D retention in bulk Be and D co-deposited in Be layers studied by 3 different thermal desorption techniques and their modelling by CRDS


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Outline

• **Be/D layer** production on W (HiPIMS)

• new Analysis device (FREDIS)

• LID (Laser-Induced Desorption) in FREDIS

• TDS in FREDIS

• Modelling

• **Bulk Be**: D implantation & TDS (ARTOSS)

• Modelling

• Summary & Outlook

• Good news: GO for new Hot Cells in Jülich
Be Layer Deposition in Bucharest

Method: High Power Impulse Magnetron Sputtering (HiPIMS) in D atmosphere by INFLPR

- pulsed magnetron plasma: several MW/m² during 3 µs
- Be layer thickness: 1 µm and 10 µm, new: 20 µm
- D content: 1-30 at% (measured by NRA)
- substrate: 5 mm polished W (IGP) by Plansee with grains elongated perpendicular to the surface
- <3E-6 hPa base pressure, 2E-2 hPa after gas inlet
- sample temperature ≈ 340 K, RT

<table>
<thead>
<tr>
<th>element</th>
<th>small sample</th>
<th>big sample</th>
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</thead>
<tbody>
<tr>
<td>Be (EDX)</td>
<td>70-72 at%</td>
<td>67-69 at%</td>
</tr>
<tr>
<td>D (NRA)</td>
<td>25-27</td>
<td>28-30</td>
</tr>
<tr>
<td>H (LID)</td>
<td>1.1</td>
<td>1.1</td>
</tr>
<tr>
<td>O (EDX)</td>
<td>1.0</td>
<td>1.9</td>
</tr>
<tr>
<td>C (EDX)</td>
<td>1.8</td>
<td>0.6</td>
</tr>
<tr>
<td>N (EDX)</td>
<td>0.4</td>
<td>0.4</td>
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</tbody>
</table>
Microstructure of co-deposited Be/D layer

HiPIMS layers

1.2-1.8 at% D
10 µm

27-30 at% D
1 µm

tokamak layer

layer from JET

Member of the Helmholtz Association
Analysis Device in Jülich: FREDIS
(Fuel Retention Diagnostic Setup)

LID desorption efficiency on 10 µm Be layer with 1.6 at% D

below Be melting:
up to 50% D desorption with a single laser pulse

with Be melting:
>99% D desorption with a single laser pulse

NRA in laser spot centre shows that up to 99% of D is desorbed

for 1 µm Be layer with 30 at% D:
>99% D desorbed by one laser desorption without melting
TDS spectra: 1 µm, 25-30 at.% D vs 10 µm, 1 at.% D

high temperature TDS peak could be the reason for the lower LID efficiency below melting

ultra-sharp TDS jump: instantaneous (within 0.4 s), by factor ~30 only in thick layers (fast heating of 10 µm, all 20 µm) reason: layer detachment ???
TDS spectra: at least 6 TDS peaks observed

- narrow Low Temperature (LT) peak: consists of 2-3 peaks
- small peak 4
- broad High Temperature (HT) peaks: often several peaks

Be/D layer on W
TDS spectra: general observations

Be/D layer on W
Coupled Reaction-Diffusion Systems (CRDS)

The code is similar to TMAP-7, TESSIM, MIMPS and others diffusion-trapping codes

Basic equations:

\[
\frac{\partial_t c_i(x, t)}{D_i(T(t))\frac{\partial^2}{\partial x^2}c_i(x, t)} + \frac{R_i([c], T(t))}{\text{reactions (trapping,etc)}} + \frac{S_i(x, t)}{\text{sources}}
\]

\[
d\sigma = j_{bs} - j_{des} \quad \text{surface coverage}
\]

\[
\frac{\partial x c_H}{x=0} = \frac{j_{bs}}{D(T(t))} \quad \text{boundary condition}
\]

\[
j_{bs} = \gamma \left(1 - \frac{\sigma}{\sigma_{\text{max}}}ight) c_H(0, t) \quad \text{bulk->surf}
\]

\[
j_{des} = 2\kappa \sigma^2 \quad \text{desorption from surface}
\]

Modes of operation:

1. Equivalent to TMAP-7 with either immediate desorption \(c_{|x=0} = 0\) or with molecular desorption flux \(\Gamma = \kappa c_{|x=0}^2\)

2. Accounting for actual surface coverage with saturation effects: \(0 \leq \sigma \leq \sigma_{\text{max}}\)

Further features:
- multiple-occupancy of traps
- trap mobility
Modelling of D release from Be/D co-deposited layers

Sharp low temperature release

\[ s(t) = \frac{1 - \text{erf}\left(\frac{t-t_0}{\tau}\right)}{2} \] switch off re-trapping (e.g. trap annealing)

collective de-trapping, e.g. release from gas bubbles by their opening or percolation through a porous network at threshold temperature \( t_0 \)

High temperature release

- Sample 19 10 \( \mu \)m D/Be=0.1 12 K/min
- CRDS 5 traps w/o re-trapping 12 K/min
- CRDS 2 traps with \( \nu_{\text{dt}} = 4 \times 10^5 \, \text{s}^{-1} \) 12K/min
- Sample 12 20 \( \mu \)m D/Be=0.01 3 K/min
- CRDS 5 traps w/o re-trapping 3 K/min
- CRDS 2 traps with \( \nu_{\text{dt}} = 4 \times 10^5 \, \text{s}^{-1} \) 3 K/min
- CRDS 3 traps ~JET for comparison
Ramp & Hold experiment: nature of low-temperature peaks

release is not governed by de-trapping;
permanent supply of hydrogen [Baldwin et. al: “hydrides”]
alternatively: strongly surface limited release
Material Change

- INFLPR/FREDIS: sputtered Be layers in D gas atmosphere → co-deposited D layers on W

  Material change: Still Be but …

- ARTOSS: Bulk Be (Be SC, Be polycrystal) with implanted D
AROSS (name could mean... “All Relevant Techniques Of Surface Science”)

- Mass & energy separated ion source 0.1-20 kV
- Sputter cleaning ion source, 1-5 kV
- Thermal atomic H source
- Electron beam evaporator
- Accelerator beam: NRA, RBS, ...
- QMS mass spectrometer for TPD, TDS
  - TDS heating by e⁻ beam
- XPS: X-ray source and electrostatic analyser
- Base pressure < 5 \cdot 10^{11} \text{ mbar}
- Beryllium compatible
Beryllium single crystal fluence scan

- High-temperature peak shifting
  - Multitrapping in single vacancies?
  - Depth effect?
- Low-temperature peak splitting
  - LT-peak consists of min. 2 sharp peaks
  - Cannot be explained with Arrhenius release
- Pre-LT-Peak
  - Release of solute Deuterium

implantation: D$_3^+$ (1 keV/D), 1E18 /m$^2$/s
TDS heating rate: 0.01 K/s

Beryllium polycrystal fluence scan

implantation: $D_3^+$ (1 keV/D), 1E18 /m$^2$/s
TDS heating rate: 0.01 K/s

- High-temperature peak shifting
  - Similar behaviour as single crystal
- Low-temperature peak splitting
  - LT-peak consists of min. 3 sharp peaks
    (0.43 eV, 0.67 eV and 0.82 eV)
  - Forms single dominant peak at high fluence
- Pre-LT-Peak
  - Visible as „shoulder“

Compare [Baldwin et. al: “only 1 LT peak”]

Modelling of new ARTOSS data (ongoing)
Defect evolution during implantation is taking into account directly in simulations
Slow down of defect creation (net) and defect saturation are reproduced

Modelling of D retention in crystalline Be

Qualitative agreement
- high temperature peak shift (effect of re-trapping) and shoulder (multiple-trapping)
- low temperature peak threshold (surface occupation after bulk saturation)

CRDS

Experimental data: M. Reinelt, NJP 2009
Hypothesis:

different adsorption surfaces for D
e.g. blisters

Be polycrystal after implantation of $\text{D}_3^+$ at
2 keV/D. Blisters are formed on the surface,
which are partially cracked open on
the top while others are peeled off or in the
process of flaking
Summary

LID (Laser-Induced Desorption within ms):

• for 1 µm HiPIMS Be/D layer with 30 at% D: complete D desorption (i.e. >99%) possible by a single laser pulse without melting
• for 10 µm HiPIMS Be/D layer with 1.6 at% D: up to 50% desorption until melting, nearly complete D desorption (ca. 99%) possible with melting

key for understanding probably in TDS spectra

TDS (slow Thermal Desorption Spectrometry):

• Be: 1st time observation of up to 3 LT subpeaks
• LT peak contribution increases with D concentration/implantation flux
• LT peak very narrow → cannot be simulated by standard diffusion-trapping model
  need to introduce: collective de-trapping, switch off re-trapping, …

• HT peak (multipeak) widens with D layer thickness/D depth
• qualitatively similar TDS spectra for bulk Be (single crystal, polycrystal) and sputtered Be layers
• ultra-sharp peak → ???
Outlook

- Is the laser desorption temperature determined or diffusion dominated?
  → analysis of increasing heating by LID on same position

- **modelling** of TDS and LID ongoing to transfer the results
  - dominant effects
  - sensitivity of desorption on binding energy, diffusion activation energy, …

- new TDS setup: i-TDS (inductive TDS): ca. 2000 K in ca. 1 min; between LID and classical TDS

- LID and TDS of our 1\textsuperscript{st} JET samples with thick Be co-deposited layers in FREDIS
  - tokamak layers behaviour compared to HiPIMS layers (are they reactor relevant?)
  - differences between T and D desorption?

- future application and modelling of LID in JET and ITER
few weeks ago: funding granted for building of new hot cells

for:
• linear plasma device: JULE-PSI
• high heat flux test device: JUDITH-3
• analysis labs, …

to handle activated material and beryllium

Thanks for your attention