Radiation damage of Heavy ions and H irradiated Tungsten – Some Experimental Results

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2^{nd} RCM of the CRP on Irradiated Tungsten, Seoul, Korea, 8-11 Sept. 2015

Plan

- Objective
- Experimental details
- Characterization of Un-irradiated samples
- Characterization of Irradiated samples

  XRD, PAS, ERDA, AFM, SEM (Stress, Defects, H retention, morphological changes)
- Summary

Objective

1. To study the Structural , microstructural and Morphological changes in Tungsten after low (0.1 MeV), Medium (7.5 MeV) and High (80 MeV) energy Heavy ion irradiations.

2. Hydrogen trapped/retained in un-irradiated and pre-irradiated Tungsten

Experimental Details

- Two types of samples are used for Experiments

1. Samples cut from plate supplied by Plansee –

Sample size : 8mm x 8 mm x 2 mm and 10 mm x10mm x 2mm

Samples cut out from Hot rolled sheet (5 mm thick), 99.95 % pure, Rolling direction parallel to the plate surface and final sample surface

2. Samples provided by Dr. Linke (Supplier Plansee) –

Sample size : 8mm x 8 mm x 2 mm

Samples cut from a Rod , 99.97 % pure, elongated grains parallel to the rod axis and perpendicular to final sample surface

- Heavy ions Au and W have been used for irradiation with 80 MeV, 7.5 MeV and 100 KeV in Pristine and Annealed (at 900C) samples.

Ion Irradiation in Tungsten

- **The 15 UD pelletron at IUAC, New Delhi –**

Energy – 80 MeV, Charge state – 7 $^{+}$					
SL No	**Ions**	**Atomic Mass**	**Sample treatment**	**Fluence (ions/cm^2) x 10^{14}**	**Current (nA)**
1	**W**	**184**	**Annealed 900 C**	**1**	**7.0**
2	**Au**	**197**	**Annealed 900 C**	**6.26**	**11.5**
3	**Au**	**197**	**Annealed 900 C**	**1**	**7.0**
4	**Au**	**197**	**Pristine**	**6.26**	**11.5**

Used for 7.5 MeV W in W

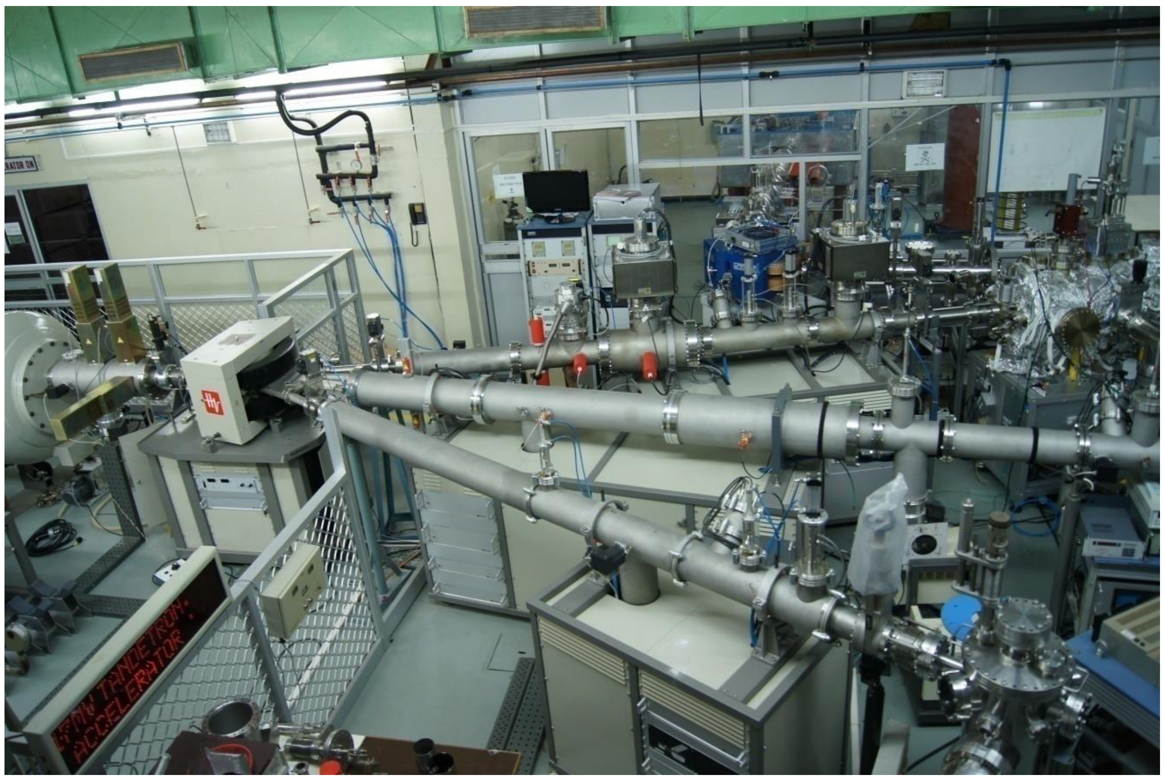


Illustration of the 1.7 MV tandetron accelerator at IGCAR along with the beam lines which is used for tungsten irradiation and ERDA analysis.

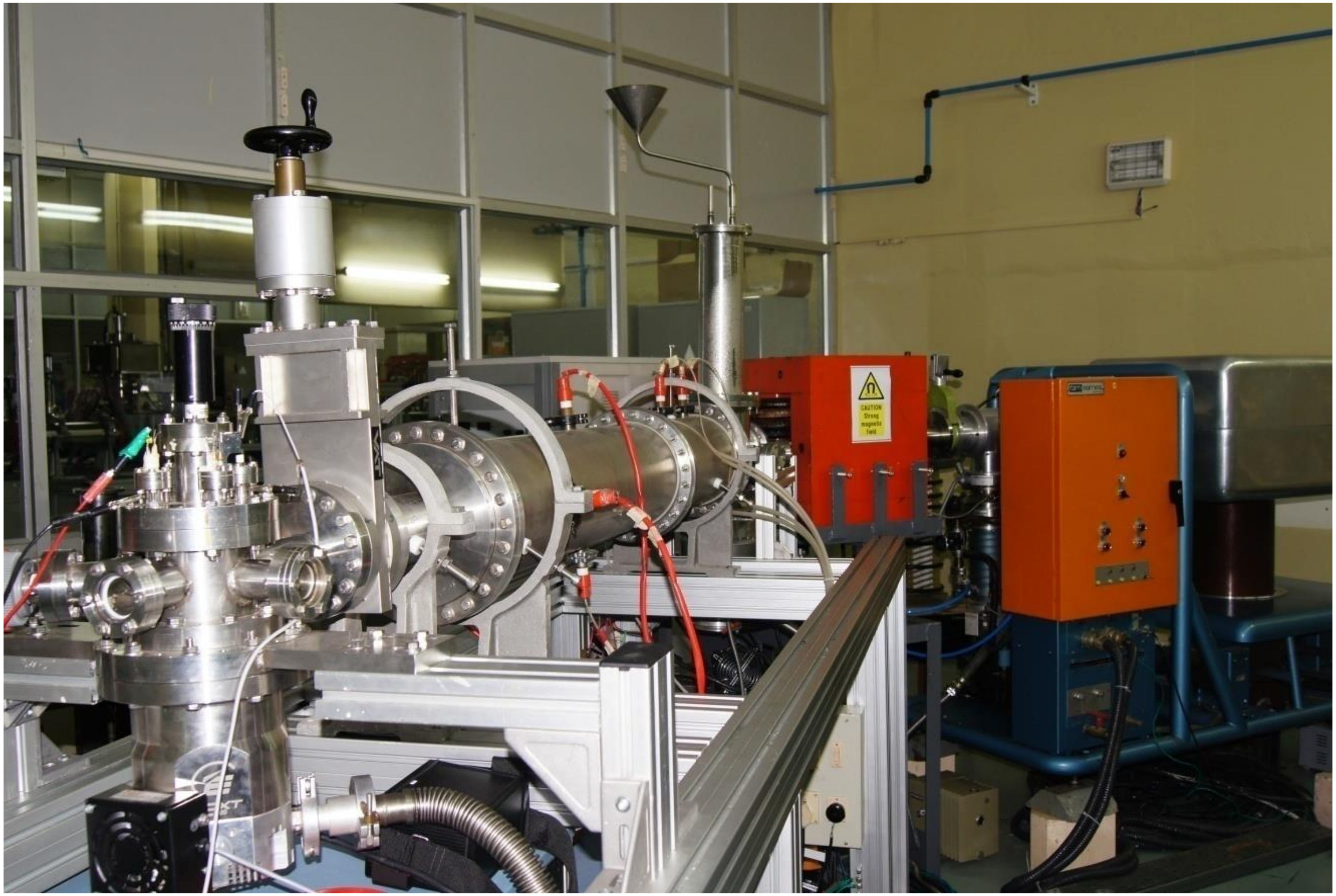


Illustration of the 150 kV accelerator along with the beam lines at IGCAR which is used for hydrogen implantation.

Characterization of Unirradiated samples

- Pristine and samples annealed at 900 C have been analyzed for their structure, microstructure, particularly for grain size and orientation using XRD and EBSD.

- SEM and AFM techniques have also been used for un-irradiated samples to compare the microstructural and morphological changes after irradiations.

EBSD Results

Sample ID: Linke. Scan on surface

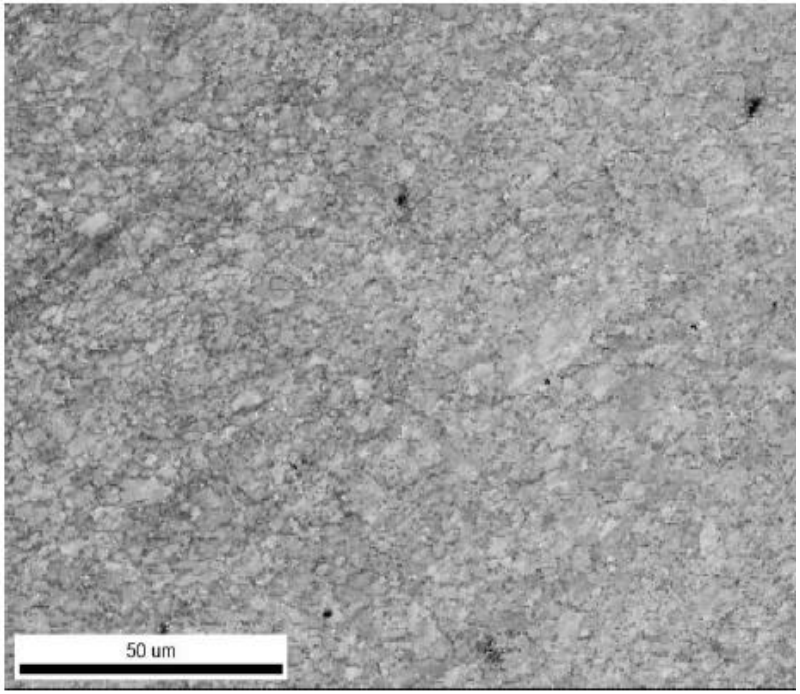
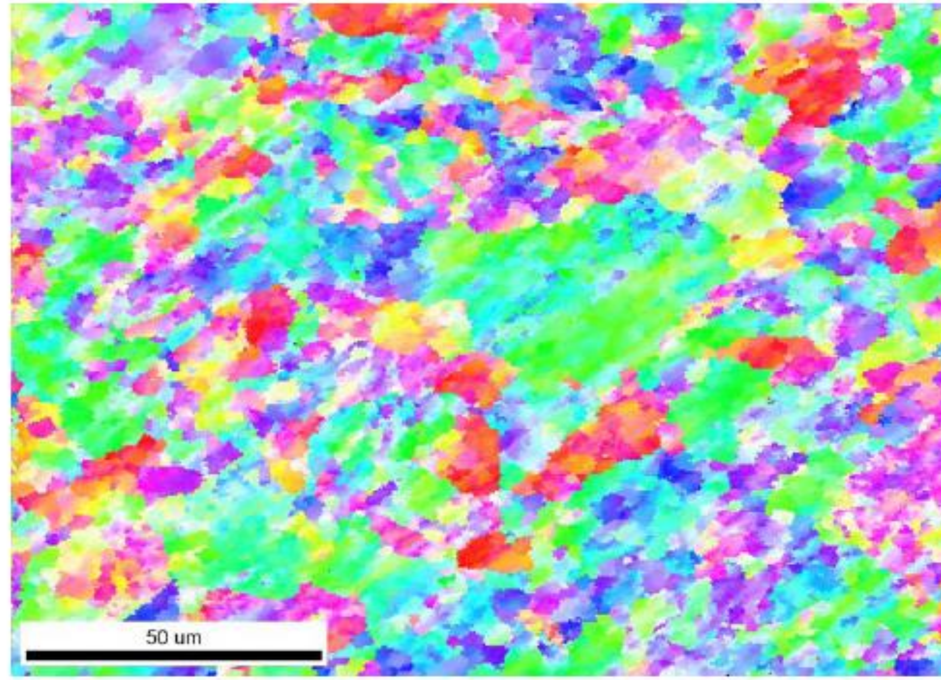
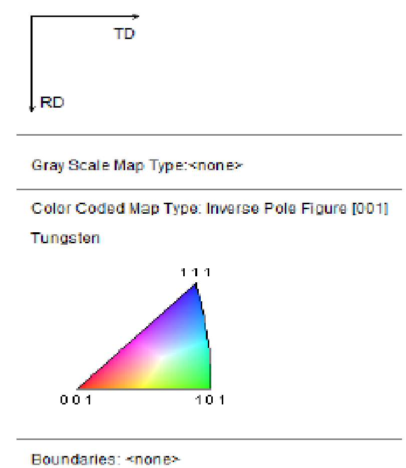


Image quality map



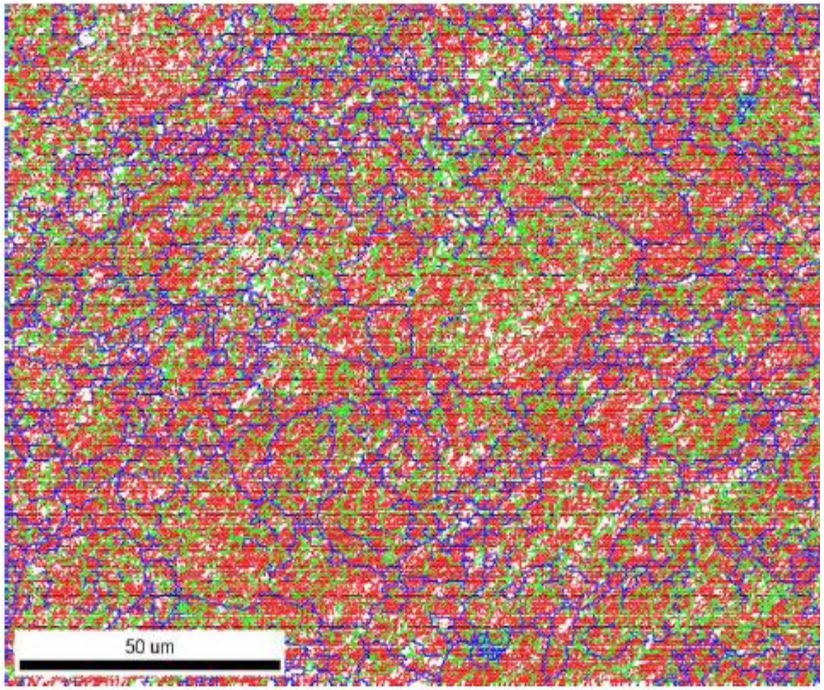
Crystal orientation map



Inverse pole figure

Observation: Grains are more equiaxed

Sample ID: Linke. Scan on surface



TD

RD

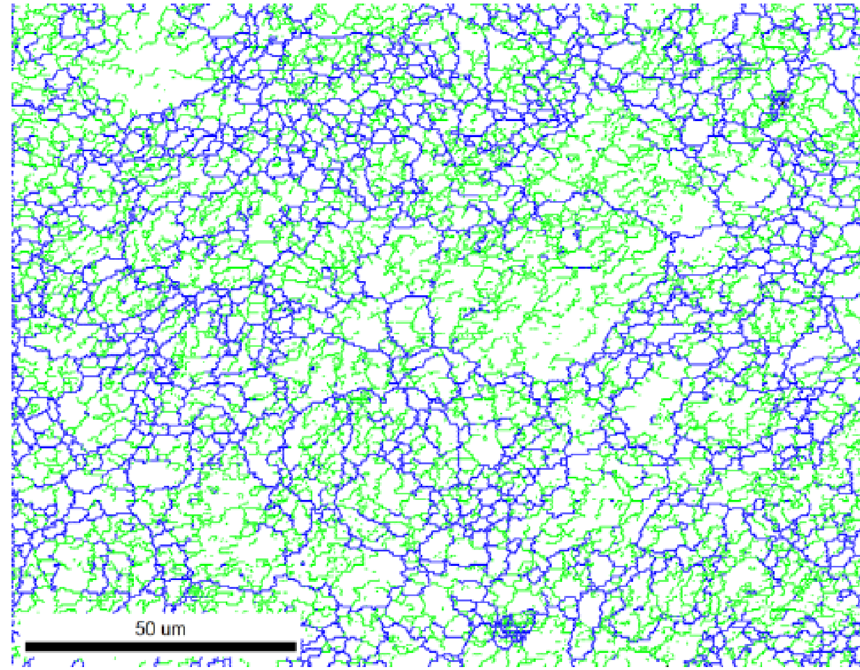
Gray Scale Map Type:<none>

Color Coded Map Type: <none>

Boundaries: Rotation Angle

	Min	Max	Fraction	Number	Length
red	1°	5°	0.657	132690	3.83 cm
green	5°	15°	0.196	39639	1.14 cm
blue	15°	180°	0.146	29543	8.53 mm

*For statistics - any point pair with misorientation exceeding 1° is considered a boundary total number = 201872, total length = 5.83 cm)

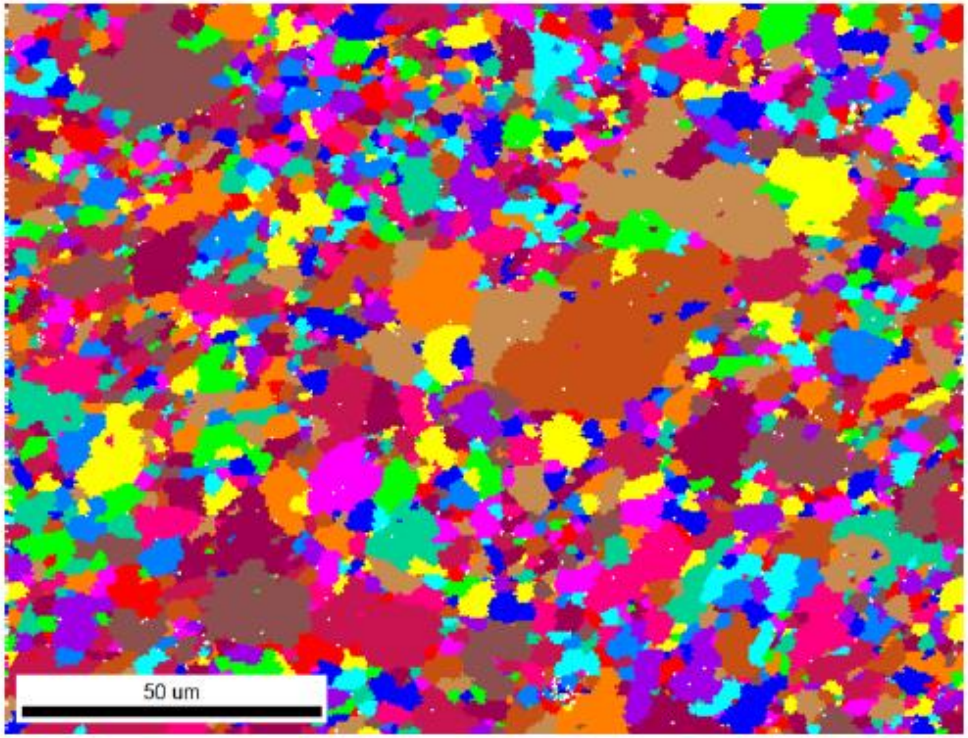


Grain boundary maps

Step size: 0.5 µm

Observation: Grains are more equiaxed

Sample ID: Linke. Scan on surface



Unique grain map

Step size: 0.5 µm

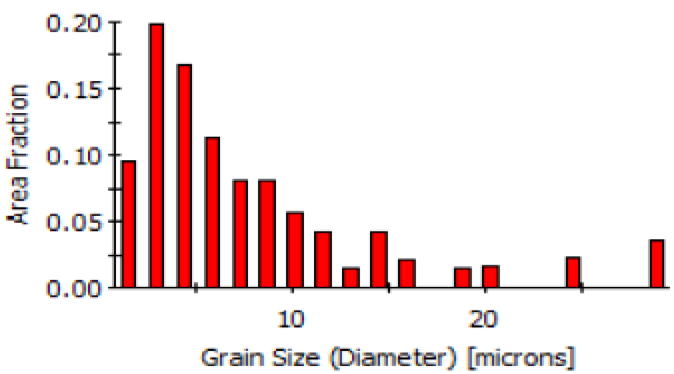
Chart: Grain Size (diameter)

Edge grains included in analysis

Diameter [microns]	Area Fraction
1.46298	0.0952196
2.90391	0.198588
4.34484	0.168422
5.78577	0.112625
7.2267	0.0814062
8.66763	0.0799982
10.1086	0.0558302
11.5495	0.0421055
12.9904	0.0139465
14.4313	0.041141
15.8723	0.0209862
17.3132	0
18.7541	0.0149332
20.1951	0.01684
21.636	0
23.0769	0
24.5178	0.0228155
25.9588	0
27.3997	0
28.8406	0.0351433

Average	
Number	2.57584
Area	7.73768

Grain Size (diameter)



Observation: Grains are more equiaxed

Sample ID: Linke. Scan-1 on cross-section

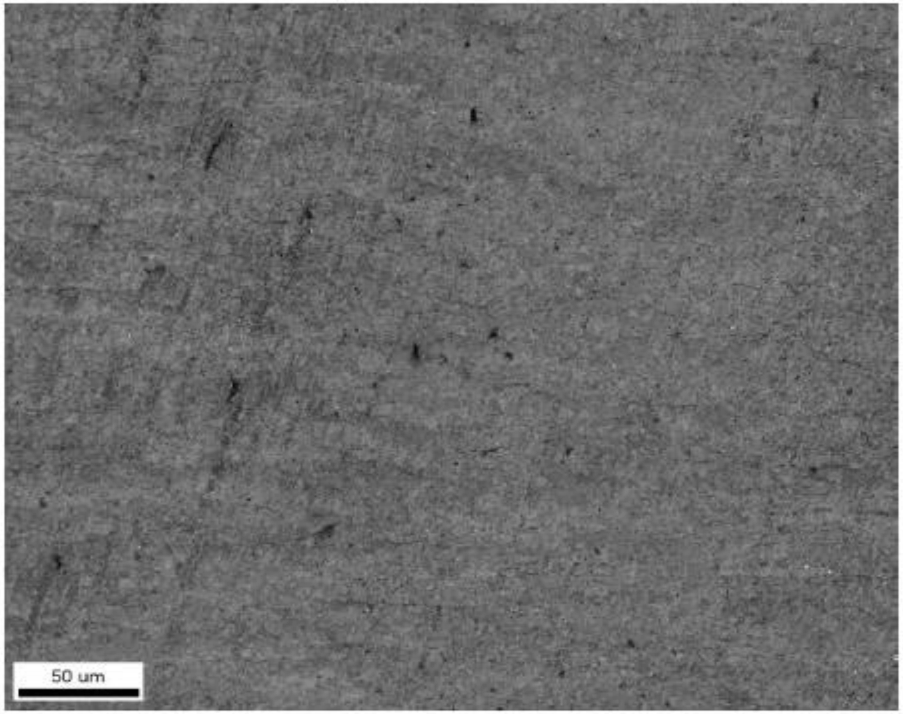
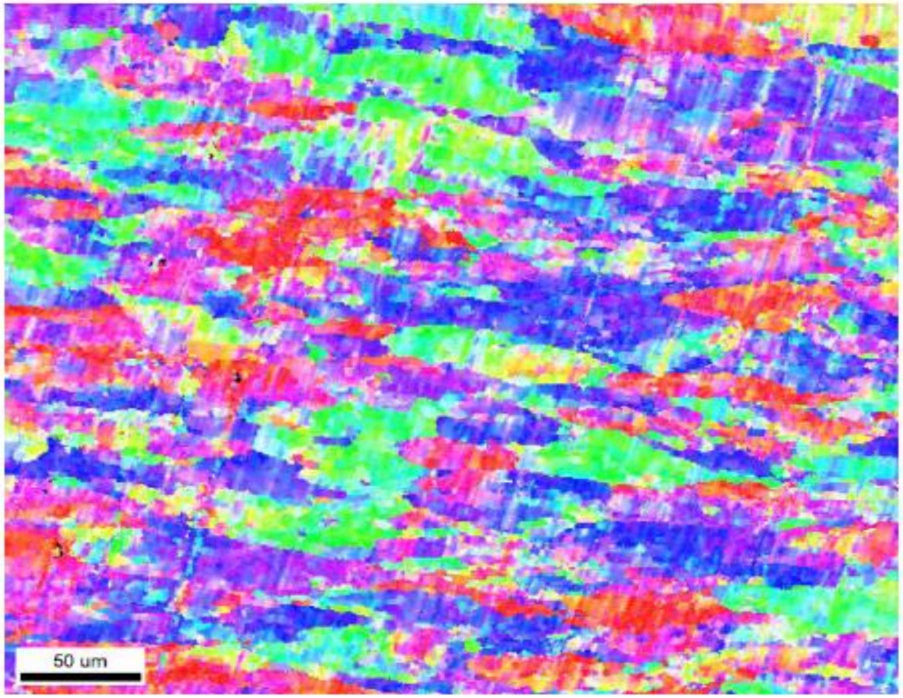


Image quality map

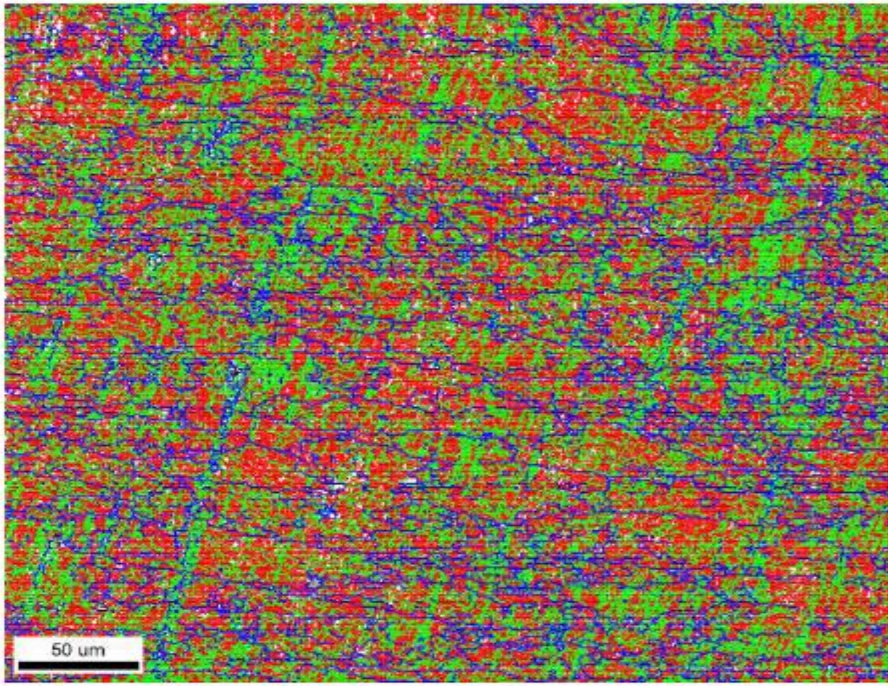


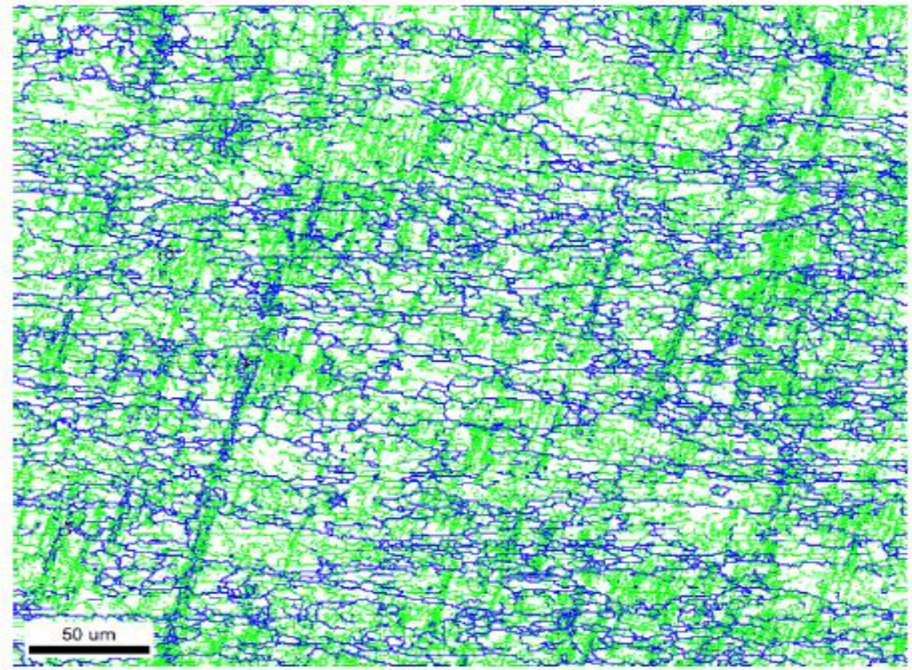
Crystal orientation map

Step size: 1 μm

The columnar nature of the grains is evident in these maps

Sample ID: Linke. Scan-1 on cross-section





Gray Scale Map Type:<none>

Color Coded Map Type: <none>

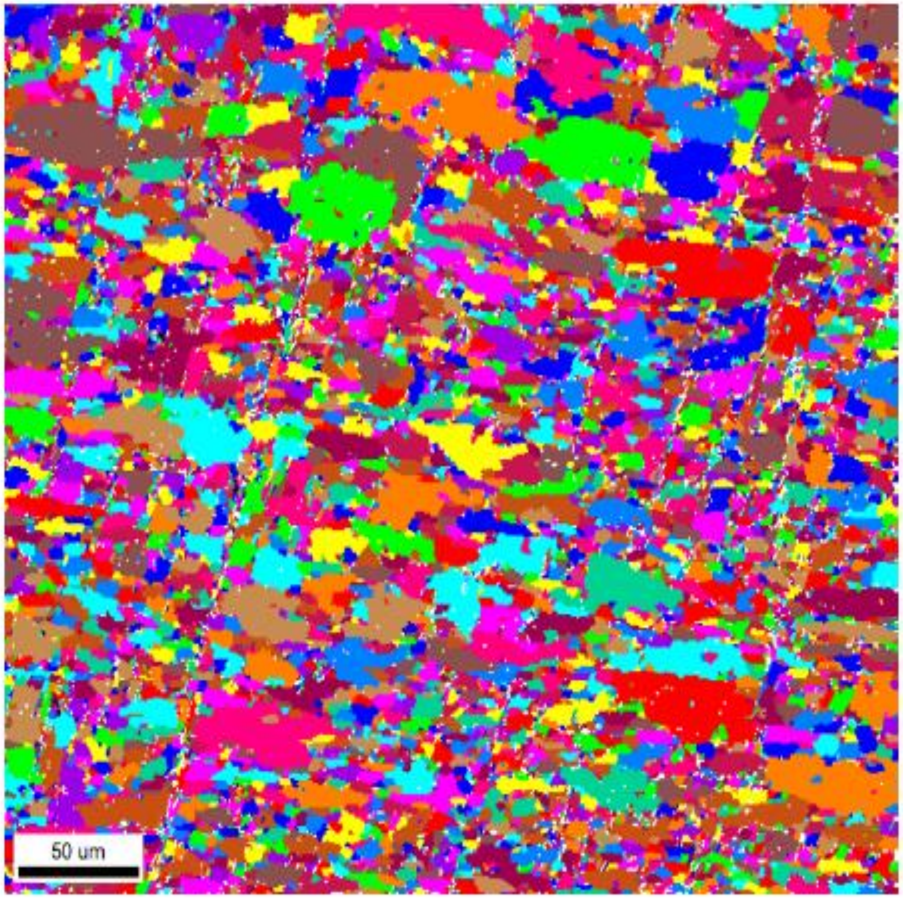
Boundaries: Rotation Angle

	Min	Max	Fraction	Number	Length
red	1°	5°	0.567	212718	12.28 cm
green	5°	15°	0.283	106103	6.13 cm
blue	15°	180°	0.151	56526	3.26 cm

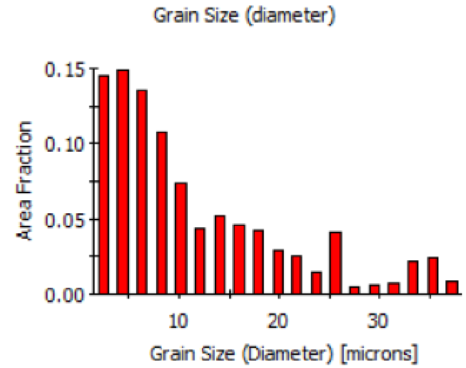
*For statistics - any point pair with misorientation exceeding 1° is considered a boundary
total number = 375347, total length = 21.67 cm)

The columnar nature of the grains is evident in these maps

Sample ID: Linke. Scan-1 on cross-section



Unique grain map



Step size: 1 µm

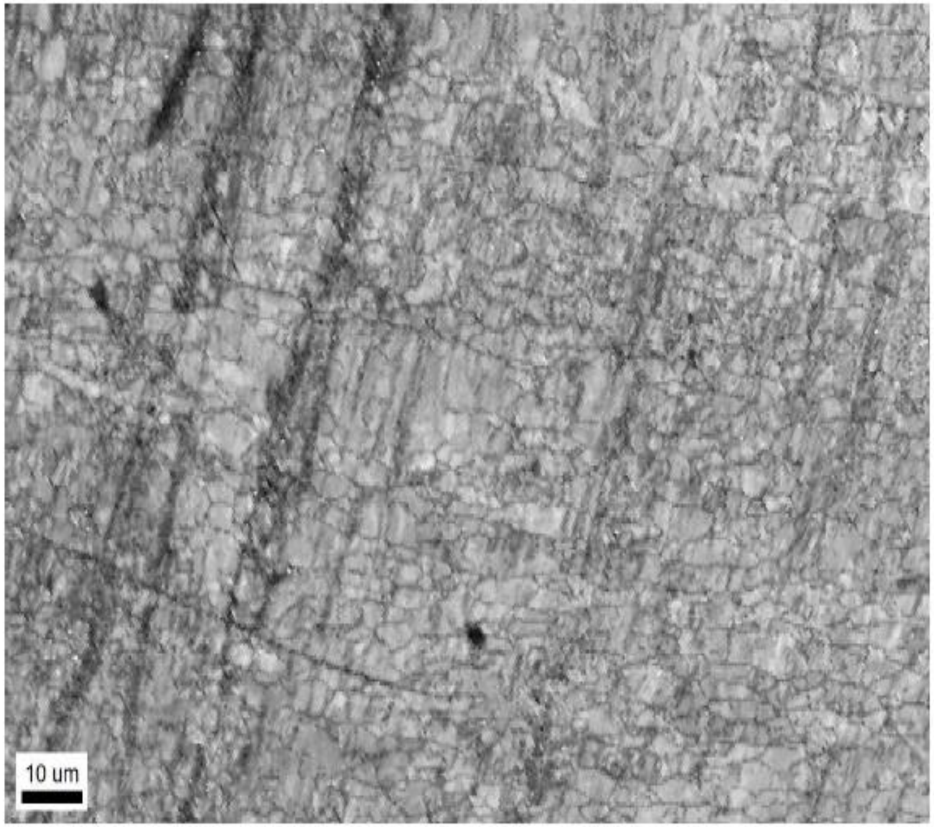
Chart: Grain Size (diameter)

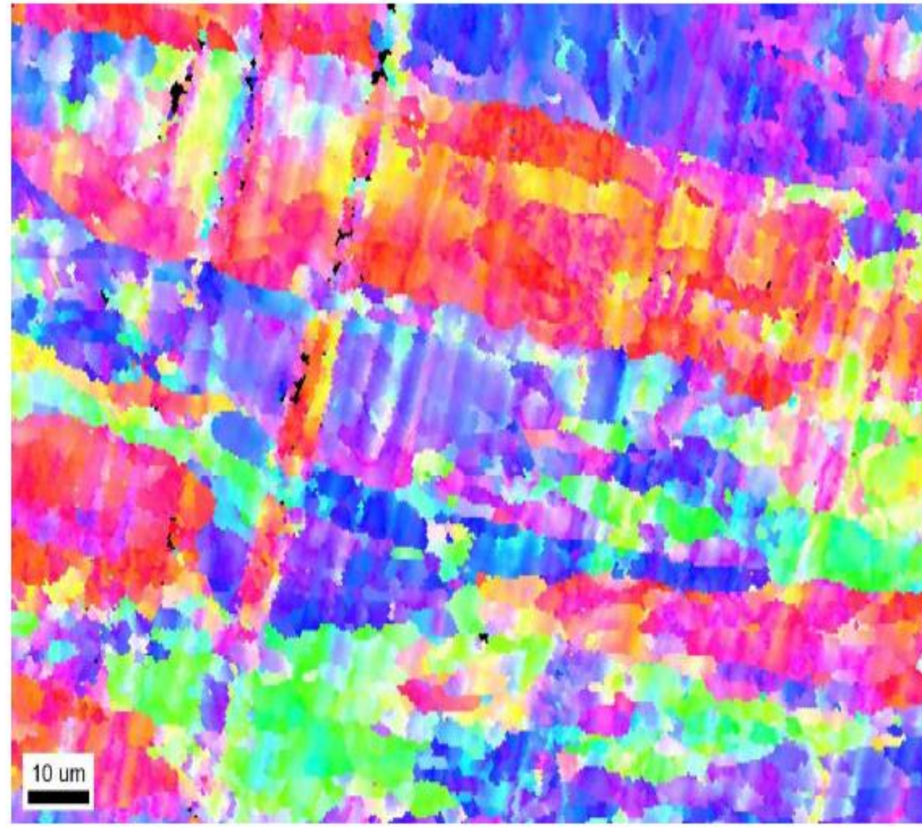
Edge grains included in analysis

Diameter [microns]	Area Fraction
2.44927	0.145318
4.37775	0.149294
6.30623	0.136018
8.2347	0.107789
10.1632	0.0740054
12.0917	0.0430326
14.0201	0.0521553
15.9486	0.0454802
17.8771	0.0421426
19.8056	0.0294598
21.7341	0.025336
23.6625	0.0146409
25.591	0.0408001
27.5195	0.00540689
29.448	0.00559973
31.3764	0.00697185
33.3049	0.0220651
35.2334	0.0243125
37.1619	0.00889281
39.0904	0.021279

Average	
Number	3.46422
Area	12.0068

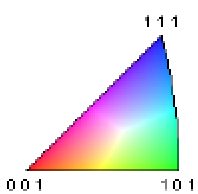
Sample ID: Linke. Scan-2 on cross-section





Color Coded Map Type: Inverse Pole Figure [001]

Tungsten

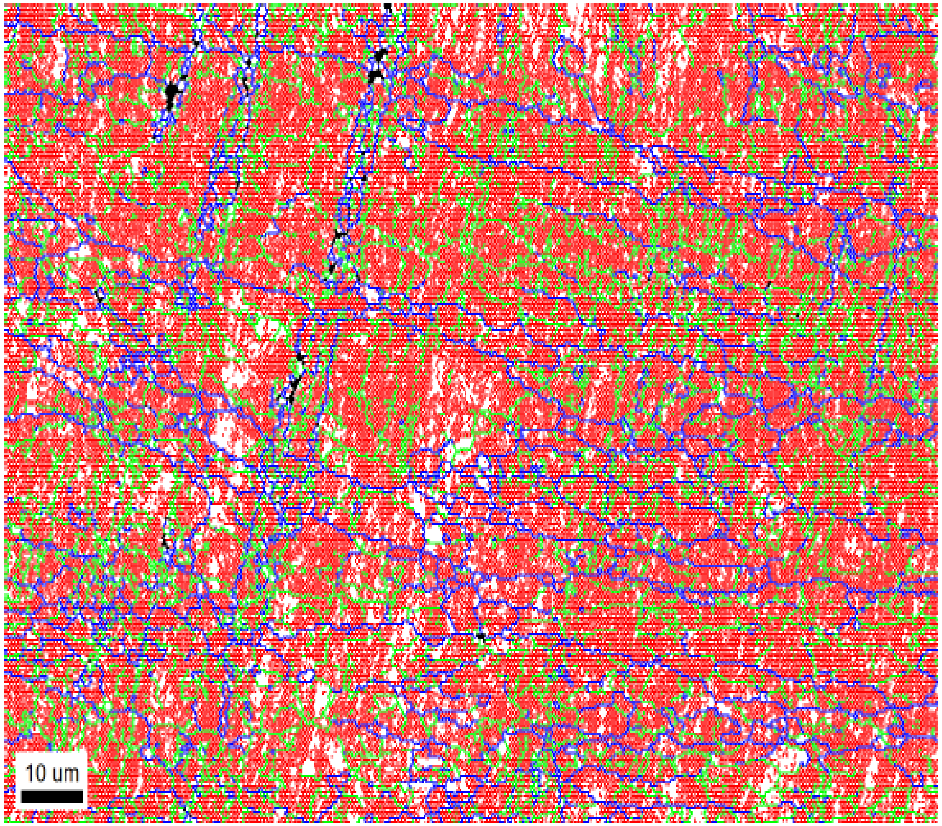


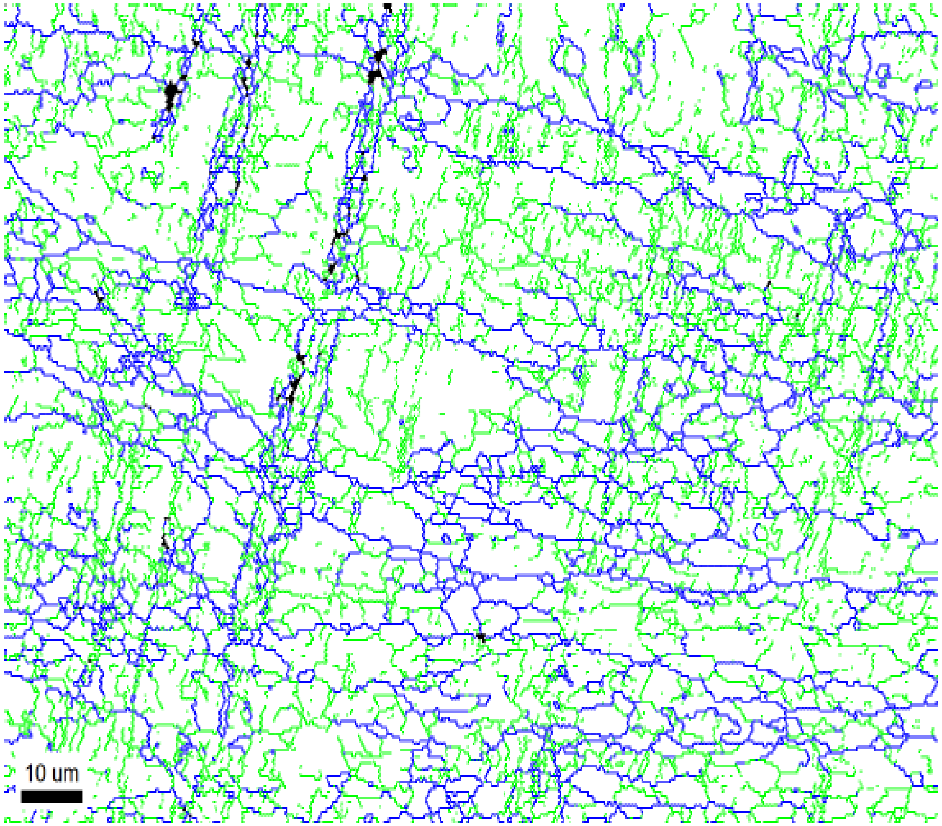
Boundaries: <none>

Inverse pole figure

The columnar nature of the grains is evident in these maps

Sample ID: Linke. Scan-2 on cross-section





Grain boundary maps

Boundaries: Rotation Angle

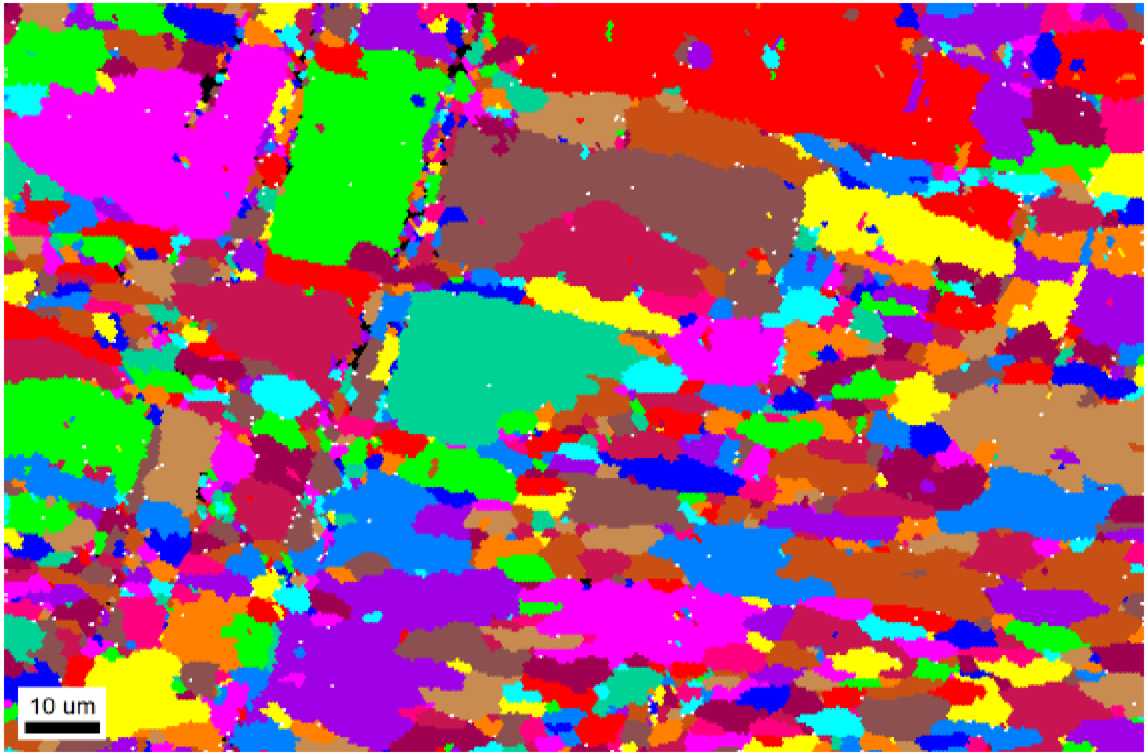
	Min	Max	Fraction	Number	Length
red	1°	5°	0.720	125967	3.64 cm
green	5°	15°	0.168	29407	8.49 mm
blue	15°	180°	0.112	19607	5.66 mm

*For statistics - any point pair with misorientation exceeding 1° is considered a boundary total number = 174981, total length = 5.05 cm)

Step size: 0.5 µm

The columnar nature of the grains is evident in these maps

Sample ID: Linke. Scan-2 on cross-section



Unique grain map

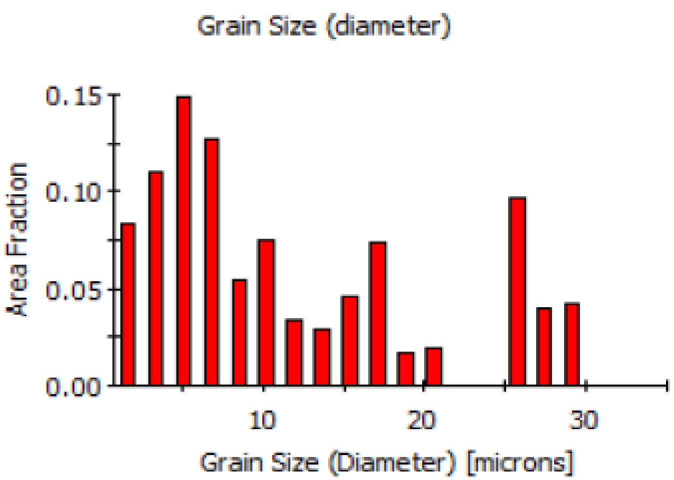
Chart: Grain Size (diameter)

Edge grains included in analysis

Diameter [microns]	Area Fraction
1.60417	0.0832771
3.32749	0.110586
5.0508	0.149069
6.77412	0.12741
8.49743	0.0543009
10.2207	0.0754509
11.9441	0.03446
13.6674	0.0291552
15.3907	0.0460201
17.114	0.0735908
18.8373	0.01754
20.5606	0.0195792
22.2839	0
24.0073	0
25.7306	0.096656
27.4539	0.0399713
29.1772	0.0429337
30.9005	0
32.6238	0
34.3471	0

Average	
Number	2.71367
Area	11.6825

Step size: 0.5 µm



Plansee samples- Surface scans

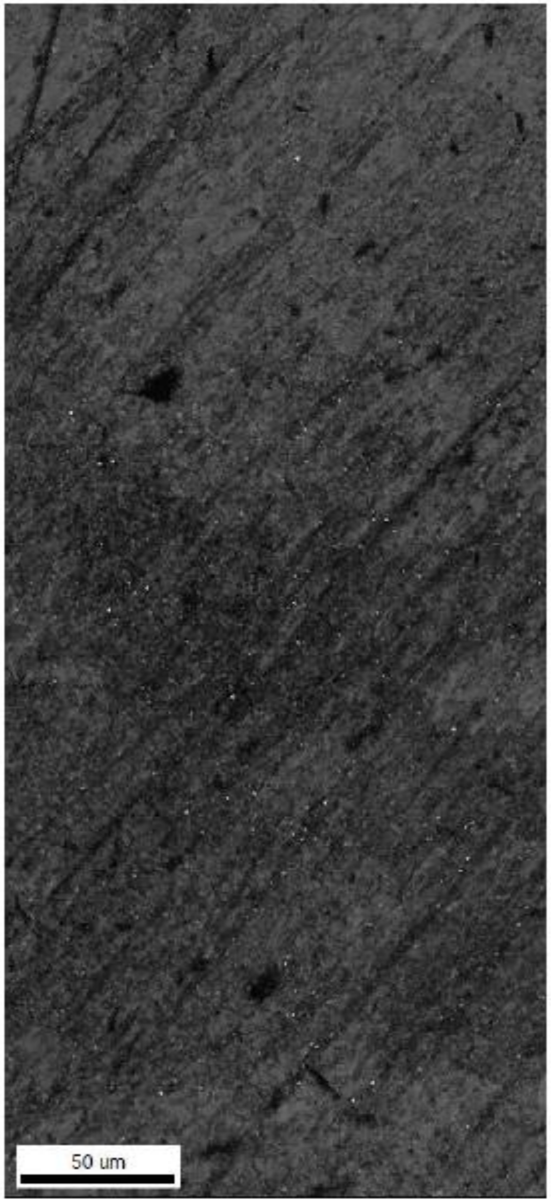
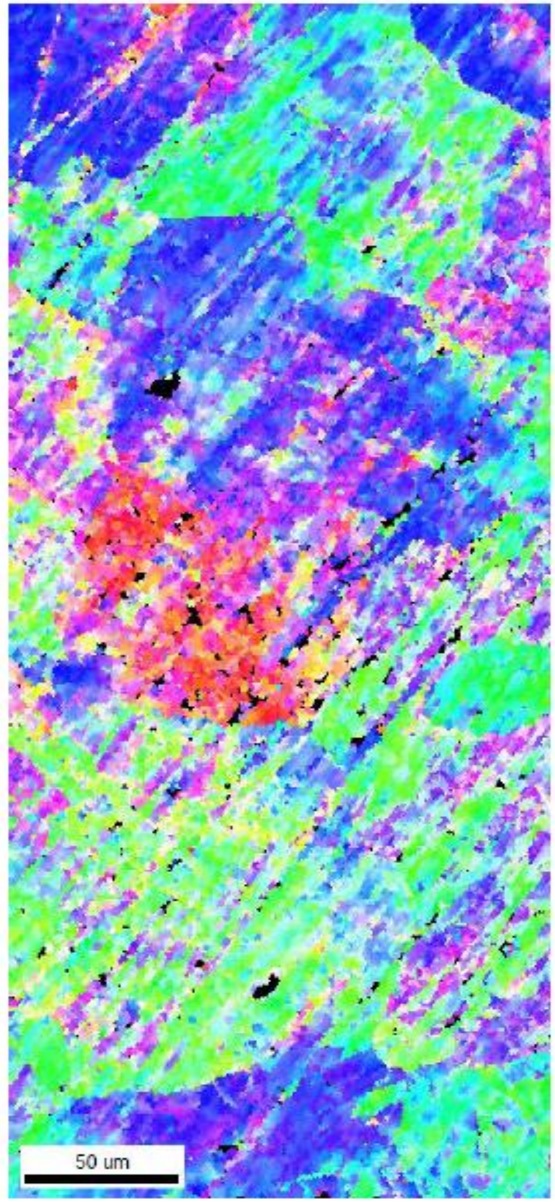
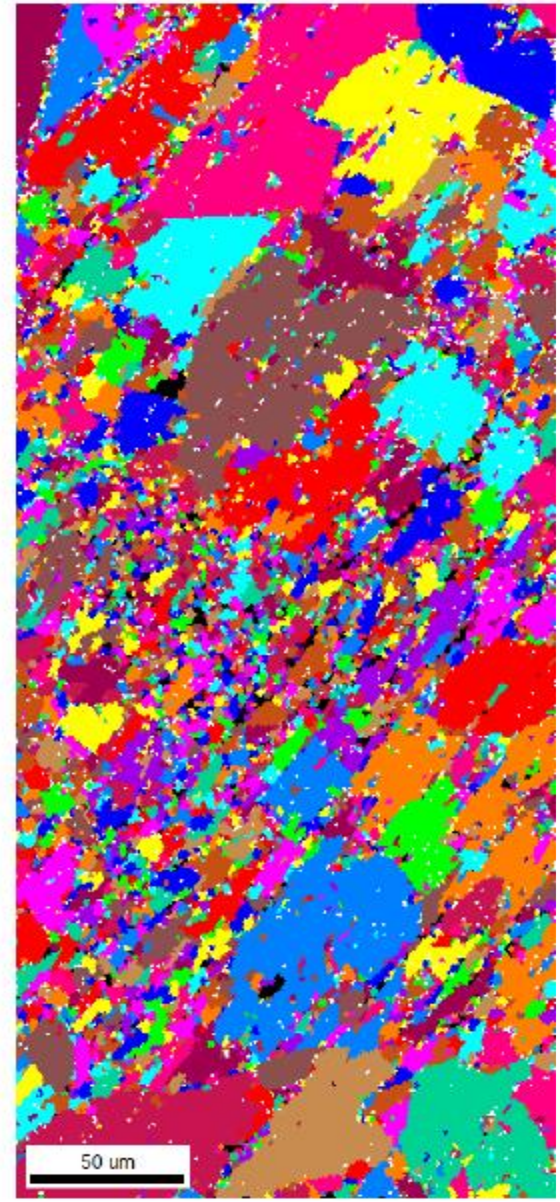


Image quality map



Crystal orientation map

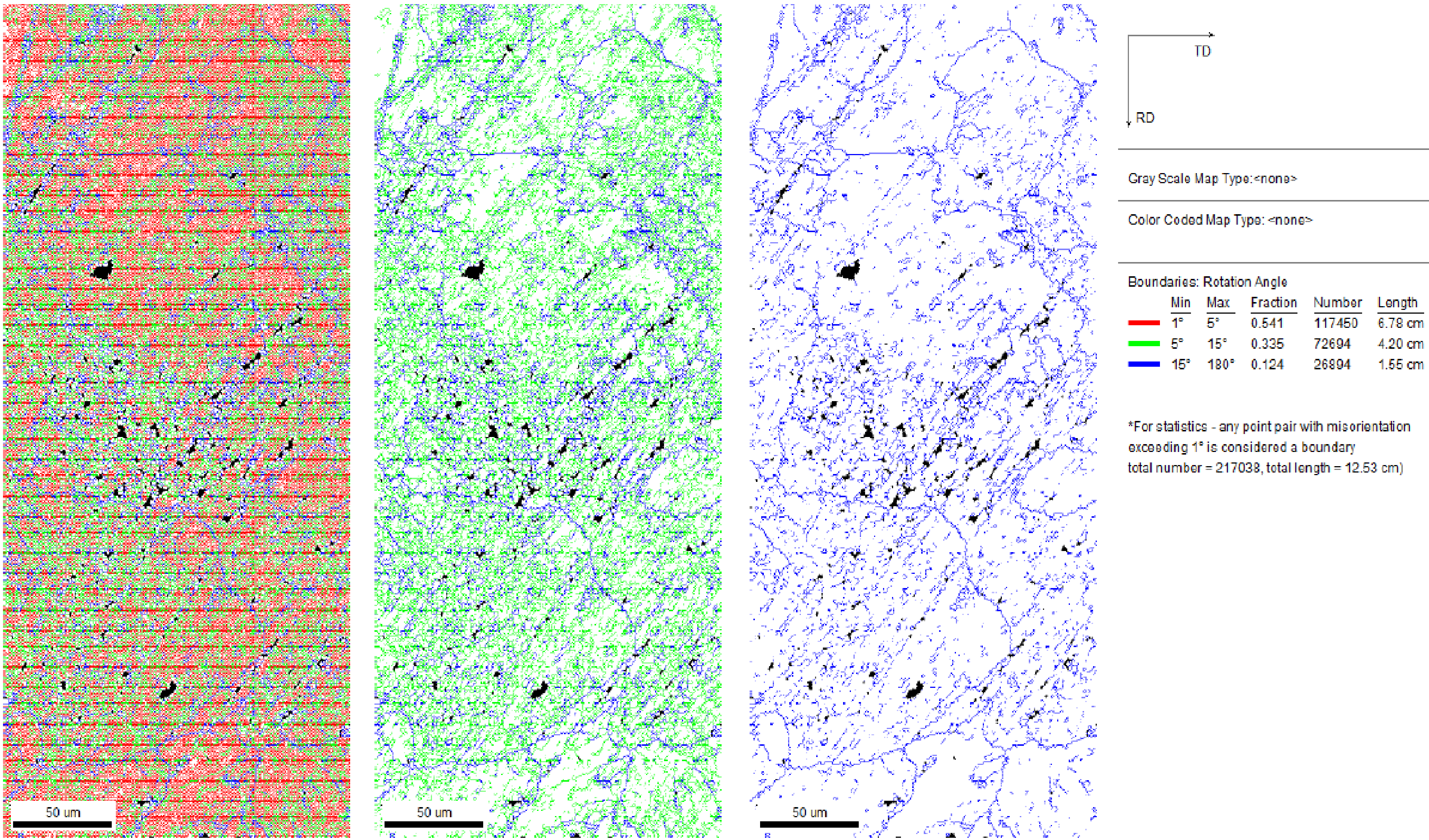


Unique grain map

Larger grains compared to the sample marked'Linke'

Step size: 1 µm

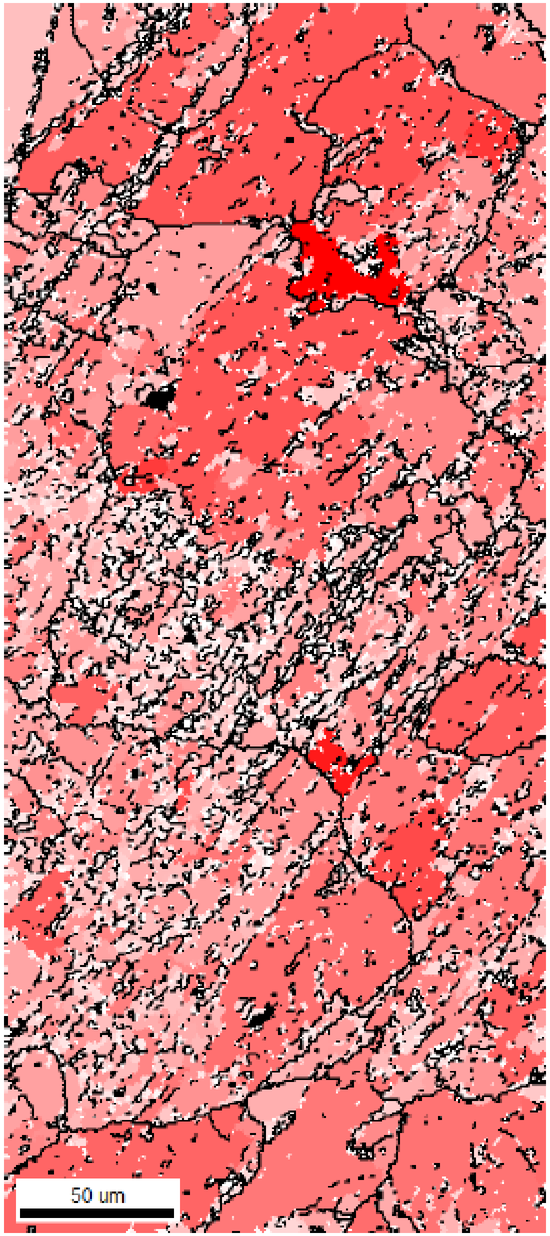
Plansee samples- Surface scans



Grain boundary maps

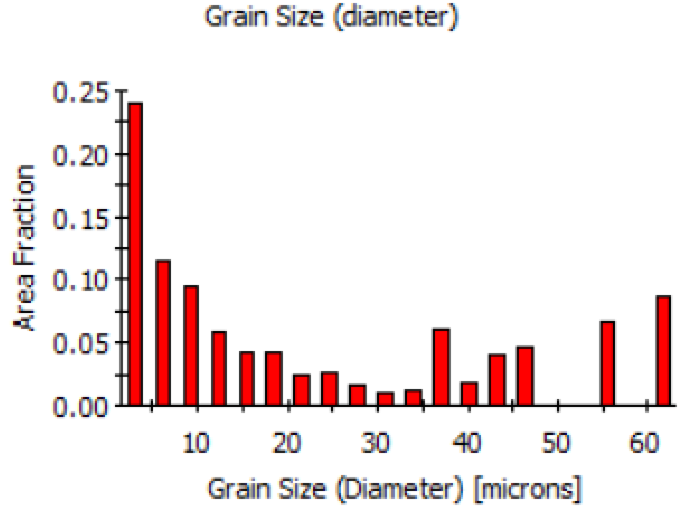
Larger grains compared to the sample marked'Linke'

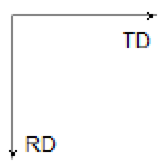
Plansee samples- Surface scans



Grain orientation spread

High angle grain boundaries





Gray Scale Map Type: <none>

Color Coded Map Type: Grain Orientation Spread

	Min	Max	Total Fraction	Partition Fraction
	0	13.8584	0.959	0.970

Boundaries: Rotation Angle

	Min	Max	Fraction	Number	Length
	15°	180°	0.124	26894	1.55 cm

*For statistics - any point pair with misorientation exceeding 1° is considered a boundary
total number = 217041, total length = 12.53 cm)

Chart: Grain Size (diameter)

Edge grains included in analysis

Diameter [microns]	Area Fraction
3.02913	0.240294
6.11734	0.114966
9.20554	0.0941945
12.2937	0.0592462
15.3819	0.041796
18.4701	0.0429032
21.5584	0.0234072
24.6466	0.0254038
27.7348	0.0168003
30.823	0.00953137
33.9112	0.0115171
36.9994	0.0600645
40.0876	0.0180037
43.1758	0.0409536
46.264	0.0473319
49.3522	0
52.4404	0
55.5286	0.0661299
58.6168	0
61.705	0.0864563

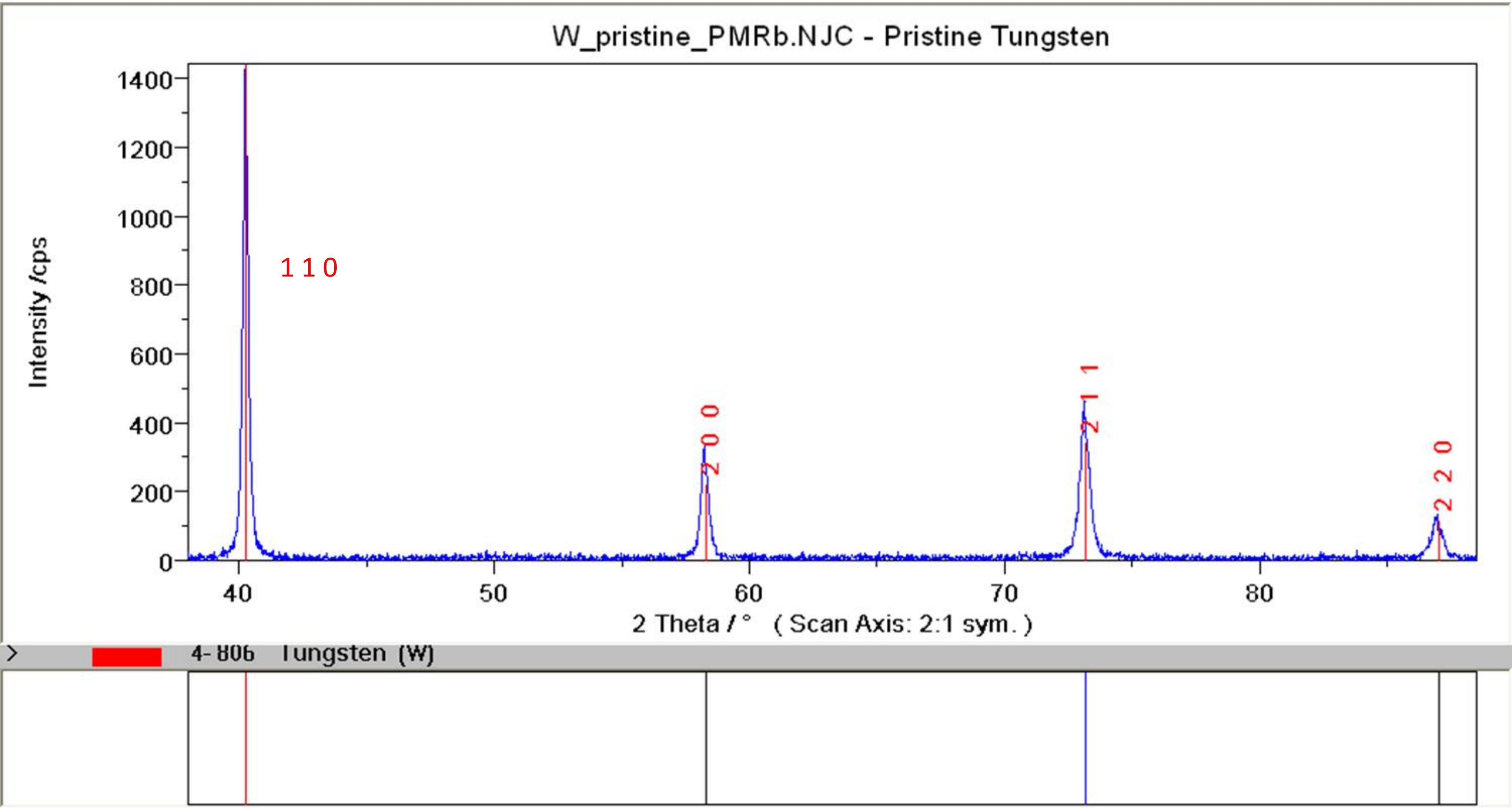
Average	
Number	3.01253
Area	22.6512

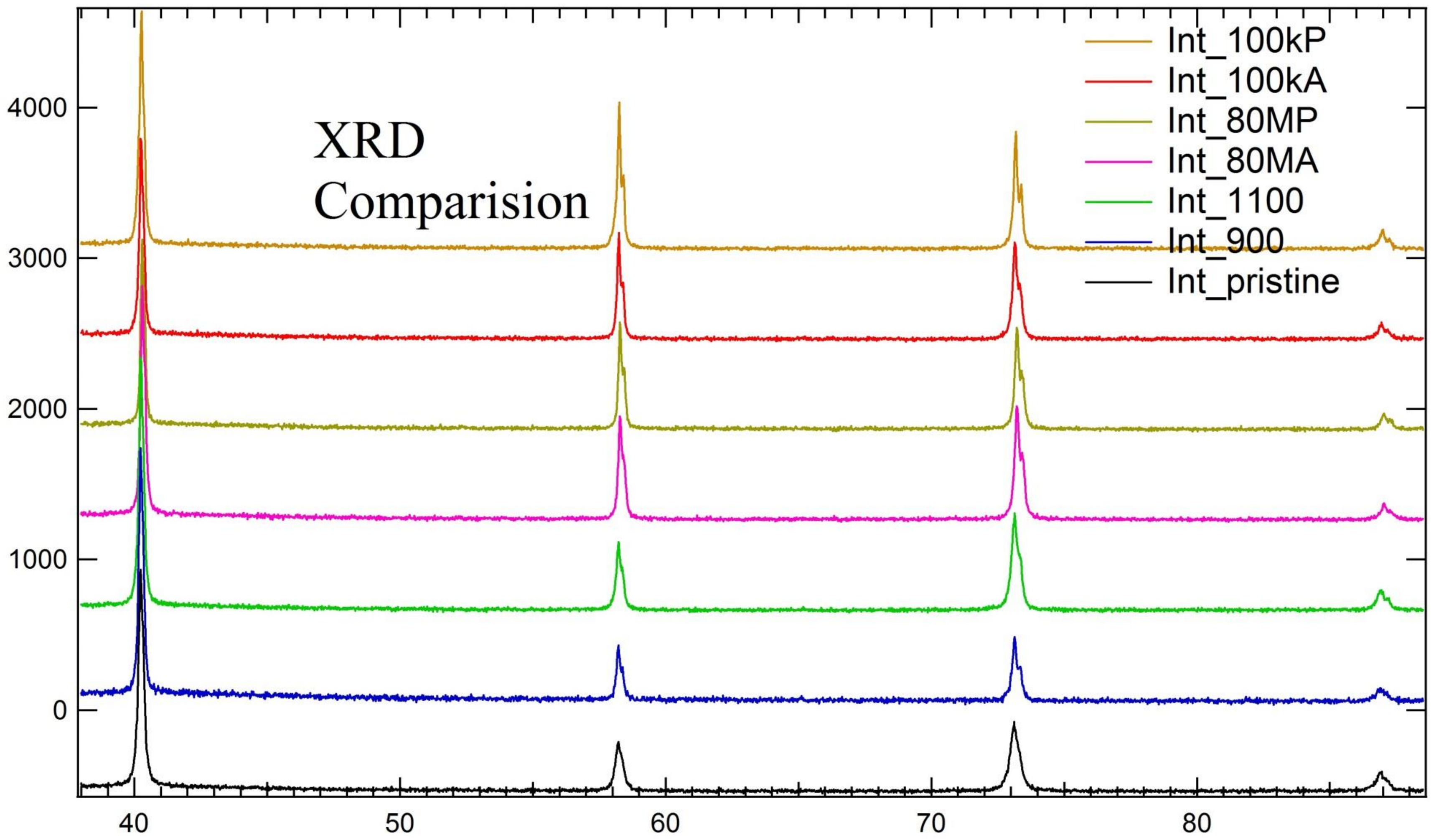
Strained Lattice

XRD study

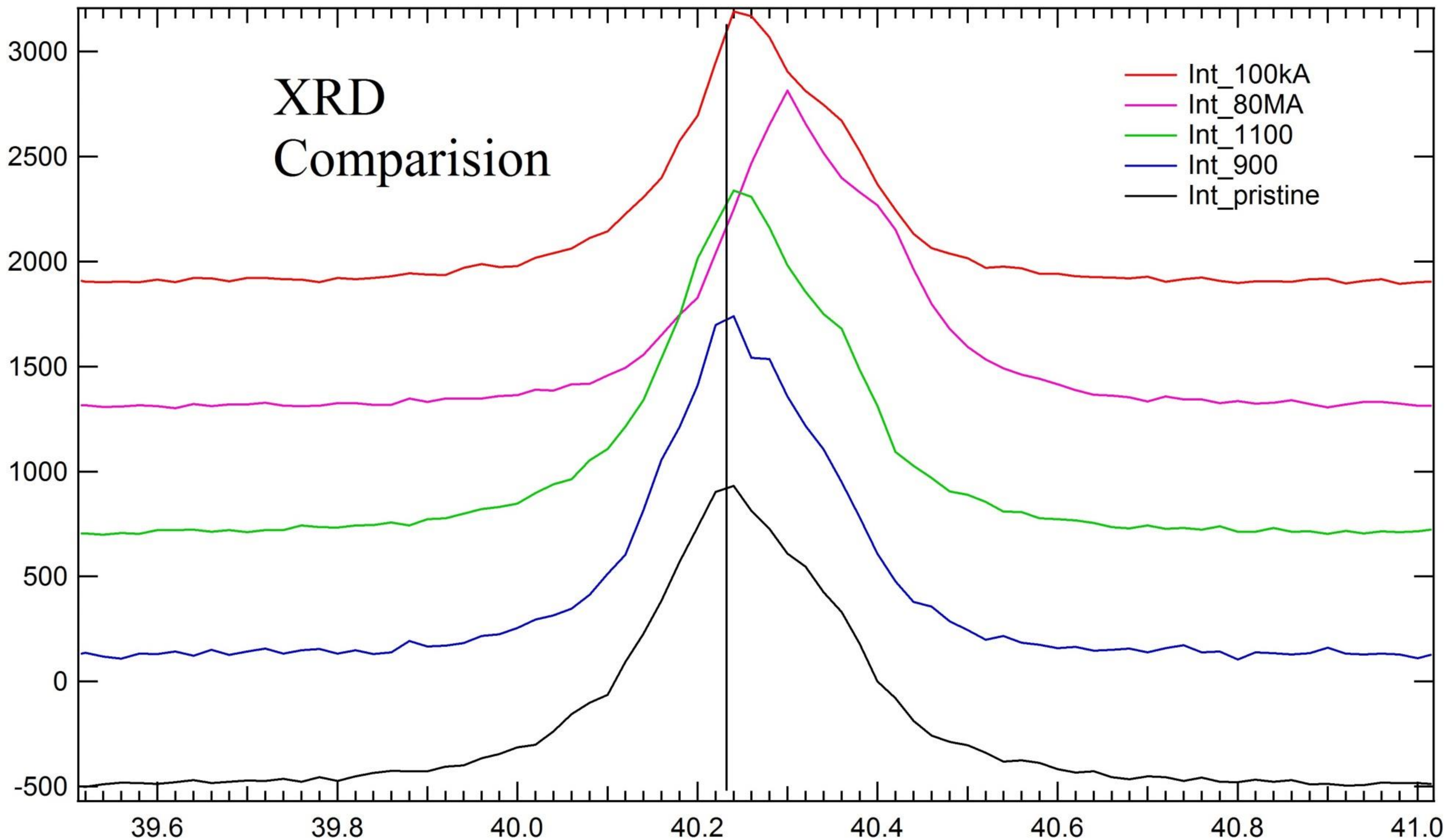
- XRD spectra were recorded for the Pristine and Annealed Tungsten samples irradiated with 80 MeV and 100 KeV and compared with the Pristine and annealed (900 C and 1100 C) samples.

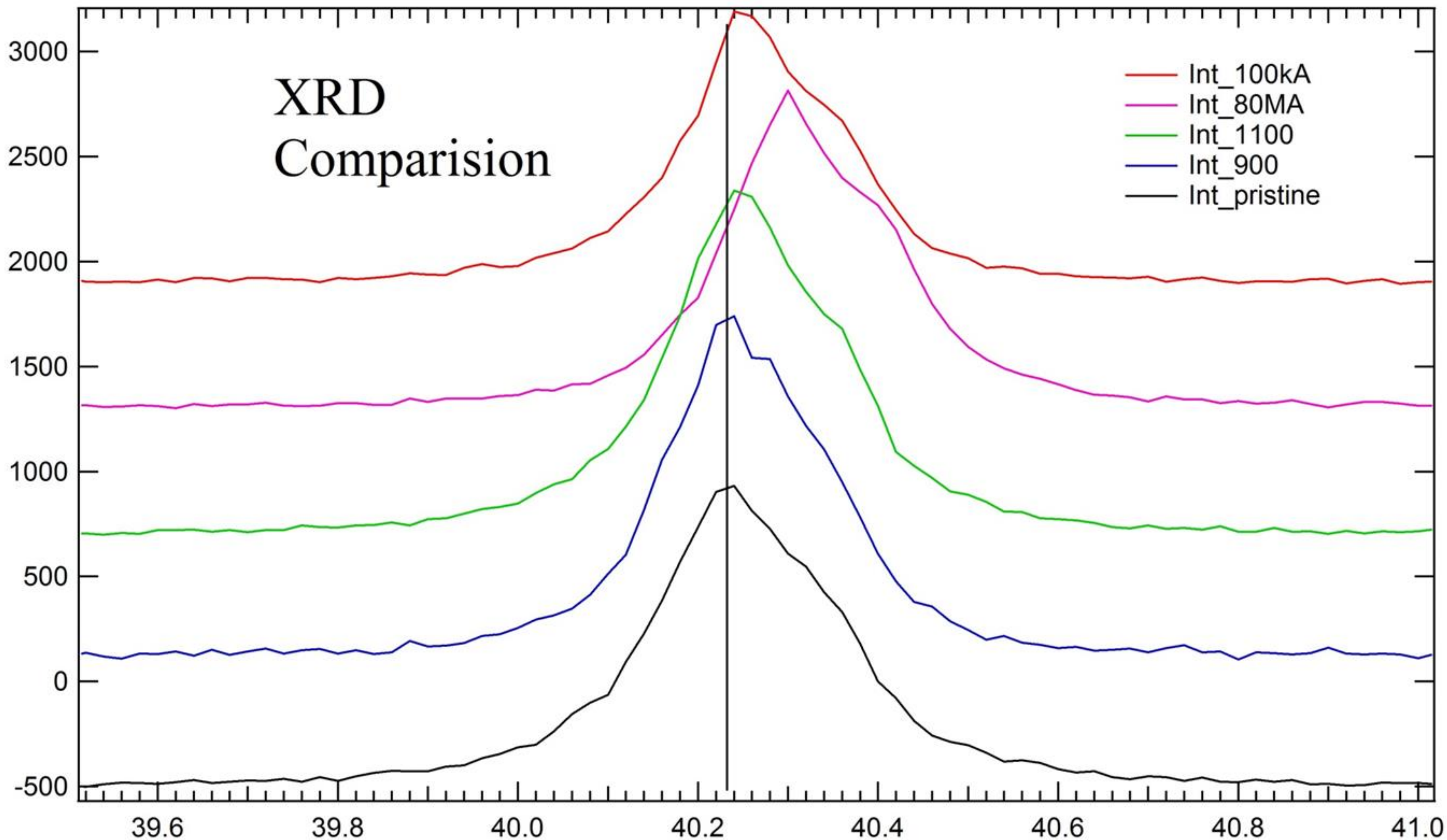
- The changes in the Relative Peak Intensity, Peak Width and peak positions are attributed to Crystallographic Texture, Micro-strain and Macro-strian respectively.

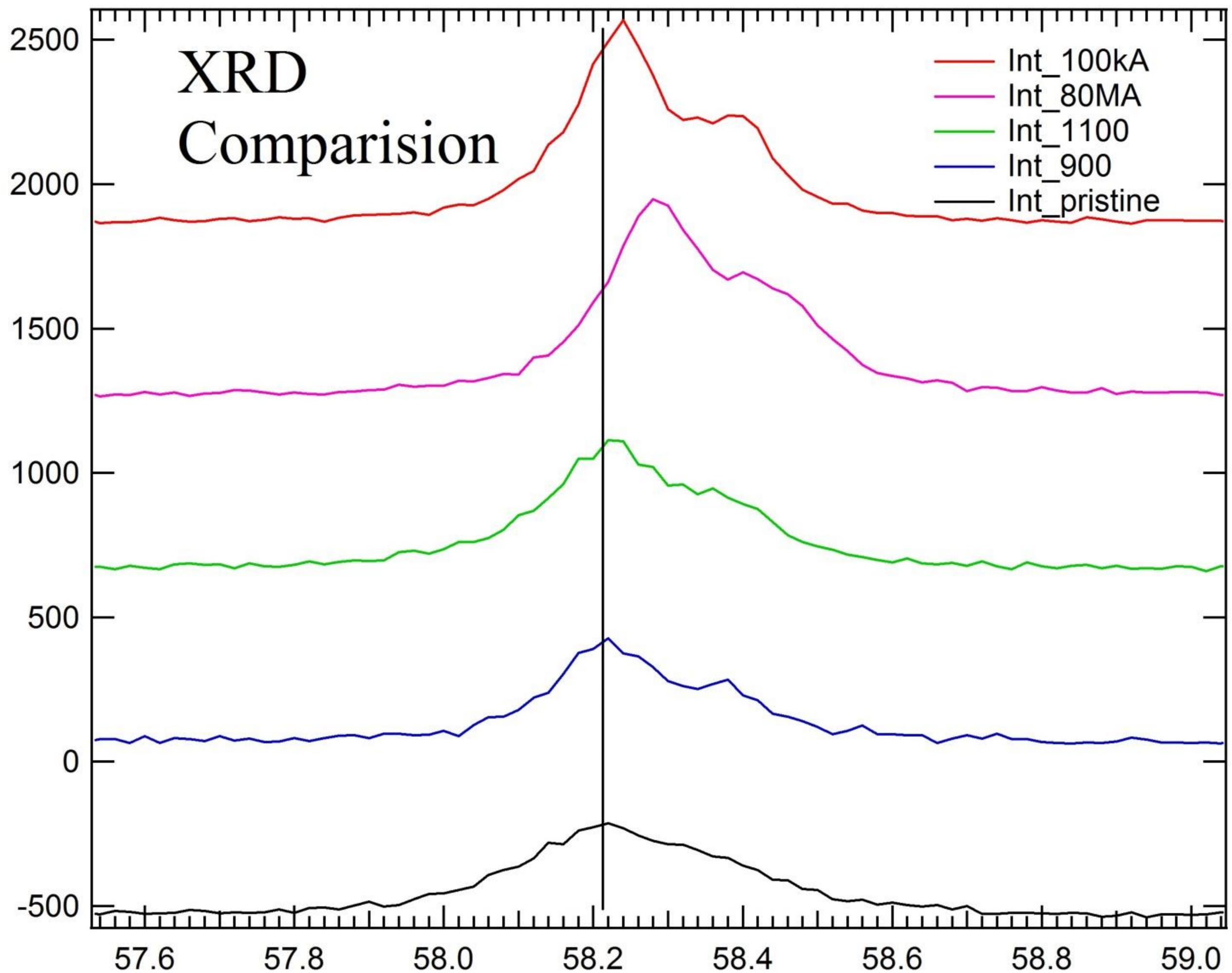

W_pristine_PMRb.NJC - Pristine Tungsten
Intensity /cps
1400
1200
1000
800
600
400
200
0
1 1 0
2 0 0
2 1 1
2 2 0
40
50
60
70
80
2 Theta / °   (Scan Axis: 2:1 sym.)
4- 806   Tungsten (W)

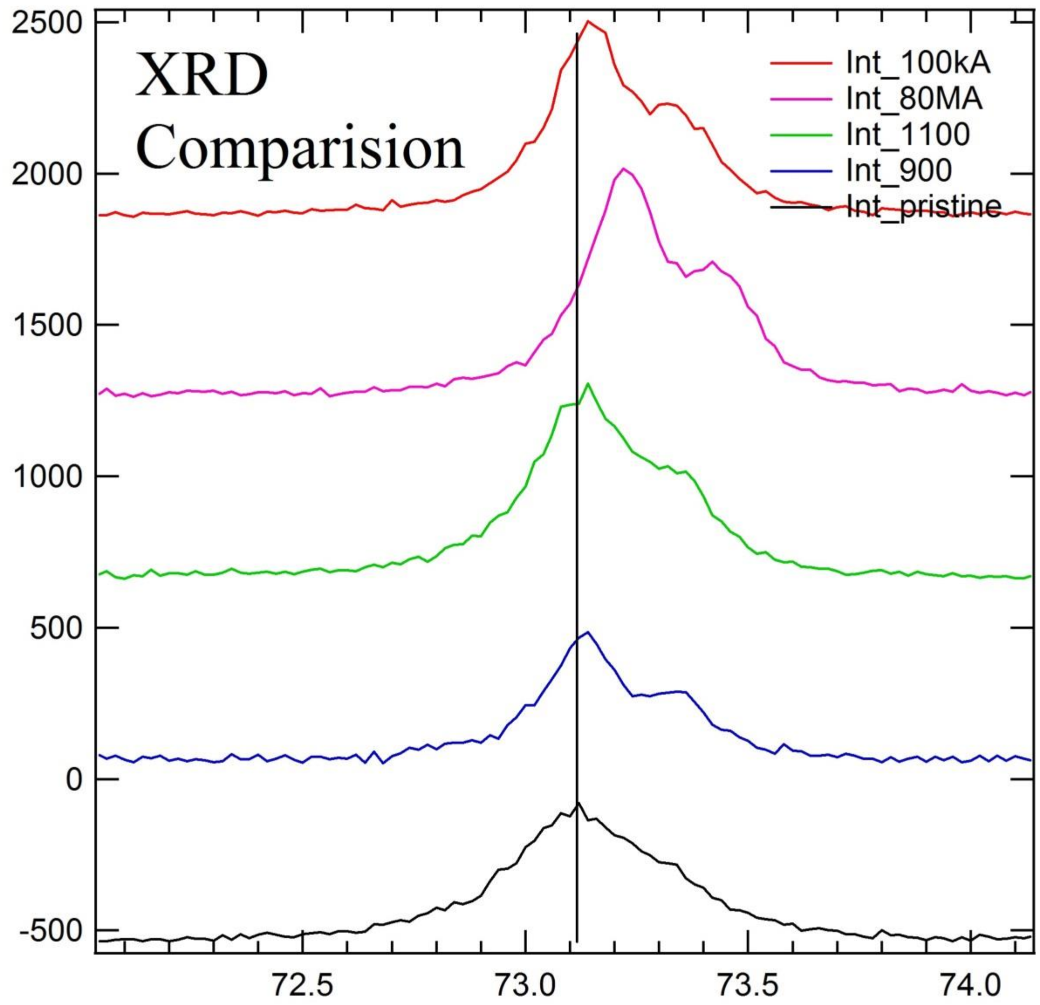


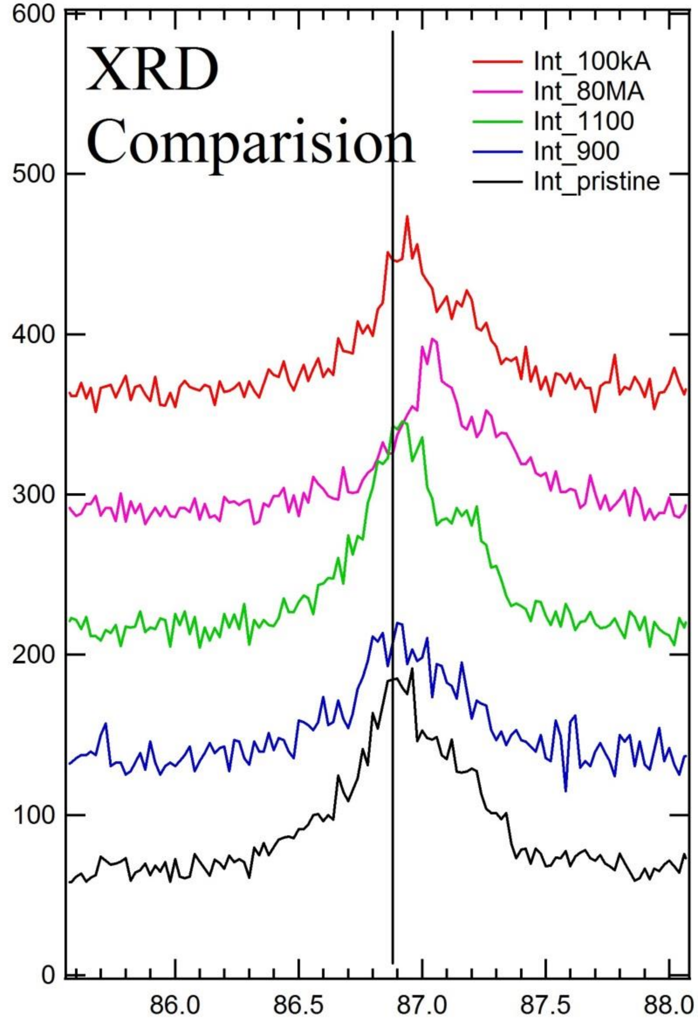
XRD
Comparision
Int_100kP
Int_100kA
Int_80MP
Int_80MA
Int_1100
Int_900
Int_pristine
4000
3000
2000
1000
0
40
50
60
70
80


XRD
Comparision
Int_100kA
Int_80MA
Int_1100
Int_900
Int_pristine
3000
2500
2000
1500
1000
500
0
-500
39.6
39.8
40.0
40.2
40.4
40.6
40.8
41.0


XRD
Comparision
Int_100kA
Int_80MA
Int_1100
Int_900
Int_pristine
3000
2500
2000
1500
1000
500
0
-500
39.6
39.8
40.0
40.2
40.4
40.6
40.8
41.0


XRD
Comparision
Int_100kA
Int_80MA
Int_1100
Int_900
Int_pristine
2500
2000
1500
1000
500
0
-500
57.6
57.8
58.0
58.2
58.4
58.6
58.8
59.0

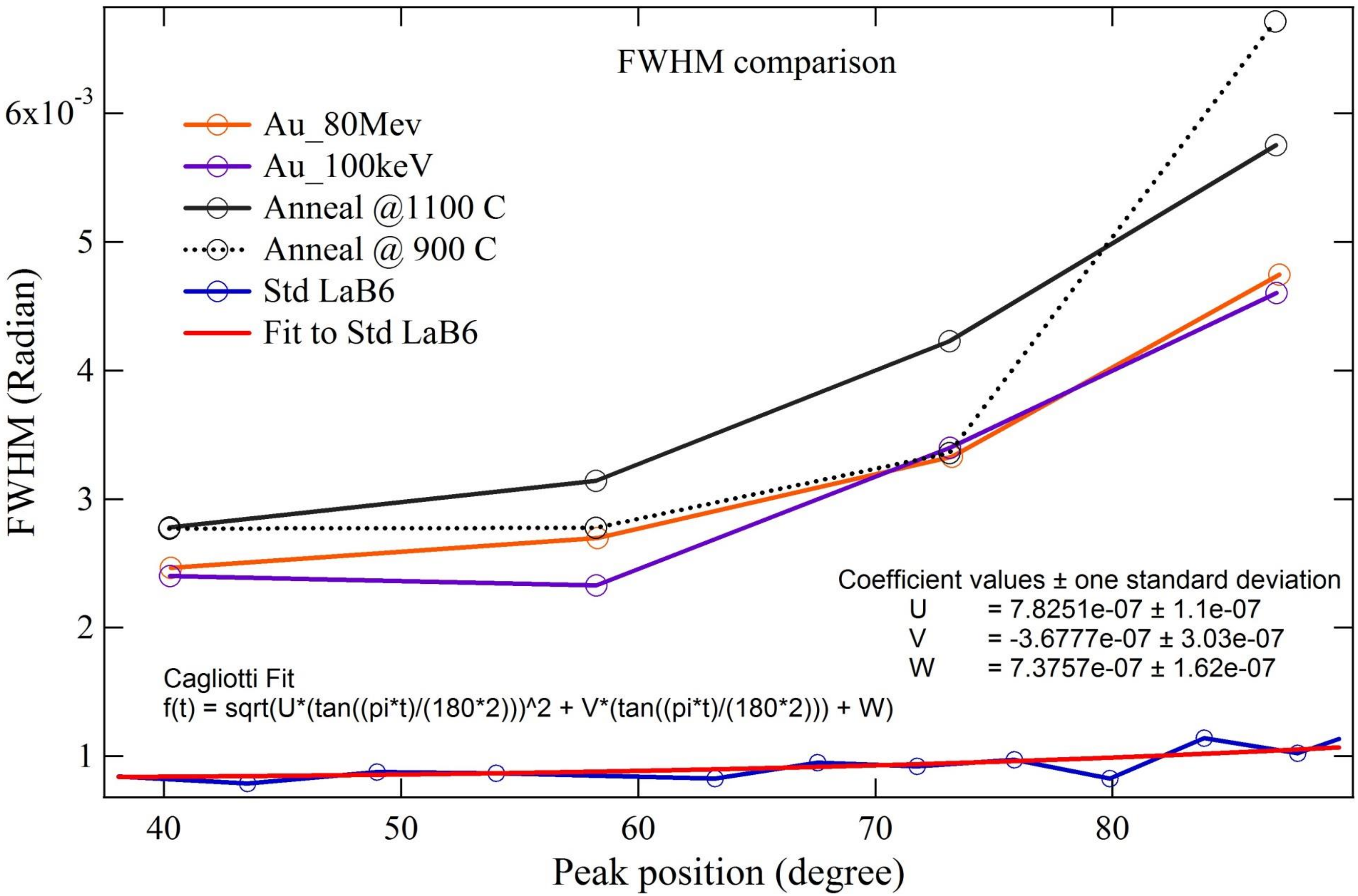

XRD
Comparision
Int_100kA
Int_80MA
Int_1100
Int_900
Int_pristine
2500
2000
1500
1000
500
0
-500
72.5
73.0
73.5
74.0

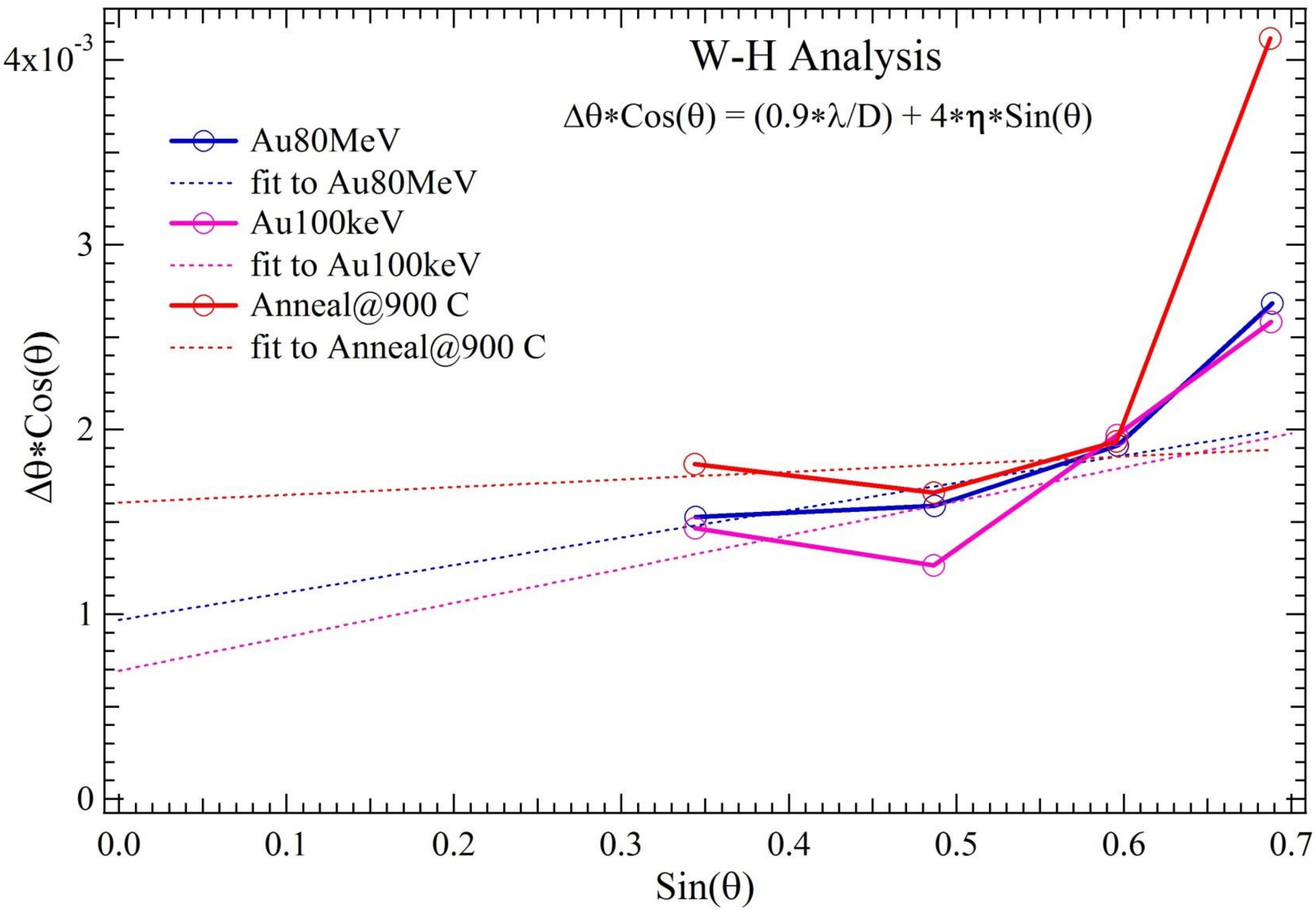

XRD
Comparision
Int_100kA
Int_80MA
Int_1100
Int_900
Int_pristine
600
500
400
300
200
100
0
86.0
86.5
87.0
87.5
88.0

Irradiation reduces the lattice stress

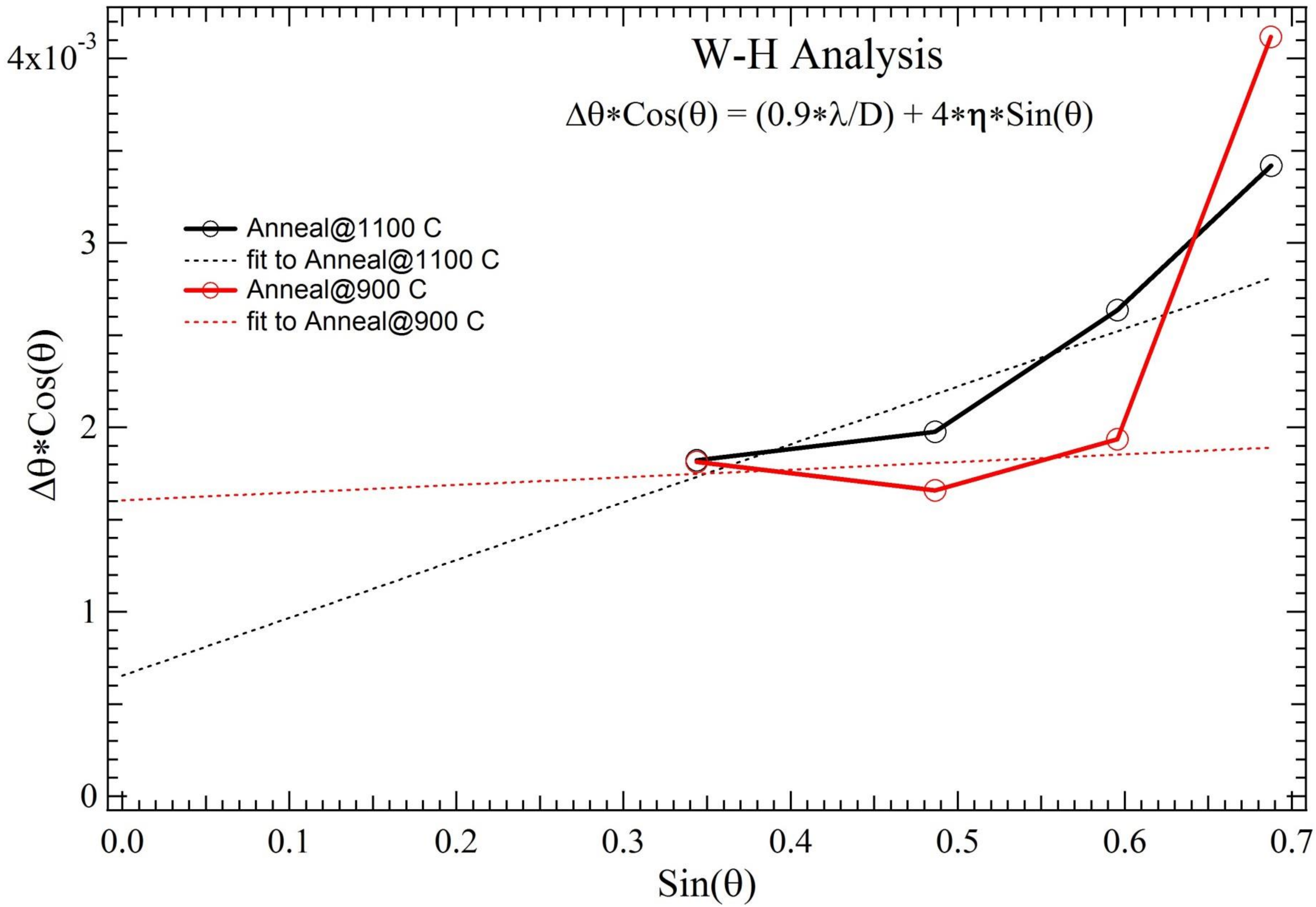
Sample	2*theta	theta	delta	d (Å)	a (Å)	strain(std)	Stress (GPa)
W110_S	40.265	20.1325	2.2397	3.1674			
W110_A	40.23	20.115	0.1593	2.2416	3.1701	8.48E-03	**0.3537**
W110_A80	40.29	20.145	0.141	2.2384	3.1656	5.80E-04	**0.2356**
W110_A100	40.25	20.125	0.138	2.2405	3.1686	3.57E-04	**0.1428**

•Young's modulus 400 GPa for polycrystalline W at 20 C (Ref:Tungsten, Lassner & Schubert)


FWHM comparison
6x10^{-3}
5
4
3
2
1
40
50
60
70
80
FWHM (Radian)
Peak position (degree)
Au_80Mev
Au_100keV
Anneal @1100 C
Anneal @ 900 C
Std LaB6
Fit to Std LaB6
Coefficient values ± one standard deviation
U = 7.8251e-07 ± 1.1e-07
V = -3.6777e-07 ± 3.03e-07
W = 7.3757e-07 ± 1.62e-07
Cagliotti Fit
f(t) = sqrt(U*(tan((pi*t)/(180*2)))^2 + V*(tan((pi*t)/(180*2))) + W)



	Annealed @ 900 C	Au irradiated @ 100 keV	Au irradiated @ 80 MeV
Crystallite Size	96 ± 0.31x nm	Not determinable	142 ± 0.36x nm
Micro strain η	~ 0	-	0.00037 ± 0.48x



	Annealed @ 900 C	Annealed @ 1100 C
Crystallite Size	96 ± 0.31x nm	Not determinable
Micro strain η	~ 0	-

PALS characterization

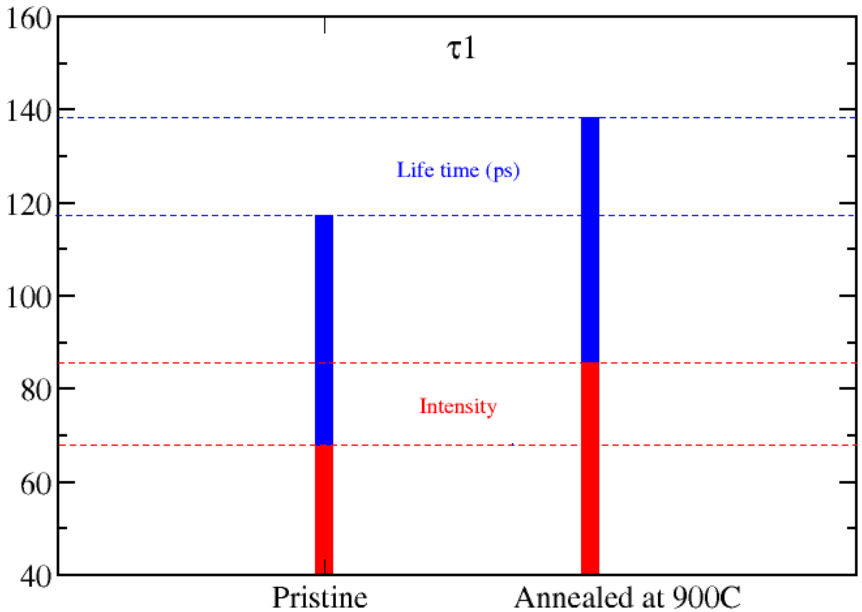
Details of Au and W irradiated samples and PALS characterization

Sr. No.	Identification (Sample details)	Ion species	Energy	Fluence
1.	80MPWAu6p214	Au7+(197)	80 MeV	$6.2x10^{14}$
2.	100KPWAu117	Au+1	0.1 MeV	$1x10^{17}$
3.	80MAWW114	W7+ (184)	80 MeV	$1x10^{14}$
4.	80MAWAu6p214	Au7+ (197)	80 MeV	$6.2x10^{14}$
5.	80MAWAu114	Au7+ (197)	80 MeV	$1x10^{14}$
6.	100KAWAu117	Au+1	0.1 MeV	$1x10^{17}$

Positron lifetime in bulk W – 105 ps

Sample	τ_1 (ps)	I_1	τ_2 (ps)	I_2
Plansee pristine	117	67.40	289	31.50
Plansee annealed (900C)	138	85.30	384	13.95
Linke annealed (900C)	133	78.20	347	20.80
80MeV annealed W on W ($1x10^{14}$ cm^{-2})	140	73.90	313	25.14
80MeV annealed Au on W ($1x10^{14}$ cm^{-2})	145	70.32	301	28.20
80MeV annealed Au on W ($6.2x10^{14}$ cm^{-2})	147	64.68	308	34.51
100keV annealed Au on W ($1x10^{17}$ cm^{-2})	145	69.20	297	29.94
80MeV pristine Au on W ($6.2x10^{14}$ cm^{-2})	142	64.97	302	34.10
100keV pristine Au on W ($1x10^{17}$ cm^{-2})	125	75.98	289	23.01

Effect of annealing in positron life time – Enhanced mono-vacancy like defects and vacancy clustering – Effect of stress in positron life time



First life time component (τ_1) for pristine and annealed samples along with respective intensities

The life-time of perfect crystal W ~105 ps

Pristine sample ~117 ps, much lower than mono-vacancy life time (also reported by Shengyun et.al , JNM, 343 (2005) 330)

Could be an indication of stress in the lattice (also seen in XRD measurements)

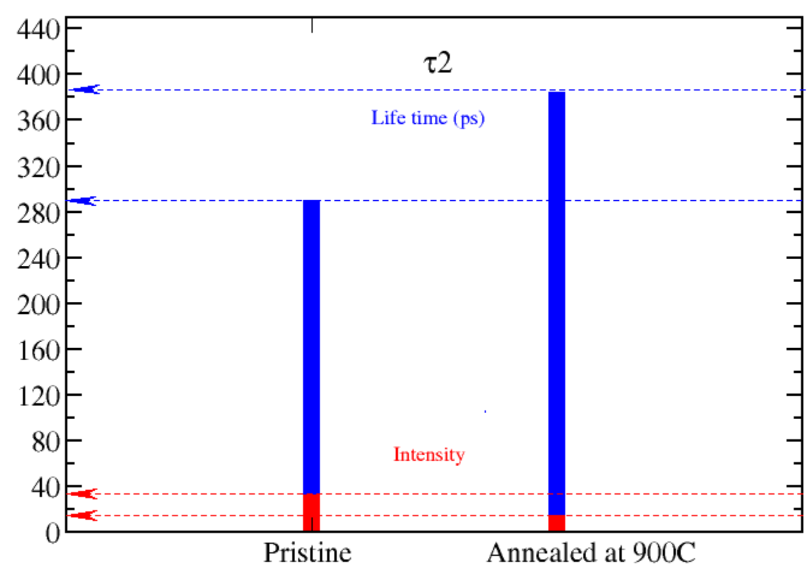
Stress can give intermediate life times

Annealing shows mono-vacancy like defects (τ_1) ~140 ps (higher intensity compared to pristine)

Annealing increases (τ_2) from 280 ps to 384 ps → Larger vacancy clusters

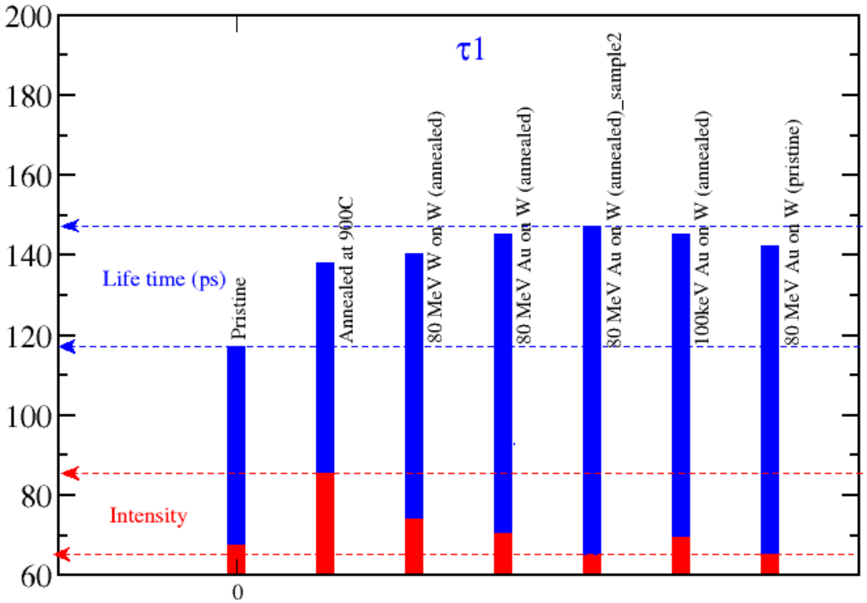
Lower intensity of (τ_2) → decomposition of vacancy clusters into mono-vacancy like defects

Effect of stress relieving ?



Second life time component (τ_2) for pristine and annealed samples and their intensities

Effect of irradiation – reduction in mono vacancy formation – Stress/dislocation effects



The first lifetime is nearly same (~140 ps) for all irradiated samples. Although the irradiation dose (1x1014 and 6.2x1014) shows a slight variation, they are still within the experimental errors

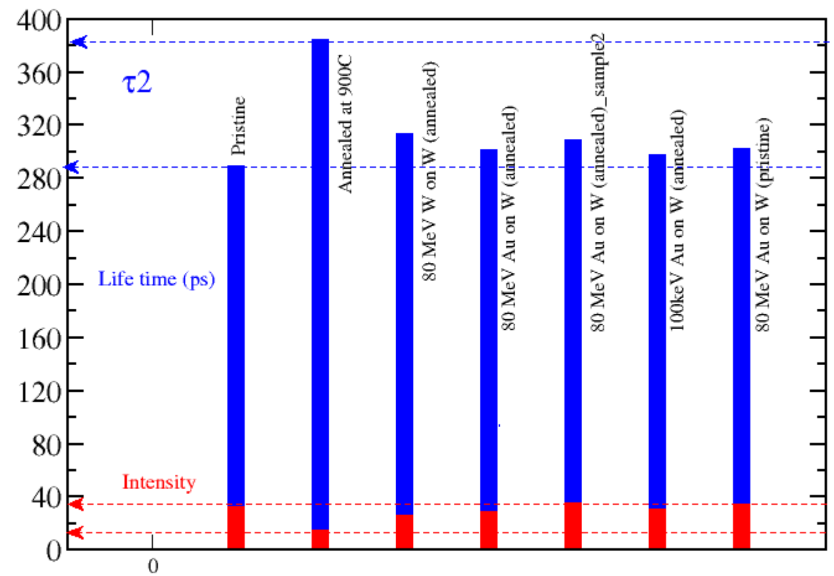
The intensity of the mono-vacancy like defects reduces due to irradiation

80 MeV irradiation in pristine sample also seem to have relieved the stress in positron range

τ2 is reduced from 384 ps to ~300 ps due to ion bombardment independent of the ion energy.

Also radiation induced stresses and dislocations in the lattice can give rise to intermediate positron life times (Kuramoto etal, JAERI conf. 2003-001)

Further investigations are ongoing to confirm this.



Detail of 7.5 MeV W and 50 MeV H_2 irradiation of Tungsten

Sample	Sample ID	W implantation fluence (ions /cm²)	Damage due to W ion implantation at surface(peak) (dpa)	Hydrogen concentration (ions/cm²)	Damage due to H ion implantat. at surface(peak) (dpa)
Plansee	P1 mask	-	-	1×10^{18}	0.09(0.74)
Plansee	P1	4.5×10^{15}	10 (17)	1×10^{18}	0.09(0.74)
Linke	L1	4.5×10^{15}	10 (17)	1×10^{18}	0.09(0.74)
Plansee	P2	4.5×10^{14}	1(1.7)	1×10^{18}	0.09(0.74)
Linke	L2	4.5×10^{14}	1 (1.7)	1×10^{18}	0.09(0.74)

The displacement damage on the sample due to tungsten and hydrogen implantations as given in the Figures have been calculated using SRIM program using quick damage routines; The quick damage routine invokes the Kinchin Pease model where the number of displacements

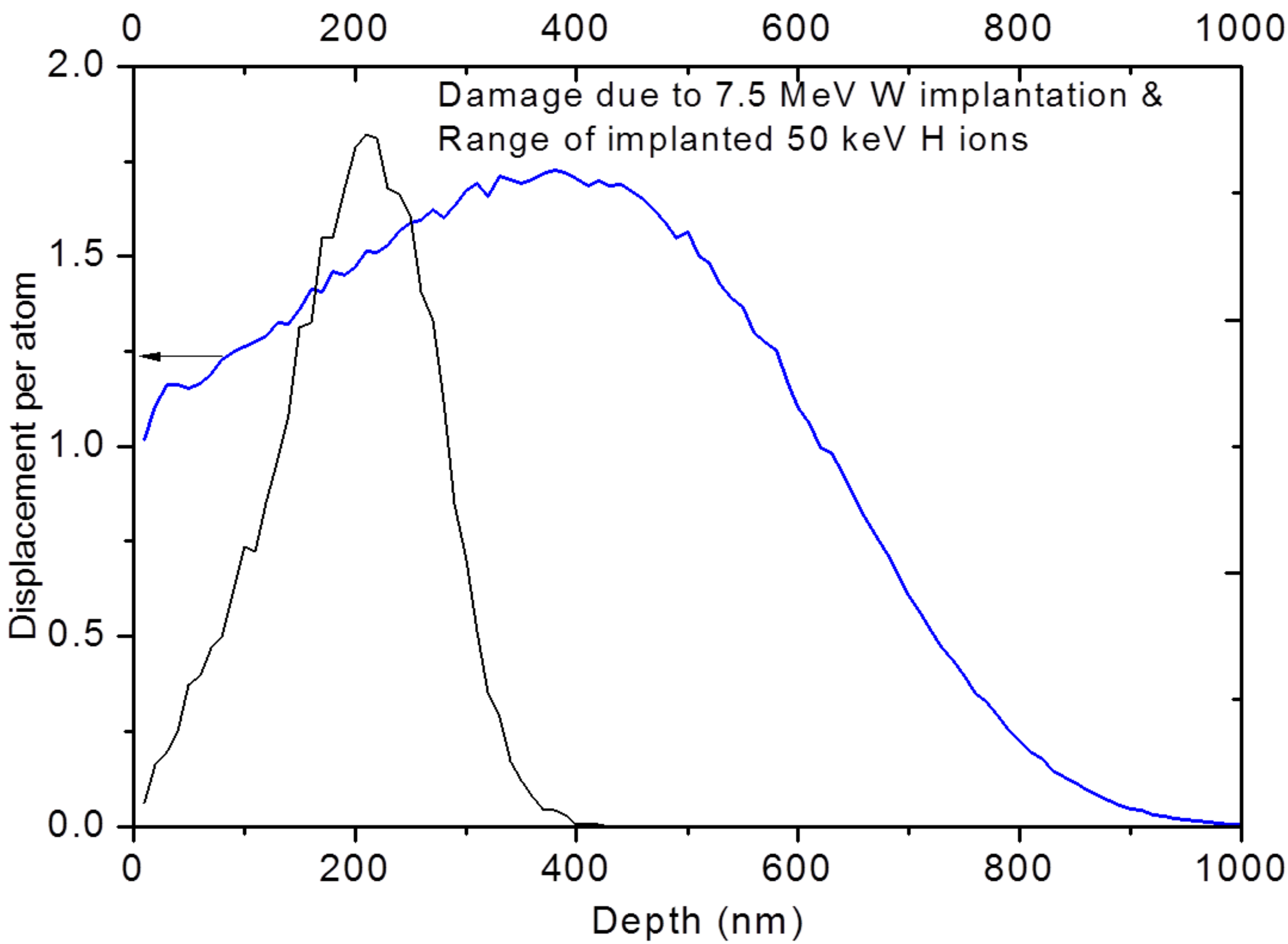
ND = 0.8 Edamage/ 2 Ed

Where ND = total number of displacements

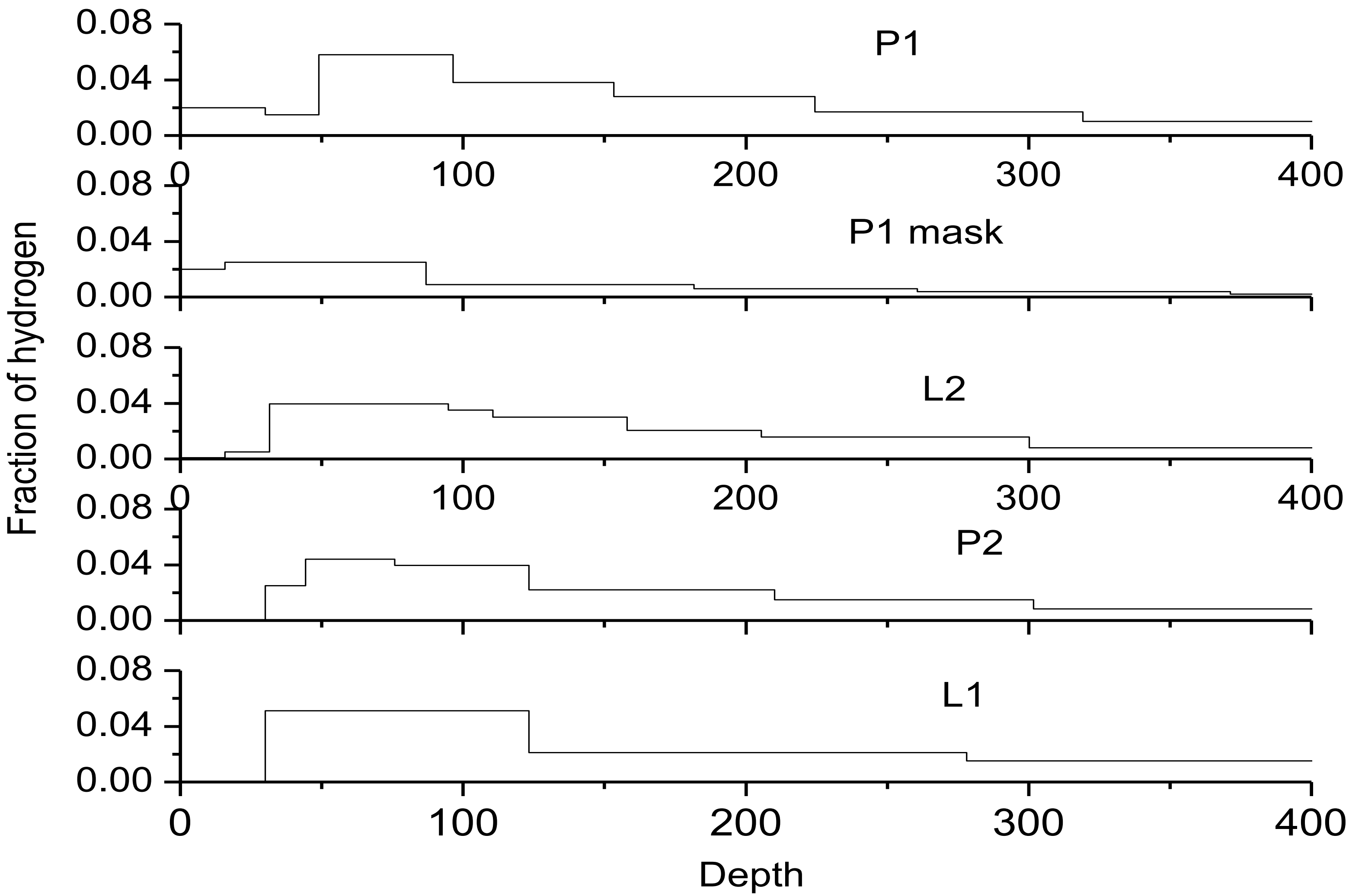
Edamage = fraction of energy used in creating displacement damage

Ed = displacement energy of 90 eV.

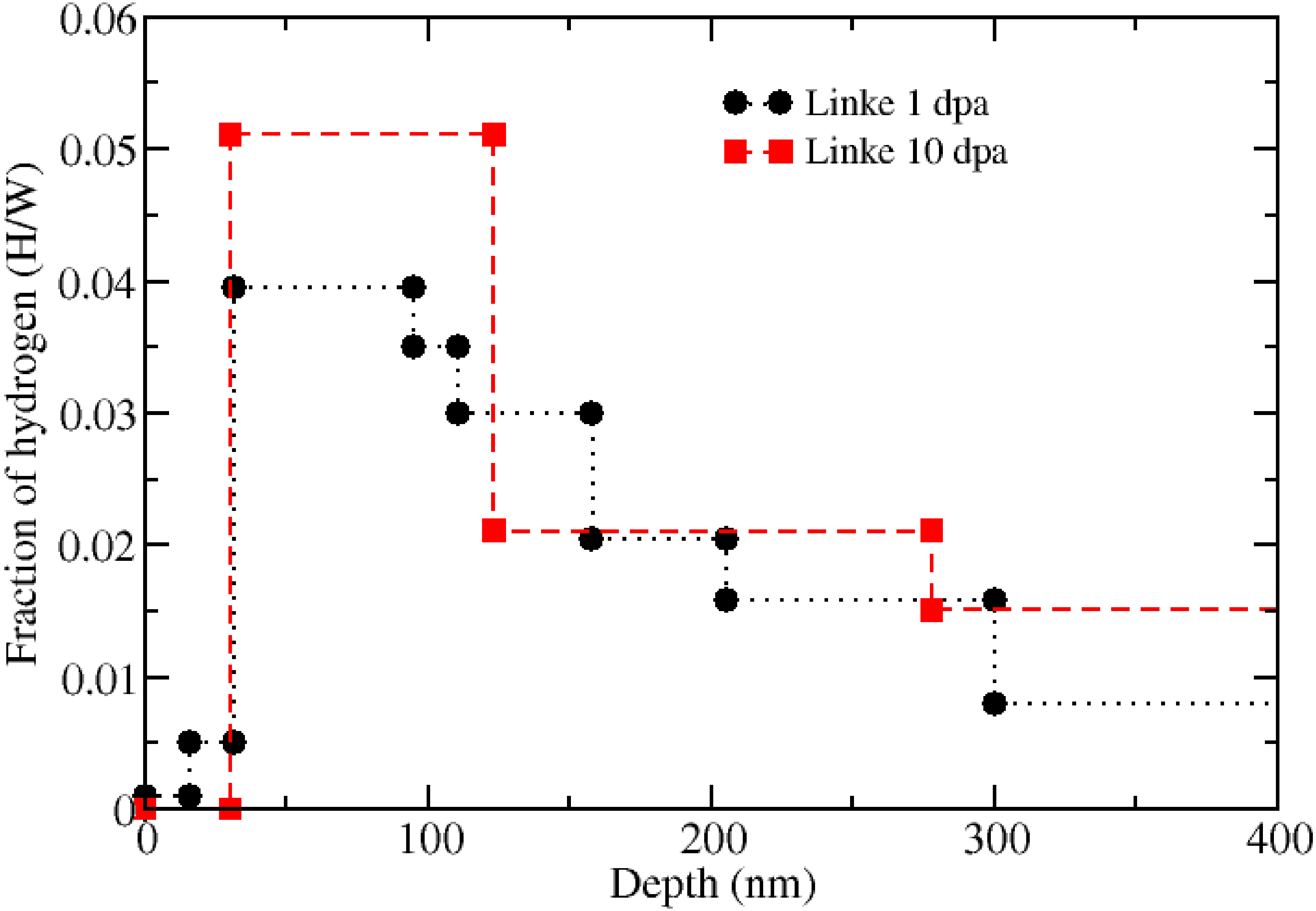
We have calculated the fluence such that L1 and P1 samples are exposed to 10 dpa at the surface; Whereas the samples L2 and P2 are exposed to 1 dpa at the surface.



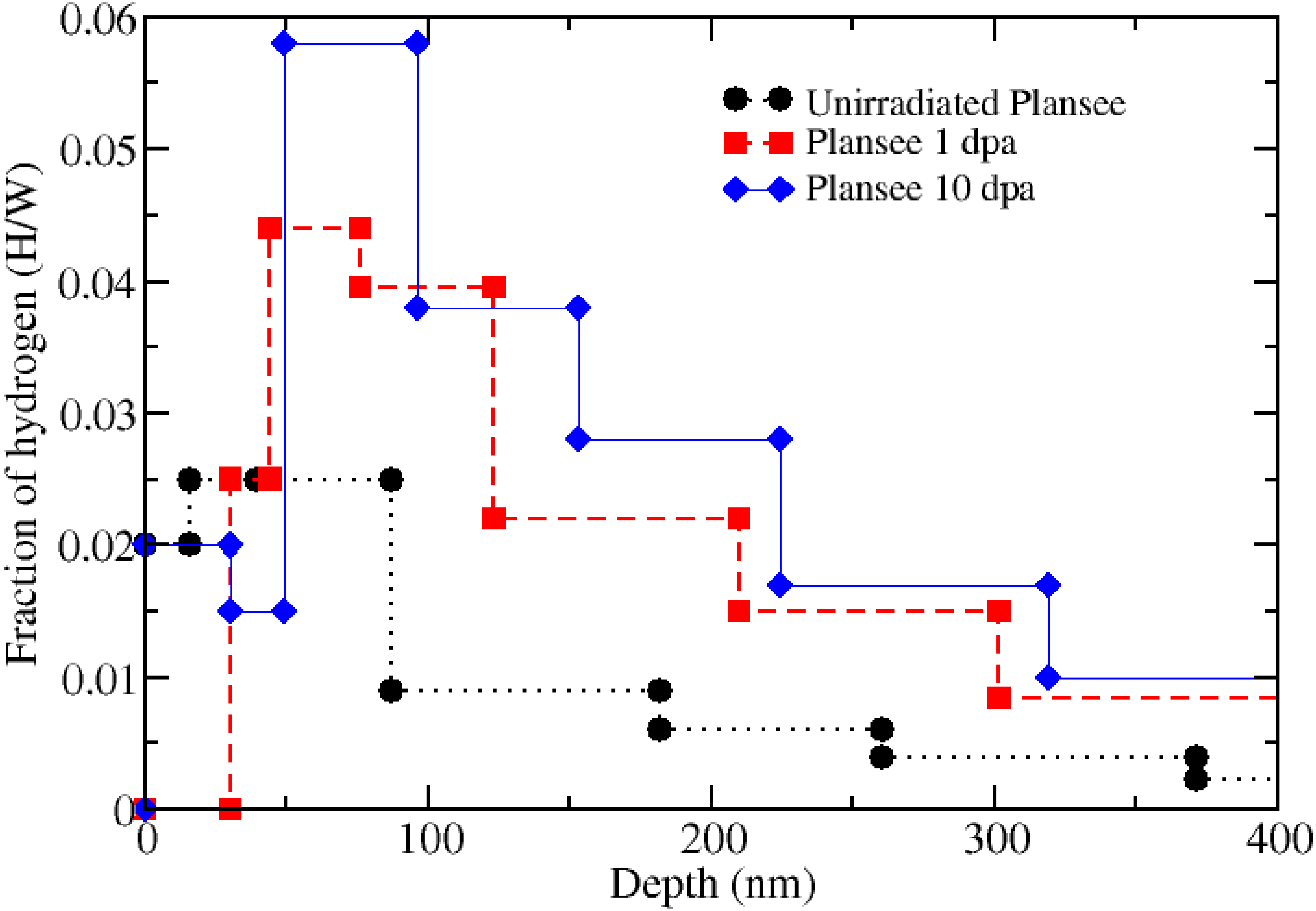
Displacement damage due to implantation of 7.5 MeV W implantation to a fluence of 4.5×10^{14} W ions /cm^2, as obtained from SRIM program.



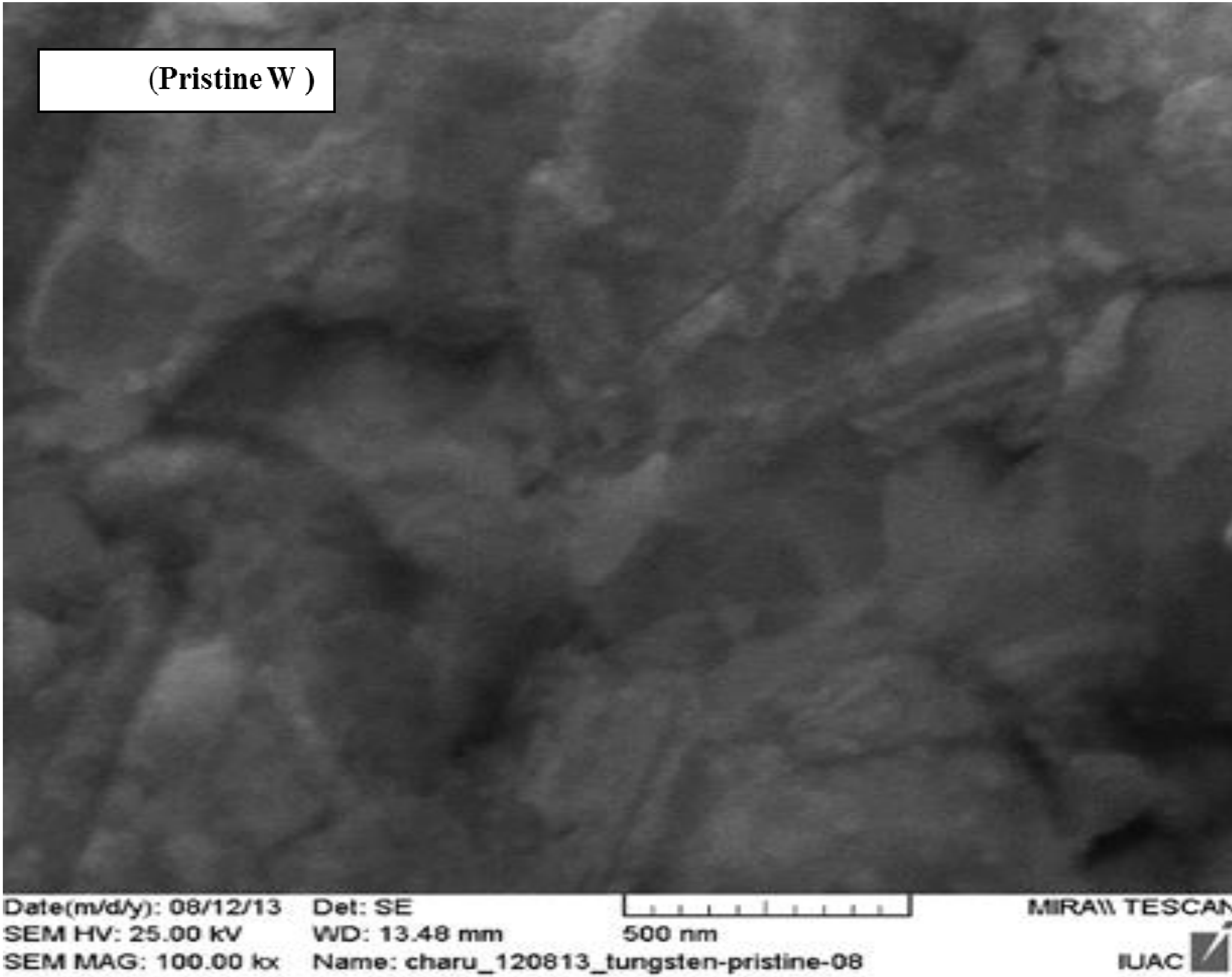
The hydrogen depth profiles obtained using SIMNRA program.

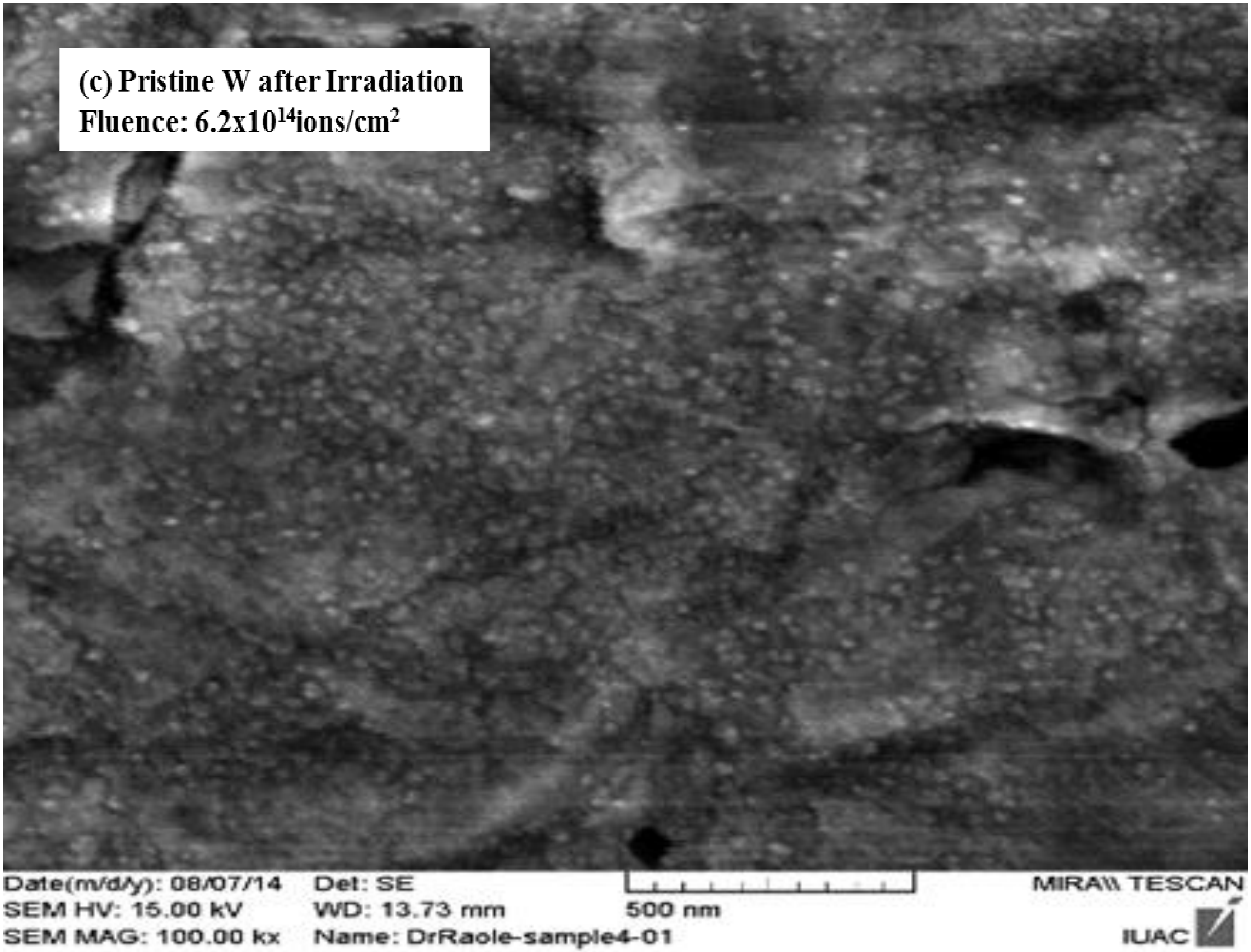


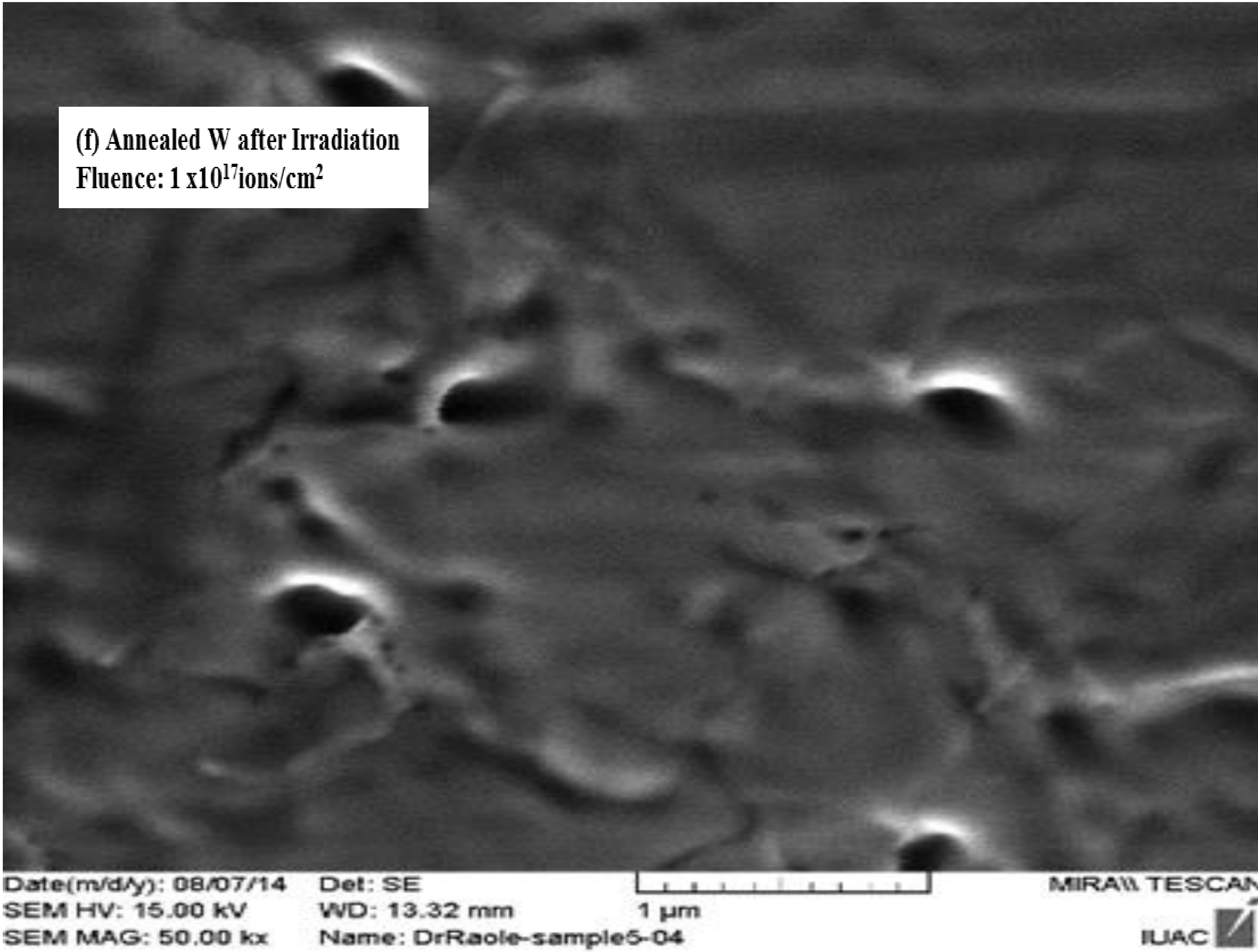
Linke 1 dpa
Linke 10 dpa
0.06
0.05
0.04
0.03
0.02
0.01
0
0
100
200
300
400
Fraction of hydrogen (H/W)
Depth (nm)



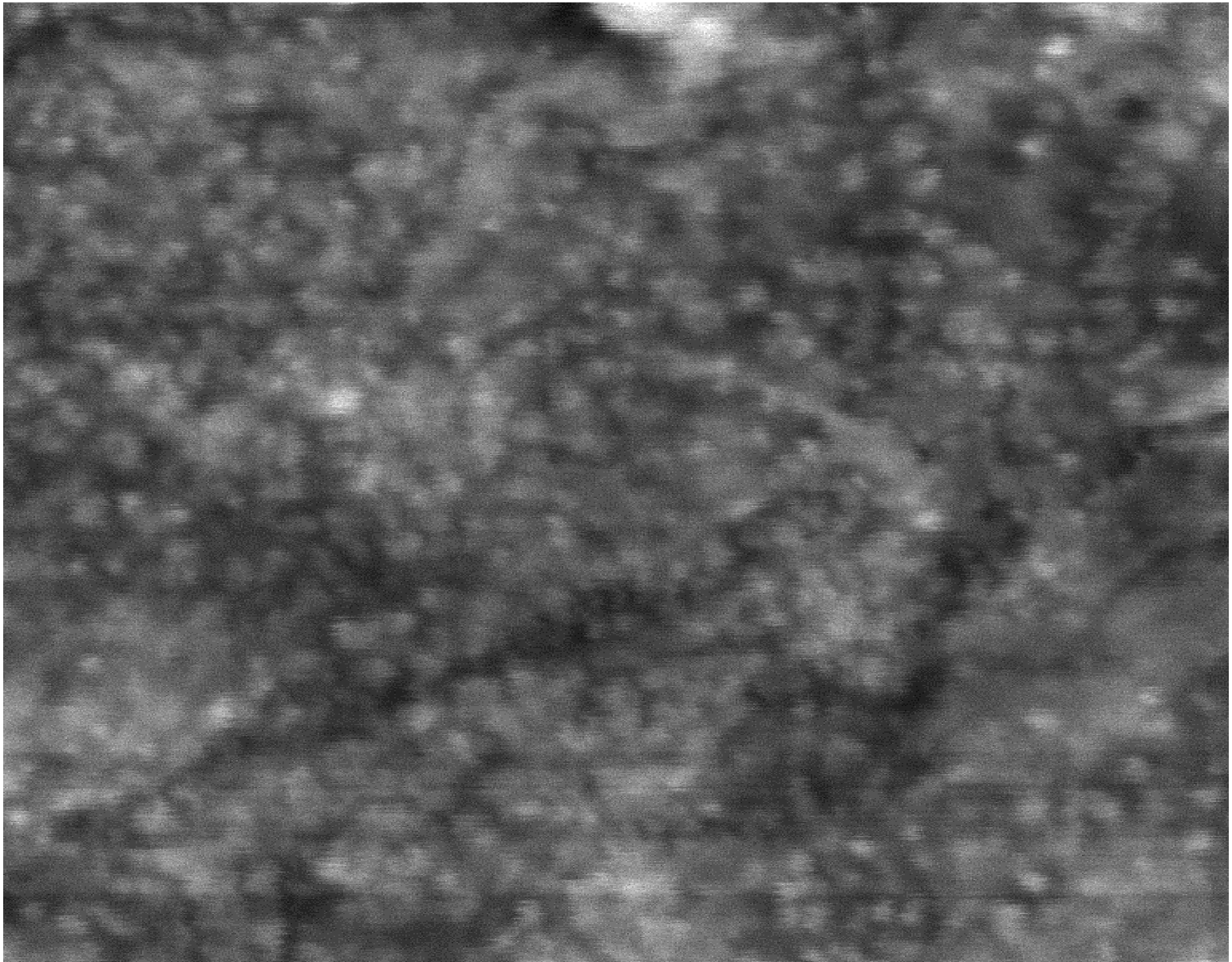
Unirradiated Plansee
Plansee 1 dpa
Plansee 10 dpa
Fraction of hydrogen (H/W)
Depth (nm)
0
0.01
0.02
0.03
0.04
0.05
0.06
0
100
200
300
400


(Pristine W)
Date(m/d/y): 08/12/13
SEM HV: 25.00 kV
SEM MAG: 100.00 kx
Det: SE
WD: 13.48 mm
Name: charu_120813_tungsten-pristine-08
500 nm
MIRA\\ TESCAN
IUAC

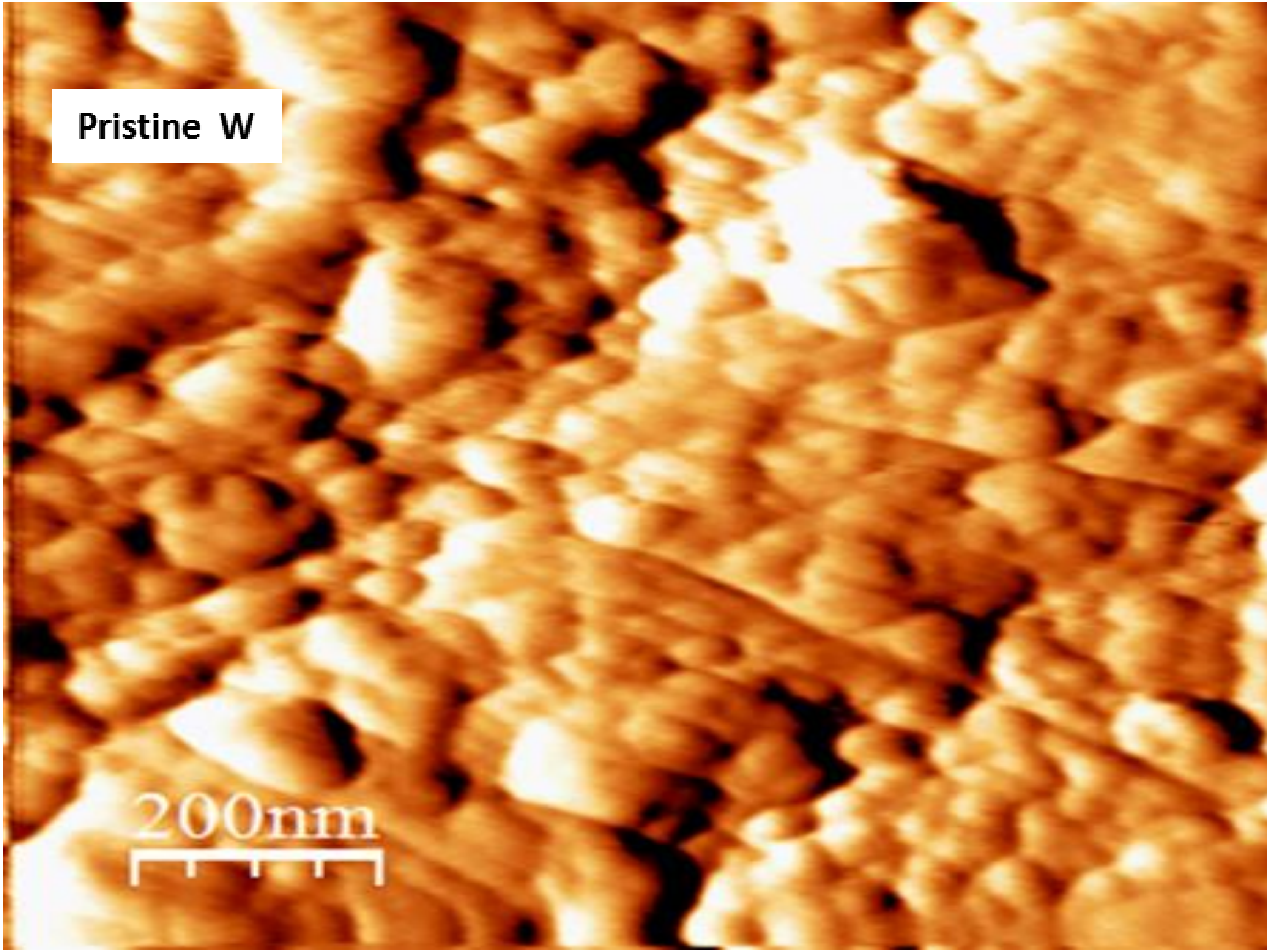

(c) Pristine W after Irradiation
Fluence: 6.2x10^{14}ions/cm^2
Date(m/d/y): 08/07/14
SEM HV: 15.00 kV
SEM MAG: 100.00 kx
Det: SE
WD: 13.73 mm
Name: DrRaole-sample4-01
500 nm
MIRA\\ TESCAN
IUAC

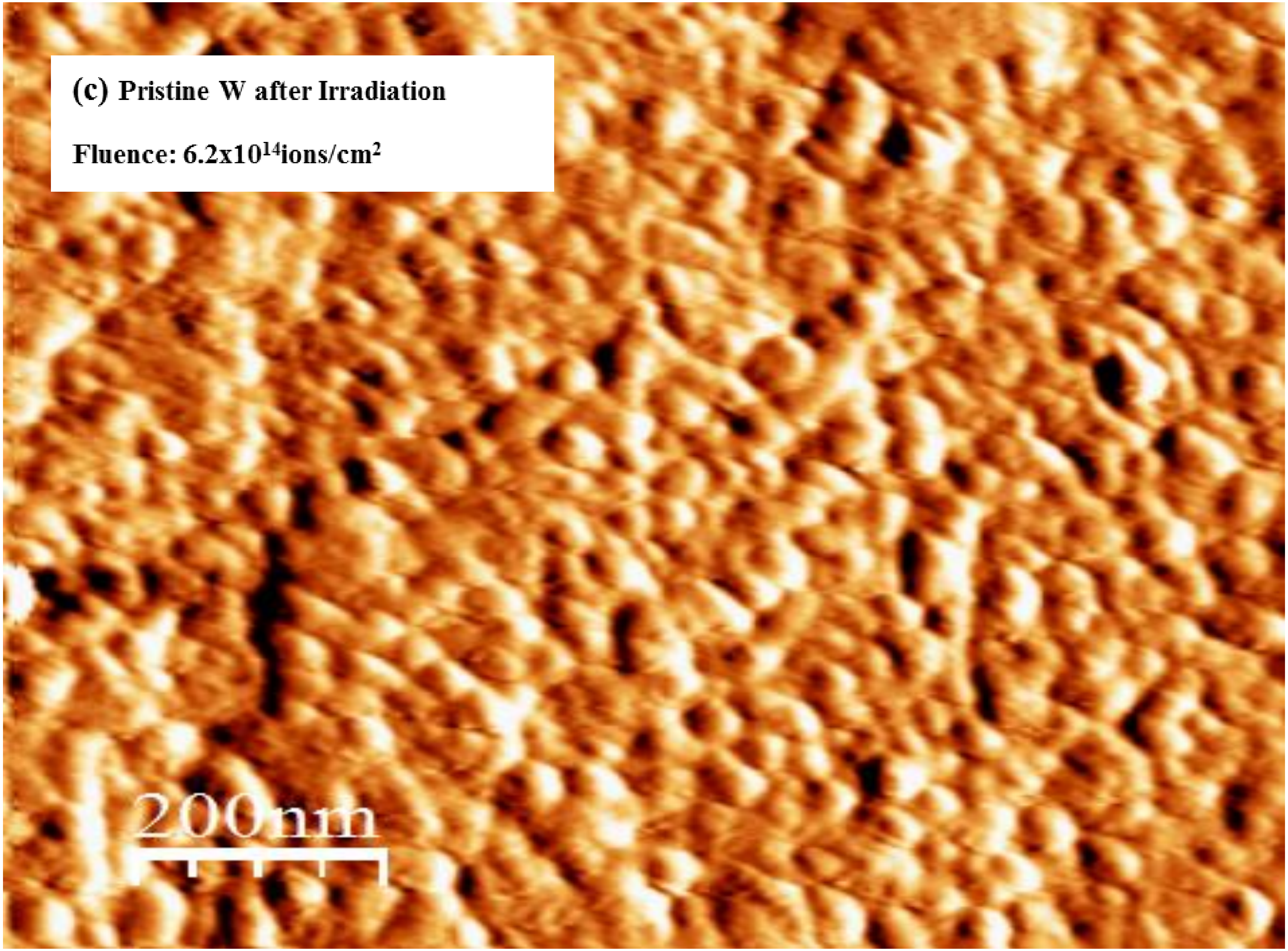

(f) Annealed W after Irradiation
Fluence: 1 x10^17 ions/cm^2
Date(m/d/y): 08/07/14
SEM HV: 15.00 kV
SEM MAG: 50.00 kx
Det: SE
WD: 13.32 mm
Name: DrRaole-sample5-04
1 μm
MIRA\\ TESCAN
IUAC

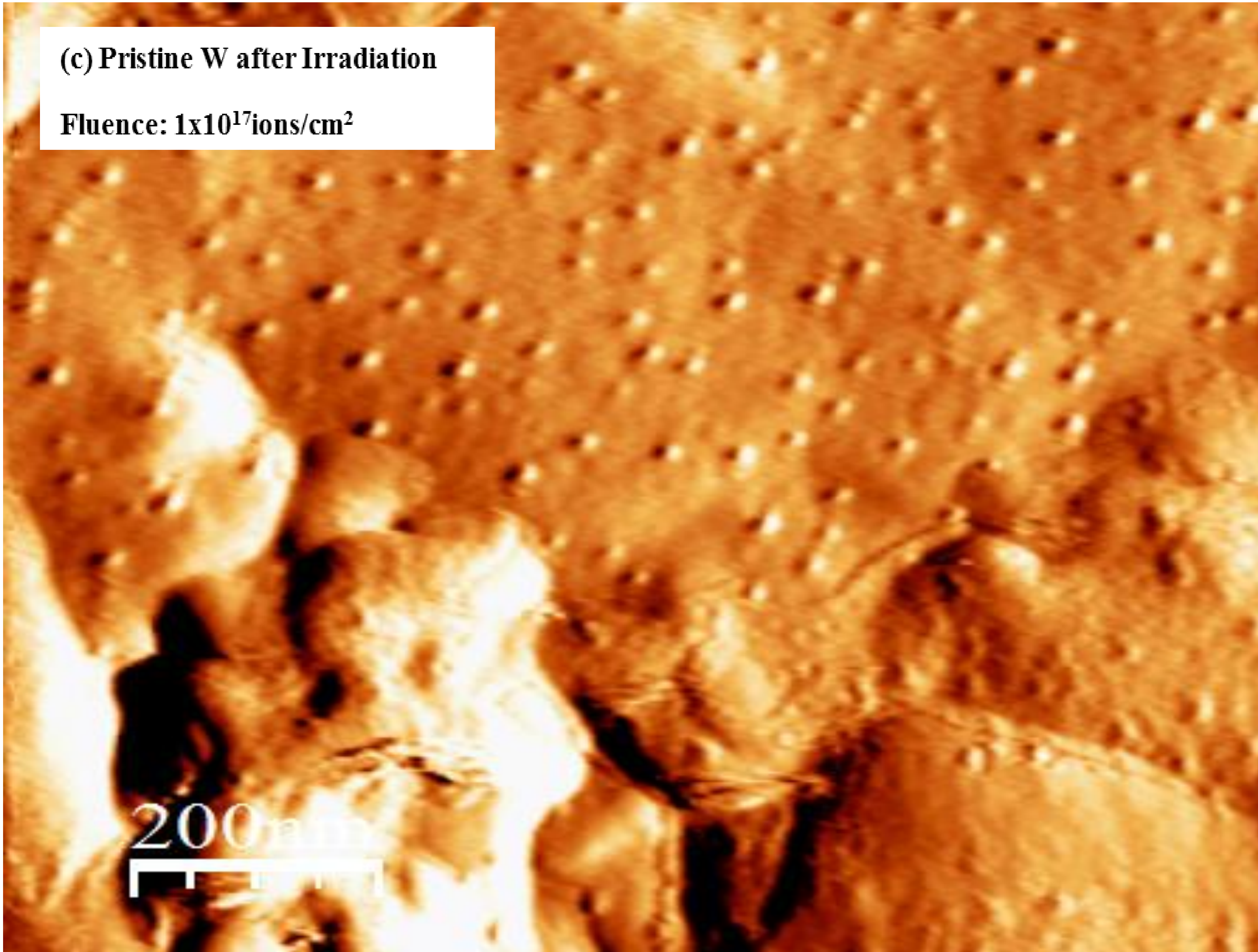
80MPWAu6p214 Image 2

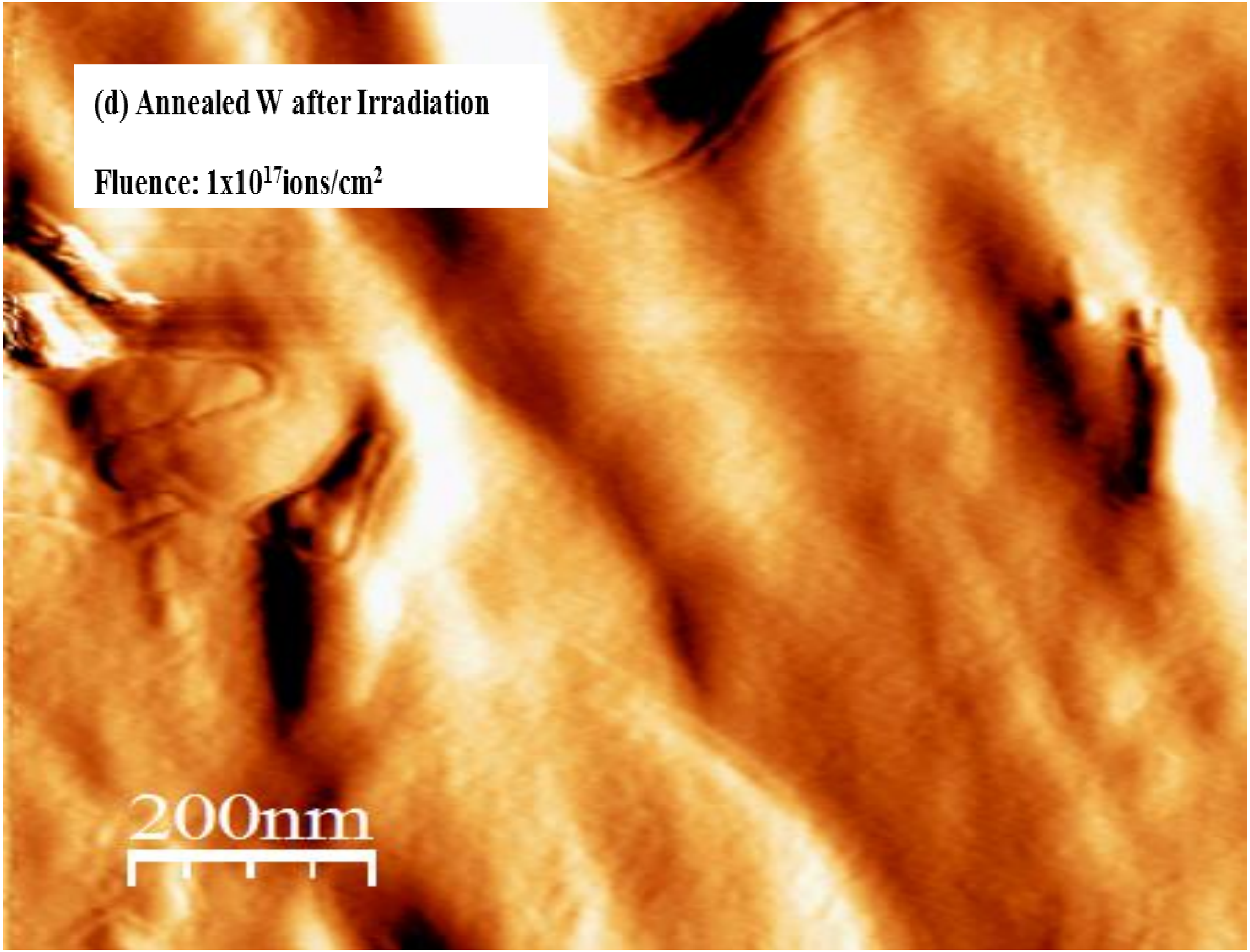


Date(m/d/y): 08/07/14 Det: SE MIRA\\ TESCAN
SEM HV: 15.00 kV WD: 13.73 mm 200 nm
SEM MAG: 200.00 kx Name: DrRaole-sample4-02 IUAC


Pristine W
200nm


(c) Pristine W after Irradiation
Fluence: 6.2x10^{14}ions/cm^2
200nm


(c) Pristine W after Irradiation
Fluence: 1x10^17 ions/cm^2
200nm


(d) Annealed W after Irradiation
Fluence: 1x10^17ions/cm^2
200nm

Summary

- EBSD results show that Tungsten (Linke-Plansee) samples have equiaxed grains on the surface. However, in the cross section columnar grains are seen. In the Plansee samples there are larger grains and lattice is strained.
- The pristine and 900 C annealed samples are tensile strained with respect to standard relaxed tungsten matrix d values. It gets progressively relieved with 1100 C anneal, 100 keV irradiation and 80 MeV irradiation.
- All irradiated samples show that irradiation tends to increase preferred orientation in (200) and (211) direction, and the effect must be in substantial thickness of 10s of microns.
- Increase in the crystallite size is observed in 80 MeV irradiated gold.
- PALS results show the effect of annealing in terms of enhanced mono-vacancy like defects and vacancy clustering . The effect of irradiation is seen in the reduction in mono vacancies.
- ERDA results show that as the W irradiation dpa value incresaes, small increase in the H trapping is seen.
- Nanostructuring is observed in 80 MeV Au irradiated Tungsten.

Thank You