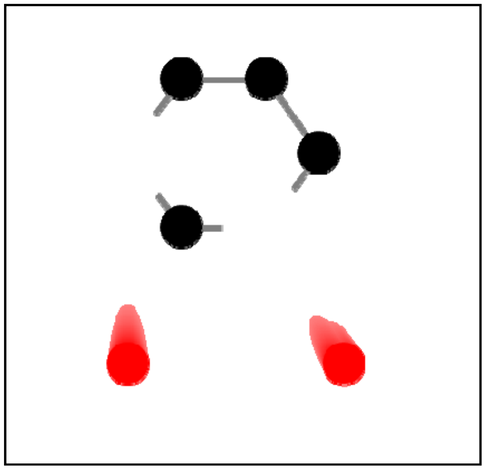

Experimental investigation of beryllium: plans and current results within the RaDIATE collaboration

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May 21, 2014

UNIVERSITY OF OXFORD

Fermilab

Science & Technology Facilities Council
Rutherford Appleton Laboratory

Content

- Context of the research
- Materials, point of interest
- Microstructural investigation proton irradiation vs ion implantation
- Mechanical properties
- Conclusions

http://www-radiate.fnal.gov

RaDIATE Collaboration

Radiation Damage In Accelerator Target Environments

Investigation of the radiation response of structural window and target materials in new highly intensity proton accelerator particle sources

Beryllium **is a promising candidate because of:**

- good "nuclear" properties;
- appropriate mechanical properties
- good "thermal" properties (conductivity, specific heat, melting point);
- high oxidation resistance;
- positive experience from existing facilities

Where will Beryllium be used?

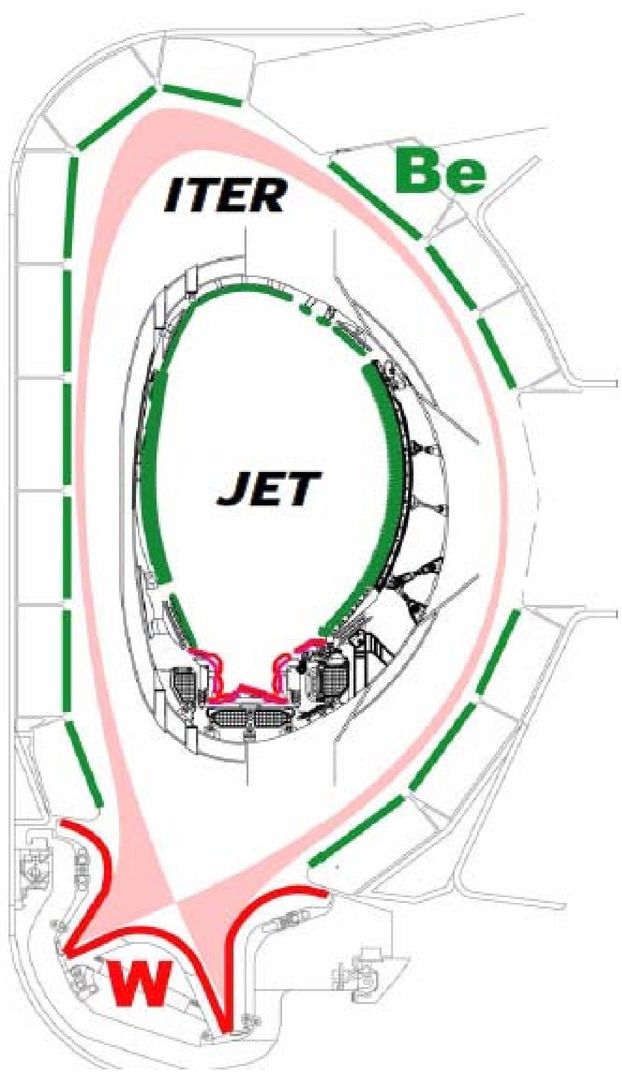
Long-Baseline Neutrino Experiment (LBNE)

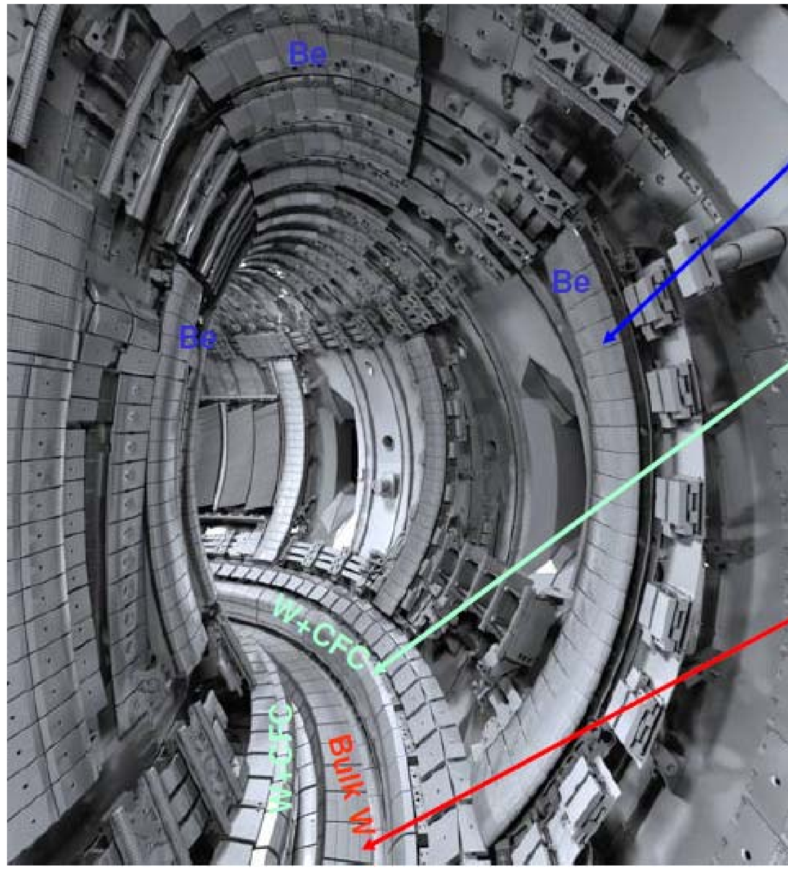
Application	Operating conditions					Proton beam parameters
	Avg. T (°C)	Peak T (°C)	Total DPA	Gas production (appm/DPA)		
				He	H	
Beam window (vacuum to air)	200	300	~ 0.23/yr	1030	2885	700 kW; 120 GeV; ~1 Hz; σ_{rms} = 1.3 mm
Target	375	450	~ 0.23/yr	1030	2885	700 kW; 120 GeV; ~1 Hz; σ_{rms} = 1.3 mm

Size:
Target: L = 950 mm, D = 15.3 mm (48 sections)
Window: 25.4 mm diameter, 0.25 mm thick

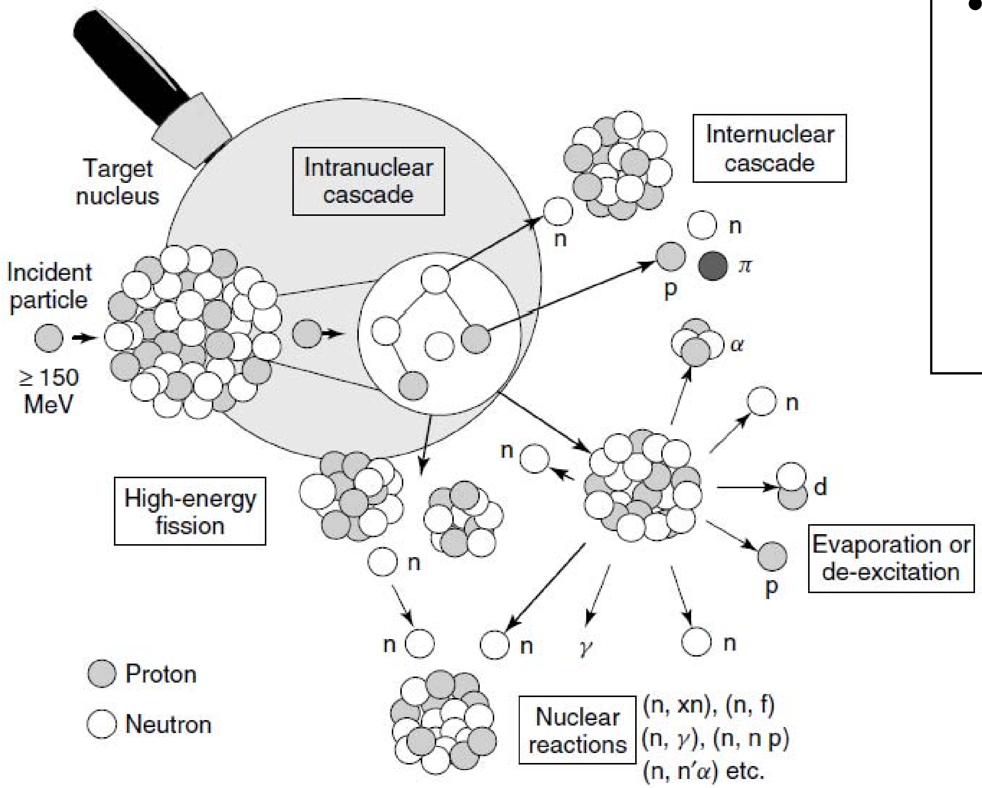
Environment: elevated temperature + radiation + pulsing loads

From: Matthews (CCFE). Overview of the JET ITER Overview ITER--like Wall first results and scientific programme first programme. APS Salt Lake City, November 2011APS 2011





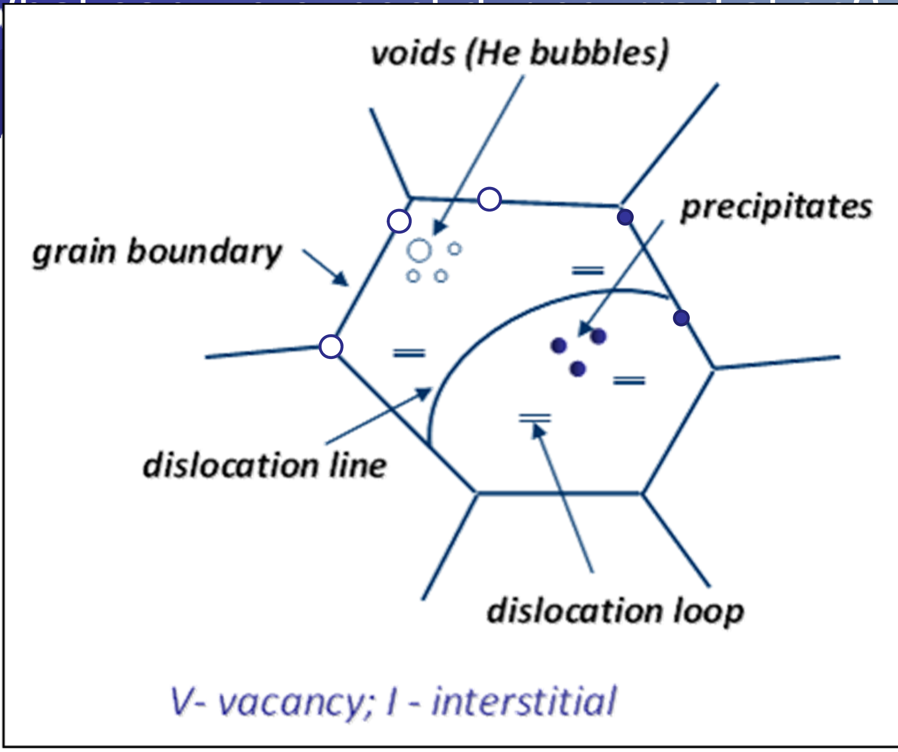
Experience exchange with fusion community



From D. Filges, F. Goldenbaum, in:, Handb. Spallation Res., Wiley-VCH Verlag GmbH & Co. KGaA, 2010, pp. 1–61.

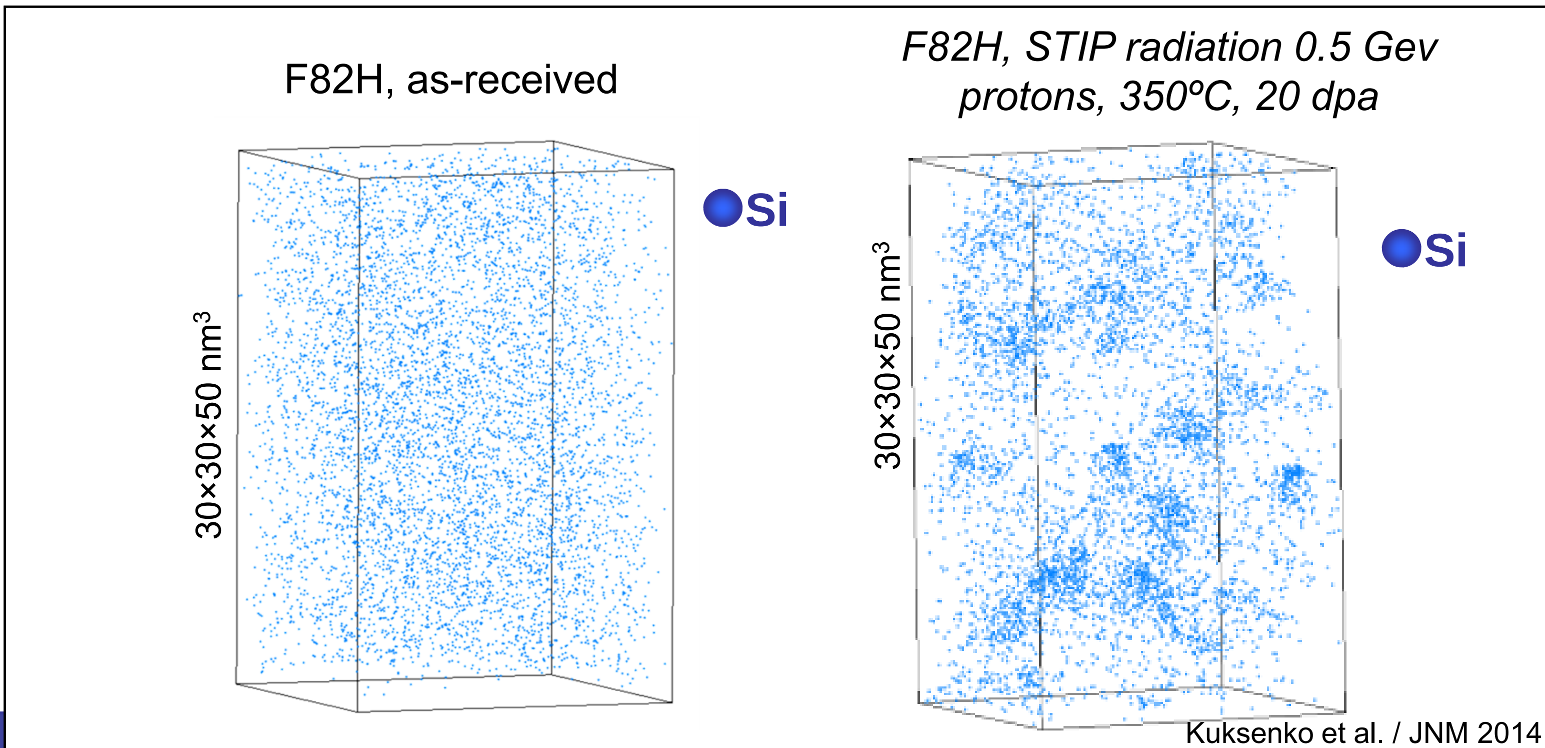
Microstructural response:

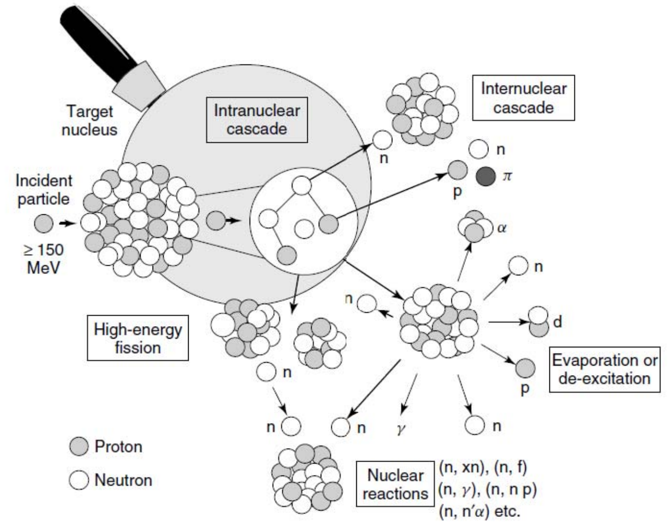
- *creation of transmutation products;*



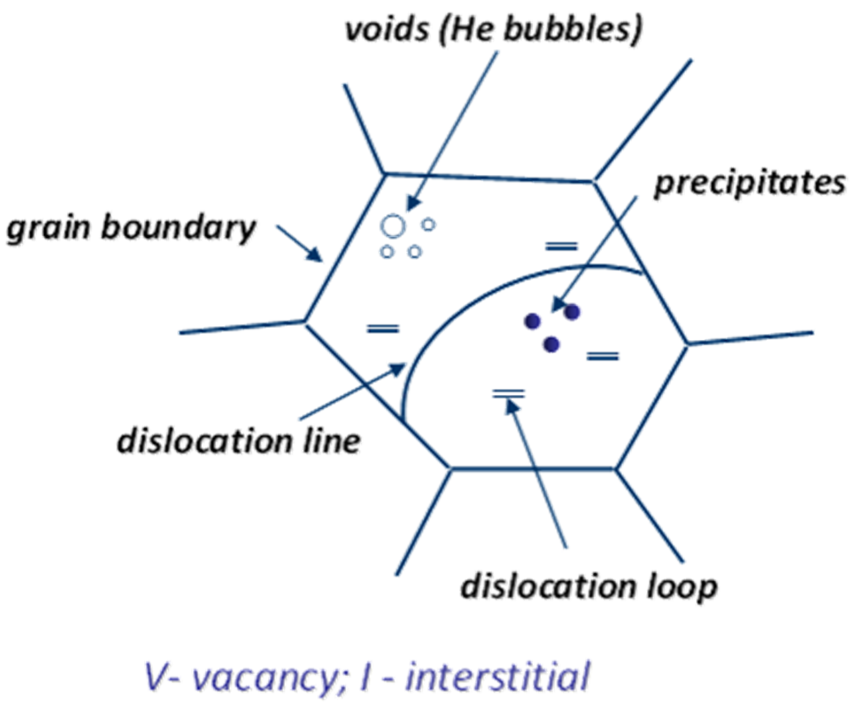
Microstructural response:

- *creation of transmutation products;*
- *creation and agglomeration of point defects;*
- *segregation (precipitation) or depletion on point defect sinks*





From D. Filges, F. Goldenbaum, in:, Handb. Spallation Res., Wiley-VCH Verlag GmbH & Co. KGaA, 2010, pp. 1–61.



Microstructural response:

- *creation of transmutation products;*
- *creation and agglomeration of point defects;*
- *segregation (precipitation) or depletion on point defect sinks*

Possible irradiation effects:

- *reduction of fracture toughness*
- *irradiation induced hardening*
- *reduction of ductility*
- *reduction of thermal conductivity*

PF-60

	Max impurities, appm
Al	170
C	450
Fe	130
Mg	810
O	2900
Si	130
N	195
Be	balance

S-200-F

	Max impurities, appm
Al	335
C	1130
Fe	210
Mg	130
O	5445
Si	195
Be	balance

S-65

	Max impurities, appm
Al	170
C	680
Fe	130
Mg	15
O	3260
Si	145
Be	balance

Method of manufacture: vacuum hot pressing

Beryllium is of industrial purity

How can we predict the radiation effect?

Investigation of the as-received Be

Investigation of the existing proton Be window

- “real” GeV proton irradiation;
- irradiated volume is big enough for microstructural investigations and micromechanical tests

But: **radioactivity of the sample**

Simulation with ion irradiation experiments

- flexibility of irradiation conditions
- observations of the evolution of the microstructure structure;
- reasonable correspondence of He/dpa ratio.

Low energy in-situ irradiation:

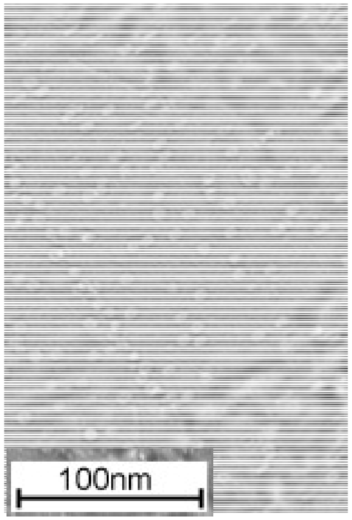
- easy variation of irradiation parameters;

High-energy irradiation + PIE

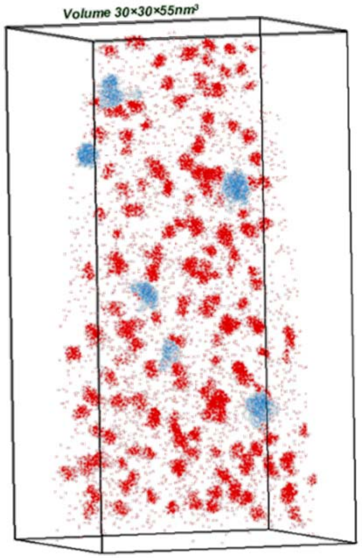
- microstructural and micromechanical tests data will be available

But: **validity of the simulation should be confirmed**

Which experimental technique should be used?

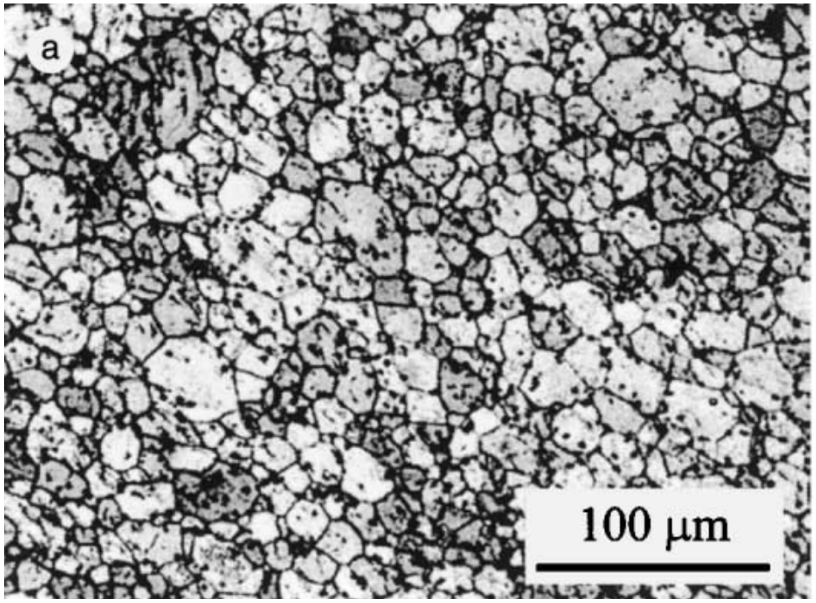


TEM → *defect clusters and He bubbles, precipitates stability*

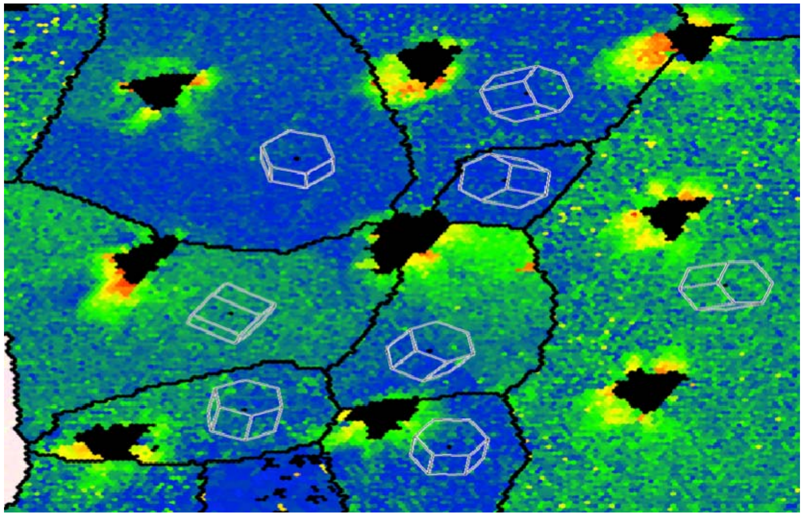


APT → *behaviours of impurities (precipitations, segregations at point defect sinks*

Micromechanical tests → *changes of mechanical properties*



TE-56 beryllium, Chakin and Ostrovsky / JNMm 2002



Local misorientation around indents made in pure Zr measured using EBSD
From http://energy.materials.ox.ac.uk/nuclear-projects/previous-projects/hydride-cracking-in-zirconium.html

Phases effect (thermal ageing data):

Fe-Al rich precipitates can:

- affect **ductility** and **creep strength** (*Jones et al. J. Common Met. 1964*)
- be preferential sites for **corrosion** pit initiation (*Punni and Cox, Corros. Sci. 2010*)

Fe-Be precipitates can

lock dislocation and increase **hardness** (*Morozumi et al. Trans. Jpn. Inst. Met. 1969*)

Al and Mg can

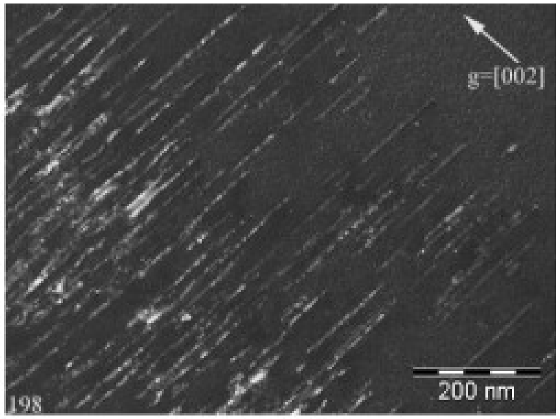
- form **low melting point eutectics** (*Kleykamp, JNM 2001*)

Precipitates should be investigated
Irradiation can produce much bigger variety of phases

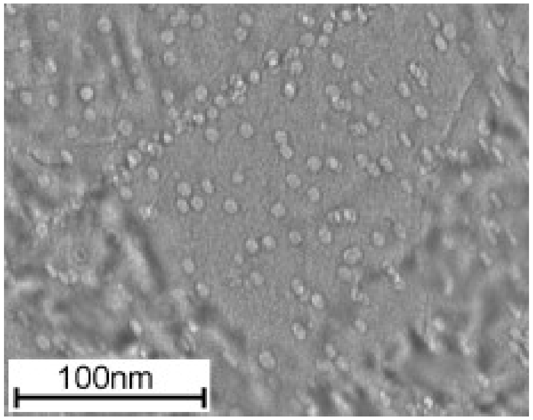
n-irradiation:

- At low T_{irr} below ~ 200ºC (Chakin et al. JNM 2009) or 400ºC (Gelles et al. JNM 1994): "black dots" and dislocation loops.
- At higher T: mainly He babbles

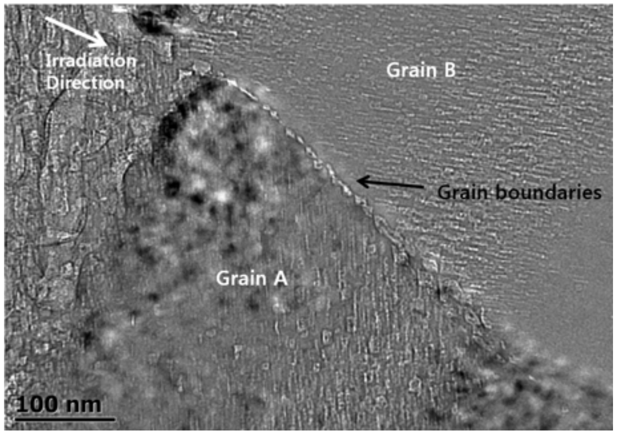
Implantation of He and H:
bubbles can dominate even at RT



Irr. Be, TEM, DF, dislocation loops, Tirr.=70 °C, $F = 6\times10^{22}$ cm^{-2} (E > 0.1 MeV) (Chakin et al. JNM 2009)



Irr. Be, TEM, BF, He bubbles loops, Tirr.=413 °C, $F = 6.5\times10^{21}$ cm^{-2} (E > 1 MeV) (Klimenkov et al. JNM 2013)

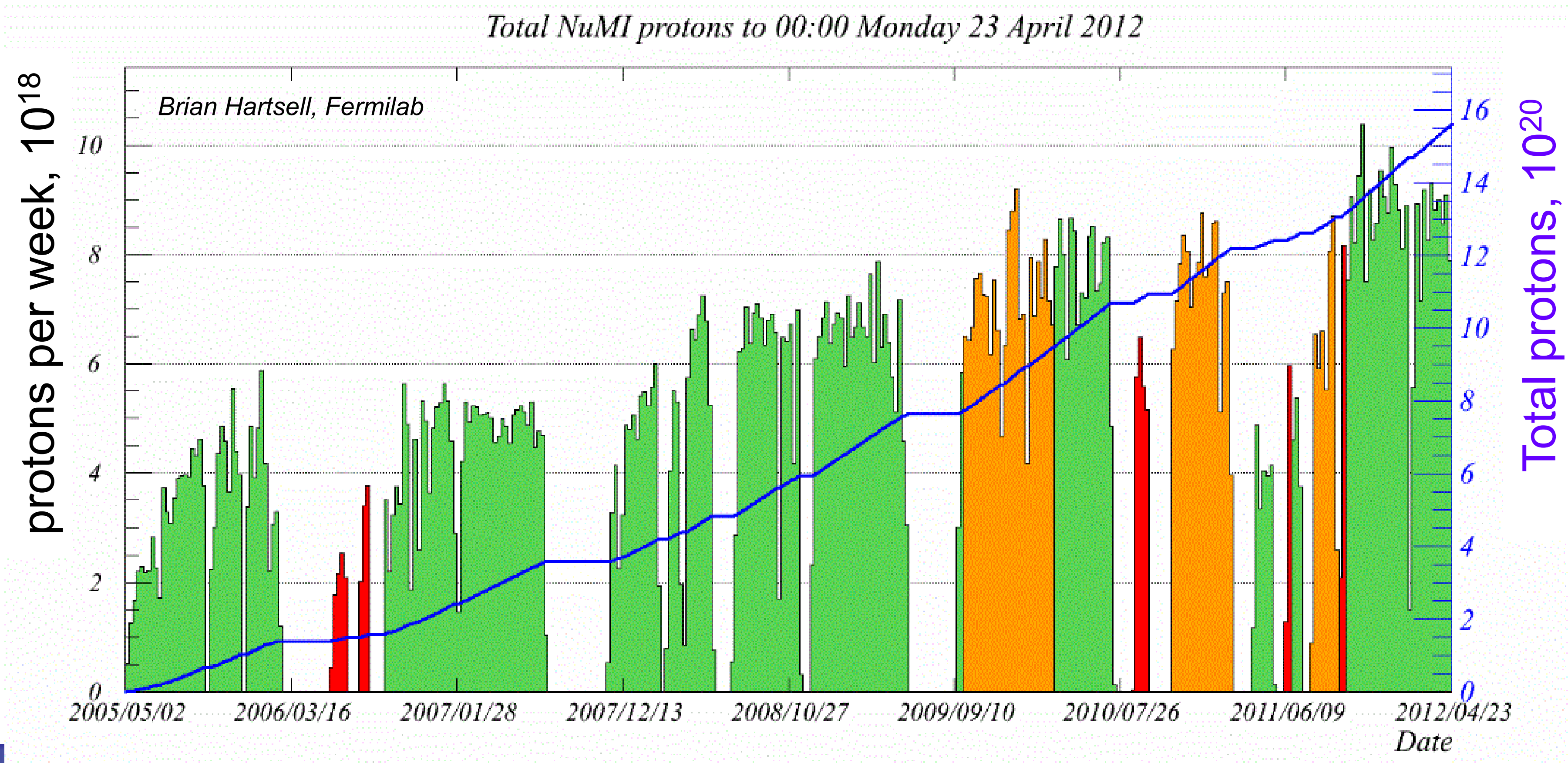


S-200-F, proton irradiation (120keV, RT. 2×10^{18} ions/cm^2), (from Kang et al. Journal of the Korean Physical Society, 63, 2013)

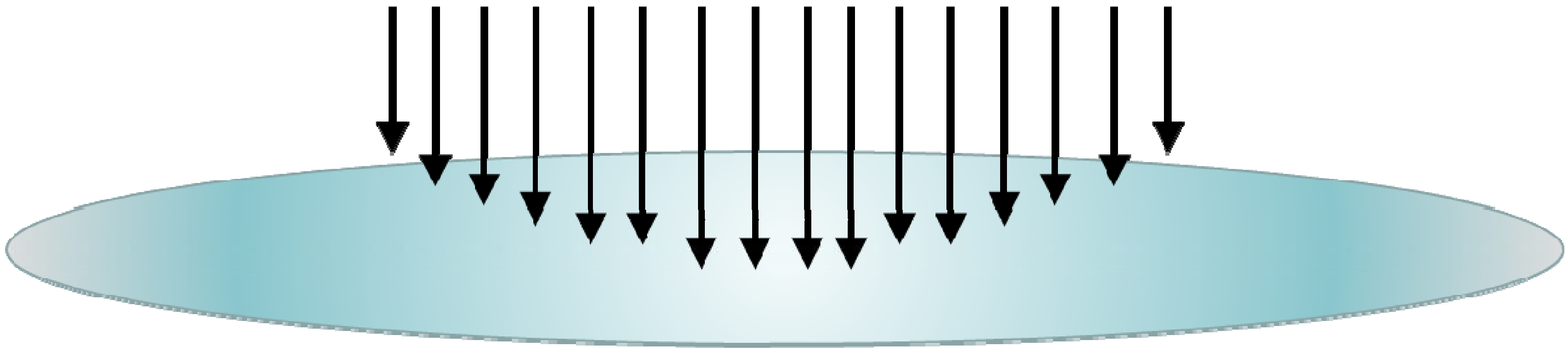
Irradiation Source	He gas production in Be (appm/DPA)
Mixed spectrum fission reactor	10-500
High energy proton beam	4000

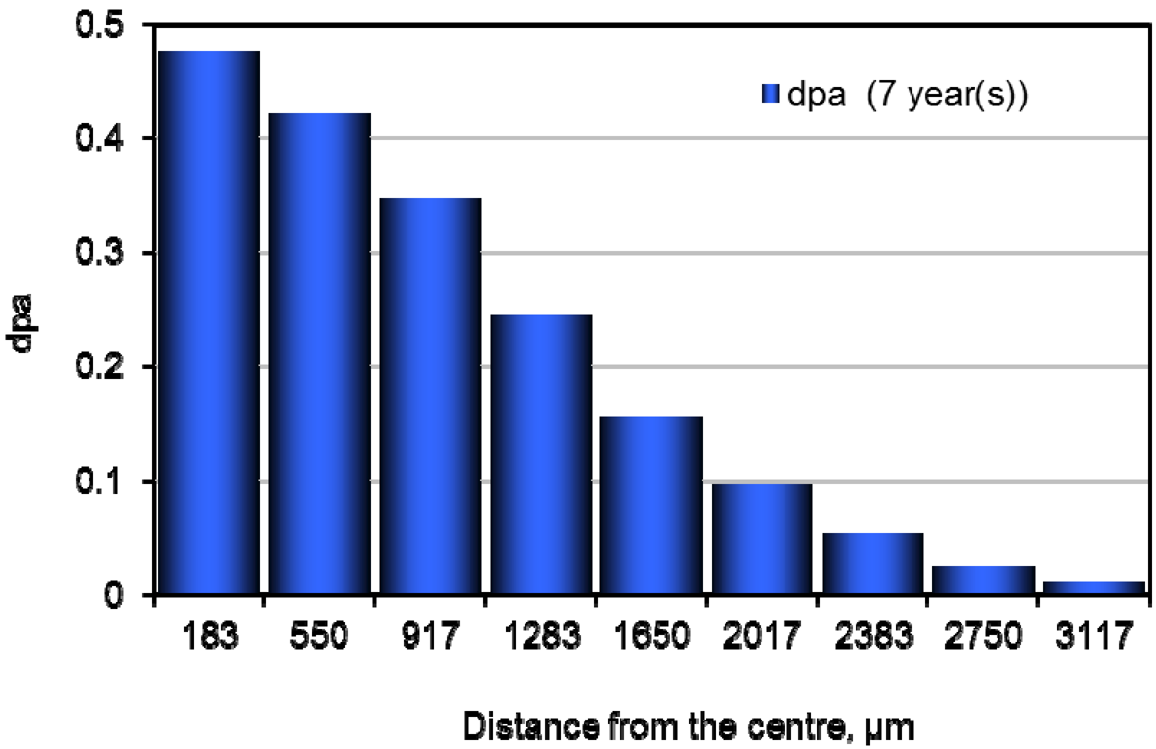
What can we expect from GeV protons?

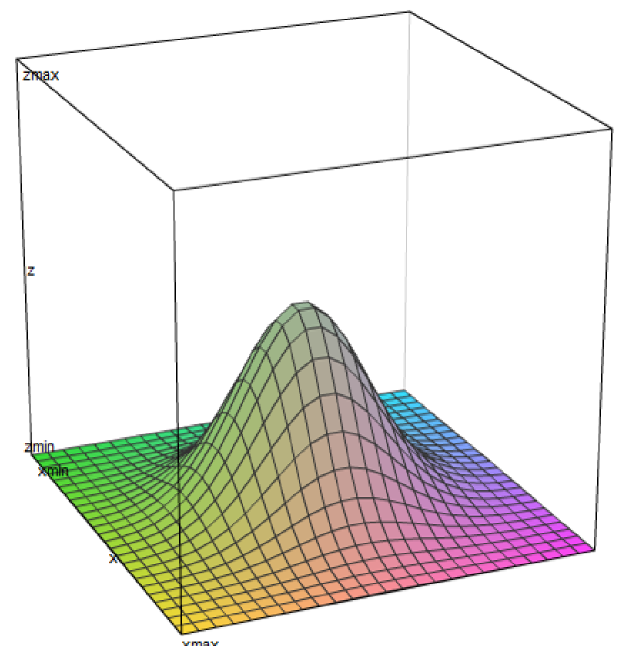
300 kW NuMI beam window
(MARS calculations of Brian Hartsell, Fermilab)
- 120GeV proton beam
- about 3×10^{13} protons per pulse, 0.5 Hz
- 1.57×10^{21} protons during its lifetime
- 1.1mm beam sigmas, X and Y
- T ≈ 200ºC



Gaussian distribution of the beam

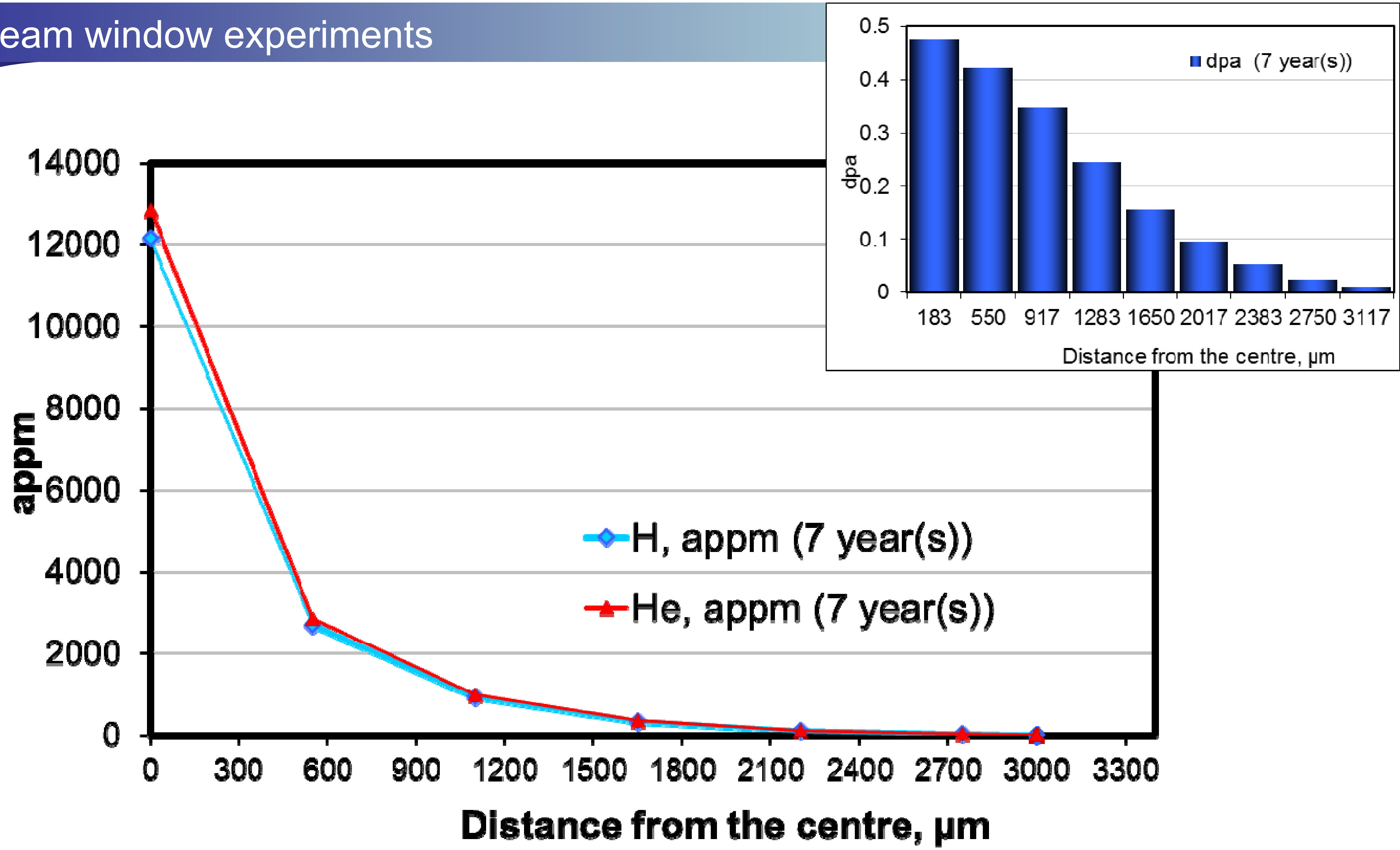






http://www.livephysics.com

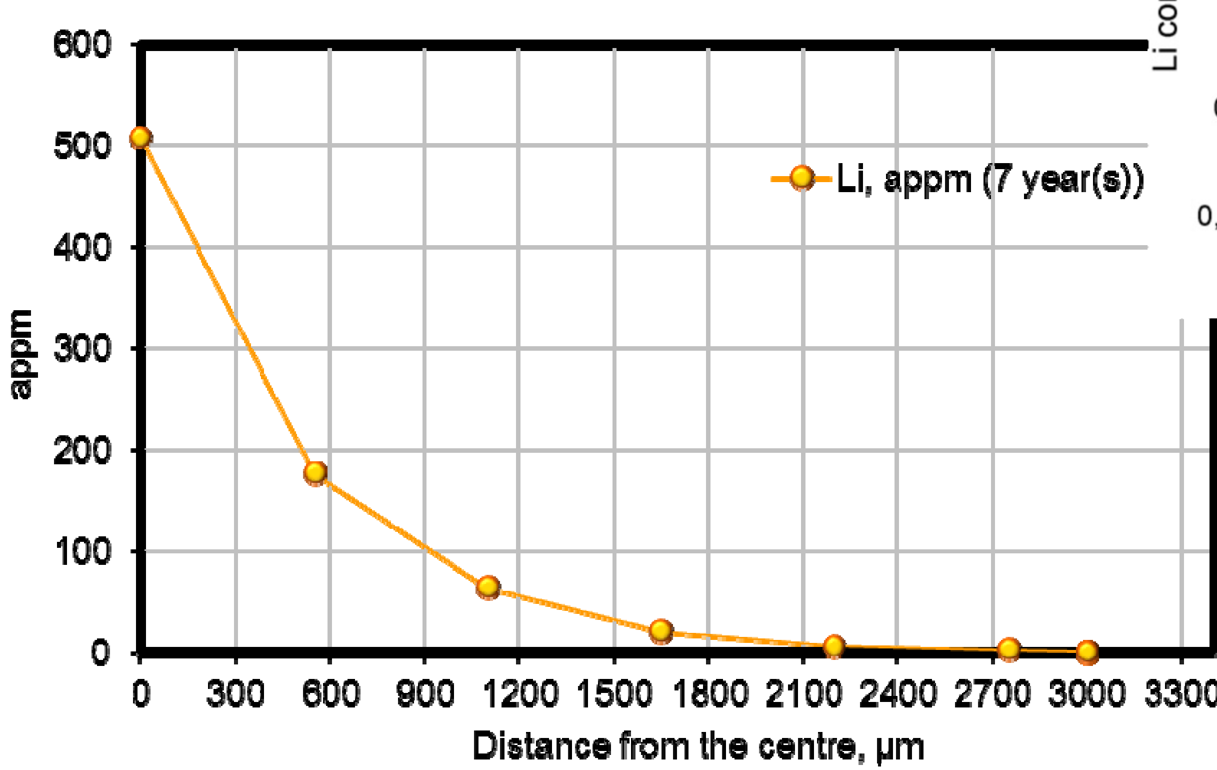
- **Radiation damage distribution is not monotonic**

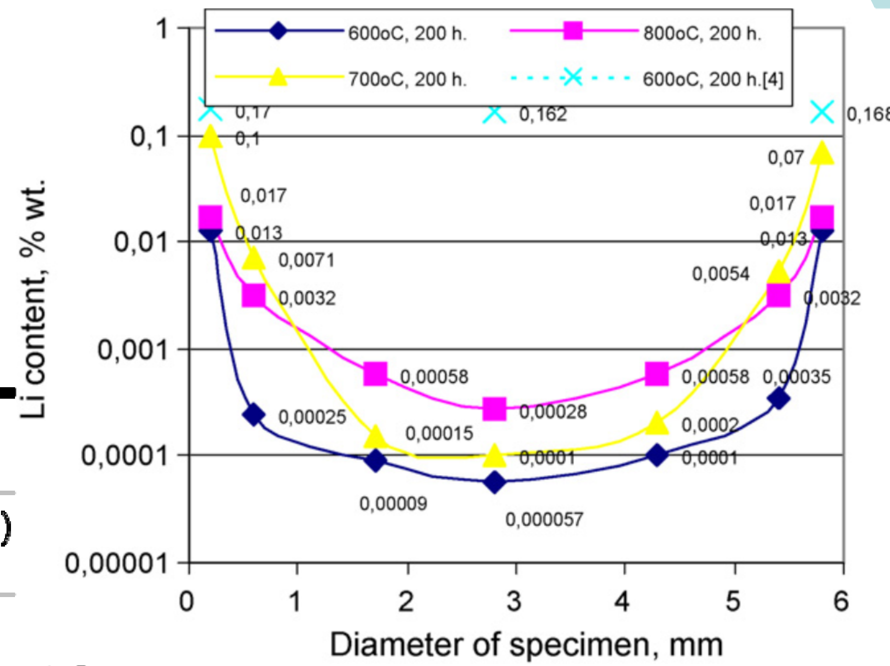


The main transmutation products are He and H

Large difference of dpa and transmutants production is likely to produce non-homogeneous changes across the surface of Be window.

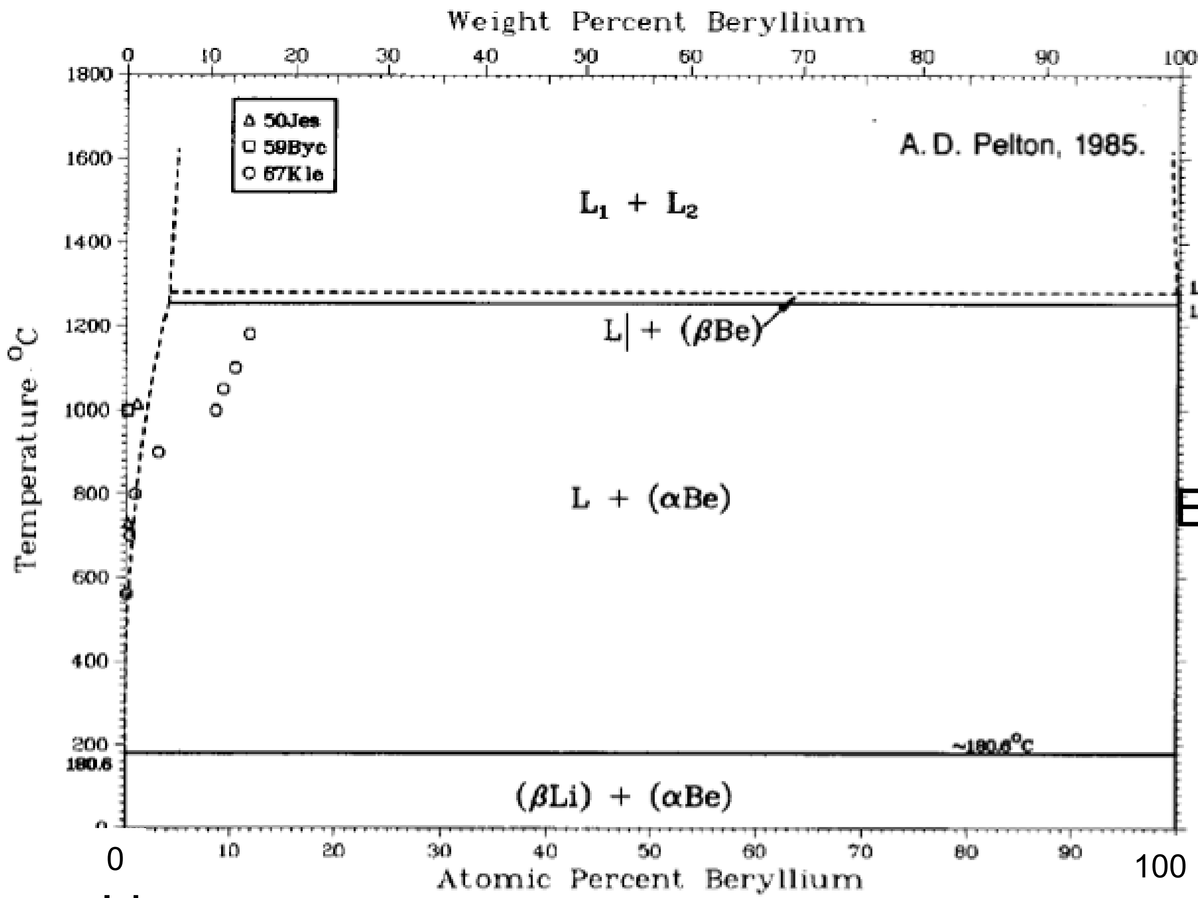
300 kW NuMI beam window
(MARS calculations of Brian Hartsell, Fermilab)
- 120GeV proton beam
- 1.57×10^{21} protons during its lifetime





Be samples annealed in contact with liquid Li. Distribution of Li concentration in beryllium along a diameter of specimens. Penetration of Li into beryllium can cause the degradation of mechanical properties.
I.B. Kupriyanov et al. / Fusion Engineering and Design *2010*

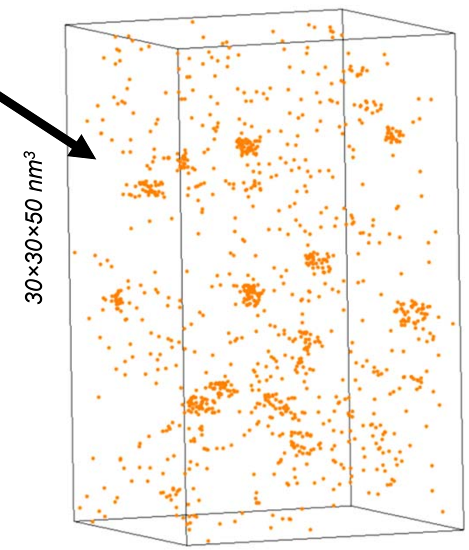
- The quantity of Li is not negligible (up to 500 appm in the centre)
- APT for experimental validation of MARS code



Li-Be phase diagram

Solubility of Li in Be: 130 appm at 700ºC and 40 appm at 600ºC (from Kupriyanov et al. / Fus. Eng. and Des. 2010)

Example



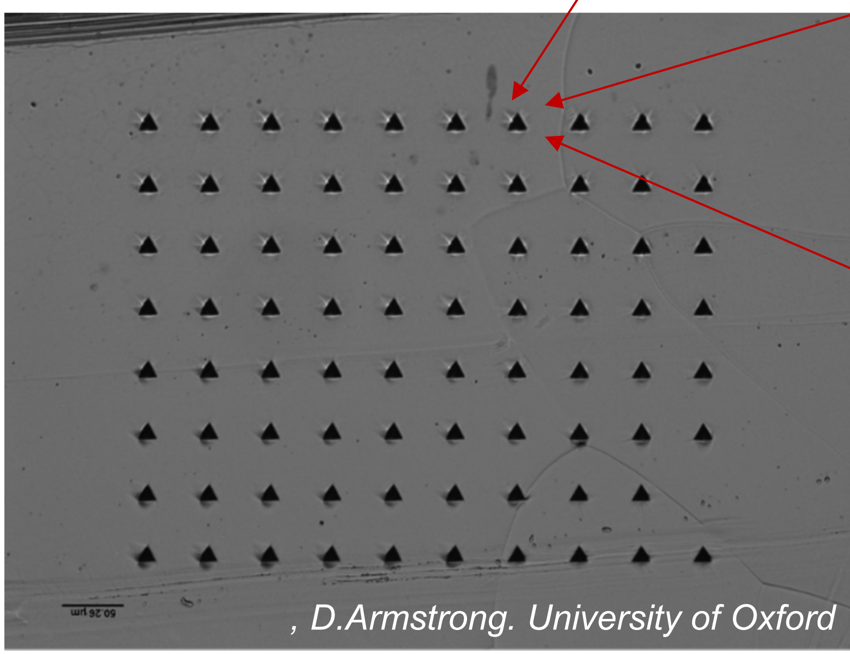
F82H, irradiation 0.5 Gev protons, 350ºC, 20 dpa, 370 appm of Ca created
Kuksenko et al. / JNM 2014

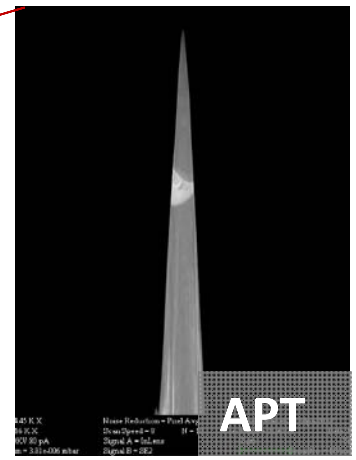
- Li is not soluble in Be. Will it segregate?
- T_{melt}(Li) = 181ºC (for bulk lithium). Can we expect the creation of liquid phase in the window?

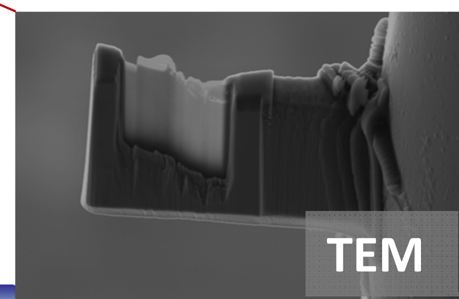
Nano-hardness measurements:

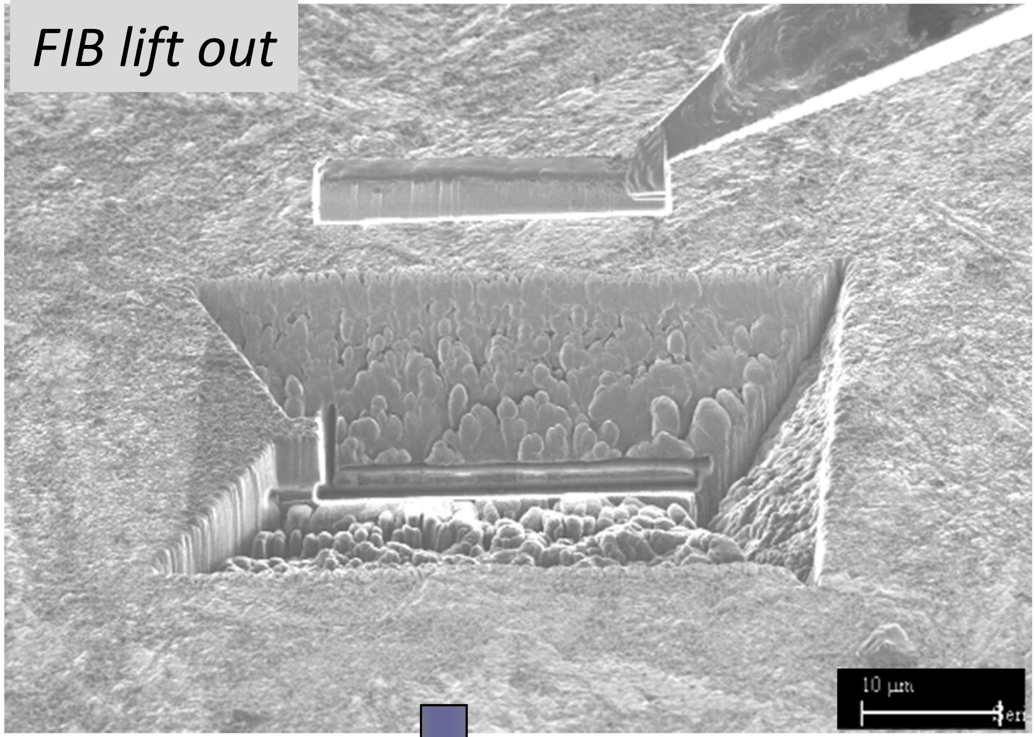
- to find the Gaussian peak
- to estimate the irradiation effect

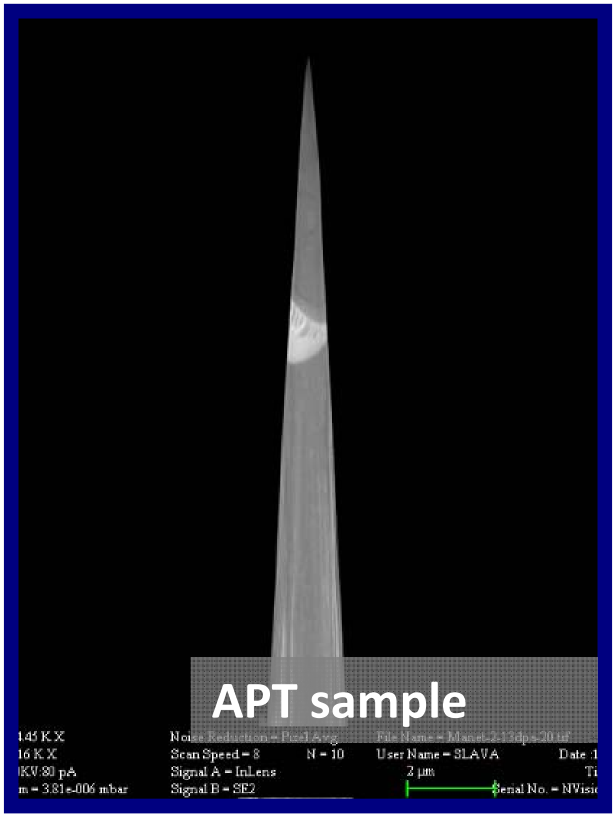
Local microstructural investigations

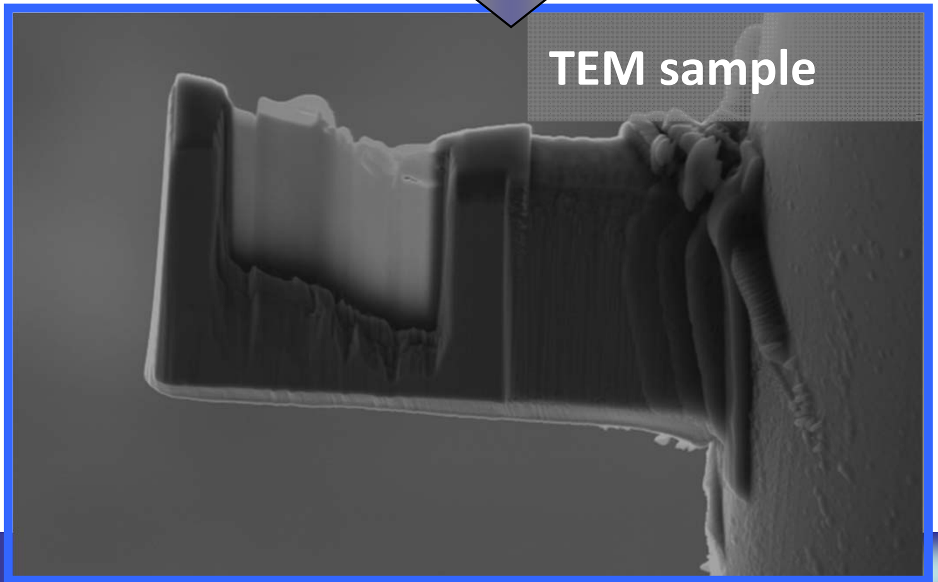






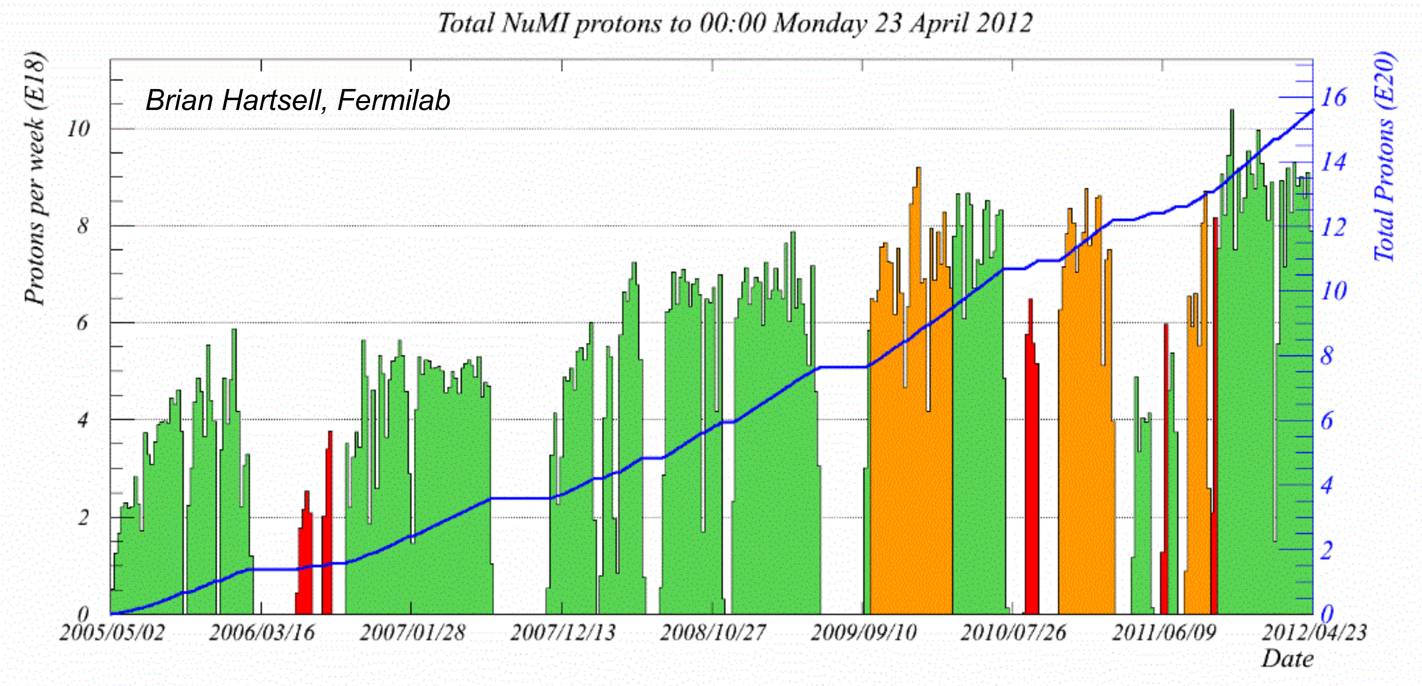






FIB lift-out

- superpose the microstructural data with the dpa, appm and hardness data
- minimize the activity of samples
- minimize the toxicity of samples



We need to know the evolution of radiation effects over the time

Collaboration with HiRadMat project (poster of Kavin AMMIGAN)

Ion irradiation experiments

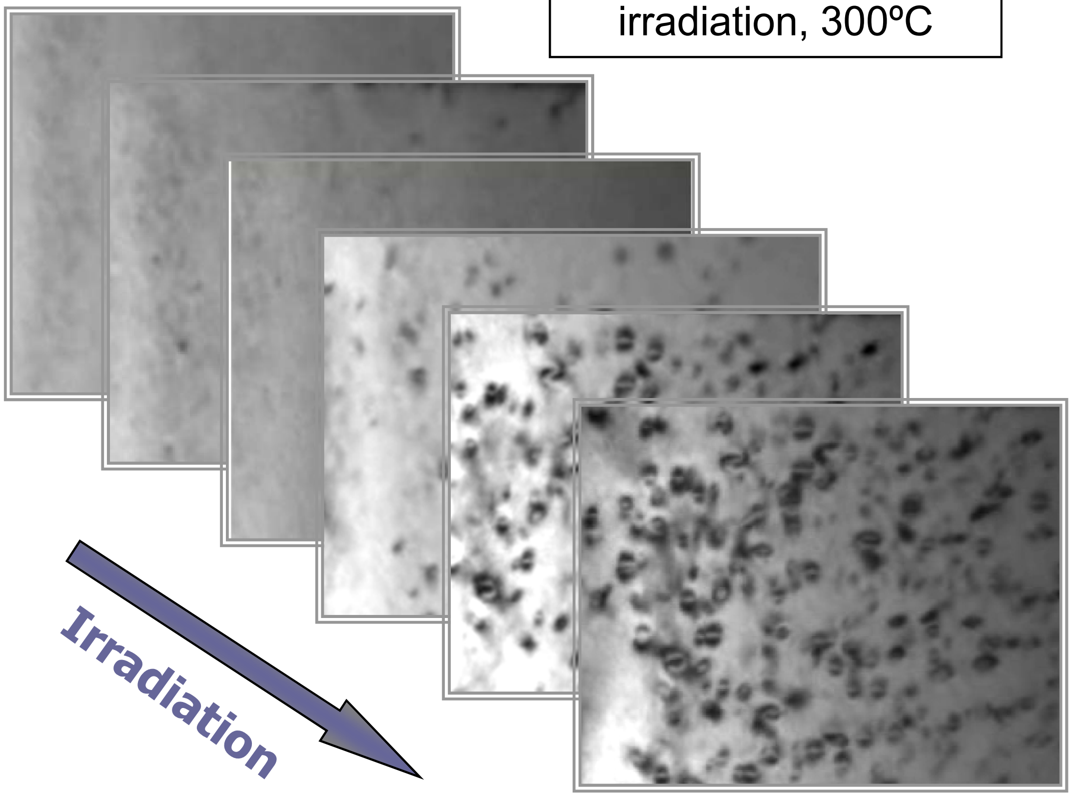
University of Huddersfield

Microscope and Ion Accelerator for Materials Investigations facility (MIAMI) University of Huddersfield , UK
(collaboration with Prof. S E Donnelly)


From http://www.hud.ac.uk/research/researchcentres/emma/miami/

Ions: He+
Beam energy: ~ 10keV => peak of damage in the middle of TEM foil (SRIM)
Dose: up to 1 dpa
Temperature: 200ºC (300ºC, 600ºC)

Fe-9Cr alloy,
150keV Fe^{+} ions
irradiation, 300ºC



In-situ observations of the evolution of the microstructure

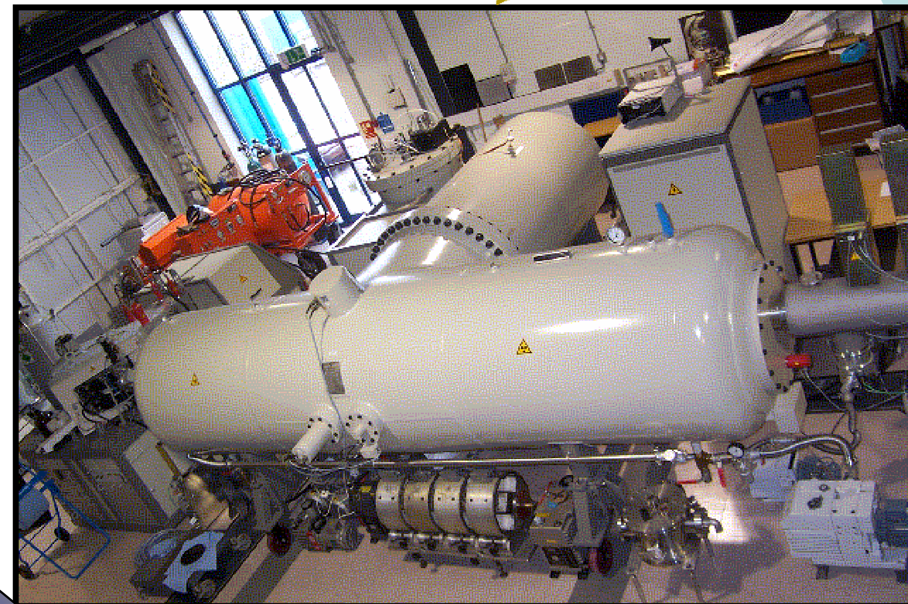
- evolution of number density and size of dislocation loops and/or He;
- estimation of mobility of point defect clusters
- Burgers vector and loops nature determination*

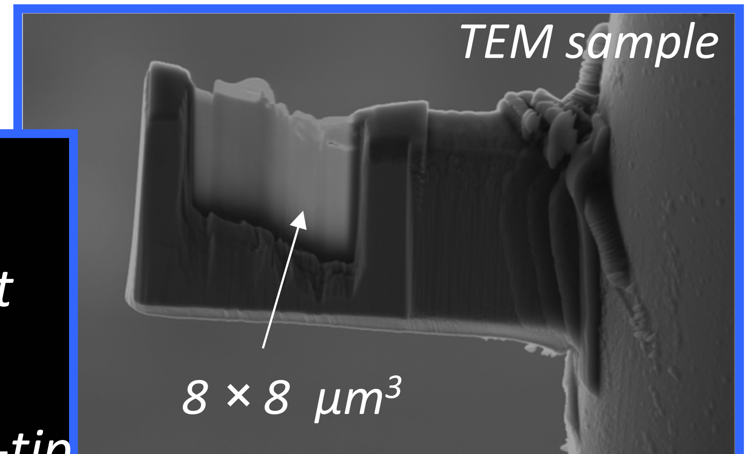
But: effect of the surface

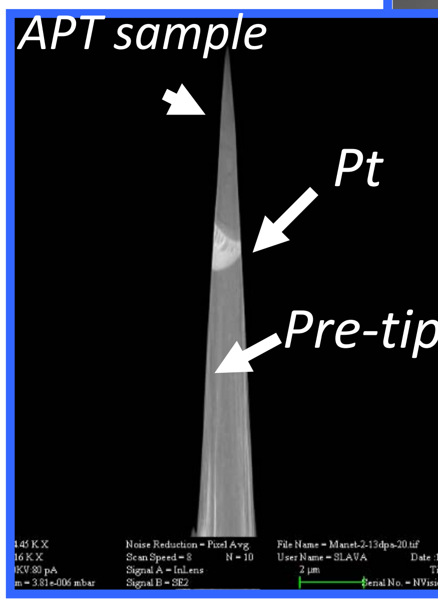
Irradiation of APT tips?

Surrey Ion Beam Centre, UK
(collaboration with Prof. R.Gwilliam)

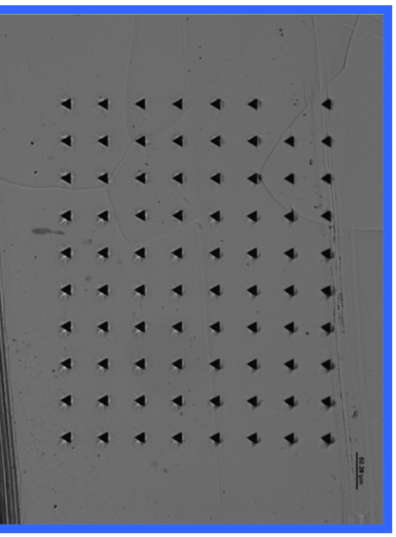
Ions: He+
Maximum beam energy: 2 MeV => 7.5µm implantation depth (SRIM)
Dose: up to 1 dpa
Temperature: 200ºC (100ºC, 400ºC)

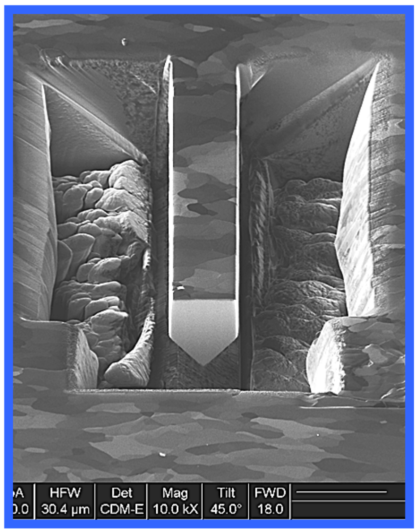




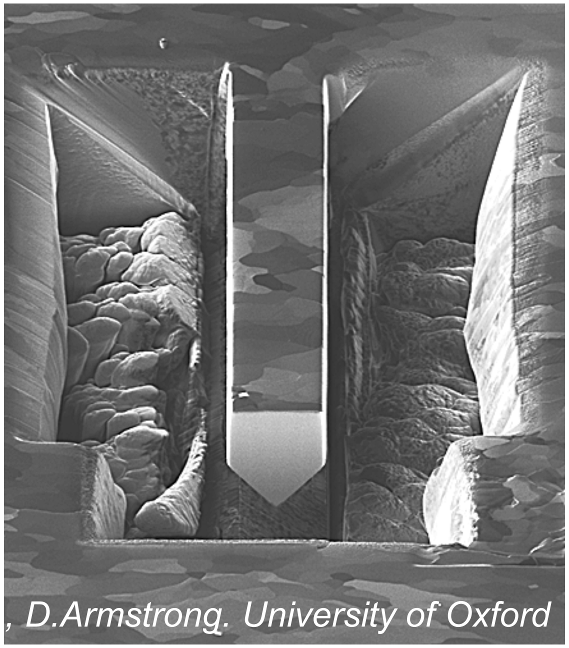


Micromechanical tests

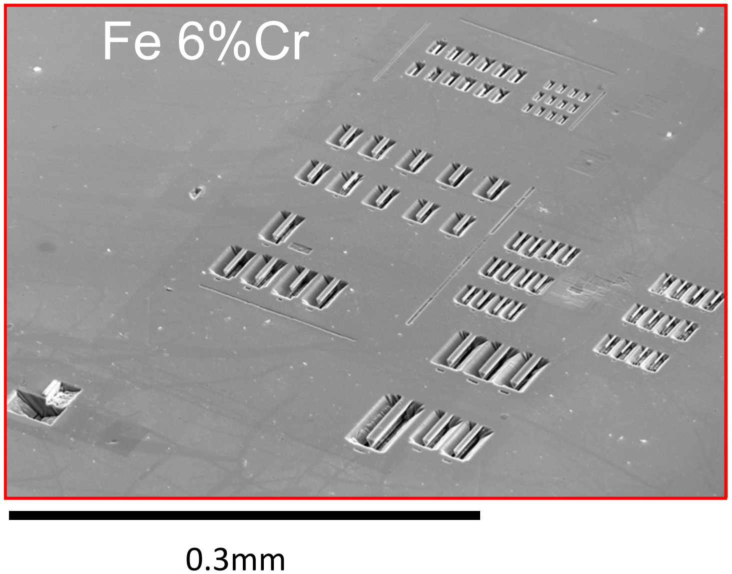


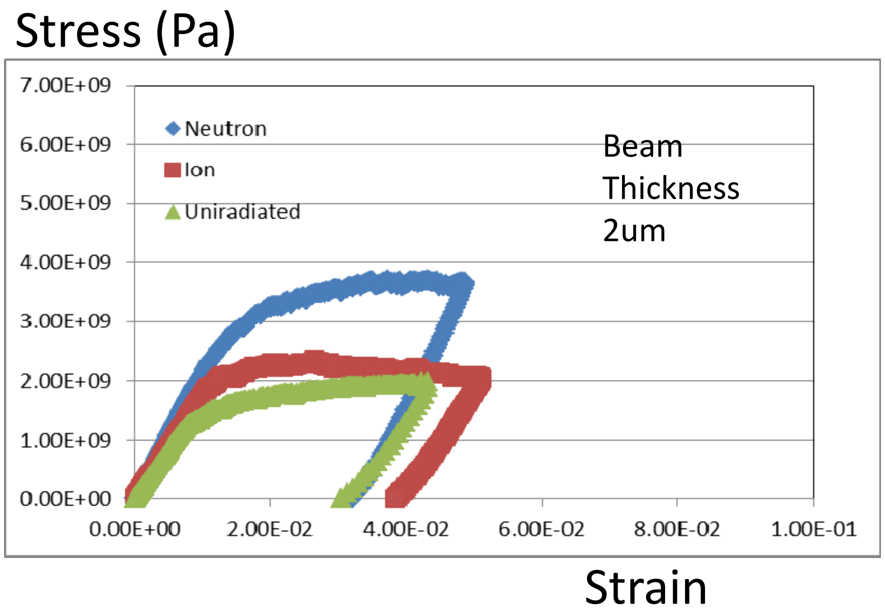


- Useful where only small samples are available (implanted layer)
- Need for a sample design that can be machined in surface of bulk samples
- Geometry that can be manufactured quickly and reproducibly



Chris Hardie
University of Oxford





Conclusions

Experimental database of the high-energy proton irradiation effects in Be is very limited

Experimental investigation of beryllium within Radiate project should cover 3 main goals:

- characterization of existing GeV proton irradiated Be samples;
- simulation of proton irradiation effect by ion implantation experiments;
- prediction of the microstructural evolution for new irradiation conditions.

Thank you for your attention!

What do we know?

enhancement of phase transformation

$$D^* = \alpha_V D_V C_V^* + \alpha_x D_x C_x^*$$

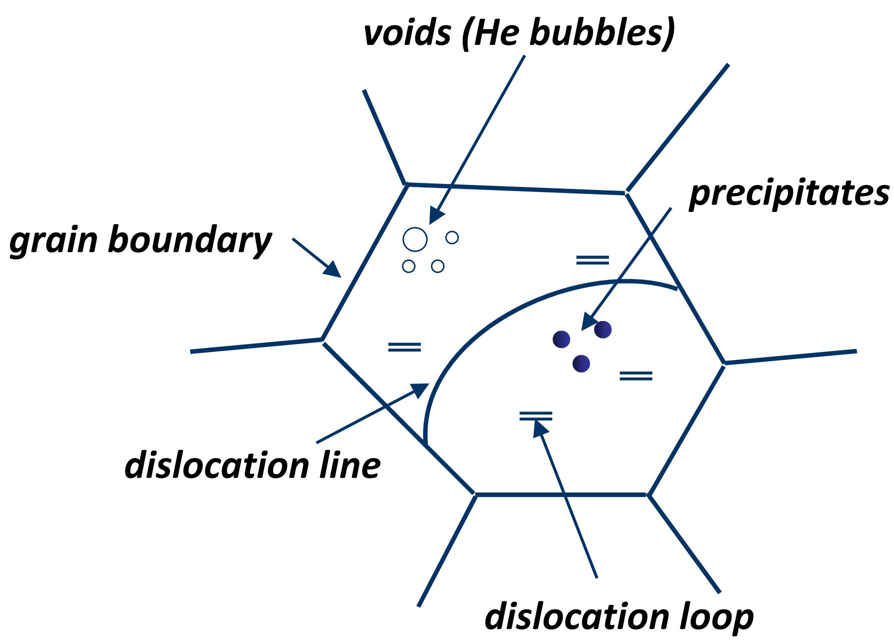
$$C_V^* > C_V^T$$

X - self interstitial atom; clusters of point defects

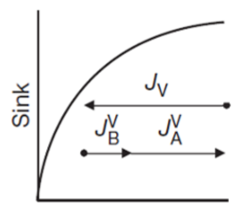
agglomeration of point defects

- self-interstitials
  - clusters;
  - dislocation loops.
- vacancies
  - voids;
  - dislocation loops.

segregation (precipitation) or depletion on point defect sinks

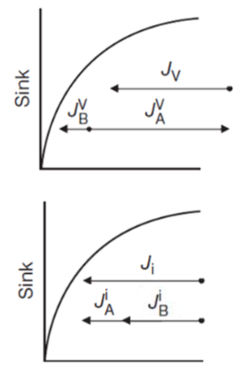


V- vacancy; I - interstitial



inverse Kirkendall effect

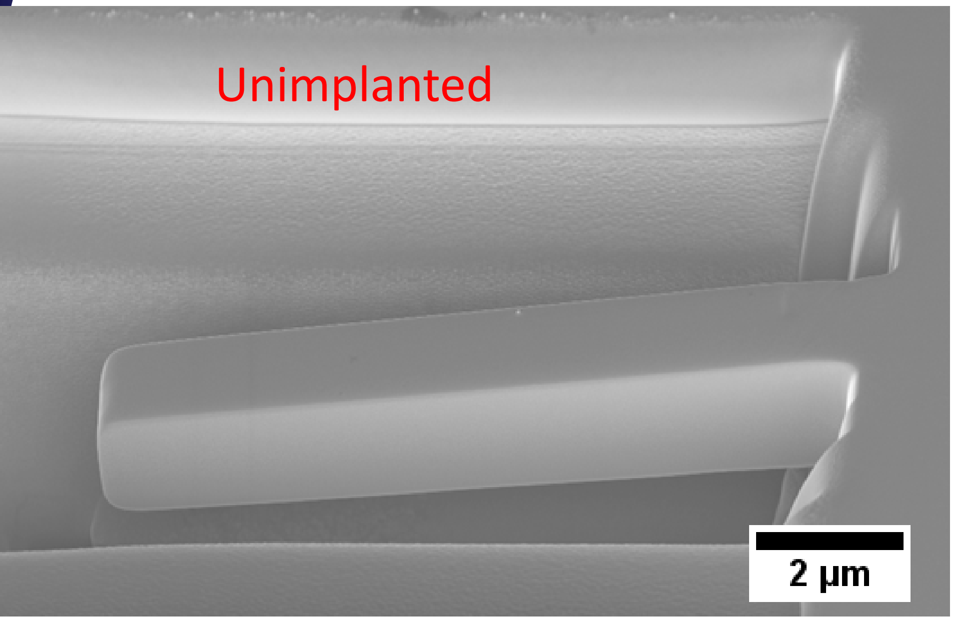
If $D_B^V < D_A^V$ ⇒ depletion of A atoms

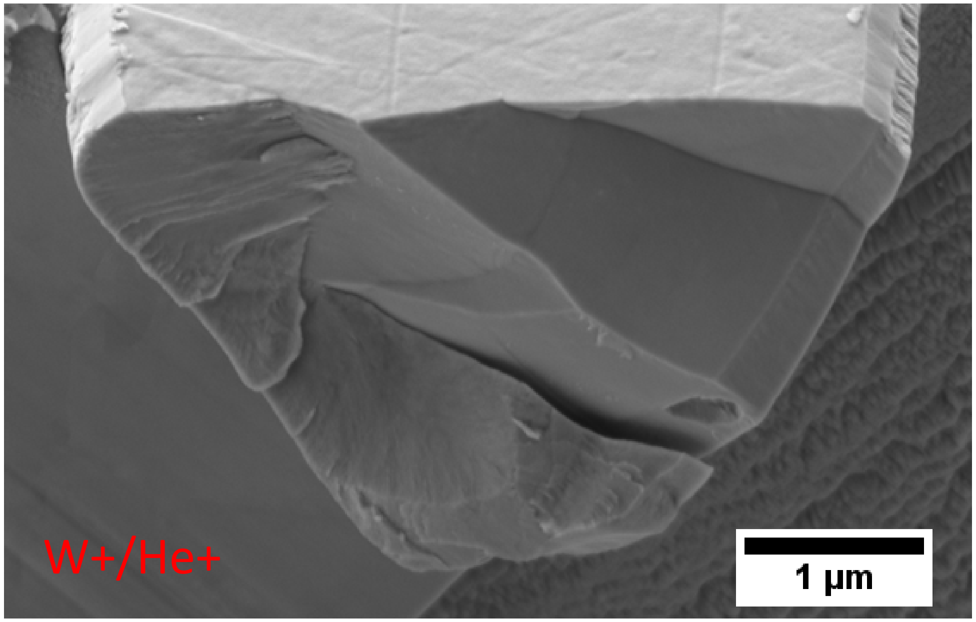


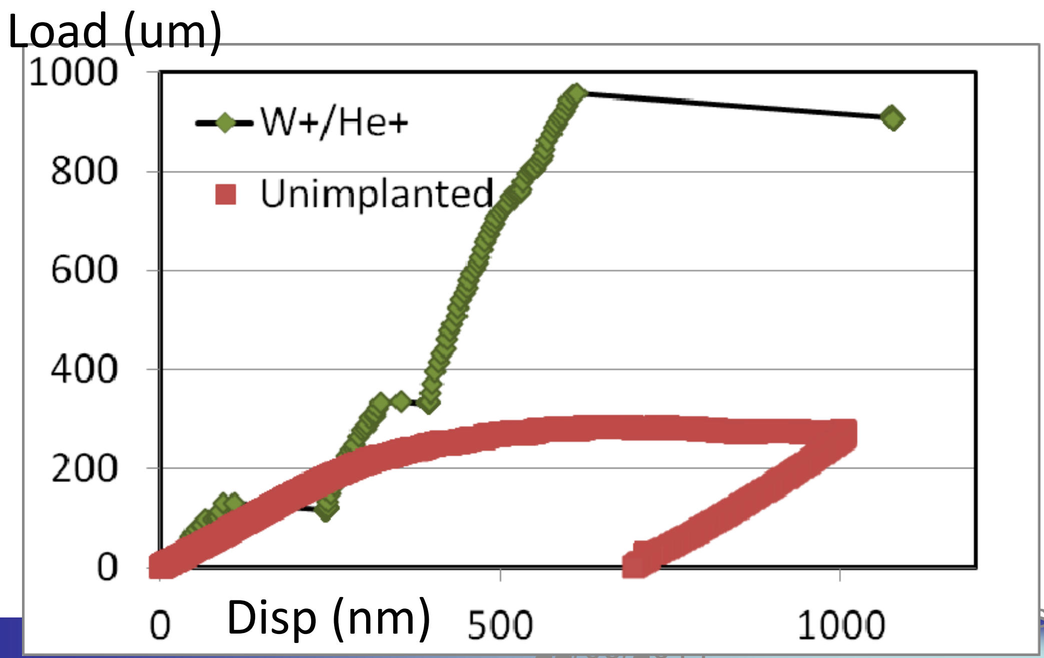
drag effects

B-V complexes or B-I complexes ⇒ Segregation of B atoms

Tested Cantilevers



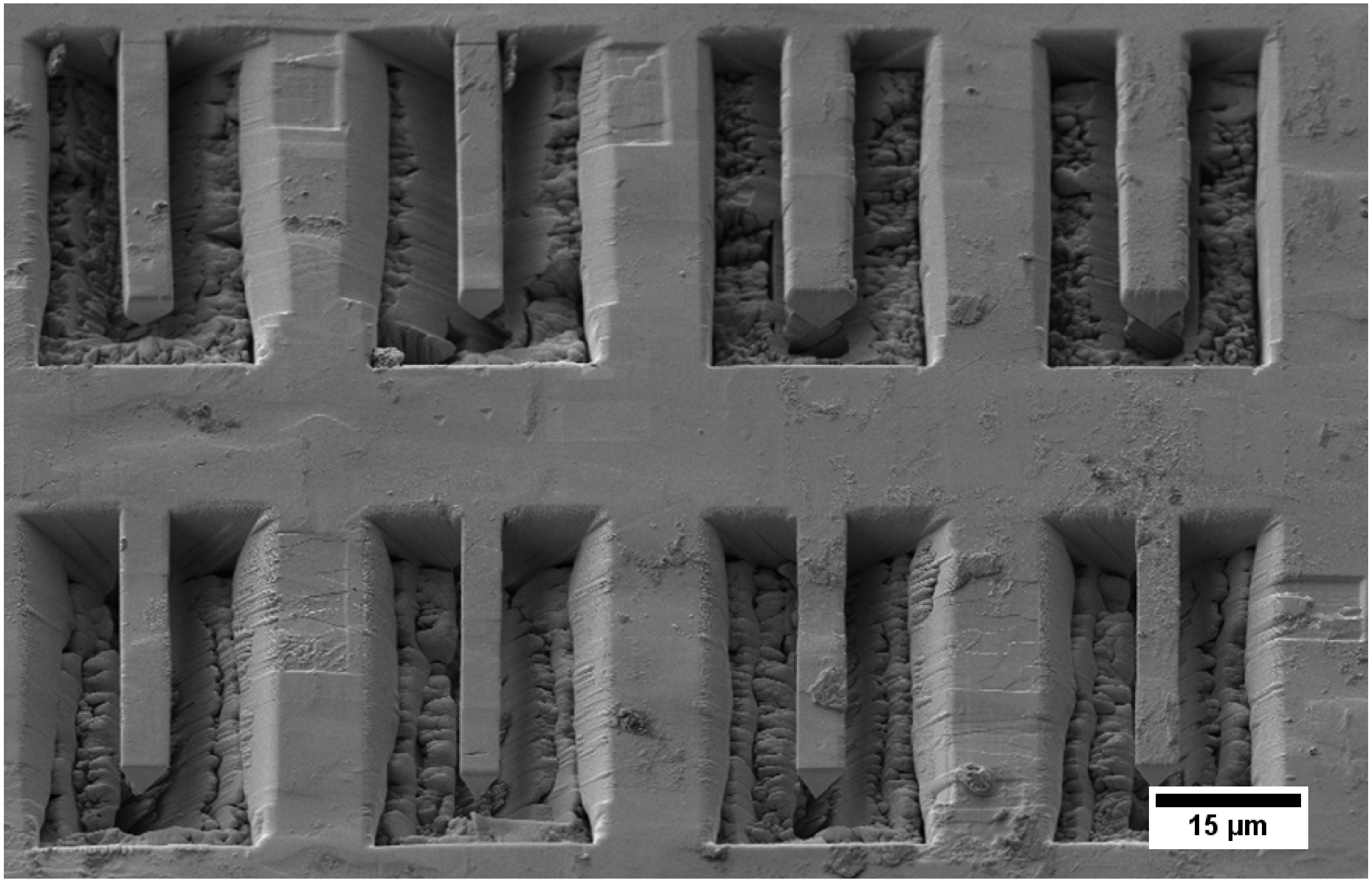




Helium has complex effects on both yield and fracture properties of tungsten

Differences between results for micro-cantilevers and nanoindentation show the difficulty of relying on one type of test

Micro Cantilevers Before Testing



Precipitates **Fe and Al rich precipitates** may affect **ductility** and **creep strength** (A.W. Jones, R.T. Weiner, J. Common Met. 6 (1964) 266.)

Grain, twin and sub-grain boundaries and dislocations can be the preferential places for precipitation of **Fe-rich phases** during ageing of Be-0.25%Fe. Dislocation can locked by precipitates leading to the increase of **hardness** (S. Morozumi, N. Tsuno, S. Koda, Trans. Jpn. Inst. Met. 10 (1969) 64.)

Intermetallic **Fe/Al/Be inclusions** are the preferential sites for **corrosion** pit initiation, some corrosion pits had also initiated at elemental **Si** and **carbide** inclusions. (J.S. Punni, M.J. Cox, Corros. Sci. 52 (2010) 2535)

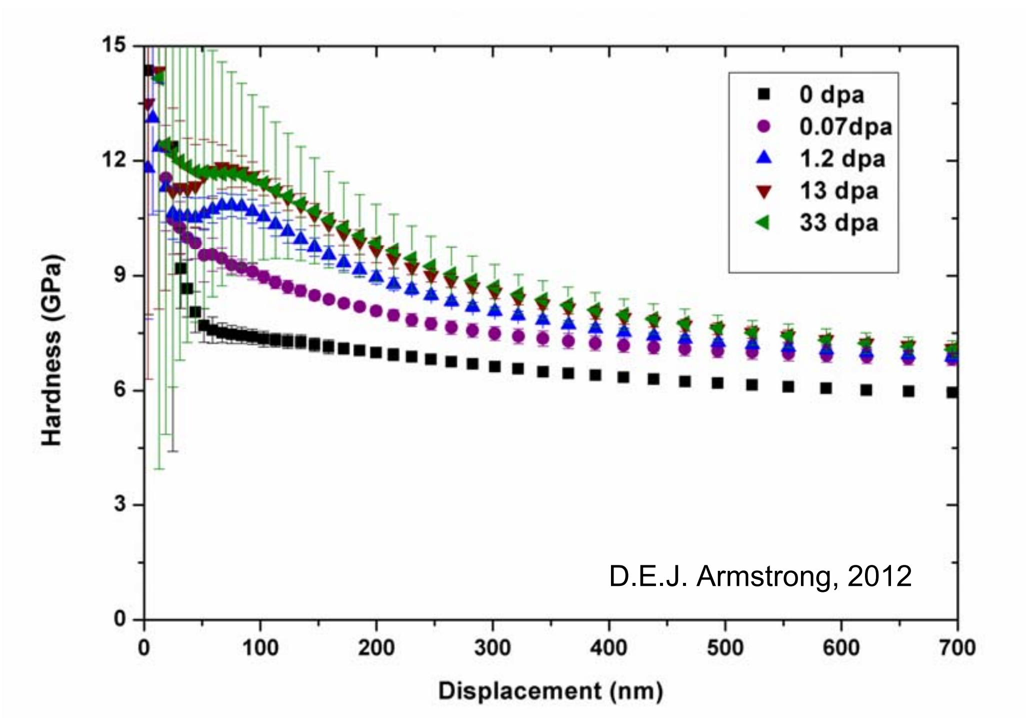
Al and Mg can form **low melting point eutectics** in Be, that might influence the mechanical behaviour of Be.

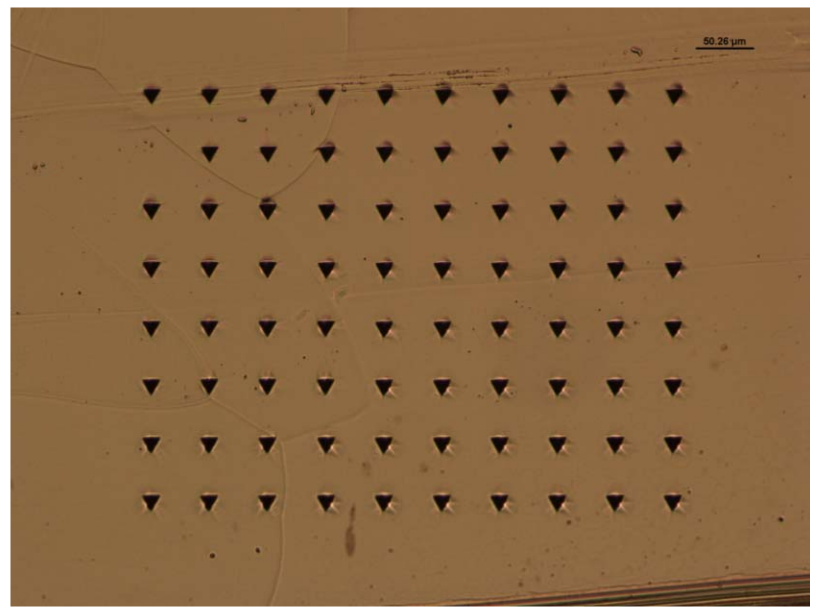
e^(-((x^2)/3+(y^2)/3))

Nanoindentation mechanical probe which allows local hardness and modulus to be measured

Micrometre

Hardness of W5Ta after self-ion irradiation





Will be used for high-energy ion irradiation samples and NuMi window (if not too "hot")