Annealing effect on the microstructure and hardness of irradiated tungsten

Barbara Horvath[1], Yong Dai[1], Yongjoong Lee[2]
[1] Paul Scherrer Institute
[2] European Spallation Source

Outline

- Introduction
- Sample preparation for (un)irradiated tungsten
- Microstructural evaluation of
  - *Unirradiated W*
  - *Irradiated W*
- Effect of annealing on irradiated W
- Effect of radiation damage amount on microstructure

Introduction

- The European Spallation Source (ESS) will be the world's most powerful neutron source facility built in Lund, Sweden
- A number of target options were reviewed for the European Spallation Source → Pure tungsten was chosen as the spallation material
- Advantages:
    - environmentally friendly compared to other target materials
    - its use in helium environment avoids corrosion issues related to water cooling
- Disadvantages:
    - low ductility
    - high ductile-to-brittle transition temperature

[1] europeanspallationsource.se

Outline

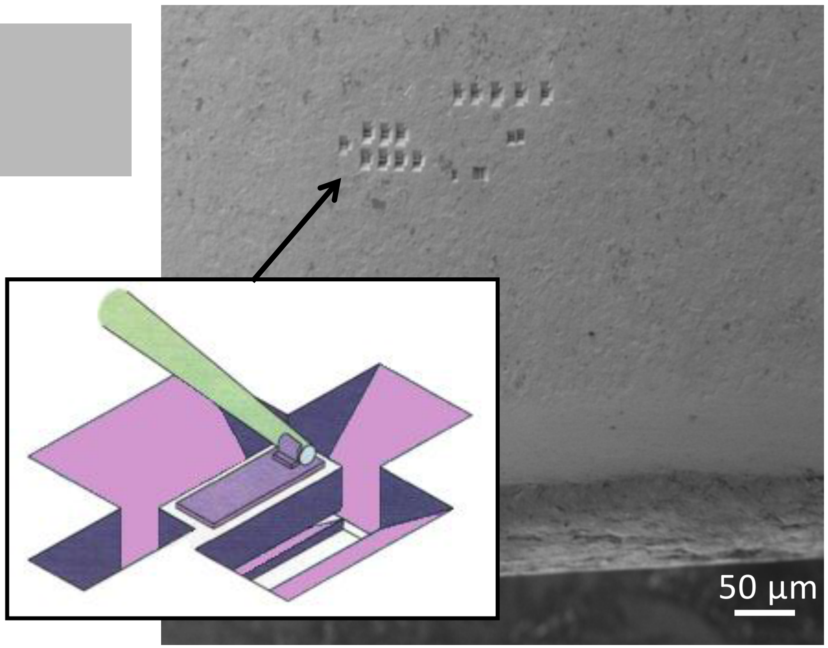
- Introduction
- Sample preparation for (un)irradiated tungsten
- Microstructural evaluation of
  - *Unirradiated W*
  - *Irradiated W*
- Effect of annealing on irradiated W
- Effect of radiation damage amount on microstructure

Sample preparation for (un)irradiated tungsten

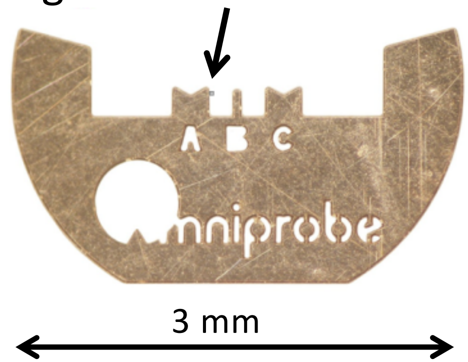
In order to examine the irradiated tungsten samples safely the following method was used:

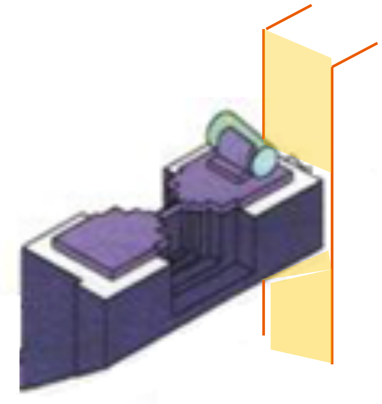
1. **Lamella with focused ion beam (FIB):**
   - The reduction of the sample size reduces the activity
   - Sample lifted out internally by a micromanipulator 8x8 μm^2 and 200 nm thick lamella is milled.

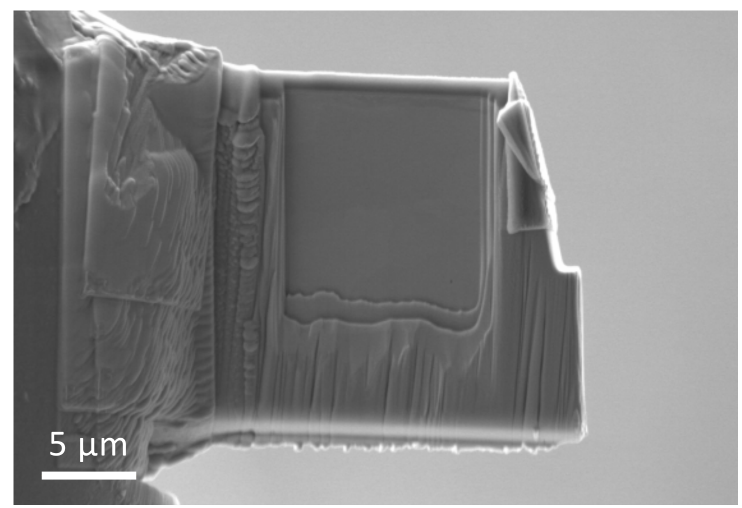
FIB sample development



TEM grid:







Creating a TEM sample with FIB:

a) etching around the sample,

b) transferring the sample to the TEM grid, thinning the membrane

Sample preparation for (un)irradiated tungsten

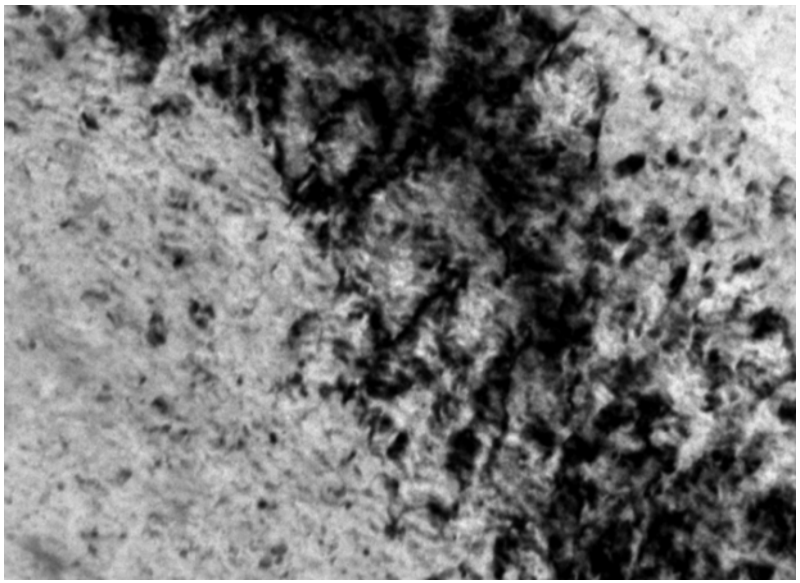
In order to examine the irradiated tungsten samples safely the following method was used:

1. **Lamella with focused ion beam (FIB):**
   - The reduction of the sample size reduces the activity
   - Sample lifted out internally by a micromanipulator 8x8 μm^2 and 200 nm thick lamella is milled.

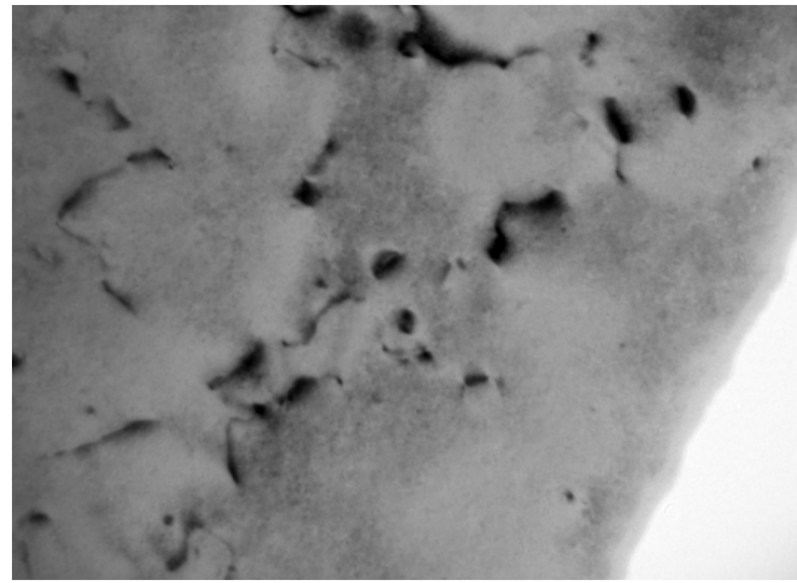
2. **Flash electrochemical polishing:**
   - The lamella is thinned down to approx. 60 nm
   - Remove the FIB induced damages on the surface.

The effect of FIB on the lamella

TEM image of a lamella



Before polishing



After polishing

The ion impact on the specimen surface leads to

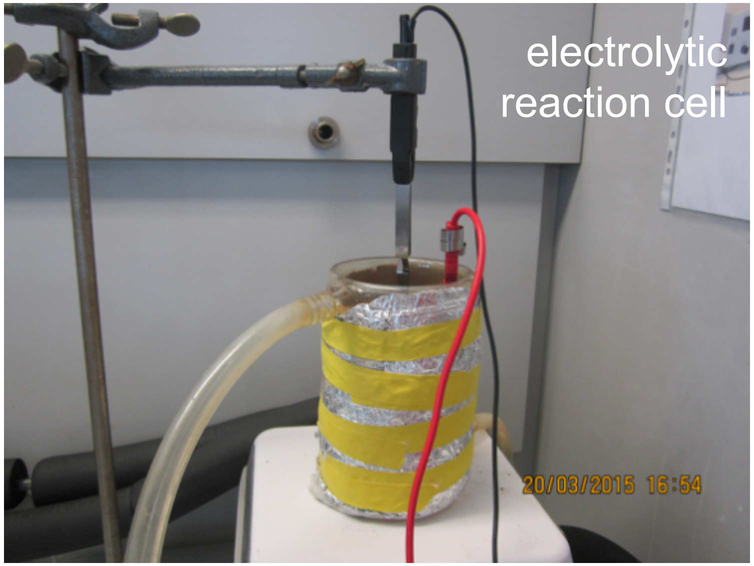
- formation of a damaged layer that may extend 10s of nm into the material
- Ga implantation into the bulk

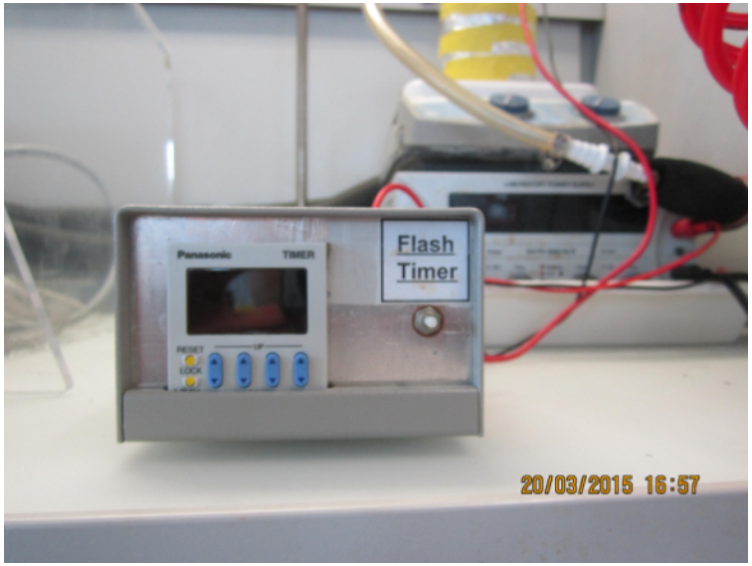
Flash electrochemical polishing

Inital lamella thickness: 200-300nm

Factors for polishing:

- Polishing Voltage
- Polishing Temperature
- Polishing solution type and percentage: NaOH 0.5-1% solution.
- TEM grid material: Mo / Cu
- Welding material for lamella to grid: C / Pt
- Polishing time: 0.001 – 0.5 second.

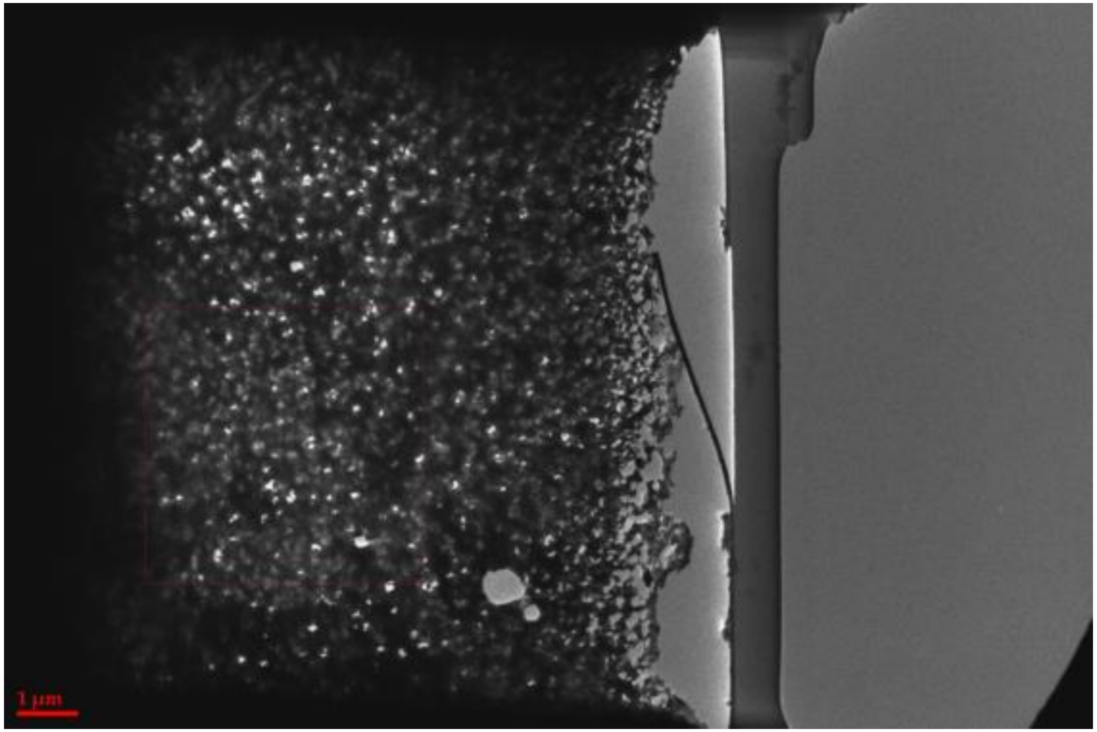




Flash electrochemical polishing

So which etching parameter should be changed…?

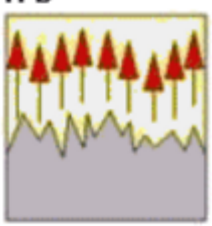
... and which direction?

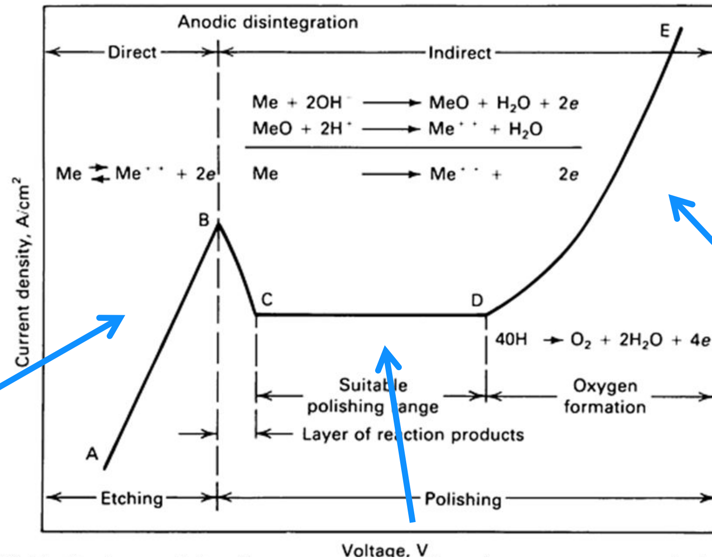


Flash electrochemical polishing

Direct anodic dissolution leads to etching

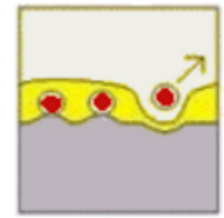
A-B

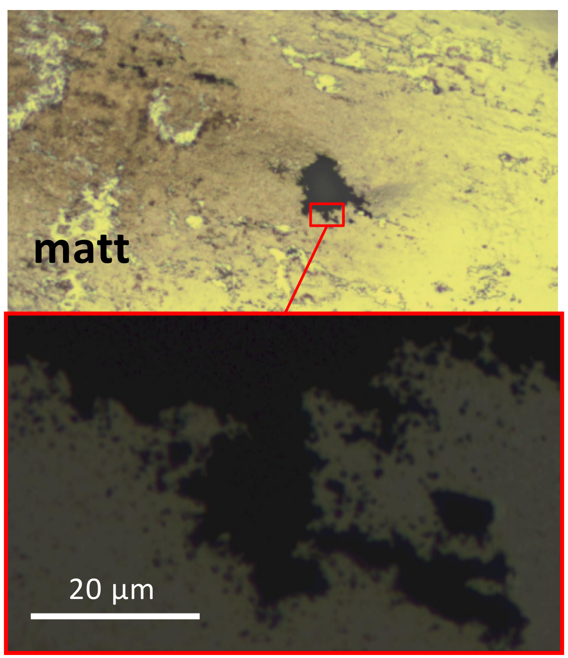


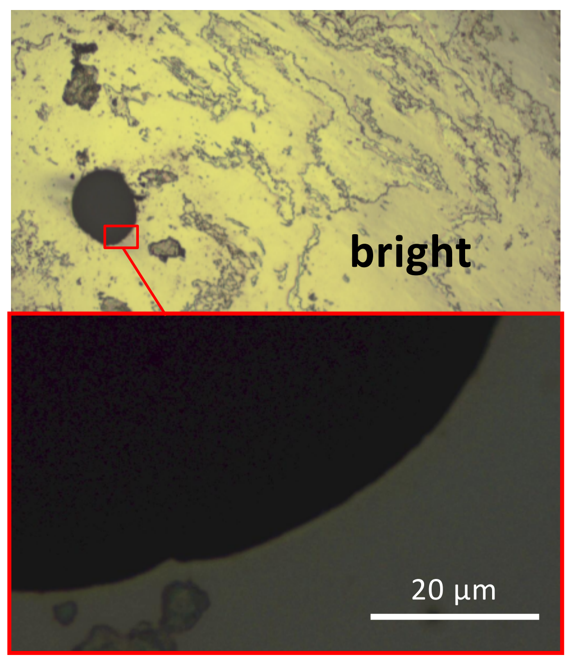


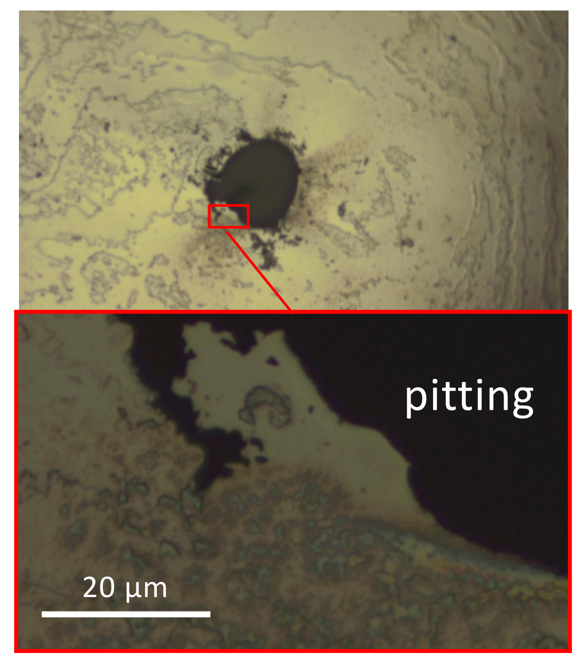
Oxygen bubbles may cause pitting

D-E



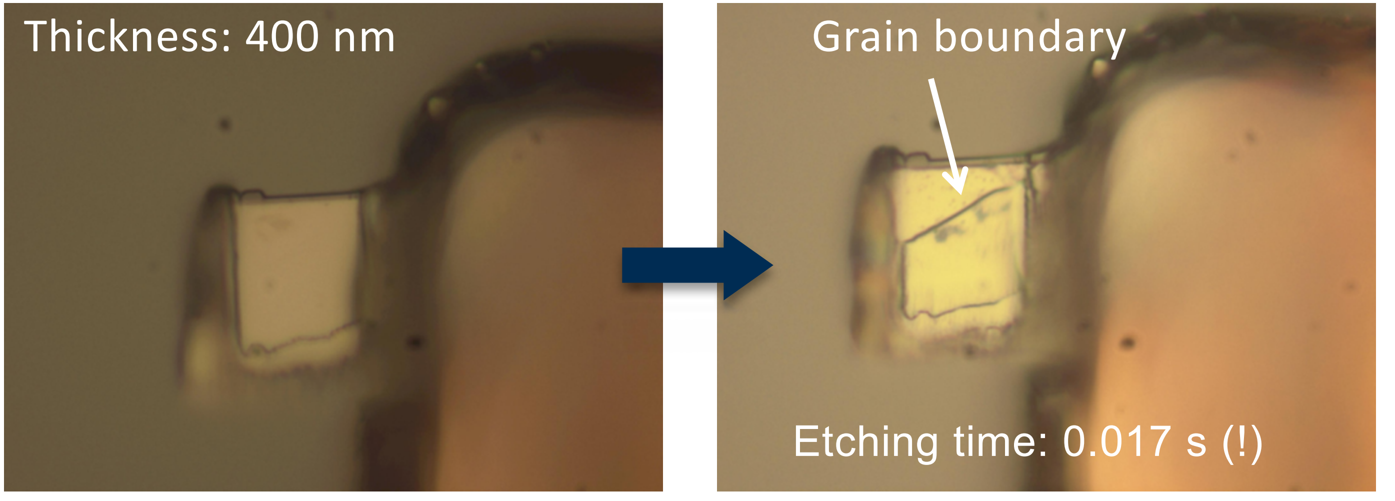






Flash electrochemical polishing a TEM lamella

Flash electropolishing



before and after flash electropolishing

Flash electropolishing conditions

Unirradiated W	Irradiated W
Cu grid, Pt weld	Cu grid, Pt weld
2 ℃	9 ℃
0.5% NaOH	0.5% NaOH
23 V	23 V

Sample preparation for (un)irradiated tungsten

In order to examine the irradiated tungsten samples safely the following method was used:

1. **Lamella with focused ion beam (FIB):**
   - The reduction of the sample size reduces the activity
   - Sample lifted out internally by a micromanipulator 8x8 μm^2 and 200 nm thick lamella is milled.

2. **Flash electrochemical polishing:**
   - The lamella is thinned down to approx. 60 nm
   - Remove the FIB induced damages on the surface.

3. **Transmission Electron Microscopy:**
   - Bright field (BF) and weak-beam dark field (WBDF) imaging conditions were used at (g, 6g), (g, 5g) or two-beam; (g=110).

Outline

- Introduction
- Sample preparation for (un)irradiated tungsten
- Microstructural evaluation of
  - *Unirradiated W*
  - *Irradiated W*
- Effect of annealing on irradiated W
- Effect of radiation damage amount on microstructure

Unirradiated tungsten

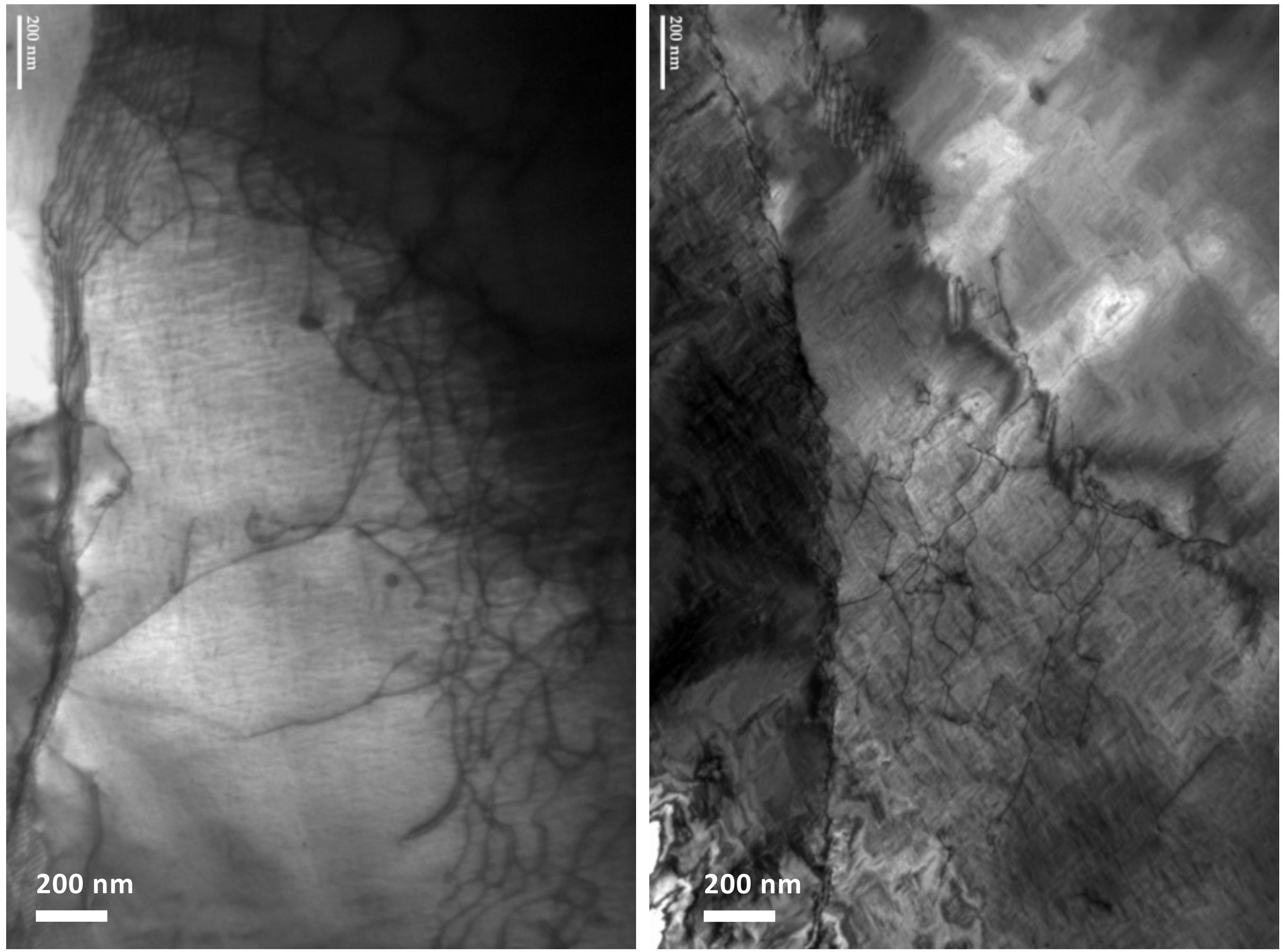
Number of dislocation lines crossing unit area in the sample

$$r_D = (n/ L^2)\ m^{-2}$$

Area of examination: 33.1 μm^2

Dislocation density:

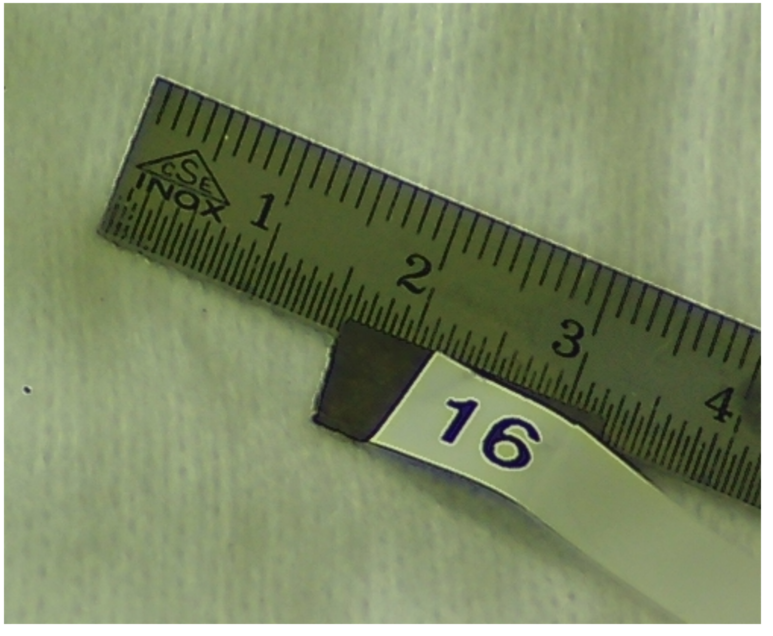
1.6×10^{13} m^{-2}



Low magnification micrographs of the dislocation structure

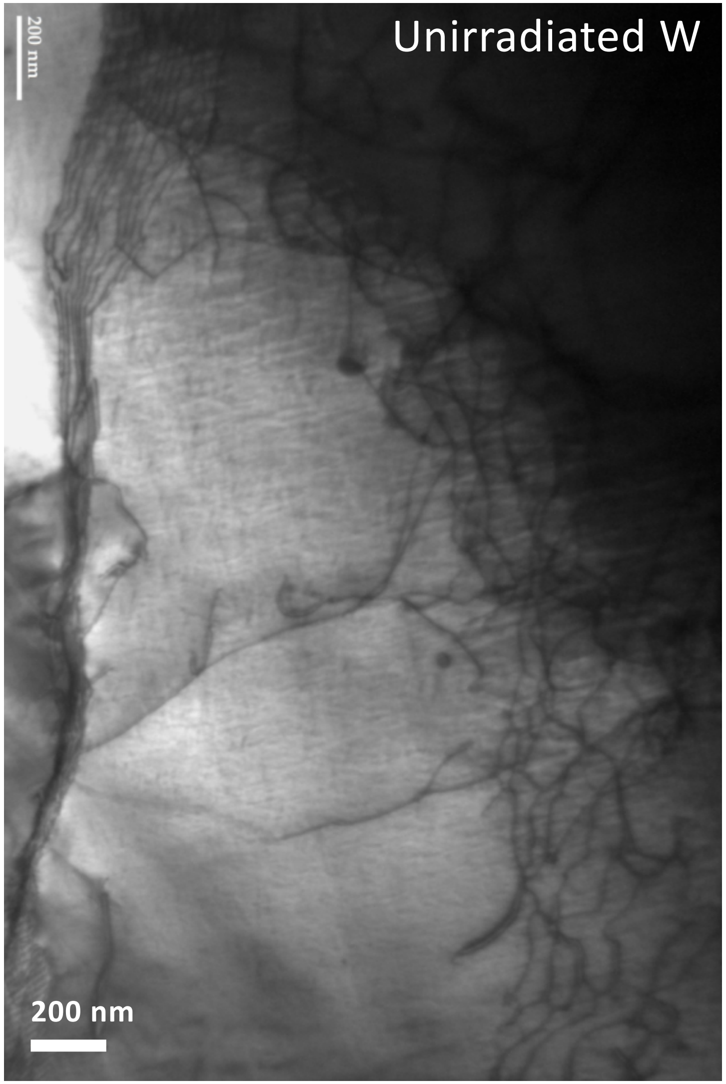
Irradiated tungsten

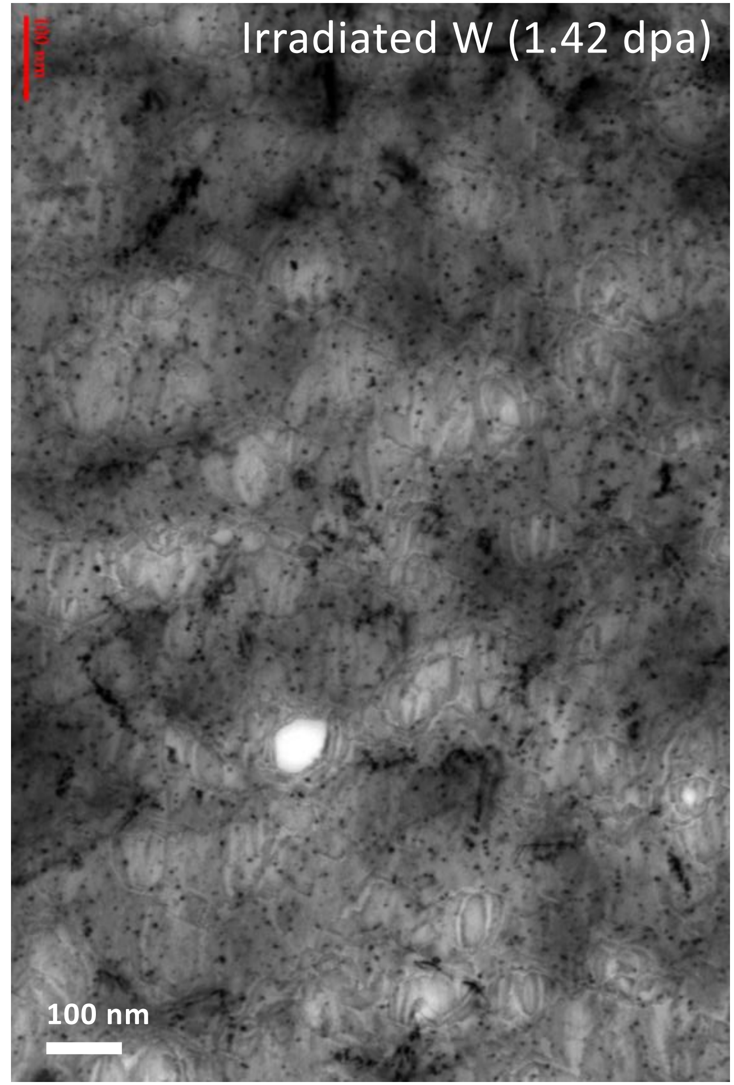
Irradiation conditions	5-SSB-W-16-1-R5
Dpa	1.42
He (appm)	37
$T_{irradiation}$	80 °C



Bending test at 450 °C previously

Effect of irradiation





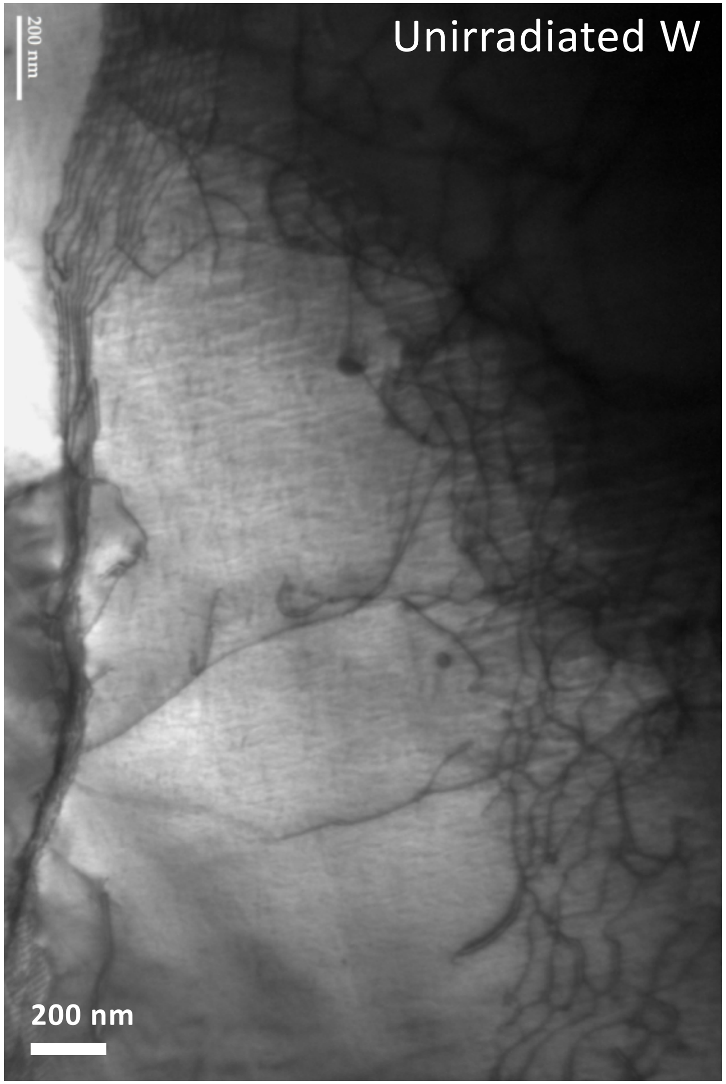
Dislocation density: 1.6×10^{13} m^{-2}

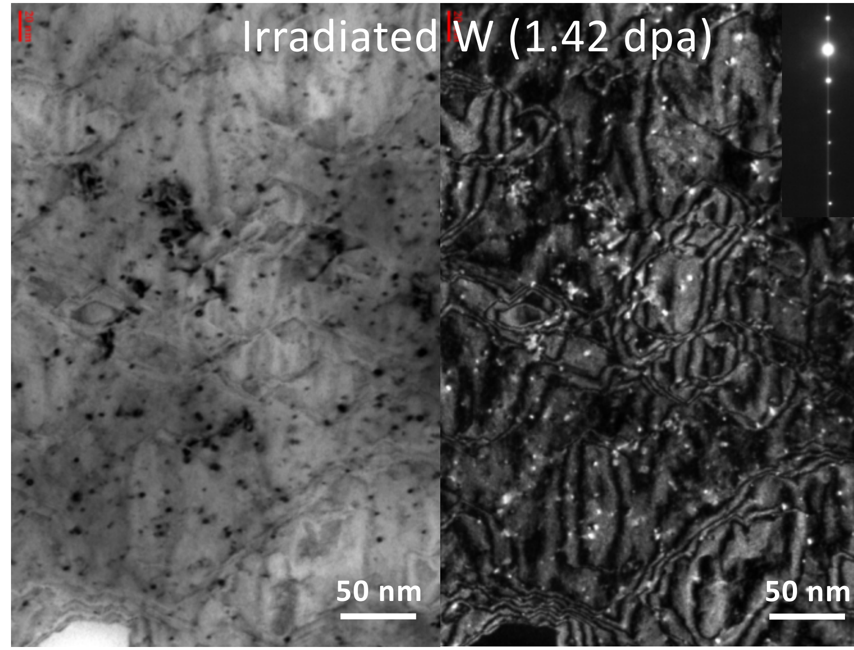
Dislocation density: 1.9×10^{13} m^{-2}

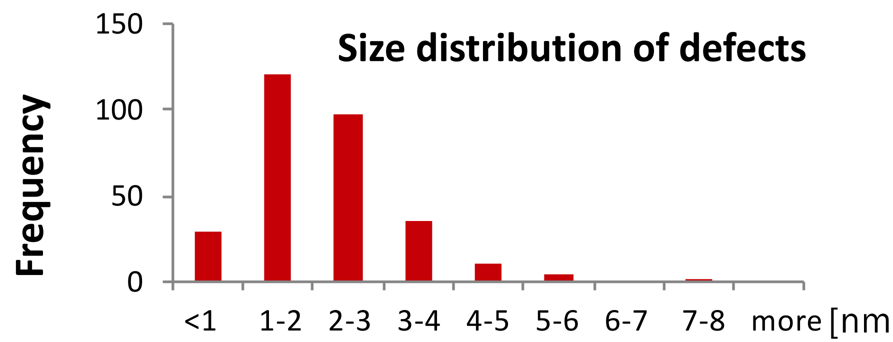
Cluster density: 3.2×10^{23} m^{-3}

Effect of irradiation

Defect clusters: aggregation of interstitial atoms through displacement damage







Dislocation density: $1.6 \times 10^{13}\ m^{-2}$

Dislocation density: $1.9 \times 10^{13}\ m^{-2}$

Cluster density: $3.2 \times 10^{23}\ m^{-3}$

Outline

- Introduction
- Sample preparation for (un)irradiated tungsten
- Microstructural evaluation of
  - *Unirradiated W*
  - *Irradiated W*
- Effect of annealing on irradiated W
- Effect of radiation damage amount on microstructure

PAUL SCHERRER INSTITUT

Effect of annealing on irradiated tungsten

Evaluate the microstructure and hardness change on 1.42 dpa irradiated W sample after annealing

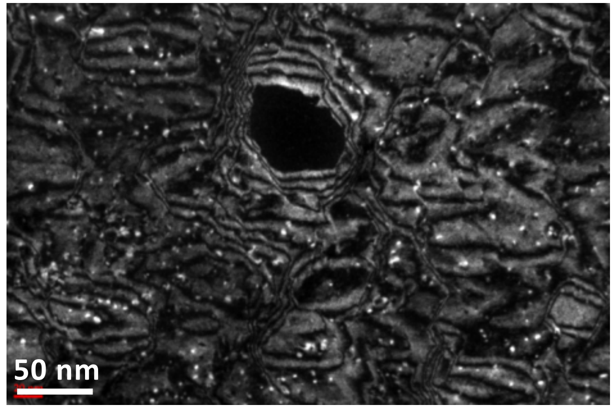
Annealing temperature:

- 500 ˚C
- 600 ˚C
- 800 ˚C
- 900 ˚C

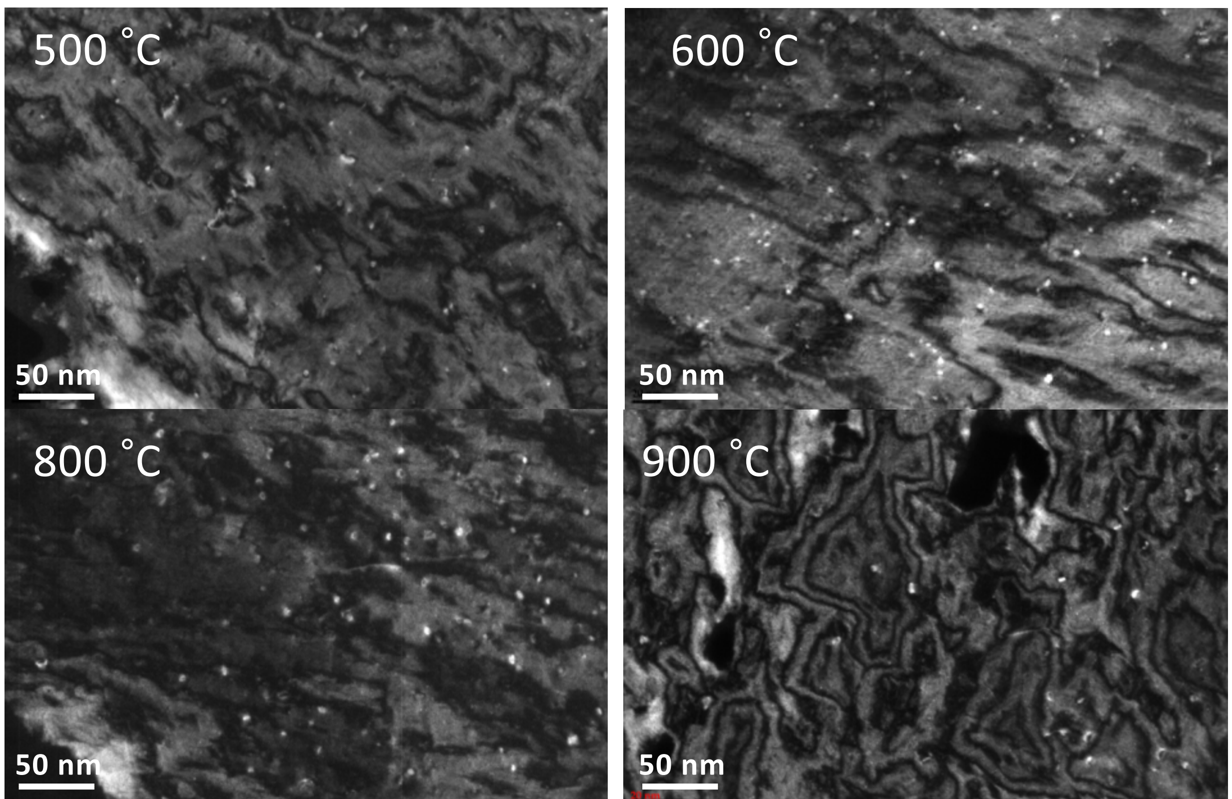
Duration: 1 hour

Counting clusters according to their sizes

Before annealing:

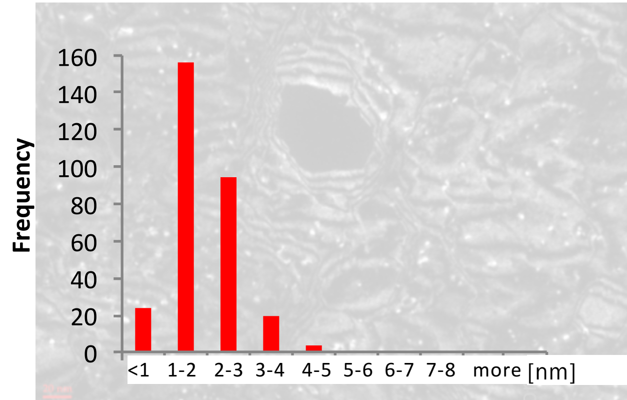


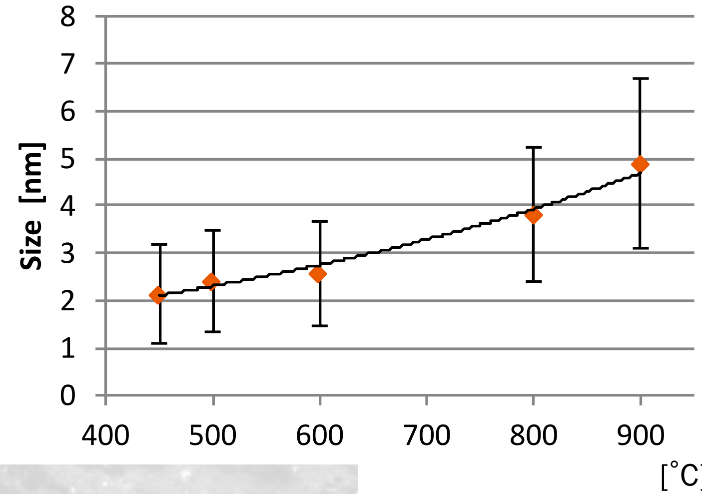
After annealing:



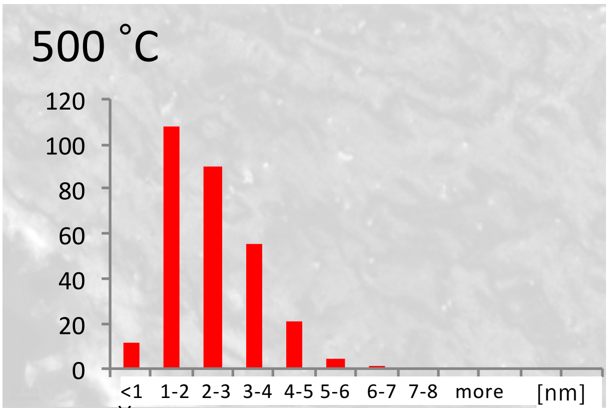
Counting clusters according to their sizes

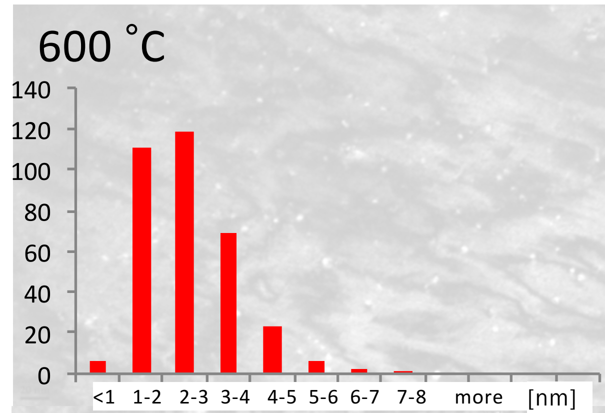
Before annealing:

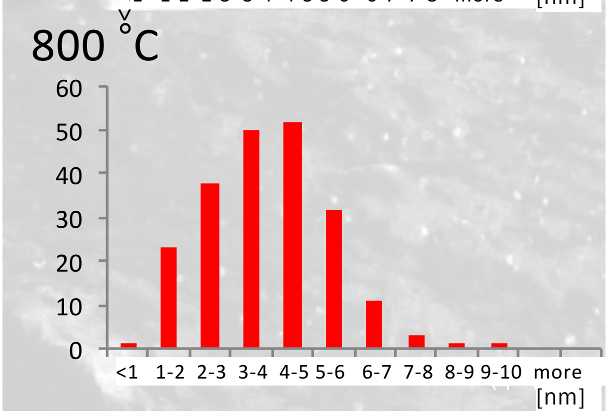


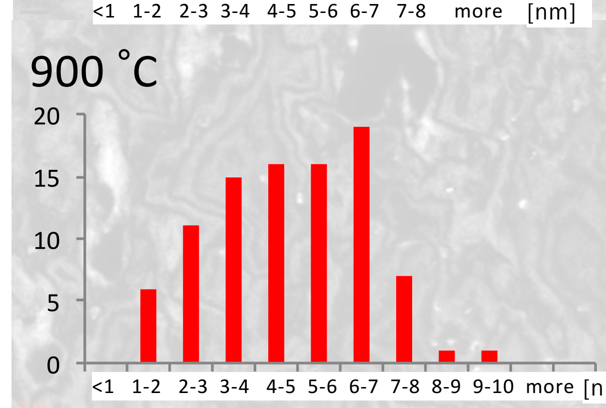


After annealing:



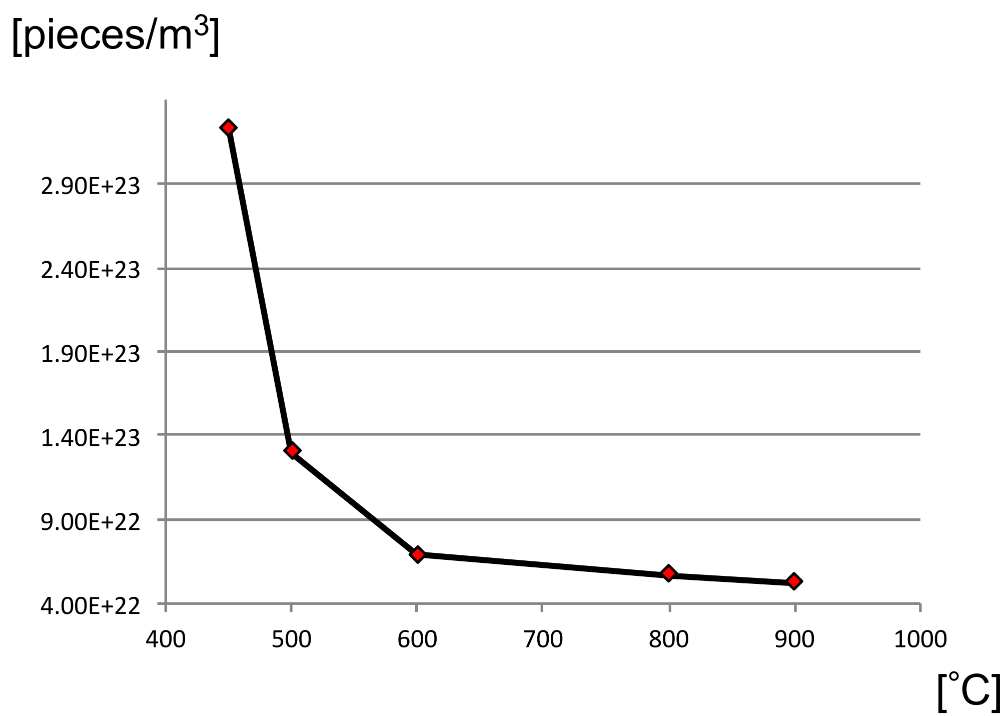






Counting cluster defects

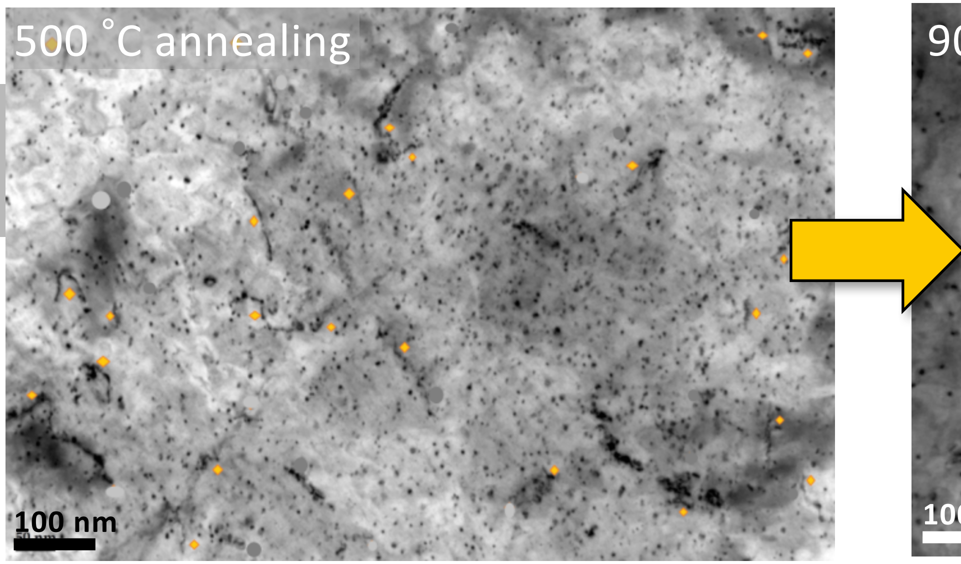
	Cluster defect density [pieces/m³]
No anneal	3.23×10^{23}
500 °C anneal	1.30×10^{23}
600 °C anneal	6.87×10^{22}
800 °C anneal	5.67×10^{22}
900 °C anneal	5.21×10^{22}

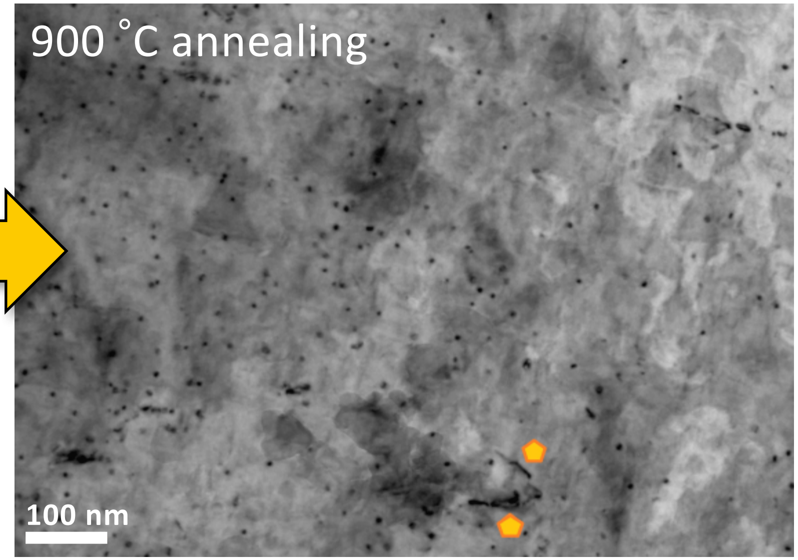


Cluster size increases
density decreases

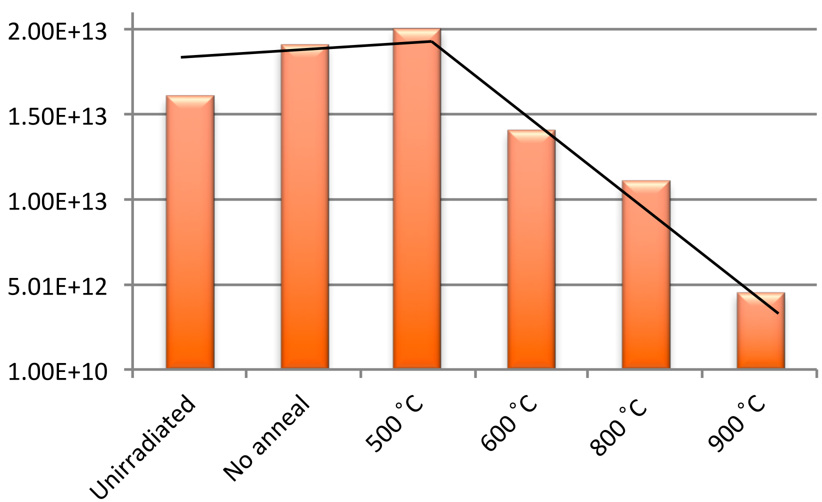
At higher temperatures they migrate and diffuse into each other

Dislocation density





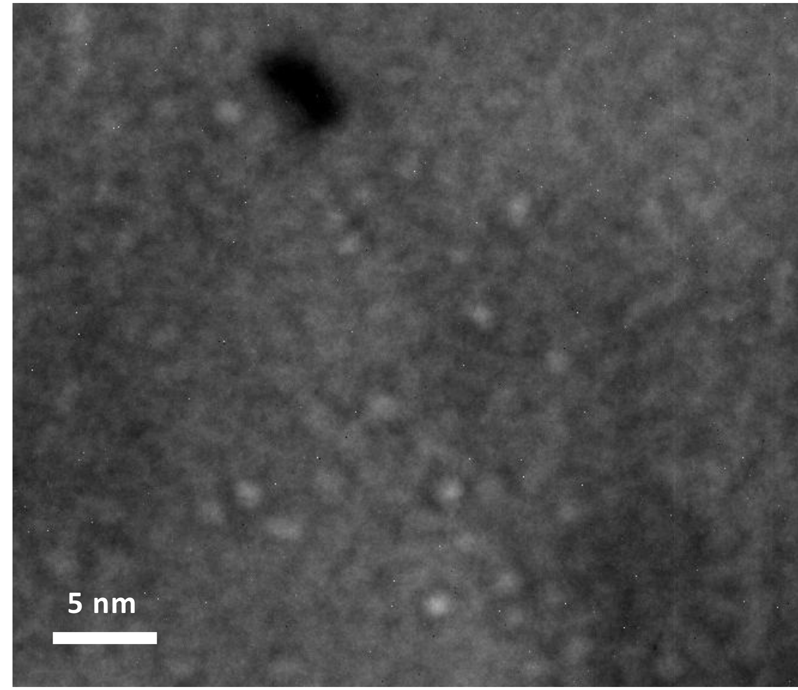
Decreasing dislocation density as annealing temperature rises



Area of examination: 3-5 μm²

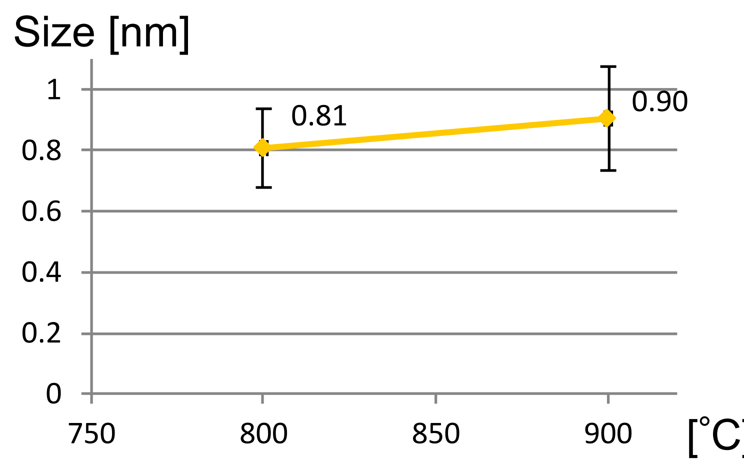
	Dislocation density [m^{-2}]
Unirradiated	1.6×10^{13}
No anneal	1.9×10^{13}
500 °C anneal	2.0×10^{13}
600 °C anneal	1.4×10^{13}
800 °C anneal	1.1×10^{13}
900 °C anneal	4.5×10^{12}

Bubble development



- First observed on 800 ˚C annealed samples
  - Neutron irradiation develop voids above a certain T
  - He developed by transmutation reactions due to irradiation, fills voids
- Bubble size increased and density decreased slightly at 900˚C

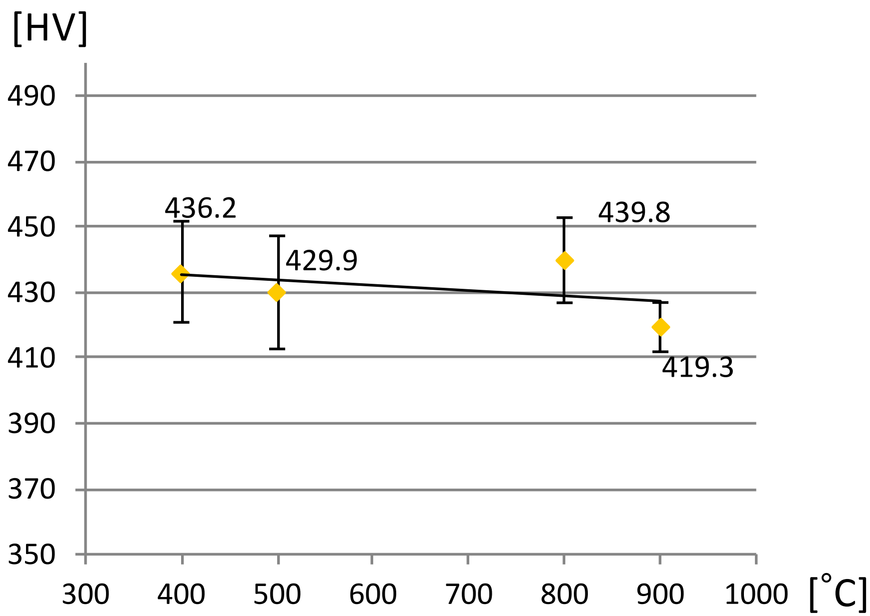
	Bubble density [pieces/m³]
800 ˚C anneal	1.27×10^{24}
900 ˚C anneal	1.18×10^{24}



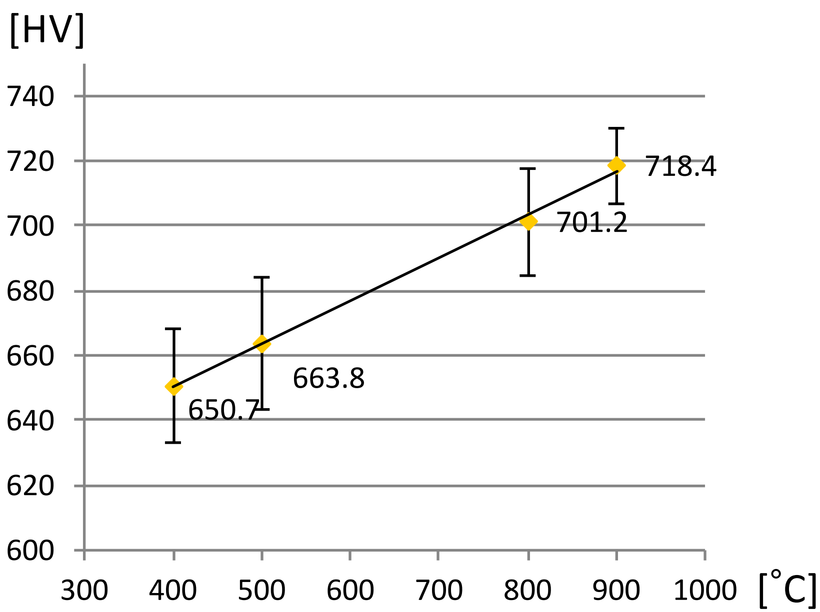
Hardness measurements

Vickers hardness test:

- diamond indenter (pyramid with square base, angle 136˚), 0.5 kg load, 15 sec
- Defects and bubbles are obstacles to dislocation movement
  → hardness increases



Unirradiated tungsten



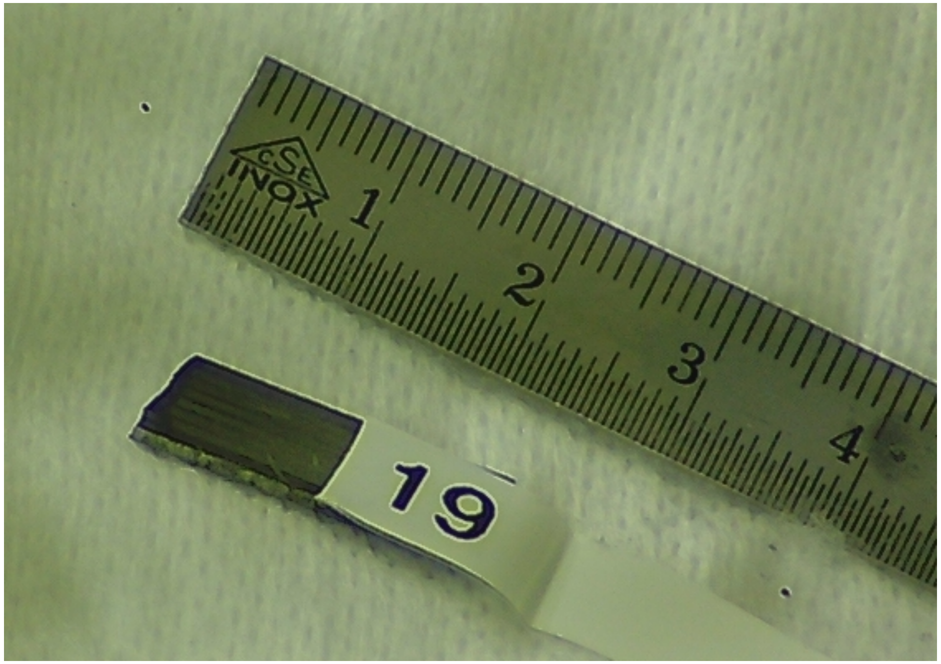
Irradiated tungsten (1.42 dpa)

Outline

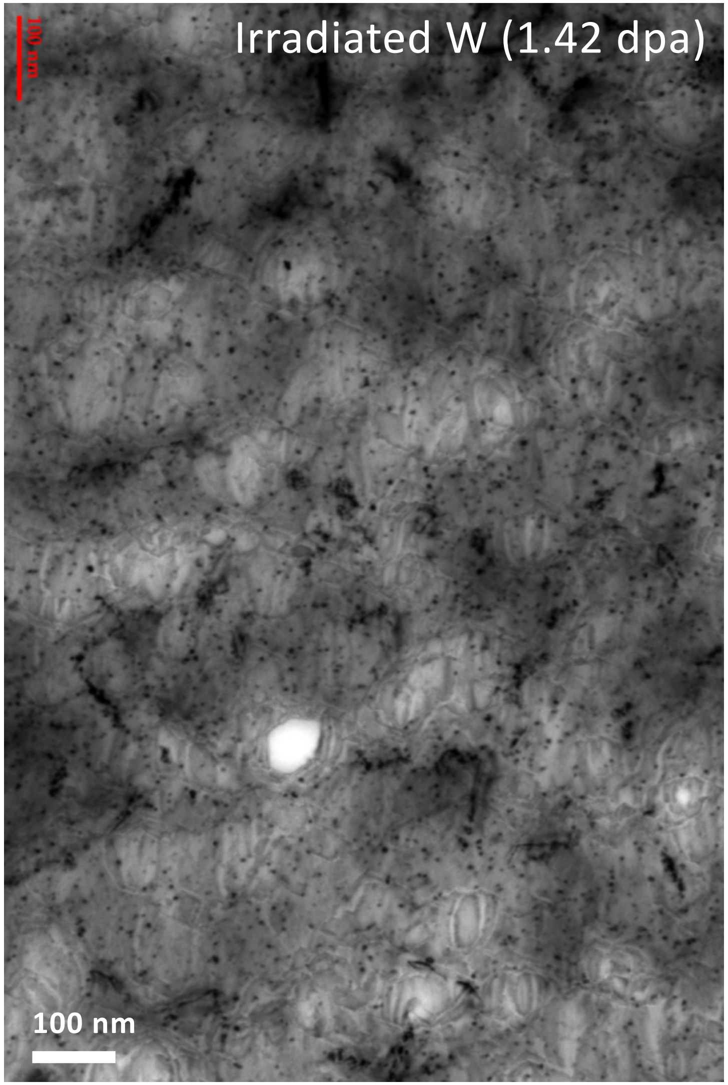
- Introduction
- Sample preparation for (un)irradiated tungsten
- Microstructural evaluation of
  - *Unirradiated W*
  - *Irradiated W*
- Effect of annealing on irradiated W
- Effect of radiation damage amount on microstructure

Effect of radiation damage amount on microstructure

Irradiation conditions	5-SSB-W-16-1-R5	5-SSB-W-19A-3-R5
Dpa	1.42	3.5
He (appm)	37	140
$T_{irradiation}$	80 °C	110 °C

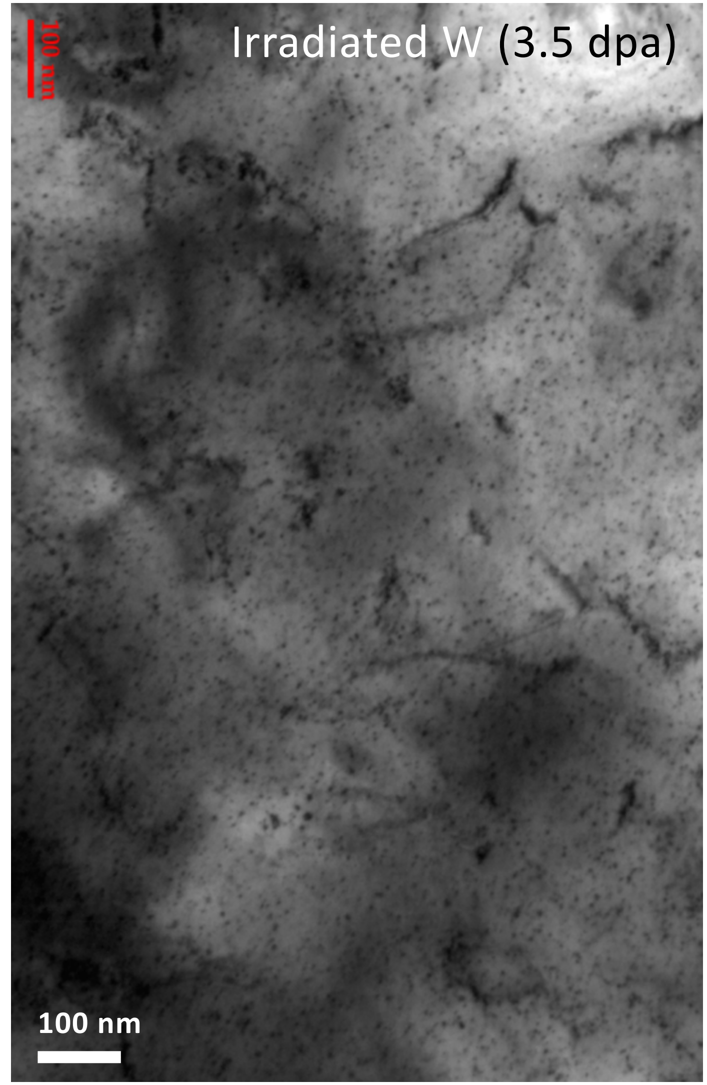


Effect of radiation damage amount



Dislocation density: 1.9×10^{13} m^{-2}

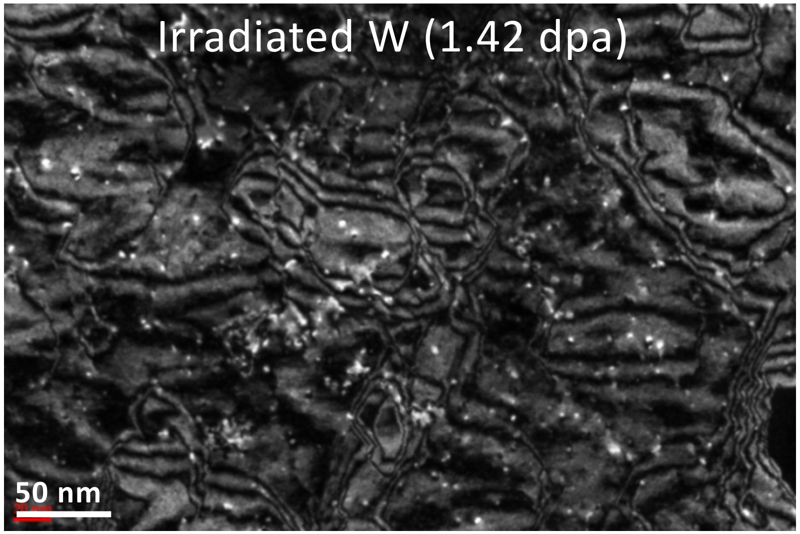
Cluster density: 3.2×10^{23} m^{-3}

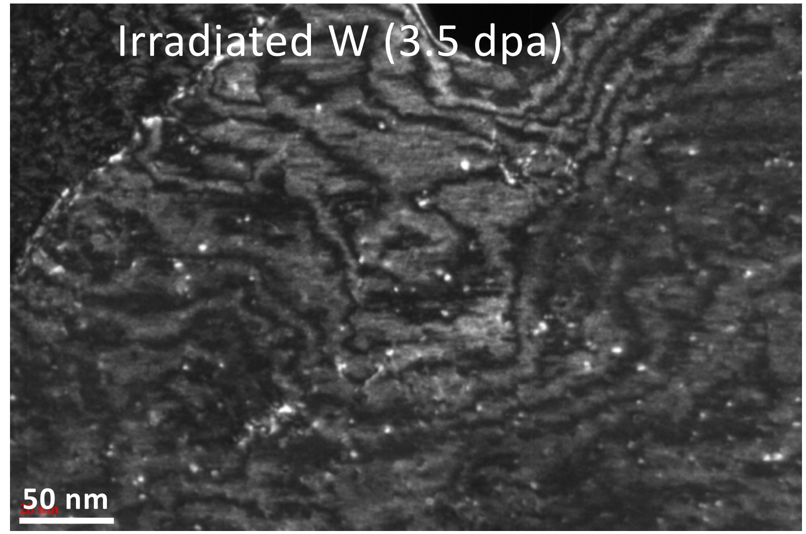


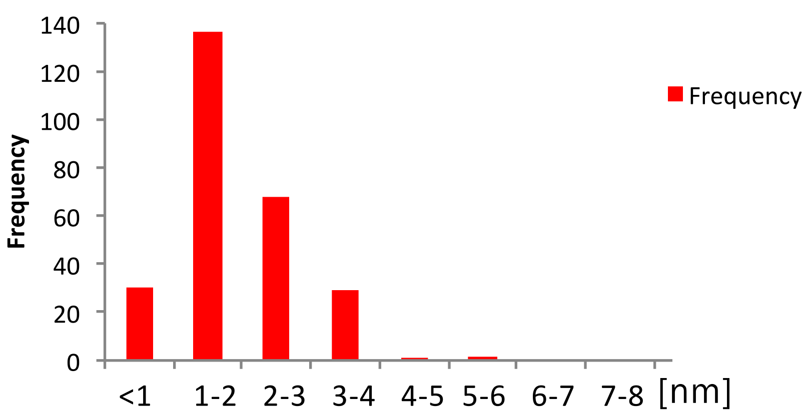
Dislocation density: 1.8×10^{13} m^{-2}

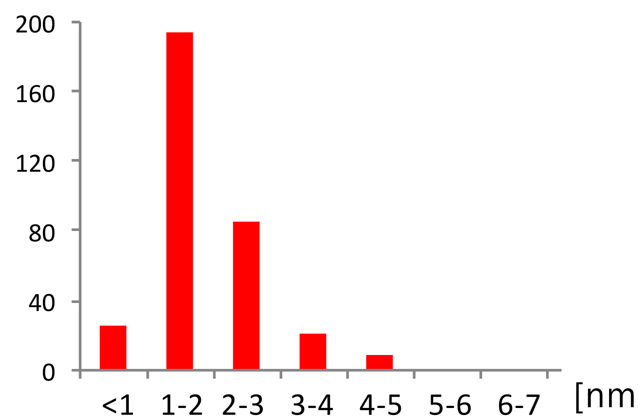
Cluster density: 3.1×10^{23} m^{-3}

Effect of radiation damage amount









Conclusion

1. The quality of flash polishing strongly depends on parameters such as voltage, temperature or the material of the grid and welding. Irradiating W shifts the optimal flash polishing parameters from 2 to 9 ˚C.
2. The structure of W changed due to irradiation. The dislocations shortened down and defect clusters appeared.
3. Annealing has an effect on dislocations and defects:
    1. No. of dislocations reduce to more than half at 900 ˚C.
    2. No. of cluster defects reduce down to $1/6^{th}$ at 900 ˚C
    3. Their sizes grow to approx. 5 nm from the initial 2 nm.
    4. He bubble development is observed from 800 ˚C
4. Radiation damage change from 1.42 to 3.5 dpa had no effect on the W structure

Thank you for your attention!

Barbara Horvath
barbara.horvath@psi.ch