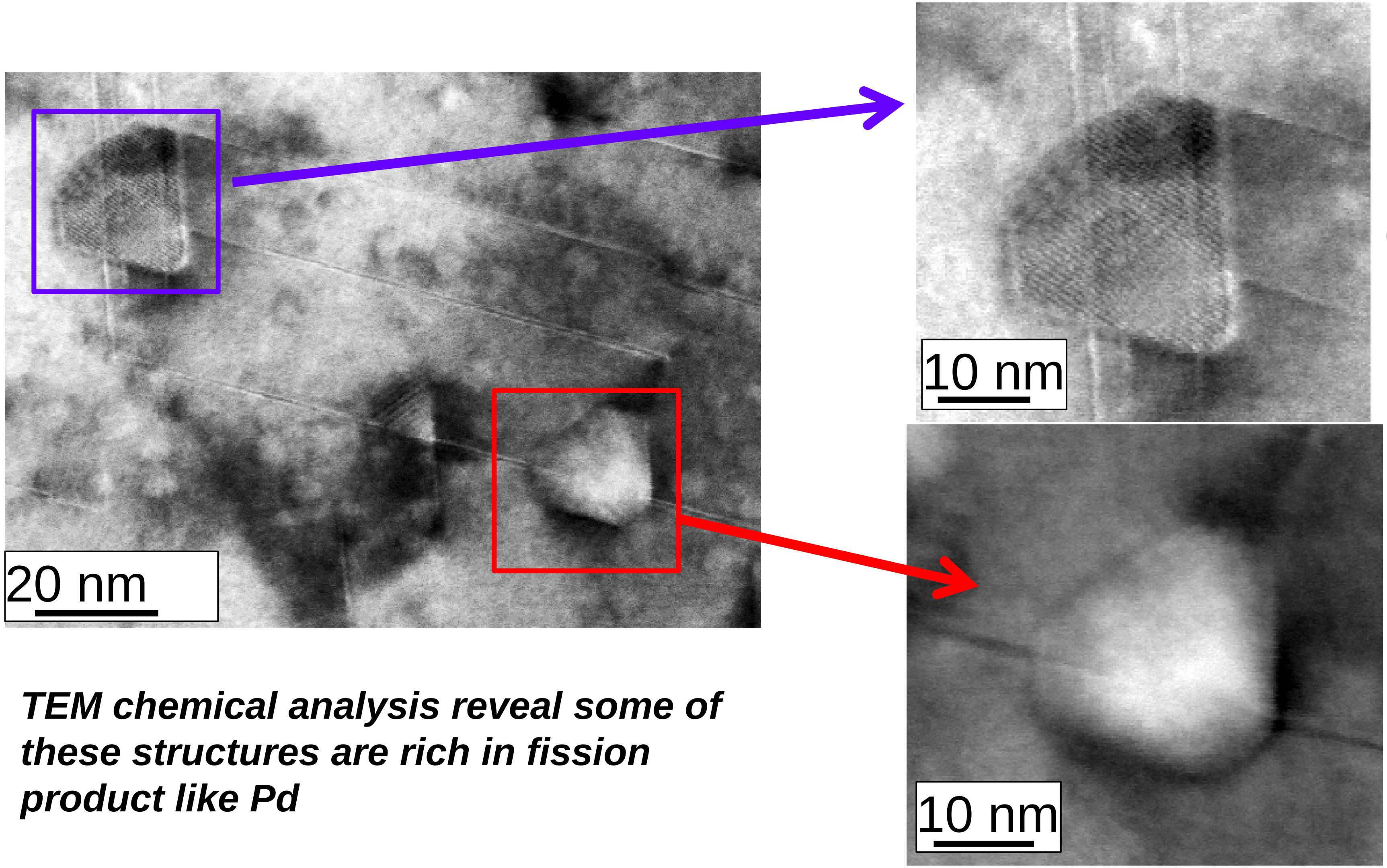
For the consistency of the study, all of the TEM were carried out along <001> zone axis of b-SiC matrix



Blue arrows: structures at the end of Frank loops along {111} planes
Red arrow: structure with one of its edge at a stacking fault

Frank loop: *are linear defects, introduced due to neutron irradiation here*
Stacking fault: *are planar defects, often present in close packed materials such as SiC*

Neutron Induced Phase Transformation of SiC



Only Si and C

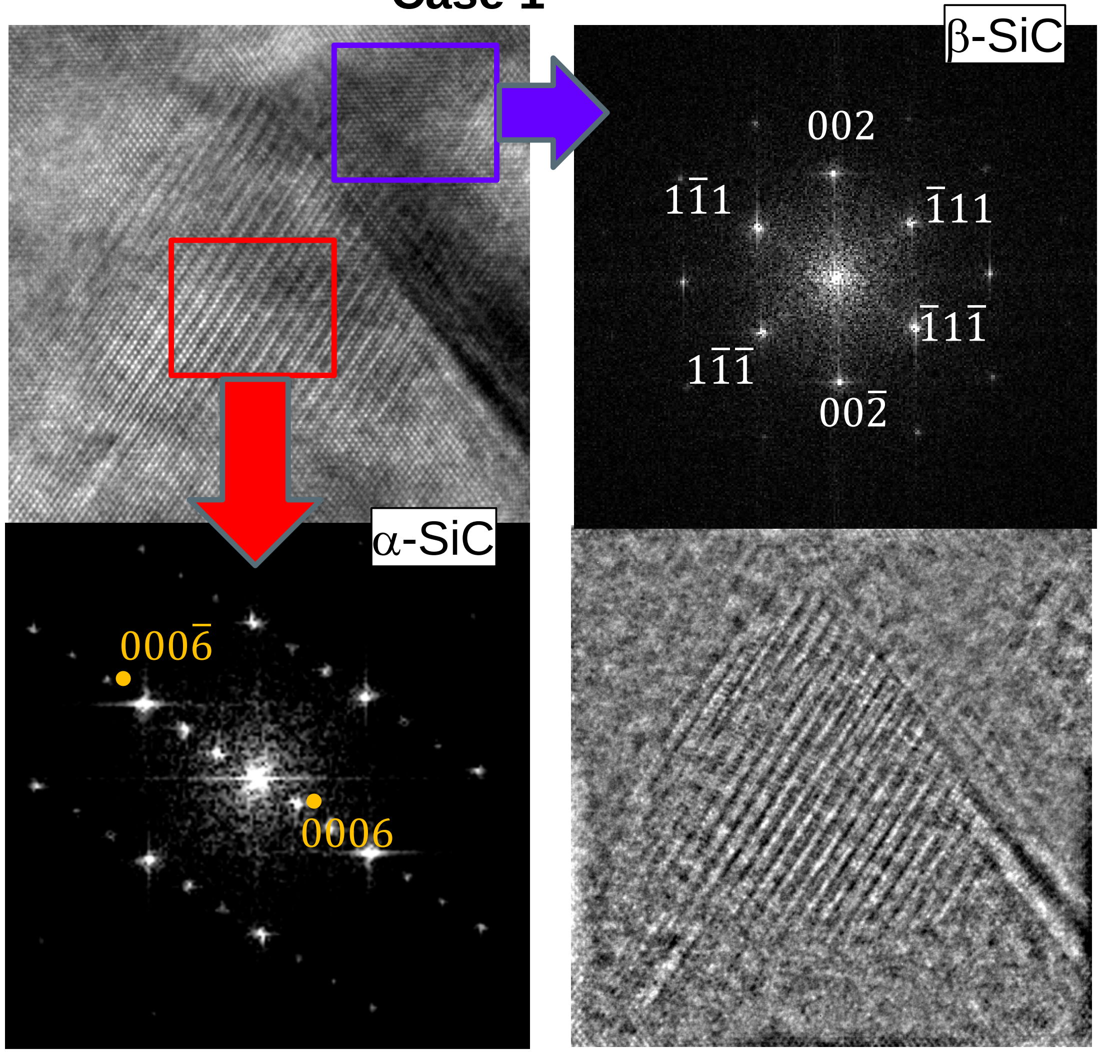
Si, C and **Pd**

TEM chemical analysis reveal some of these structures are rich in fission product like Pd

Neutron Induced Phase Transformation of SiC

High Resolution (HR)TEM: for structural analysis

Case 1



$\{111\}_{\beta}$ $||\{0006\}_{\alpha}$

α-SiC precipitate appear to nucleate at the edge of a Frank loop

Some of the structures were confirmed to be irradiation induced low temperature ***α-SiC***

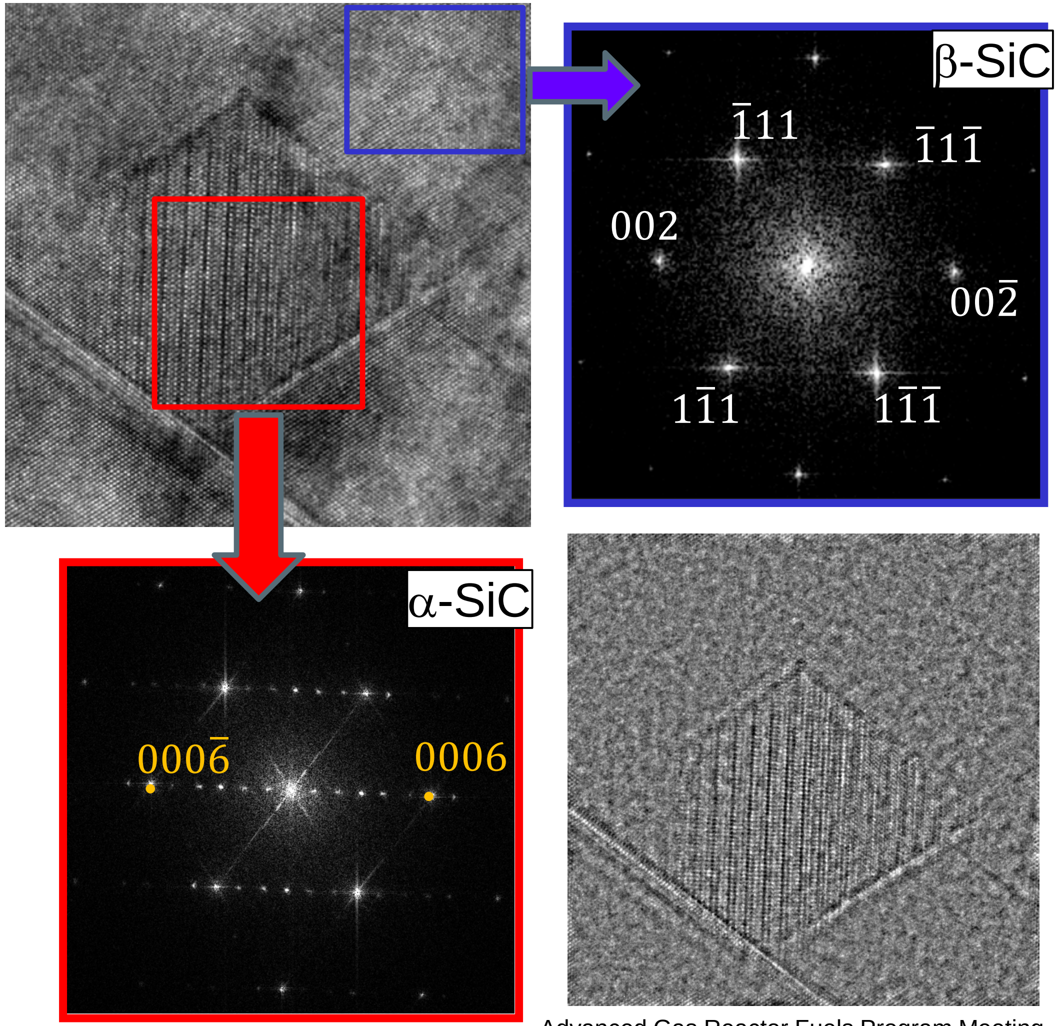
Also described by: [Chad M. Parish, Takaaki Koyanagi, Sosuke Kondo &Yutai Katoh, Irradiation-induced β to α SiC transformation at low temperature]

[Tan TY et al On the diamond cubic to hexagonal phase transformation in SiC Phil. Mag. A 44: 127-140, 1981

Ute Kaiser, Nanocrystal formation in hexagonal SiC after Ge+ ion implantation, Journal of electron microscopy 50(3): 251-263 (2001)

Neutron Induced Phase Transformation of SiC

Case 2



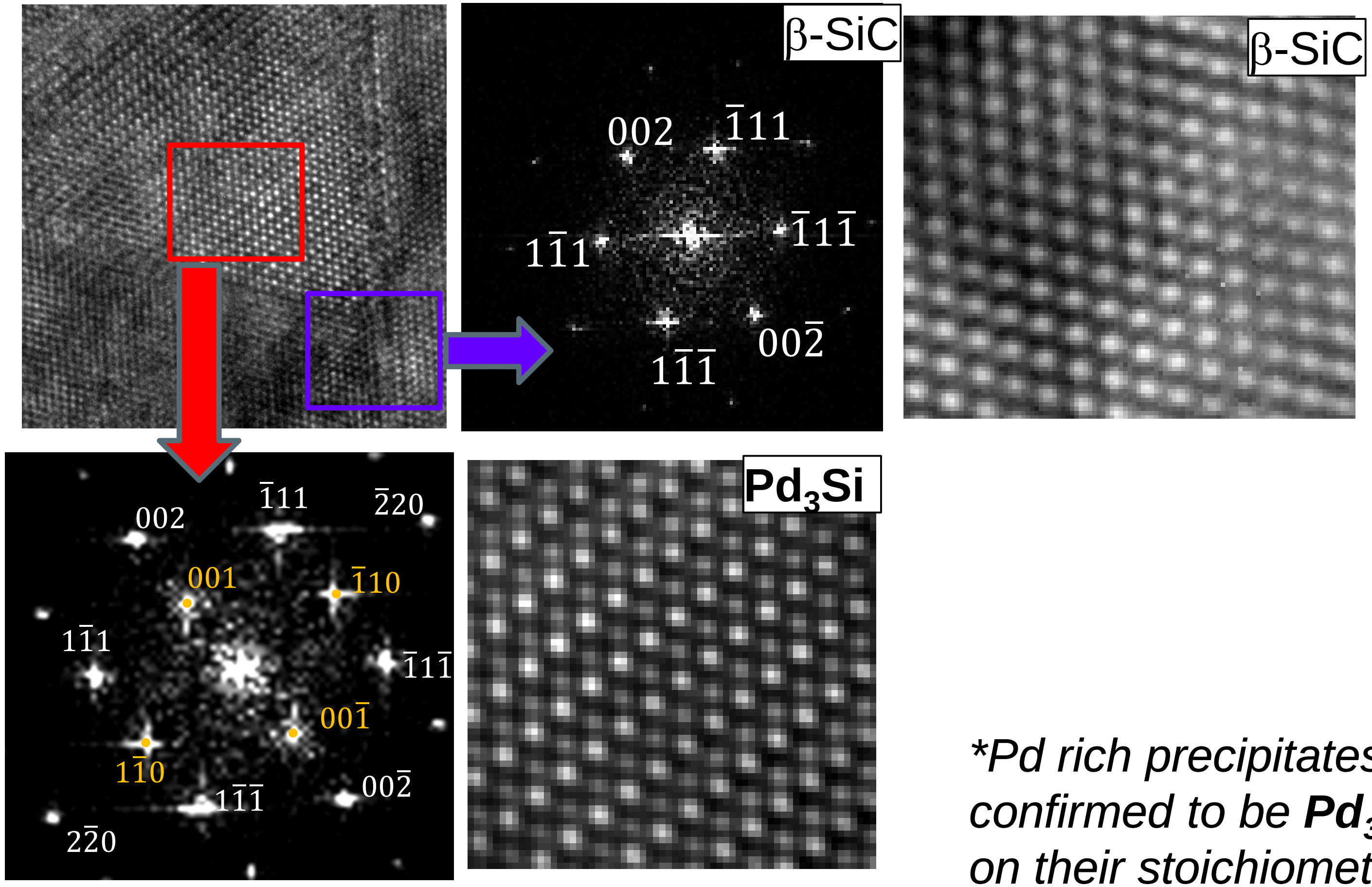
Surprisingly,
$\{002\}_{\beta}$ $||\{0006\}_{\alpha}$

α-SiC lies on a stacking fault

α-SiC appears have multiple orientation with β-SiC

Neutron Induced Phase Transformation of SiC

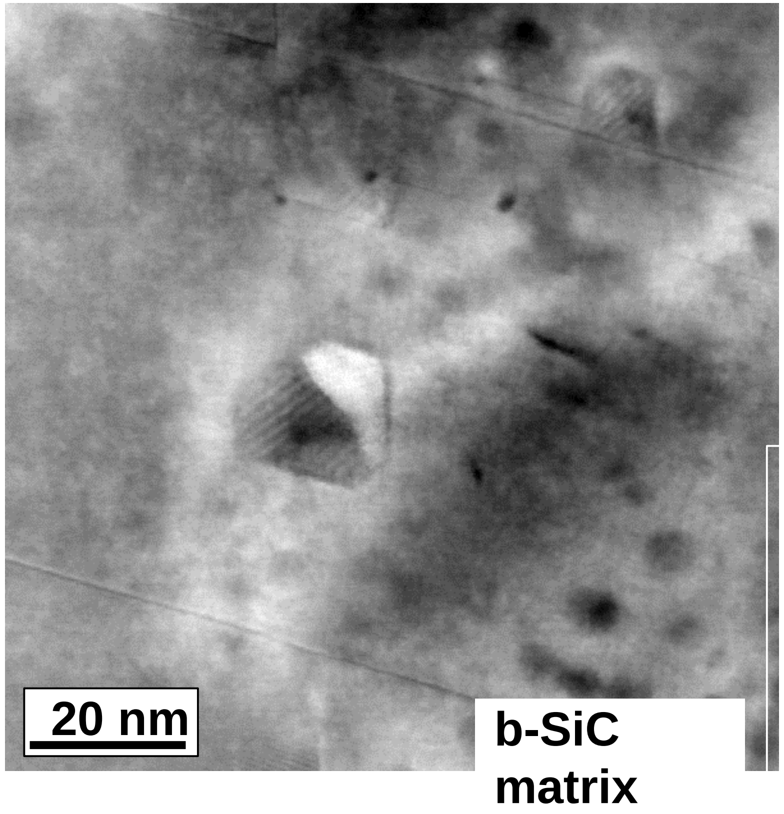
High Resolution (HR)TEM of Pd rich precipitates

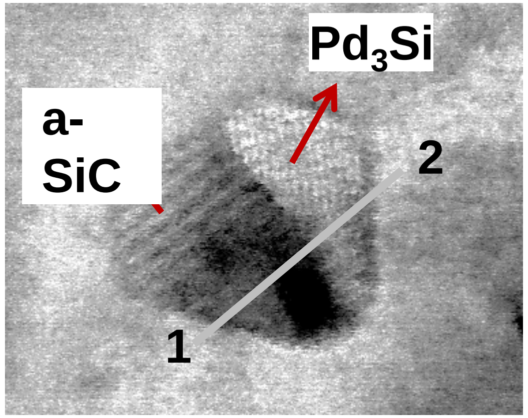


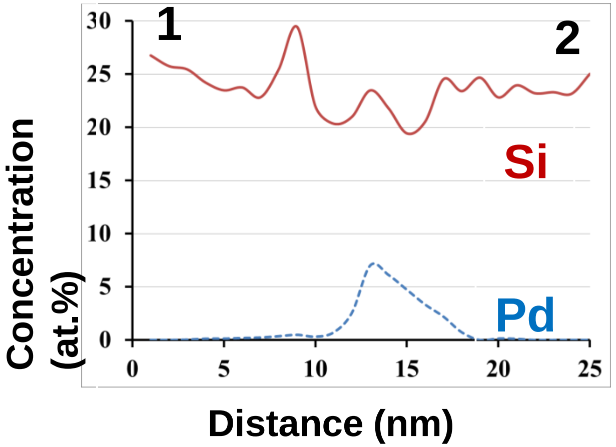
$L1_2$ structure of precipitate corresponds to **Pd_3Si** stoichiometry

Pd rich precipitates were confirmed to be **Pd_3Si, based on their stoichiometry*

Intragranular presence of Pd in SiC

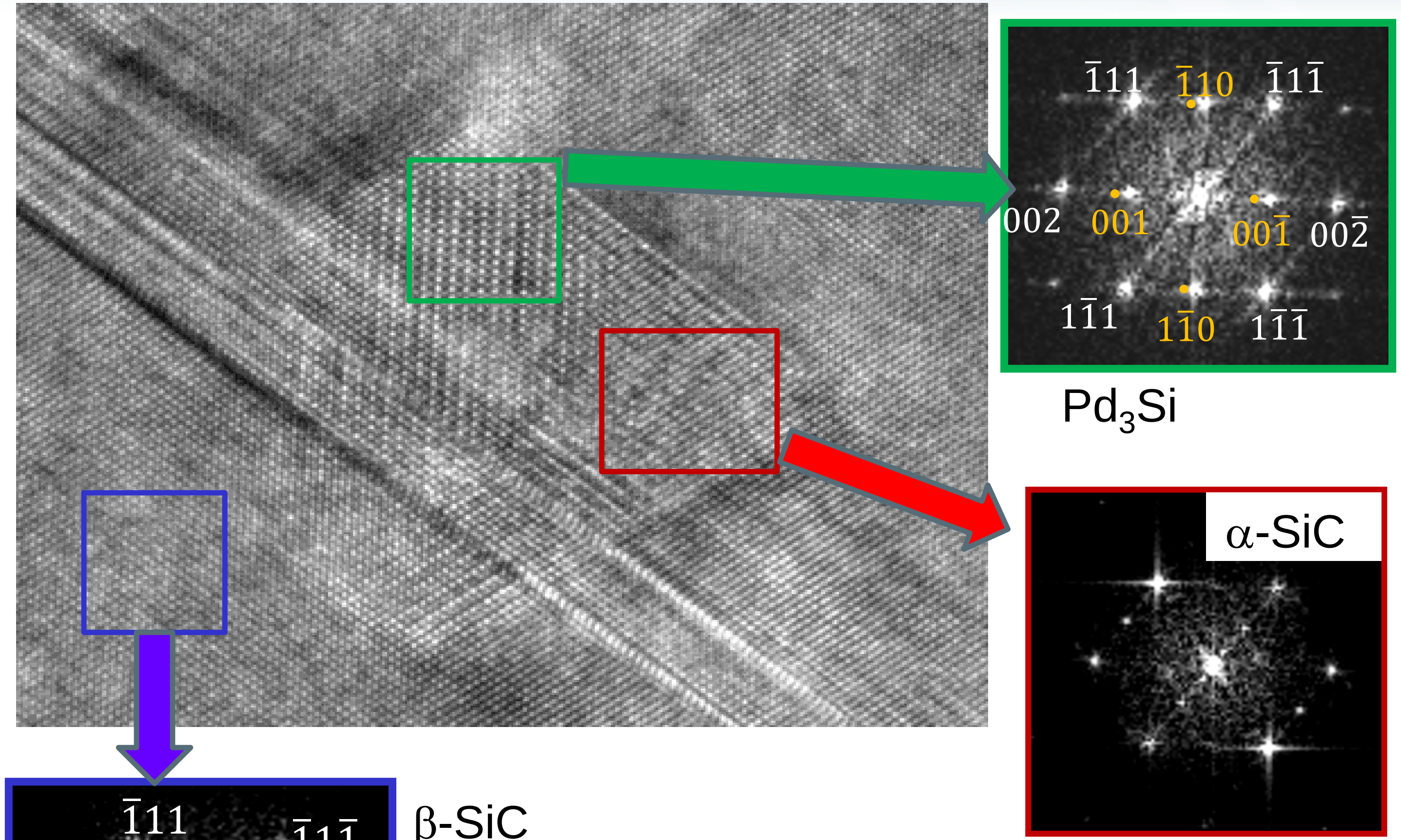


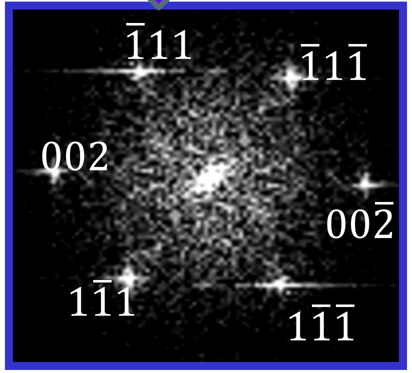




- STEM image showing the different crystallography and mass contrast within the hexagonal structure
- It has been reported that reaction of Pd with α-SiC is easier than β-SiC, hence
- Pd_3Si adopts the morphology of parent surrogate a-SiC: Metamorphosis

Intragranular presence of Pd in SiC



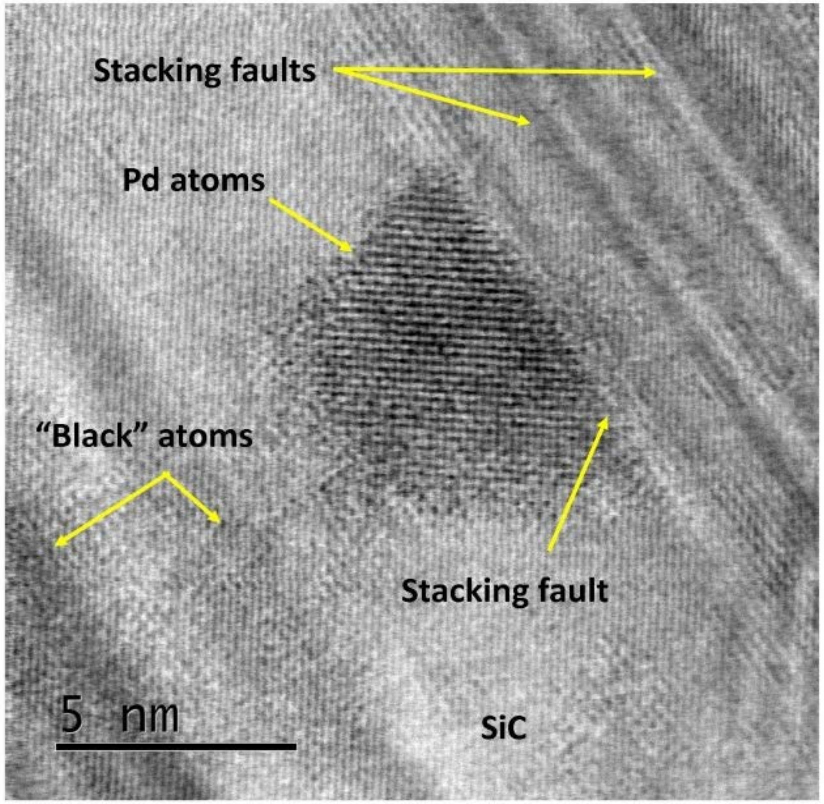


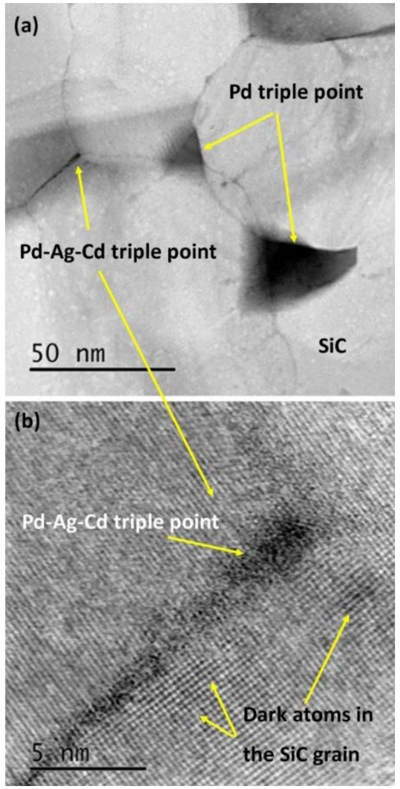
β-SiC

HRTEM captures the transformation of a-SiC into a Pd_3Si precipitate

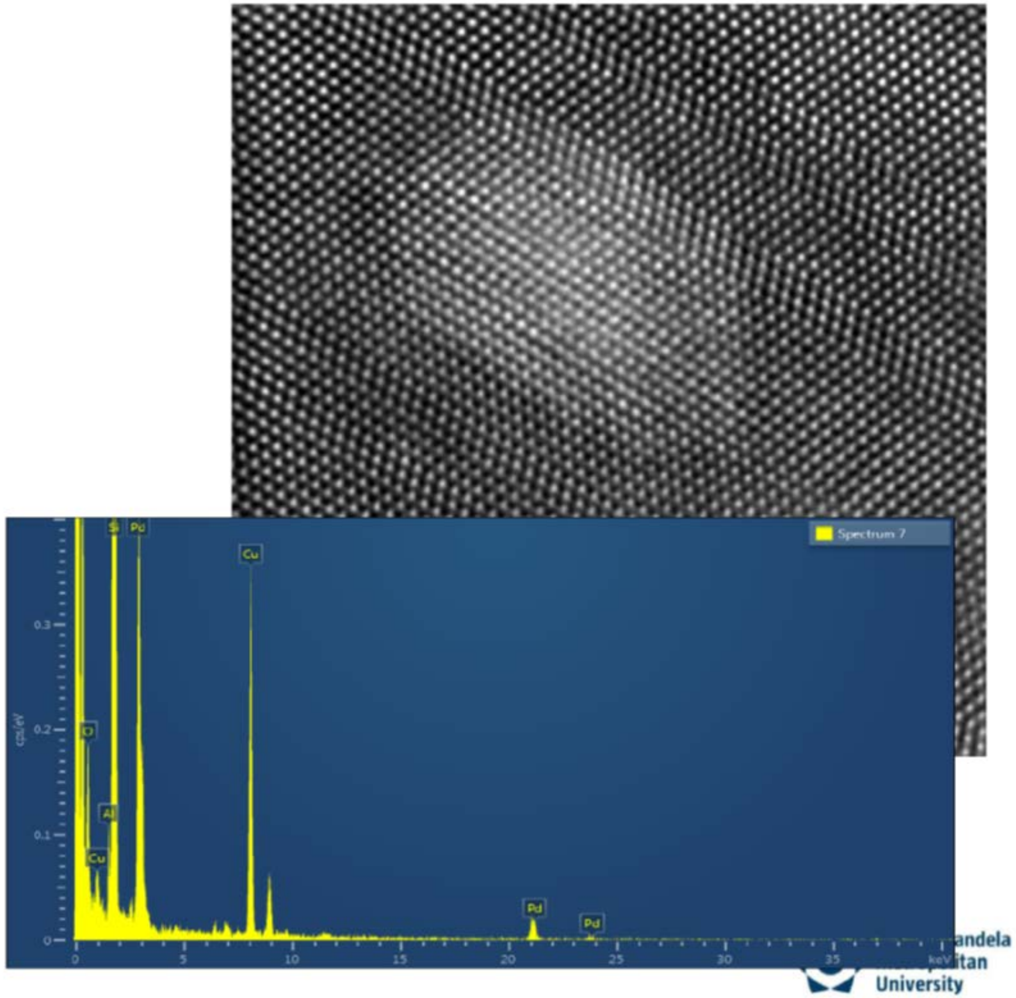
Particle AGR1-411-030

Intragranular Presence of Pd in SiC: Particle AGR1-411-030



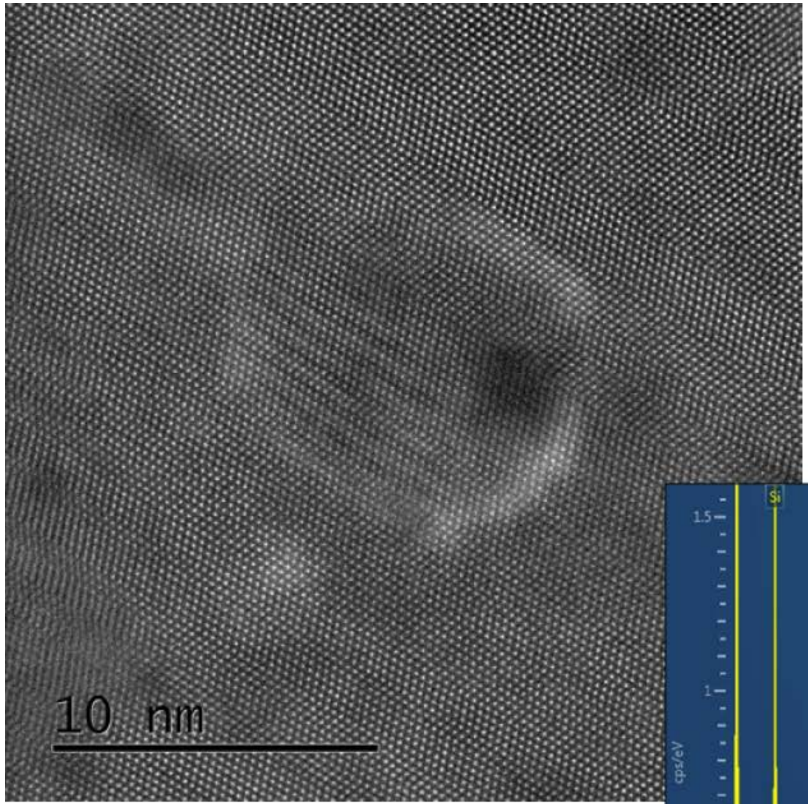


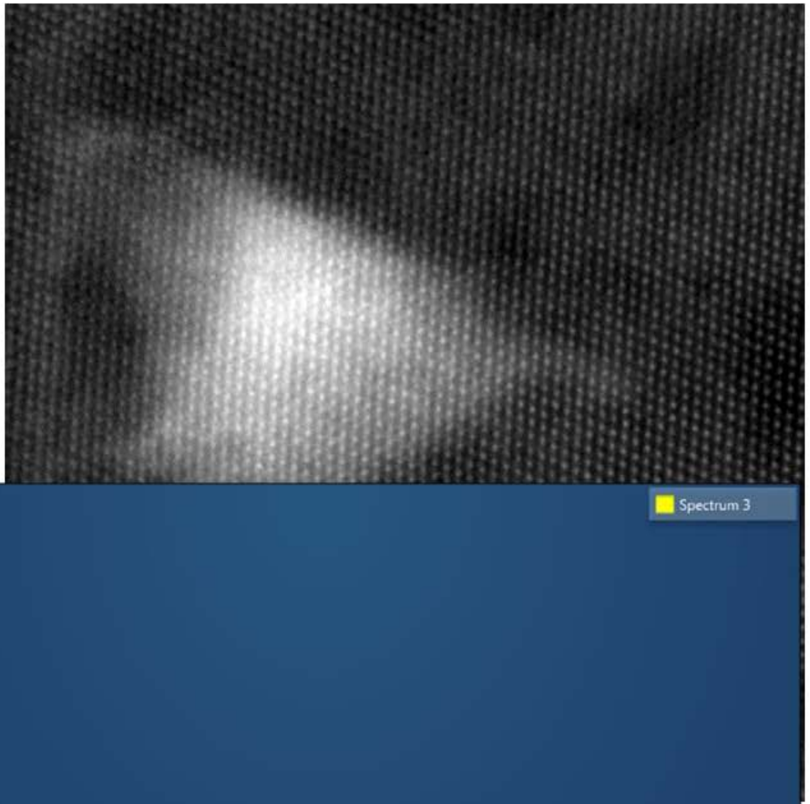
[Van Rooyen,I. J.,E. J. Olivier and J. H Neethling, "Investigation of the Fission Products Silver, Palladium and Cadmium in Neutron Irradiated SiC using a Cs Corrected HRTEM", Journal of Nuclear Materials, 476 (2016) 93 – 101]

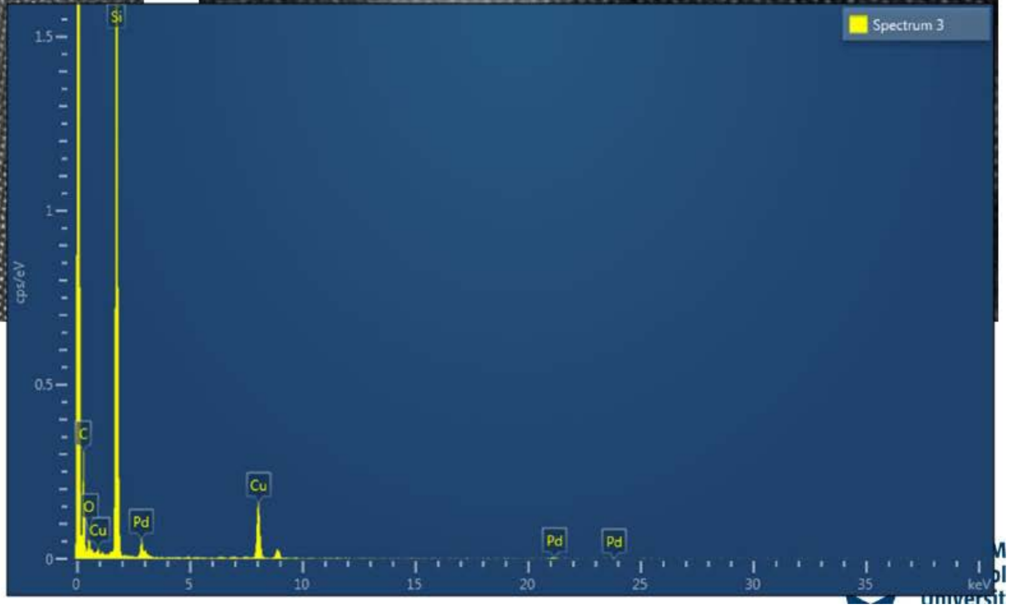


[E J Olivier, J H Neethling, I J van Rooyen, Cs-corrected STEM and EDS Investigation of Pd and Ag transport along SiC grain boundaries and dislocations, Baotou China, 4th SiC workshop]

Intragranular Presence of Pd in SiC: Inclusions with void-like nature





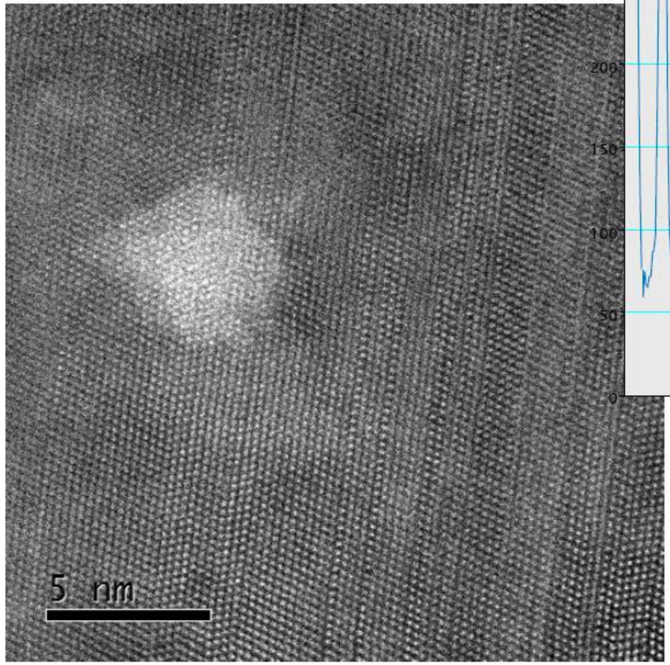


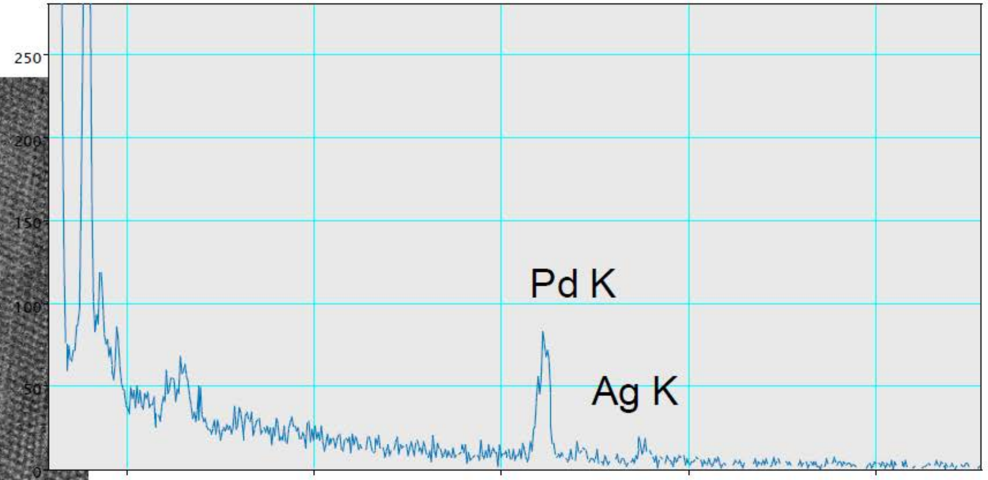
Particle AGR1-411-030

Predominantly Pd rich

[E J Olivier, J H Neethling, I J van Rooyen, Cs-corrected STEM and EDS Investigation of Pd and Ag transport along SiC grain boundaries and dislocations, Baotou China, 4th SiC workshop]

Intragranular Presence of Ag in SiC: Particle AGR1-411-030





Intergranular Ag detected by prolonged EDS acquisition using low dose. Use of Principal Component Analysis for post acquisition processing

[E J Olivier, J H Neethling, I J van Rooyen, Cs-corrected STEM and EDS Investigation of Pd and Ag transport along SiC grain boundaries and dislocations, Baotou China, 4th SiC workshop]

Nelson Mandela Metropolitan University
for tomorrow

Conclusions

- Neutron induced phase transformation of SiC:
  - α-SiC regions found at Frank loops and Stacking Faults of both AGR-1 and AGR-2 particles
  - α-SiC regions appears to have multiple orientations with β-SiC matrix

- No α-SiC regions and Frank loops identified in unirradiated AGR-1 and AGR-2 particles

- Some Transformed regions contain Pd
  - Although Pd is found as the main intragranular fission product, the possibility of Pd assisted transport of other elements cannot be ruled out.
  - Pd rich precipitates confirmed to corresponds to **Pd_3Si**

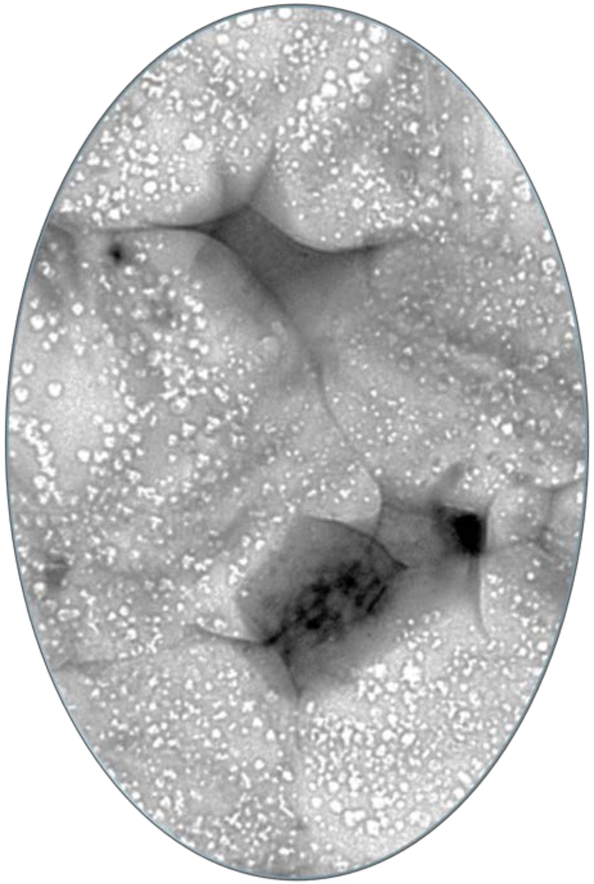
Recommendations: Future Work

- Expand the neutron damage work currently performed by correlating neutron-induced microstructural findings:
  - defect density,
  - volume fraction, and
  - morphology

  with neutron irradiation parameters (i.e., neutron fluence and temperature (based on microstructural features of this study)

- Analyze the defect density and distribution in the vicinity of fission product precipitates

- Integrate PED, neutron damage, chemical composition and structural information for fission product mechanisms



Acknowledgements

- Scott Ploger and Jason Harp: Mount and decontamination preparation for electron microscopy examination and micro-analysis
- Jim Madden: FIB-STEM
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Questions??

What now??

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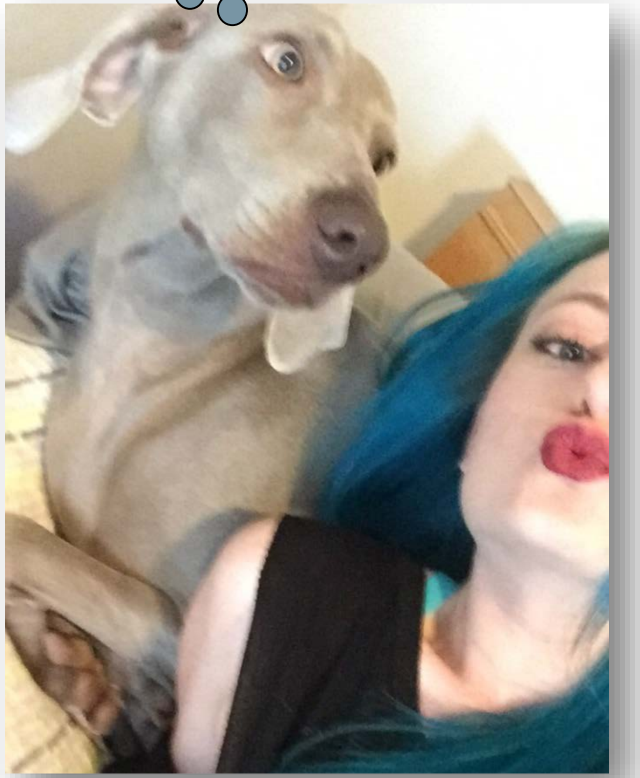

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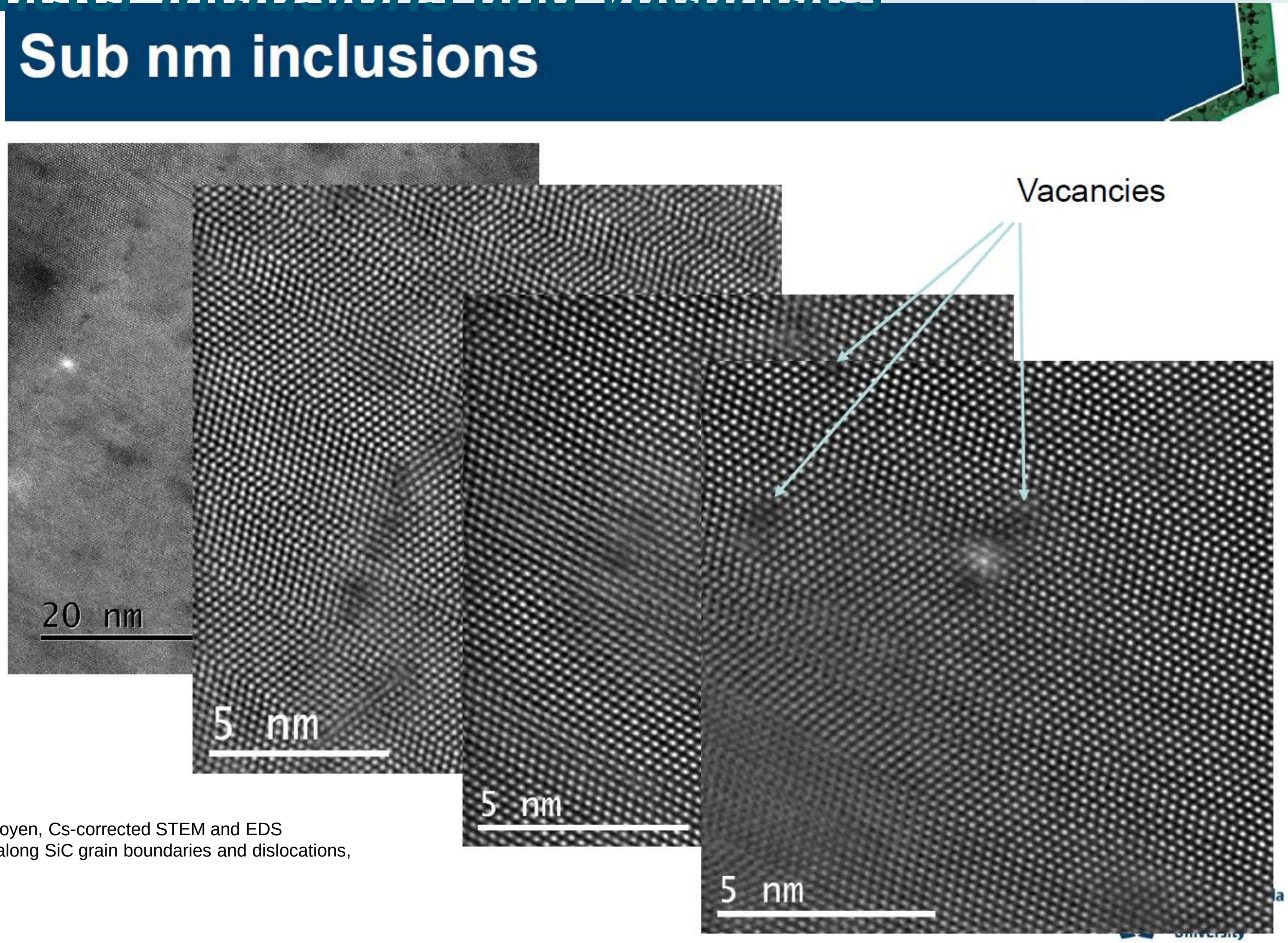
Technique Acronyms

Acronyms	Description
APT	Atom Probe Tomography
EDS	Energy Dispersive Spectroscopy
EBSD	Electron Back Scattered Diffraction
EELS	Electron Energy Loss Spectroscopy
EFTEM	Energy Filtered TEM
EPMA	Electron Probe Micro-Analysis
FIB	Focused Ion Beam
HRTEM	High Resolution Transmission Electron Microscopy
SAD	Selected Area Diffraction
SEM	Scanning Electron Microscope
STEM	Scanning Transmission Electron Microscopy
TEM	Transmission Electron Microscope
t-EBSD	Transmission-EBSD
TKD	Transmission Kikuchi Diffraction
WDS	Wavelength Dispersive Spectroscopy

Sub nanometer inclusions and vacancies



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