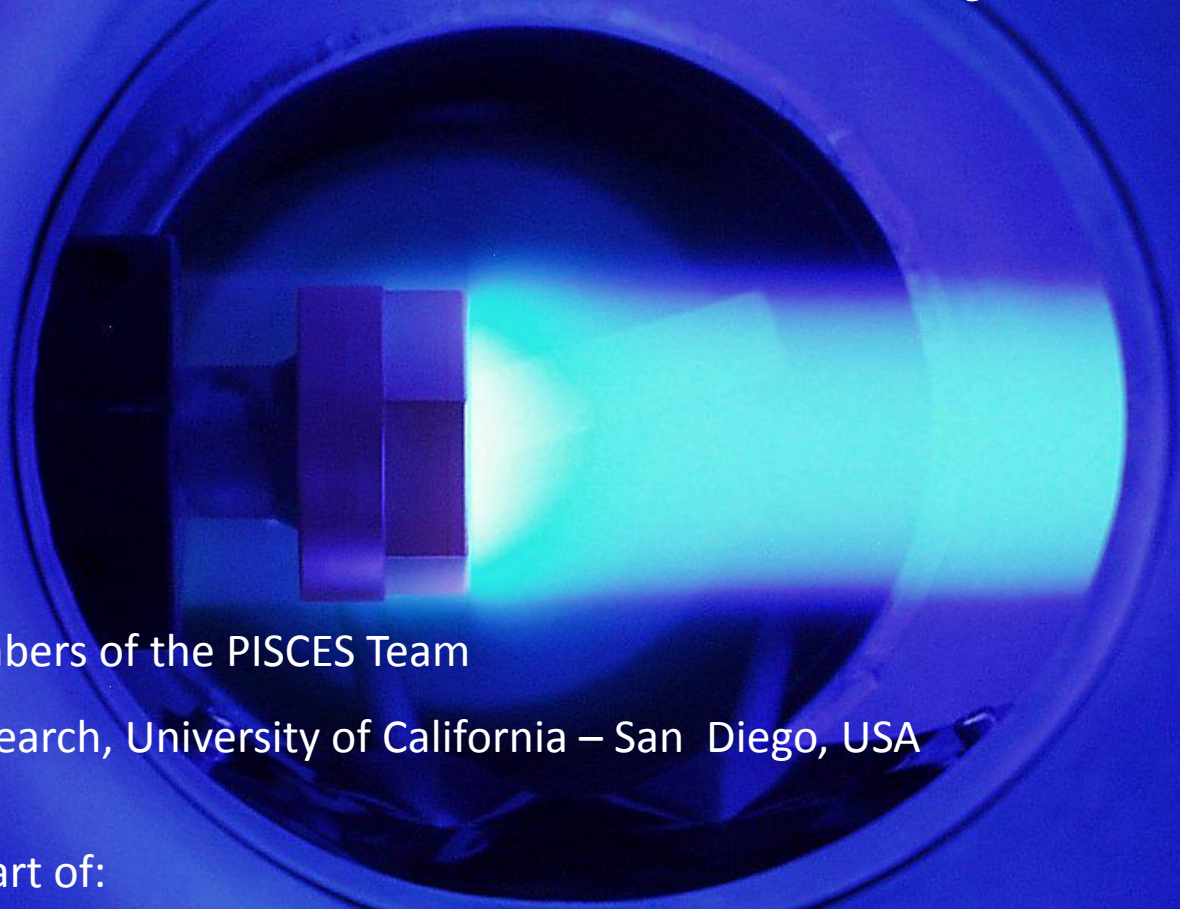
Plasma interactions with Be surfaces



R. P. Doerner for members of the PISCES Team

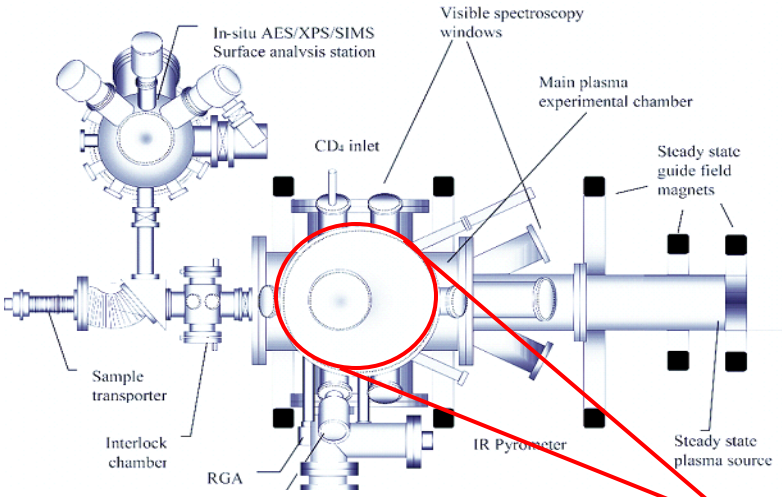
Center for Energy Research, University of California – San Diego, USA

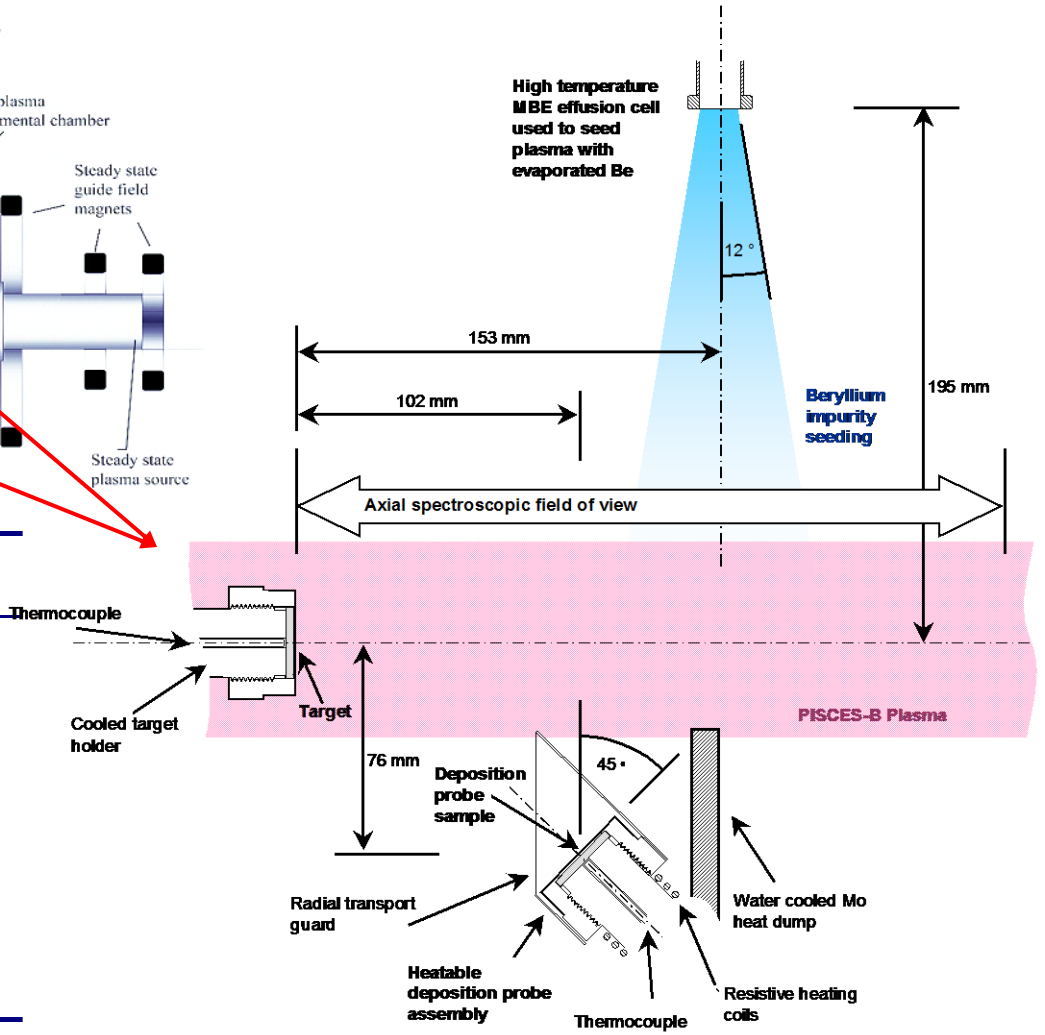
Work performed as part of:

US-EU Collaboration on Mixed-Material PMI Effects for ITER

US- Japan Technology Exchange Program

PISCES-B experimental arrangement

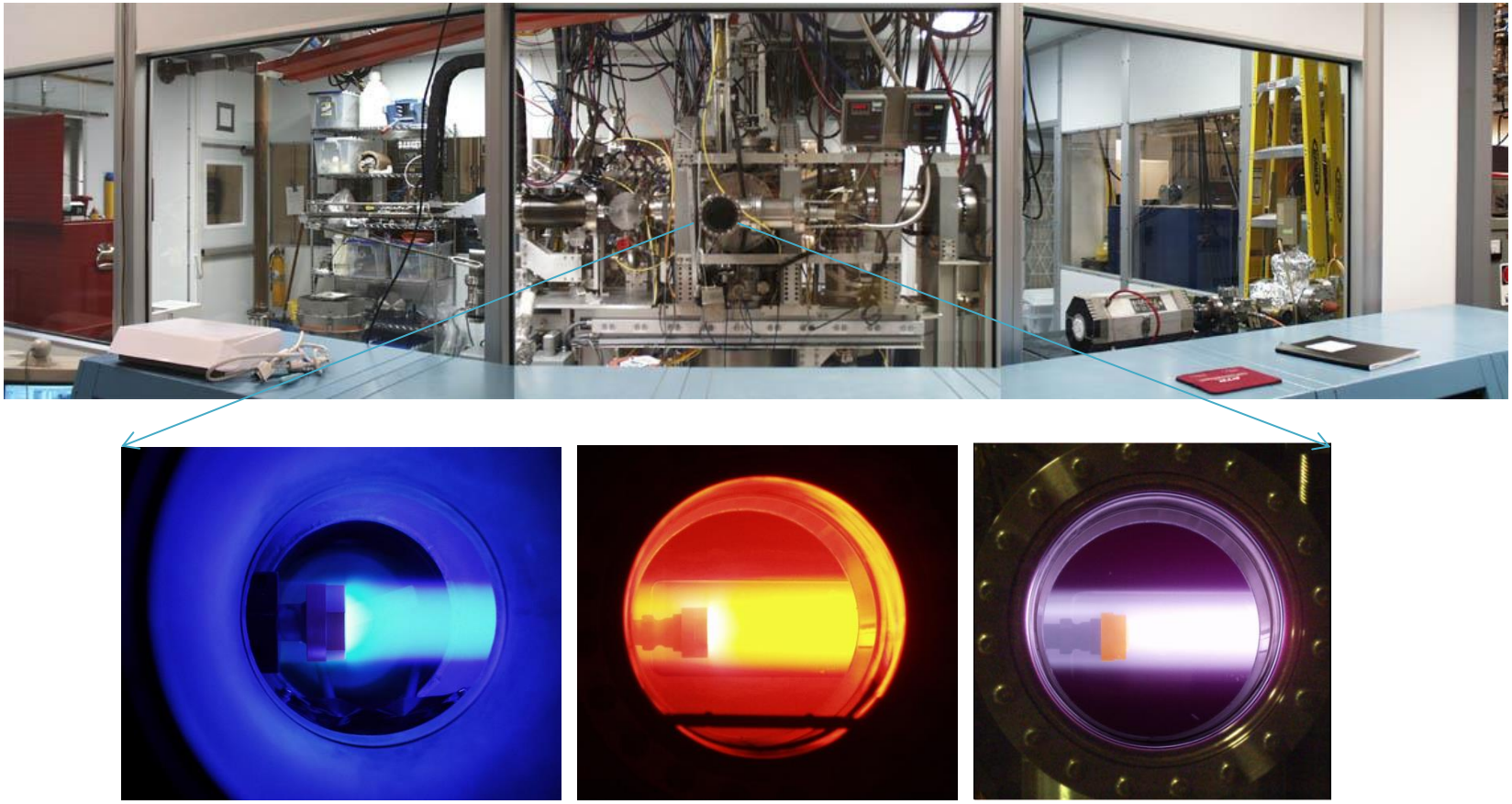




PISCES

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

PISCES-B linear plasma facility at UCSD



Many pure elemental species plasmas (Ar, Ne, D) and mixed species plasmas (D/He, D/Ar) are possible in the PISCES devices

Summary of Be PMI R&D in PISCES

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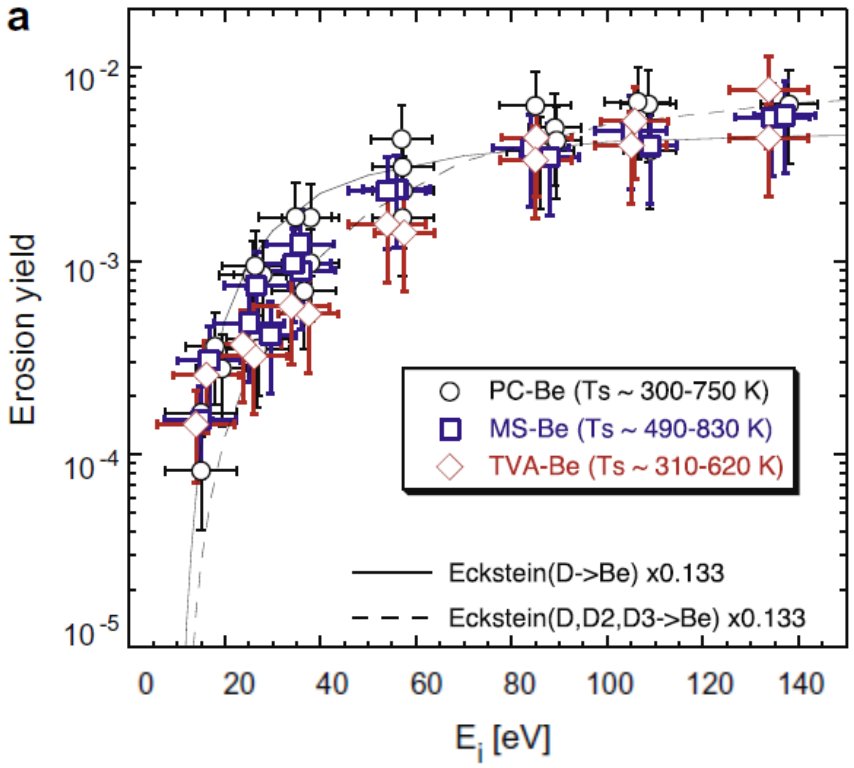
Pure D, He, or mixed D/He plasma bombardment produces less Be sputtering than Eckstein's TRIM predicts

D. Nishijima et al., JNM 390-391(2009)132.



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

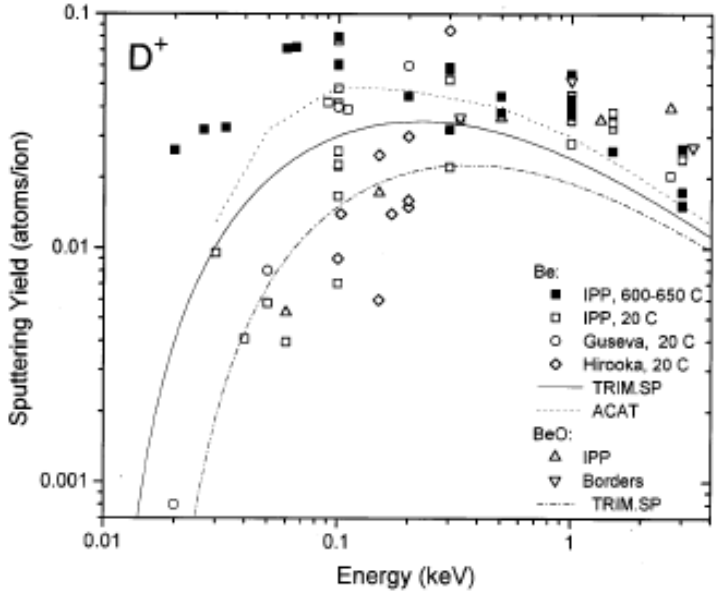
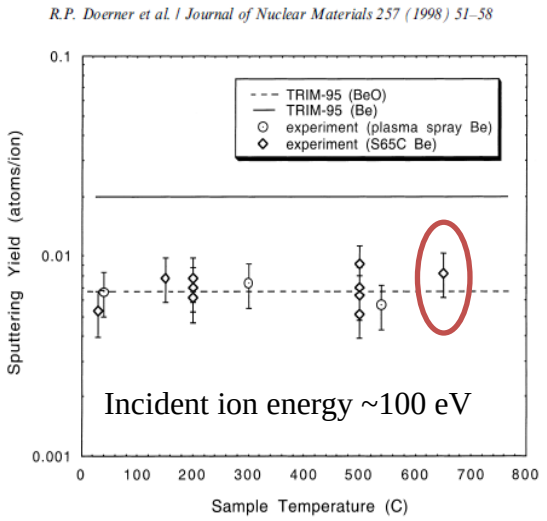
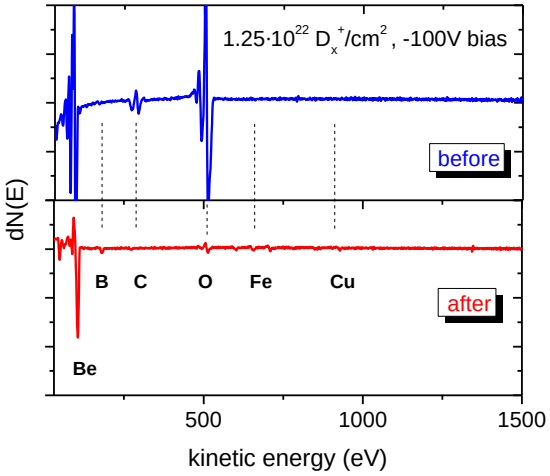
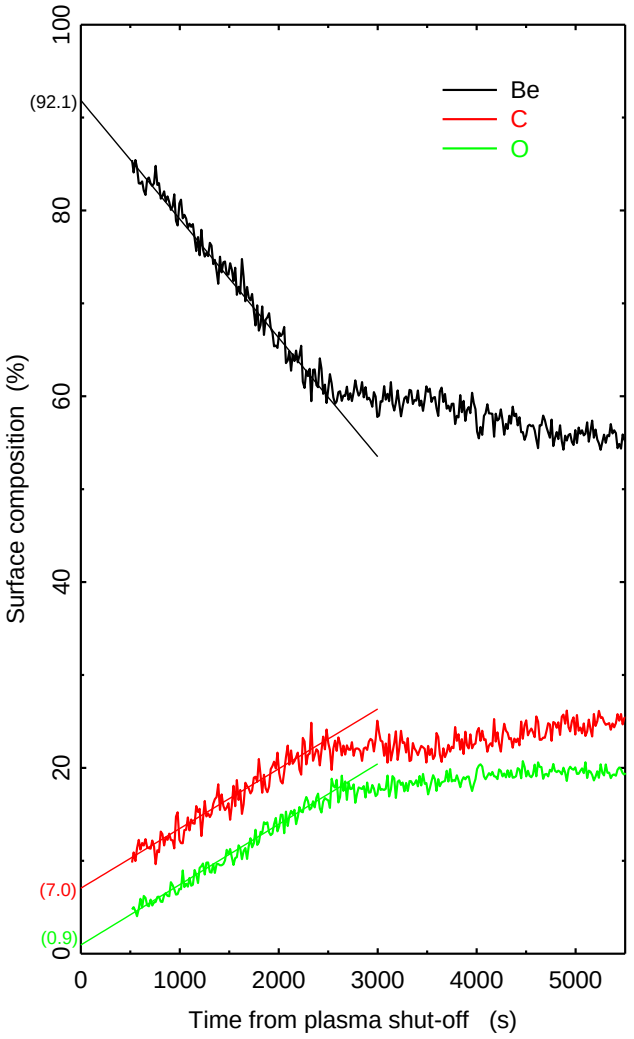


Fig. 2. Energy dependence of the sputtering yield of Be and BeO bombarded with D at normal incidence. Experimental data [6,13,23–26] and results obtained with computer simulation [6,22].

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

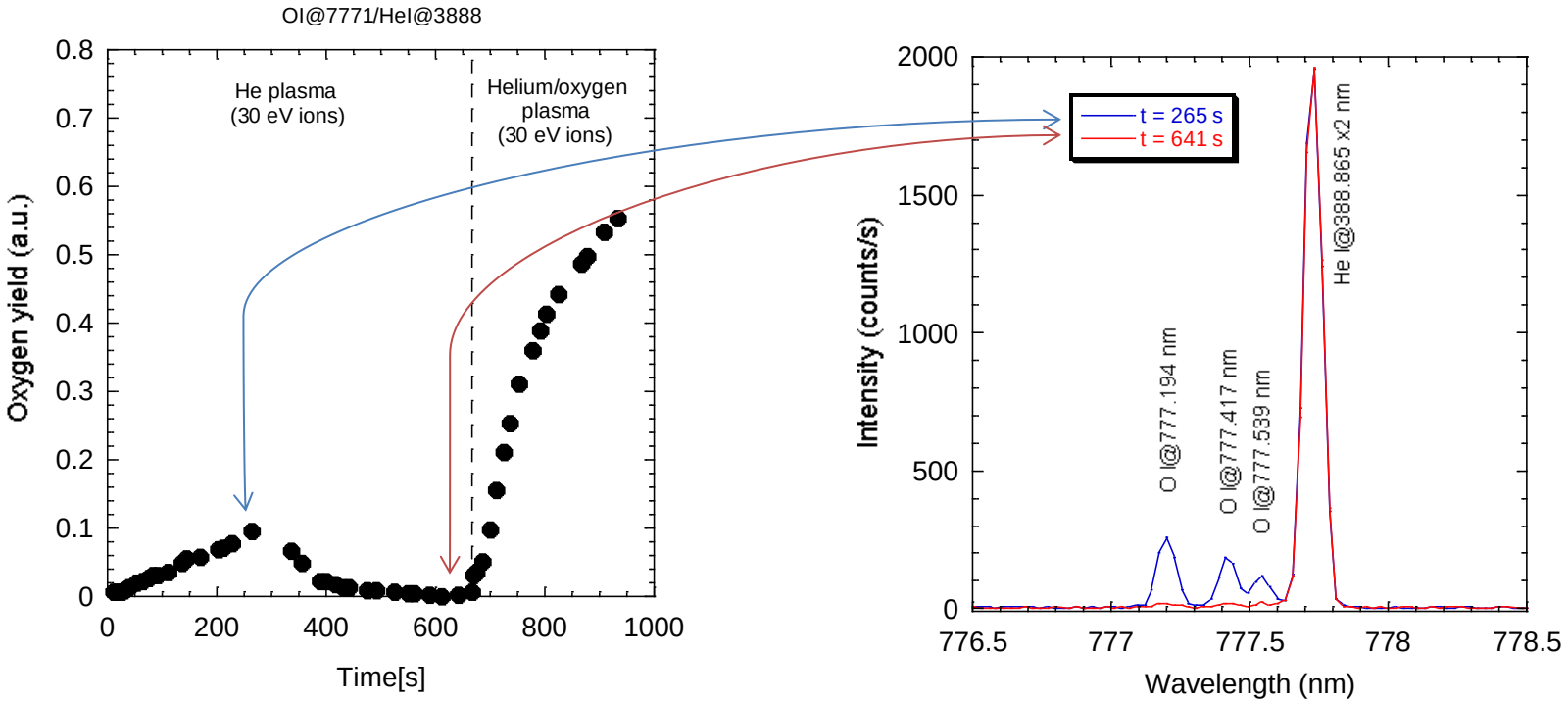






Native beryllium oxide surface is removed early during the plasma exposure

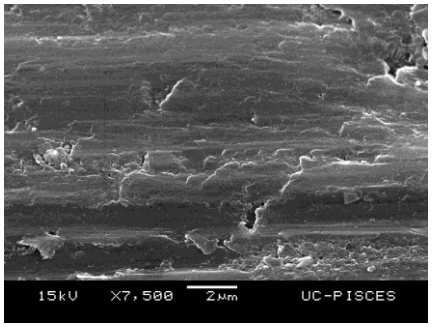
PISCES



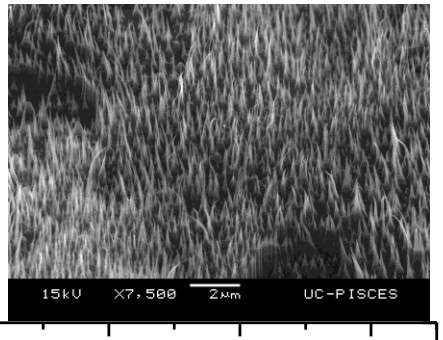
- Distinctive oxygen lines near 777 nm can monitor erosion of O from surface
- Background helium plasma does not change (second order He I line)
- Larger ion energy or flux will remove oxide layer quicker
- BeO (0,0) molecular band emission (@ 470.86 nm) is not detectable

Development of surface morphology decreases erosion

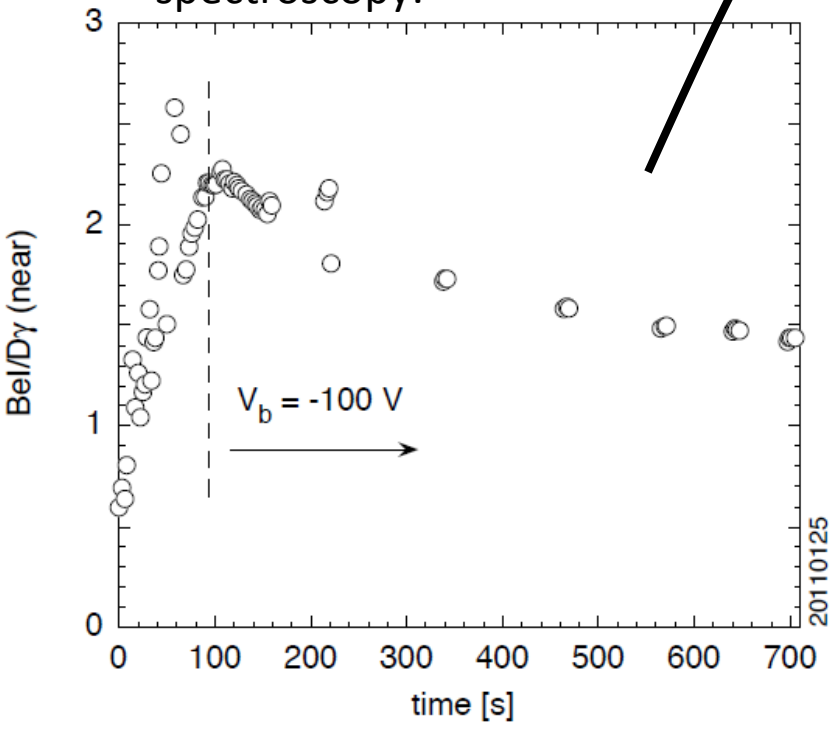
Be surface before plasma exposure



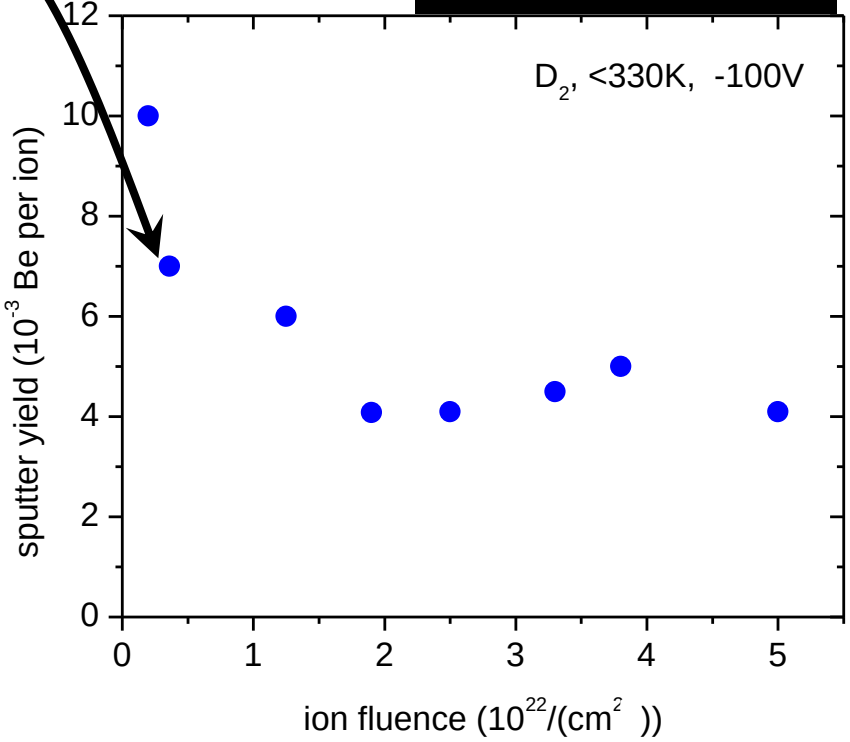
after



spectroscopy:



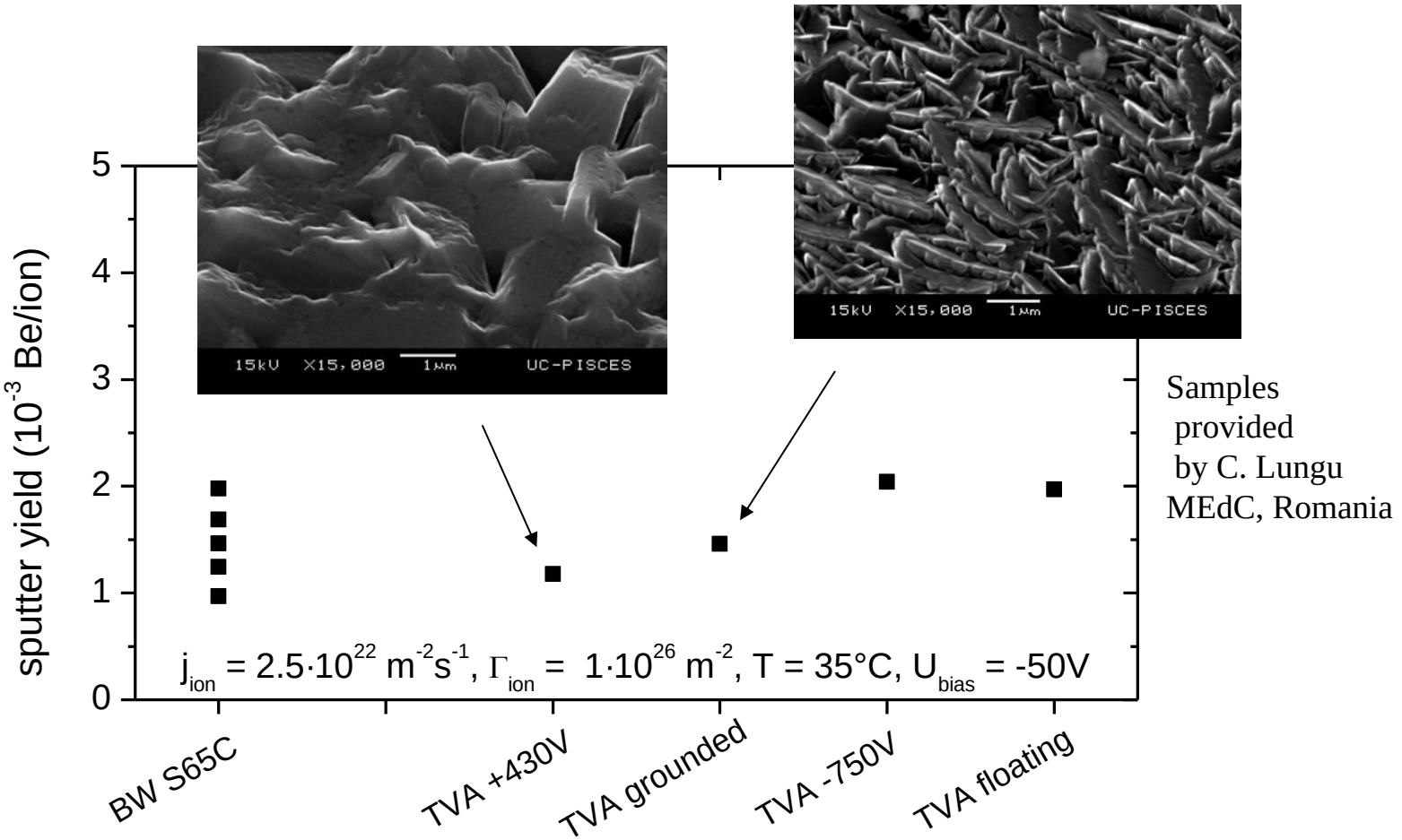
mass loss:



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

Initial surface morphology does not significantly affect erosion

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

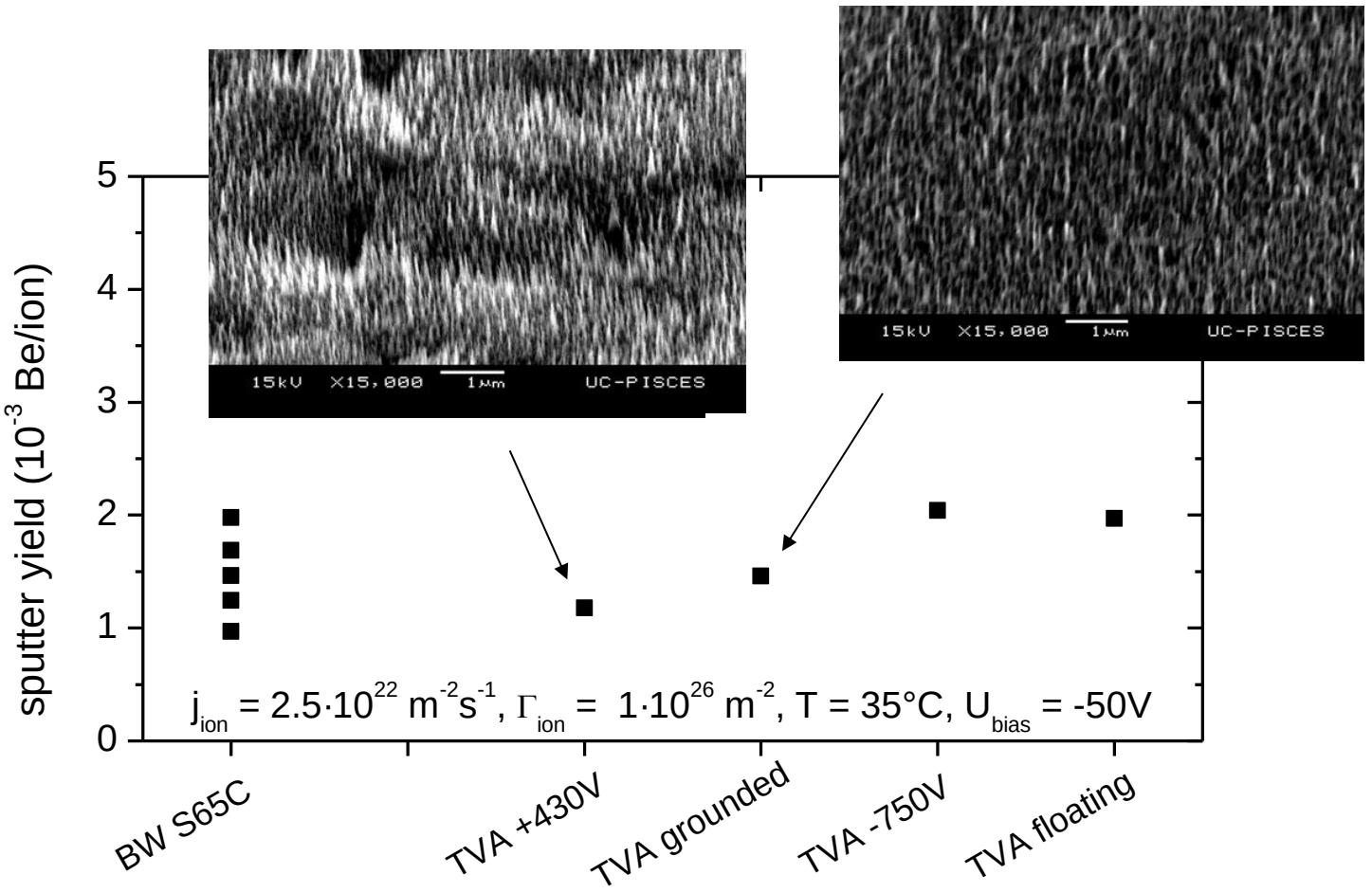


Samples provided by C. Lungu MEdC, Romania

⇒ **no influence within accuracy of the measurement**

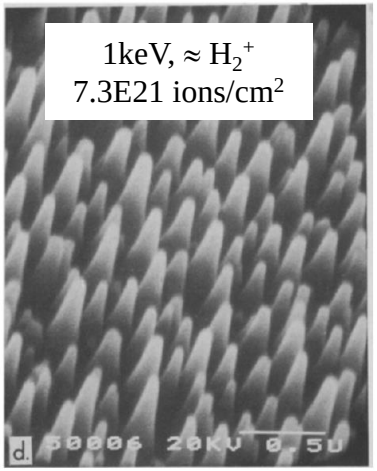
Initial surface morphology does not significantly affect erosion

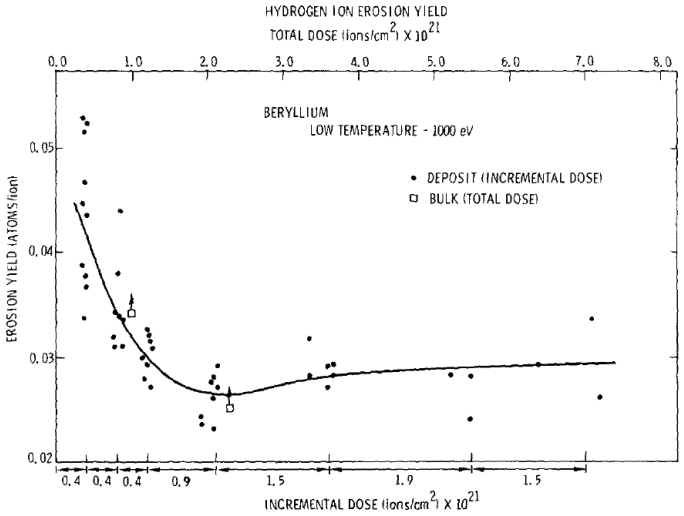
D exposure -50V, < 330K: $j_D = 1 \cdot 10^{22}\ D_x^+/cm^2$



⇒ **sub micron morphology develops during exposure**

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



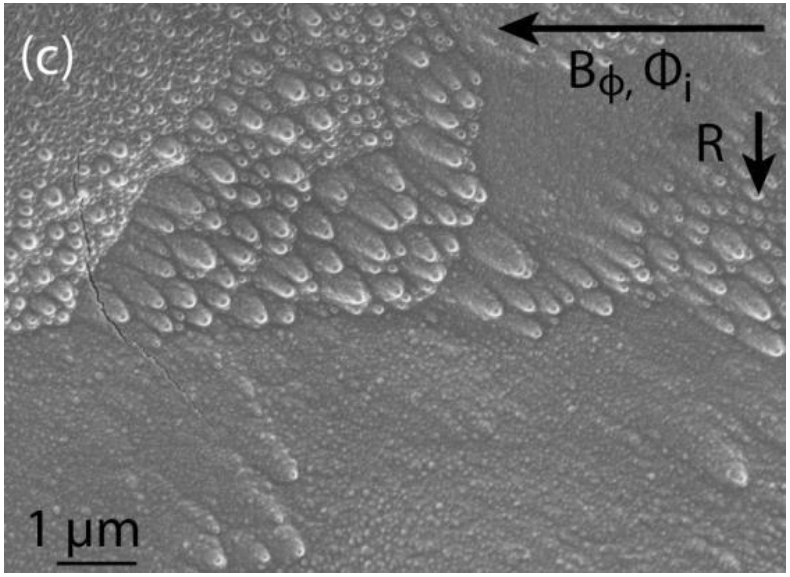


High flux ion beam
Mattox and Sharp, JNM 80(1979)115

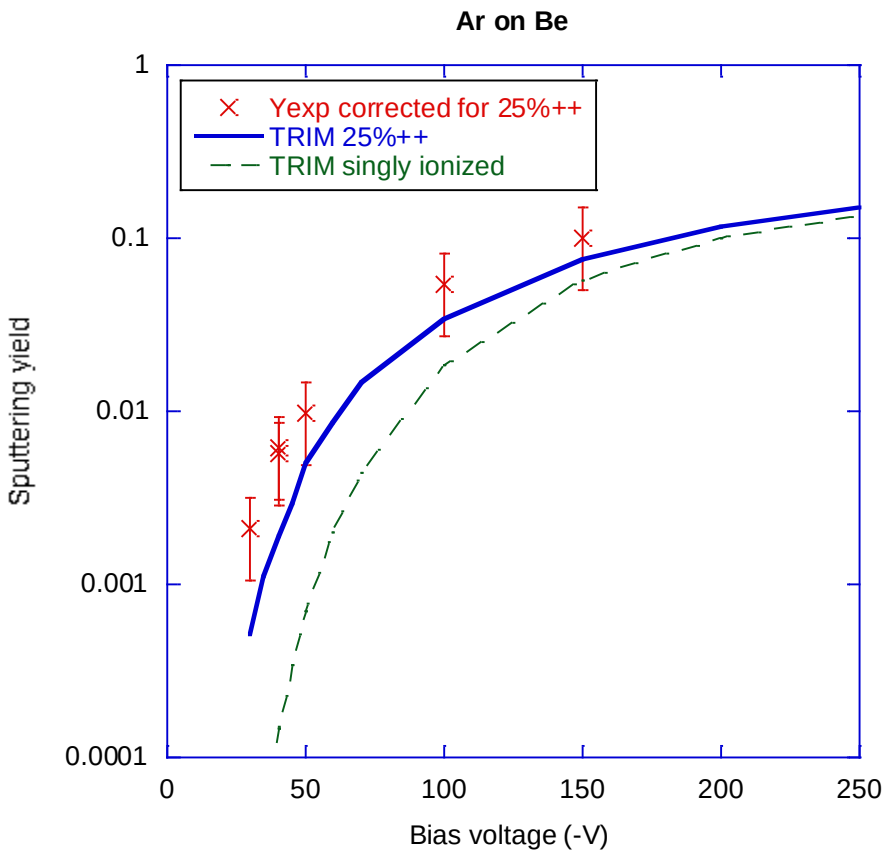
JET-ILW

H. Bergsaker et al., JNM 463(2015)956

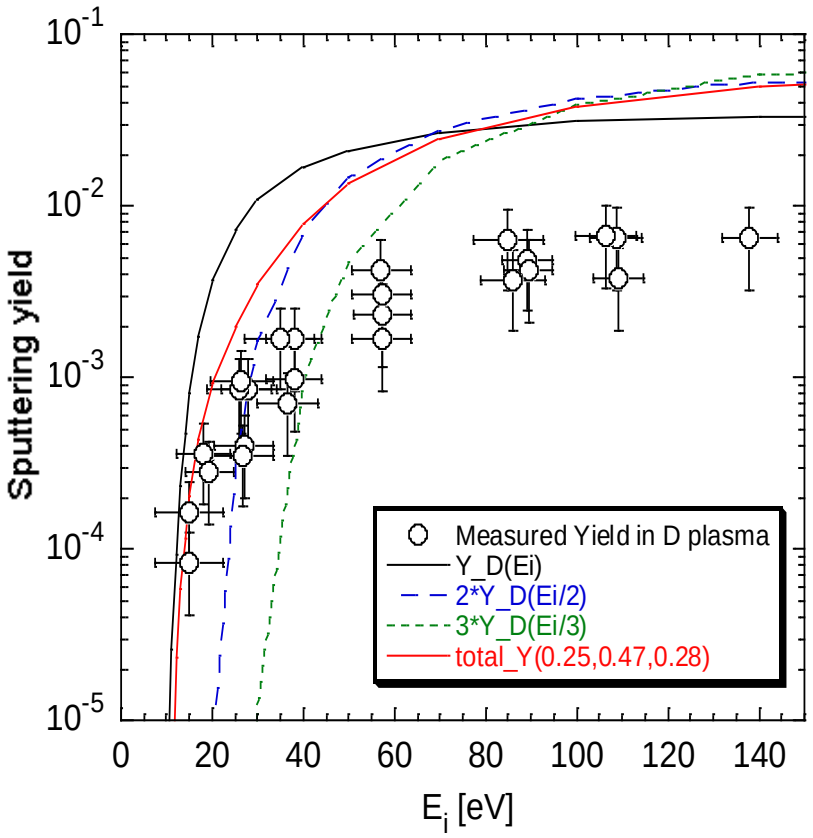
Figure 5. SEM images of the surfaces at 1/10 (a) 1/8 (b) and 3/6 (c). All surfaces show elongated structures that are pointing in a direction 15-20° downwards, with respect to the toroidal direction. The region (c) is slightly depressed with respect to the surrounding layer and has about 4 µm Be deposited, so it seems likely that the cone like structure consists mainly of Be.



D (& He) plasma erosion rates consistently lower than TRIM, Ar plasma sputter data agrees with TRIM

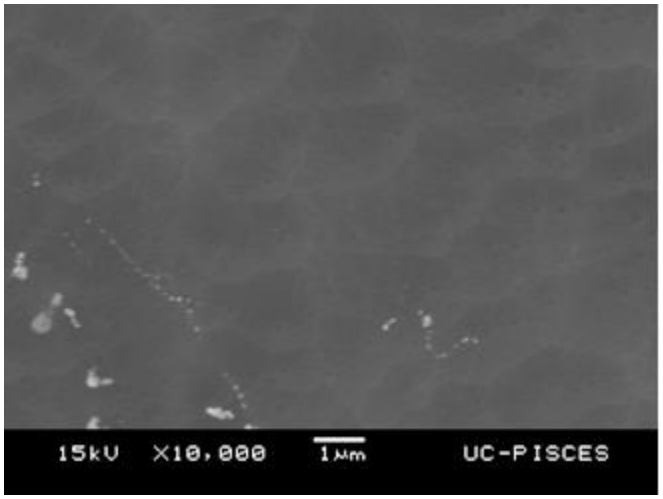


D on Be
[from Doerner et al., PSI20]

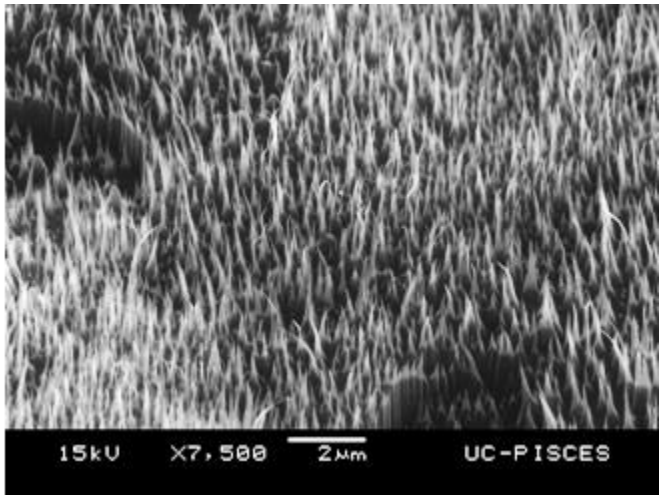


Distinct surface morphology differences exist after high and low mass sputtering of a Be target

After Ar plasma sputtering (~75 eV)



After D plasma sputtering (~80 eV)

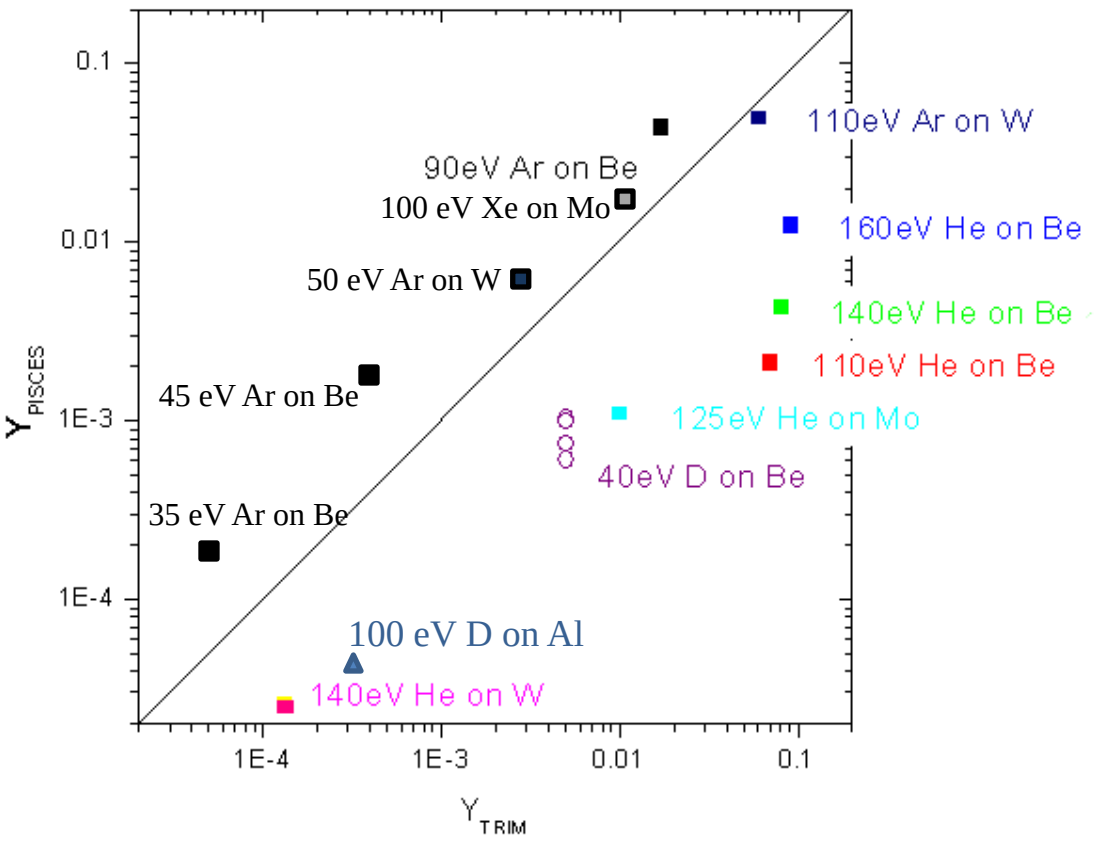


- Smooth Be surface exists after Ar plasma sputtering
- No significant decrease in Be erosion with fluence during Ar sputtering

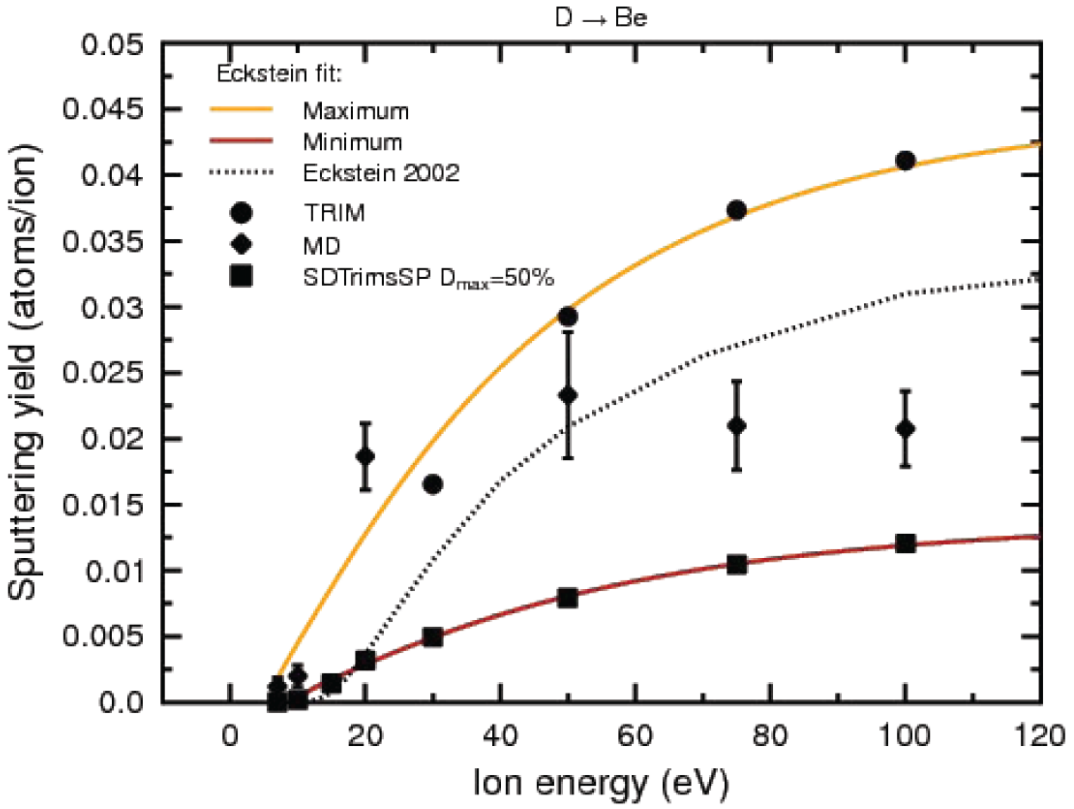
Low-Z vs. high-Z sputtering discrepancy is seen from a variety of metals exposed to plasma

PISCES

- Reason for this behavior is not understood
- Fuel atoms within the stopping distance of surface may influence sputtering/reflection
- Effect is measured for a variety of substrate materials
  - PISCES (Be, Fe, Mo, W, Al)
  - Pilot (Sn)
  - TPE (Be)



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)

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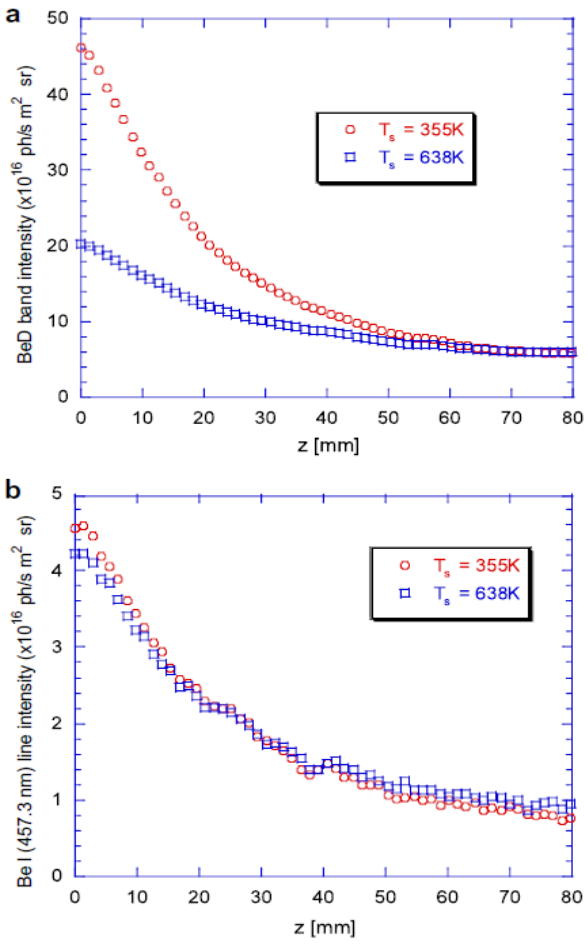
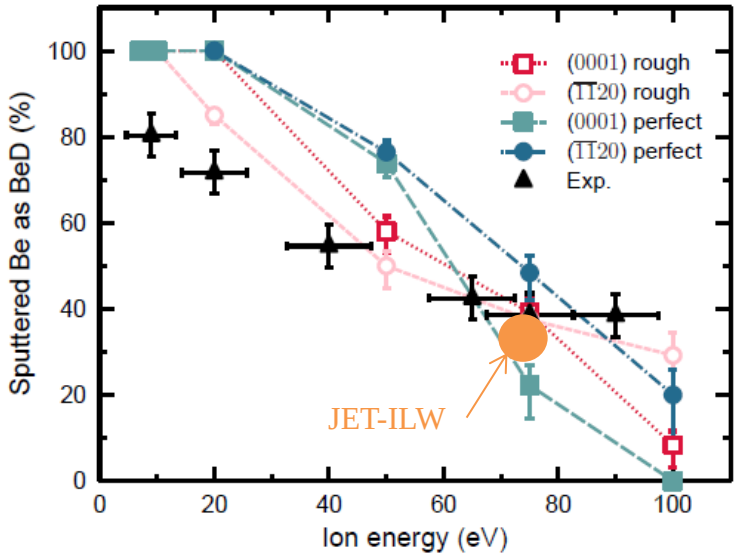
Chemically-assisted physical sputtering of BeD is temperature dependent (influence of He is being studied)



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.

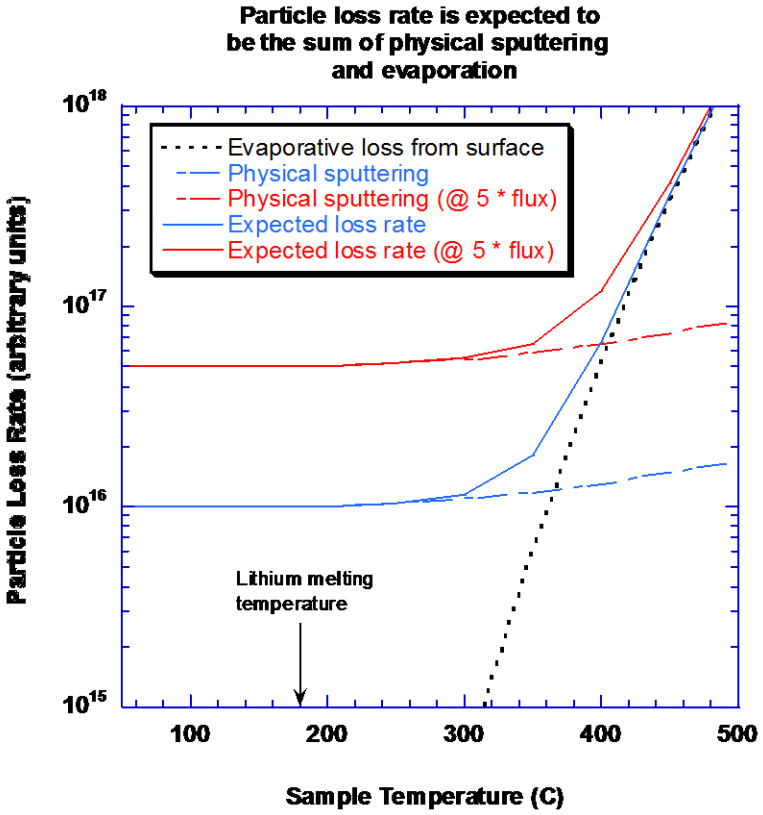
MD simulations of D on Be predicted subsequent erosion measurements

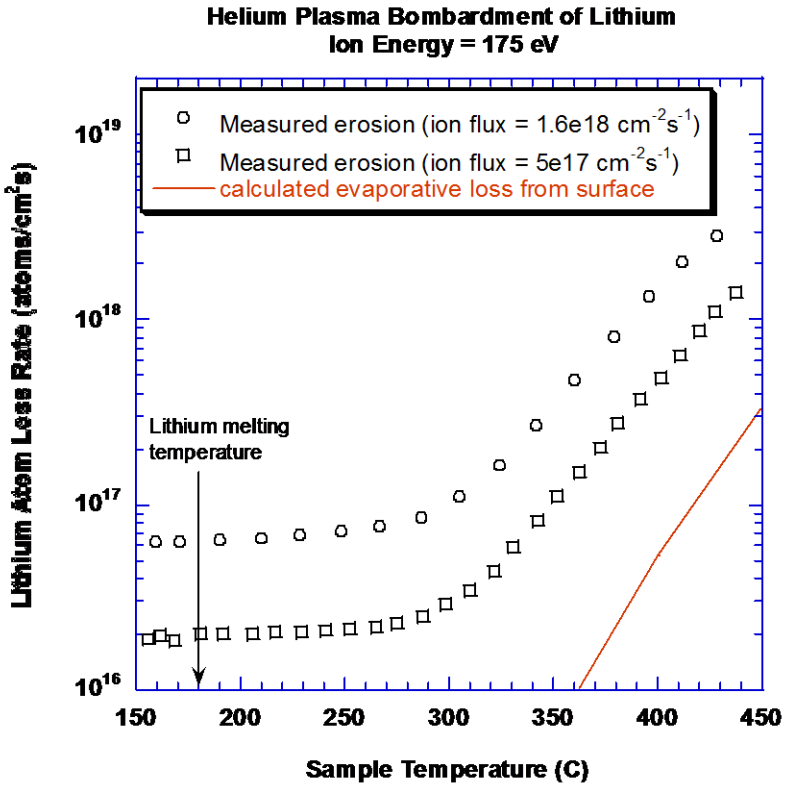


Exp. from D. Nishijima et al, PPCF 50(2008)125007.
Sim. from C. Bjorkas et al., New J. Physics (2009).

Temperature Dependent Sputtering :

PISCES experiments show qualitative agreement with expectations, but quantitatively show enhanced erosion at high temperature.

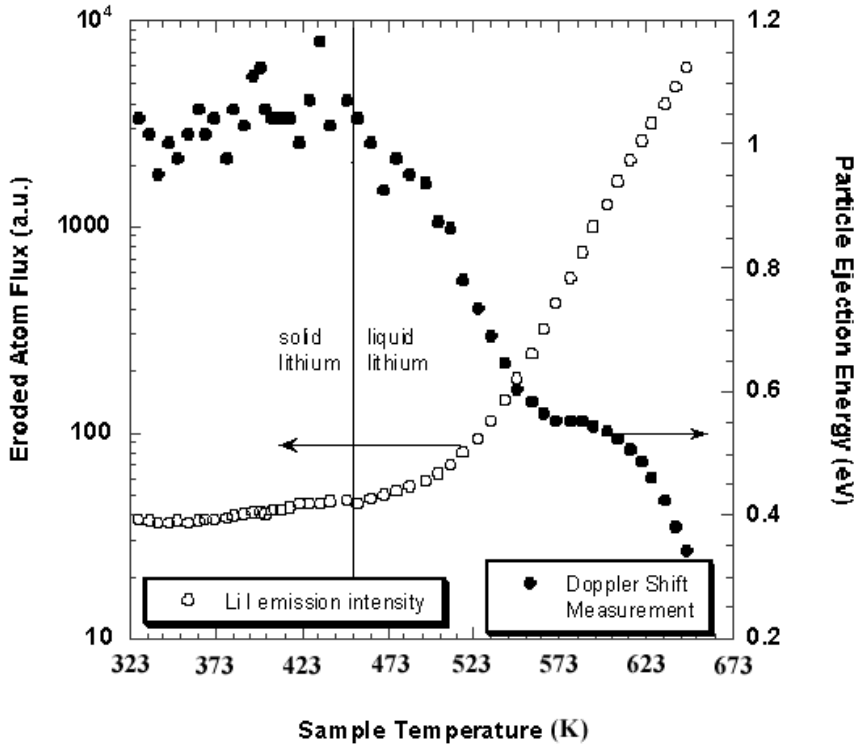




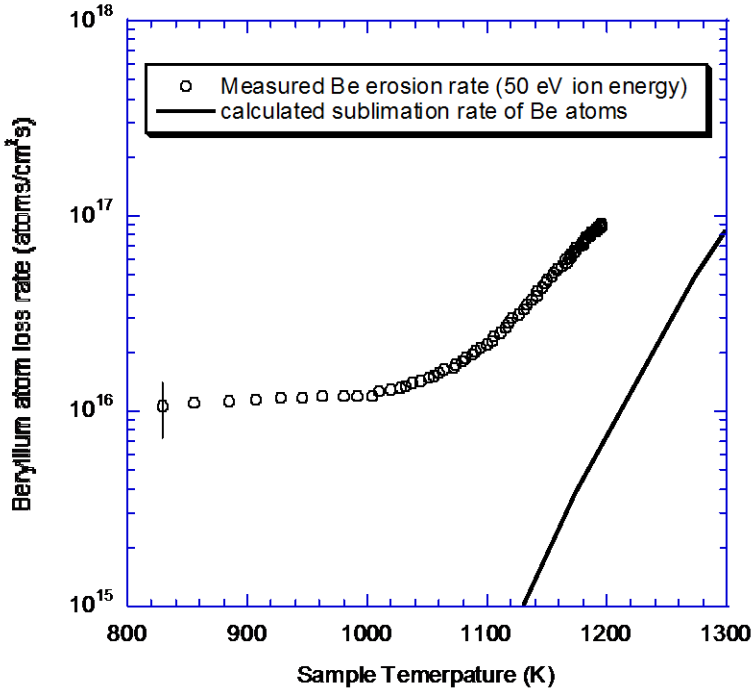
Temperature Dependent Sputtering :

Decrease in average ejection energy of particles accompanies the increase in erosion rate, implying enhanced evaporation. Also true in solids.

PISCES



Solid Beryllium Samples also Exhibit Enhanced Erosion at High Temperature



Energetic particle bombardment of surfaces results not only in sputtering, but also in the creation of surface adatoms.

- Adatoms are less strongly bound to the surface and can sublimate at lower temperature, before recombining at surface defect sites [R. P. Doerner et al., J. Appl. Phys. 95 (2004) 4471.]

The total atom loss rate from a surface is written

$$J_{total}(T) = J_{pl}Y_{ps} + J_o(T) + J_{ad}(T)$$

where, $J_{pl}Y_{ps}$ is the sputtered particle flux

J_o is the flux of lattice atoms sublimated $\quad J_o = K_o n_o \exp^{(-E_o/T)}$

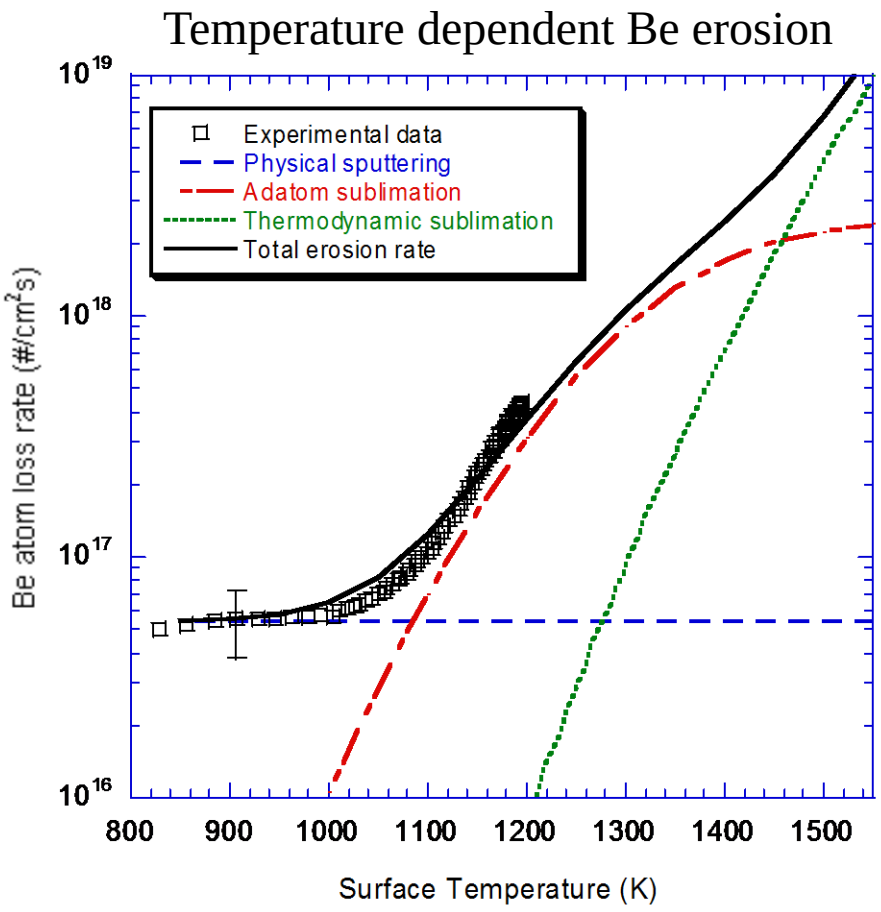
J_{ad} is the adatom sublimation flux $\quad J_{ad} = n_{ad} / t_{sub}$

And the areal density of adatoms is

$$n_{ad} = \frac{Y_{ad} J_{pl}}{\left(\frac{1}{t_{rec}} + \frac{1}{t_{sub}}\right)}$$

Three terms of the total loss rate can be plotted.

$$J_{total}(T) = J_{pl}Y_{ps} + J_o(T) + J_{ad}(T)$$



- Adatom loss term saturates as the fraction sublimated approaches the number created
- At higher temperature the total loss rate is dominated by 'normal' thermal sublimation
- The temperature range where different terms dominate depends on experimental conditions

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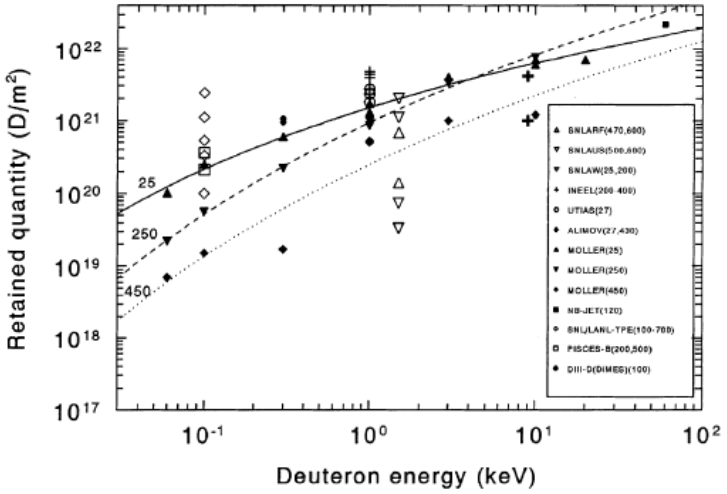
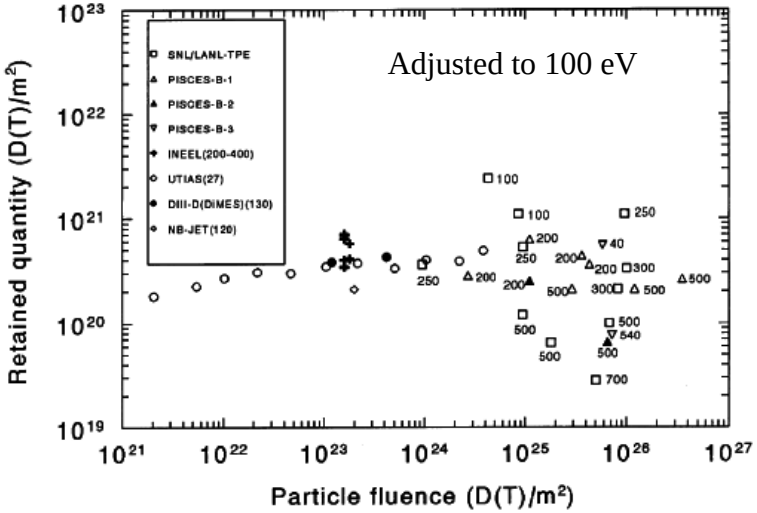
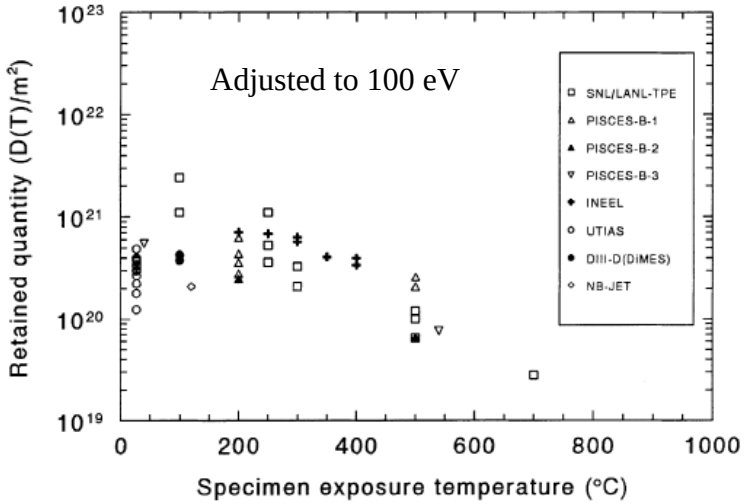
Anderl et al.[JNM 273(1999)1] reviewed D retention in Be



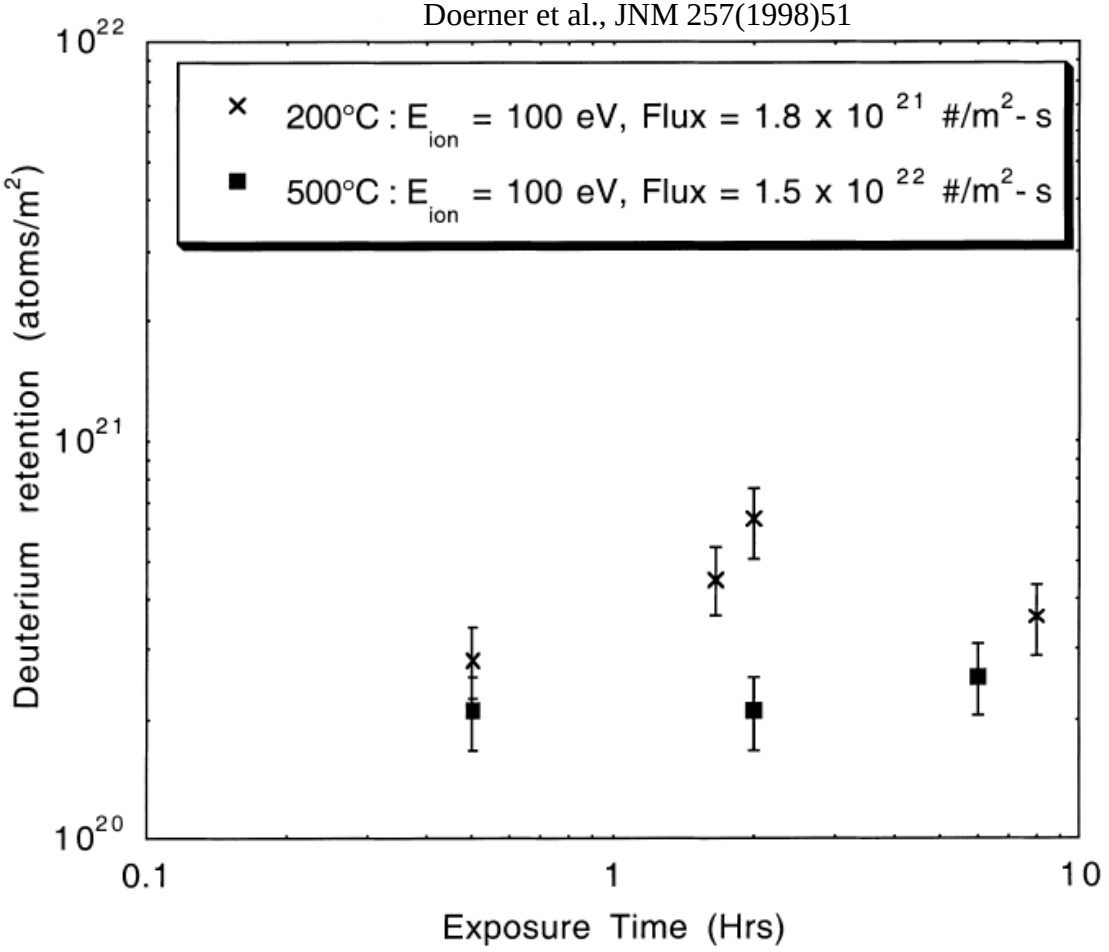


This review is still considered the definitive publication on D retention in Be.



At elevated temperatures, there is little D retention dependence on plasma fluence

Doerner et al., JNM 257(1998)51



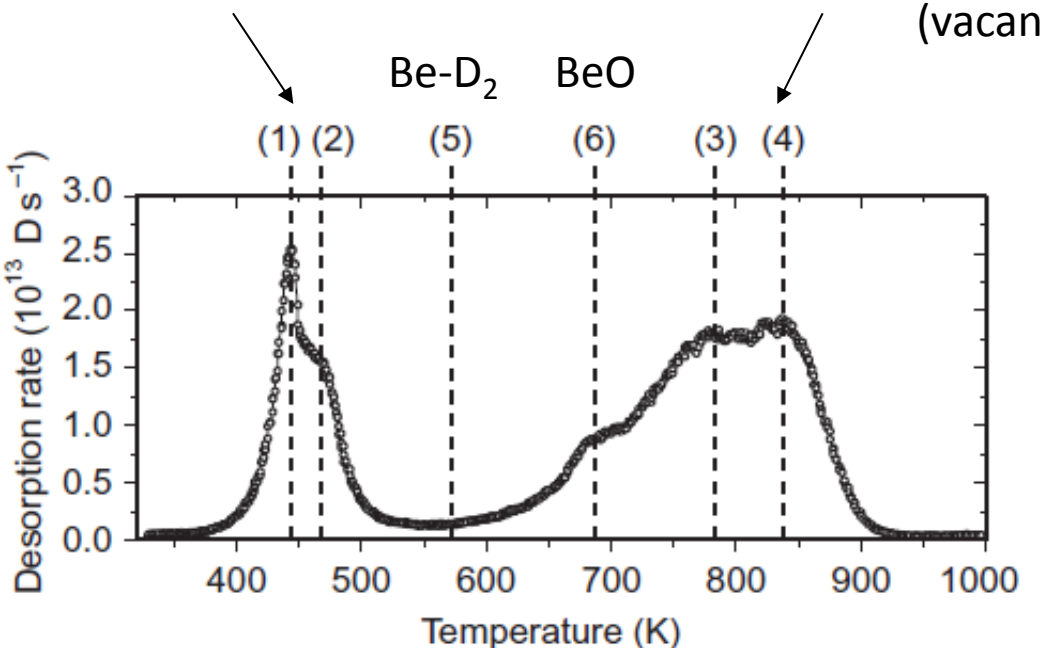
D retention in clean Be (UHV subML O coverage)

Reinelt et al. New Journal of Physics 11 (2009)
Oberkofler et al. Nuc.l Instrum. Methods 267 (4) (2009)

low flux/fluence implantation at 0.3-3keV with 10^{15}D cm^{-2} s^{-1}:

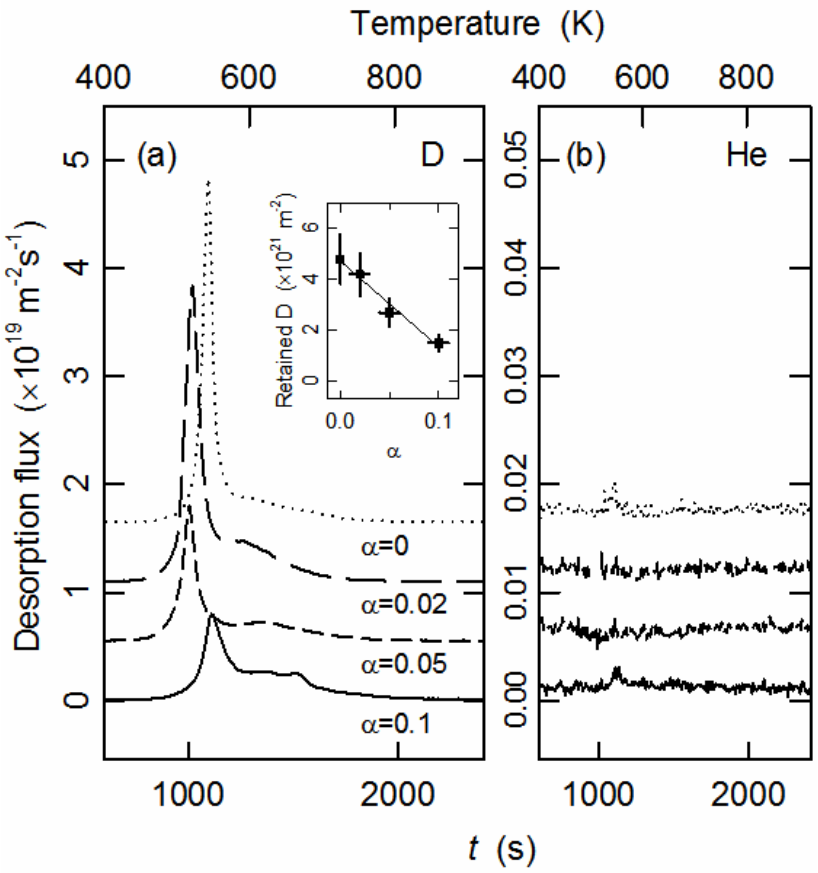
structural modifications, supersaturated
(D_2 in bubbles / amorphous Be-D, D/Be up to 1)

defects created by the implantation cascade (vacancies)

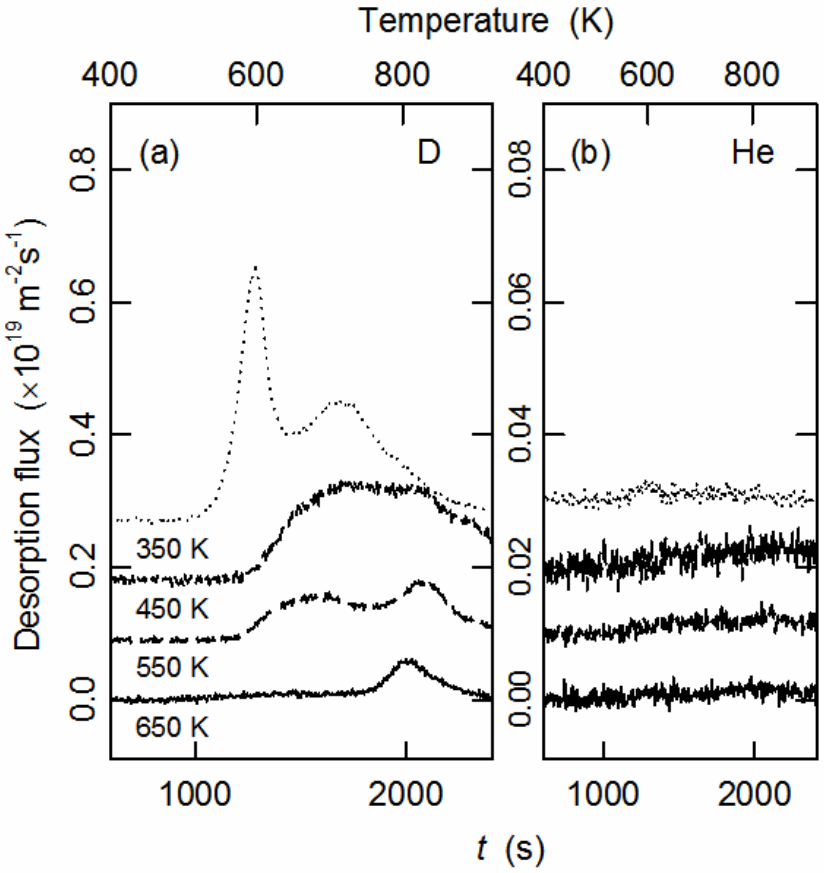


Only low temperature release peak (~500K) is affected by mixed D/αHe plasma

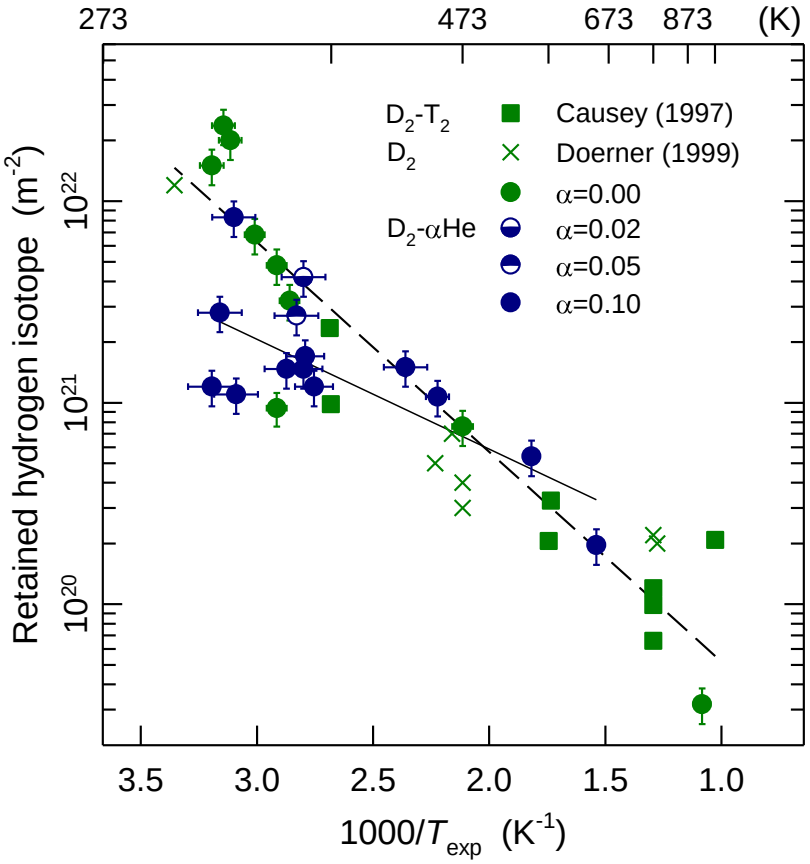
Sample temperature ~330K, F= 1e26 m^{-2}

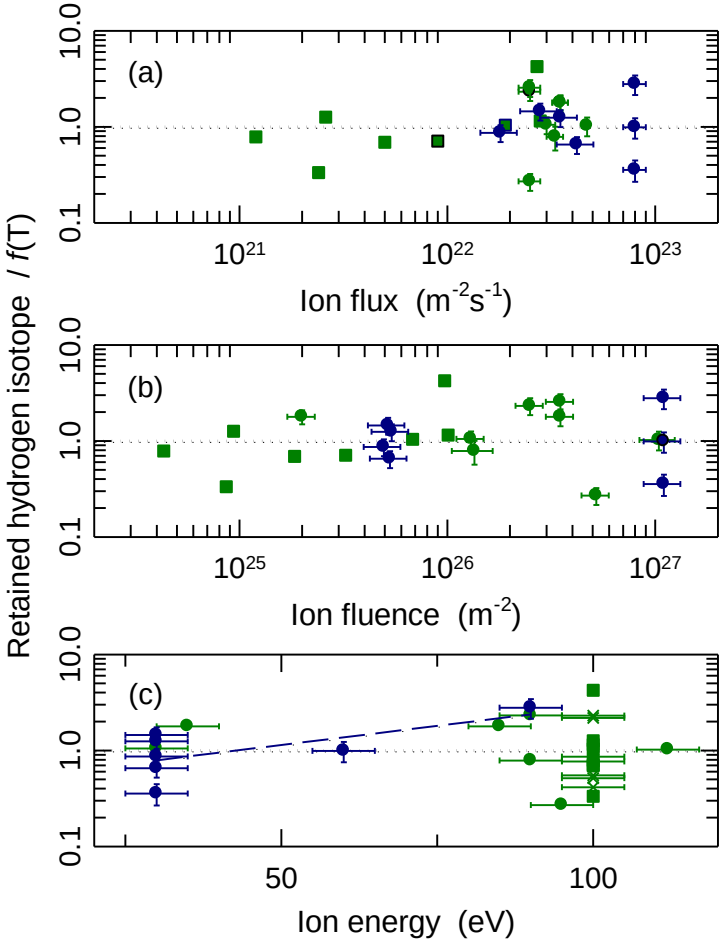


α=0.1, F= 5e25 m^{-2}

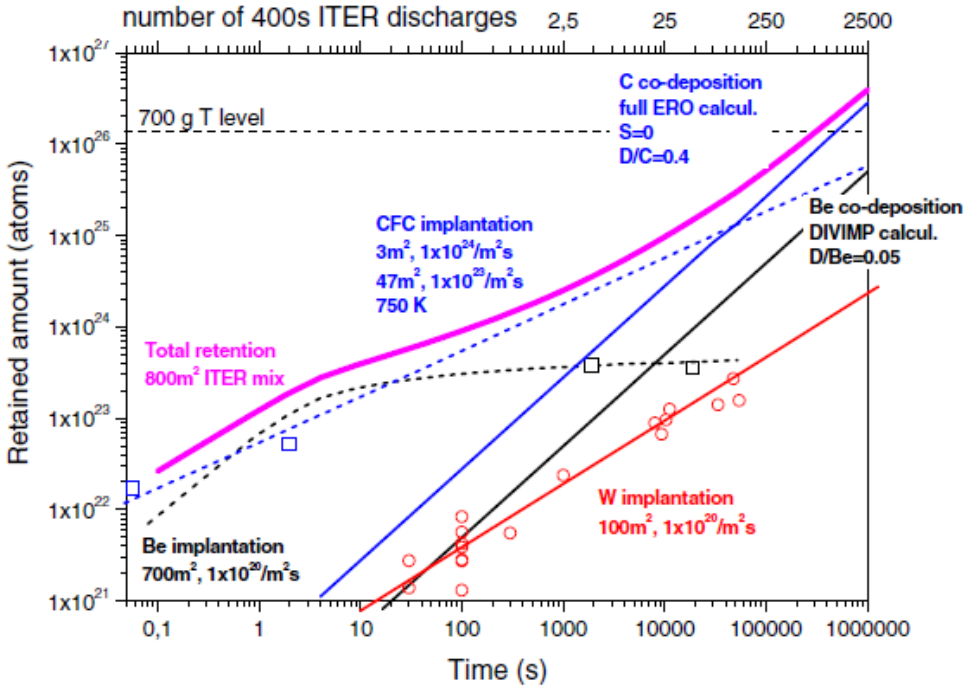


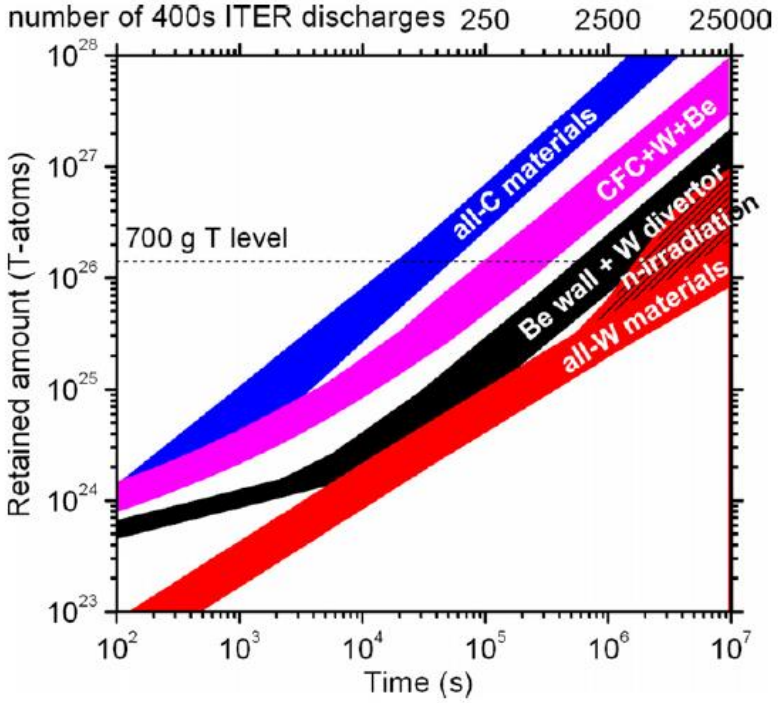
Influence of D/He plasma on D retention shows affects only at low exposure temperature





Retention in Be codeposits will dominate inventory in ITER

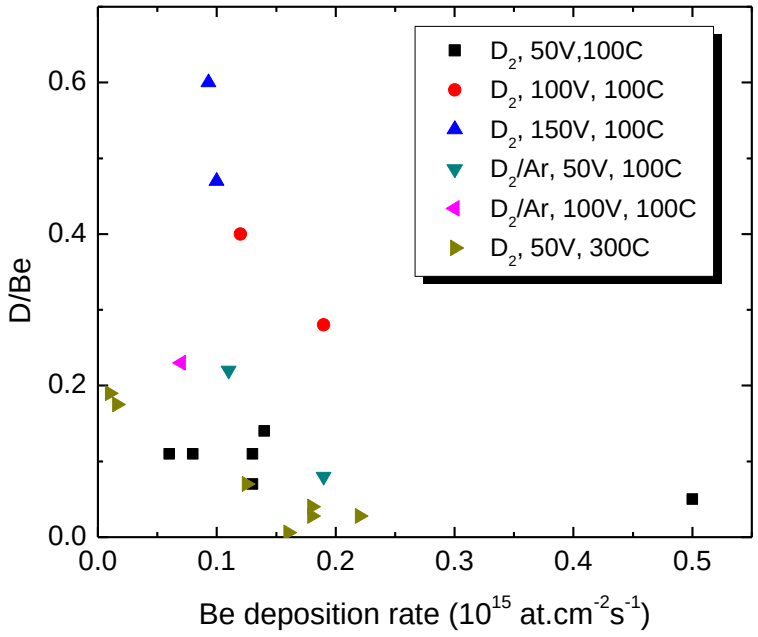


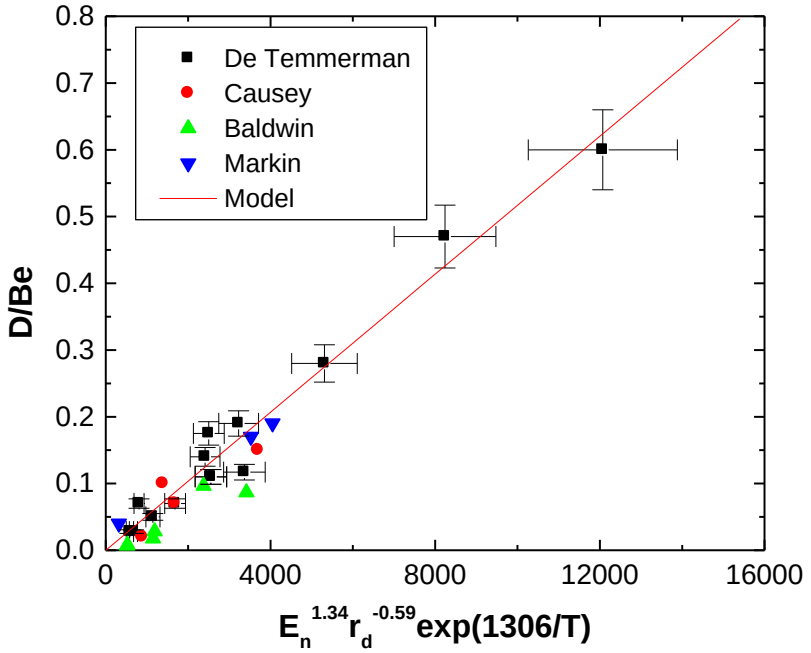


[from Roth et al., PPCF 50(2008)103001]

Retention in codeposited Be can be predicted based on codeposition conditions

$$D/Be = (5.82 \times 10^{-5}) E_n^{1.17 \pm 0.2} (\Gamma_D/\Gamma_{Be})^{-0.21 \pm 0.1} \exp^{(2273 \pm 311/T_C)}$$





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

Summary of Be PMI R&D in PISCES

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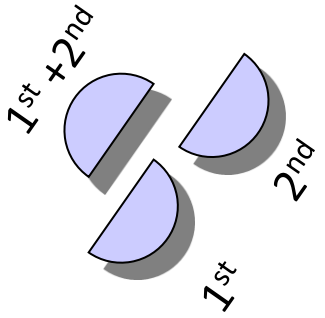
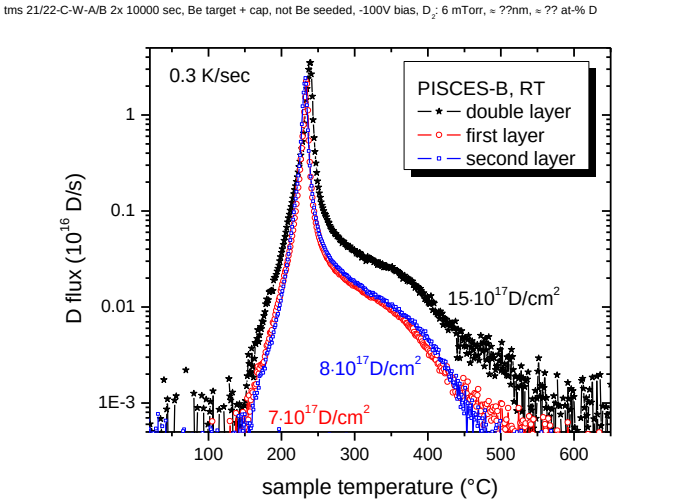
- Be mixed-material PMI studies
  - Be/W layers
  - Response to transient heating events
- Discussion

Release of D from Be codeposits : passivating oxide layer at interface has little impact on release behavior

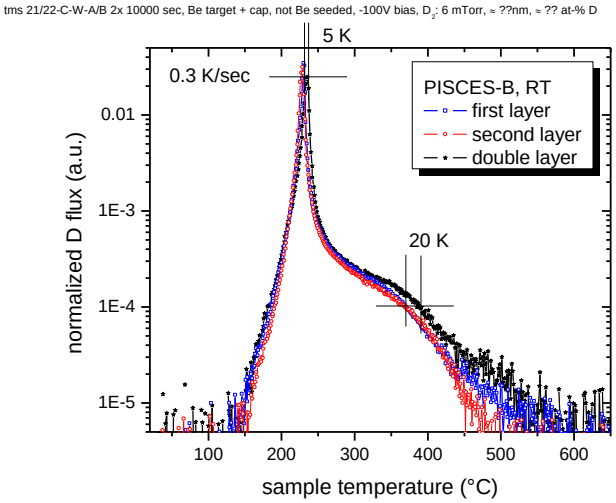
- Double layer codeposit release agrees with sum of two single layers (thickness ~ 100 nm) collected on PISCES-B witness plates
- Slight increase in release temperature with increasing codeposit thickness
- Difficult to model due to small differences in each codeposit



D release from single and double layer, water cooled codeposits

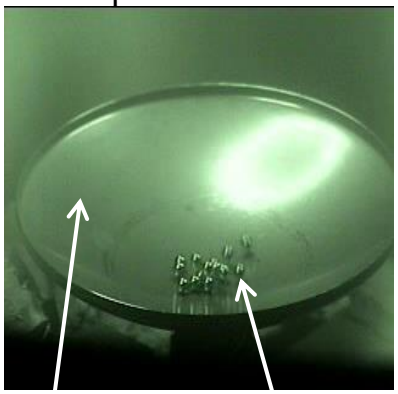


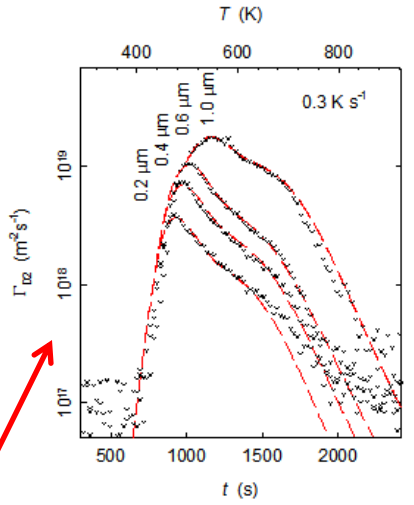
D release from single and double layer, water cooled codeposits

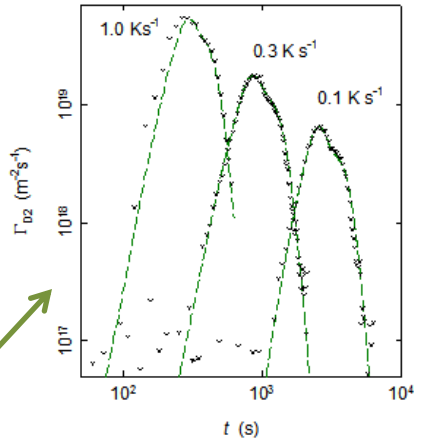


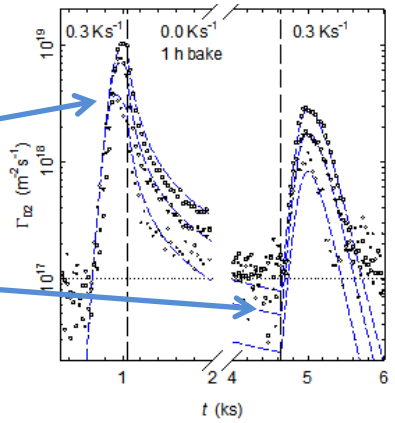
A single TMAP model describing Be-D co-deposit release agrees with experiments over a wide range of parameter space

3 Be sputter guns coat spheres with Be/D



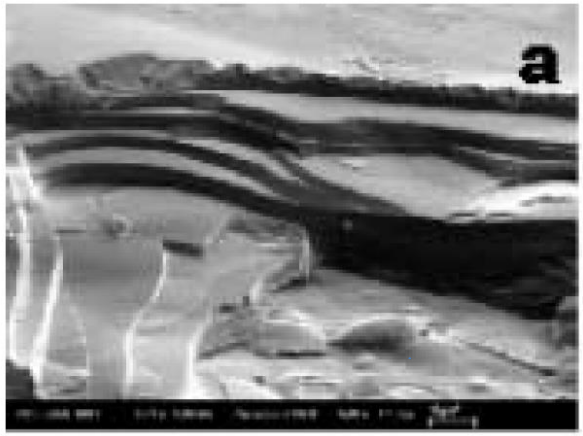




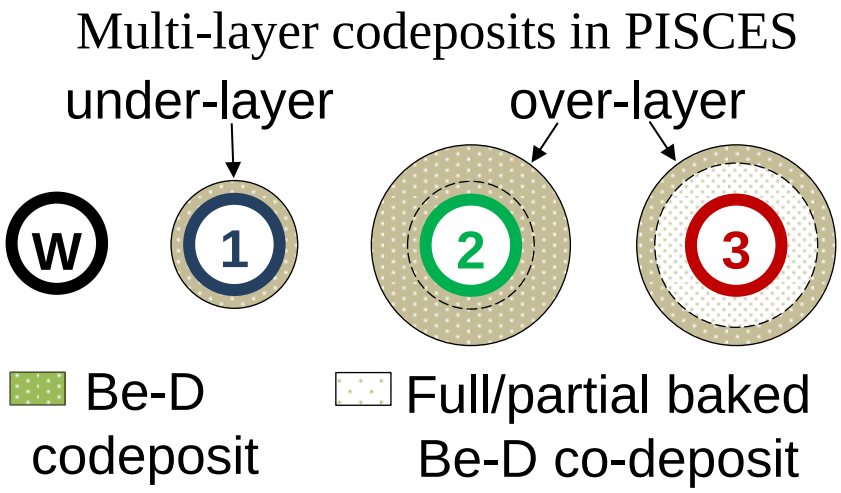


- Identical Be-D co-deposits on W spheres is ideal for modeling D release
  - Reproduces shift in release to higher temperature with increasing layer thickness (TMAP red)
  - Reproduces change in release during differing heating rate bakes (TMAP green)
  - Reproduces release decrease during temperature holds
  - Reproduces release behavior during subsequent temperature increase after hold (TMAP blue)

ITER will likely encounter depleted codeposit under layers covered by saturated codeposit over layers

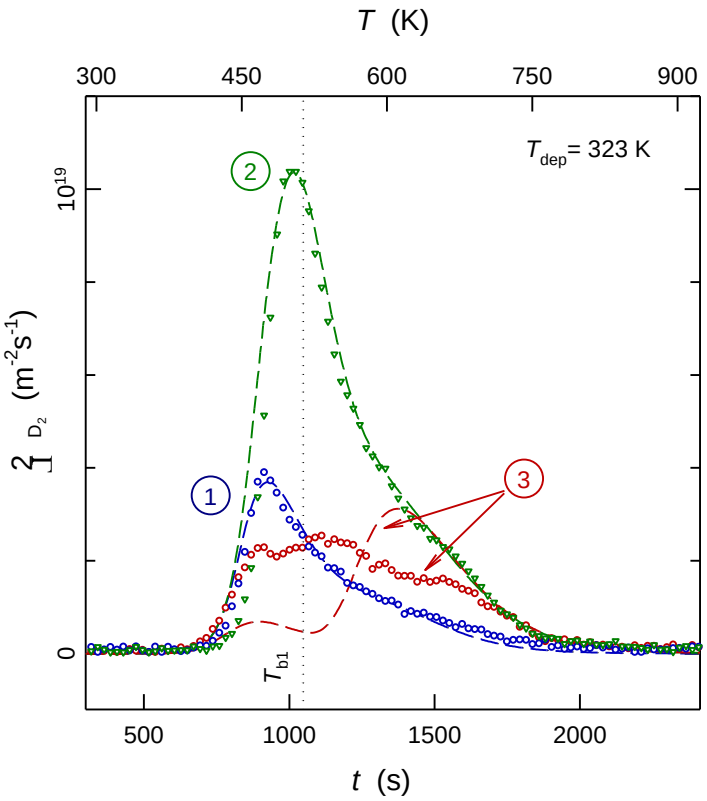


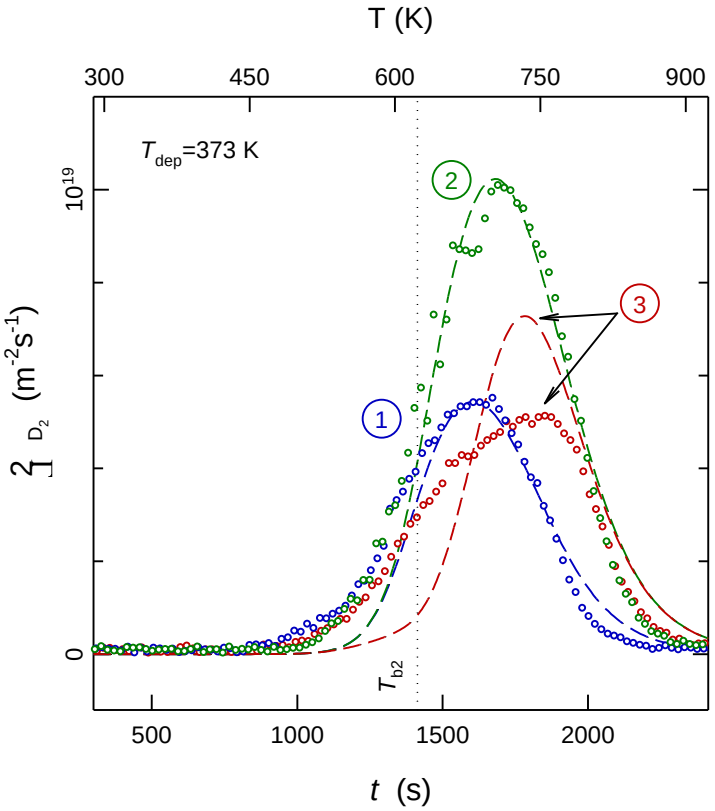
Multi-layered co-deposit in TEXTOR
from M. Rubel et al., Phys. Scr. T103(2003)20.



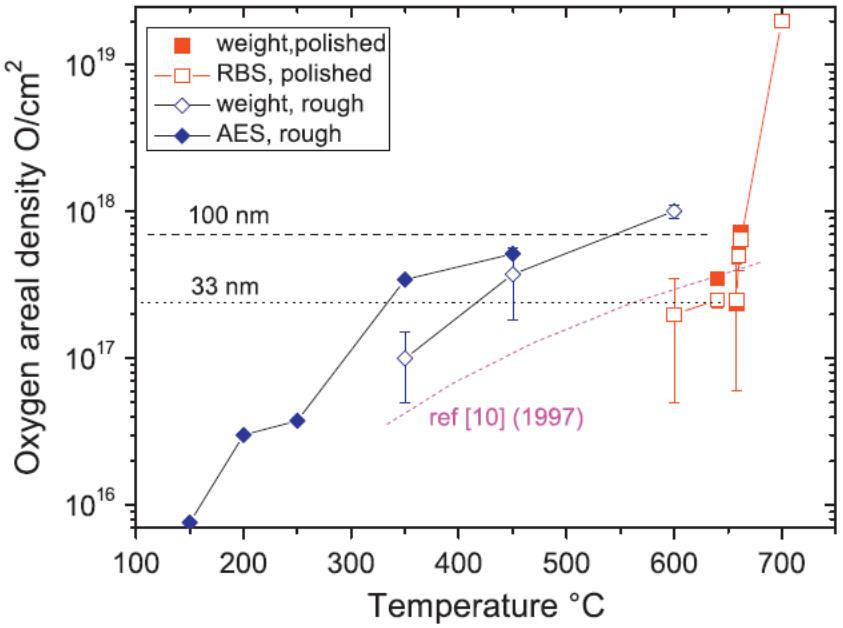
- Periodic baking of ITER will be used to control in-vessel tritium accumulation (depleting codeposits) [definition – under layers]
- Subsequent operation will lay down new, saturated Be-D/T codeposits [definition – over layers]
- Under layers could be fully, or partially, depleted of D/T
- How does depleted under layer affect D/T release from over layer, or does T diffuse and retrap in previously depleted trap sites?

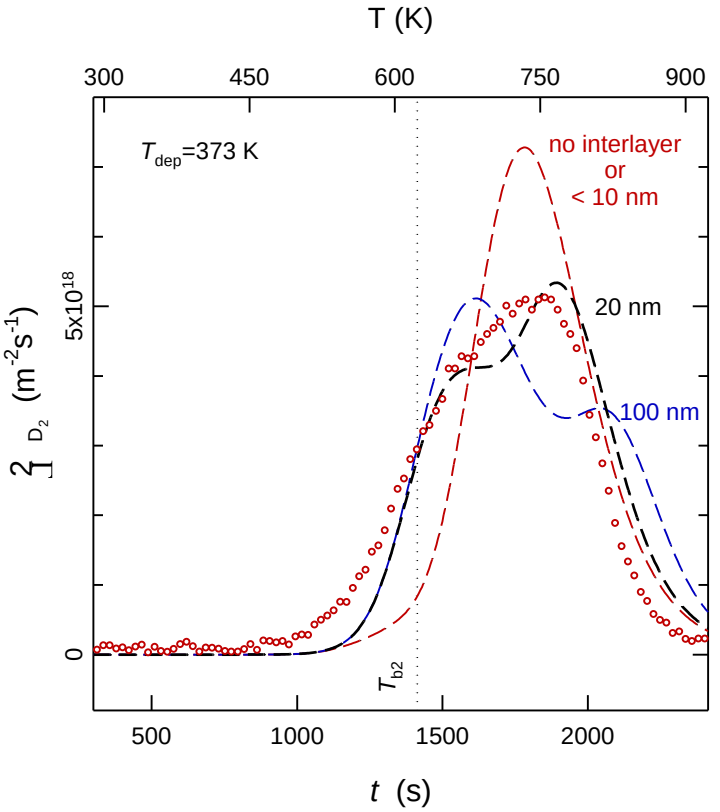
Agreement between the standard TMAP model and experiment is no longer achieved





Formation and growth of an oxide layer during baking has a profound effect on D release behavior





- Oxide thickness on Be after 6 hours of baking in atmosphere @350°C : 10 ~ 60 nm

$$D_{iii} = 1.3 \times 10^{-9} \exp\left(-0.62[\mathrm{eV}]/\mathrm{k}T\right)\ [\mathrm{m}^2\mathrm{s}^{-1}]$$

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- Be mixed-material PMI studies
  - Be/W layers
  - Response to transient heating events
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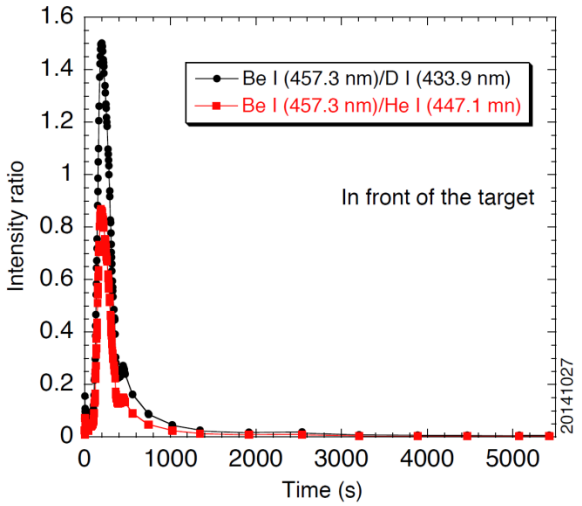
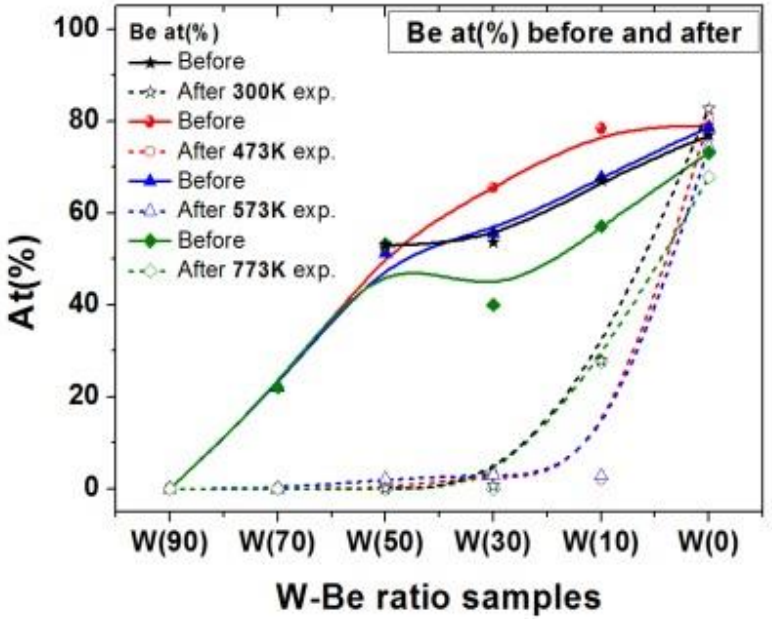
Preferential loss of Be from the surface of mixed Be-W targets (from Romania) occurs during D and D/He plasma exposure

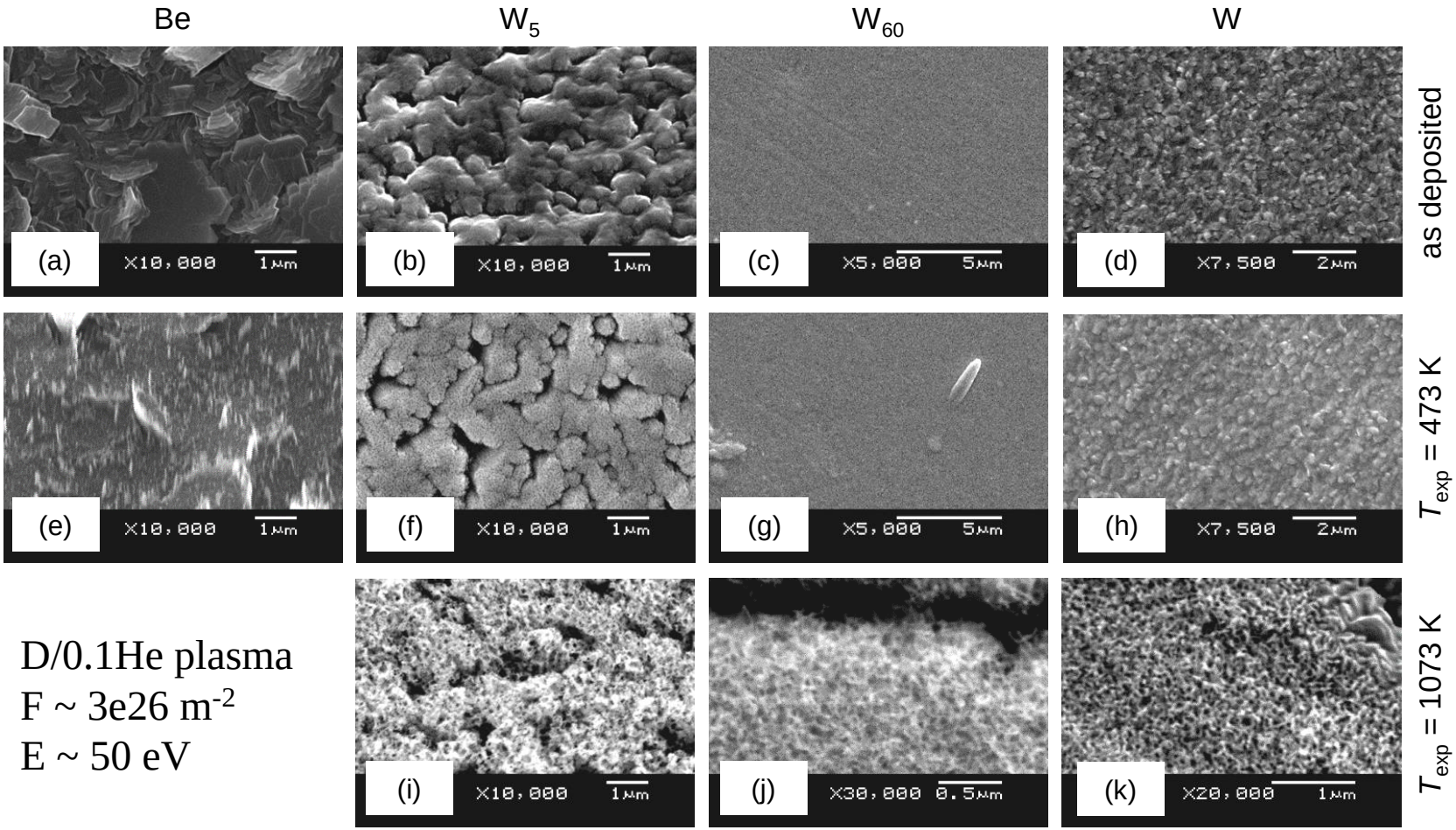
- Samples prepared by thermo-ionic arc (TVA) process in Romania
- Ion energy is above Be sputtering threshold, but below W sputtering threshold
- Be I line emission quickly decreases from exposed targets
- W-rich surfaces result after D or D/He plasma exposure regardless of initial surface composition
- Similar to Fe/W behavior in RAFM steels





Loss of surface Be results in W fuzz growth at high temperature

PISCES

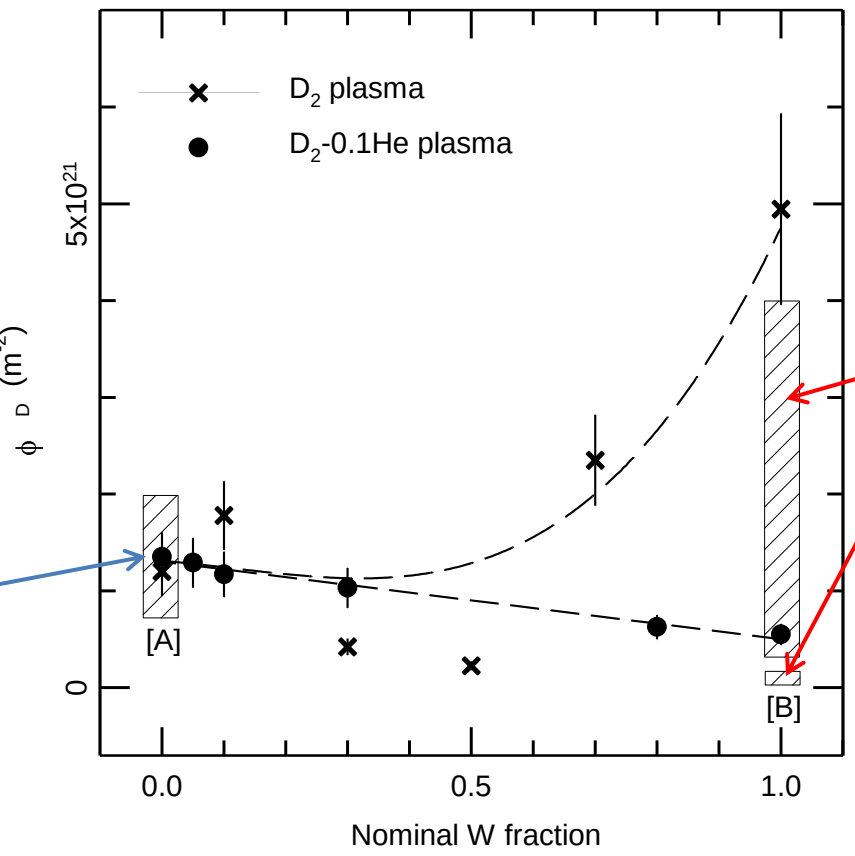


D/0.1He plasma
F ~ 3e26 m^{-2}
E ~ 50 eV

[from I. Jepu et al., submitted to JNM 2016]

D retention in Be-rich TVA layers follows that of bulk Be, whereas W-rich TVA layers follows that of bulk W

- D retention measured by TDS after 473 K exposure
- No measureable D desorbed after 1073 K exposure



Retention in bulk Be after D, or D/0.1He, plasma exposure [Baldwin PSI 2016]

Retention in bulk W after D, or D/0.1He, plasma exposure [Baldwin NF 51(2011)103021]

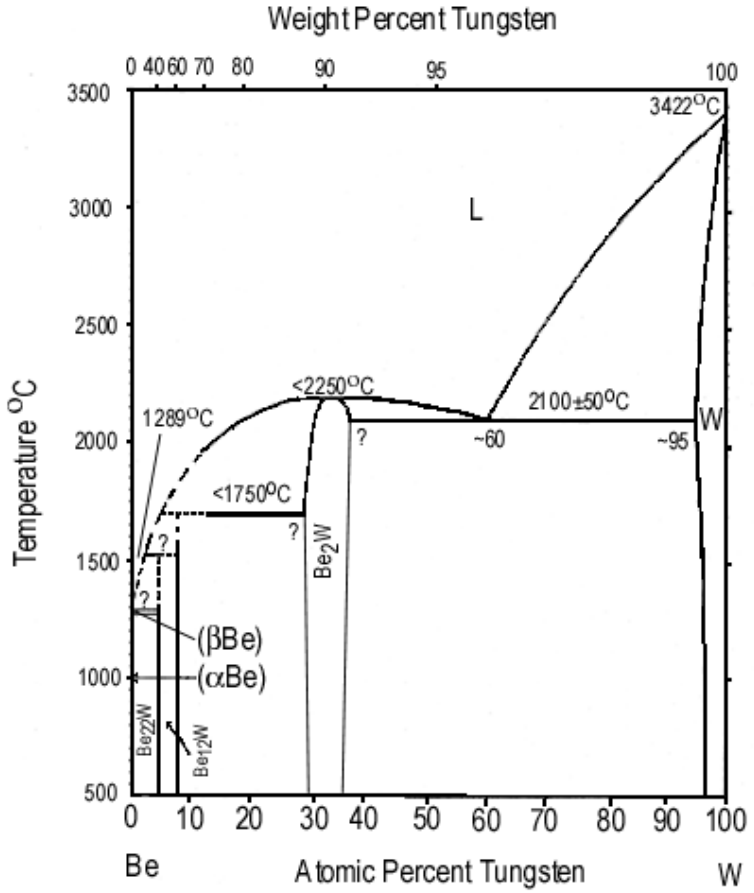
[from I. Jepu et al., submitted to JNM 2016]

Understanding pre-created Be/W surface layers is important, but plasma-created Be/W layers will also form when the incident plasma on W contains Be.

In PISCES-B, this can be investigated by using the Be oven to seed controlled amounts of be impurities into the plasma column.

Two cases are possible, when the incident flux of Be is less than the erosion rate of Be from the surface, or when the incident flux is greater than erosion.

What will be the result of Be layers on plasma exposed W surfaces?



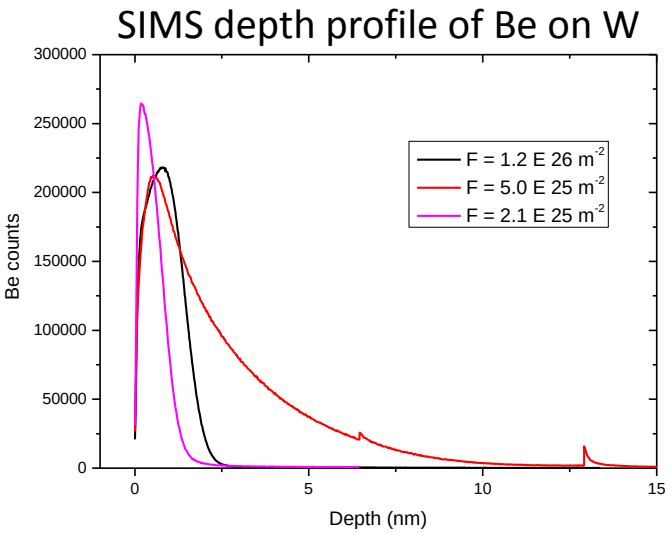
From H. Okamoto and L.E. Tanner, in "Phase Diagrams of Binary Tungsten Alloys", Ed. S.V. Naidu and P. Rao,Indian Institute of Metals, Calcutta, 1991.

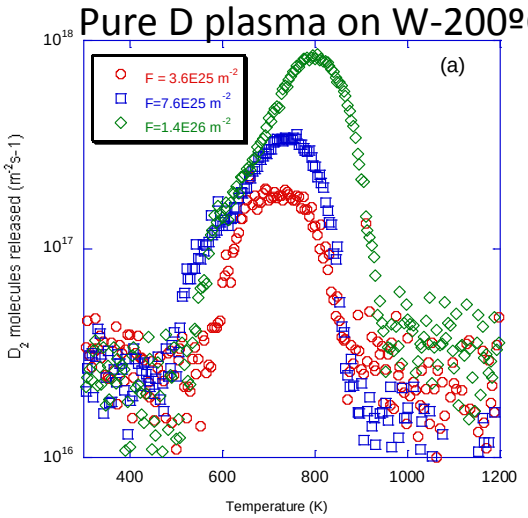
- Be can alloy with W
- Resulting alloys ($Be_{22}W$ and $Be_{12}W$) have much lower melting temperatures (~1500°C)
- Be_2W alloy also has somewhat lower melting temperature (<2250°C)

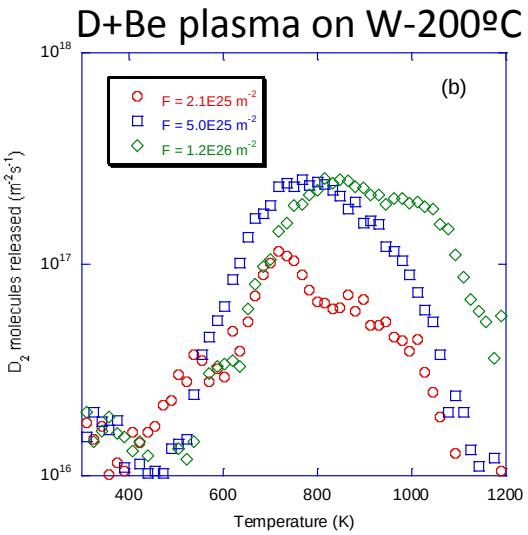
Be-containing plasma on W forms a thin Be_2W surface layer but does not drastically increase retention in W

PISCES

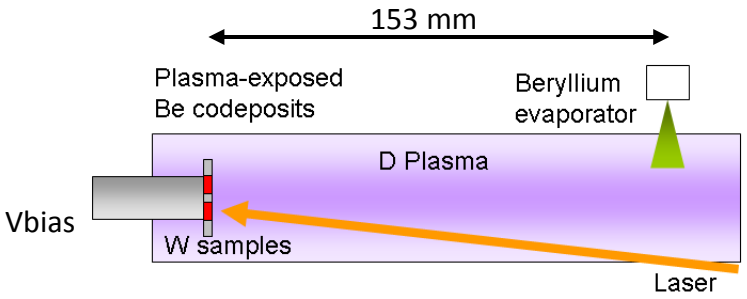
A thin (few nm) Be_2W layer forms on the surface of W exposed to D+Be plasma (when the Be flux is smaller than erosion), which does not change retention significantly, but may act to inhibit release of implanted D







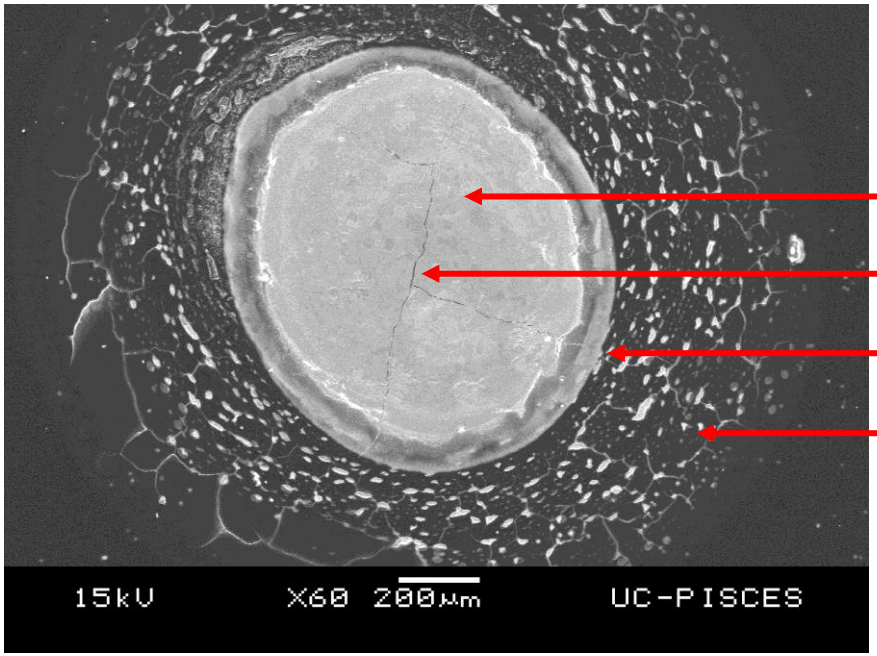
Laser transients on Be-coated W create a rich variety of morphology changes



Macroscopic Be layers grow on W targets when the Be arrival rate is greater than the erosion rate

Large type I ELM

70 MJ/m^2/s$^{1/2}$ D plasma, ~3 micron Be layer



Ablation of Be, recrystallization of W

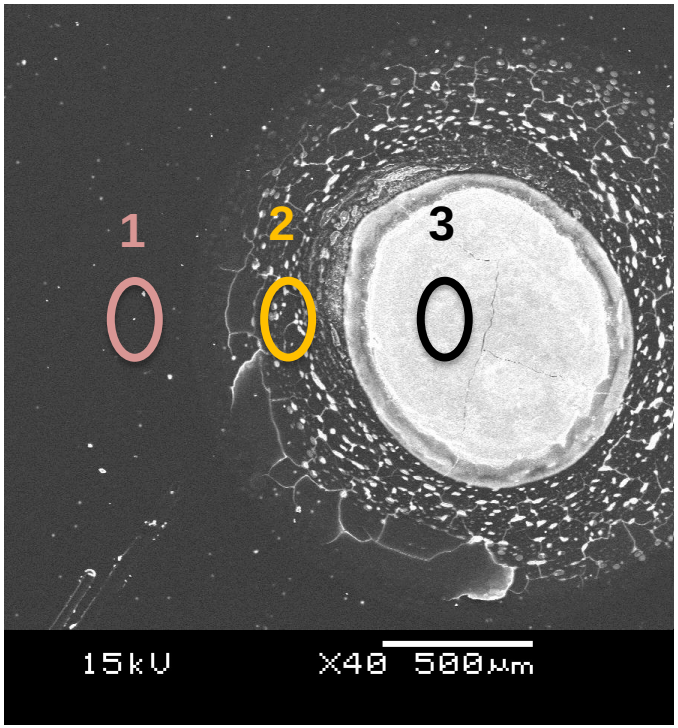
Be-W alloying and cracking

Be melt flow

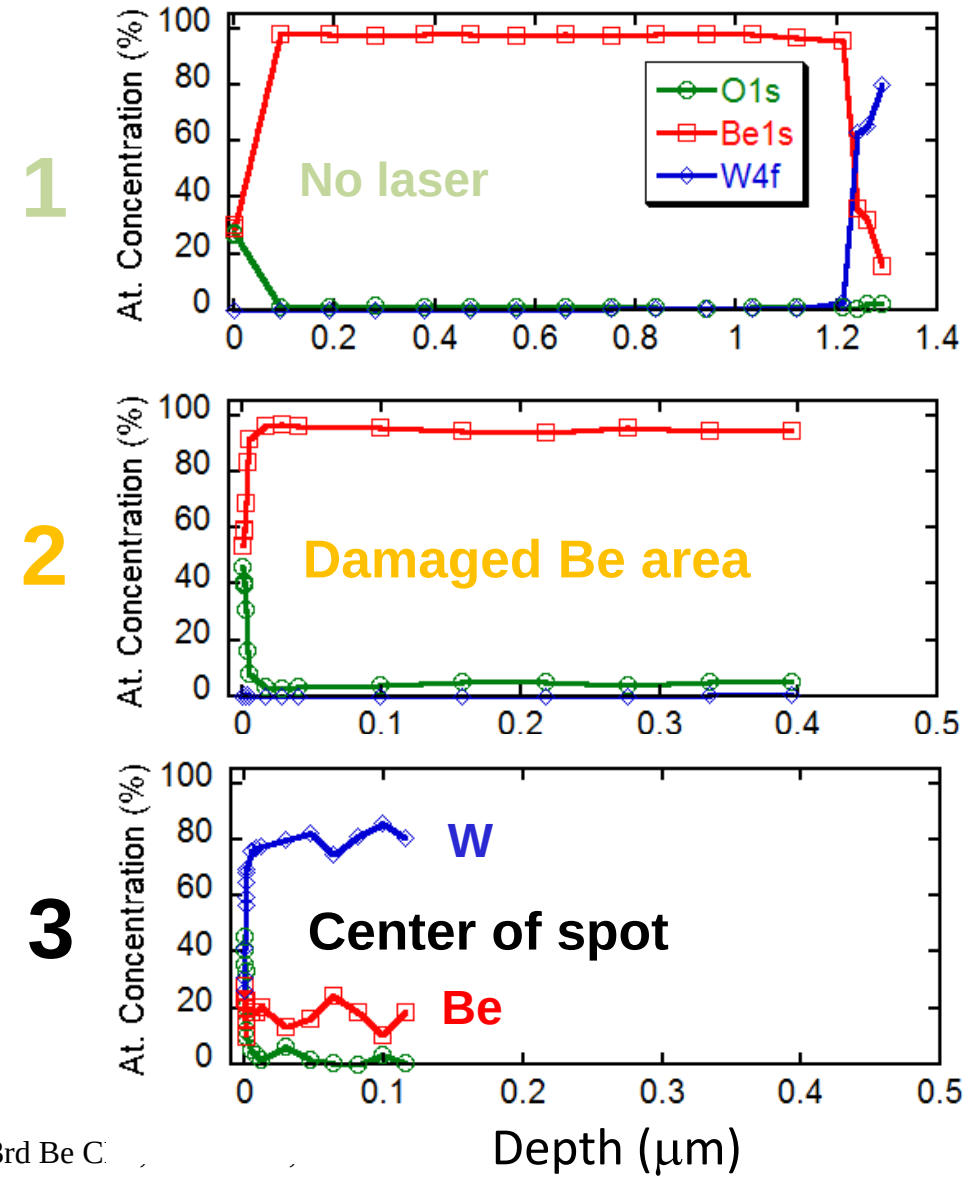
Delamination and holes in Be coating

Sputter XPS depth profiles show Be-W mixture at center of spot

70 MJ/m^2/s$^{1/2}$



T. Höschen



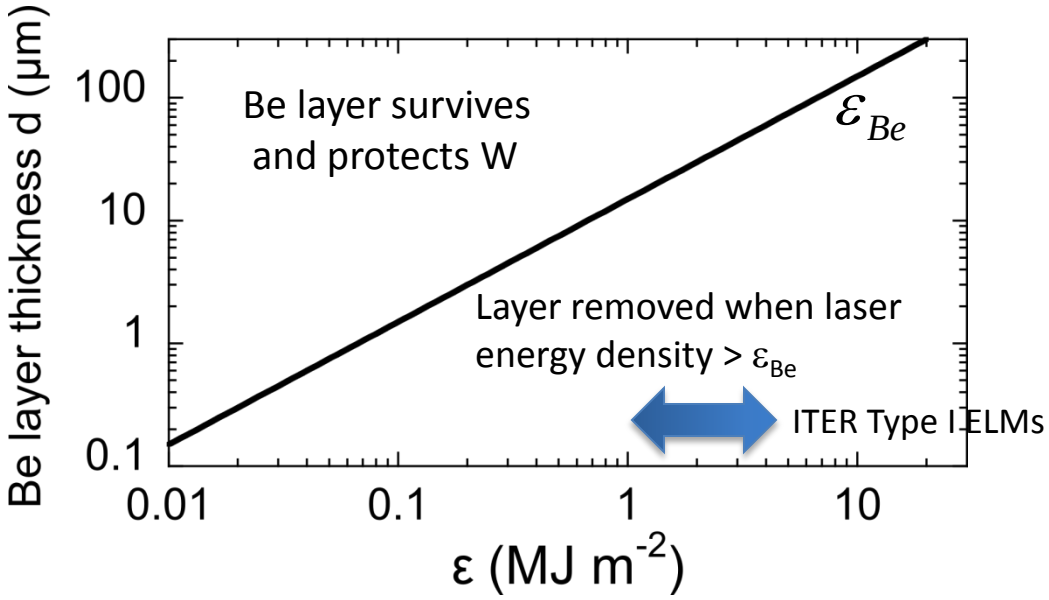
How much energy is needed to remove the Be coating?

Sacrificial Be layer

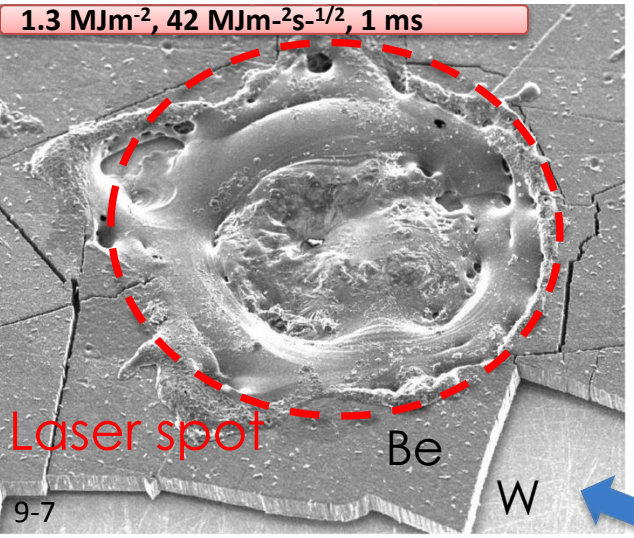
Can eroded wall material that is deposited on the divertor provide protection to the underlying W? How thick does Be layer need to be?

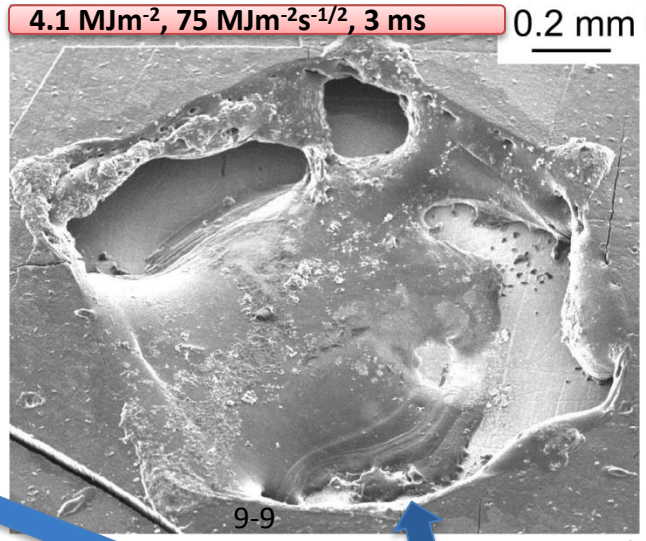
Energy density required to sublimate (or equivalently, melt and evaporate) a layer of Be with thickness *d* : $\varepsilon_{Be} = \Delta H \rho d / \mu$ "Be formation energy density"

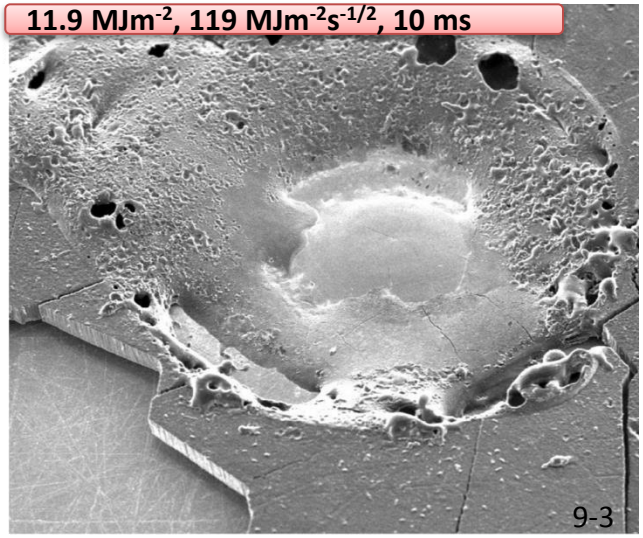
ρ is density, μ is molar mass, and enthalpy of formation for Be is ΔH = 324 kJ/mol (*CRC Handbook)*

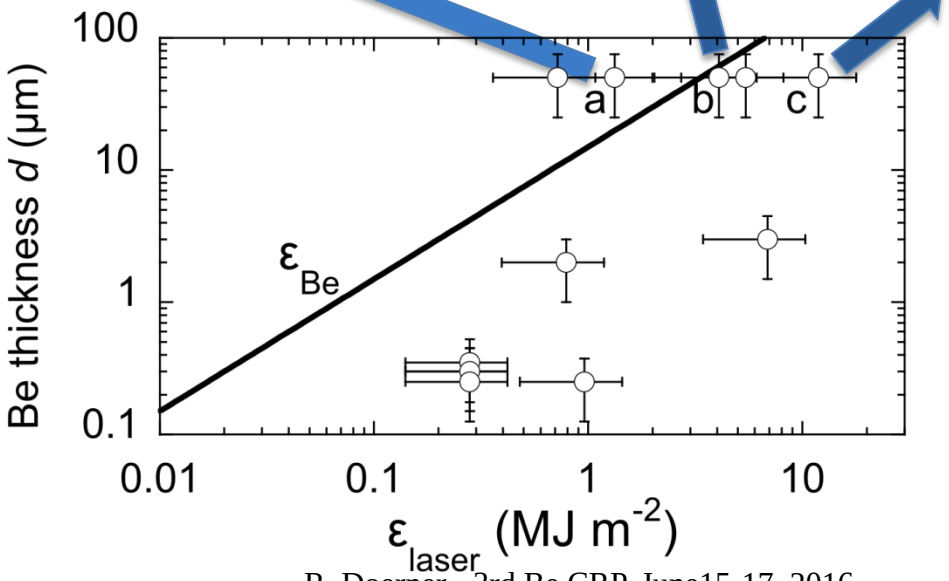


SEM images support simple estimate for Be layer removal









Be is removed by laser when the absorbed laser energy is greater than the enthalpy of formation of Be layer,

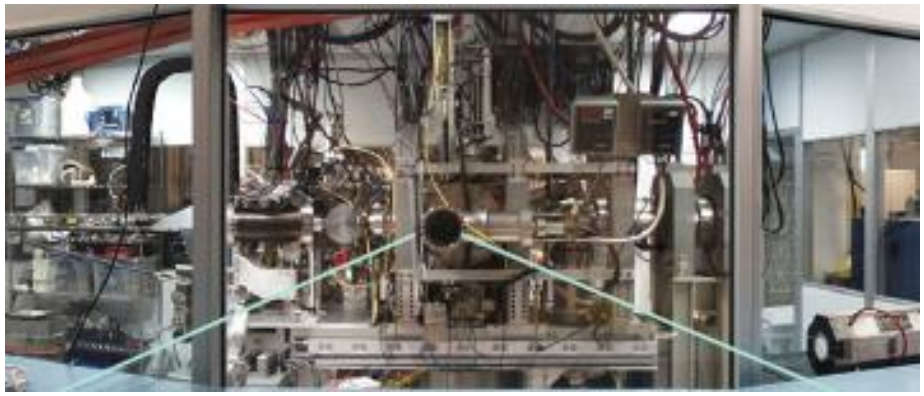
$\varepsilon_{laser} > \varepsilon_{Be}$

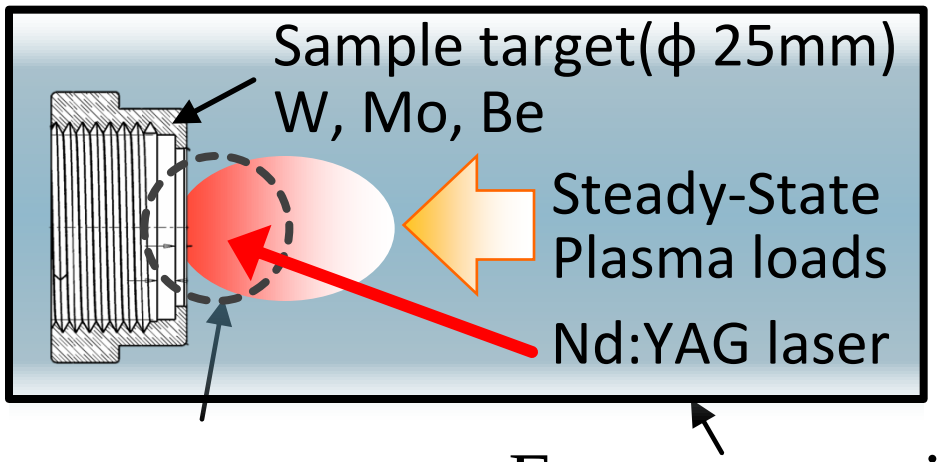
Plasma response to laser heating of Be targets differs from that of W, Mo targets

- Be, W and Mo targets were irradiated with 20 msec long laser pulses (max. energy 50 J) and the response of the plasma was observed
- Penetration distance (or decay length) of ablated material was measured along with background plasma line emission
- PIC simulations show Mo and W atoms are localized close to the targets due to their slow ejection velocity and higher ionization cross-section, whereas ablated Be penetrates more deeply into the plasma column causing cooling of the plasma

[from K. Ibano et al., PSI 2016]

Overview of experiments at PISCES-B





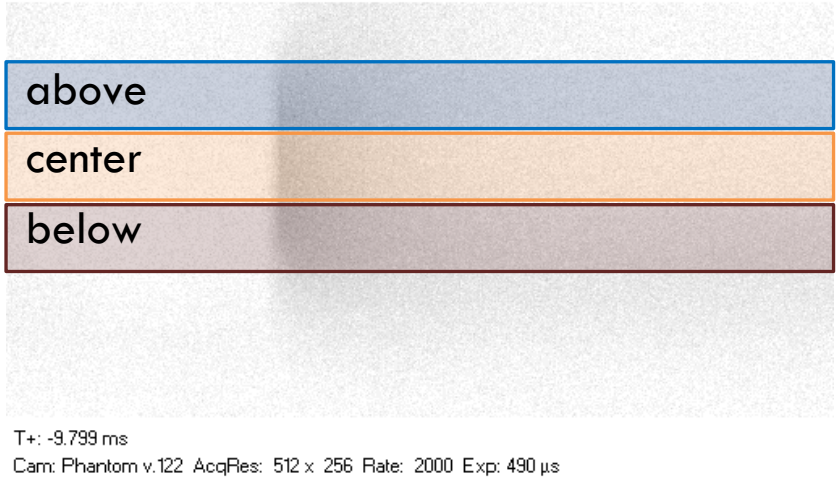
Plasma parameters	
T_e	6.5 eV
n_e	1.3×10^{19} m^{-3}
Flux	10^{23} m^{-2}s^{-1}
Gas	~5 mTorr (Ar, He)

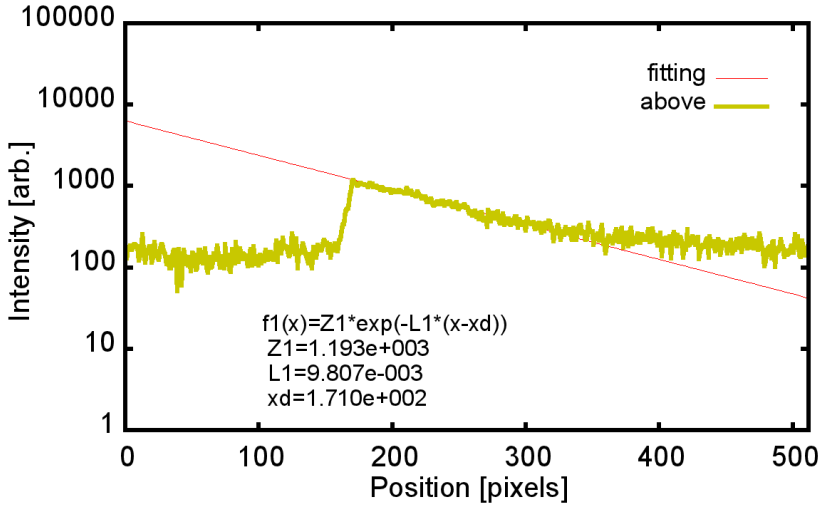
Experimental parameters	
E_{laser}	<50 J (10-20ms)
V_{bias}	-50 ~-100 V
T_{target}	< T_{fuzz}

Samples were irradiated with shots of the maximum laser energy.

➔ Once a shielding behavior was observed, smaller energy shots were tested.

Analysis of experimental results





$$f_{(x)} = I_{\mathbf{peak}} * e^{-\lambda\, x_{\mathbf{peak}}}$$

Longer Decay length

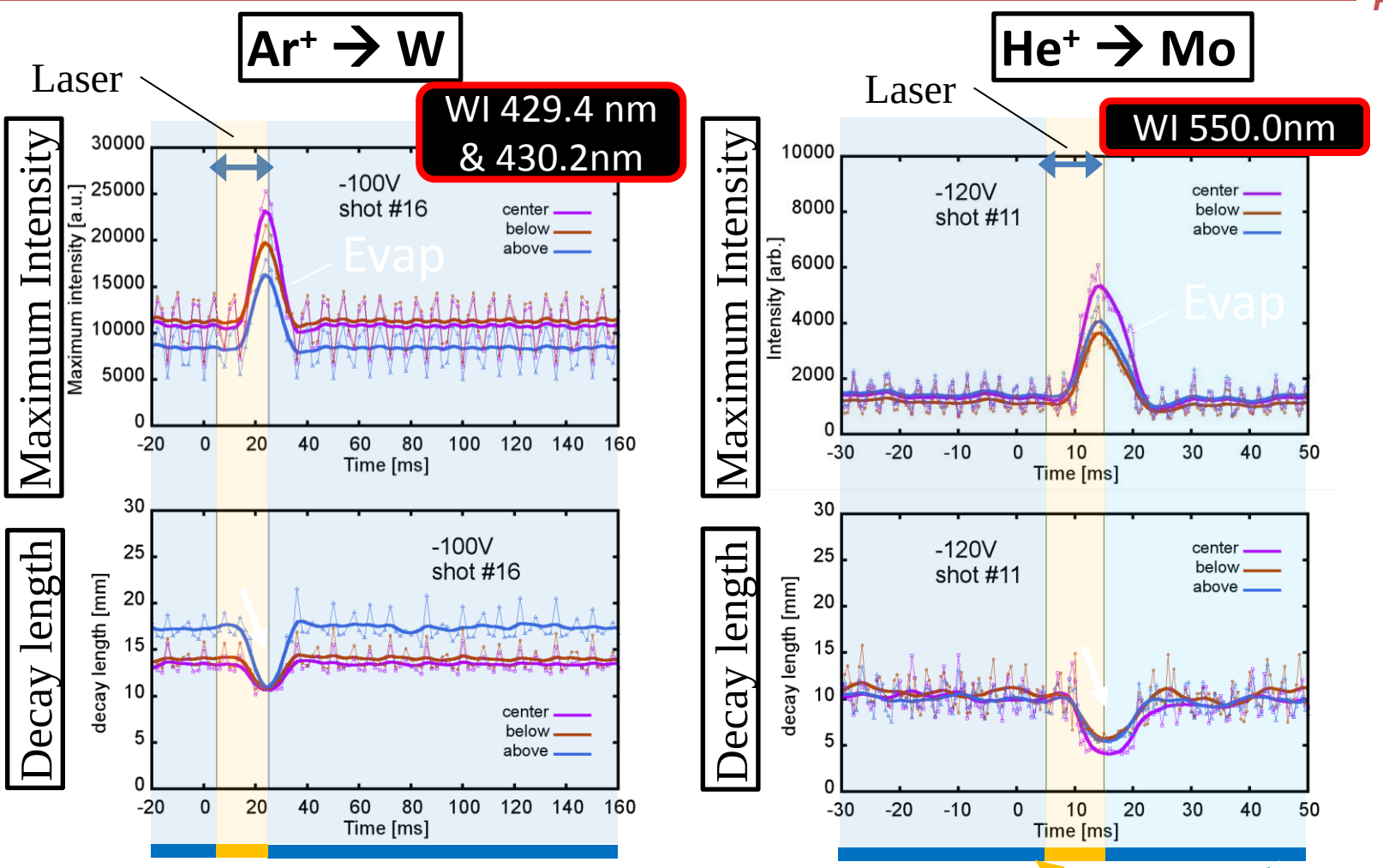
➔ Longer mean free path

→ Faster particle ejection

and/or

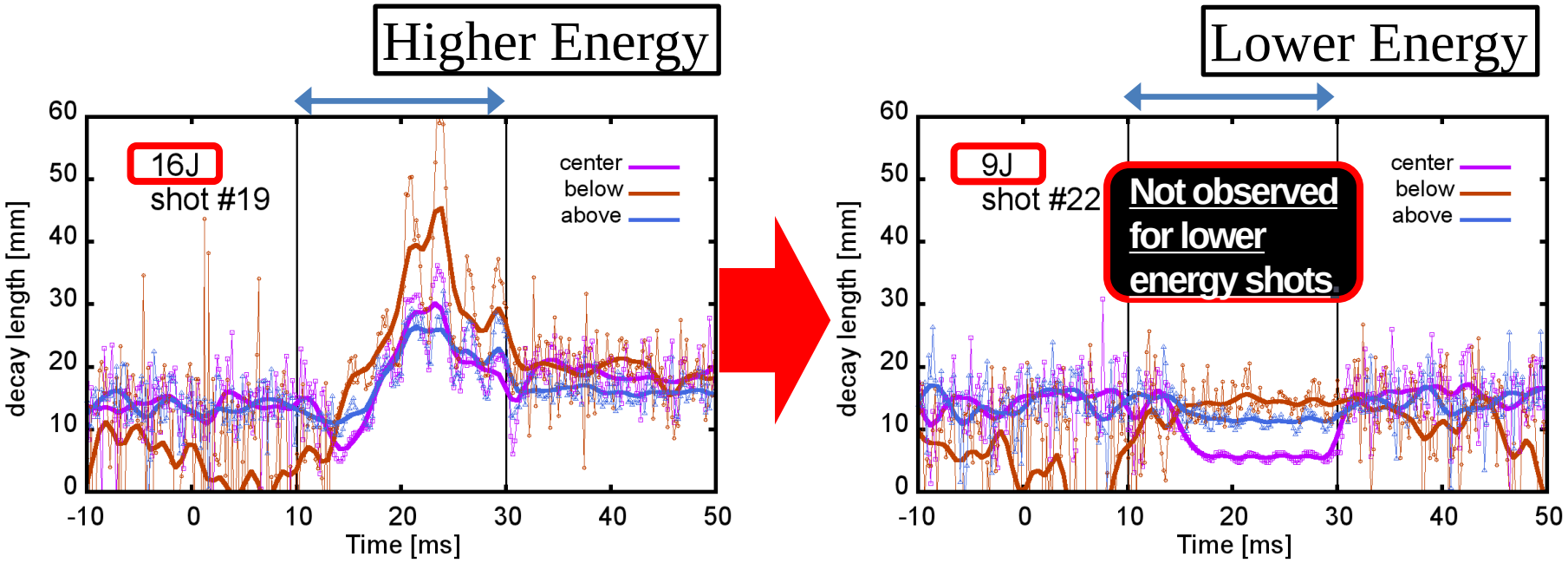
→ Lower plasma temperature

Experimental observations: W and Mo



Smaller ejection energy of evaporation than sputtering makes decay length shorter.

Experimental observations: Beryllium

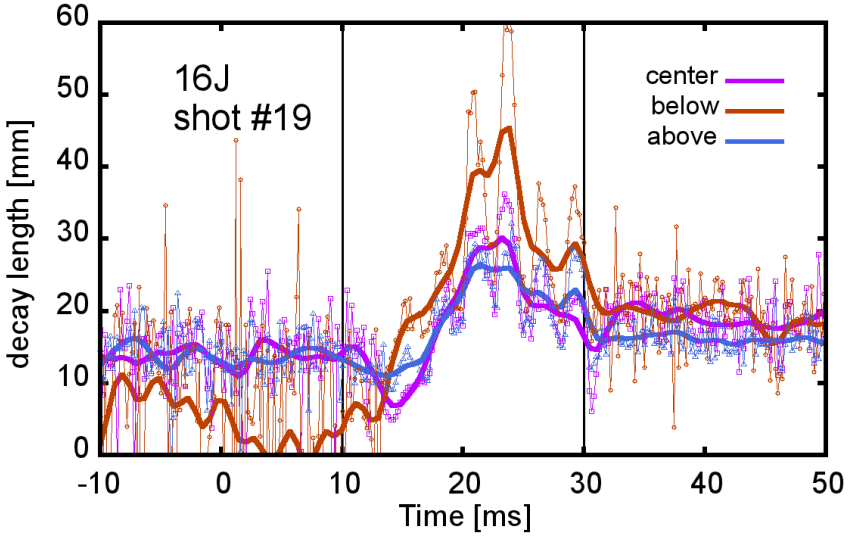


Longer decay length during laser shots means longer mean free path of Be vapor, but lower vapor velocity implies lower plasma temperature.

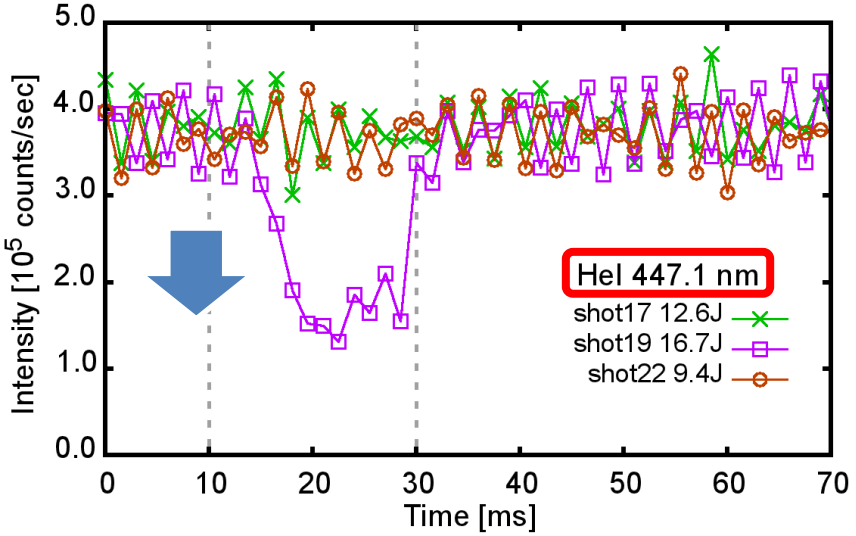
(Vapor Shielding)

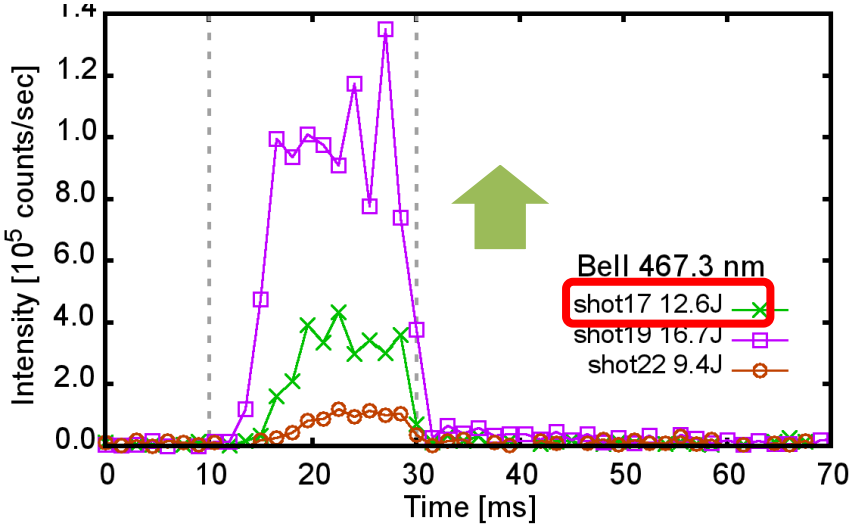
Experimental observations: Beryllium

High Speed Camera



Spectroscopy





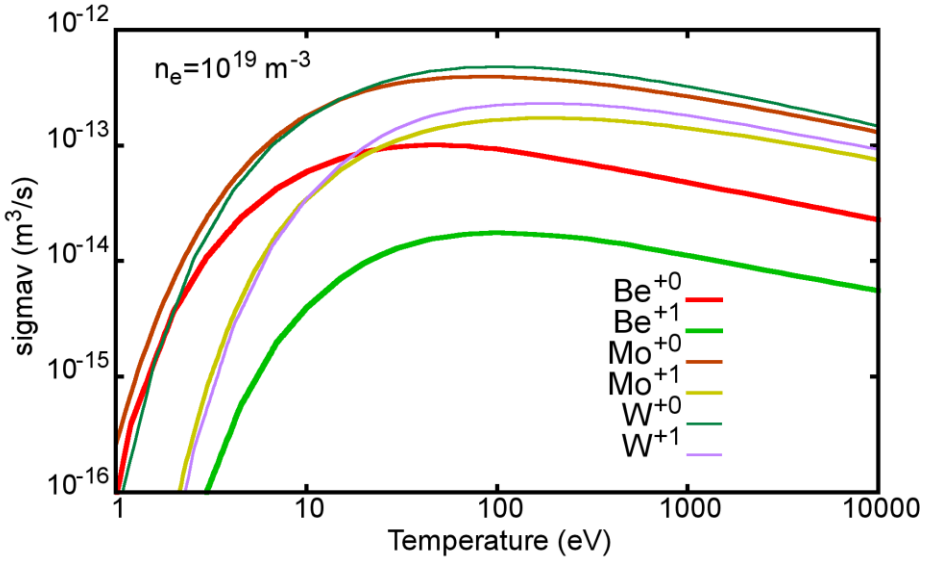
During laser shots,
- HeI intensity drops
- BeII intensity increases

➔ Plasma cooling by Be vapor.

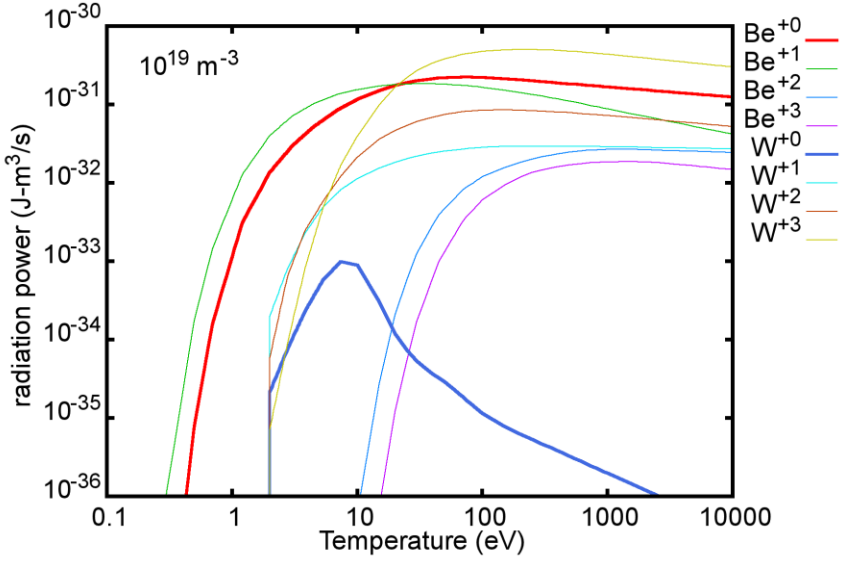
<u>No apparent cooling was observed for W & Mo.</u>

In addition to ablation velocity, atomic physics differs between W and Be

Ionization cross section

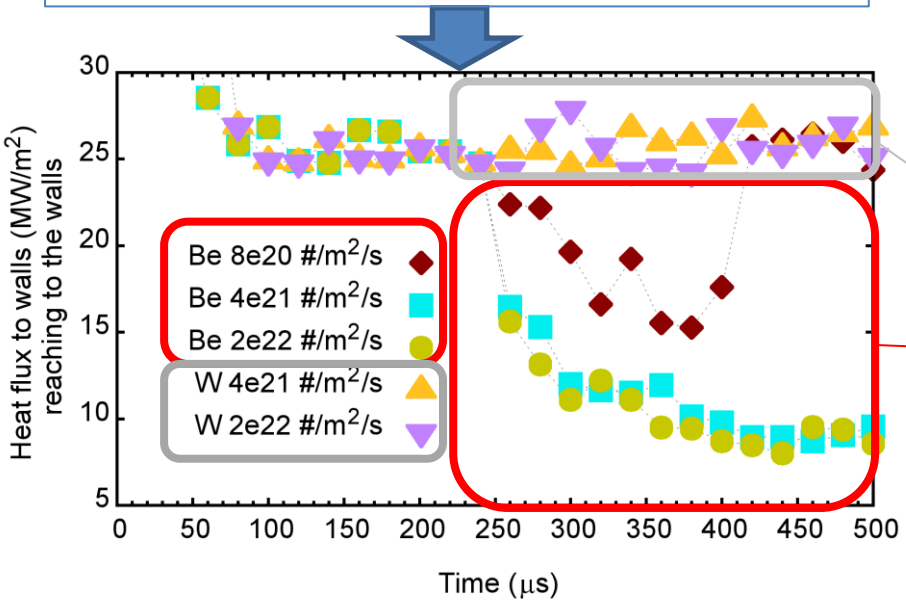


Radiated power



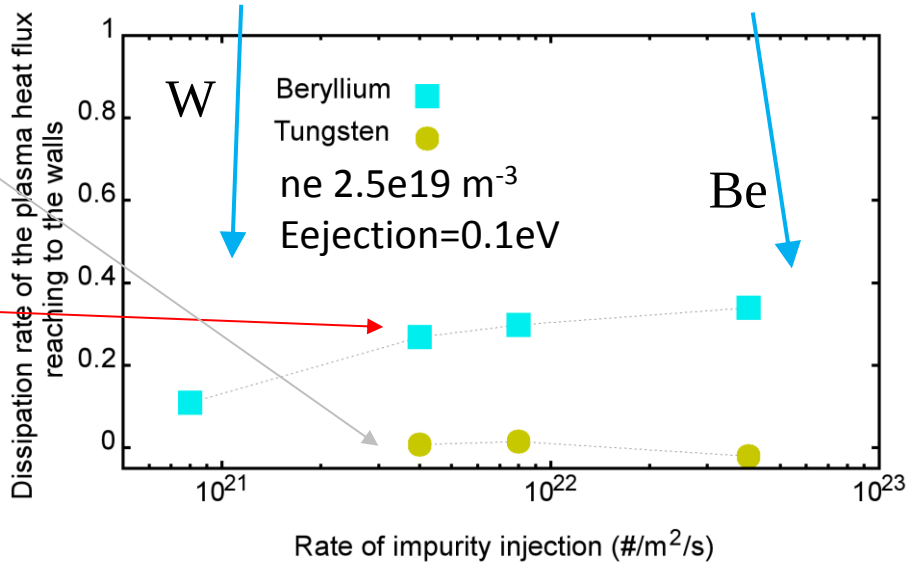
PIC simulations predict more energy dissipation in PISCES by Be than W

Impurity ejection starts at this point



Heat flux reaching the wall

vapor pressure at melt point



Heat flux dissipation rate as a function of impurity amounts.

- Be impurities significantly reduce the heat flux to the wall.
- In comparison, no apparent reduction was observed for W.

Summary of Be PMI R&D in PISCES

- Erosion of Be in a high-flux plasma environment
  - Comparisons with TRIM estimates
  - Chemically assisted physical sputtering of BeD
  - Temperature dependence of physical sputtering
- Fuel retention in Be
  - Retention in plasma exposed surfaces
  - Retention in Be codeposits
- D release behavior from Be
- Be mixed-material PMI studies
  - Be/W layers
  - Response to transient heating events
- Discussion

Summary/Discussion

- High-flux of low-z ions still appears to reduce erosion of surfaces (JET-ILW sees lower Be erosion than PISCES-B) chemistry and temperature dependent losses may be important
- Fuel retention will be dominated by codeposition with Be and estimates seem robust, although impurities could alter values
- Release of D from Be is well modeled by TMAP (not including impurity effects)
- Be/W mixed material formation on hot divertor plates is still an open issue during both steady-state as well as transient events
- The response of the plasma to particle ejection from various materials has shown differences between Be and W

While much progress has been made in PMI with Be, there are still many details to be investigated