Radiation induced formation gas bubbles in beryllium after neutron irradiation up to 6000 appm helium production

Michael Klimenkov, Ute Jäntsch, Vladimir Chakin, Nikolai Zimber and Pavel Vladimirov

Karlsruhe Institut of Technology, Hermann-von-Helmholtz-Platz 1 76344 Eggenstein-Leopoldshafen, Germany

The current interest in mechanical properties and microstructure of neutron irradiated beryllium refers to its planned application in the Helium-Cooled Pebble Bed (HCPB) European concept of a breeding blanket of DEMO. Irradiation experiments in high-neutron flux nuclear research reactors yield information about microstructural evolution of beryllium under conditions relevant to fusion (temperature, damage dose, helium and tritium productions) excluding 14 MeV neutrons impact which is not present in the neutron spectra of fission reactors. The HIDOBE-02 irradiation campaign accomplished at the HFR, Petten corresponds to 1246.5 Full Power Days at a reactor power level of 45 MW in the temperature range from 410°C to 680 °C. Transmission electron microscopy (TEM) has been to study the evolution of voids during neutron irradiation at different temperatures. The target preparation of specimens was performed using focused ion beam (FIB).

TEM study shows the formation of radiation induced hexagonal flat gas bubbles inside the grains, however at the lowest irradiation temperature of 410° the pebbles show the uniform shape. The diameters of the bubbles increase from a few nanometers for 410°C to more than hundred nanometers for 680 °C. The number density of bubbles decreases, correspondingly, by more than two orders of magnitude. The preferable formation of bubbles along the grain boundary and dislocation lines was observed. Analytical investigations using electron energy loss spectroscopy show the presence of He and H23 inside bubbles. Also the Si and Fe segregation on the voids was detected [2].

EDX mapping shows that the precipitates inside the grains and on the GBs have increased iron and aluminum content, indicating the formation of an Fe-Al-Be phase. In the material irradiated at 440°C, most of the precipitates also have Fe-Al-Be composition, while several other single- and multiphase precipitates were found. The Fe-Al-Be phase is observed as 10-15 nm precipitates within the grains and as 200 nm particles bound to a gas bubble at the GB. The present study shows detailed microstructural changes induced by neutron irradiation in beryllium.

[1] M. Klimenkov, et al. Journal of Nuclear Materials 455 (2014) 660–664
[2] M. Dürrschnabel, et al. Scientific Reports, 11, 7572 (2021)

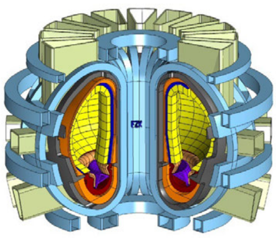
Corresponding Author:
Dr. Michael Klimenkov
michael.klimenkov@kit.edu
Karlsruhe Institut of Technology, Hermann-von-Helmholtz-Platz
1 76344 Eggenstein-Leopoldshafen, Germany

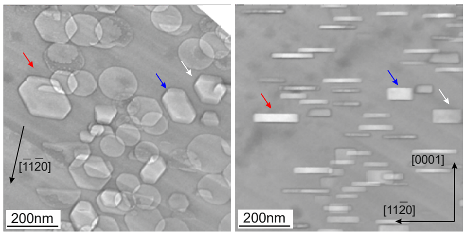
Karlsruhe Institute of Technology

Radiation induced formation gas bubbles in beryllium after neutron irradiation up to 6000 appm helium production

M. Klimenkov, **P. Vladimirov, N. Zimber, U. Jäntsch and A. Möslang**

Institute for Applied Materials - Applied Materials Physics, Karlsruhe Institute of Technology (KIT), Karlsruhe, Germany.





KIT – The Research University in the Helmholtz Association

www.kit.edu

Content

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- Introduction
- Microstructural examination of neutron irradiated beryllium at IAM-AWP (HIDOBE I and HIDOBE II)
- Detection of He and ^{3}H inside bubbles
- Distribution of impurity atoms in irradiated Be
- Conclusions

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Beryllium application in fusion technology

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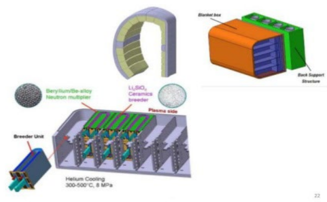
- Application as a "First Wall" material in ITER.
- As neutron multiplier material in different tritium-breeding blanket concepts for the future demonstration fusion power plant DEMO.

Helium Cooled Pebble Bed Blanket



Production of helium and tritium (^3H) by transmutation reactions

$^9Be + n \rightarrow 2\,^4He + 2n$ (E_n>2.7 MeV)

$^9Be + n \rightarrow {^7Li} + {^3He}$ (E_n>10.7 MeV)

$^7Li + n \rightarrow {^4He} + {^3H} + n$

$^9Be + n \rightarrow {^6He} + {^3H}$ (E_n> 0.6 MeV)

$^6He \rightarrow {^6Li} + b^- + \nu$

$^6Li + n \rightarrow {^4He} + {^3H}$

Prediction of irradiation resistance of beryllium pebbles under close-to-fusion conditions.

- operation temperature,
- accumulated damage dose,
- amount of helium and tritium generated by neutron-induced transmutation.

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Neutron irradiation programs 2005 - 2016

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High Dose Beryllium irradiation program **(HIDOBE-I) (2005-2007)**

High-Flux Reactor (HFR), Petten, Netherlands

Dose 18 dpa, 3000 appm ^{4}He, **300** appm ^{3}H

Irradiation temperatures: 410°C, 480°C, 590°C, 700°C

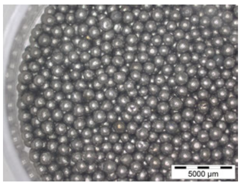
High Dose Beryllium irradiation program **(HIDOBE-II) (2005-2011)**

High-Flux Reactor (HFR), Petten, Netherlands

38 dpa displacement per atom **5900** appm ^{4}He, **640** appm ^{3}H

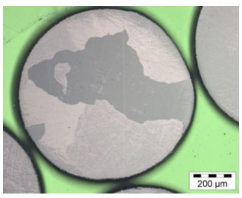
Irradiation temperatures: 370°C, 440°C, 540°C, 650°C

Be pebbles



∅ 1mm
∅ 2mm

1mm pebble



100-300µm grain size

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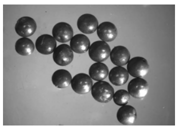
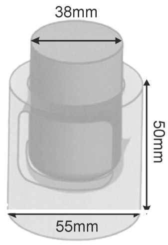
HIDOBE-I – TEM sample preparation

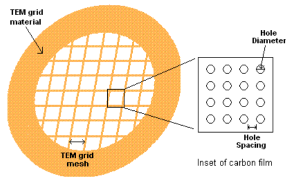
Irradiated beryllium pebbles

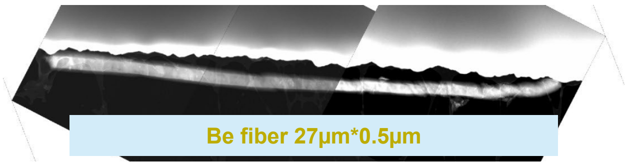


crushing tool for preparation of Be powder

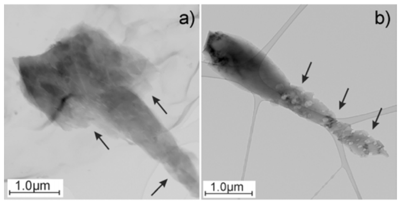


After grinding of Be pebble small pieces were deposited on the copper grid covered by carbon film.





TEM images of small powder particles



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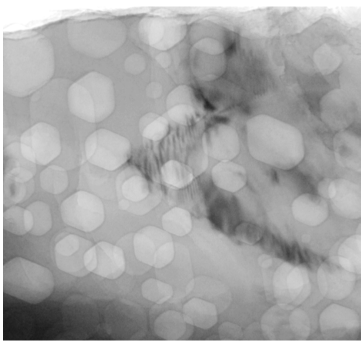
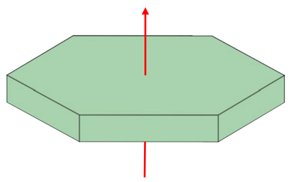
HIDOBE-I post irradiation examination

Bubbles – hexagonal shape



Bubbles – rectangular shape

c-axis



prismatic-disc shape of bubbles in beryllium

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HIDOBE-I - post irradiation examination

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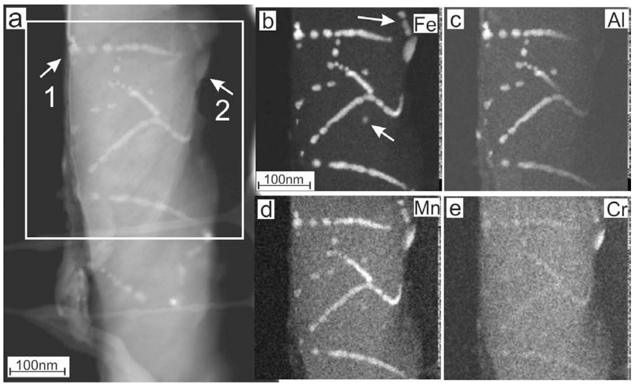
Pecipitates 480°C



The FeAlMnCr pecipitates located in a fiber of 350 nm thickness.

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HIDOBE-I post irradiation examination

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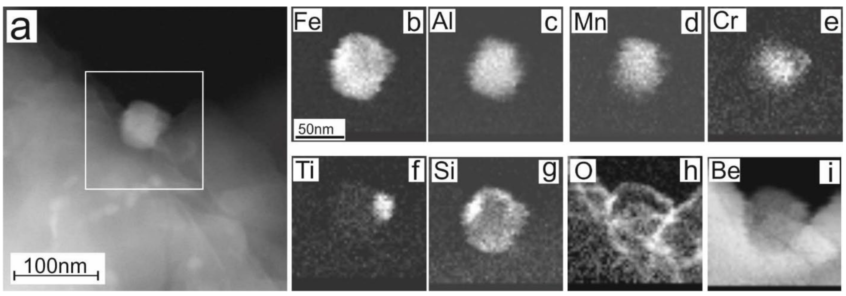
Precipitates contains of FeAlMnCrTi beryllides



- M.Klimenkov, V.Chakin, A.Moeslang, R.Rolli, Journal of Nuclear Materials 443 (2013) 409-416
- M.Klimenkov, V.Chakin, A.Moeslang, R.Rolli, Journal of Nuclear Materials 455 (2014) 660–664

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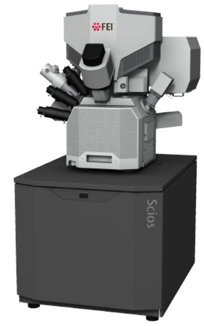
HIDOBE-II

High Dose Beryllium irradiation program **(HIDOBE-II)** **(2005-2011)**
High-Flux Reactor (HFR), Petten, Netherlands
38 dpa displacement per atom **5900** appm ^{4}He, **640** appm ^{3}H
Irradiation temperatures: 370°C, 440°C, 540°C, 650°C

Microstructural examination:
2016-2021



TEM lamellae were prepared from irradiated tungsten using focused ion beam (FEI SCIOS) in the FML at KIT.

Transmission Electron Microscope
Talos F200X G2 / 200 kV FEG

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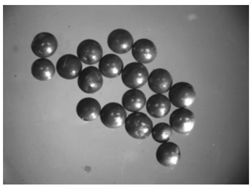
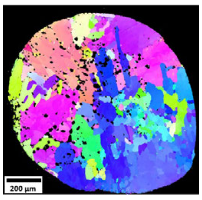
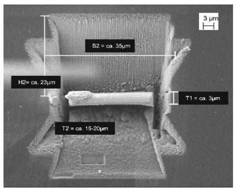
Specimen preparation using Foccused Ion Beam

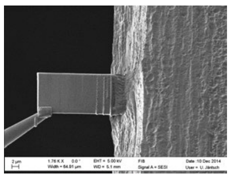


Removal of larger amounts of surrounding material using Ga ion beam.

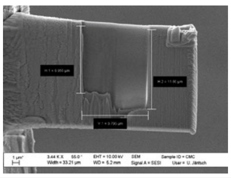


Deposition of prepared lamellae on the Cu greed

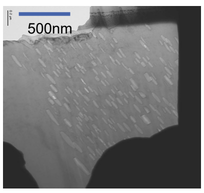




Preparation of thin transparent window for TEM analysis



The thickness varried from 120 to 350nm mean path in beryllium ≈0,6-2.1λ



lamella is ready for TEM analysis

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Microstructure of irradiated beryllium

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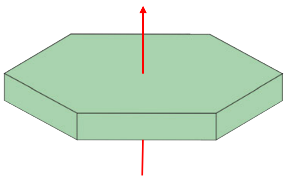
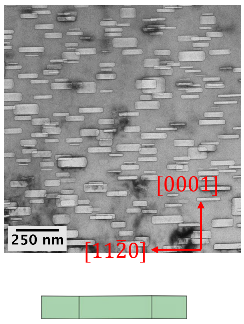
- Bubbles show hexagonal prismatic-disc shape in the transmission electron microscope (TEM)

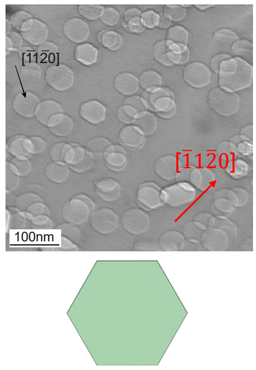
c-axis



prismatic-disc shape of bubbles in beryllium



view on prismatic face



view on basal face

Tomography in TEM

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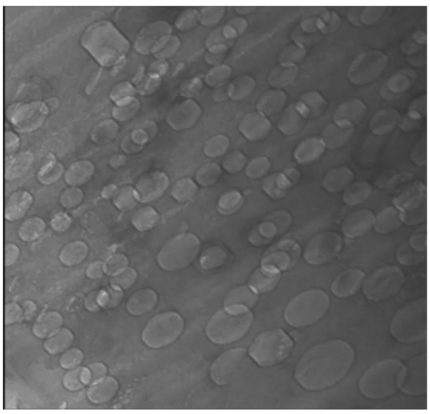
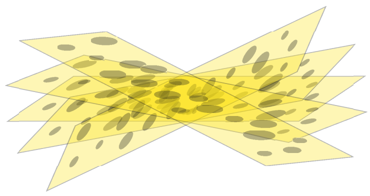
66° total tilt (from -32° to +34°)

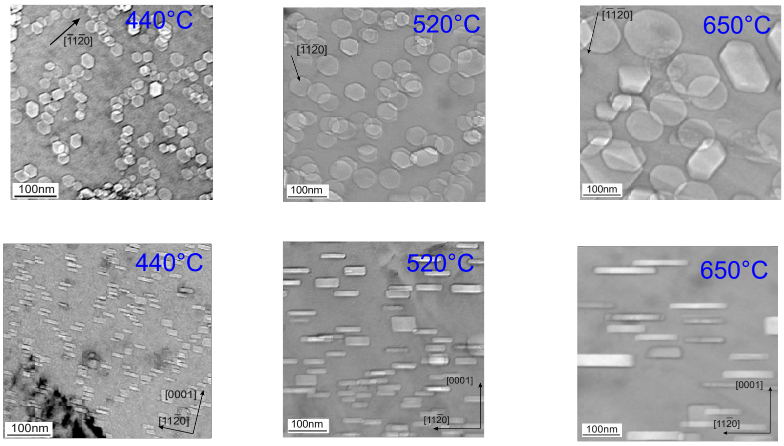


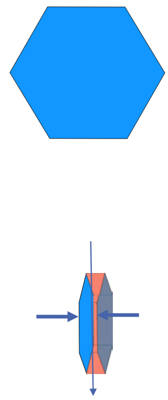


step ~3°
23 images

Bubbles in beryllium irradiated at different temperatures

Images of the bubbles in the basal and prismatic orientations





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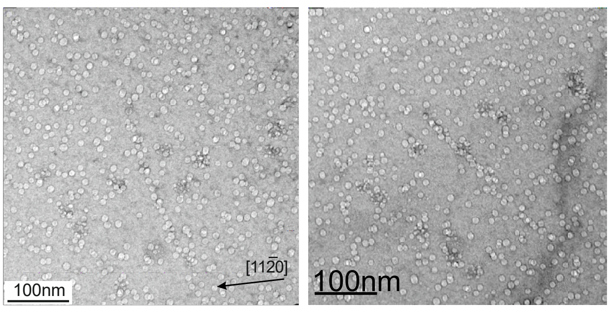
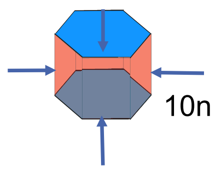
Bubbles in beryllium irradiated at 370°C

Tilt of 60° between these images



A uniform bubble shape



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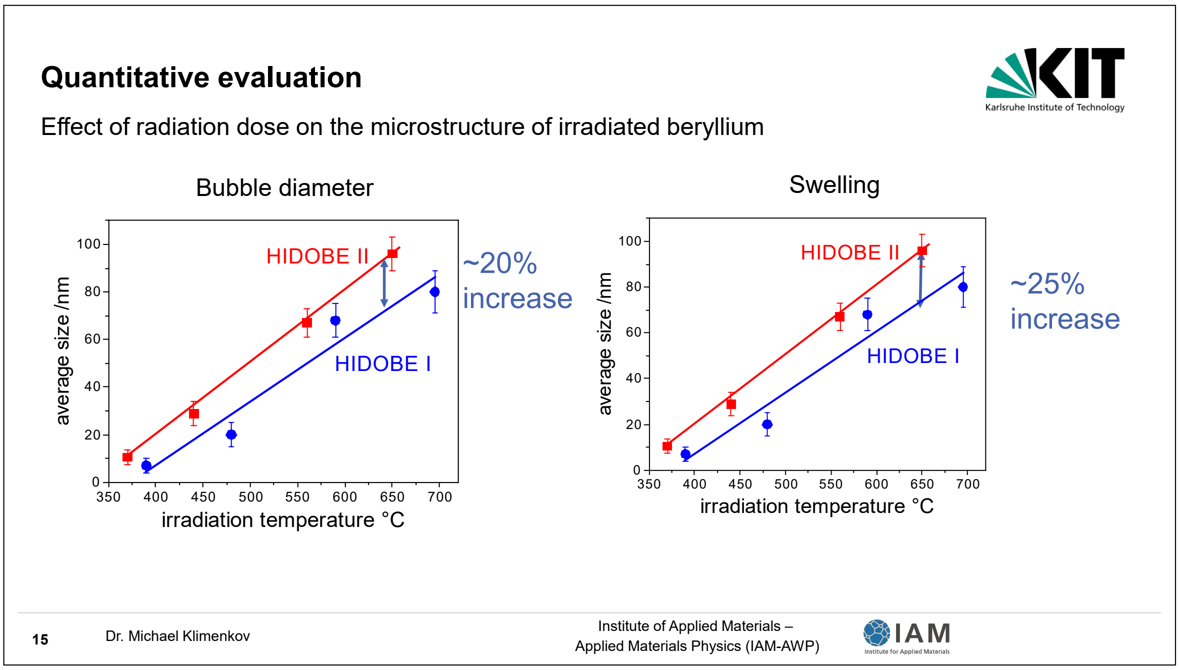
Quantitative evaluation
Effect of radiation dose on the microstructure of irradiated beryllium
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Bubble diameter
Swelling
HIDOBE II
HIDOBE I
~20% increase
~25% increase
average size /nm
irradiation temperature °C
15
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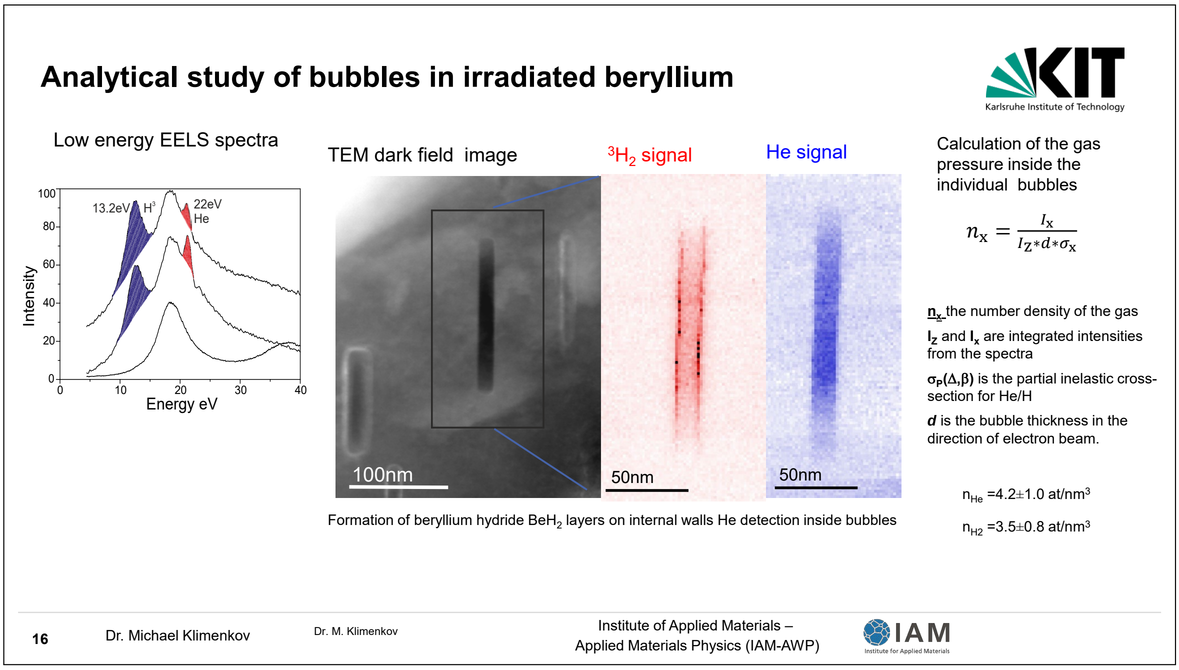
Analytical study of bubbles in irradiated beryllium
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Low energy EELS spectra
13.2eV H
22eV He
Intensity
Energy eV
TEM dark field image
$^3H_2$ signal
He signal
100nm
50nm
50nm
Formation of beryllium hydride BeH_2 layers on internal walls He detection inside bubbles
Calculation of the gas pressure inside the individual bubbles
$n_x = \frac{I_x}{I_Z * d * \sigma_x}$
n_x the number density of the gas
I_Z and I_x are integrated intensities from the spectra
$\sigma_P(\Delta,\beta)$ is the partial inelastic cross-section for He/H
d is the bubble thickness in the direction of electron beam.
n_{He} =4.2±1.0 at/nm³
n_{H2} =3.5±0.8 at/nm³
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Dr. M. Klimenkov
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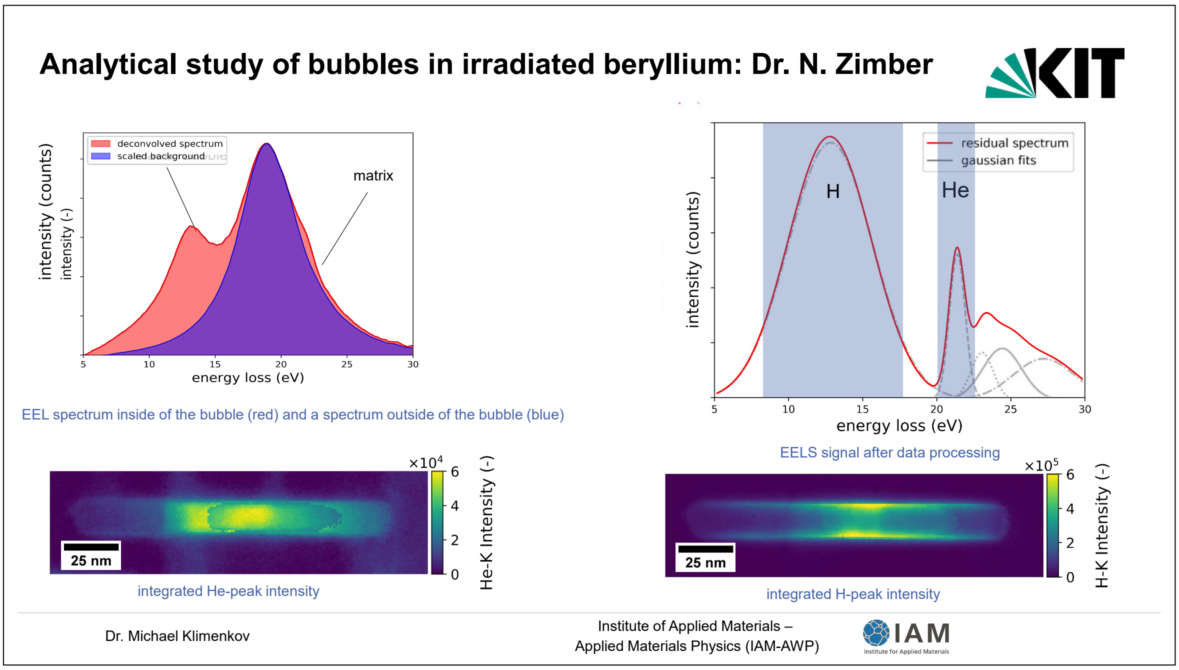

Analytical study of bubbles in irradiated beryllium: Dr. N. Zimber
deconvolved spectrum
scaled background
matrix
intensity (counts)
energy loss (eV)
EEL spectrum inside of the bubble (red) and a spectrum outside of the bubble (blue)
residual spectrum
gaussian fits
H
He
EELS signal after data processing
25 nm
He-K Intensity (-)
integrated He-peak intensity
H-K Intensity (-)
integrated H-peak intensity
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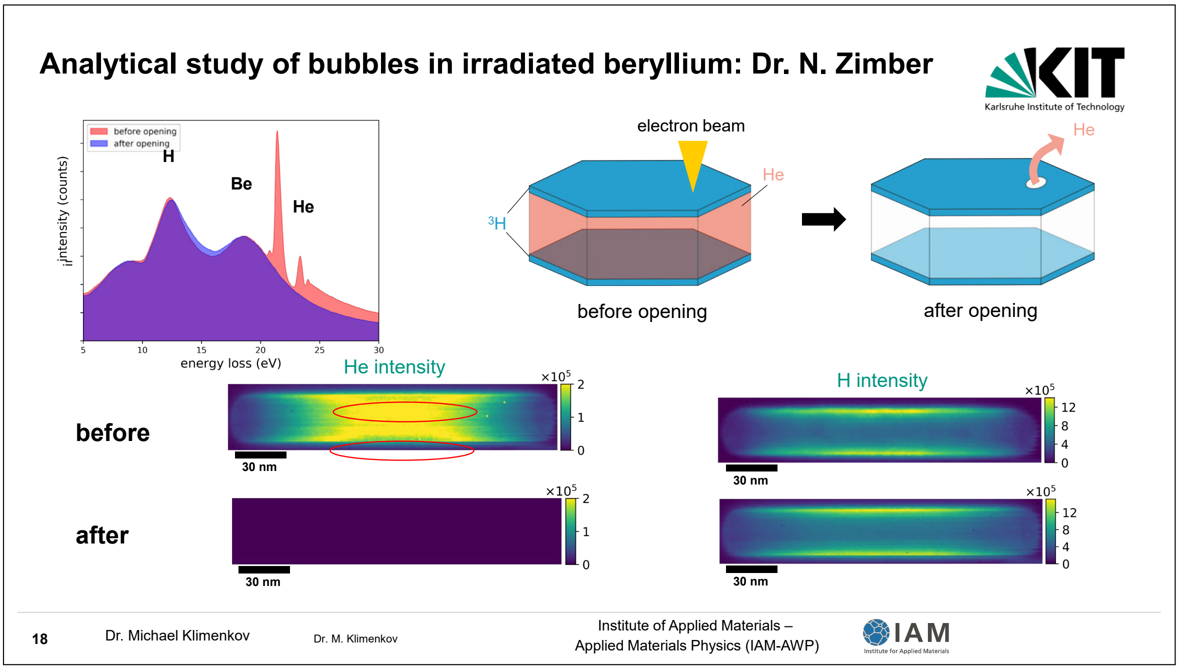

Analytical study of bubbles in irradiated beryllium: Dr. N. Zimber
Karlsruhe Institute of Technology
before opening
after opening
intensity (counts)
H
Be
He
energy loss (eV)
electron beam
³H
before opening
after opening
He intensity
H intensity
before
after
30 nm
18
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HIDOBE II publications

- M. Klimenkov, P. Vladimirov, J. Hoffmann, N. Zimber, A. Möslang, V. Kuksenko "First simultaneous detection of helium and tritium inside bubbles in beryllium" Micron **127** (2019) 102754
- M. Klimenkov, P. Vladimirov, U. Jäntsch, V. Kuksenko, R. Rolli, A. Möslang and N. Zimber "New insights into microstructure of irradiated beryllium based on experiments and computer simulations" Scientific Reports (2020) 10:8042
- N. Zimber, P. Vladimirov, M. Klimenkov, V. Kuksenko "Investigation of a high-dose irradiated beryllium microstructure" Journal of Nuclear Materials **540** (2020) 152374
- N. Zimber, P. Vladimirov, "The role of grain boundaries and denuded zones for tritium retention in high-dose neutron irradiated beryllium" Journal of Nuclear Materials **568** (2022) 153855

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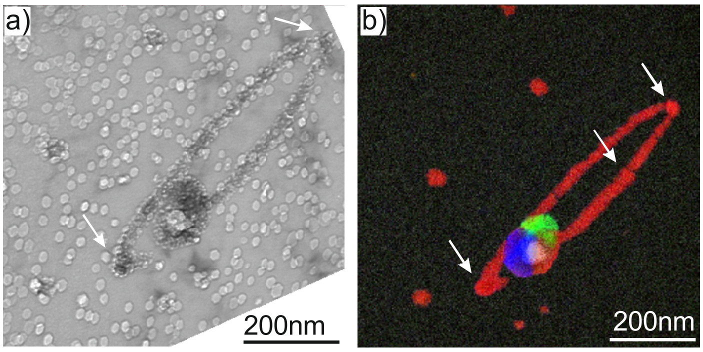
Distribution of impurity elements

presence of pecipitates at 370°C and 440°C



A loop decorated by a segregation of Fe-Al-Be phase pinned by a complex phase precipitate (a) observed at 643 K (370 °C).

Various phases in EDX map are colored as follows

- AlFe
- MnSi
- CrTi
- UFe

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Summary

- Successful microstructural examination of neutron irradiated beryllium at IAM-AWP over the past 10 years (6 publications in peer reviewed journal).
- Detection of He and H_2 in bubbles by TEM in neutron irradiated Be was reported for the first time.
- Recent study were focused on the characterization of gas bubbles located inside grains and at the GBs as well as on the study of spatial distribution and composition of secondary phase precipitates.
- **Recent results (HIDOBE II):**
- Helium bubbles were found inside grains at all irradiation temperatures, which is consistent with previous literature. Their shape is spherical at 370°C and changes to a flat hexagonal prism at higher irradiation temperatures. The bubble size increases strongly with irradiation temperature. The apparent swelling estimated from the TEM data reaches ~9% at 650°C
- Precipitation of Fe-Al-Be phase within grains were detected at low irradiation temperatures (370°C and 440°C)
- EELS spectroscopy was used for detection and analysis of He and H_2 gases trapped inside flat hexagonal bubbles formed on the basal planes of beryllium under neutron irradiation.
- The number densities of both gases inside the bubbles were calculated using atomic scattering cross-section and the intensity of the zero-loss peak. The values He=(4.2±1) at/nm^3 and nH_2=(3.5±1.2) molecules/nm^3 were determined for a bubble with a diameter of 160nm.

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