Multi-method approach towards a detailed understanding of Be for fusion

13th August 2014 | M. Köppen
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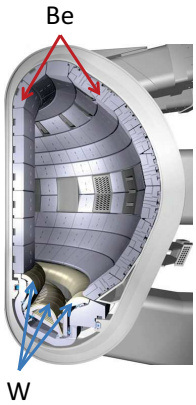
Outline

1. Motivation
2. Surface experiments and methods
   - General approach
   - Experimental capabilities with examples
   - Planned experiments II
   - Work plan
3. Laser-based techniques
4. High heat flux experiments on beyllium and beryllium components
   - Steady state loads
   - Transient heat loads

Motivation

- ITER material mix: Be and W
  - 700 m² Be
  - W in divertor region
- Plasma impurities
  - Seeding gas, e.g. Nitrogen
  - Contaminations, e.g. Oxygen
- Material transport by:
  - Erosion
  - Plasma transport
  - Re-deposition



Why is compound formation of interest?

- Elevated temperatures and ion irradiation lead to solid state reactions
  - e.g. Be_2W alloy formation in divertor region
- Be-based Mixed Materials have new properties compared to Be:
  - Isolating / different conductivity
  - Altered Hydrogen retention
  - Co-deposits
  - Ammonia formation
- Physical and Chemical Data needed as input data for e.g. ERO, SOLPS, WallDyn to predict first wall properties for future devices

	Be	BeO	Be_3N_2	W	WO_2	WO_3
Melting Point [K]	1551	2780	2473	3683	1773	1746
$\Delta_f H°$ [kJ mol^{-1}]	--	-609	-558	--	-590	-843

Surface experiments and methods for Be and Be-based mixed materials

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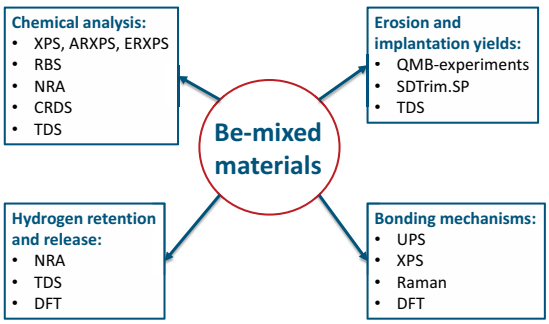
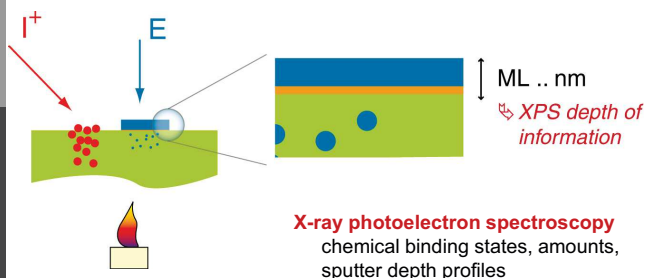
Multi-method approach: Solutions



Experimental approach



X-ray photoelectron spectroscopy
chemical binding states, amounts, sputter depth profiles
synchrotron: chemical depth profiles

Accelerator-based techniques
amounts, depth profiles
hydrogen isotope analysis (amounts, profiles)

Modeling approach: Description of chemistry
The CRDS-model

1. Define all substances

Be
W
C
Be_2C
W_2C
WC
Be_2W
$Be_{12}W$
Be_{gas}
BeO
O_{ads}
WO_3
$WO_{3,gas}$

+ Initial areal densities [#/m²]

2. Elementary reactions

$2\,Be + C \rightarrow Be_2C$
$Be_2C \rightarrow 2\,Be + C$
$W + C \rightarrow WC$
$WC \rightarrow W + C$
$2\,W + C \rightarrow W_2C$
$W_2C \rightarrow 2\,W + C$
$W + 2\,Be \rightarrow Be_2W$
$Be_2W \rightarrow W + 2\,Be$
$W_2C \rightarrow WC + W$
$WC + W \rightarrow W_2C$
$Be + O \rightarrow BeO$
$BeO \rightarrow Be + O$
$W + 3\,O \rightarrow WO_3$
$WO_3 \rightarrow W + 3\,O$

Sublimation:
$Be \rightarrow Be_{gas}$
$WO_3 \rightarrow WO_{3,gas}$

O-Adsorption:
$O_{2,gas} \rightarrow O_{ads}$
$O_{ads} \rightarrow O_{2,gas}$

3. Define equations for reaction fluxes

$$\Gamma_1\left[\frac{1}{m^2 s}\right] = [Be]^2[C]k_1 \exp\frac{-\Delta E_1}{kT} \quad \text{with} \quad k_1\left[\frac{m^4}{s}\right]$$

$$\Gamma_2\left[\frac{1}{m^2 s}\right] = [Be_2C]k_2 \exp\frac{-\Delta E_2}{kT} \quad \text{with} \quad k_2\left[\frac{1}{s}\right]$$

...

4. Define balances

Change of areal density of substance =
+ all formation reaction fluxes
− all destruction reaction fluxes
± all "non-chemical" fluxes

$$\frac{d[Be]}{dt} = -2\Gamma_1 + 2\Gamma_2 + \ldots \qquad \frac{d[Be_2C]}{dt} = +\Gamma_1 - \Gamma_2 + \ldots$$

→ Set of ordinary differential equations

$$y'(t) = f(t, y(t)), \quad \text{with initial value} \quad y(t_0) = y_0$$

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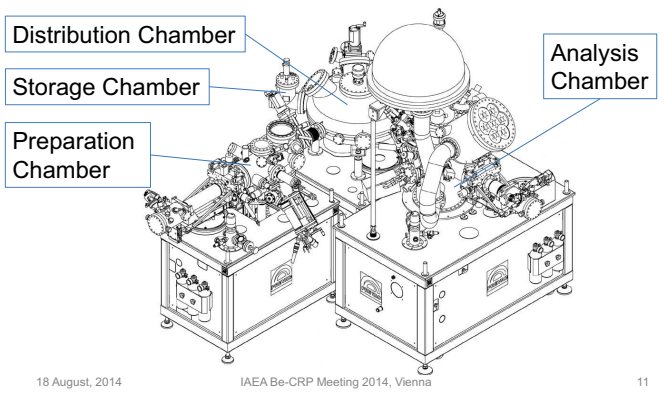
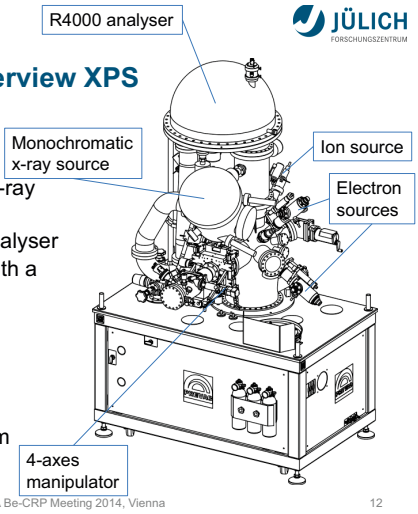
Experiments: Overview XPS



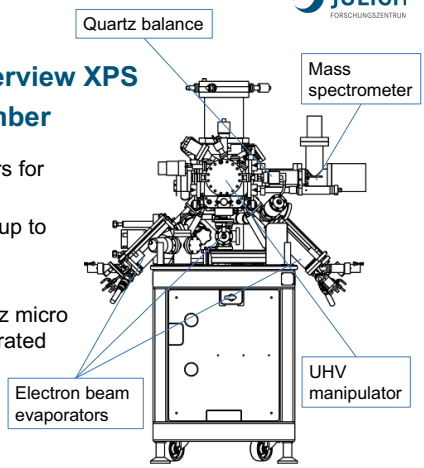
Experiments: Overview XPS
Main chamber

- XPS
  - Monochromatic x-ray source
  - Hemispherical analyser
- 2 ion sources, one with a Wien Mass Filter
- 2 electron sources
- UV source
- Sample size:
  - heating: 2,5 cm
  - Non-heating: 5 cm



Experiments: Overview XPS
Preparation Chamber

- 3 electron evaporators for W, C, Be
- Manipulator, heating up to 2500 °C
- Mass spectrometer
- High-resolution quartz micro balance can be integrated



Experiment: Methods in XPS

Be-mixed materials

Chemical analysis:
- XPS, ARXPS, ERXPS
- RBS
- NRA
- CRDS
- TDS

Erosion and implantation yields:
- QMB-experiments
- SDTrim.SP
- TDS

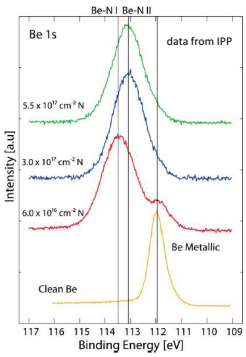
Hydrogen retention and release:
- NRA
- TDS
- DFT

Bonding mechanisms:
- UPS
- XPS
- Raman
- DFT

Example X-ray photoelectron spectroscopy: N-Implantation in Be

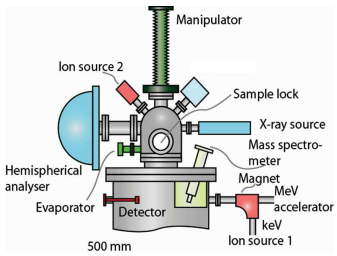
- Implantation experiment of nitrogen ions into clean beryllium at different fluences up to 5.5 x 10^{17} cm^{-2}
- Deconvolution of spectra shows three species:
  - Metallic Be
  - Be-N-phase II at 113.1 eV
  - Be-N-phase I at 113.8 eV
- Be-N-Phase I is an intermediate compound at low fluences
- Be-N-Phase II is a metastable compound

N-fluence [10^{17} cm^{-2}]	Be 111.8 eV	Be-N I 113.1 eV	Be-N II 113.8 eV
0.6	15.1 %	54.5 %	30.4 %
3.0	3.4 %	4.7 %	91.9 %
5.5	2.2 %	3.1 %	94.7 %



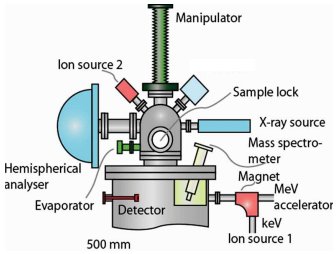
ARTOSS

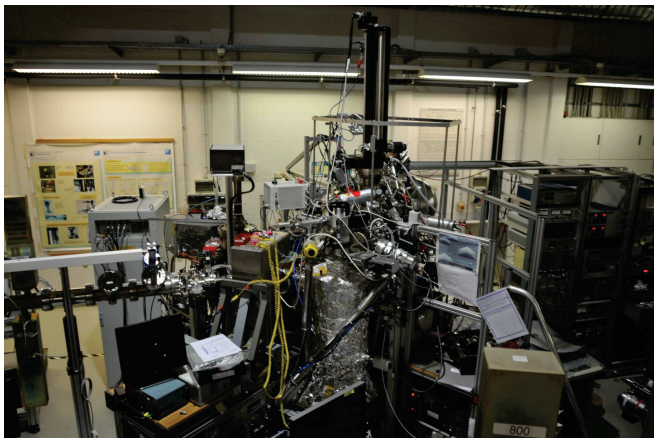
- Ion beam analysis
  - Nuclear reaction analysis (NRA)
  - Rutherford backscattering spectroscopy (RBS)
- Mass-spectrometer for thermal desorption spectroscopy (TDS)
- Standard-X-ray Source



ARTOSS

- Mass-seperated low energy ion source (0.1-10 keV)
- Thermal atomic H-source
- Base Pressure: < 5 x 10^{-11} mbar
- Electron beam evaporator





Experiment: Methods in ARTOSS

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Erosion and implantation yields:
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Hydrogen retention and release:
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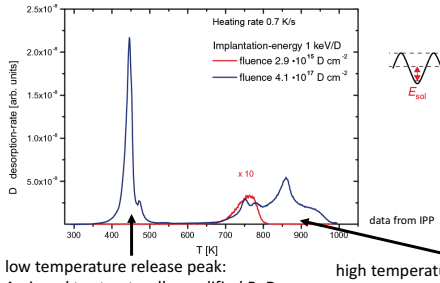
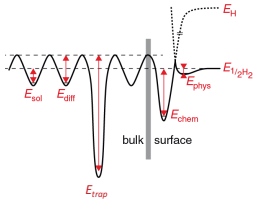
- DFT

Bonding mechanisms:
- UPS
- XPS
- Raman
- DFT

Example: Thermal desorption spectroscopy of D in Beryllium

Deuterium-release and retention of Be(0001)



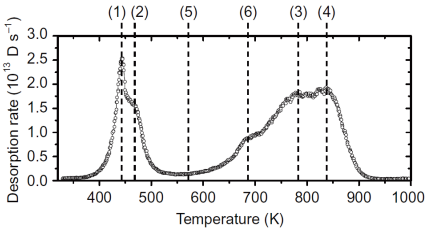


low temperature release peak:
Assigned to structurally modified BeD_2
From DFT: BeD_{2-x}

high temperature release peak:
Retention in ion induced defects

TPD of D implanted Be

Full TPD spectra of D implanted in Be



sc Be (11$\bar{2}$0)

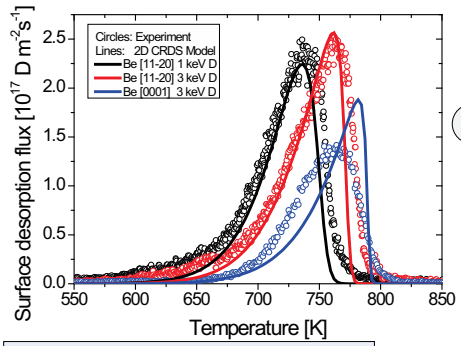
1 keV/D
$\Phi = 2\times10^{17}$ cm^{-2}

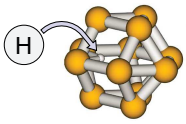
TDS 1K/s

Peaks 1 and 2 – structural changes (over-saturation with D, fluence threshold)
Peaks 3 and 4 – trap sites produced by collision cascades
Peak 5 – hydride formation (BeD_2), decomposition at ~500-600K
Peak 6 – release from an oxidized Be surface layer

M. Reinelt, NJP 11 (2009) 043023

D-release anisotropy at low fluences on Be single crystals





H trapped in a monovacancy DFT-calculation by A. Allouche

temperature shifts in D release peaks due to
- Different implantation depth
- Different crystal orientation (release ∥ and ⊥ to Be planes)
- Retained amount of D smaller for Be(0001)

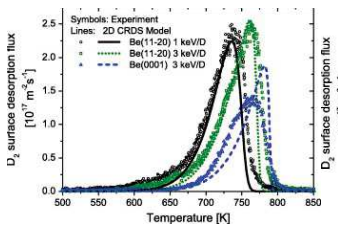
Fluences:
Be(11-20) 1 keV/D: 2.7× 10^{15}cm^{-2}
Be(11-20) 3 kev/D: 2.9× 10^{15}cm^{-2}
Be(0001) 3 kev/D: 2.7× 10^{15}cm^{-2}

Thanks to M. Reinelt and M. Oberkofler

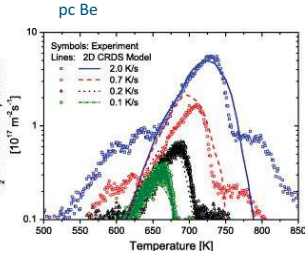
Modelling of D retention: CRDS

- Assumption: desorption diffusion limited !
- Flux at both side surfaces of the sample: $\Gamma_{TPD} = D_H^z \frac{\partial\rho((z=z_0, z_{max}), x, t)}{\partial z}$

sc Be, different orientations



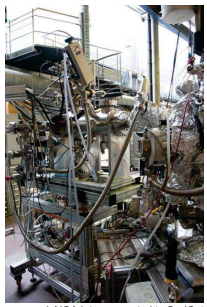
pc Be



R. Piechoczek, JNM 438 (2013) S1072

Experiment: LAICA

- Experiments at HZB-BESSY II at "SurlCat"
- Preparation chamber "LAICA" is interconnected
  - 3 Evaporators: Be, C and W
  - Heater for temperatures up to 1370 K
  - 5 kV Ion gun for implantation and sputtering
  - Auger system
  - Base pressure: 5 x 10^{-10} mbar
- ERXPS mesurements in SurlCat
  - Excitation energies from 30 to 1300 eV
  - Base pressure: < 1 x 10^{-10} mbar



LAICA interconnected to SurlCat

Experiment: Methods in LAICA

Be-mixed materials

Chemical analysis:
- XPS, ARXPS, ERXPS
- RBS
- NRA
- CRDS
- TDS

Erosion and implantation yields:
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Hydrogen retention and release:
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- DFT

Bonding mechanisms:
- UPS
- XPS
- Raman
- DFT

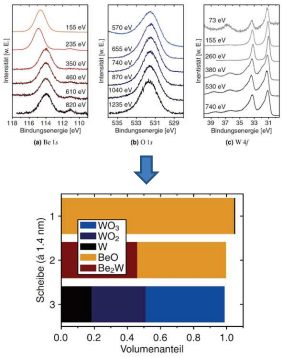
Example: Energy-resolved XPS at synchrotron

- Information depth depends on the kinetic energy of the photoelectron
- The kinetic energy depends on the energy of the photon

⇒ Variation of photon energy varies information depth

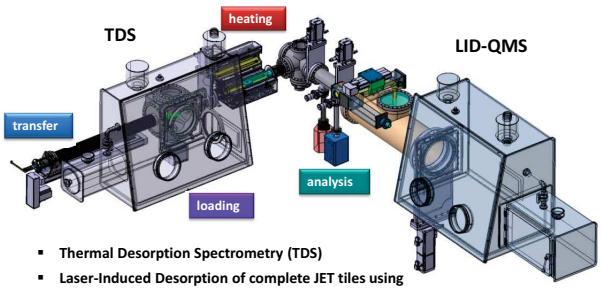
⇒ Information depths of different core levels is equal

⇒ **Allows chemical depth-profiling of the first nanometers**



Versatile beryllium analysis facility

Design and construction of a JET-oriented beryllium analysis facility (inside the controlled area of HML but outside the hot cell)



- **Thermal Desorption Spectrometry (TDS)**
- **Laser-Induced Desorption of complete JET tiles using Nd:YAG laser**
- **Surface composition analysis by glow discharge spectroscopy**

Already operational

Will be moved to hot cells asap

Experiment: Methods in LID/QMS

Be-mixed materials

Chemical analysis:
- XPS, ARXPS, ERXPS
- RBS
- NRA
- CRDS
- TDS
- LIDS

Erosion and implantation yields:
- QMB-experiments
- SDTrim.SP
- TDS

Hydrogen retention and release:
- NRA
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Bonding mechanisms:
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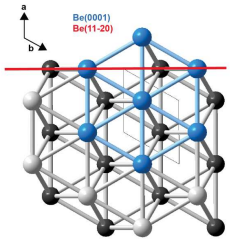
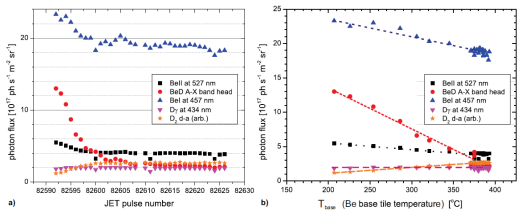
Isotope exchange

- few experimental data on H isotope exchange in Be
- Beryllium has **not an "ideal" hcp** structure. Be-Be distances are closer in <0001> than perpendicular to it (e.g. <11-20>) → different diffusion constants
- What role does dissociation play

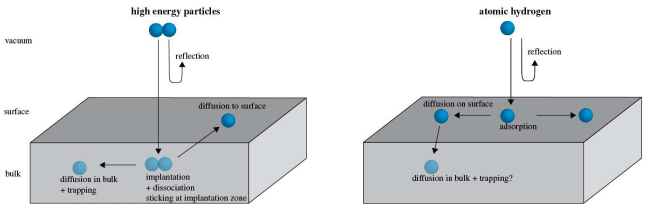


Confirmation of BeD_x species

- Observation of specific BeD line (OES) in JET, PISCES ...
- Predicted by calculations (C. Björkas 2009)
- Existence not yet proven in laboratory experiments
- sputtering of BeD or BeD_2



Planned experiments: ion vs. atom



- Until now only experiments with D ion implantation performed → trapping of D in trap-sites produced during implantation
- Molecular hydrogen does not adsorb on Be → atomic hydrogen needed
- *Open questions:*
  - Temperature dependency (absorption barrier)?
  - Retention in intrinsic defect sites?
  - Does He/N etc. ion bombardment create additional trap-sites for D in Be?

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Work Plan

- Investigation of the system Be-N
- Investigation of hydrogen retention and release behaviour of Be-N
- Investigation oxidation behaviour of Be-N
- Experiments on the interaction of He-ions with Be-N
- Isotope exchange in Be-based mixed materials
- Understanding BeD_x-formation and properties
- Investigation of different behaviour of atomic and ionic hydrogen

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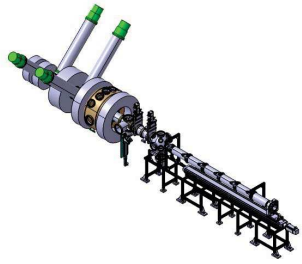
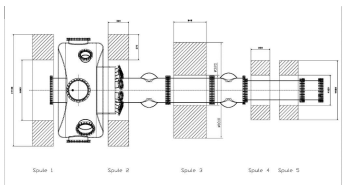
Laser-based techniques

Conceptual design of
JULE-PSI linear plasma device

- **Steady state linear plasma generator (B = 0.2 T) with target exchange and analysis chamber, inside Hot Cell**
- **Loading conditions (deuterium plasmas), with target biasing**
  - $\Delta_{flow\ channel}$ ~ 6 cm
  - $n_e = 10^{17} - 10^{19}\ m^{-3}$
  - T_e up to 20 eV
  - E_{ion} = 10-200 eV (biasing)
  - $\Gamma_{ion} = 10^{21} - 10^{23}\ m^{-2}s^{-1}$
  - Fluence: $10^{27}\ m^{-2}$ in 4 h
  - q = 0.1 - 2 MW m^{-2}, simulation of transients by laser irradiation (40 J / 1ms)
- **Procurement of components is ongoing**

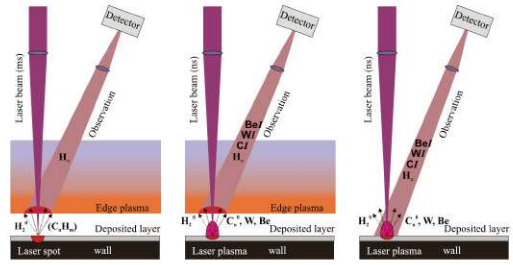




Laser-based surface analysis overview

Laser-based diagnostics:

- Laser-induced desorption spectroscopy (LIDS),
- Laser-induced ablation spectroscopy (LIAS) and
- Laser-induced breakdown spectroscopy (LIBS)



a) LIDS — Idea: only fuel desorption, no material ablation

b) LIAS — Material ablation

c) LIBS — Material ablation

Future Program

- Time resolved physics of LIBS plasma
- Investigation of LIBS plasma physics under:
  - vacuum
  - gaseous (Ar, N, He) environments
  - magnetic field
- Impact of the laser wavelength on the LIBS plasma
  - Combination of two laser beams (Dual LIBS)
- Ablation physics: Energy distribution (ToF), species distribution, formation of cluster, reproducibility (particles/pulse),
- Modelling of the laser-material interaction

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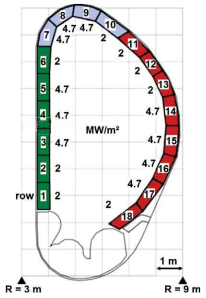
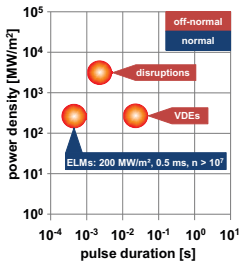
High heat flux tests on Be-components

ITER FW power loads



Steady state power load capability for different first wall rows



Expected transient power loads (off-normal events have to be considered and tested as well)

[1] R. Mitteau et al., Fusion Eng. Des. 88 (2013), pp. 568–570

[2] M. Merola, et al., Fusion Eng. Des. (2014), http://dx.doi.org/10.1016/j.fusengdes.2014.01.055

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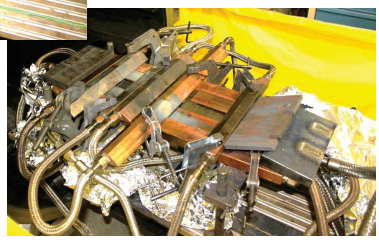
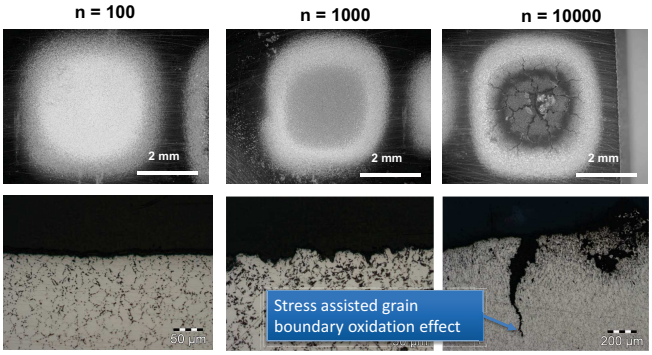
Assembly of the mock-up in JUDITH 2

- 2 mock-ups are tested at once
- For the thermal insulation both mock-ups are fixed with sharpened screws
- Each mock-up is equipped with 2 thermo-couples

- For the protection of the base plate and mock-up holder beam-dumps are used



Repeated thermal shocks on beryllium



Energy density: $E = 1.0\ MJ/m^2$ Pulse duration: $\Delta t = 5\ ms$
Base temperature: $T_0 = 250°C$ Heat flux factor: $P \cdot \sqrt{(\Delta t)} = 14\ MW/m^2 s^{1/2}$

Outline

1. Motivation
2. Surface experiments and methods
   - General approach
   - Experimental capabilities with examples
   - Planned experiments II
   - Work plan
3. Laser-based techinques
4. High heat flux experiments on beryllium and beryllium components
   - Steady state loads
   - Transient heat loads

High heat flux test facility JUDITH 1

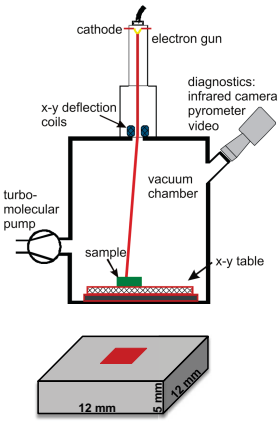
Electron beam facility JUDITH 1 is used to simulate transient high heat loads

Electron beam parameters

max. power:	60 kW
acc. Voltage:	120 kV
pulse duration:	≥ 1 ms
beam diameter:	~ 1 mm

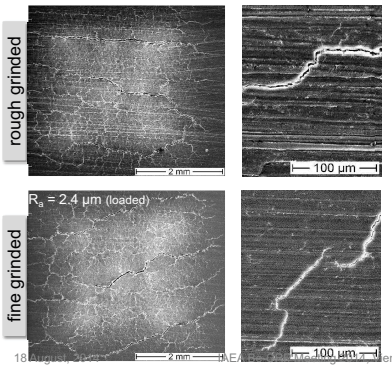
ELM-like loading conditions

power density:	200 - 800 MW/m²
pulse duration:	1 ms
pulse number:	up to 1000



Surface finish influence

abs. power density: 0.8 GW/m²
pulse length: 1 ms, pulse number: 100



No clear coincidence of cracks and lathe faced cavities observable

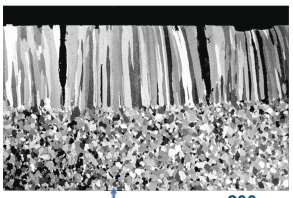
R_a [µm]	unloaded	loaded
rough	0.7	2.6
fine	0.4	2.4

Cracks might coincide with grain orientation/microstructure
➡ Further investigations valuable

Thermal shock loading of beryllium

E_{abs} = 5.2 MJm^{-2}, Δt = 5 ms, T_0 = 20° C (to simulate plasma disruption)

S65C



Hot pressed in vacuum

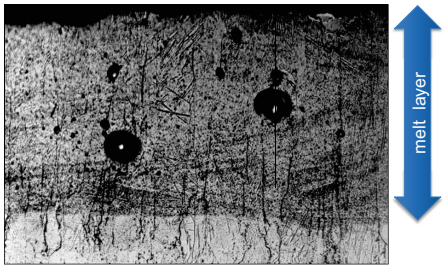
TR-30

Punched, relative high oxygen content

J. Linke et al, 3rd IEA Intern. Workshop on Be Techn. for Fusion, 1997.

Thermal shock testing of neutron-irradiated Be samples

Neutron irradiation:	T_{irr} = 700°C, 0.35 dpa
Electron beam loading:	E_{inc} = 15 MJm^{-2}, Δt = 5 ms, n = 5



M. Rödig et al, J. of Nuclear Materials, 2002.

- Beryllium is transmuted into tritium and helium during irradiation
- Pore formation during electron beam loading of neutron irradiated beryllium
- He concentration is 55 ppm