

Reaction-diffusion based modelling of hydrogen retention in beryllium



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In place of motivation

PWI + diffusion + chemistry = multi-scale

- Simultaneous occurrence of different processes
- Continuous surface composition evolution
- Discrete description not always applicable: long computational times, error propagation
- Can continuous description be faster, easier and more flexible?

Hydrogen retention in plasma-facing materials

- Implantation, diffusion, de-/trapping, release
- Modelling as a system of coupled rate equations
- Limitation of the TMAP code [10]:
Number, occupancy and mobility of trap sites

Reaction-Diffusion System

Mass conservation: continuity equation:

$$\frac{\partial \rho_i}{\partial t} + \nabla \cdot \Gamma_D = (\text{Formation} - \text{Destruction}) + (\text{Sources} - \text{Sinks})$$

Diffusion flux: Fick's law :

$$\Gamma_D = -D \nabla \rho_i \quad D = D(T) = D_0 \exp\left(-\frac{E_a}{k_B T}\right)$$

Chemical reactions: Arrhenius eq'n:

$$R_i = -k_i \exp\left(-\frac{\Delta E_i}{k_B T}\right) [\rho_A]^a [\rho_B]^b \quad (\text{ex.: } aA + bB \rightarrow cC)$$

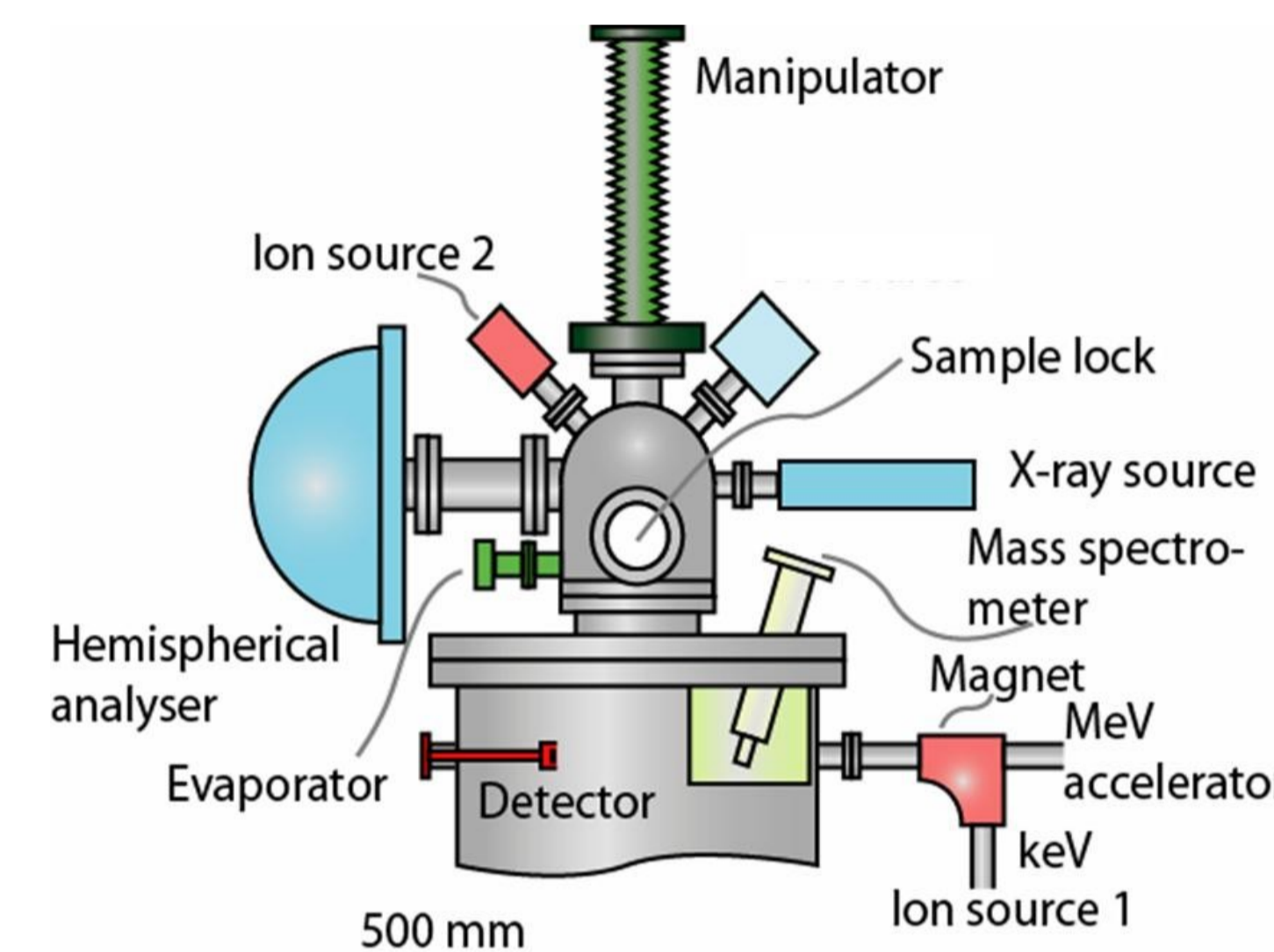
Additional non-chemical source/sink terms

- implantation / deposition
- erosion / transport from the domain

Master equation in 1D for each component (A,B,C,...):

$$\frac{\partial \rho_A(x,t)}{\partial t} = \frac{\partial}{\partial x} \left(D(T) \frac{\partial \rho_A(x,t)}{\partial x} \right) + \sum \Gamma_{\text{Formation}}(\rho_{A,B,C\dots}, x, t) - \sum \Gamma_{\text{Dissociation}}(\rho_{A,B,C\dots}, x, t) + \sum \Gamma_{\text{Sources}}(\rho_{A,B,C\dots}, x, t) - \sum \Gamma_{\text{Sinks}}(\rho_{A,B,C\dots}, x, t)$$

Application to Be experiments in ARTOSS



ARTOSS experiment [1]

(now located at Forschungszentrum Jülich):
UHV conditions for sample preparation and analysis:
In-situ surface preparation (sputtering, annealing),
D implantation, surface analysis (XPS, NRA),
and D release (Temperature Programmed Desorption)

Experiments [2-6]: implantation of D at 1, 2, 3 keV/D into Be(11-20) and B(0001) single crystals (sc) and pc-Be

Implantation profiles: D and damage (Frenkel pairs) as Gaussian fit to SDTrimSP simulation results

TPD: $T(t)$ linear at 0.7 K/s (sc Be) and 0.1–2.0 K/s (pc Be)

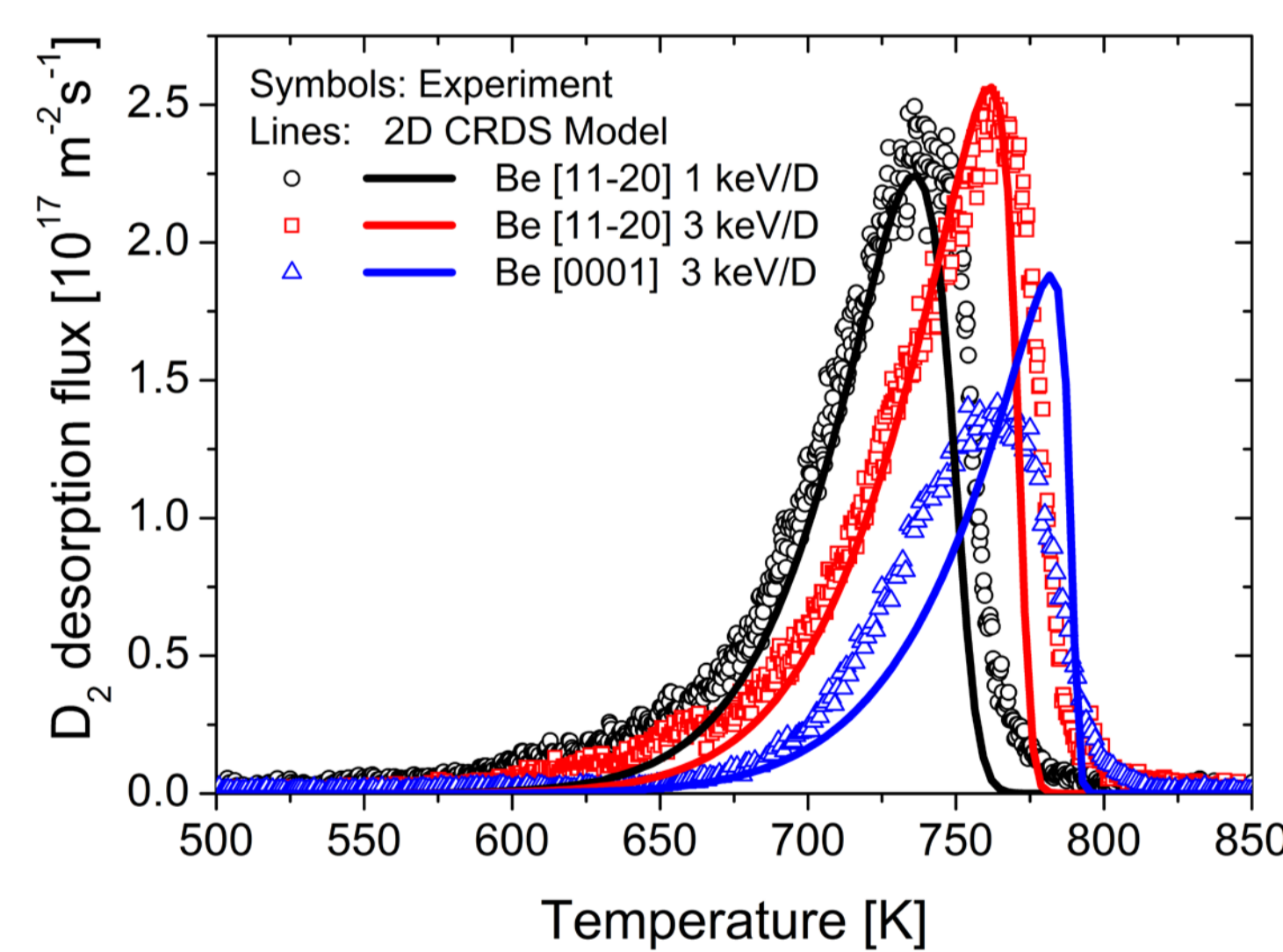
CRDS: Coupled Reaction-Diffusion System [4,5]

H – solute hydrogen (at interstitial position)
V – mono-vacancy; I – self-interstitial Be atom
HV – hydrogen-mono-vacancy complex (trapped H)

$$\text{Trapping } (H + V \rightarrow HV): R_1 = \rho_H \left(\frac{\rho_{MV}}{\rho_{Be}} \right) \frac{D_H(T)}{a_0^3} \\ \text{Detrapping } (HV \rightarrow H + MV): R_2 = \rho_{HV} v \exp\left(-\frac{E_{det}}{k_B T}\right)$$

Simulation of all phases of the experiment: implantation, relaxation, TPD
Using Wolfram Mathematica® Software
Issues: stiff system, numerical instabilities

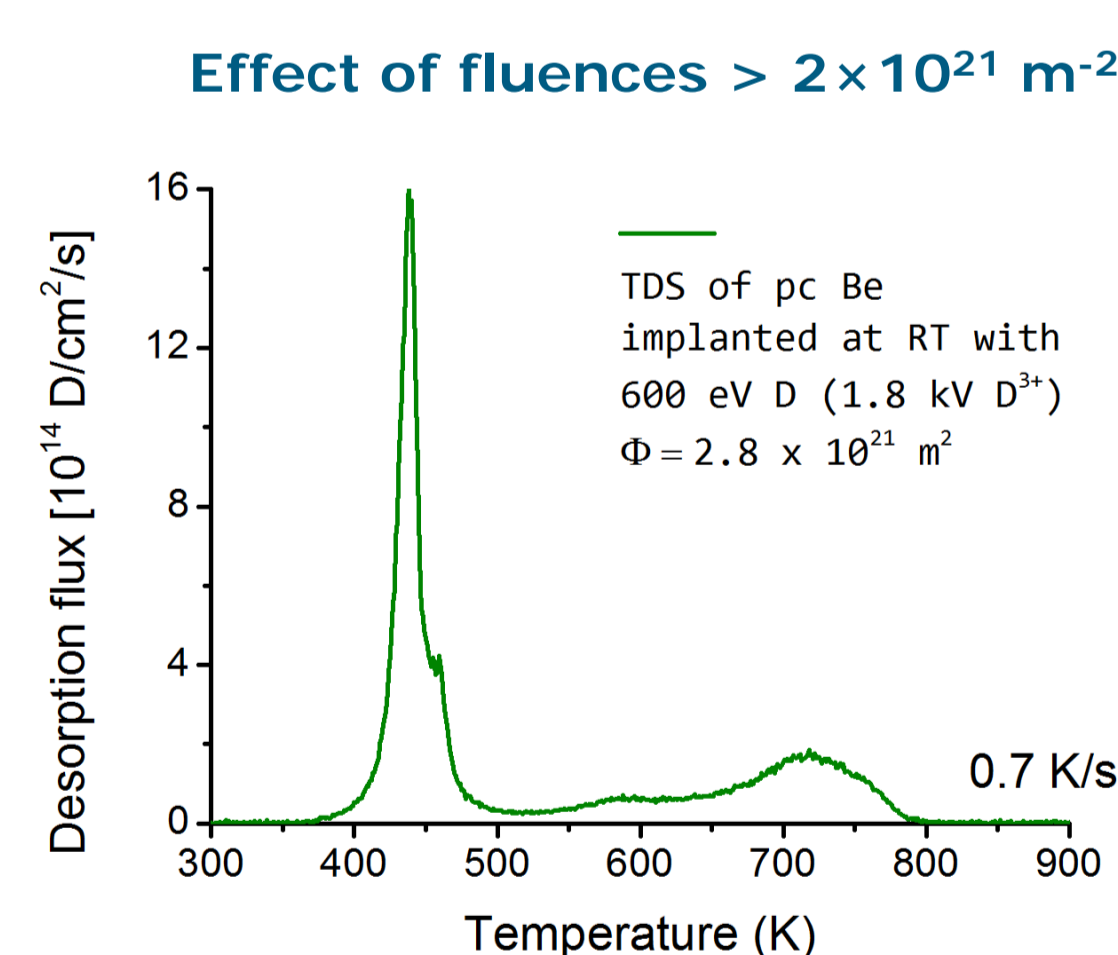
Conclusions from earlier studies [4,6]



2D CRDS: using DFT data [7] for diffusion and trapping

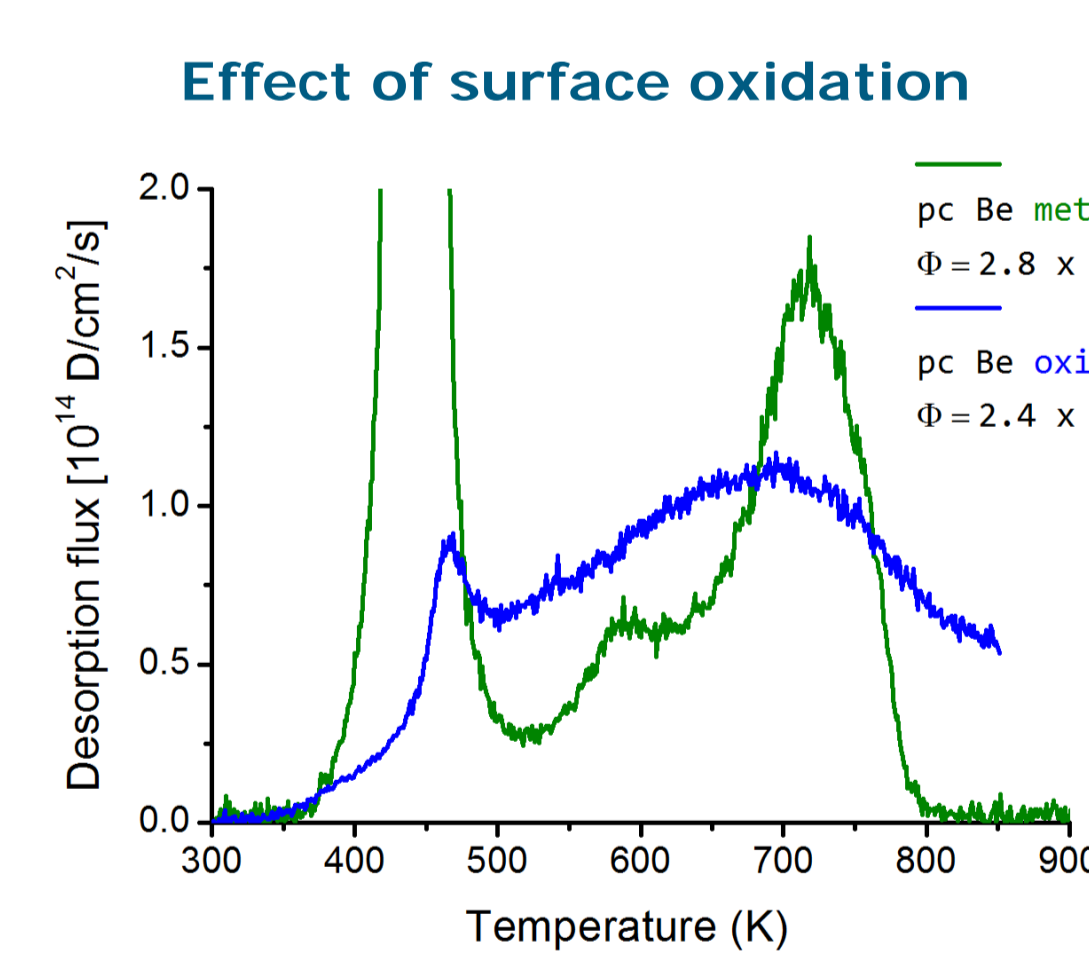
D release: re-trapping on the way to surface, TPD peak shift with D ion energy (ion range)

Effect of crystal orientation due to anisotropy of Be interstitial diffusion: faster interstitial diffusion to surface → less annihilation with vacancies → more V left as trap sites → effect of crystal orientations: Anisotropic diffusion of Be self-interstitial? [7-9]



Low-temperature peaks:

fluence threshold
structural changes due to oversaturation with D?
amorphization?
BeD₂ hydride formation?
Surface coverage effects?

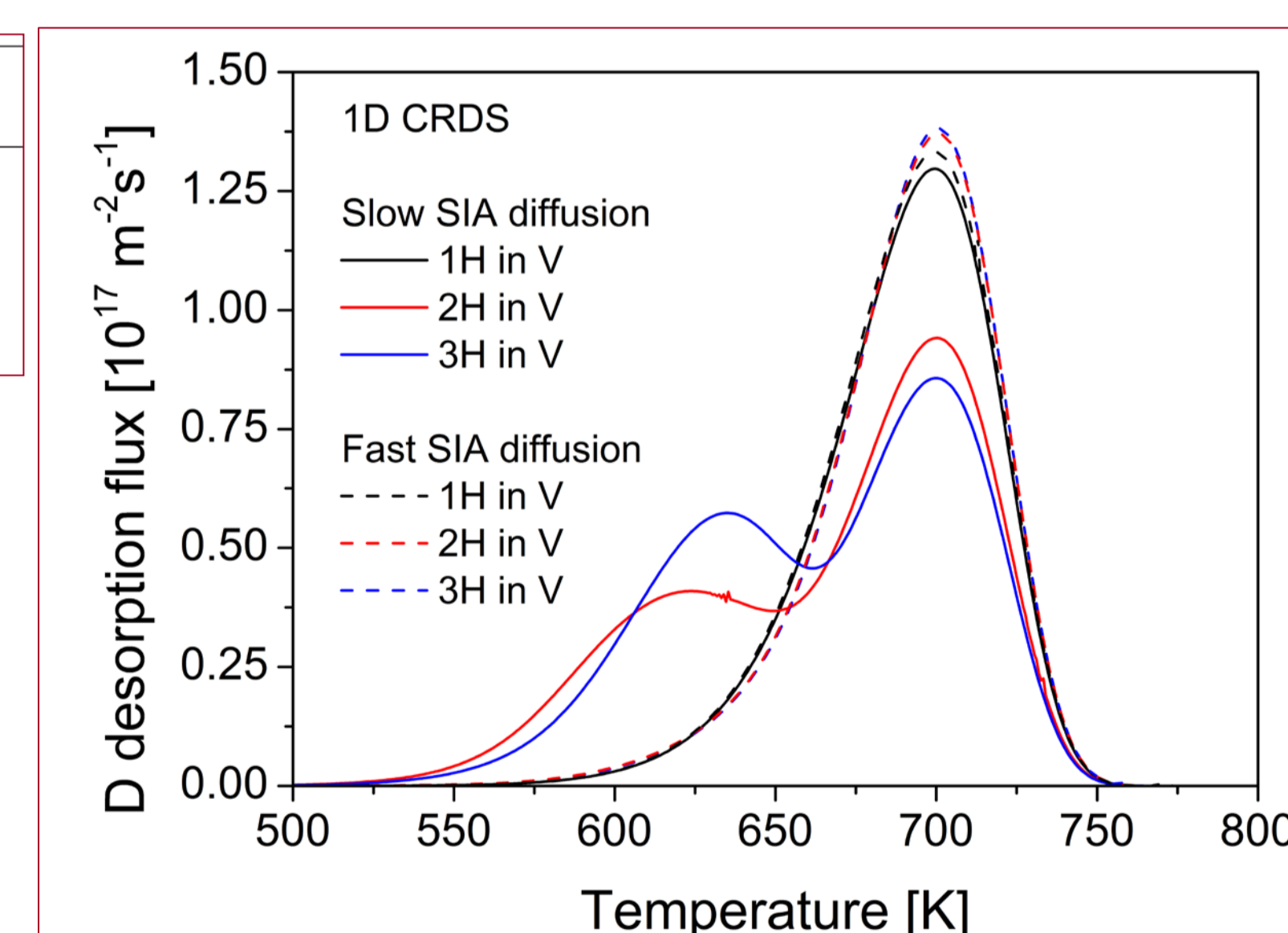


Implantation within BeO layer

similar total retention but different release behavior
low-T peak reduced
baking less efficient

Multiple trapping in vacancies [7]

nH	activation barrier (eV)	
	detrapping	trapping
1H	1.70	0.85
2H	1.35	0.52
3H	1.34	0.42
4H	1.32	0.40
5H	1.37	0.42
6H	0.98	1.37



Slow diffusion of Be self-interstitials → double peak structure → not observed experimentally → supports fast diffusion (DFT)

Fast diffusion → minor influence of multiple trapping

Note: de-trapping energy of 6th H E_{dt} ~ 1 eV [7] would correspond to a release peak in the low-T range of 400-500K in both (fast and slow Be diffusion) cases...

Outlook

CRDS

- Many parameters to control
- Not a "black box" solution

Choices to make

- Mathematica® vs Coding
- CRDS vs KMC

Will be important

- BeD₂, BeO, amorphous layers
- Surface limiting processes

New experiments in ARTOSS

- Be-N-D, Be-W-D systems
- Isotope exchange

Notes:

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