SAND2014-17969PE

Continuum-scale modeling of hydrogen and helium bubble growth in metals

R. Kolasinski[a], M. Shimada[b], D. Cowgill[a], D. Buchenauer[a], and D. Donovan[c]

[a]*Sandia National Laboratories, Hydrogen & Metallurgy Science Dept., Livermore, CA*
[b]*Idaho National Laboratory, Fusion Safety Program, Idaho Falls, ID*
[c]*University of Tennessee, Dept. Of Nuclear Engineering, Knoxville, TN*

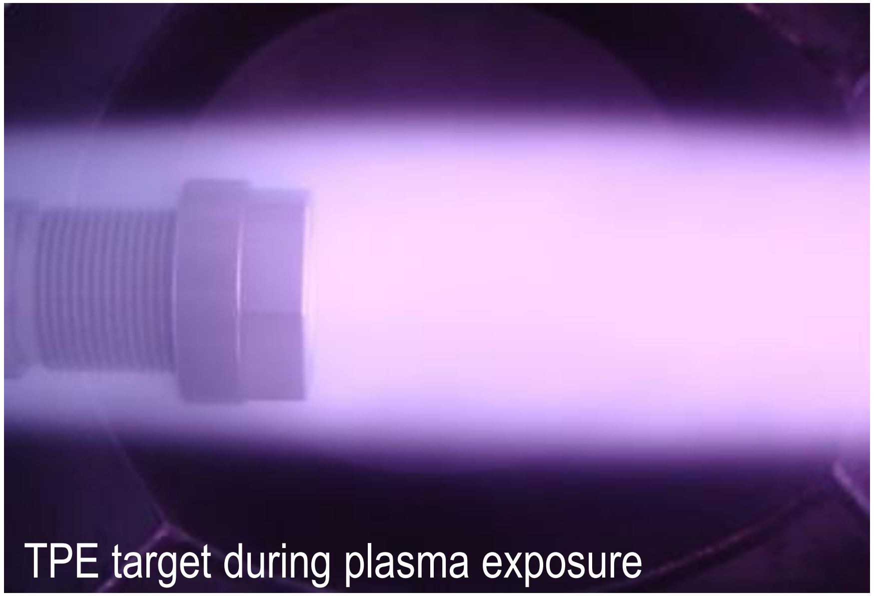
Y. Oya[d], T. Chikada[d], and K. Michibayashi[f]

[d]*Shizuoka University, Department of Chemistry, Shizuoka, Japan*
[f]*Shizuoka University, Department of Geosciences, Shizuoka, Japan*

Tritium Focus Group Meeting *September 24, 2014*

Motivation: Analysis of bubble growth in ITER-grade W samples exposed in TPE

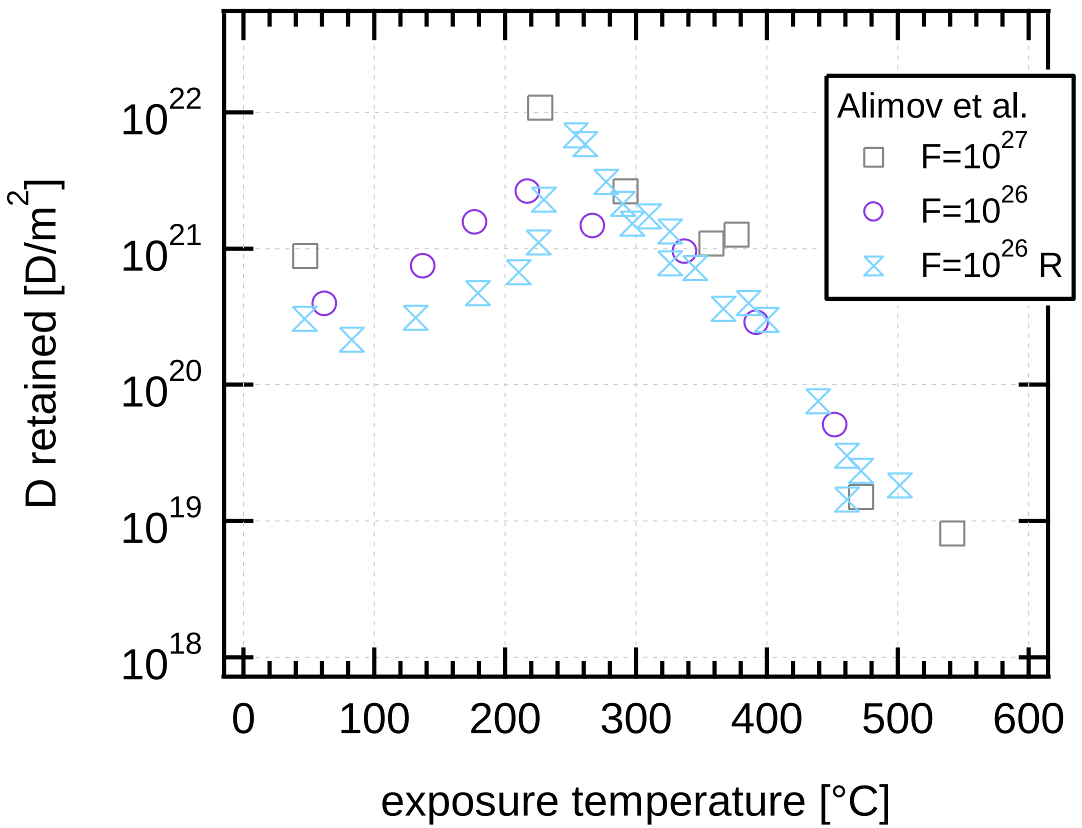
- Precipitation affects migration through material
- Bubble growth depends on microstructure
- Growth mechanisms critical to developing realistic models



exposure type	ion energy [eV]	duration [min]	flux (Γ_i) [m^{-2} s^{-1}]	fluence (Φ) [m^{-2}]
LF	100	60	4.9×10^{21}	1.8×10^{25}
HF	100	120	1.5×10^{22}	1.1×10^{26}

- TPE plasma exposures at INL
- Microscopy at Shizuoka

Retention measurements correspond closely with those obtained in other laboratories



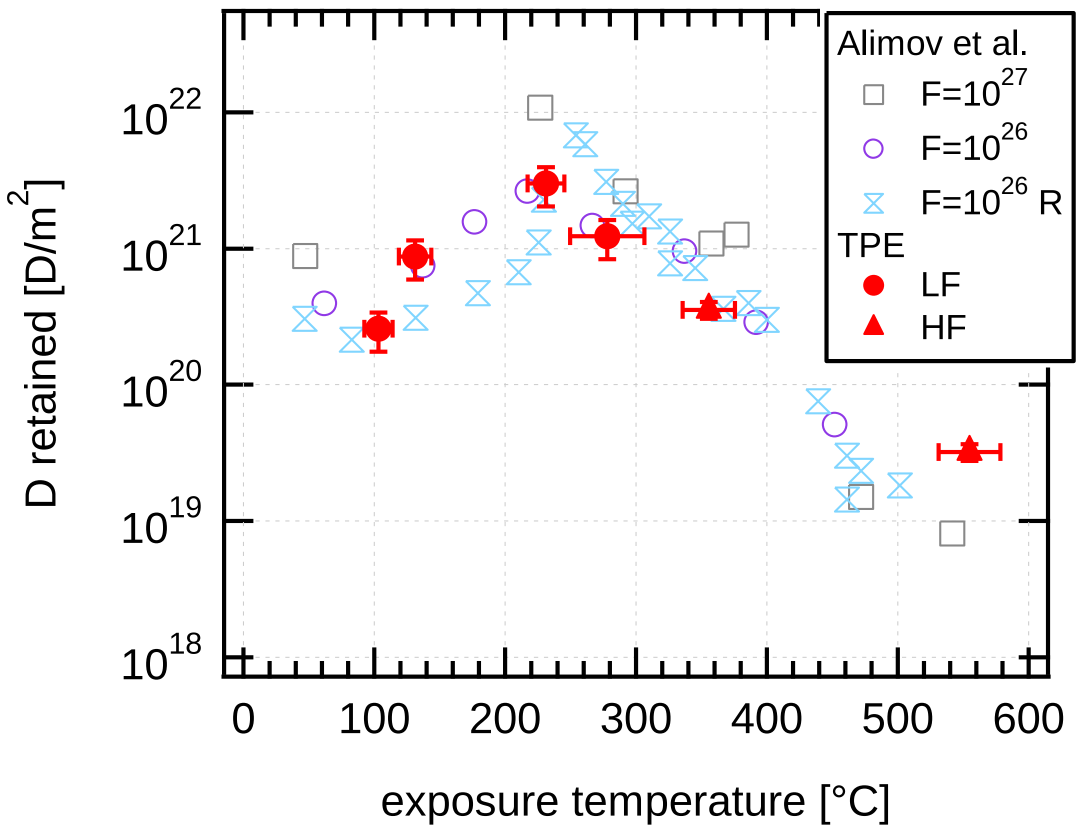
V. Kh. Alimov, et al. *J. Nucl. Mater.* **420** (2012) 519.

Previous work by Alimov et al:

- ITER-grade W
- E = 38 eV
- $\Phi = 10^{22}$ D m^{-2} s^{-1}

Comparable exposure conditions

Retention measurements correspond closely with those obtained in other laboratories



V. Kh. Alimov, et al. *J. Nucl. Mater.* **420** (2012) 519.

Previous work by Alimov et al:

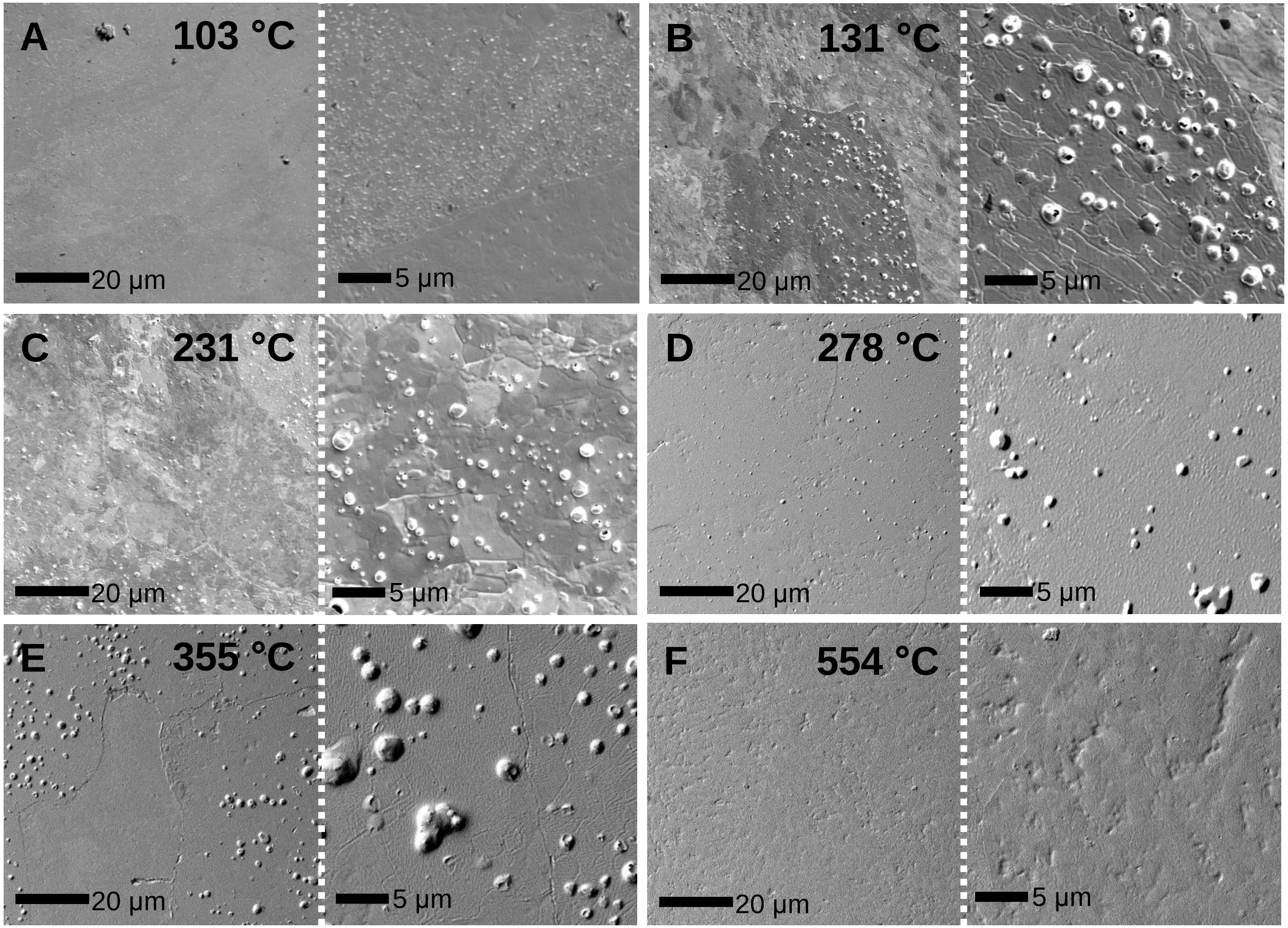
- ITER-grade W
- E = 38 eV
- $\Phi = 10^{22}$ D m^{-2} s^{-1}

Comparable exposure conditions

TPE retention measurements:

- Correspond closely with Toyama/IPP meas.
- Confirm accepted retention temp. dependence.

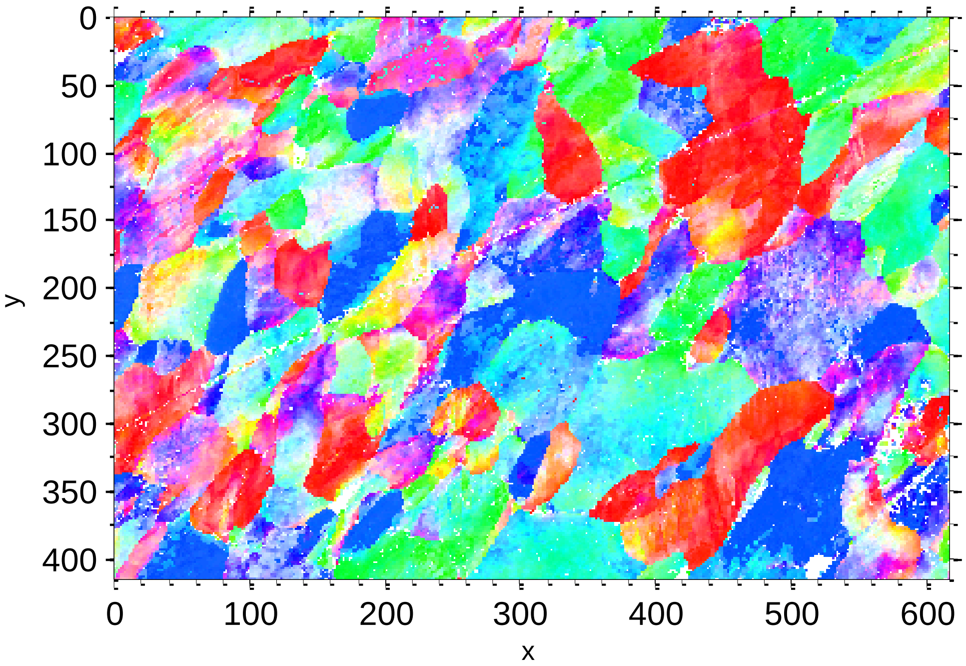
Surface morphology variation with temperature



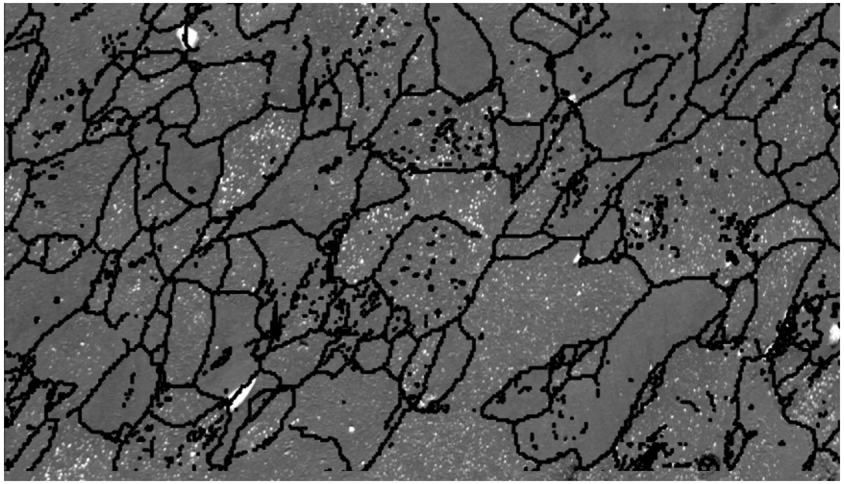
Key features:

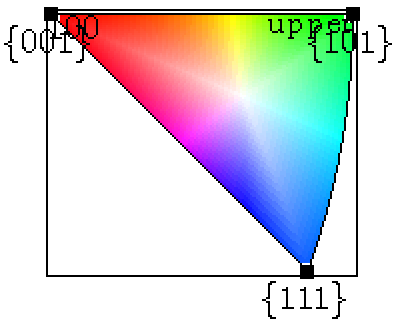
- Non-uniform coverage
- Bubbles are small (<10 μm dia.) compared with warm-rolled W material.
- Absent at temperature extrema.

EBSD measurements reveal dependence on grain orientation



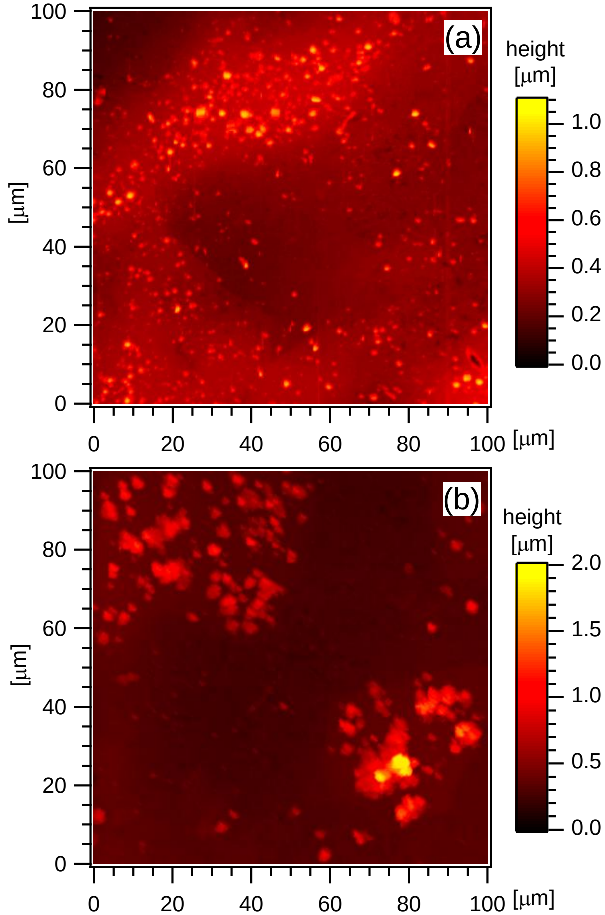
SEM image of the same area





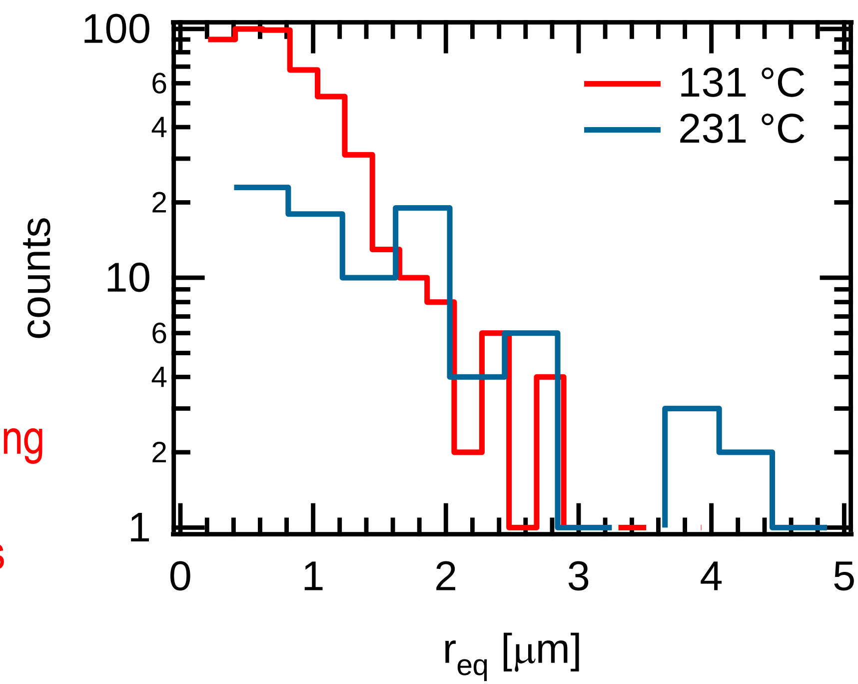
- Grain orientation indicated by inverse pole plot.
- Bubbles visible on grains with <111> and <110> directions aligned normal to surface
- Considerable distortion within individual grains
- Un-annealed sample showed increased distortion

Atomic force microscopy reveals details of surface structure



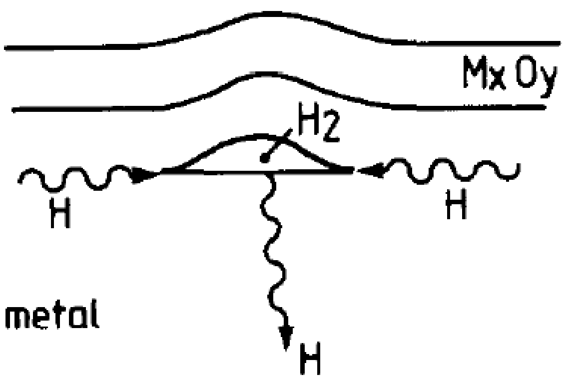
- Atomic force microscopy provides information on the shape of the deformed surface.
- Individual bubbles identified and analyzed automatically.

corresponding bubble size distributions

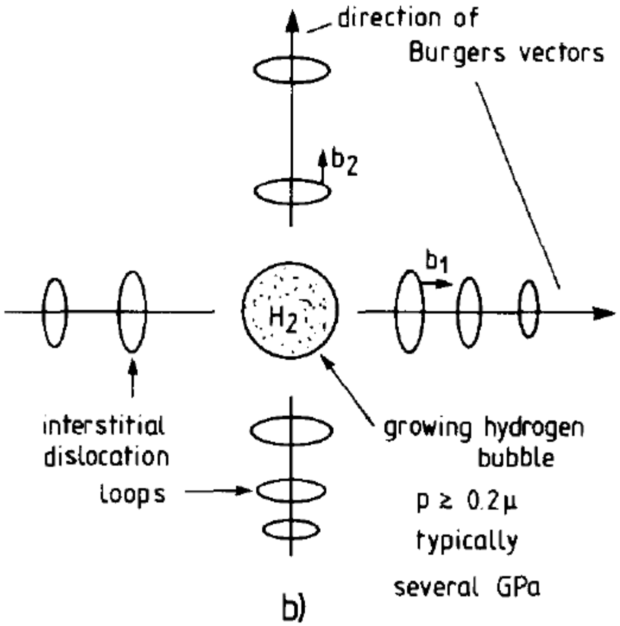


What bubble growth mechanisms are active in W during plasma exposure?

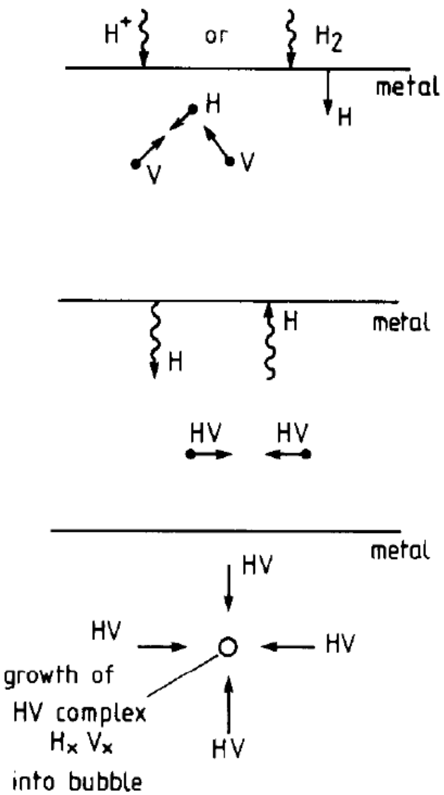
near-surface plastic deformation



dislocation loop punching



vacancy clustering



Figures from: J. B. Condon & T. Schober, *J. Nucl. Mater.* **207** (1993) 1.

Far from the free surface, dislocation loop punching is favored

Three bulk precipitate growth mechanisms considered:

- Dislocation loop punching

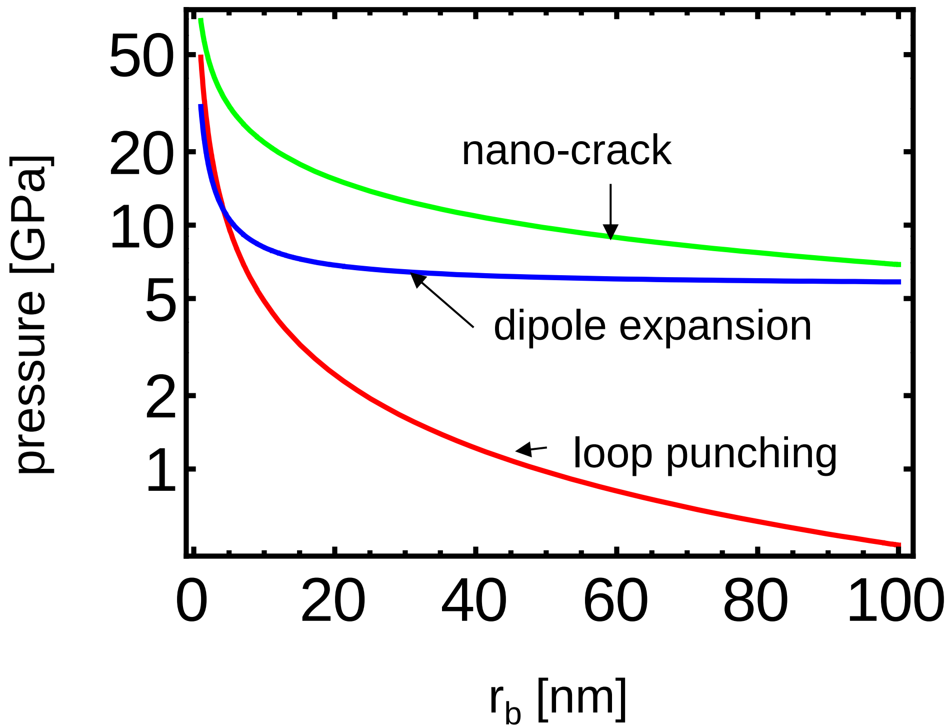
$$p_{LP} \geq \frac{2\gamma}{r} + \frac{\mu b}{r} \sim \frac{1}{r}$$

- Griffith nano-crack extension

$$p_{NC} \geq \sqrt{\frac{\pi\mu\gamma}{(1-\vartheta)r}} \sim \frac{1}{\sqrt{r}}$$

- Dislocation dipole expansion

$$p_{DE} \geq \frac{2\gamma}{s} + \frac{\mu d}{2r} \sim \frac{1}{r} + c$$



Based on methods developed in:
D. F. Cowgill, "Physics of He Platelets in Metal Tritides," in *Effects of Hydrogen on Materials* (2009).

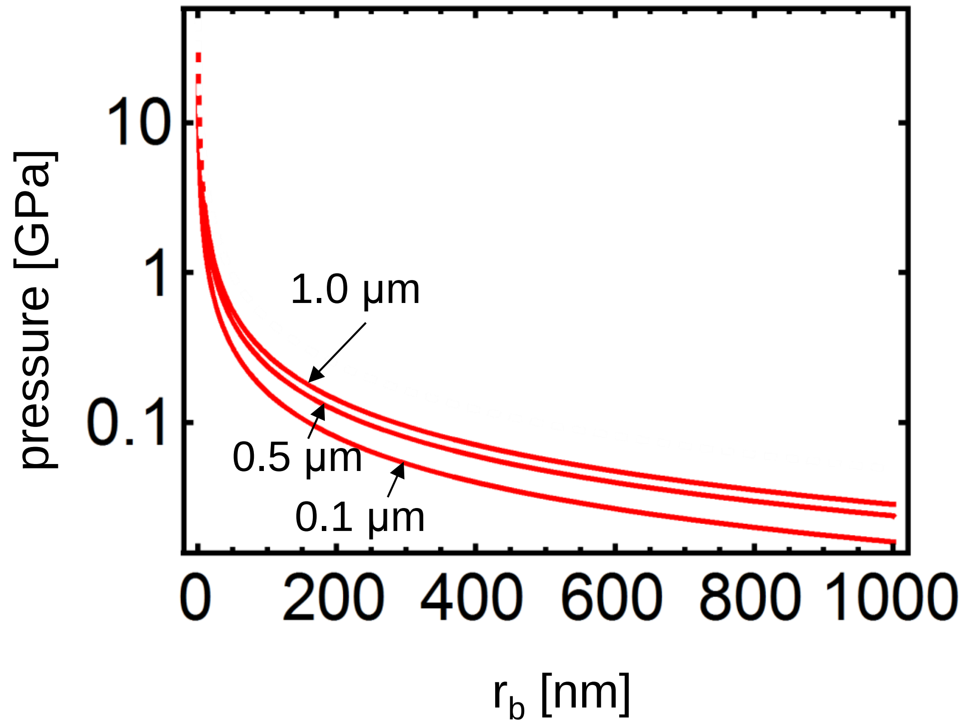
Near the free surface, bubbles may grow by crack extension

Crack extension competitive with loop punching near surface:

$$p_B \geq \frac{1}{r}\left(\frac{4\gamma(Eh)^{1/3}}{5C_1C_2}\right)^{3/4} \sim \frac{1}{r}$$

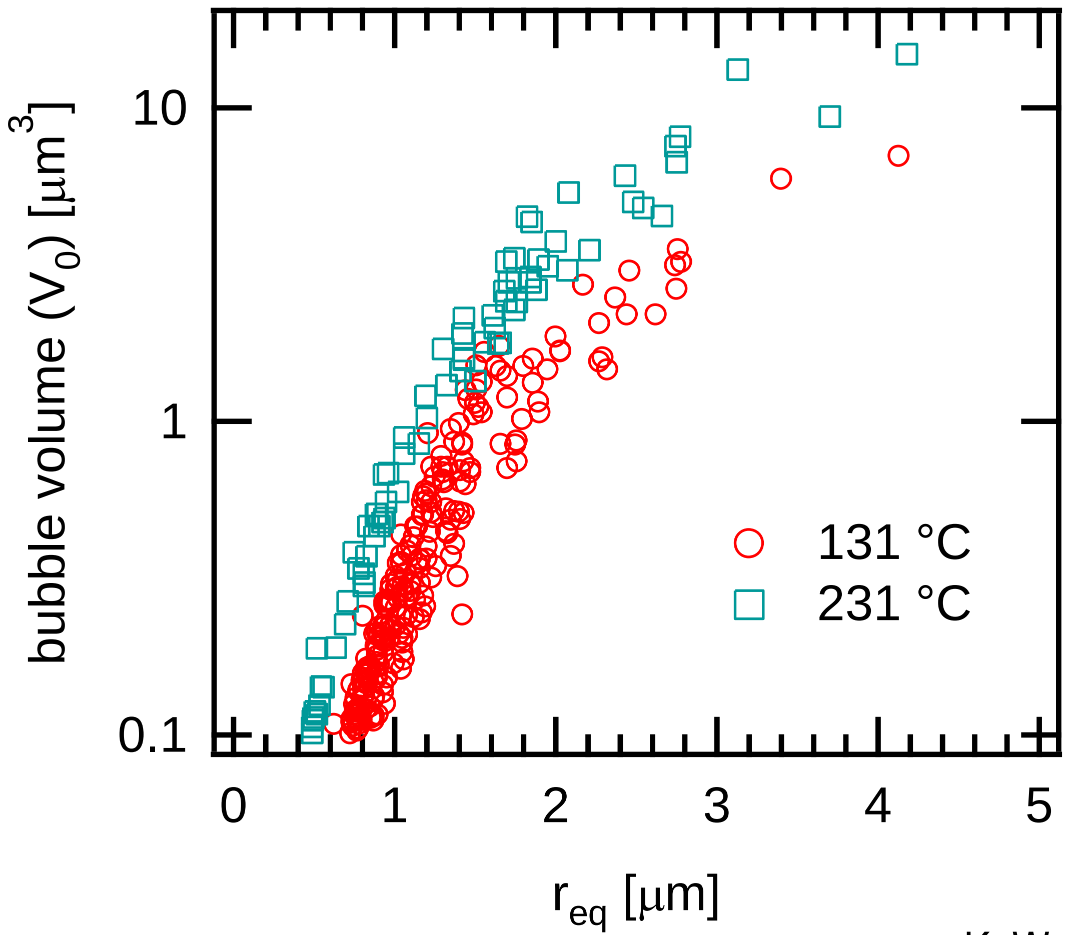
Limitations:

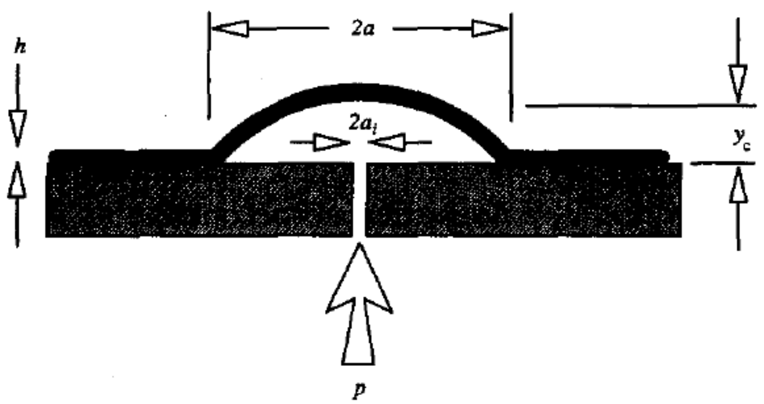
- Correction for thick blisters
- Effect of plasticity (blunting of crack tip)
- Hydrogen effects



Stress calculations based on calculations by K. Wan & Y. Mai, *Acta metall. mater.* **43** (1995) 4109.

Bubble volumes measured with AFM correlate well with blister model



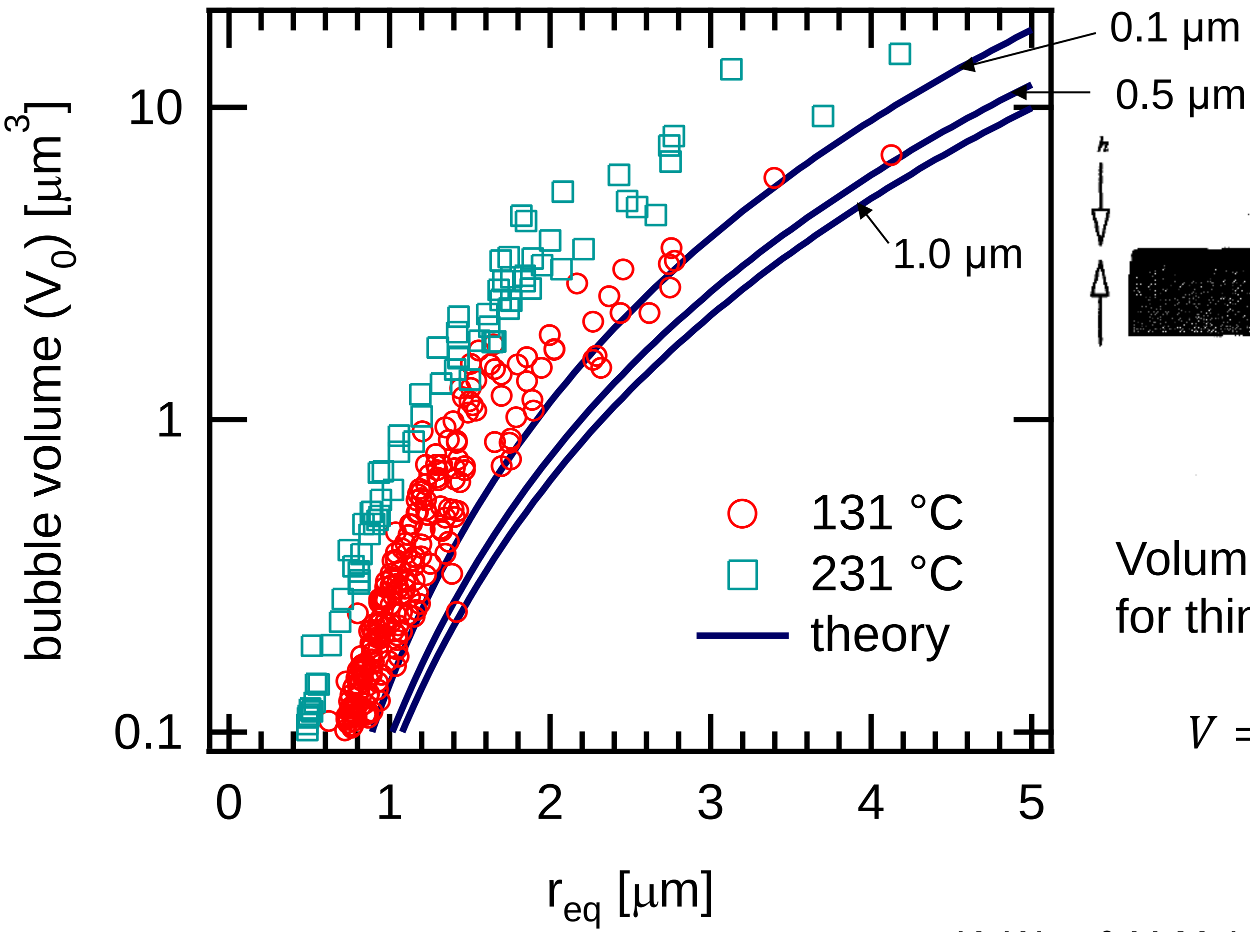


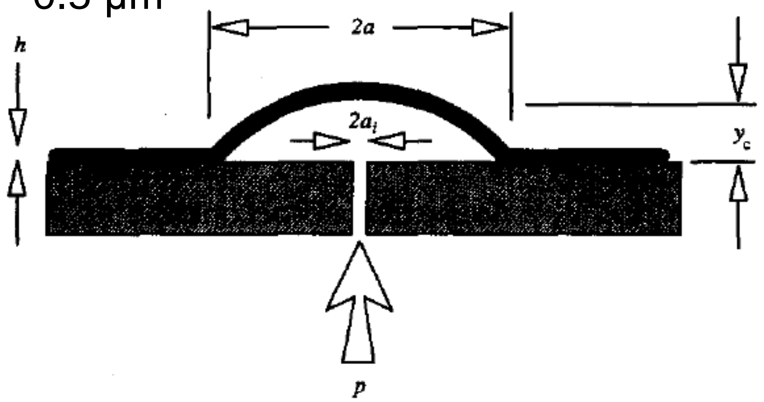
Volume modeled using blister test for thin film adhesion:

$$V = \int y(r) 2\pi r dr = C_1 \pi a^2 y_c$$

K. Wan & Y. Mai, *Acta metall. mater.* **43** (1995) 4109.

Bubble volumes measured with AFM correlate well with deflection model





Volume modeled using blister test for thin film adhesion:

$$V = \int y(r) 2\pi r dr = C_1 \pi a^2 y_c$$

K. Wan & Y. Mai, *Acta metall. mater.* **43** (1995) 4109.

Diffusion and trapping modeled with a continuum-scale approach

Diffusion: 1-D, uniform temperature:

$$\partial u(x,t)/\partial t = D(t)\,\partial^2 u(x,t)/\partial x^2 - q_T(x,t) - q_B(x,t)$$

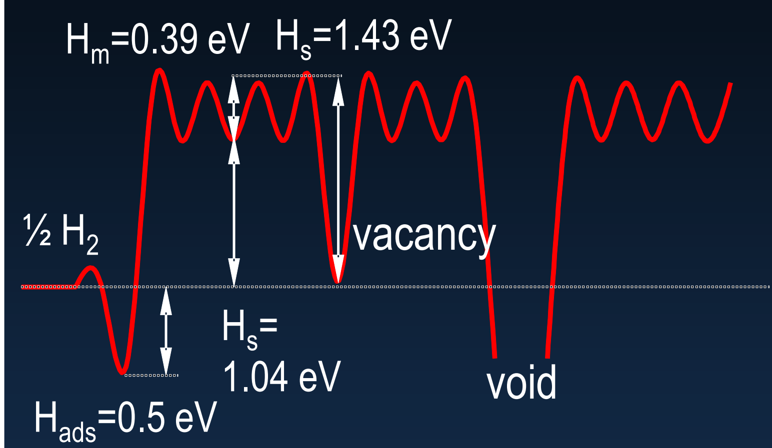
Point defects:

- 1.4 eV saturable traps, no nucleation.
- Used approach of Ogorodnikova [*J. Nucl. Mater.* (2009)] to address trapping and release.

Bubbles:

Modeled using a approach of Mills [J. Appl. Phys. (1959)].

$$q_B(x,t) = \partial u_B(x,t)/\partial t = 4\pi D(t) r_B(x,t) N_B(x)[u(x,t) - u_{eq}(x,t)]$$



Enthalpies for H migrating through W.

Dissolution of H in W is highly endothermic.

H equation of state takes into account non-ideal gas effects

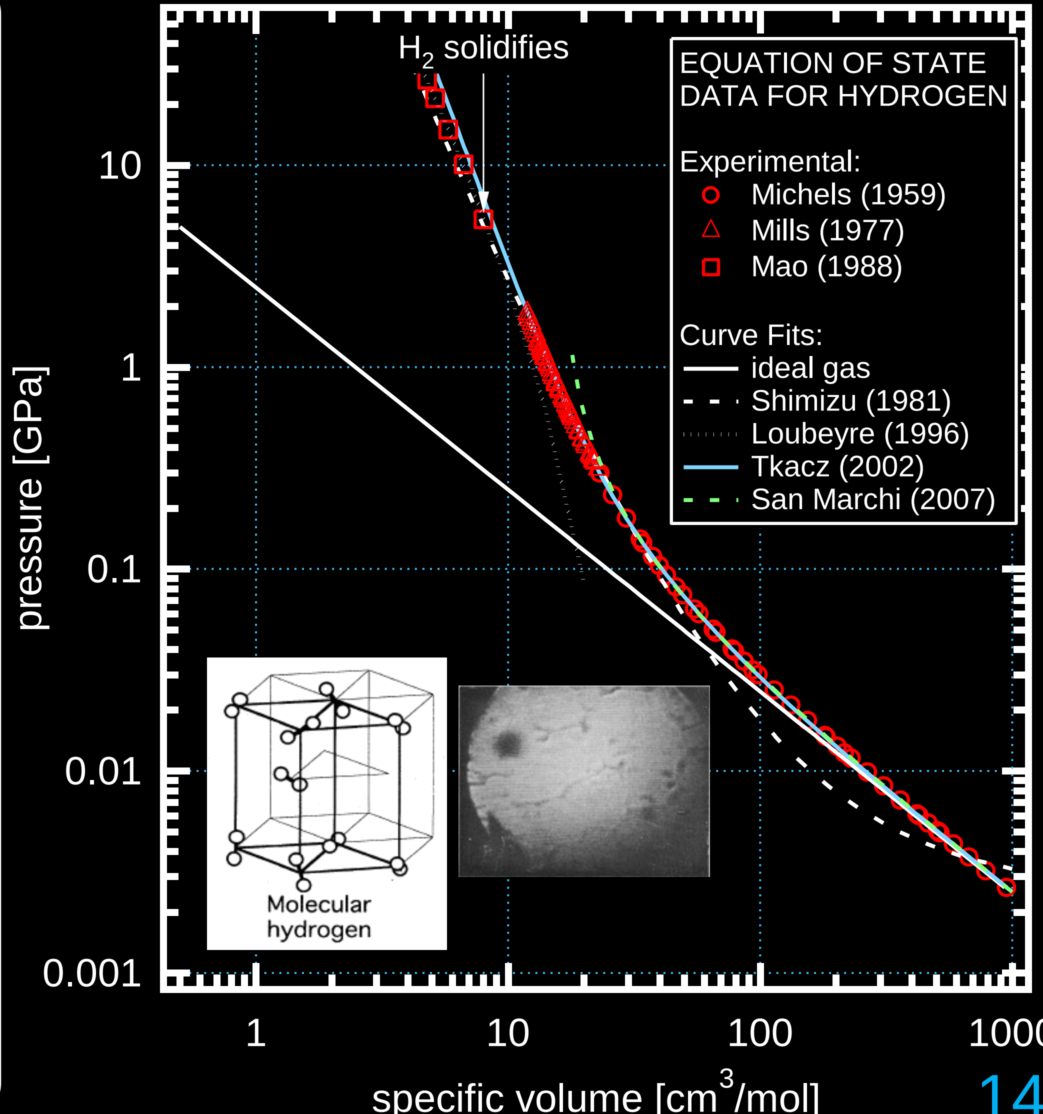
H_2 equation of state (EOS):

- P > 1 GPa expected within small bubbles.
- At 300 K, H_2 solidifies at p=5.7 GPa.
- Tkacz's [J. Alloys & Compounds (2002)] EOS to provide the best fit:

$$v = Ap^{-1/3} + Bp^{-2/3} + Cp^{-4/3} + (D + ET)p^{-1}$$

- San Marchi's simplified EOS better at low pressure:

$$v = \frac{RT}{p} + \mathrm{b}$$



When is bubble growth favorable?

Calculation of equilibrium press.

When is precipitate in equilibrium with mobile conc.?

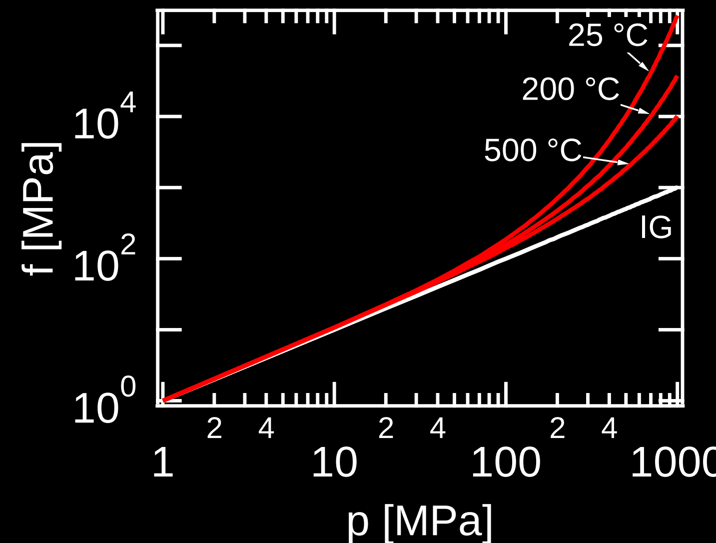
- Equate chemical potentials of gas and solution phase.
- Calculate fugacity to account for non-ideal behavior:

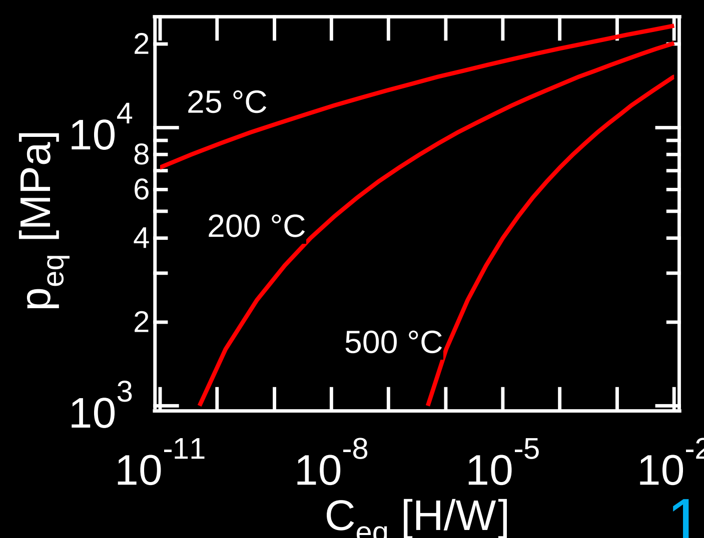
$$\ln(f/p) = \int_0^p \left(\frac{v(p,T)}{RT} - \frac{1}{p}\right) dp$$

- Equilibrium conc. given by:

$$u_{eq} = \sqrt{f} S_0 \exp(-H_S/RT)$$

S_o and H_s from Frauenfelder [JVST, 1969].





Summary of surface morphology findings

- ITER-grade W sample exposed in TPE show similar retention to Toyama/IPP studies.
- Analysis of surface morphology:
  - XPS shows implanted C reduced considerably
  - SEM/EBSD illustrate non-uniform bubble growth over surface
  - Bubble grow on (110) and (111) crystal planes
  - AFM analysis provide bubble volumes
- Modeling of bubbles:
  - Thin film adhesion model adapted to model blister grown on tungsten.
  - Model reproduces bubble sizes observed with AFM

Acknowledgements

We would like to express our appreciation to:

- Our collaborators at INL and Sandia/CA:
  - Brad Merrill, Robert Pawelko, Lee Cadwallader
  - Richard Nygren, Josh Whaley, Jon Watkins, Thomas Felter

Sandia National Laboratories is a multi-program laboratory managed and operated by Sandia Corporation, a wholly owned subsidiary of Lockheed Martin Corporation, for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-AC04-94AL85000.

This work was prepared for the U.S. Department of Energy, Office of Fusion Energy Sciences, under the DOE Idaho Field Office contract number DE-AC07-05ID14517.