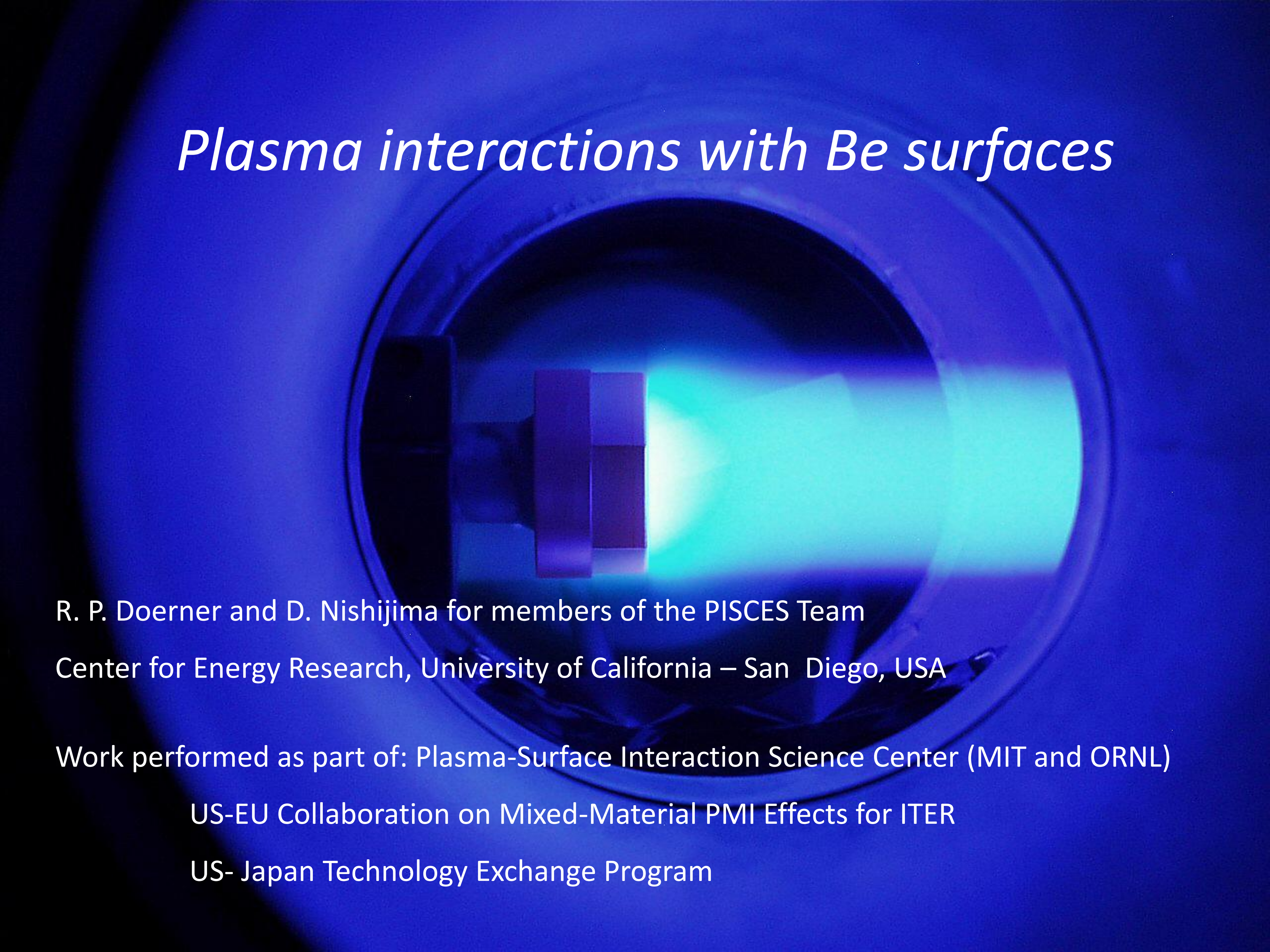


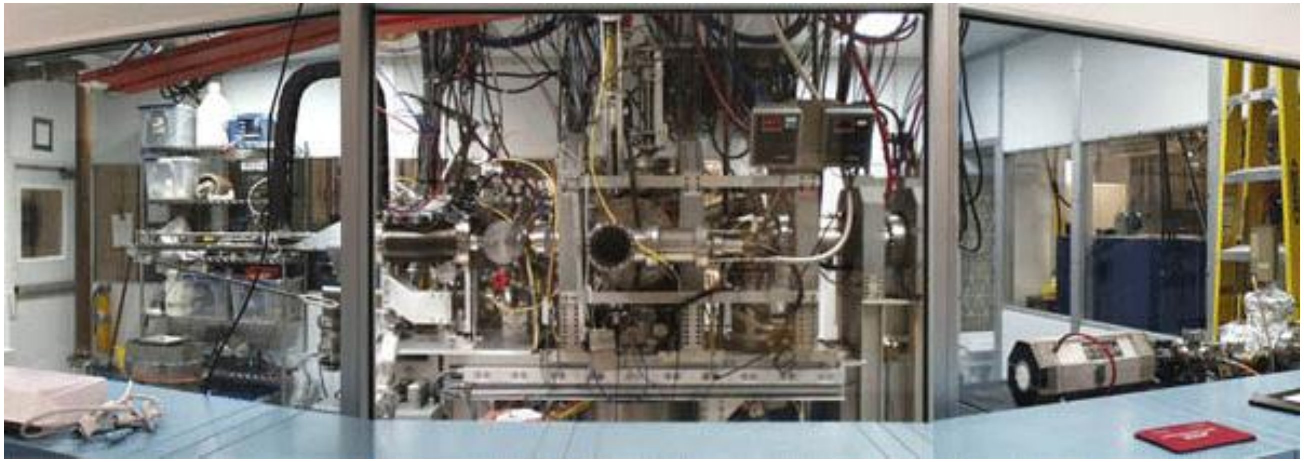
Plasma interactions with Be surfaces

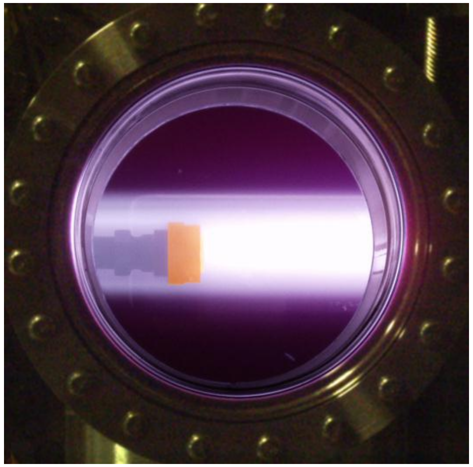
R. P. Doerner and D. Nishijima for members of the PISCES Team

Center for Energy Research, University of California – San Diego, USA

Work performed as part of: Plasma-Surface Interaction Science Center (MIT and ORNL)
US-EU Collaboration on Mixed-Material PMI Effects for ITER
US- Japan Technology Exchange Program

The PISCES-B divertor plasma simulator is used to investigate ITER mixed materials PSI.



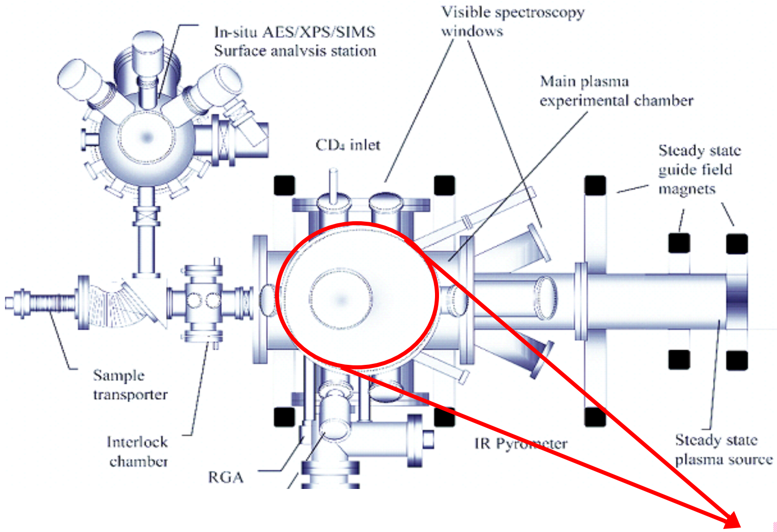


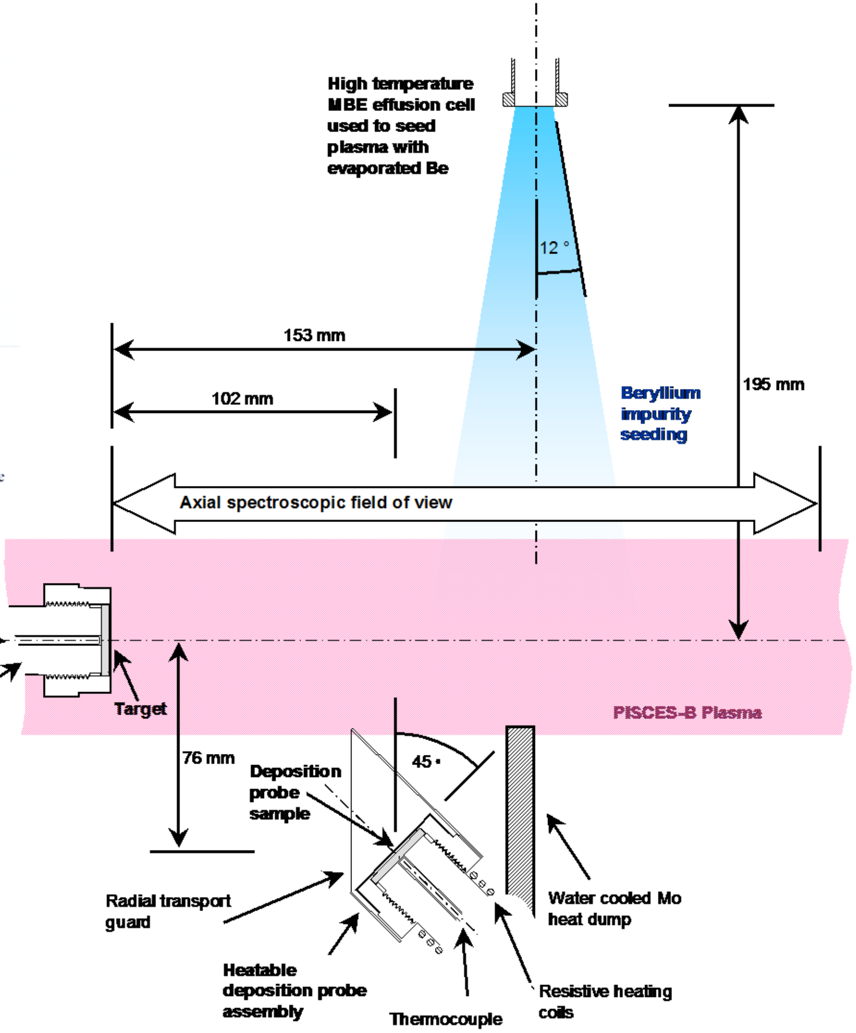
- PISCES-B is contained within an isolated safety enclosure to prevent the release of Be dust.

	PISCES	ITER (edge)
Ion flux (cm^{2}s^{-1})	**10^{17}–10^{19}**	**~10^{19} - 10^{20}**
Ion energy (eV)	**20–300 (bias)**	**10–300 (thermal)**
T_e (eV)	4–40	1–100
n_e (cm^{-3})	**10^{12}–10^{13}**	**~10^{13}**
Be Imp. fraction (%)	**Up to a few %**	**1–10 (ITER)**
Pulse length (s)	**Steady state**	**1000**
PSI materials	C, W, Be	C, W, Be ..
Plasma species	H, D, He	H, D, T, He

PISCES-B has been modified to allow exposure of samples to Be seeded plasma

PISCES





P-B experiments simulate Be erosion from ITER wall, subsequent sol transport and interaction with W baffles or C dump plates, as well as investigation of codeposited materials using witness plates

Outline of Technical Presentation

- Retention and release
  - Retention in plasma exposed Be
  - Retention in Be-rich codeposits
  - Release due to flash heating and long-term bakes
- Erosion in the plasma environment
  - Be erosion from D, He and Ar plasma
  - Chemical sputtering of BeD
  - Redeposition/sticking efficiency
- Be-containing mixed materials (W, C, N, O) have not been included in this presentation
- Spectroscopic issues for Be
- Discussion Points
- [A couple slides on Be:H LEIS from Sandia – R. Kolasinski]

Retention in implanted (ion beam and plasma) beryllium saturates

- Retention exhibits an energy (or ion range) dependence
- Once normalized to an energy of 100 eV, the spread in the database is greatly reduced
- During low fluence ion beam measurements retention increases linearly up to ~ 1e21 D/m^2 then saturates
- During high flux plasma measurements, retention quickly saturates at ~ 1e21 D/m^2 up to fluences exceeding 1e26 m^{-2}

Data normalized to 100 eV

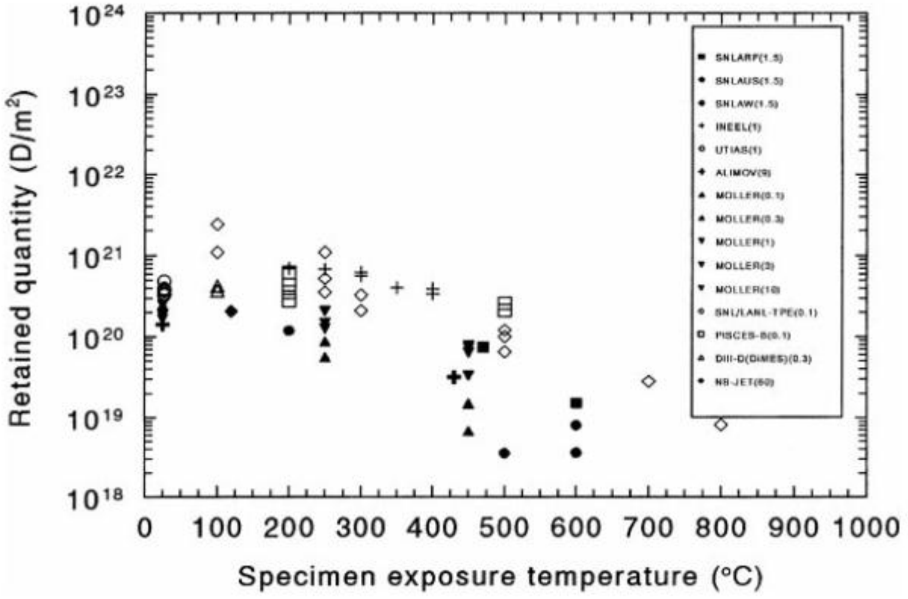


Fig. 5. Comparison of *adjusted* deuterium retention data as a

From: R.A. Anderl et al., JNM 273(1999)1.

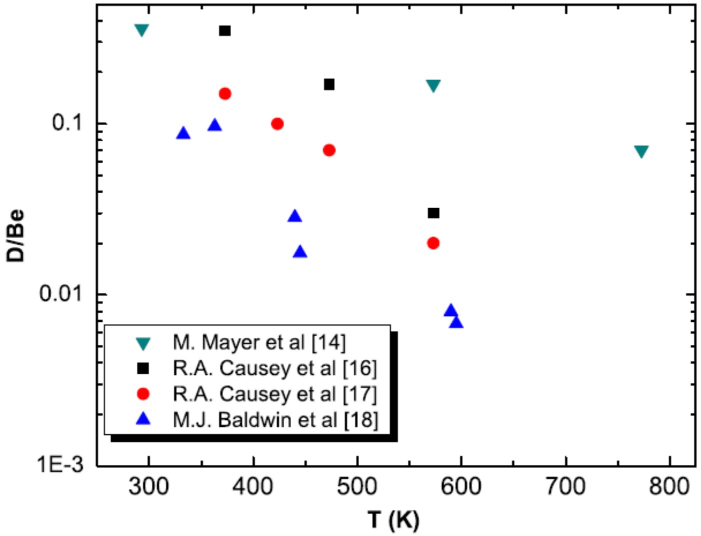
Retention in Be codeposits does not saturate, level depends on deposition conditions

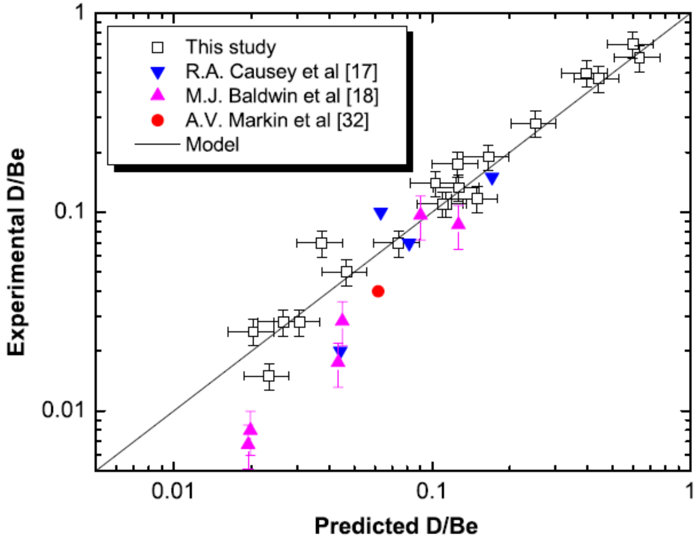
- Since thickness grows with time, retention does not saturate
- D/Be level becomes the figure of merit and depends on deposition conditions (T (surface), E (D atom), deposition rate)

$$\frac{D}{Be} = 2.94 \times 10^{-5} \times r_d^{-0.59\pm0.1} \times E_n^{1.34\pm0.15} \times e^{\frac{1306\pm190}{T}}$$

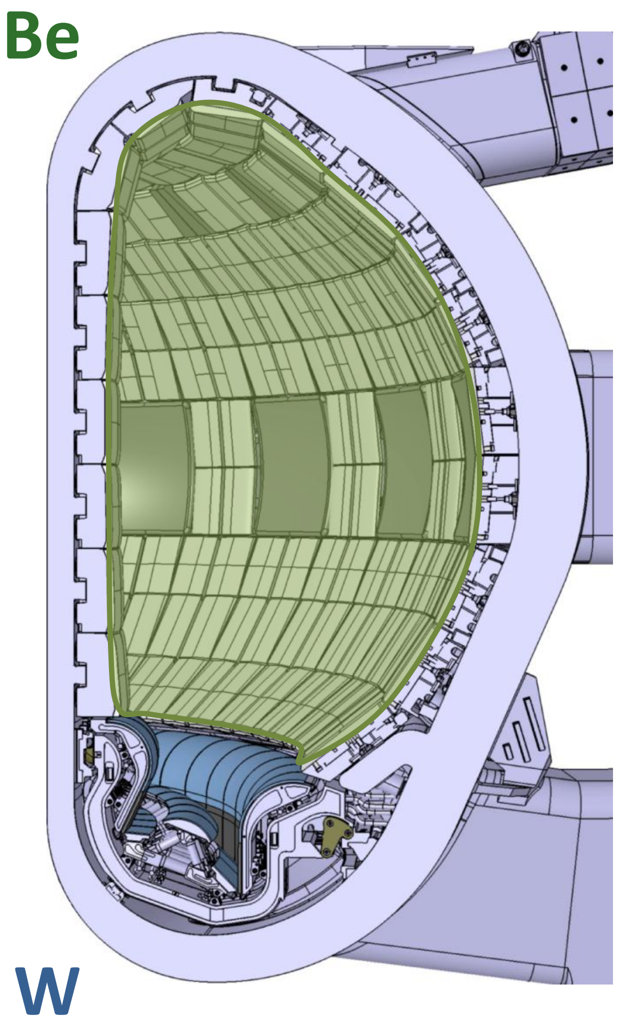
[From: G. De Temmerman et al., NF 48(2008)075008]

- Scaling laws are only valid over certain ranges in parameter space
- D/Be does not exhibit a dependence on O content, but is strongly influenced by C content in the codeposit





T accumulation in ITER will be dominated by Be codeposits

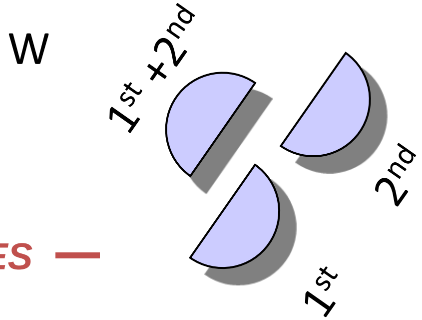


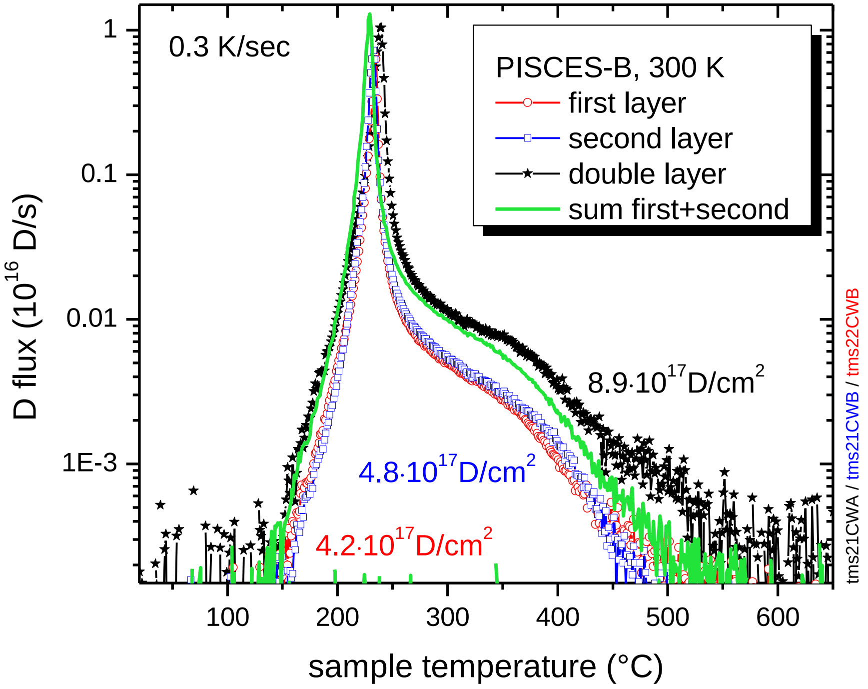
- ITER will have 700 sq. m of Be first wall and start-up limiters.
- Nuclear licensing requires low T in vessel inventory, 700 g (mobilizable).
- In the absence of C, T accumulation in ITER will be driven by co-deposition of T with eroded Be.

- Inventory control options:
  1) Transient thermal loads, rapid (< 10 ms) surface heating to high *T* during controlled plasma termination. **HOW MUCH IS RELEASED?**
  2) Bulk PFC bake-out, 513 K (main wall), 623 K (divertor). **HOW LONG TO BAKE?**
  3) Remote probes (inefficient, last resort).
  4) Component replacement (when all else fails).

Internal BeO layers do not influence D release

PISCES

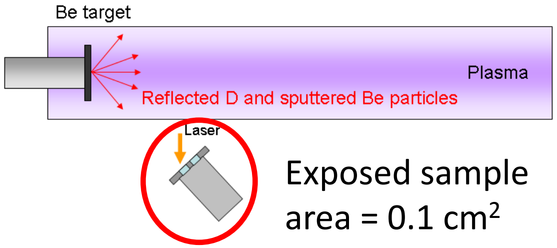




- Two Be codeposits were collected while venting to replacing one half the sample between codepostion runs
- Several nm thick BeO will exist between subsequent codeposits
- Release behavior of the multilayer codeposit is almost identical to the sum of the individual codeposits
- Conclusion is that internal BeO layers will not impact the knowledge gained from studying pure Be codeposits

Be/D codeposits can be made in several locations

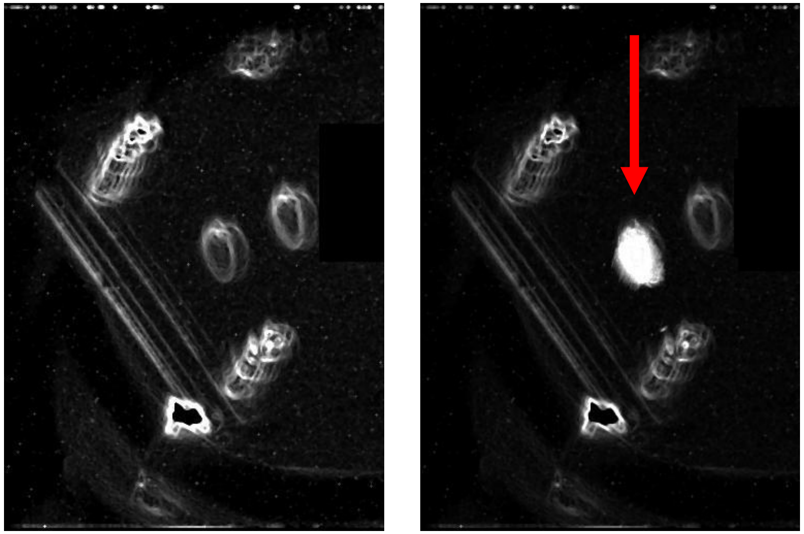
Collection plate is located outside plasma interaction region



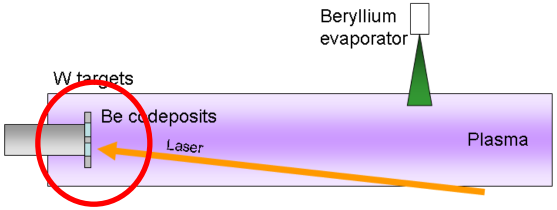
Exposed sample area = 0.1 cm^2

No laser

Laser

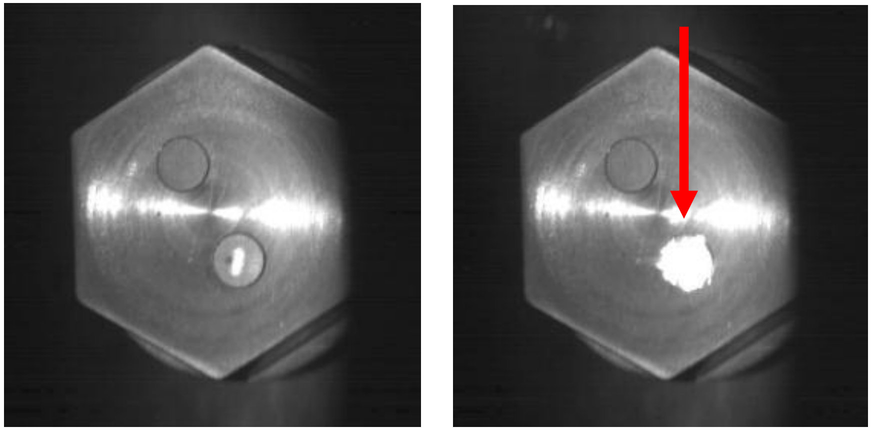


Be-seeding of the plasma can grow codeposits on a floating target



Exposed sample area = 0.28 cm^2

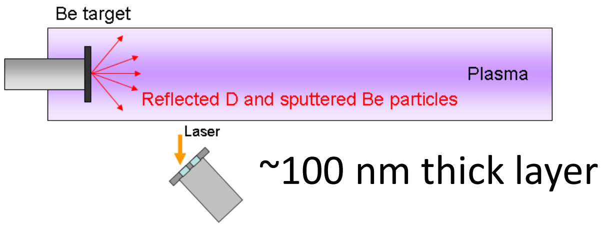
Laser



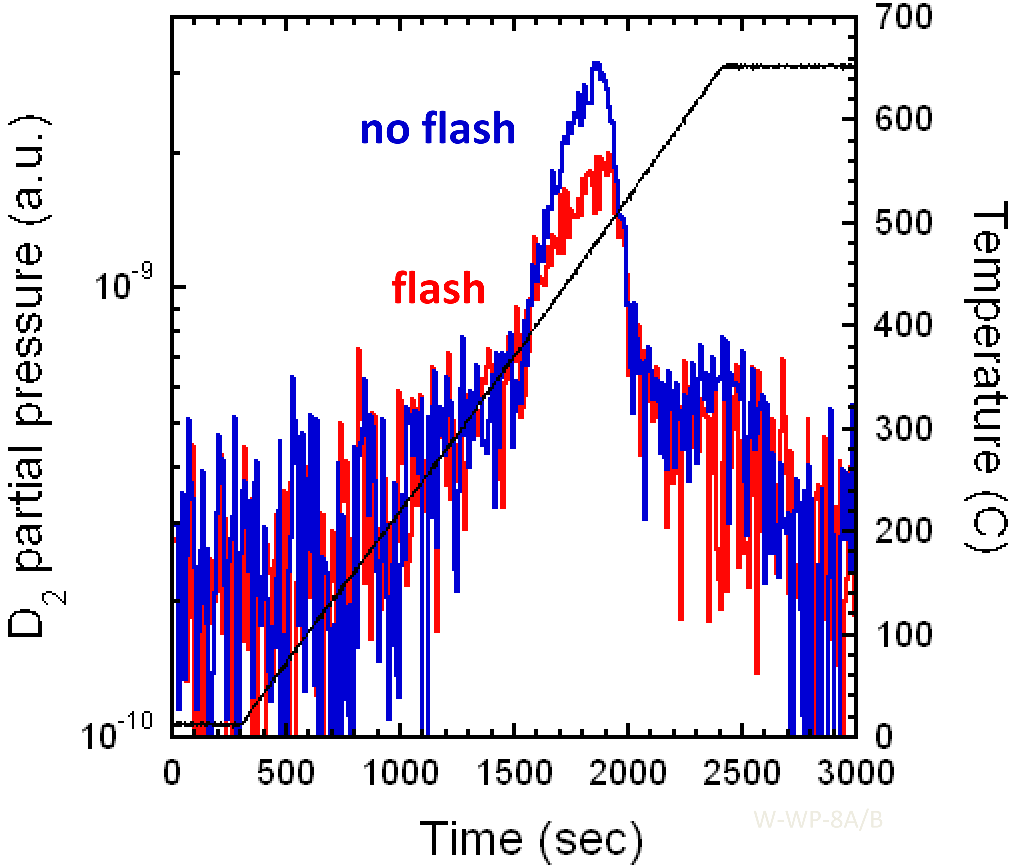
Laser flash heating procedure

- Two side-by-side samples loaded with D (co-deposition or implantation)
- Plasma shut off, then one sample hit with welding laser (1064 nm, 50 J, 10 ms duration, 1 to 4 flashes typically, and up 50 flashes to simulate repetitive events)
- Fast pyrometer used to measure temperature
- D retention measured in both samples separately using thermal desorption spectroscopy (TDS) → determine amount of D removed during flash

Flash heating of collector plate codeposits show little release of D



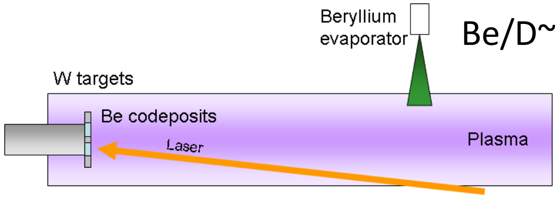
- T_{flash} = 900ºC
- Flash desorbs ~20% of retained D


TDS comparison

Flashing thicker target-generated codeposits shows even less fractional release of D

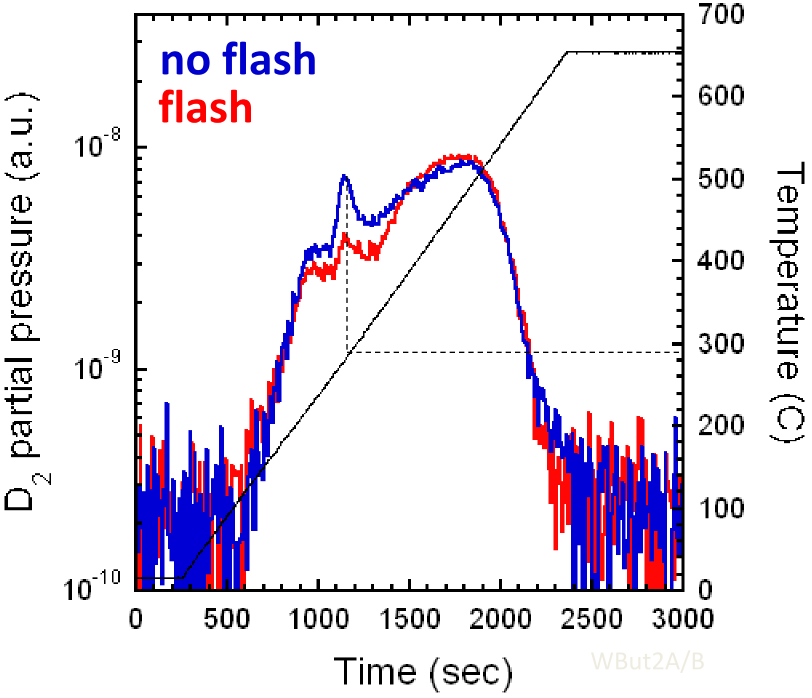
PISCES

- T_{flash} ~ 450°C
- Clear reduction in (BeD_2?) peak at 290°C

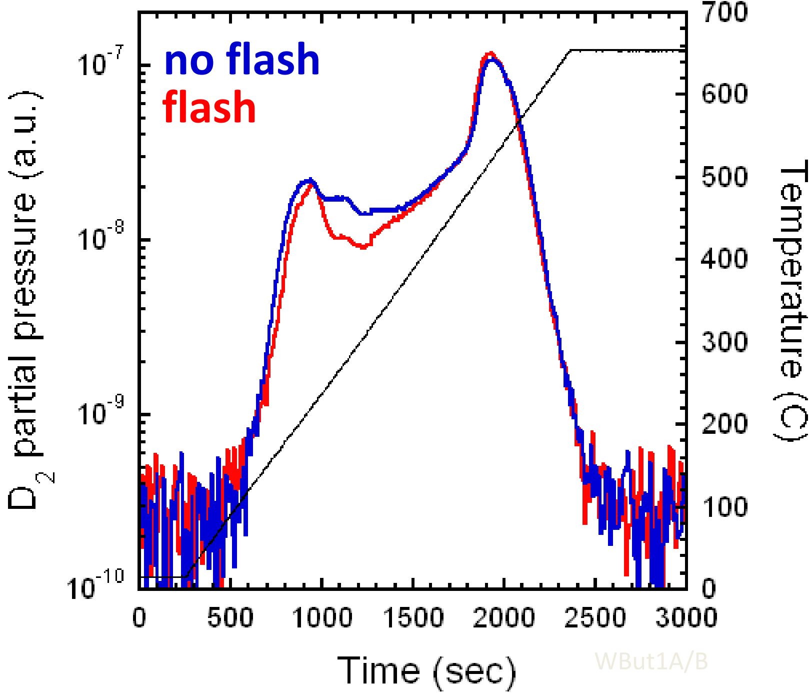


Be/D~0.5%

150 nm thick codeposit

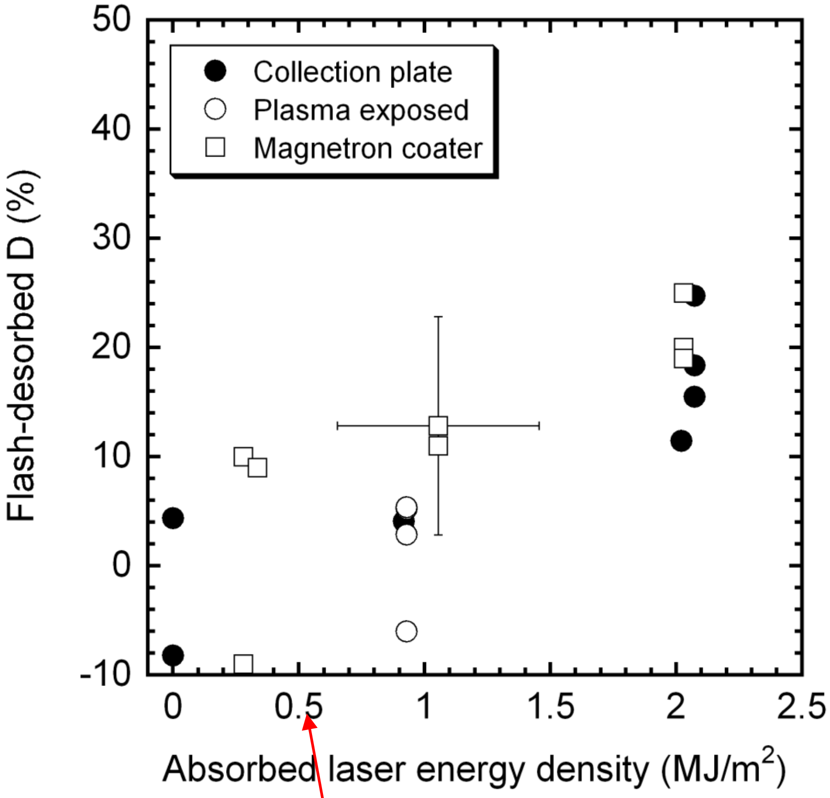


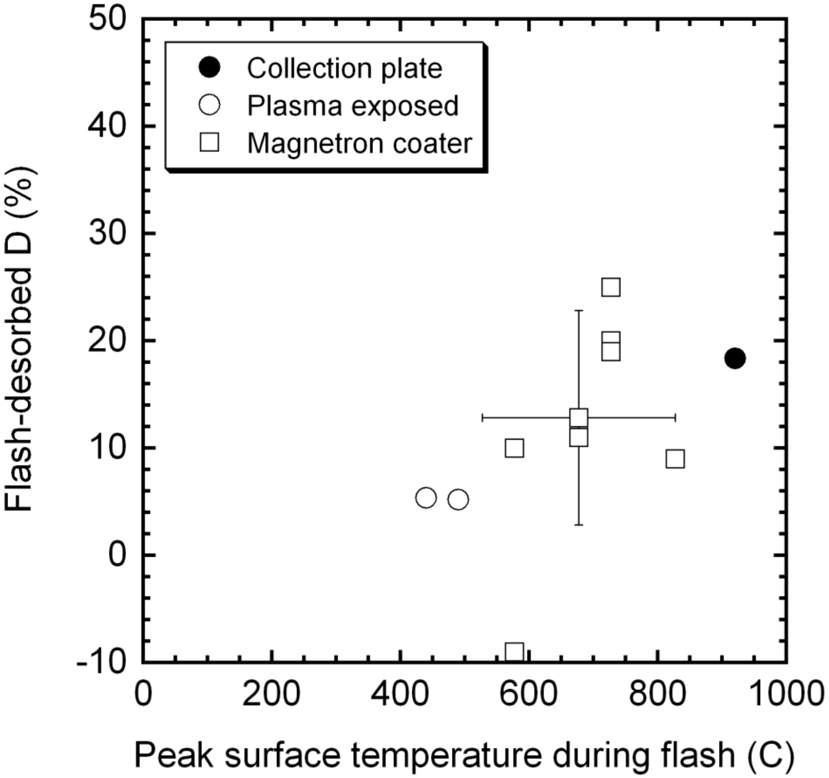
650 nm thick codeposit



Small D release (~10%) is observed at ITER-relevant energy densities

PISCES





ITER disruption flash energy density with uniform radiation distribution, assuming 350 MJ of thermal energy spread over 700 m²

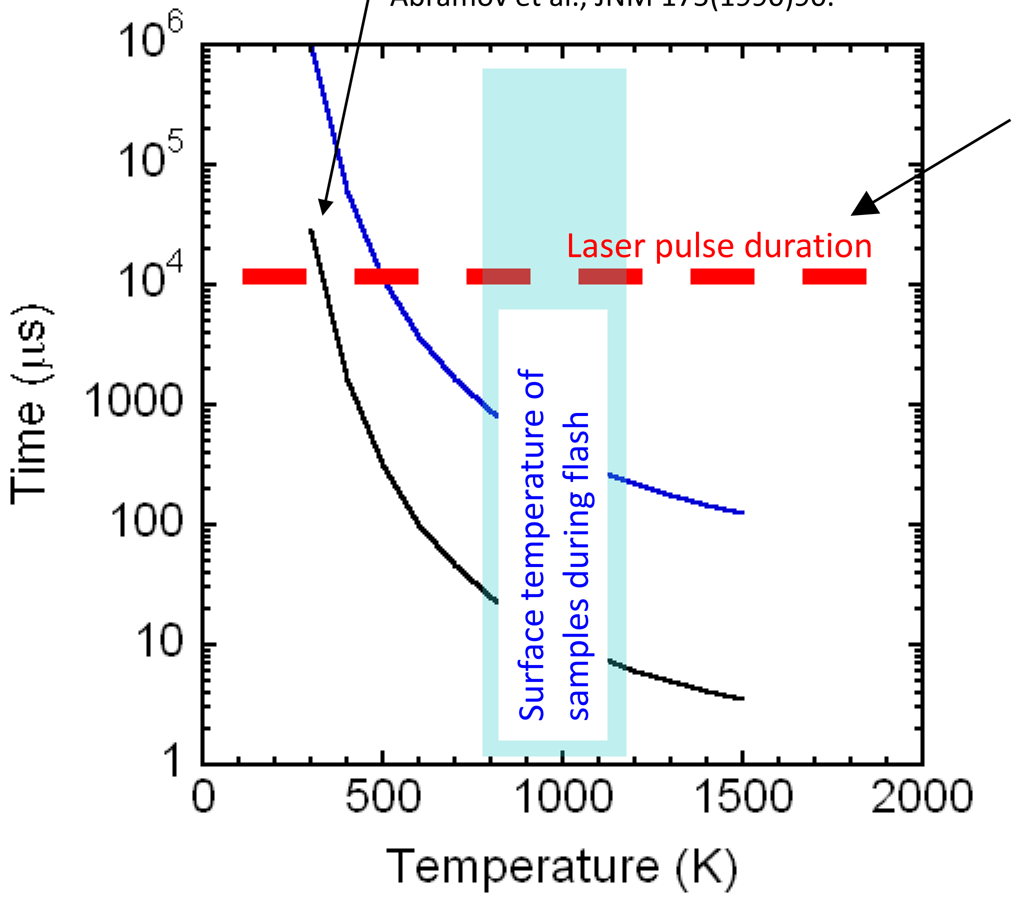
Be melting temp = 1287 C

Desorbed D fraction = $1 - \phi_{flash}/\phi_{control}$

Something is wrong with our view, diffusion is not the rate limiting process

Time for D diffusion through 100 and 600 nm thick Be lattice

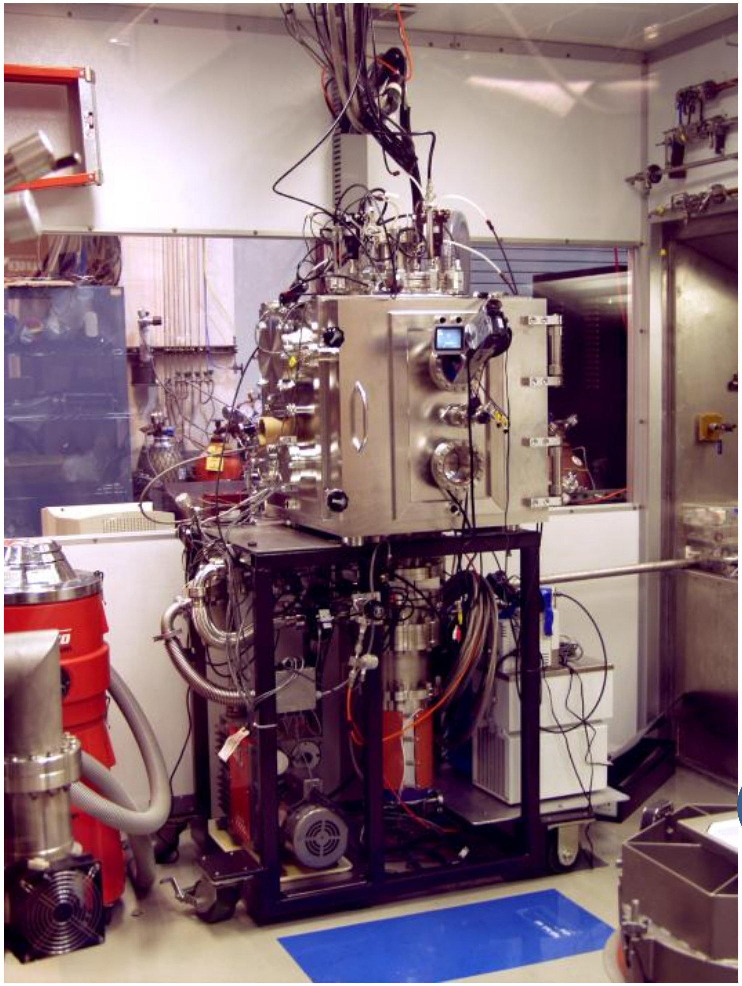
Abramov et al., JNM 175(1990)90.



Laser pulse is much larger than diffusion time, so if diffusion were the only process there would be enough time for D to random walk to the edge of the layer.

Reproducibility of Be codeposits created in PISCES-B makes detailed TMAP analysis problematic

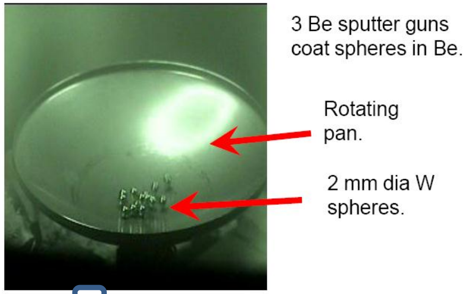
GA magnetron sputter coater produces batches of ‘identical’ co-deposits



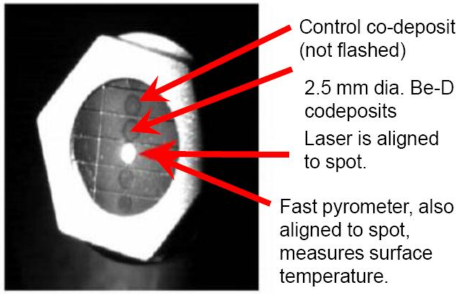
Utilizes 3, 100 W Be sputter guns, operated at 6 mTorr in 80% Ar, 20 % D_2

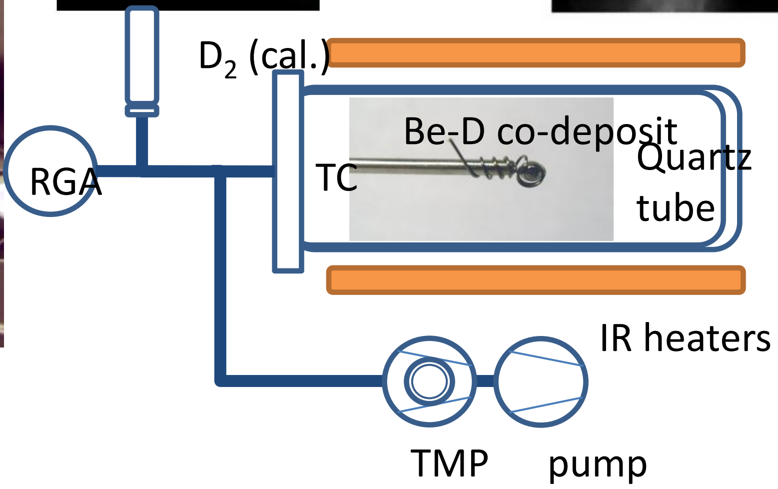
Be deposition rate 2.5×10^{15} $cm^{-2}s^{-1}$
Be-D co-deposited layers 1 μm thick

Bake-out Exp.
$T_{dep} < 323$ K



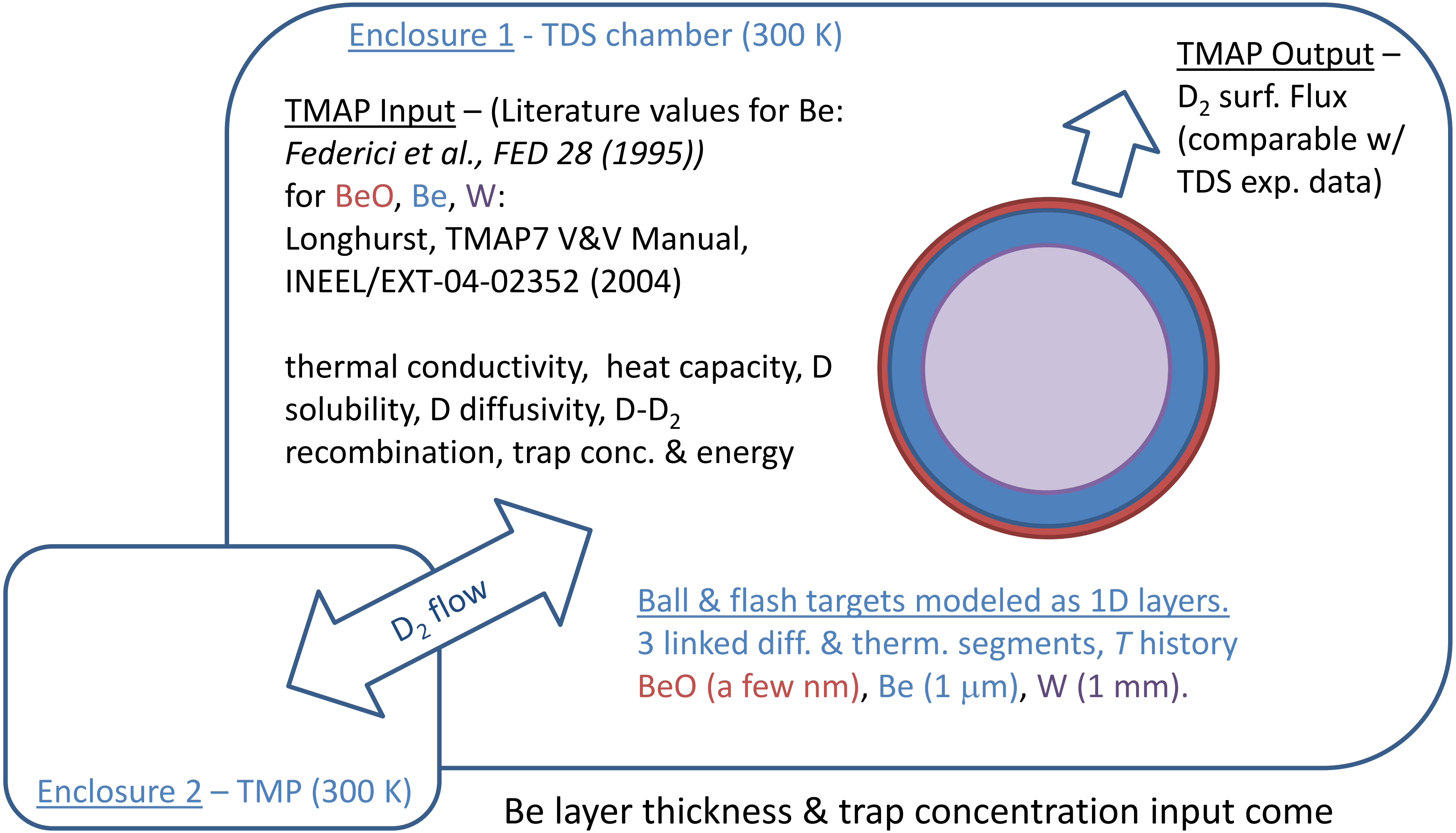
Transient Exp.
$T_{dep} \sim 500$ K





Special holder designed to give good TC contact to balls

Modeling (TMAP 7)



Enclosure 1 - TDS chamber (300 K)

TMAP Input – (Literature values for Be:
Federici et al., FED 28 (1995))
for BeO, Be, W:
Longhurst, TMAP7 V&V Manual,
INEEL/EXT-04-02352 (2004)

thermal conductivity, heat capacity, D solubility, D diffusivity, D-D_2 recombination, trap conc. & energy

TMAP Output –
D_2 surf. Flux
(comparable w/
TDS exp. data)

Ball & flash targets modeled as 1D layers.
3 linked diff. & therm. segments, *T* history
BeO (a few nm), Be (1 μm), W (1 mm).

Enclosure 2 – TMP (300 K)

Be layer thickness & trap concentration input come from experimental measurements.

Base modeling co-deposit D_2 release.

- TDS data (symbols) from a Be–D co-deposit sphere. Two trap states (α and β) with a 'tail' (γ) reminiscent of 2nd order release.

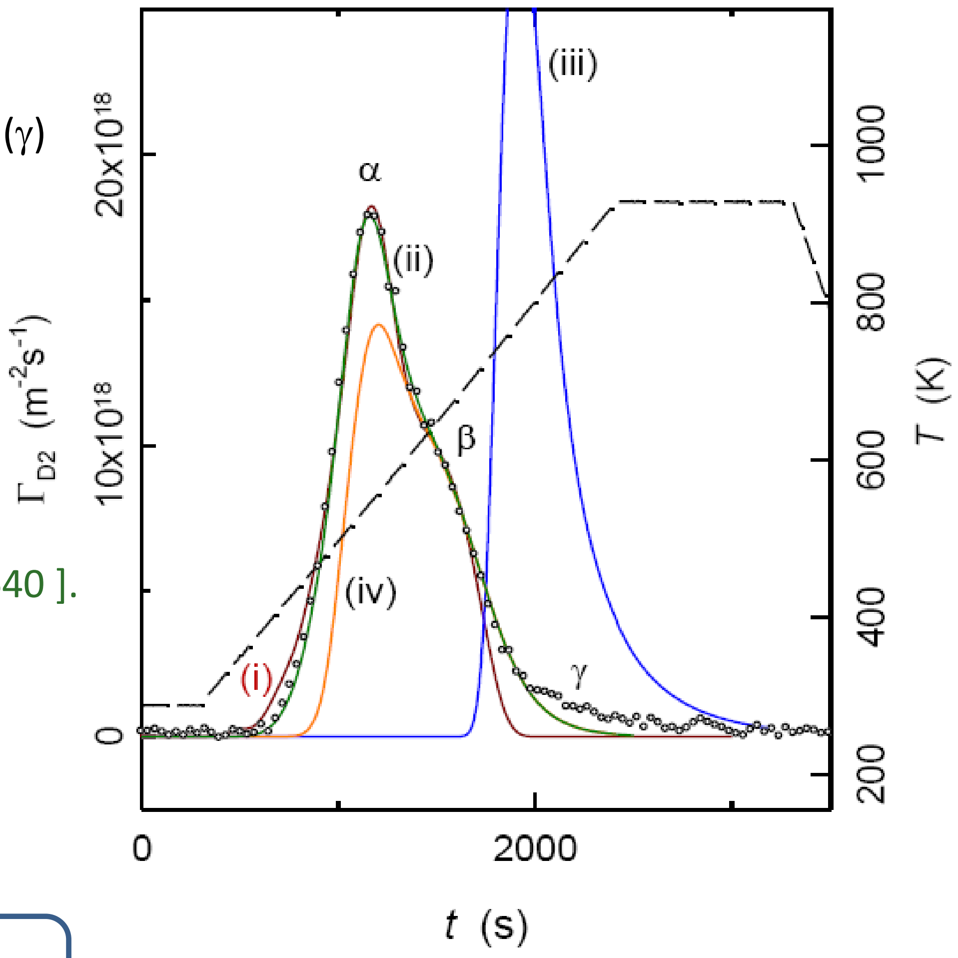
- TMAP output (i, ii, iv) - single layer Be(1 μm) model. E_{traps}: 0.80 eV & 0.98 eV.

- (i) Sieverts law release.
- (ii) k_r specified [Federici et al., FED 28 (1995)
- 136 & Longhurst et al. JNM 258–263 (1998) 640].
- (iii) Incl. 10 nm BeO surface layer.

  ~ *Result does not agree with experiment.*
- (iv) simulation (ii), following 10 ms thermal transient to 1123 K.

  ~ *Minimal desorption from low T trap only.*

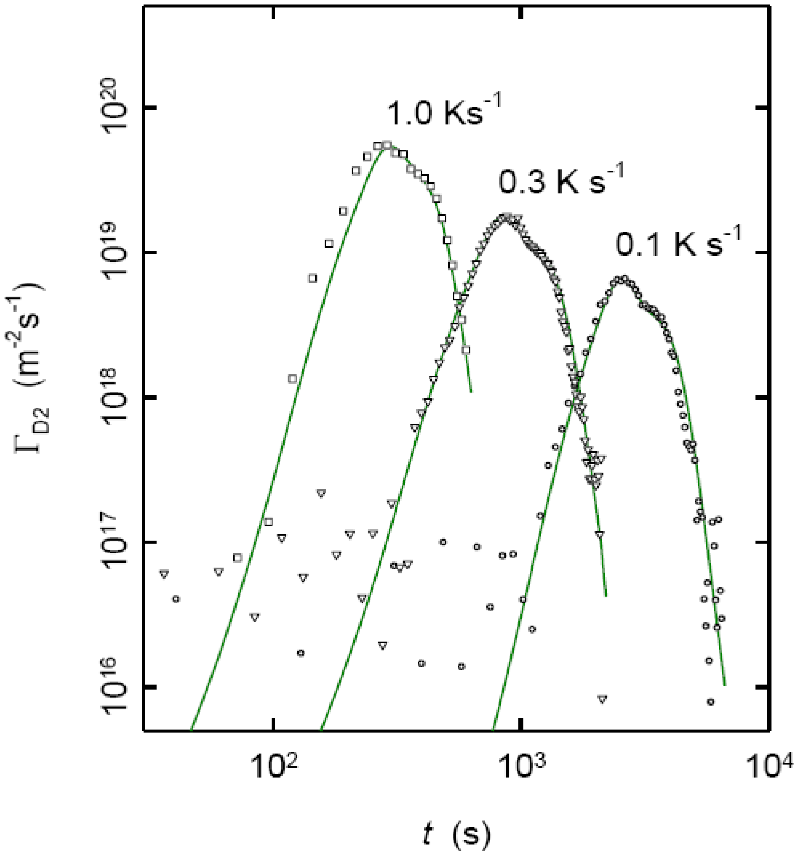
- TMAP single layer model (ii) gives best result.



Measured D/Be agrees with integrated TMAP model release

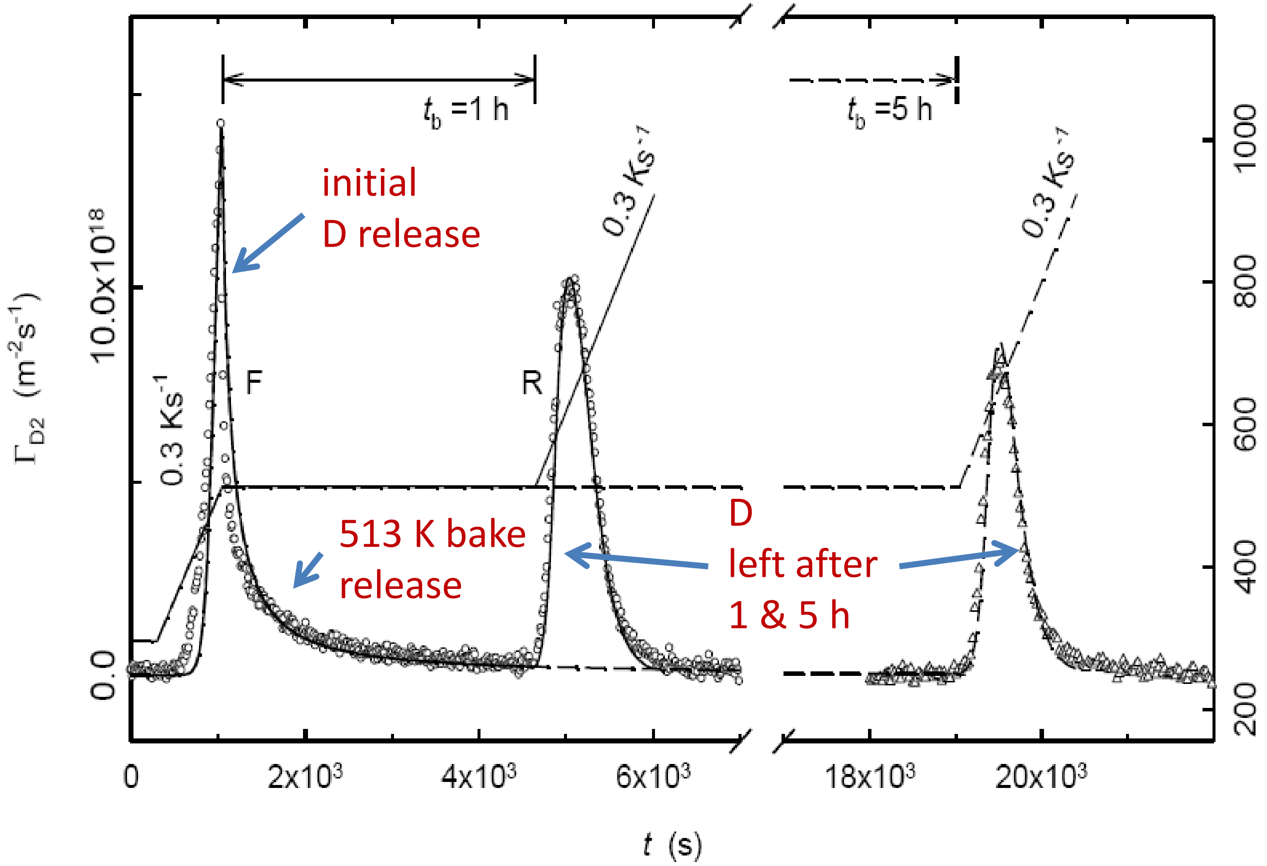
Base modeling co-deposit D_2 release (Temperature ramp rate variation)

- Good agreement between TMAP single Be layer model (ii) and TDS data acquired with different heating rates in the range 0.1 – 1.0 K.
- Identical codeposits are essential for this comparison of model and experiment



TMAP modeling of long-term bake-out.

TMAP models bake-out, but k_r *must be adjusted as in* Longhurst *et al*. JNM 258–263 (1998) 640, *by the factor, [1+exp(c_D/A)], where c_D is the D conc. in the near surface, and A is a constant. Sharp fall (F) and rise (R) are better modeled as a result.*



initial D release

513 K bake release

D left after 1 & 5 h

- TDS data (symbols) for 1 & 5 h fixed temp bake at 513 K.
- Lines – TMAP
- t_b varied up to 25 h
- Data collected for 513 K and 623 K bakes.

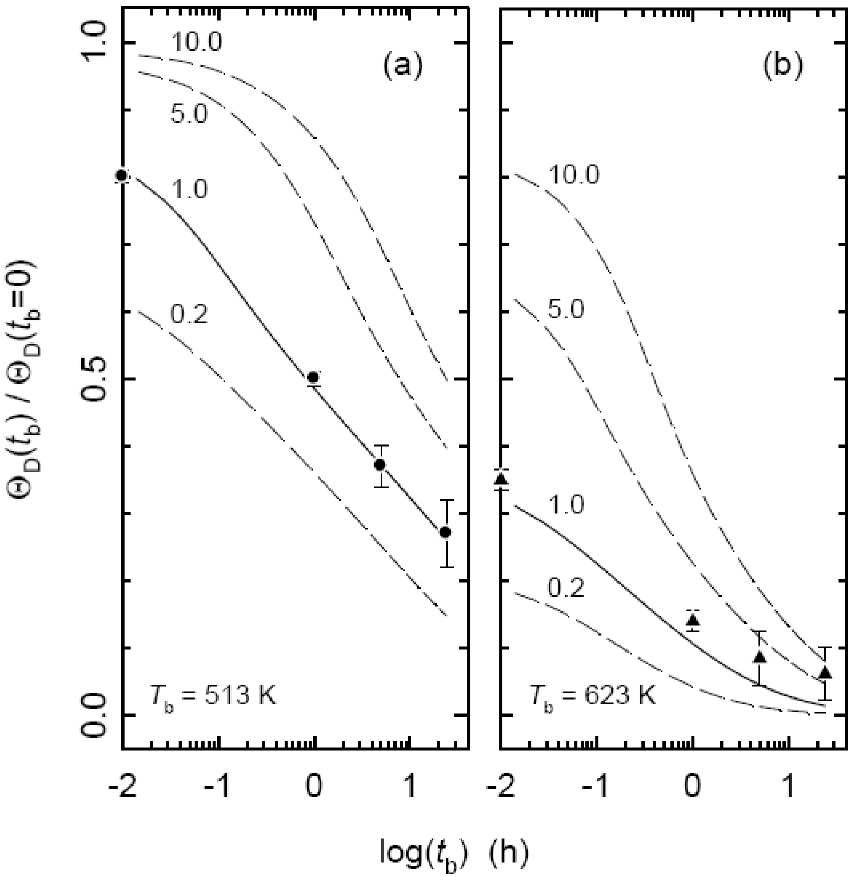
D inventory remaining after bake: Exp & TMAP.

- Experiment (symbols) & TMAP (solid line) shows remaining D in 1 μm thick co-deposit falling significantly in ~1 day at ITER bake-out temperatures of 513 K & 623 K.

- TMAP output (dashed lines) are other layer thicknesses, 0.2, 5 and 10 μm.

- Thick layers require longer bake-out in TMAP simulations as a consequence of high trap concentration (analogous to reduction in diffusivity).

- Bakes longer than ~1 day are increasingly ineffective.



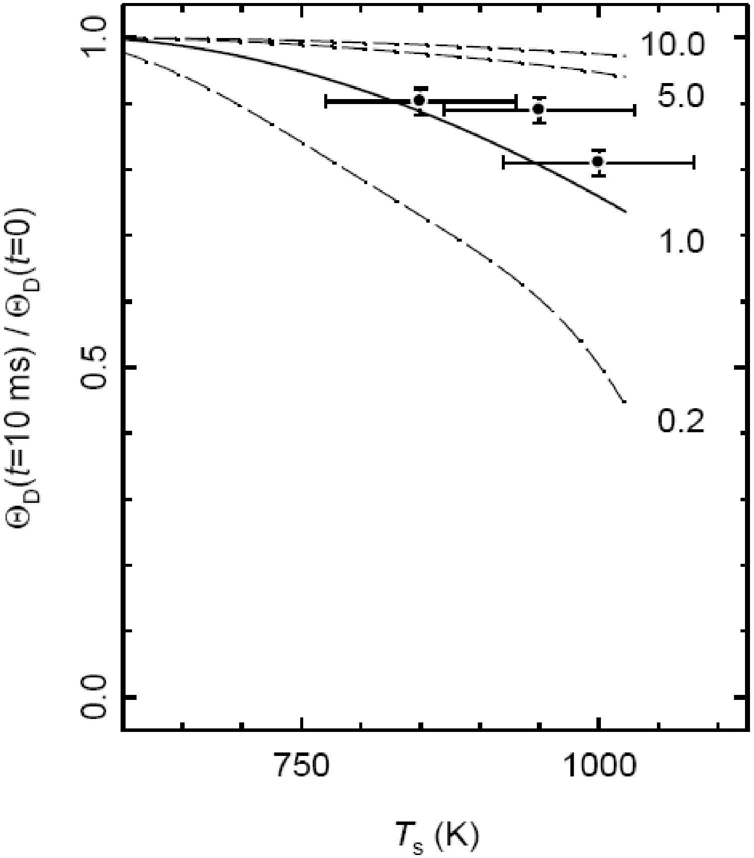
D inventory left after thermal transient to 1000 K for 10 ms: Exp & TMAP.

- Remaining D inventory in co-deposits (normalized) remains high following a 10 ms laser pulse for layer temperature up to 1000 K.

- TMAP simulation agrees reasonably well with (full line) experiment.

- Dashed lines show TMAP output for other co-deposit thicknesses of 0.2, 5 and 10 μm.

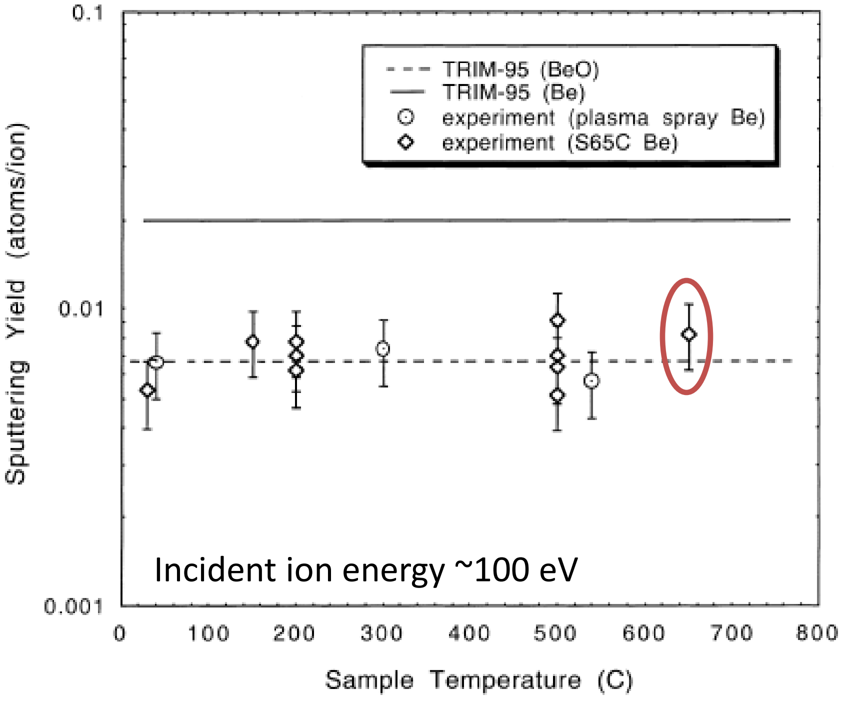
- Again, thicker co-deposits desorb less (as seen in PISCES-B codeposit data from the target location)



Significant variations in the Be sputtering yield are measured

discrepancy between - PISCES-B - Eckstein's TRIM - ion beam - JET - sputter yields
(< 0.7%) (< 3.5%) (< 8%) (< 45%*)

R.P. Doerner et al. / Journal of Nuclear Materials 257 (1998) 51–58



Incident ion energy ~100 eV

J. Roth et al., FED 37(1997)465.

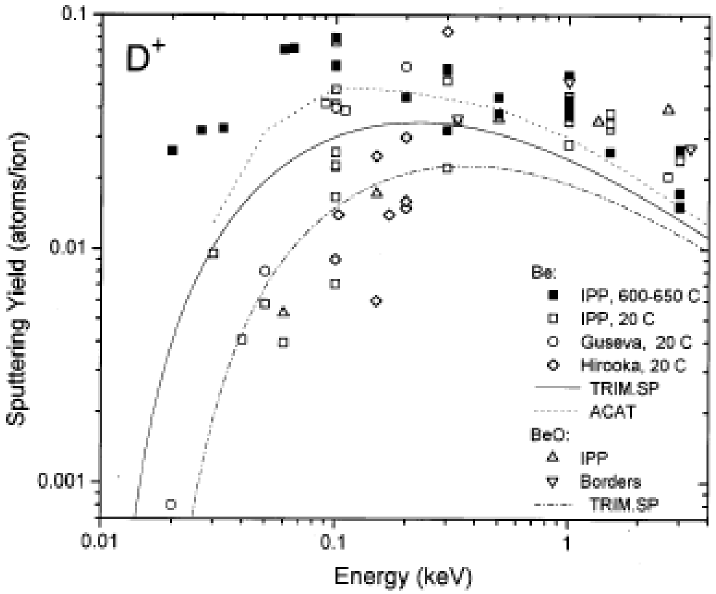
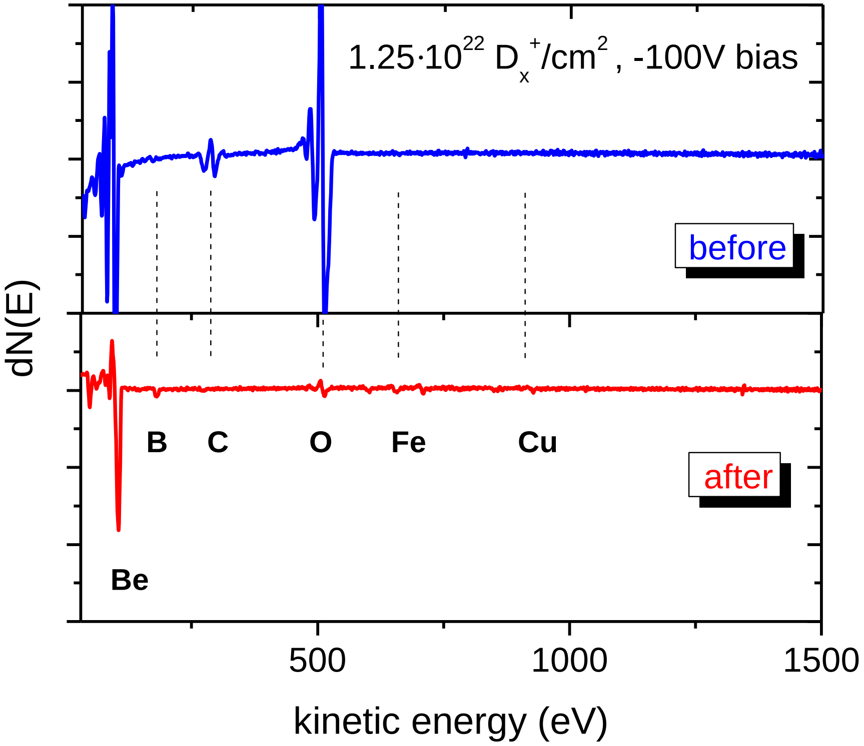
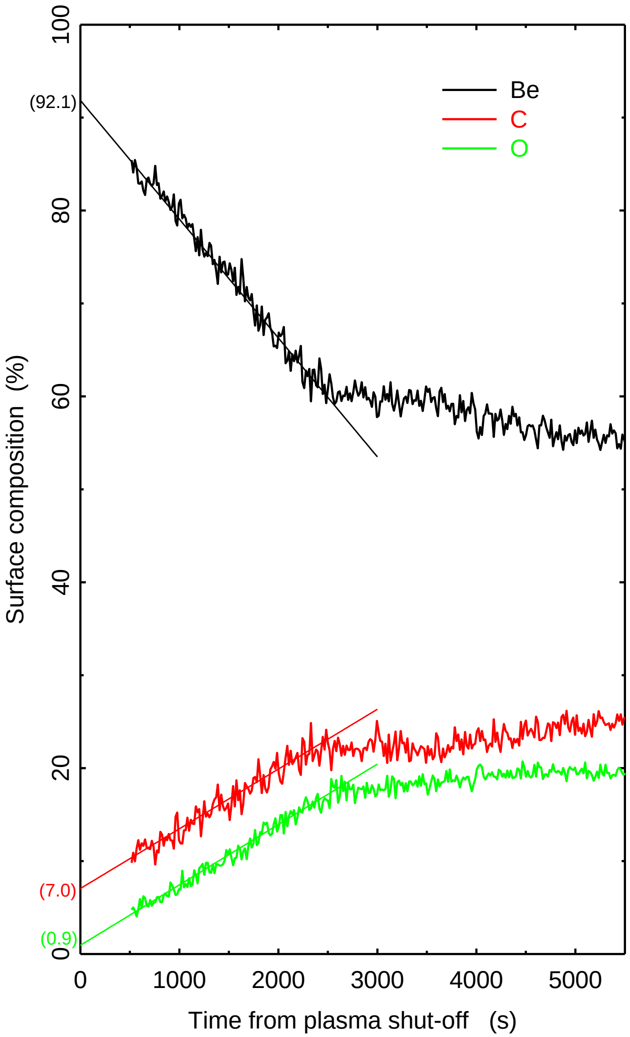


Fig. 2. Energy dependence of the sputtering yield of Be and BeO bombarded with D at normal incidence. Experimental data [6,13,23–26] and results obtained with computer simulation [6,22].

* JET data includes impurity sputtering, angle of incidence, etc.

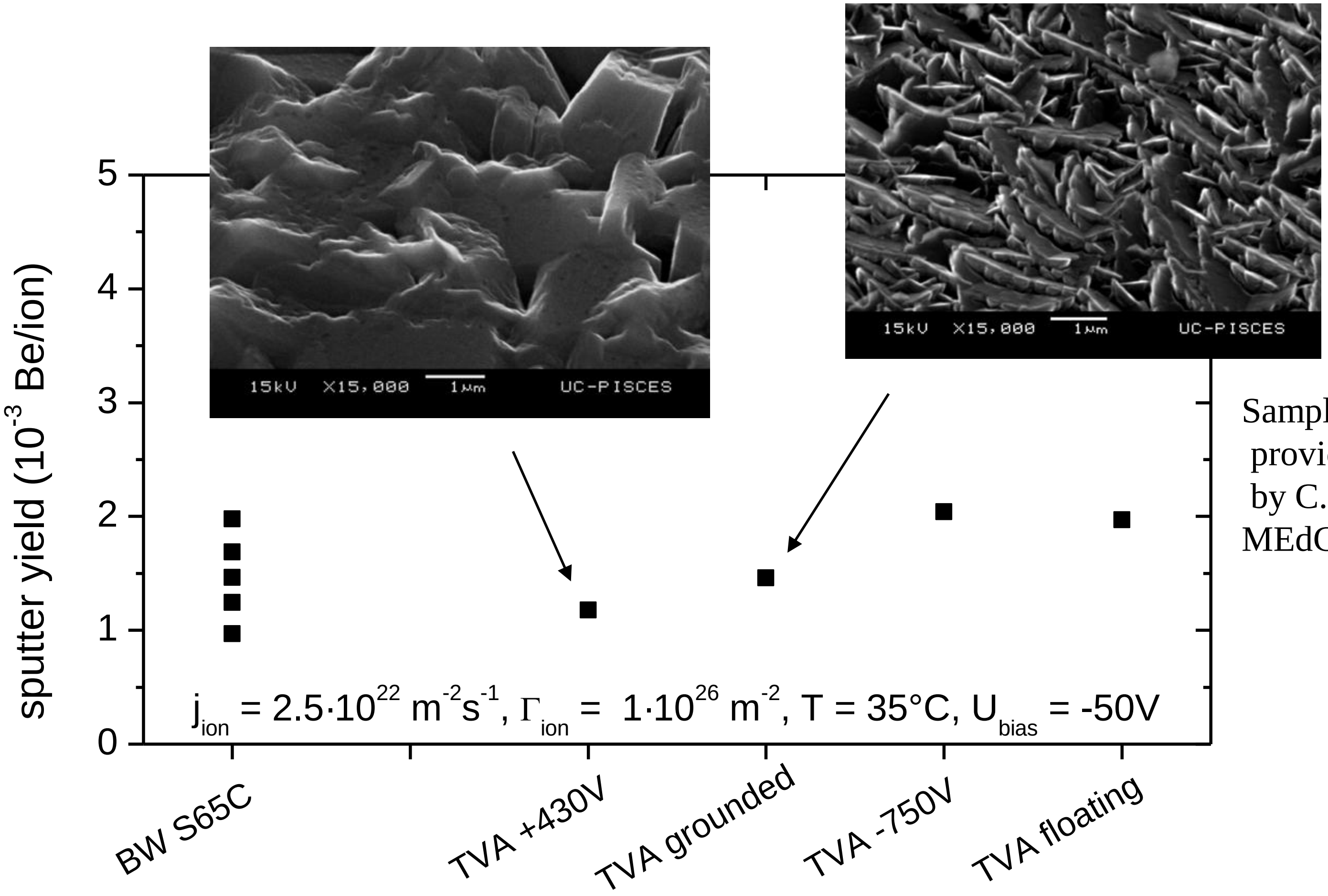
AES reveals a relatively ‘clean’ Be surface during sputtering yield measurements





sputtering of Be: influence of surface morphology

D exposure -50V, < 330K: $j_D = 1 \cdot 10^{22}\ D_x^+/cm^2$

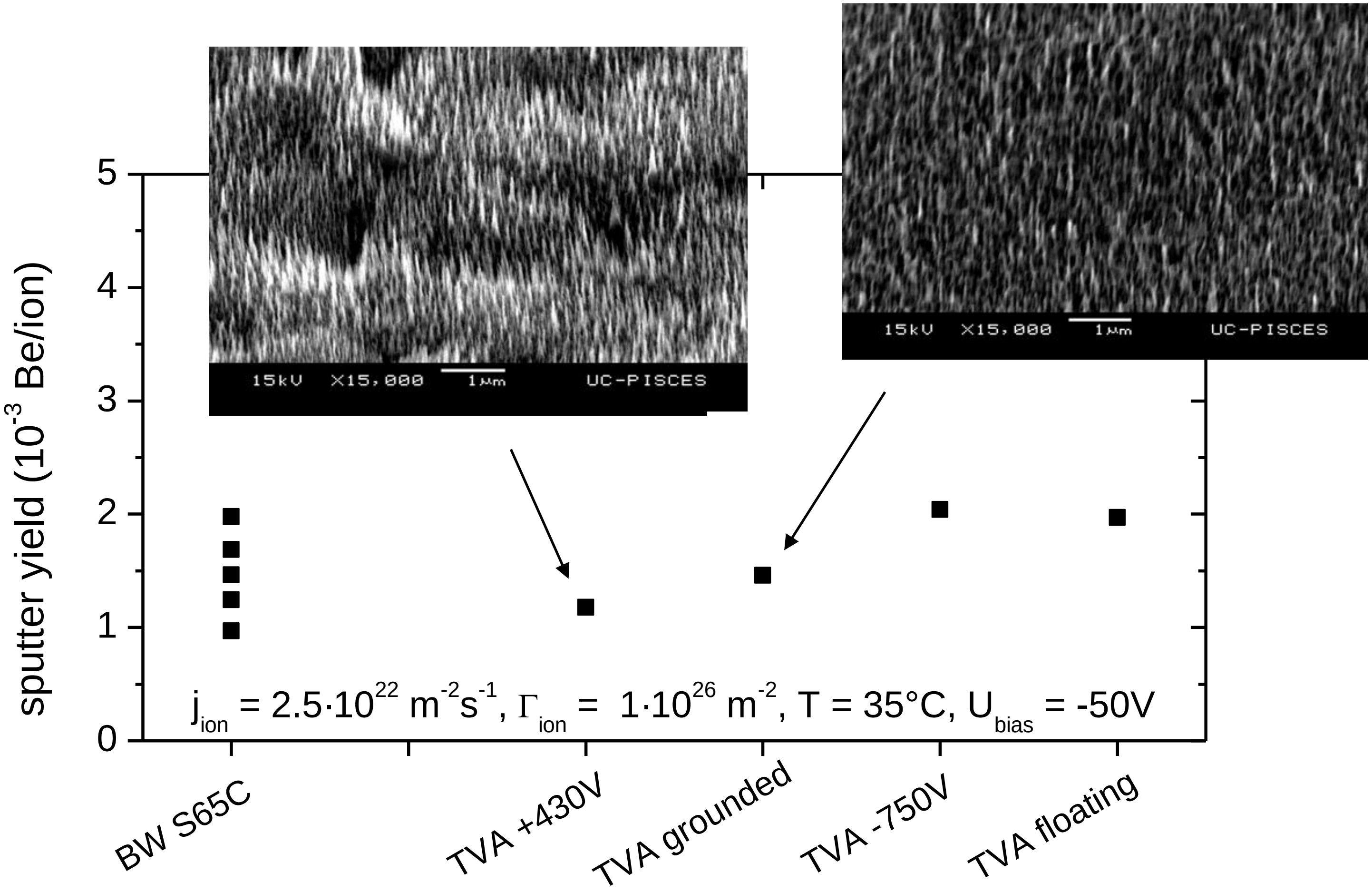


Samples
provided
by C. Lungo
MEdC, Romania

⇒ no influence within accuracy of the measurement

sputtering of Be: influence of surface morphology

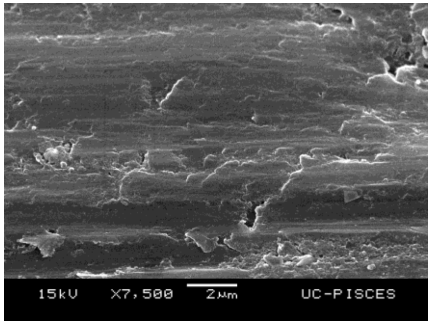
D exposure -50V, < 330K: $j_D = 1 \cdot 10^{22}$ D_x^+/cm^2



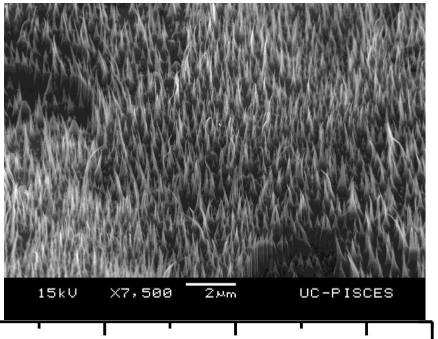
⇒ sub micron morphology develops during exposure

Surface morphology evolution with time / fluence

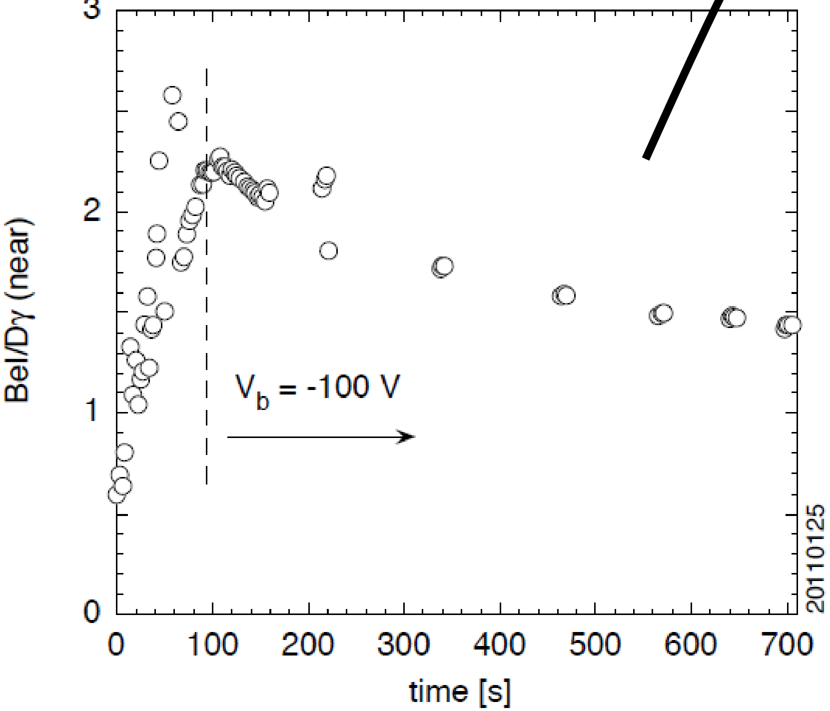
Be surface before plasma exposure



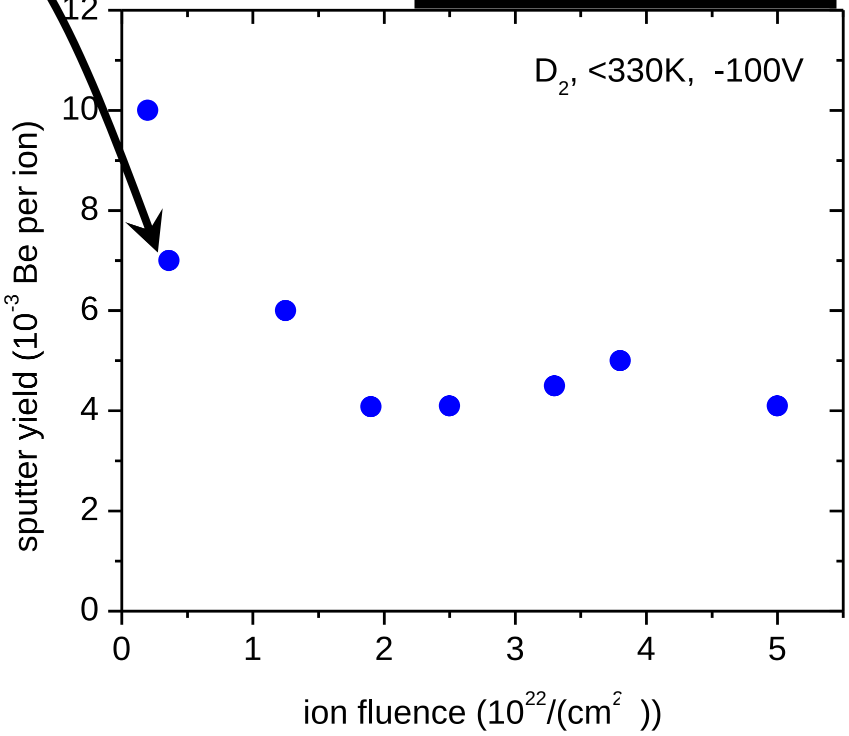
after



spectroscopy:



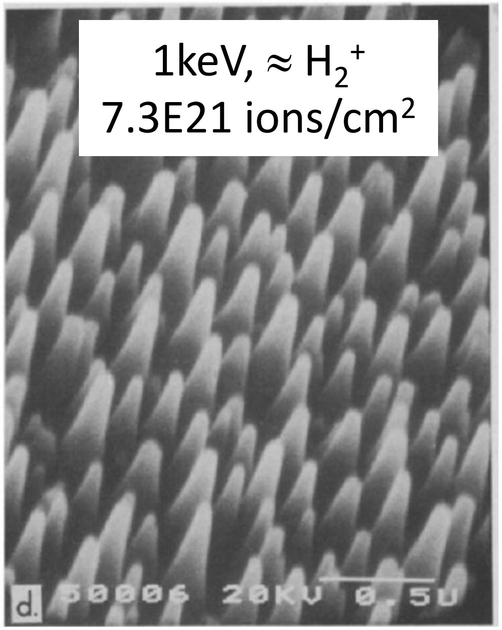
mass loss:



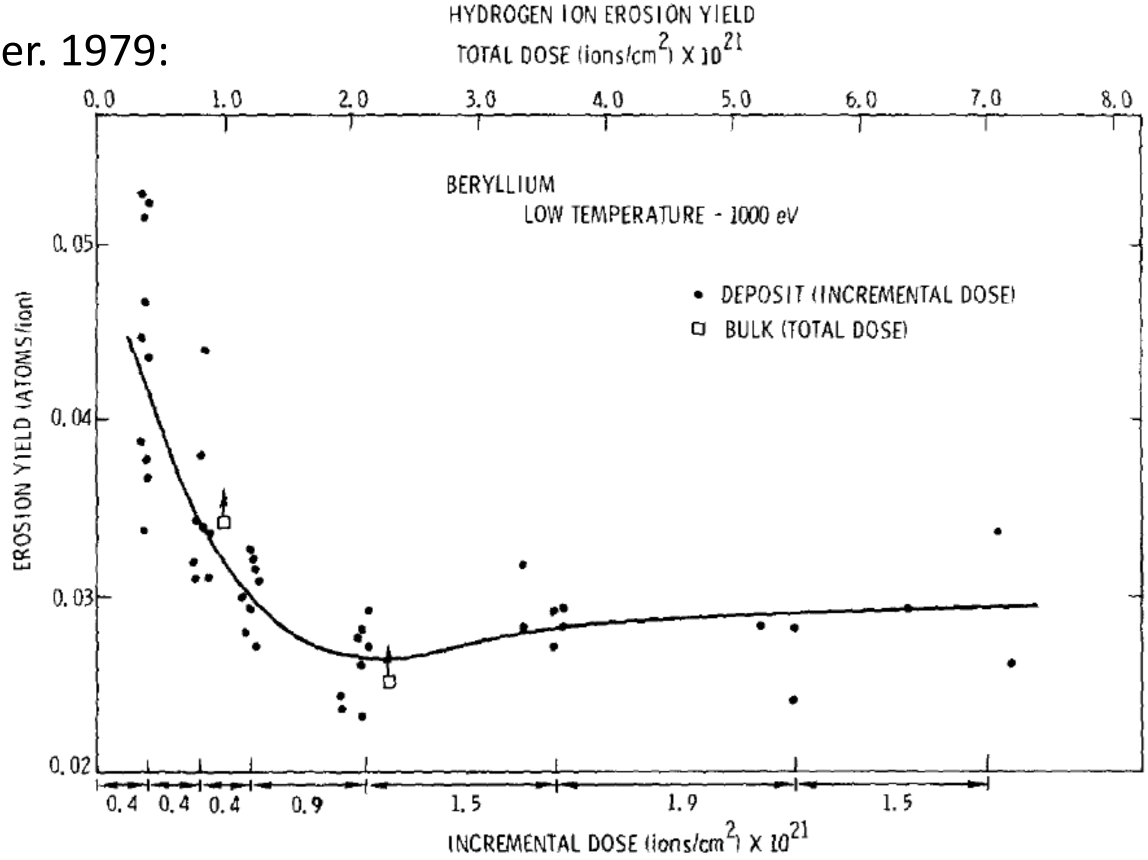
⇒ morphology change can account for a factor of 2 in reduction of the yield

Similar yield evolution with time/fluence is documented in the literature

Mattox and Sharp, J. Nucl. Mater. 1979:

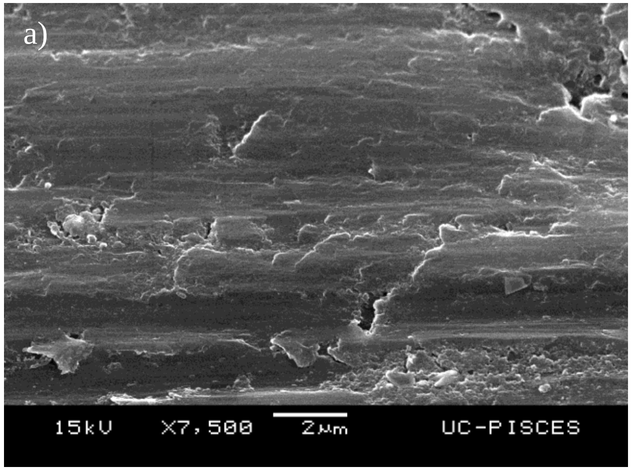


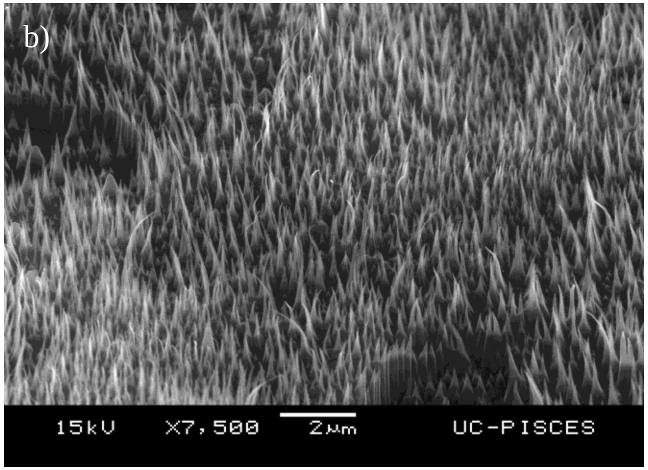
1keV, ≈ H_2^+
7.3E21 ions/cm^2



⇒ morphology change can account for a factor of 2 reduction of the yield

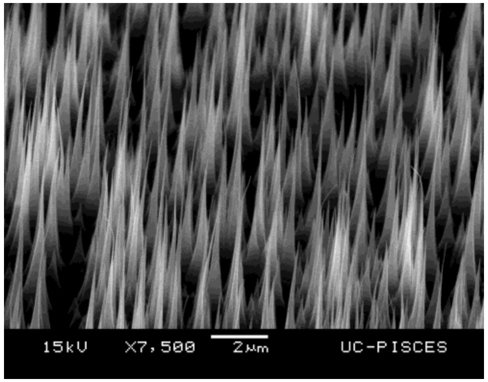
Morphology appears to saturate with fluence, feature length increases with sputtering yield



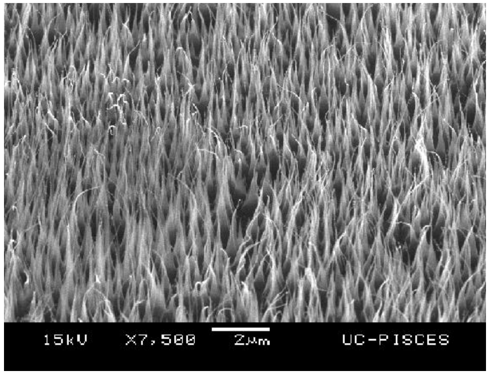


100v
0.3e22cm2
Deuterium
Plasma

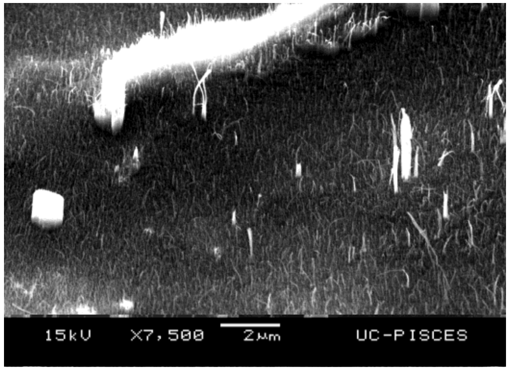
170v
3E22 CM2



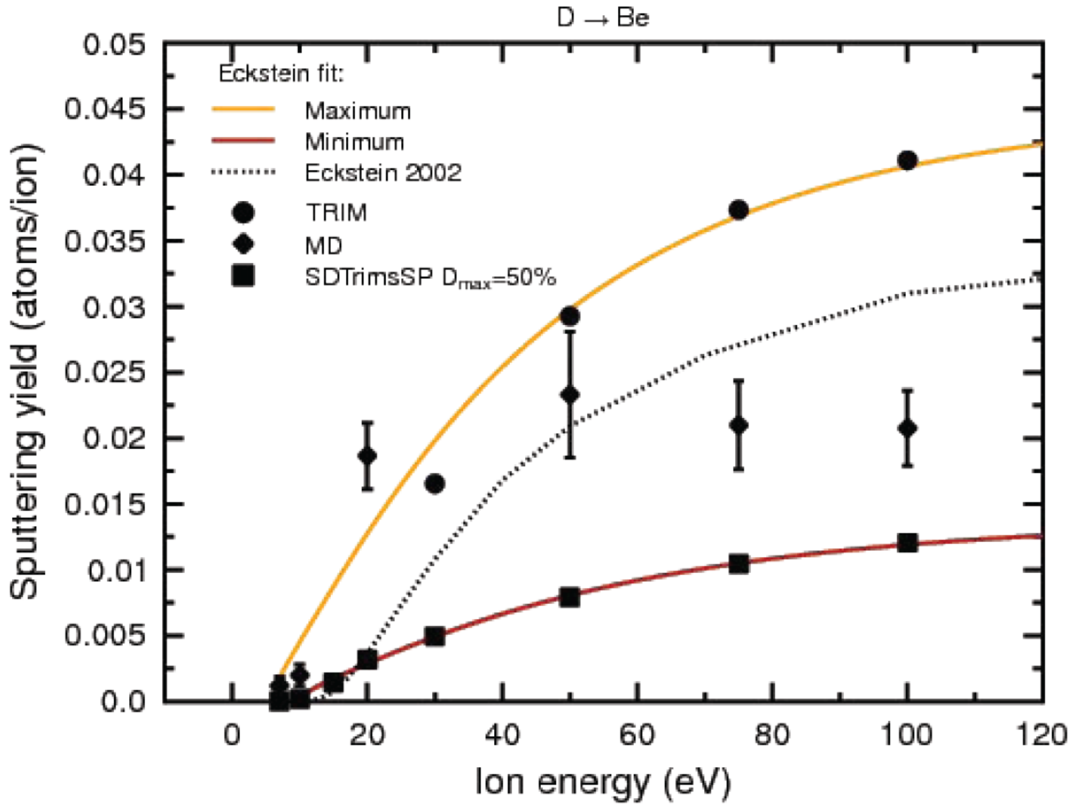
100v
3.3e22cm2



50v
3.3e22cm2



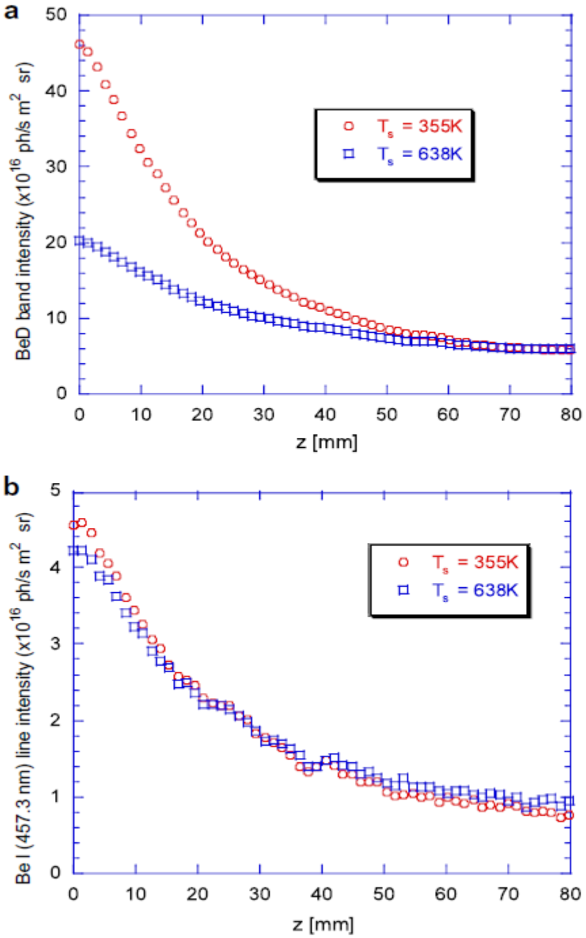
Plasma atoms remaining in the near surface also can reduce the sputtering yield by a factor of 2-4



From C. Björkas

1) “maximum” – static TRIM + MD
2) “minimum” – SDTrimSP with 50% of D (reasonable limit)

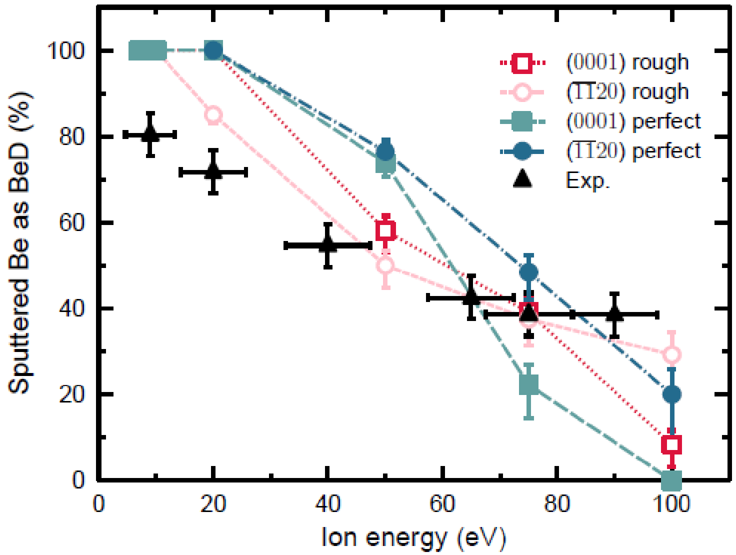
Chemically-assisted physical sputtering of BeD is temperature dependent



From R. Doerner et al, JNM 390-391 (2009) 681.

Similar e-folding distance of BeD and Be I intensity indicate BeD is physically sputtered, not chemically eroded. Beryllium deuteride is not volatile.

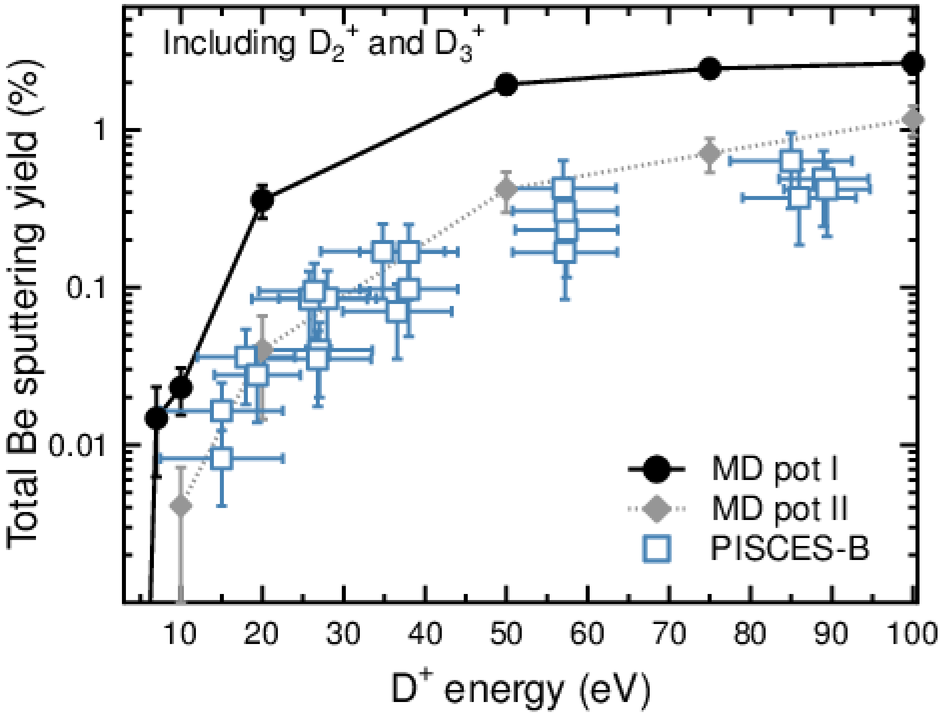
MD simulations of D on Be predicted subsequent erosion measurements



Exp. from D. Nishijima et al, PPCF 50(2008)125007.
Sim. from C. Bjorkas et al., New J. Physics (2009).

What is the appropriate potential to use for a saturated Be surface?

From: C. Björkas et al., presented at Theory of Fusion Plasmas workshop, Varenna, Italy 2012, to be published



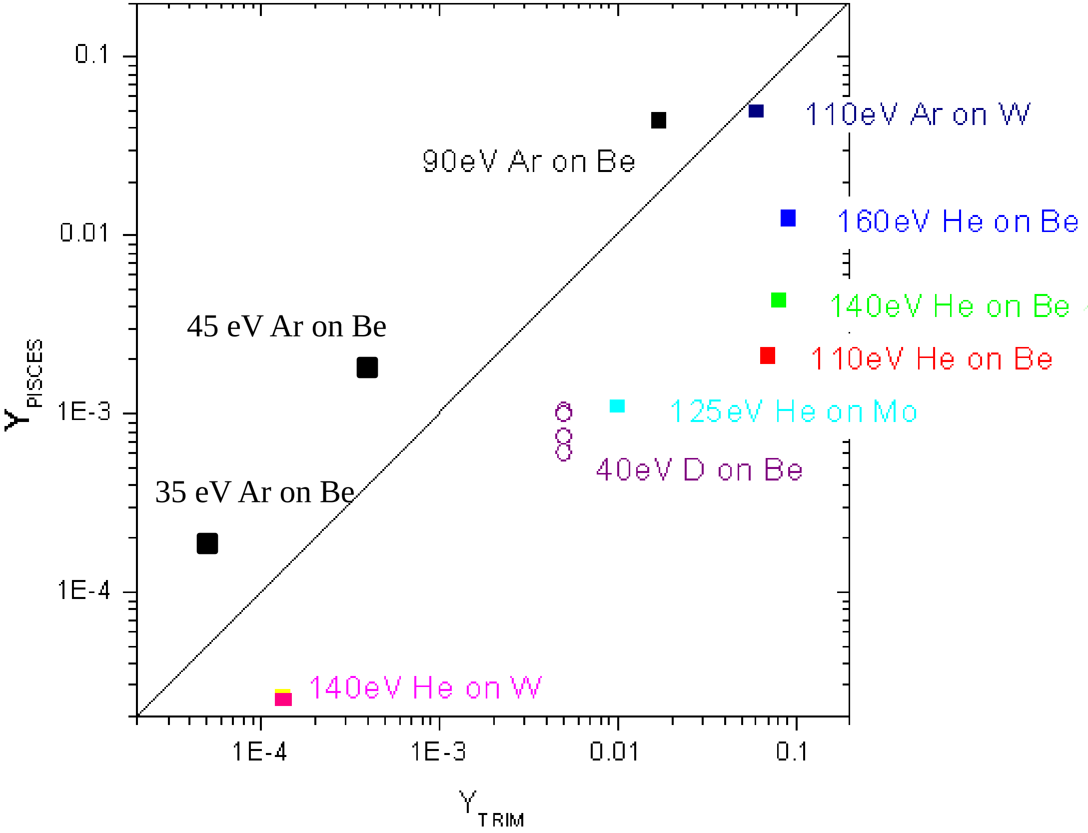
- Two similar Be-Be and Be-H potentials give markedly different results for Be sputtering
- Cohesive Energy
  - Pot I = 3.32 eV, Pot II = 3.62 eV
- Cut-off distance
  - Pot I = 2.908 A, Pot II = 2.685 A
- Binding energy at 300 K
  - Pot I = 7.8 eV, Pot II = 10.8 eV

[Pot I] C. Björkas, K.O.E. Henriksson, M. Probst, and K. Nordlund. *Journal of Physics: Condensed Matter*, 22:352206, 2010.

[Pot II] C. Björkas, N. Juslin, H. Timko, K. Vörtler, K. Henriksson, K. Nordlund, and P. Erhart. *Journal of Physics: Condensed Matter*, 21:445002, 2009.

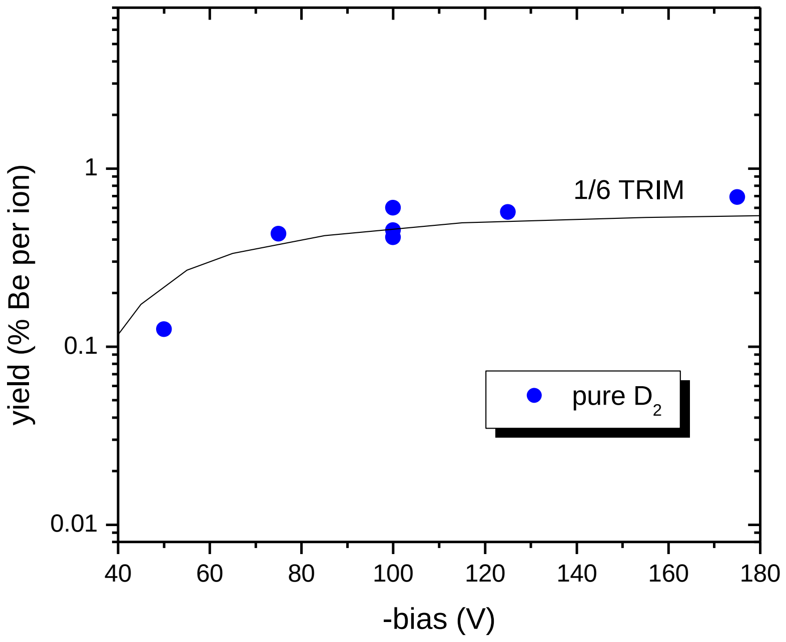
On the other hand, heavy ion bombardment yield agrees with TRIM calculations

- Reason for this behavior is not understood
- Ar on Be results in smoother surface after sputtering
- Ar implantation depth is shallower
- Reflection coefficient of Ar is lower than He or D (more momentum directed into target)
- Ar diameter is larger, perhaps less likely to reside in the near surface region
- Effect is measured for a variety of substrate materials



Erosion/deposition balance in Be seeded high flux D discharges

Mass loss

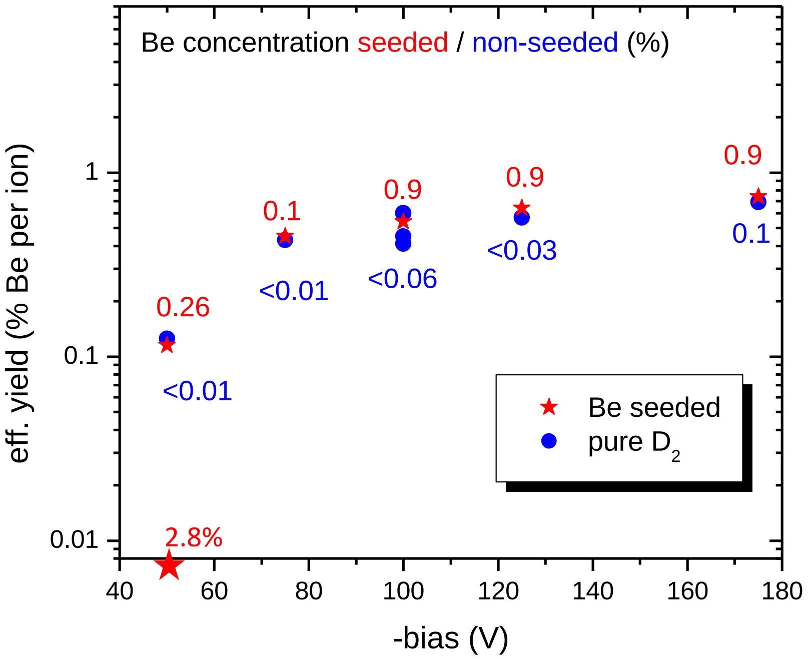


ion fluence: ≈$10^{22}/cm^2$
target temperature < 320K

- Use Be oven seeding to balance surface erosion to test input parameters of material migration models
- Mass loss measures net erosion
- Spectroscopy measures gross erosion (Be I line)
- $Y_{Be \rightarrow Be} \approx Y_{D \rightarrow Be}$, and low concentration of Be
- When incident/seeded Be ion flux = sputtered flux of Be, net erosion should = 0.

No change in mass loss is measured when Be seeding flux equals sputtering of Be by D

Mass loss



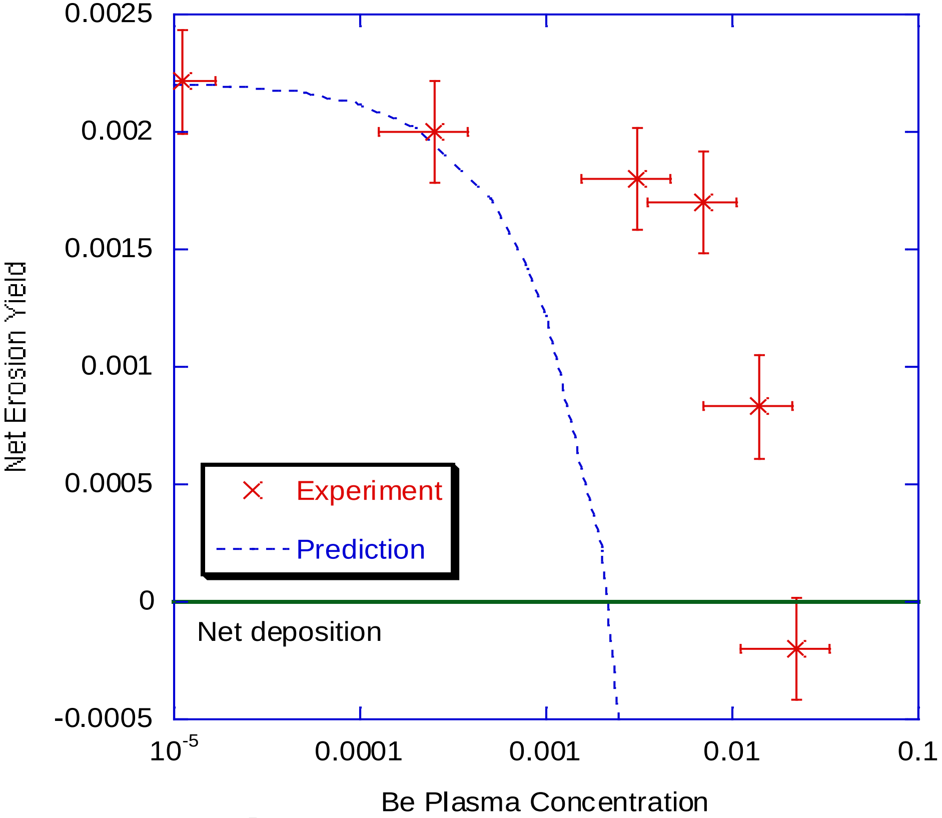
ion fluence: ≈10^{22}/cm^2
D/Be plasma
target temperature < 320K

- ADAS database is used
- Be flux from Be II (313.1 nm) and background plasma flow velocity (E. Hollman JNM, PSI-19)
- Be ion flux is verified during no bias discharges, when weight gain is measured (net deposition)
- Net erosion stays constant, implying gross erosion must increase
- Erosion yield of 0.15% can only be compensated by seeding 2.8% Be

Net erosion does not change as expected

PISCES

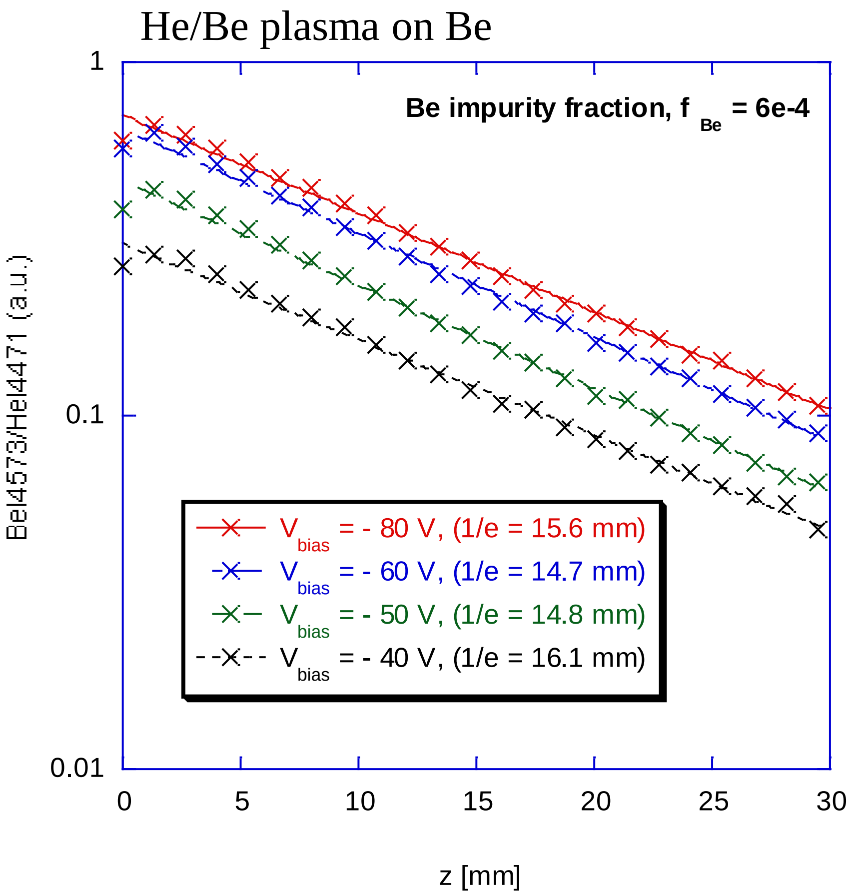
Weight loss measures net erosion



D/He/Be plasma,
50 eV ion energy

- Use reproducible plasmas while simply changing the Be oven temperature
- Net erosion remains ~ constant until Be influx >> sputtering rate
- Seeding rate must exceed erosion rate by a factor of 10 to reach net deposition
- Two possibilities, reduced sticking of depositing Be, or increased re-erosion, could explain observations

Penetration distance is independent of incident ion energy, inconsistent with particle reflection

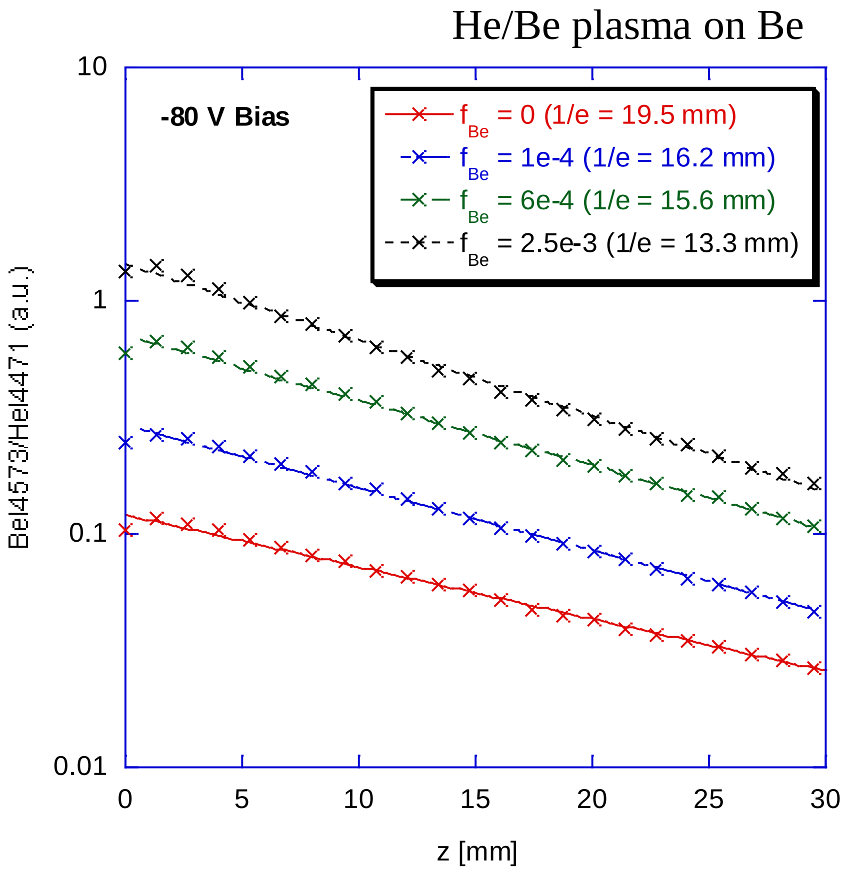


- <Sputtered Be particle energy> ~0.5 E_{bind}, or 1.6 eV for Be
- Penetration distance should increase with bias voltage if there is a large increase in reflected particles when Be oven is on

V_{bias}	$E_{Be\ ion}$	$E_{reflect\ Be}$
- 80 V	67 eV	4.2 eV
- 60 V	47 eV	3.1 eV
- 50 V	37 eV	2.5 eV
- 40 V	27 eV	2.0 eV
- 30 V	17 eV	1.3 eV

Penetration distance of Be atoms into the plasma becomes shorter when Be oven is on

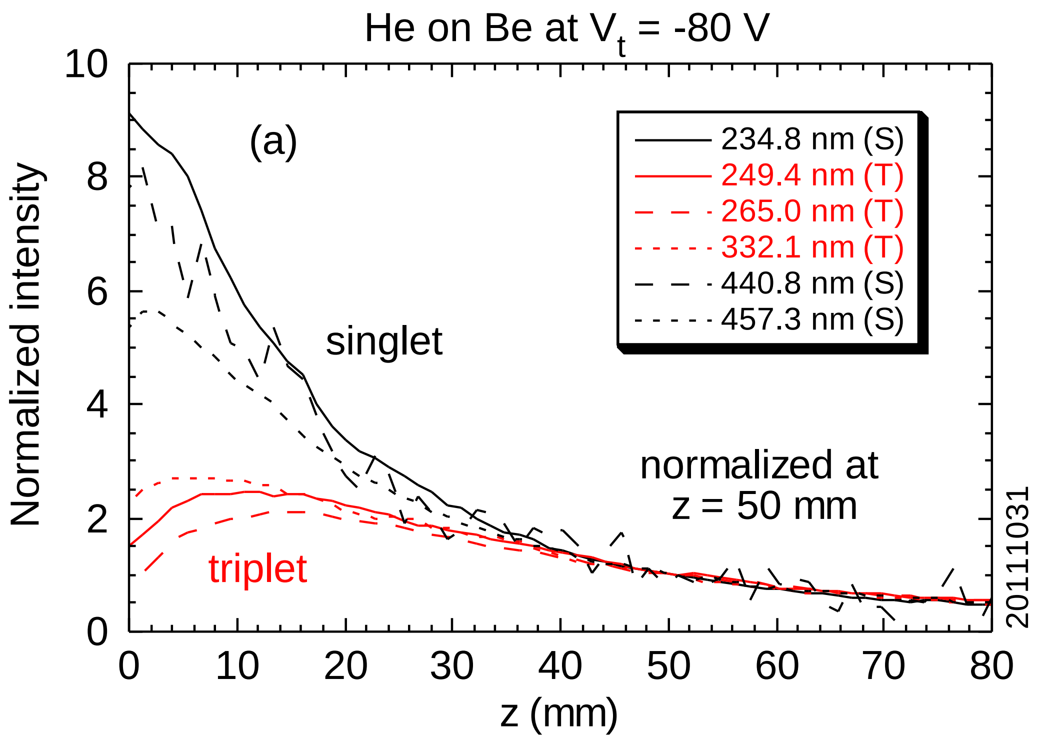
- Yield α (1/Binding Energy)
- Higher erosion rate implies a lower binding energy, which equates to a smaller surface release velocity, and therefore a shorter penetration distance
- Increased Be I intensity indicates larger Be surface atom loss rate (i.e. larger gross erosion)



Clear difference in axial profiles between Be I singlet and triplet transitions is observed.

- Axial Be I emission intensity profiles from sputtered Be atoms
- Singlet lines are more peaked in front of the target than triplet lines.

What causes this difference?



n_e ~ 20x10^{18} m^{-3}
T_e ~ 4.5 eV
Γ_i ~ 12x10^{22} $m^{-2}s^{-1}$
P_{He} ~ 13 mTorr

Are metastable atoms sputtered at a higher velocity?

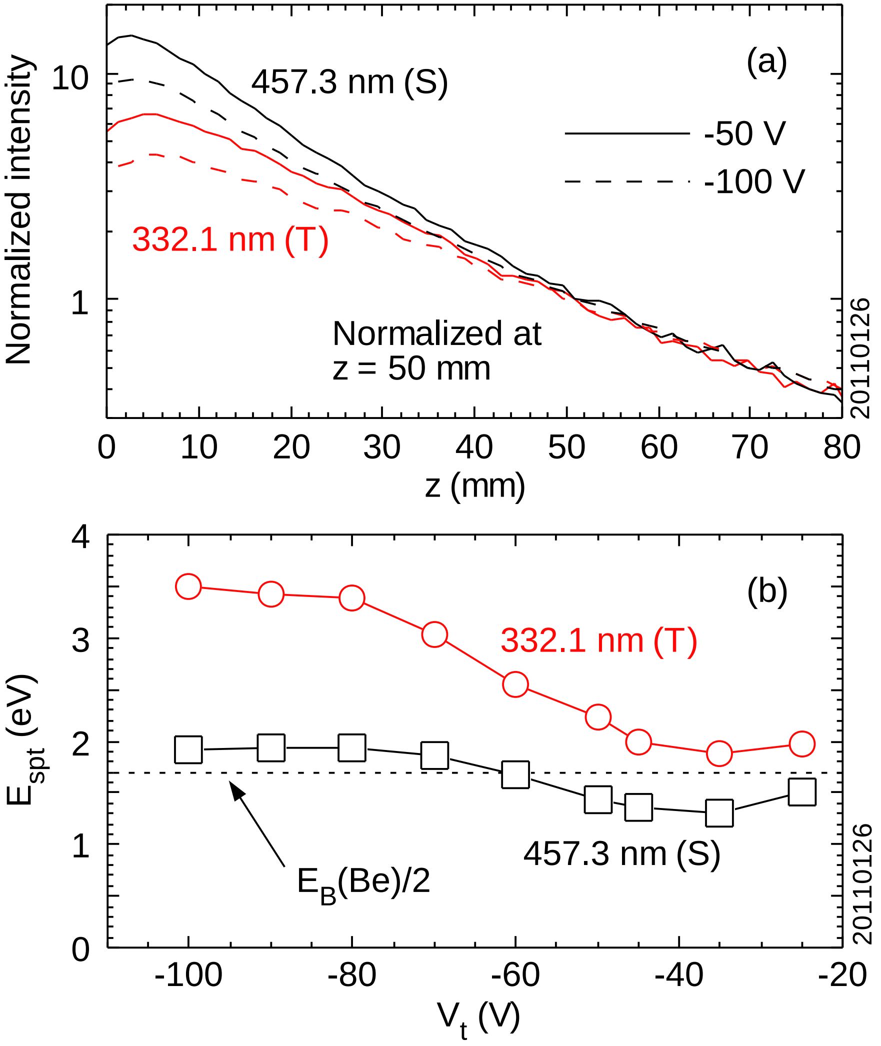
- With increasing target bias voltage V_t, emission profiles become flatter.

The sputtered energy becomes higher.

- Sputtered energy of metastable atoms is ~1.5x higher than that of ground state atoms at V_t = -100 V.

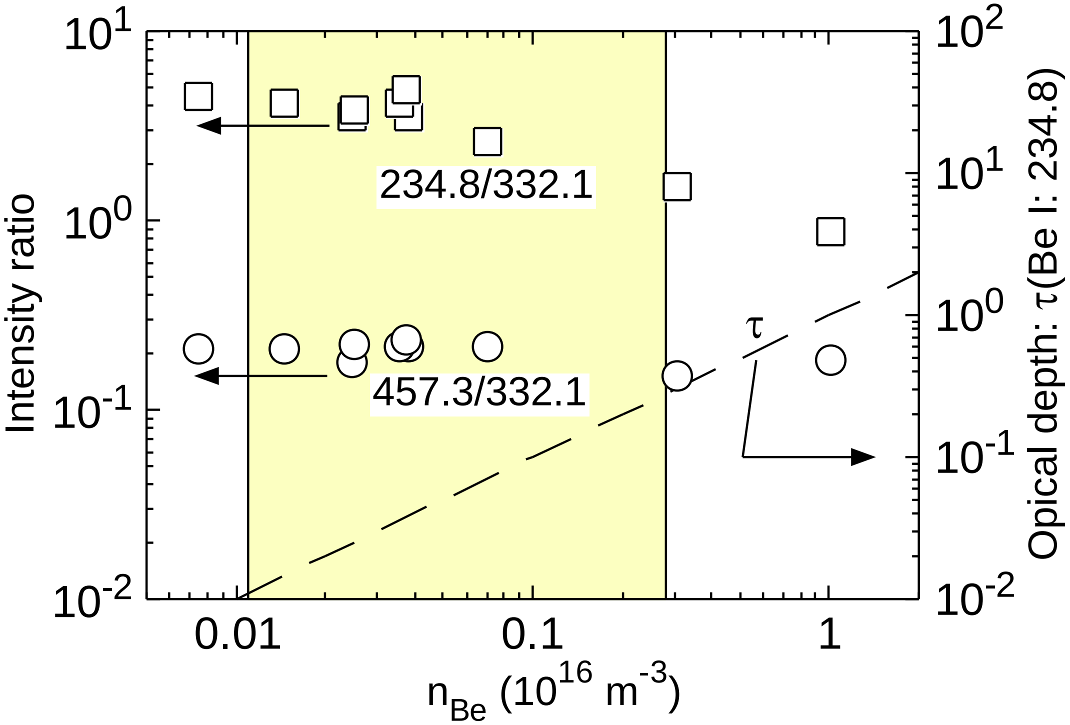
Calculated by assuming that electron impact ionization is the dominant process for the decay:

$$\lambda_{mfp} = \frac{v_{Be}}{\langle \sigma v \rangle_{Be \rightarrow Be+} n_e}$$



The resonance transition at 234.8 nm starts to be reabsorbed at n_{Be} > 0.1e16 m^{-3}.

- 234.8 nm/332.1 nm decreases with photon absorption at 234.8 nm.
- 457.3 nm/332.1 nm is expected to increase with photon absorption of resonance transitions, but not affected yet in this n_{Be} range.



Optically thick when τ > 1

(Measured in Be seeded He plasma)

Possible topics for discussion:

- Mixed materials (erosion of and retention in), impurity effects
- Impurity flow speed in a flowing background plasma
- Retention due to implantation vs. codeposition
- Quantifying the amount of gas atoms in a surface during plasma exposure, impact on erosion
- Developing MD potentials for a gas saturated surface
- Quantifying surface morphology change on erosion
- Re-erosion of deposits, reflection probability

- What are the relevant parameters for inclusion in a database
  - Erosion: Temp., Energy, Angle, Flux, Morphology, Composition, …
  - Retention: Temp., Energy, Flux, Morphology, Composition, …