IAEA CRP meeting, Vienna, Austria, 26-28.11.2013

Thermal desorption of deuterium from damaged tungsten: experiments and calculations

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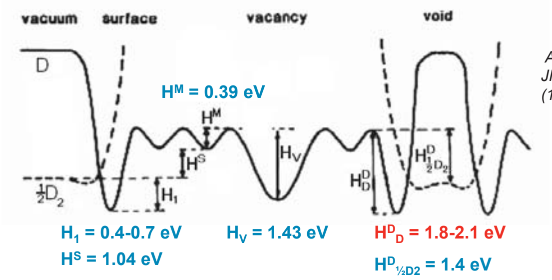
Outline:
- Introduction
- Damage by keV ion beam
- Damage by MeV ion beam (collaboration with IPP)
- Calculations (Diffusion equation)
- DFT calculations

Participants of the project

- **MEPhI**
  - Plasma Physics department (Yu. Gasparyan, A. Pisarev, E. Marenkov, V. Efimov, A. Mednikov)
  - Material Science department (M. Ganchenkova)
- **IPP** (O. Ogorodnikova, M. Mayer, K. Sugiyama)

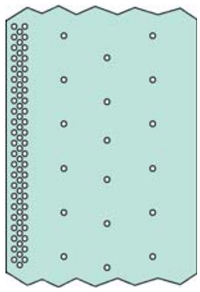
Deuterium retention in tungsten



A. Van Veen et al., JNM, 155-157 (1988) 1113-1117.

- Tungsten has a very low solubility (H_s = 1.04 eV), and rather strong traps!
- **Trapping determine accumulation of hydrogen isotopes in tungsten!**
- Traps: dislocations, grain boundary, vacancy, vacancy clusters, voids.
- One trap can accumulate more than one atom of deuterium.

Ion and neutron damage in W



- **Ion induced defects** are located in the limited area close to the surface with the concentration up to several at.%
- **Neutron induced defects** are distributed deep in the bulk of material

$100\ m^2 \times 1\ \mu m \times 10\%\ n_{trap}/n_{solid} = 3\ g$

$100\ m^2 \times 1\ cm \times 0.1\%\ n_{trap}/n_{solid} = 300\ g$

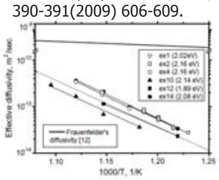
- What kind of traps do neutrons produce? (Difference from ions?)
- Hydrogen transport in damaged W? (Filling rate of traps)
- Evolution of traps with the temperature?

Filling of traps

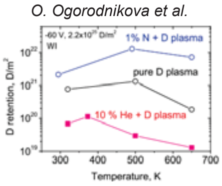
- Traps retard transport of hydrogen in the bulk of material.
The effective diffusivity can be 1-2 orders less than real Frauenfelder's value.
⇒ Even if we have a high concentration of traps in the bulk, the filing rate of these traps can be slow.

- Surface conditions are important for the filling rate! Impurities in the incident flux can significantly change the filing rate of traps.
- Different impurities can both increase and decrease the filling rate and retention

Yu.M.Gasparyan et al., JNM, 390-391(2009) 606-609.



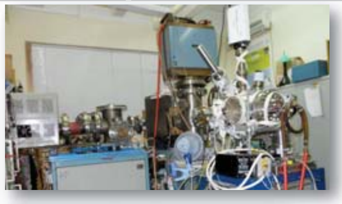
O. Ogorodnikova et al.



•Yu. Gasparyan et al, Journal of Applied Physics 110, 33303 (2011).
• O. V. Ogorodnikova et al., Phys. Scr. T145 (2011) 014034.

TDS facilities in MEPhI

UHV TDS stand
(TDS of externally exposed samples)

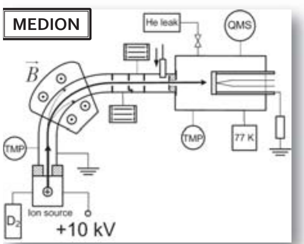


MEDION
(keV ion implantation + *in situ* TDS)

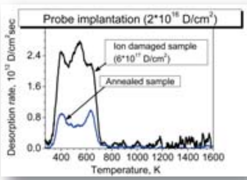
MD-2
(Co-deposition of D with materials + *in situ* TDS)

Damage by keV D_3^+ ions: Probe method

A. Rusinov, Yu. Gasparyan, et al., JNM, 415 (2011) S645-S648.

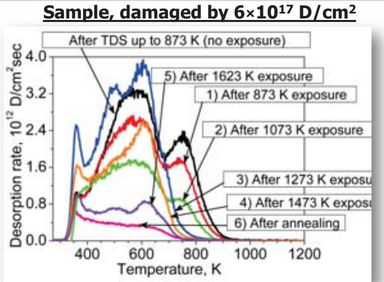


Ion beam: 10 keV/D_3^+, 10^{14} D/cm²sec, Ø3mm
TDS: 2 K/sec, 300K-1750K, ~10 min after irradiation
Probe fluence : 2×10^{16} D/cm²
Probe TDS reflects types and concentrations of defects

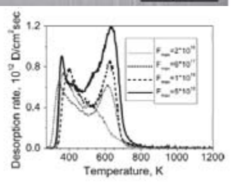


Damage by keV D_3^+ ions

Sample, damaged by 6×10^{17} D/cm²



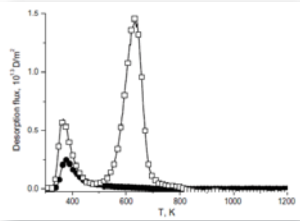
A. Rusinov, Yu. Gasparyan, et al., JNM, 415 (2011) S645-S648

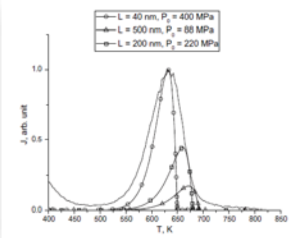


- Clear transformation at 1073-1273 K! (vacancy clusters transforms to bigger voids or single vacancies?)
- At the end of experiments small pores have been observed.

Desorption from pores: Calculations

E.D. Marenkov et al. Nucl. Instr. and Meth. B 269 (2011) 876–880.

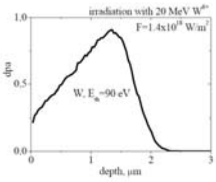


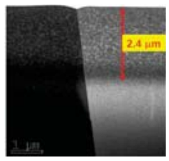


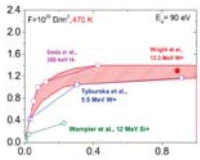
- The peak at 630 K correlated with the presence of pores in the sample.
- This peak can be described by deuterium gas release from pores distributed in the 500 nm thick surface layer with the pressure 90-400 MPa.

Damage by MeV ions (IPP, Garching)

Irradiation with 20 MeV W^{6+} ions





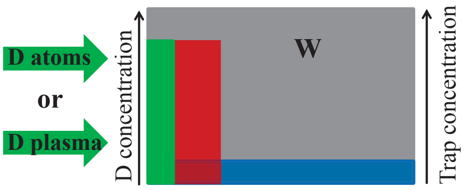


B. Tyburska et al., JNM, 415 (2011) S680-S683.
O. Ogorodnikova et al., JNM, 415 (2011) S661-S666.
L. Ciupinski et al. Nucl. Instr. and Meth. , 317 A (2013) 159

Deuterium filling of traps

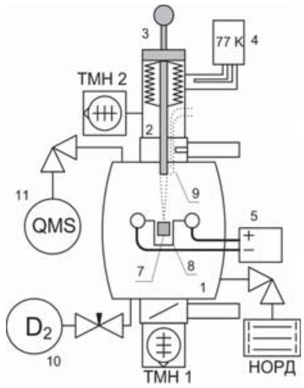
HABS (Ljublana)	PlaQ (Garching) – ECR plasma
Average atomic energy: 0.2 eV/D	Ion energy: 20-200 eV/D
Atomic flux: 3.5·10^{18} D/m²s	Ion flux: (5-10)·10^{19} D/m²s
Temperature: 300-800K	Temperature: 300-800K

S. Markelj et al., JNM, 438 (2013) S1027-S1031.
O. Ogorodnikova et al., JNM, 442 (2013) 518-527.



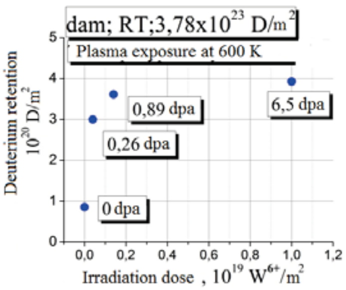
UHV TDS Stand

1 – TDS chamber
2 – Sample exchange chamber
3 – Linear feed through
4 – Nitrogen trap
5 – Power supply
7 – Sample
8 – Heater
9 – Thermocouple
10 – Calibration system
11 – Quadrupole mass-spectrometer

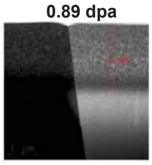


- The base pressure is $<5\times10^{-9}$ mbar
- Sample exchange chamber allows to reduce time of experiments

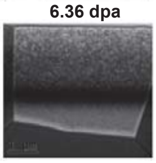
Dependence on dpa level



0.89 dpa



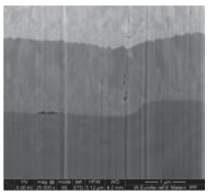
6.36 dpa

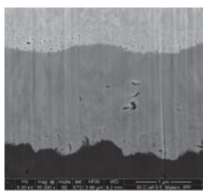


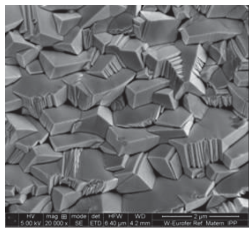
- D retention in damaged W has a trend to saturation at 0.5-1 dpa
- However slow gradual increase is observed till highest dpa, that is in agreement with TEM images

Tungsten coatings

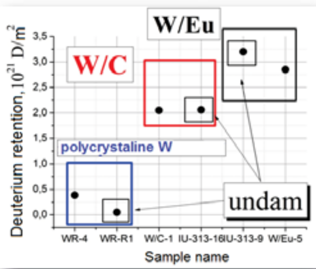
- W coatings on graphite and steel (Eurofer) substrates deposited using the method of combined magnetron sputtering and ion implantation – CMSII.
- Thickness - 10 µm.







W Coatings & Polycrystalline W



Fluence – 2.2x10^{25} D/m²

Temperature – 600 K

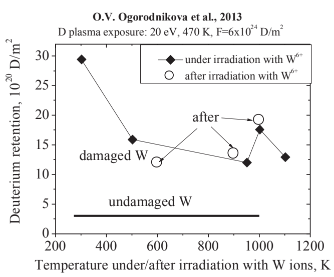
W coatings – 10 µm

Damaged zone – 2.5 µm

- Influence of radiation damages on retention in W coatings is very small, due to presence of many virgin defects.
- Higher retention in W coatings is due to 4 times deeper zone of defects.
- Coatings on Eurofer accumulates more D due to diffusion in to the bulk of substrate.

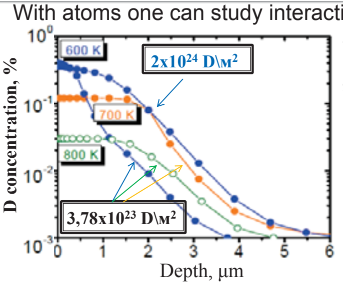
Annealing of radiation damage

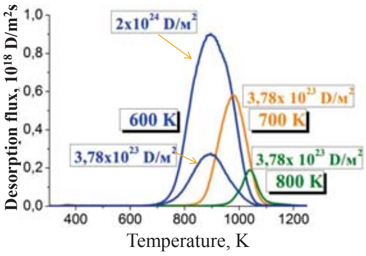
- The D retention reduces intensively in the temperature range between 300 and 700 K.
- Annealing up to 1100 K reduces only partly the density of radiation defects in W and, consequently, the D retention
- **An increase of the D retention at around 1000 K correlates with agglomeration of radiation-induced defects in clusters observed by TEM** (L. Ciupinski et al., 2013) **.**



Atomic exposure

With atoms one can study interaction with original pattern of traps.





- High D retention as in the case of plasma!
- D concentration in traps decreases with the implantation temperature.
- At 700 K and 800 K traps are filled already at the fluence of 3×10^{23} D/m^2.
- One can see the shift of the TDS peak with the increase of exposure temperature.

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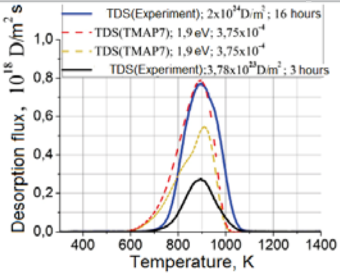
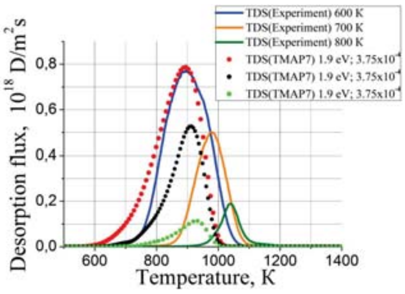
Calculations: 600 K



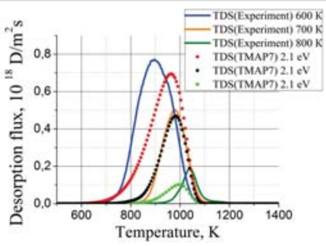
- TMAP 7 code was used.
- Frauenfelder's diffusivity
- Uniform distribution of traps in the damaged region
- Good agreement for high fluence, when traps are filled.
- Dynamics of traps filling depends on recombination coefficient at the surface.
- Even with the highest recombination coefficient the rate of traps filling is faster than in experiment ⇒ some factors reduce the transport rate

Calculations: all spectra



- If one uses the same parameters for higher exposure temperature, the calculated width of peaks becomes more than experimental one
- Peak positions are also shifted

Calculations: all spectra



- To get a better agreement with the experiment at 700 K we used higher energy of traps and inhomogeneous profile
- Probably, defect structure changes with the increase of exposure temperature. Or we use a wrong model?

Calculations: Traps with several places

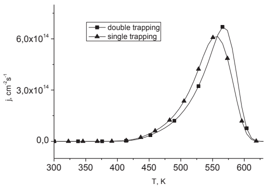
Real defects can trap several atoms. One can add to the model additional processes:

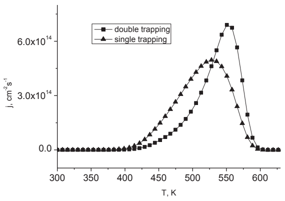
V+H=VH

VH+H=VH_2

$$\frac{\partial C_s}{\partial t} = D\frac{\partial^2 C_s}{\partial x^2} - \frac{\partial}{\partial t}(C_{t1} + C_{t2})$$

First calculations confirm that this assumption makes peaks thinner!



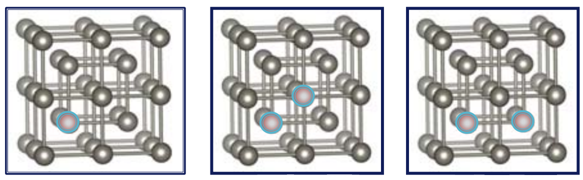


DFT calculations: Effect of strains on radiation defects in W

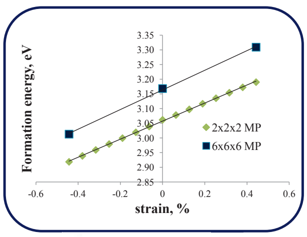
Dr. Maria Ganchenkova

Material department, MEPhI

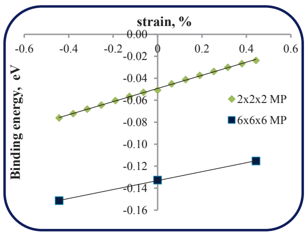
1. DFT based code VASP
2. PAW PBE
3. SCs 250 atoms
4. *k*-points 2x2x2-8x8x8 MP mesh
5. 400 eV of cut-off energy



Vacancies in W: Pressure effect



Formation energy



V2-1NN binding energy

1. Strain conditions considered: isotropic 3D strain within ± 0.44%
2. Energies change linearly within the considered interval of strain values
3. Formation energy varies within ± 0.15 eV
4. Binding energy varies within ± 0.02 eV

Summary

- Radiation damages lead to accumulation of high amount of hydrogen isotopes (up to ~1 at.%).
- Different W materials has similar maximum concentration of traps. In the case of high initial trap concentration the effect of radiation on retention is smaller.
- Both keV and MeV ion irradiation produce a high concentration of defects with the high detrapping energy (1.8-2.2 eV), which determine retention at high temperatures. These defects are, very likely, vacancy clusters.
- At 1000-1200 K vacancy clusters has transformation to bigger size clusters (voids). Large scale pores can be formed which are stable at very high temperatures.
- The filling rate of defects depends strongly on impurities on the surface and in the incident flux.