Interface-induced enhanced deuterium plasma-driven permeation in chemical vapor deposition tungsten-copper composite

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by Dr. Long CHENG, Beihang University | 2025 May 26 | Email: LCheng@buaa.edu.cn

Background: Hydrogen isotope transport in plasma-facing component

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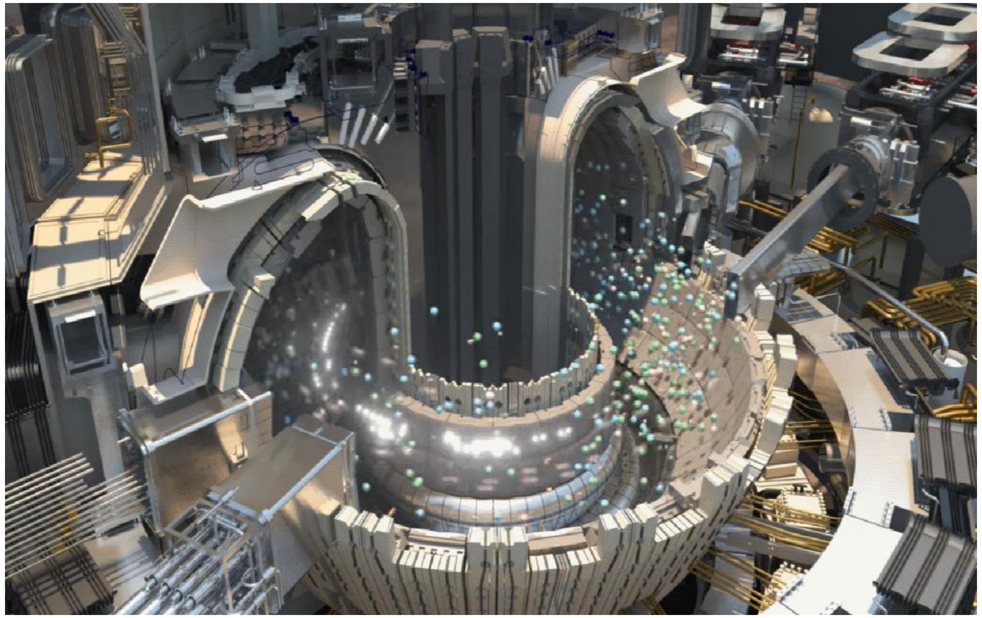

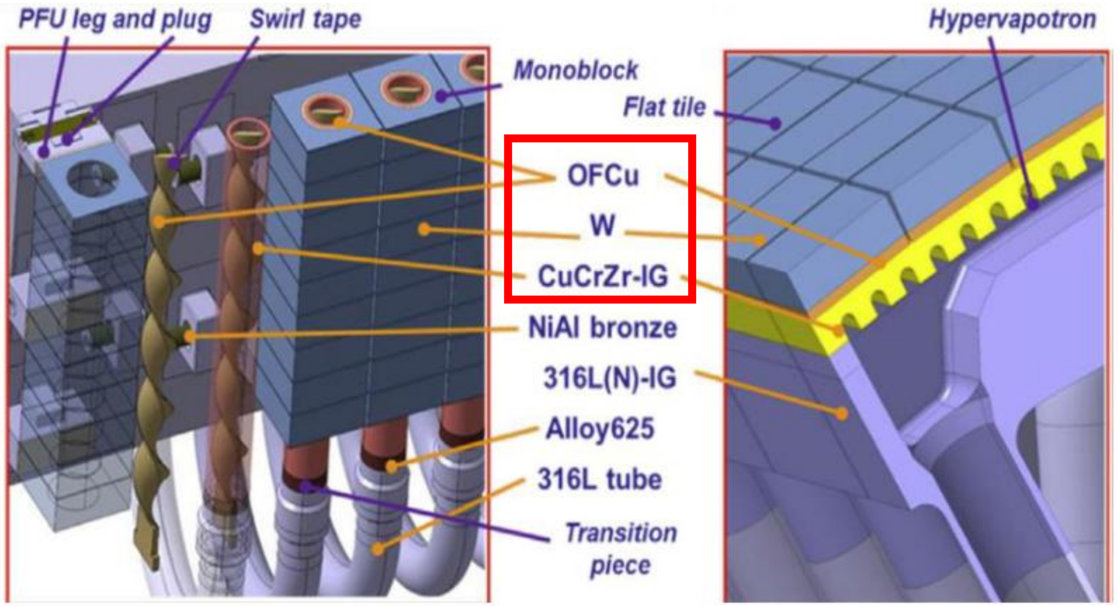

In future fusion devices, the design of robust plasma-facing components (PFCs) is essential

- Tungsten (W) is considered as the most probable plasma-facing material (PFM) due to its high melting point, high sputtering threshold and low fuel inventory
- Copper-chromium-zirconium (CuCrZr) alloy is commonly selected as the heat-sink material due to its high ductility and favorable thermal conductivity
- To mitigate the high mechanical stress in-between W and CuCrZr, a copper (Cu) interlayer is employed

Background: Hydrogen isotope transport in plasma-facing components

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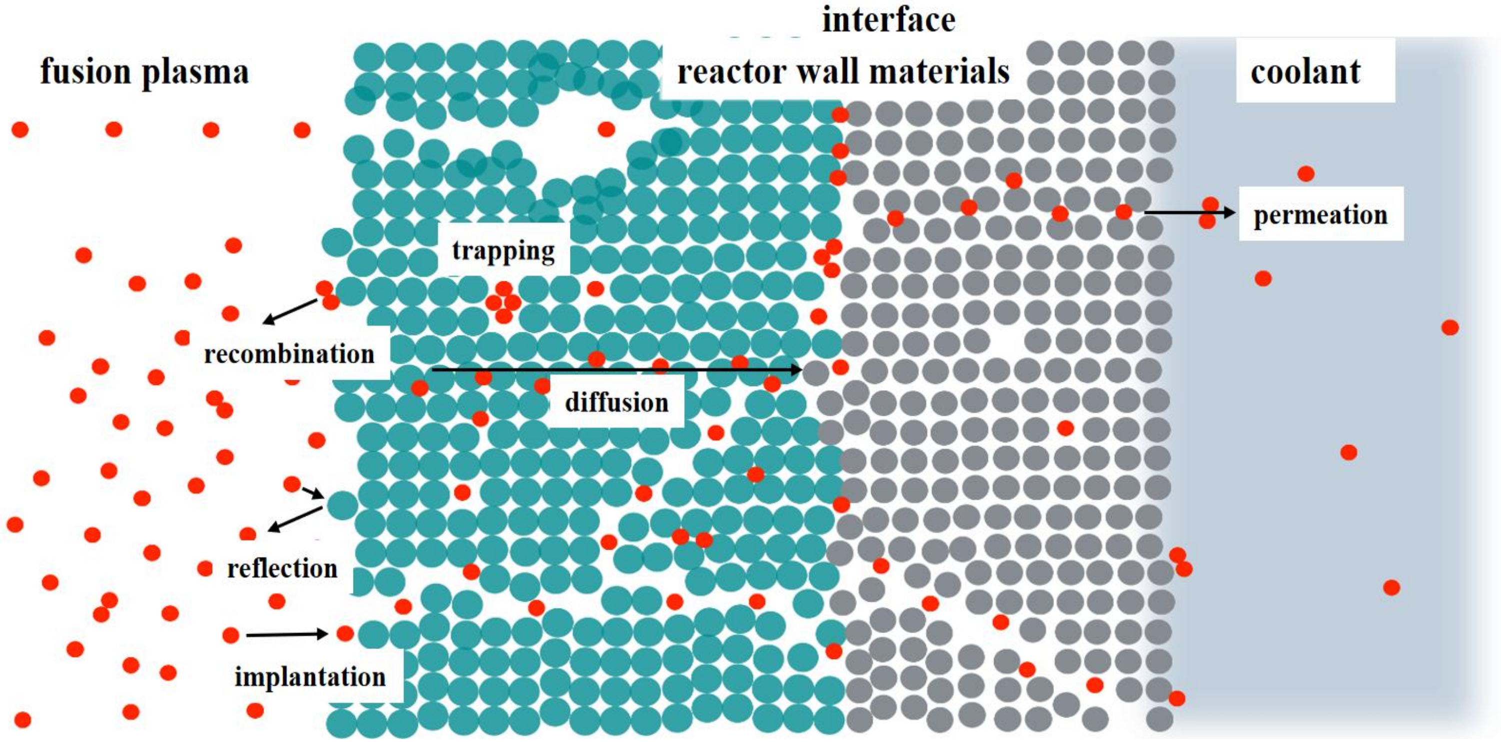


- During the operation of tokamaks, PFCs are directly exposed to the edge plasma, leading to a series of plasma-material interactions. The implanted hydrogen isotope (HI) can diffuse into and permeate through the bulk, then penetrate into the coolant, potentially forming radioactive tritiated water which may impact the self-sustaining cycle of tritium.
- Although Cu is commonly used as an interlayer in PFCs and directly bonded to W, experimental studies on D permeation in W/Cu composite remain limited

Kalle Heinola, Hydrogen Permeation in Fusion-Relevant Materials, 2020

Research Purpose & Expected Results

Purpose

- Investigate deuterium transport in W-Cu composite, including diffusion, recombination and permeation.
- Understand the difference of deuterium transport in between W-Cu composite, bare W and bare Cu under identical plasma-driven permeation conditions

Expected Results

- D diffusion coefficient of W-Cu composite should fall between that of bare W and bare Cu
- Deuterium permeation flux in W-Cu composite should be lower than in bare Cu due to the lower solubility of H in W

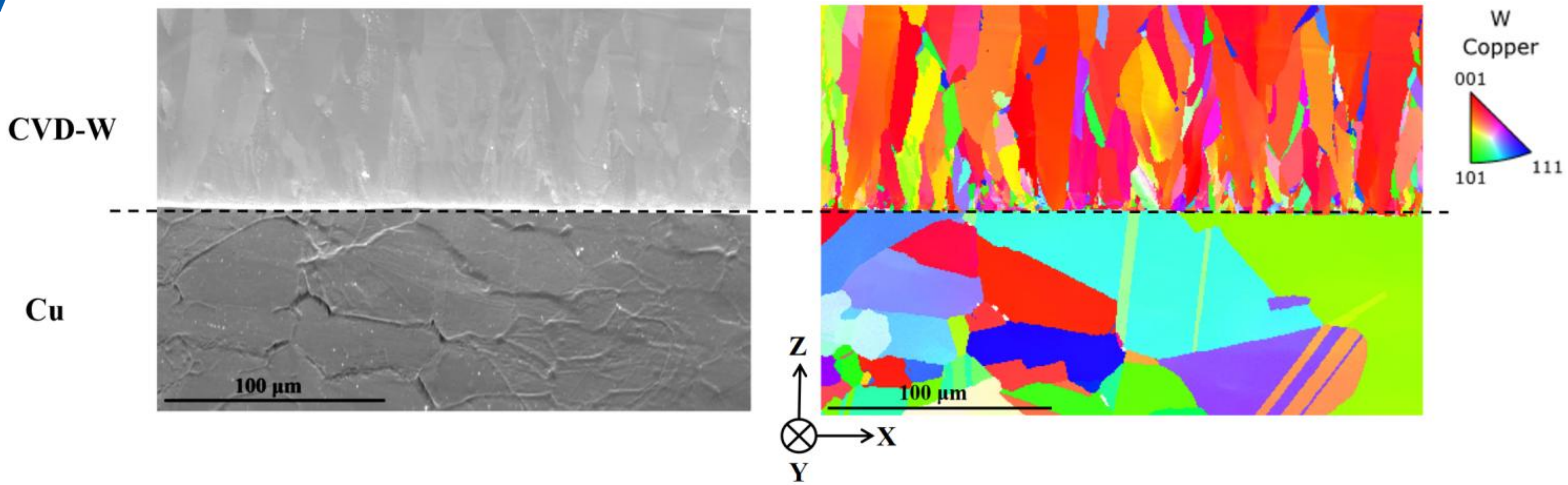
Experimental details

- In this work, three types of materials were used, including CVD-W/Cu composite, bare CVD-W (with >99.9999 wt% purity) and bare Cu (with >99.99 wt% purity)
- Deuterium plasma-driven permeation experiments were carried out in the PREFACE facility at the Institute of Plasma Physics, Chinese Academy of Sciences
- Scanning electron microscope (SEM), electron backscatter diffraction (EBSD), energy dispersive spectrometer (EDS) and Ga focused ion beam (FIB) were measured at Beihang University
- A typical W(110)/Cu(111) interface structure is fabricated in DFT calculations, using the Vienna ab initio Simulation Package (VASP)17
- D permeation in samples was analyzed using TMAP7

Sample	Size (μm)	Temperature (K)				Plasma flux (m^{-2} s^{-1})
CVD-W/Cu	Φ25 *250	741	700	660	/	~3×10^{20}
Cu	Φ20*172	742	700	620	587	
CVD-W	Φ20*100	790	742	695	643	

Sun Y., Cheng L., et al., Nucl. Fusion 65 (2025) 066003

Cross-section morphology of CVD-W/Cu interface



- CVD-W/Cu was produced using chemical vapor deposition, which has advantages of high purity, high density, excellent thermal conductivity, and preferred orientation along <001>.
- The growth direction of W grains is parallel to the normal direction of the exposure surface, exhibiting a columnar grain structure.

W and Cu distribution near the CVD-W/Cu interface

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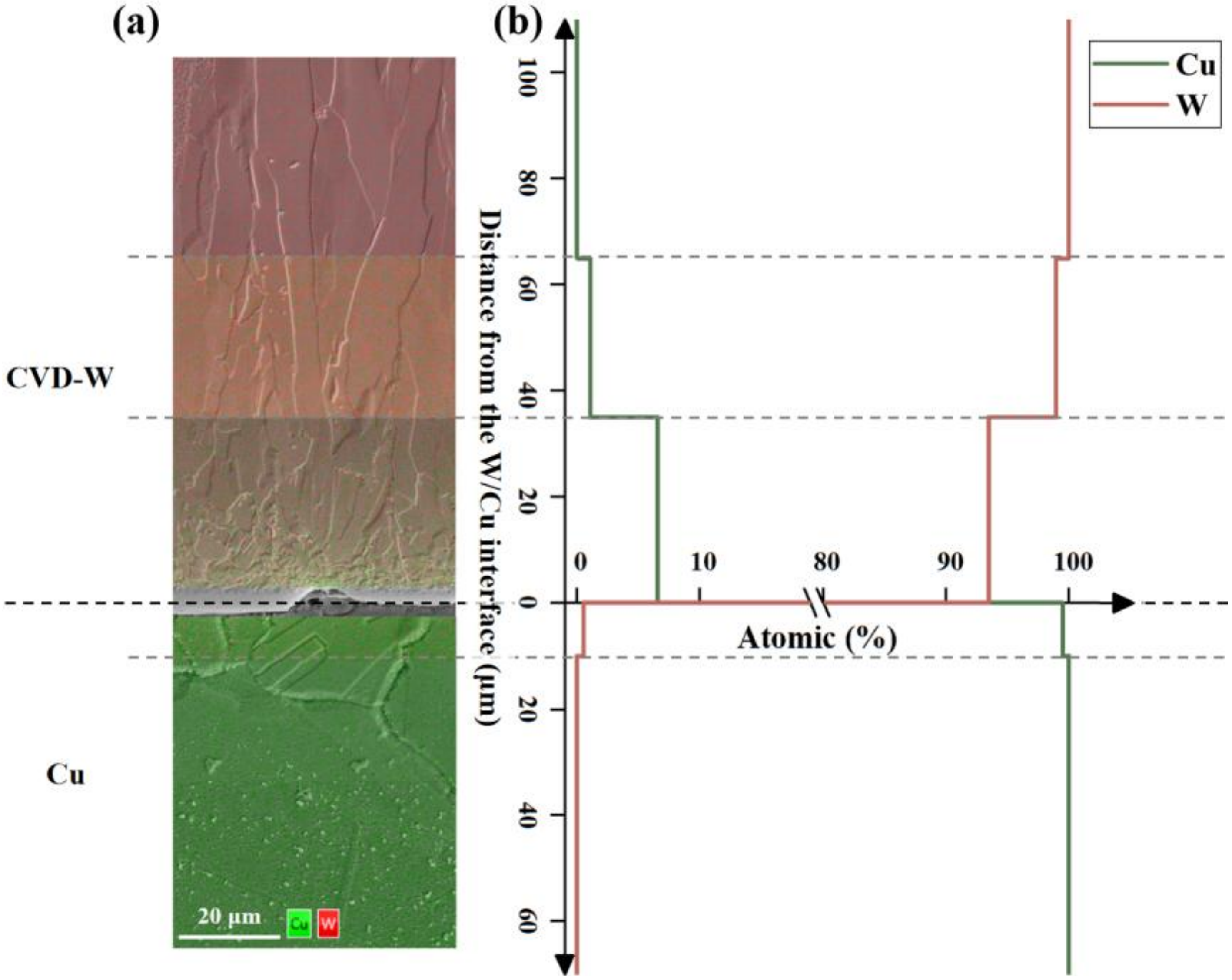
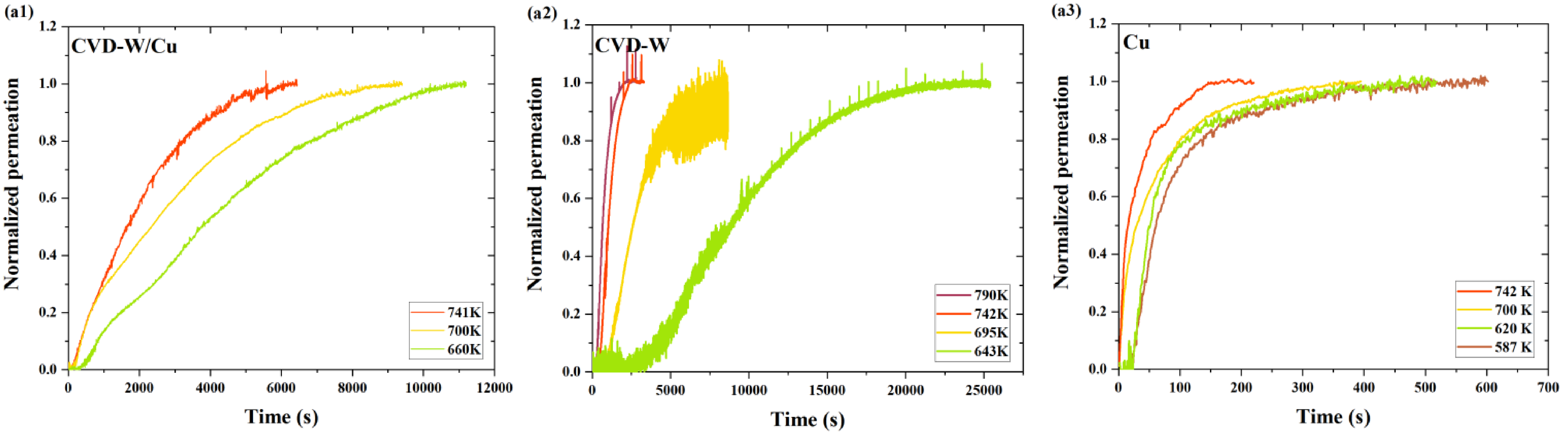


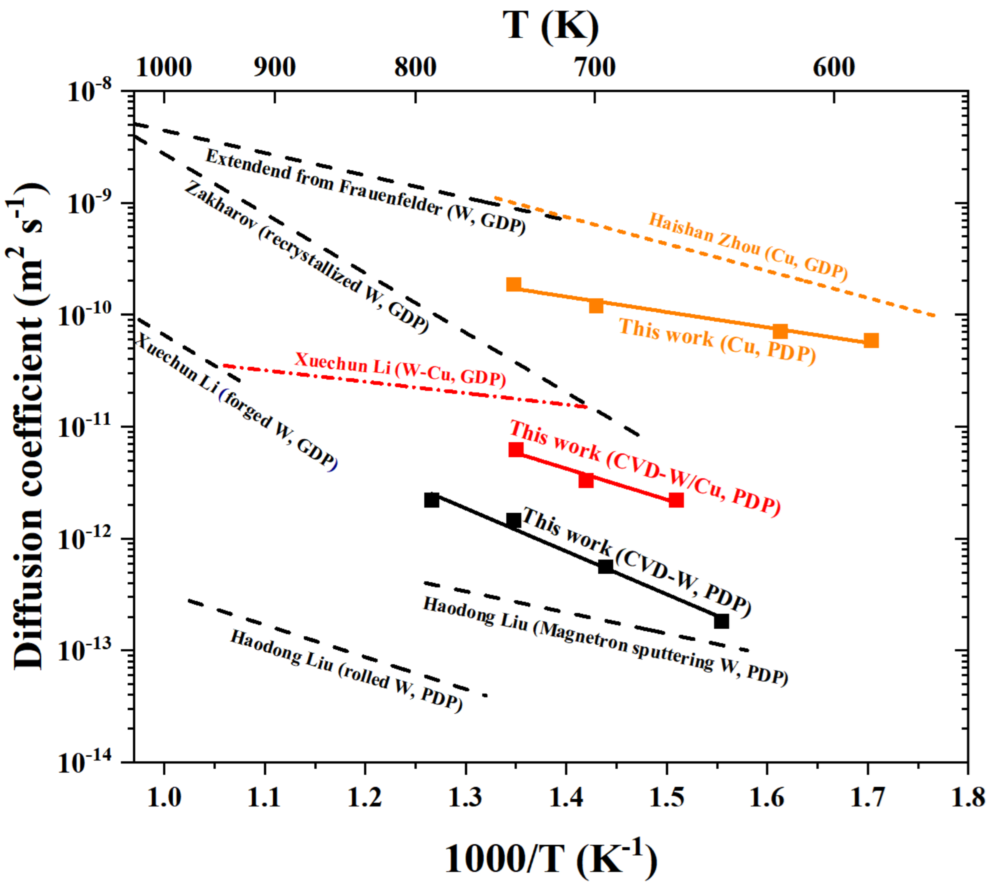
- In the CVD-W layer, the average Cu atomic concentration is 6.6 at.% within ~35 μm from the interface, and decreases to 1.1 at.% at ~35 μm-65 μm from the interface.
- In the Cu layer, only a trace amount of W (0.5 at%) was detected within ~10 μm from the interface, with no W detected outside this region.

Transient PDP flux



- Normalized permeation curves for 250 μm thick CVD-W/Cu, 100 μm thick CVD-W and 172 μm thick Cu at different temperatures.
- Break-through time in the W/Cu composite is longer than in bare W.

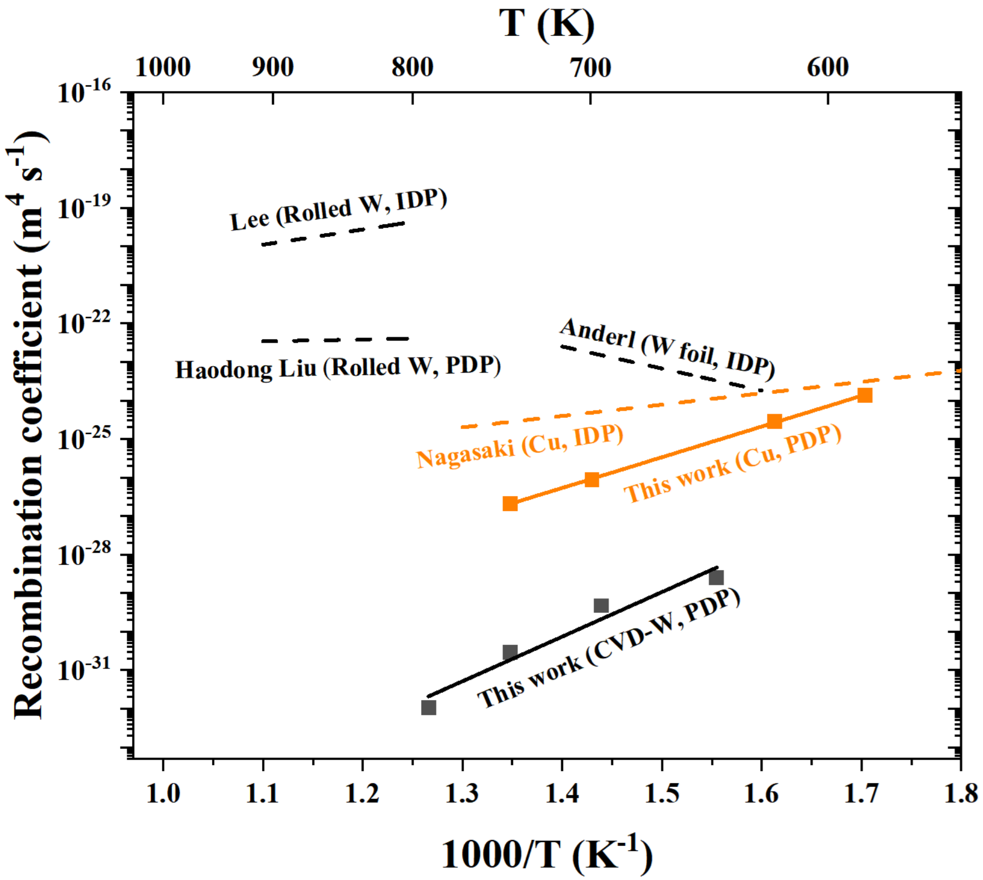
D diffusion coefficients and front surface recombination coefficient



$$D_{CVD-W/Cu} = 3.2 \times 10^{-8} exp(\frac{-0.55\ [eV]}{k_b T})$$

$$D_{CVD-W} = 1.9 \times 10^{-7} exp(\frac{-0.77\ [eV]}{k_b T})$$

$$D_{Cu} = 1.3 \times 10^{-8} exp(\frac{-0.27\ [eV]}{k_b T})$$



$$K_{CVD-W} = 4.5 \times 10^{-47} exp(\frac{2.30\ [eV]}{k_b T})$$

$$K_{Cu} = 3.8 \times 10^{-38} exp(\frac{1.58\ [eV]}{k_b T})$$

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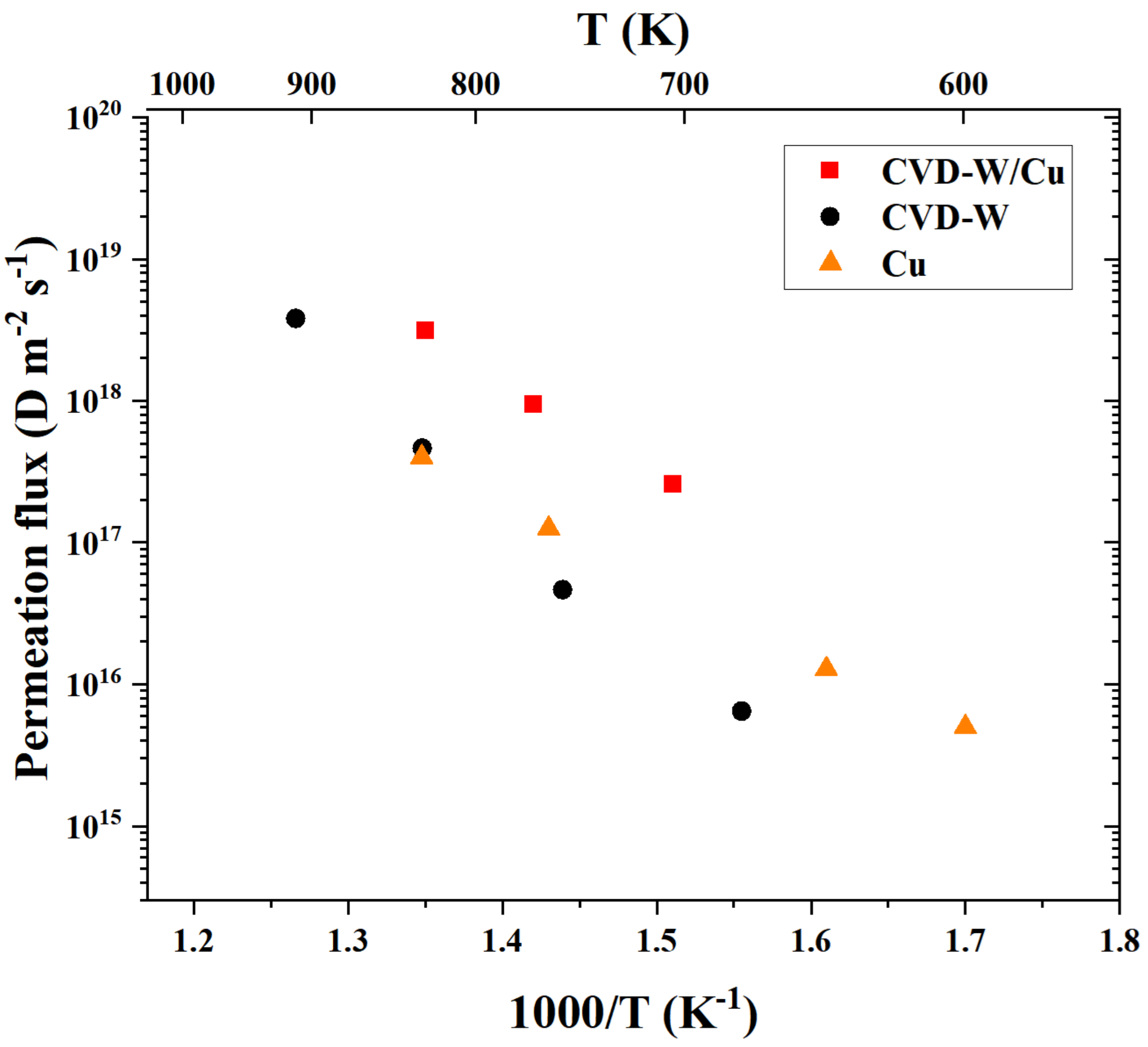
Steady state permeation flux



Steady state permeation flux as a function of temperature in CVD-W/Cu, CVD-W and Cu

- In CVD-W and Cu, the steady state permeation flux is converted by assuming a thickness of 75 μm and 175 μm, respectively.
- Notably, the steady state permeation flux in CVD-W/Cu is higher than that in bare Cu within the experimental temperature range.

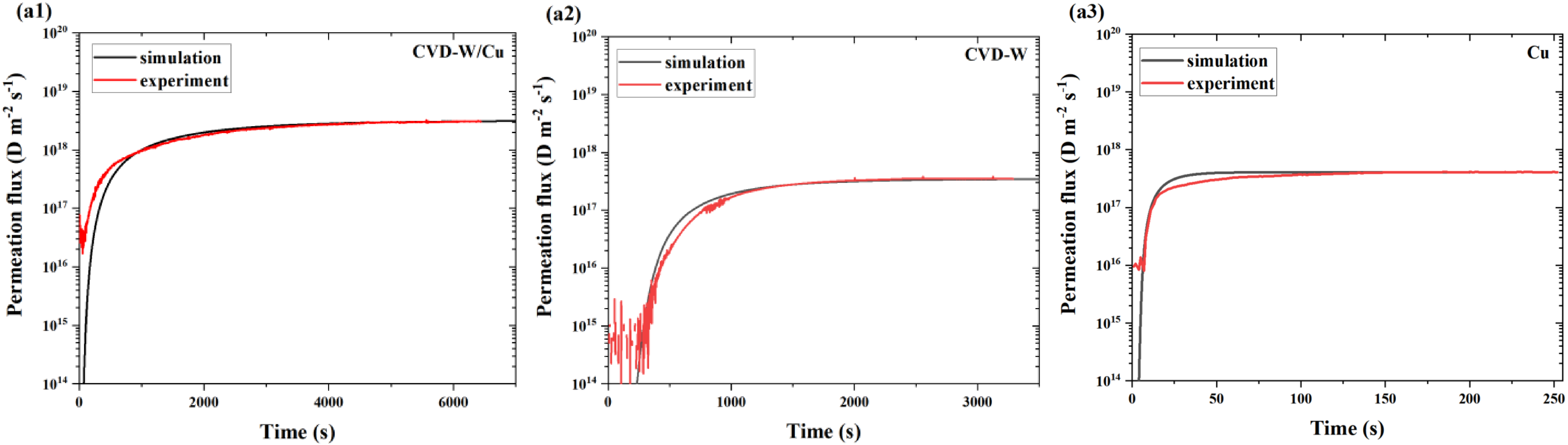
"Permeation flux in W-Cu composite should be lower than in bare Cu due to the lower solubility of H in W"

TMAP Simulation of PDP experiments

Input parameters	Values		
Temperature	741 K		
Materials	W	Cu-entry region of W	Cu
Length	**10 μm**	**65 μm**	175 μm
Solubility	$1.87 \times 10^{24} exp(\frac{-1.04\ [eV]}{k_b T})$	$\mathbf{1.87 \times 10^{24} exp(\frac{-0.6[eV]}{k_b T})}$	$3.14 \times 10^{24} exp(\frac{-0.57\ [eV]}{k_b T})$
Diffusion coefficient	$1.9 \times 10^{-7} exp(\frac{-0.77\ [eV]}{k_b T})$		$1.3 \times 10^{-8} exp(\frac{-0.27\ [eV]}{k_b T})$
Recombination coefficient	$4.5 \times 10^{-47} exp(\frac{2.30\ [eV]}{k_b T})$	/	$3.8 \times 10^{-38} exp(\frac{1.58\ [eV]}{k_b T})$
Implantation depth	1.6 nm	/	3 nm
Implantation coefficient	0.6	/	0.9

- The diffusion coefficients and recombination coefficients are taken from this work, as obtained in the experimental temperature range of ~600 K-800 K.

TMAP Simulation of PDP experiments



- Comparison between the experimental and simulated permeation curves in 250 μm thick CVD-W/Cu, 100 μm thick CVD-W and 172 μm thick Cu at 741 K.

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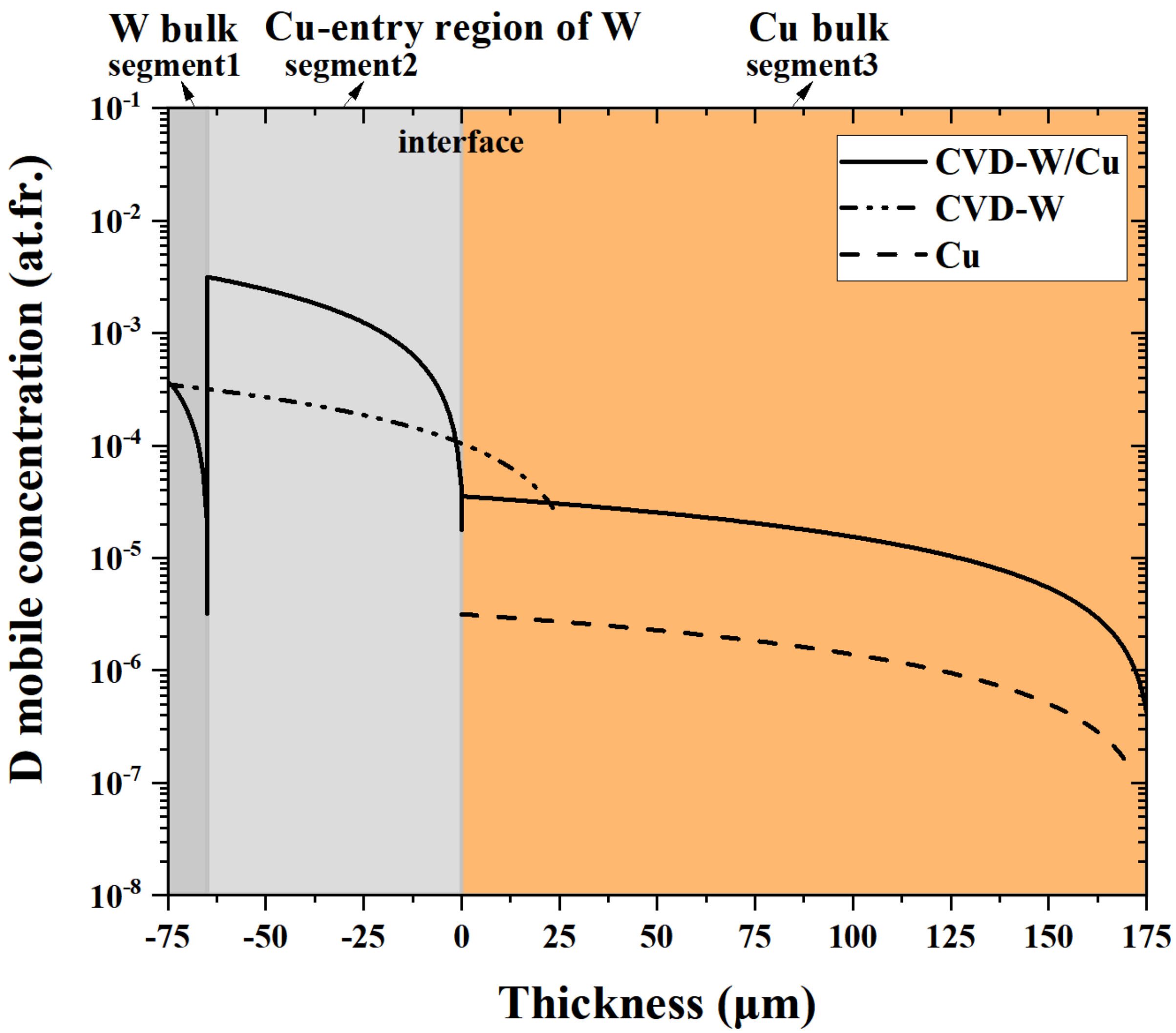
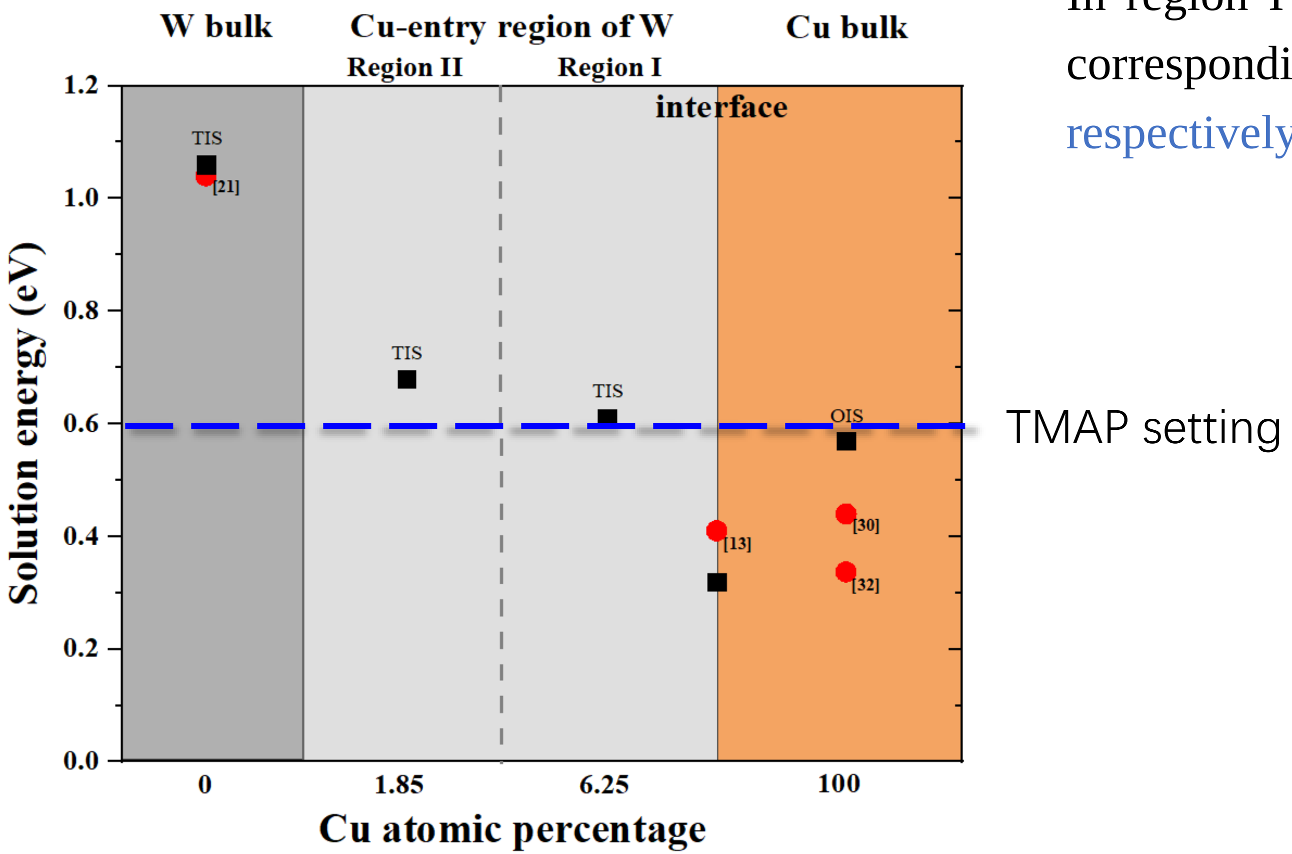
TMAP Simulation of PDP experiments



- Simulated solute D concentration distribution along the sample thickness after reaching steady state permeation at 741 K.
- Notably, because of the lower D solution energy in the Cu-entry region of W (segment2), the D mobile concentration within the W layer exhibits a discontinuity, with an abrupt increase in segment2.
- The substantial D accumulation in segment2 drives more D atoms diffusing into the Cu layer, thereby enhancing the permeation flux in W/Cu composite compared to bare Cu.

H solution energies in W/Cu composite

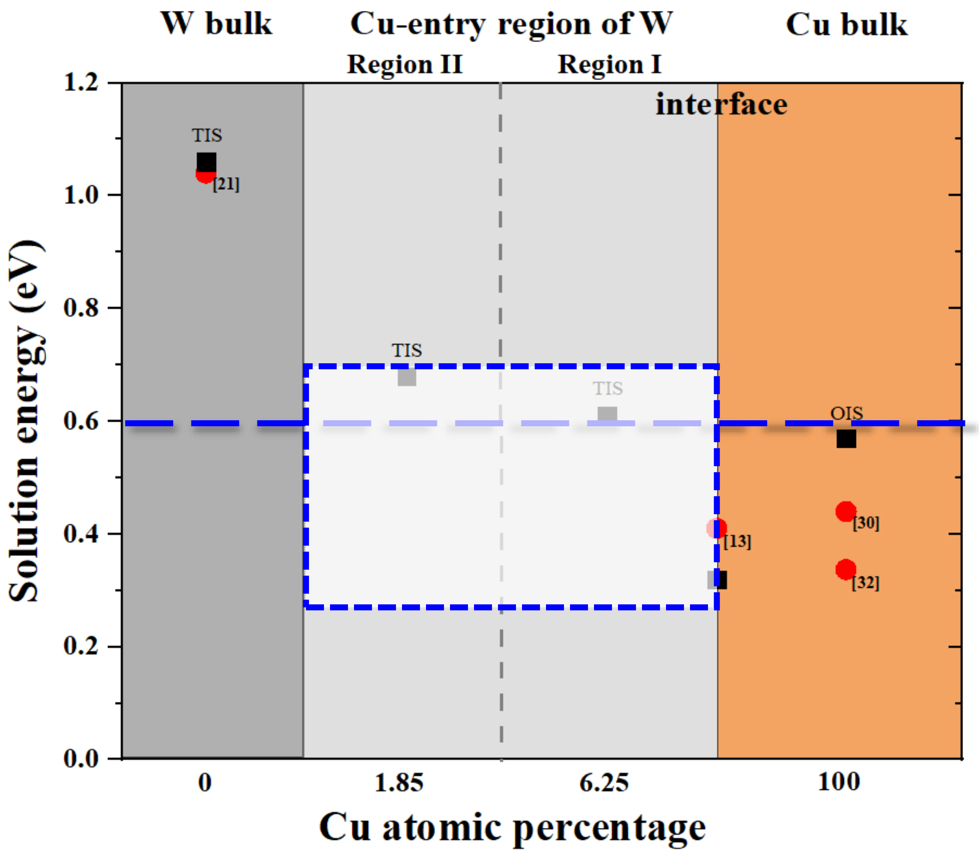
Calculated H solution energies (with zero-point energy) in the W/Cu composite using DFT



- In region I and region II, H atoms tend to dissolve near Cu atoms, corresponding to H solution energies of 0.61 eV and 0.68 eV, respectively, which are lower than the value in the W bulk.

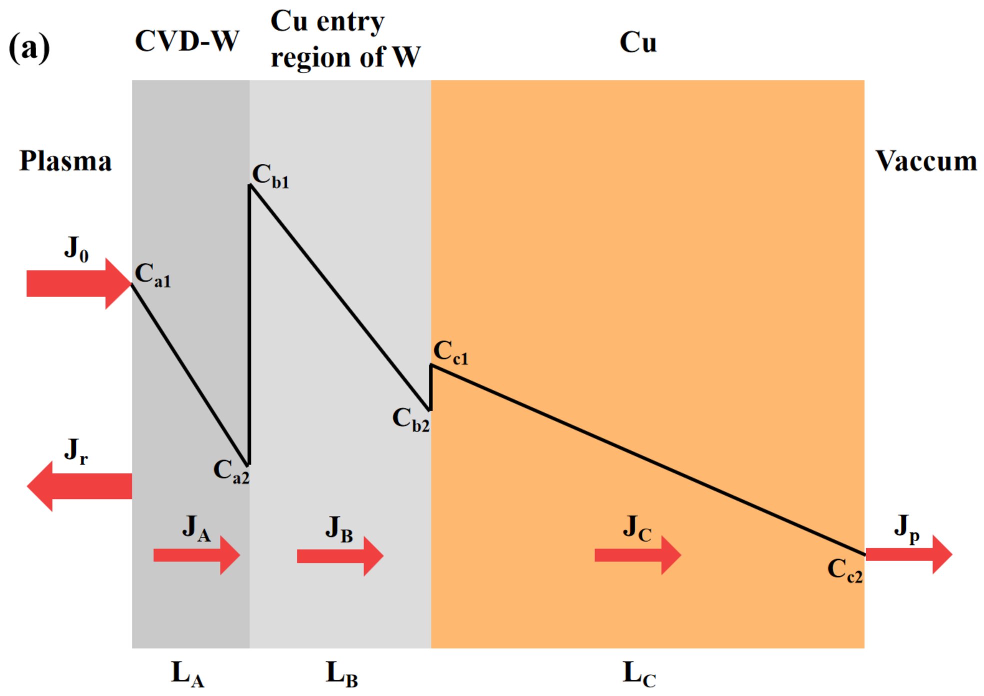
H solution energies in W/Cu composite

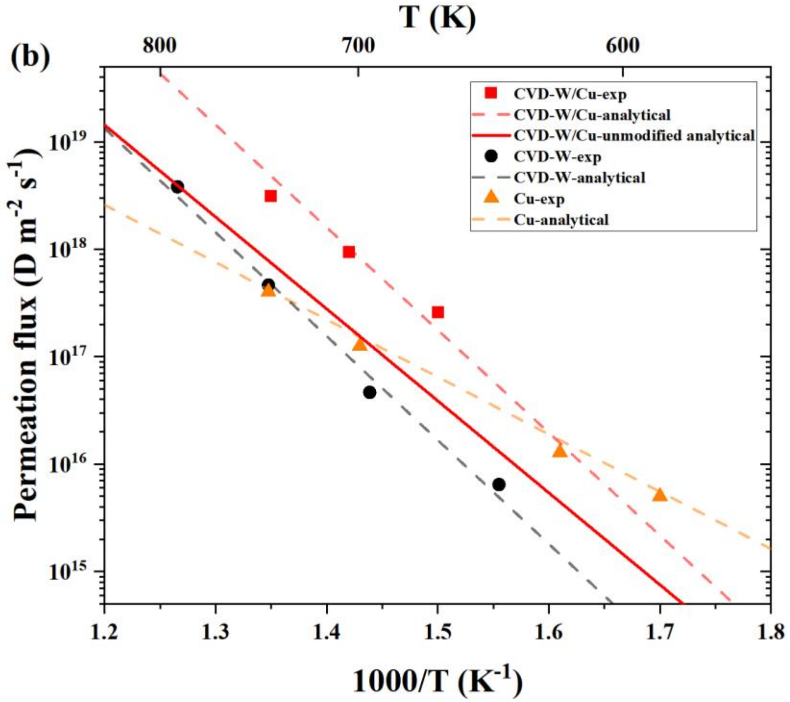
Calculated H solution energies (with zero-point energy) in the W/Cu composite using DFT



- In region I and region II, H atoms tend to dissolve near Cu atoms, corresponding to H solution energies of 0.61 eV and 0.68 eV, respectively, which are lower than the value in the W bulk.
- Actually, Cu atoms in W are possibly in agglomerated states rather than being uniformly distributed. When the number of Cu atoms is less than two, the H solution energy is ~0.68 eV. As the number of aggregated Cu atoms exceeds three, the H solution energy decreases to ≤ 0.23 eV. This reduction arises from the formation of tetrahedral structures composed of two Cu atoms and two W atoms, which enhance H atoms solution.
- The solution energy set in TMAP represents the average value for a segment. The solution energy of 0.6 eV set in the Cu-entry region of W in TMAP accounts for the combination of W, uniformly distributed Cu and the aggregated states of Cu.

Modified analytical solution for steady state permeation flux in multi-layer composite





mass conservation

$$J_0 = J_r + J_p$$

$$J_0 \approx J_r = K_{r1} C_{a1}^2$$

chemical potential continuity

$$\frac{C_{a2}}{S_A} = \frac{C_{b1}}{S_B}, \quad \frac{C_{b2}}{S_B} = \frac{C_{c1}}{S_C}$$

Fick's first law

$$J_A = D_A \frac{C_{a1} - C_{a2}}{L_A}$$

$$J_B = D_B \frac{C_{b1} - C_{b2}}{L_B}$$

$$J_C = D_C \frac{C_{c1} - C_{c2}}{L_C}$$

initial

$$J_p = \frac{D}{L}\sqrt{\frac{J_0}{K_{r1}}}$$

Modified

$$J_p = \frac{1}{\frac{L_A}{D_A S_A} + \frac{L_B}{D_B S_B} + \frac{L_C}{D_C S_C}} \sqrt{\frac{J_0}{K_{r1} S_A^2}}$$

$$J_p = \frac{1}{\sum_{n=1}^{\infty} \frac{L_n}{D_n S_n}} \sqrt{\frac{J_0}{K_{r1} S_A^2}}$$

Summary

- A series of D PDP experiments were conducted on CVD-W/Cu, CVD-W and Cu in the temperature range of ~600 K-800 K.
- Under the identical experimental conditions, an unexpected result was found that the steady state permeation flux in CVD-W/Cu was higher than that in bare Cu. Based on the rate equation simulations and density functional theory calculations, it is suggested that the reduction in H solution energy in the Cu-entry region of CVD-W led to substantial D accumulation near the interface, enhancing the steady state permeation flux in CVD-W/Cu compared to bare Cu.
- Furthermore, an analytical solution for the steady state permeation flux in a generalized three-layer composite was derived using a modified analytical equation for the fast evaluation of steady state permeation flux.

Implications of the work

- The accumulation of HI near the interface could potentially weaken the W/Cu bonding strength, increasing the risk of crack formation at the interface.
- The "realistic" state of interface may lead to modification of tritium transport modelling for ITER.

Thanks for your attention !

Comparison between the experimental and TMAP simulated permeation curves