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MEGAPIE and its post-irradiation examination

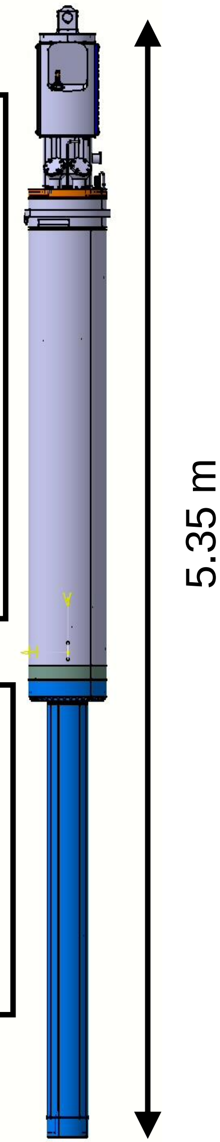
EuCard-2 Workshop on ADS, 7-9.February 2017, CERN, Geneve

MEGAPIE - MEGAwatt Pilot Experiment

- **Joint international initiative to design, build, license, operate, dismantle and explore a liquid metal LBE*) spallation target for the 1 MW beam power regime for the first time (started ~ September 1999).**

***) LBE: Lead-Bismuth-Eutectic (T_m=125°C)**

„Accelerator Driven Systems (ADS) and transmutation technologies are becoming important for the sustainable development of nuclear energy all over the world, but have technical challenges spread over a wide range of fields. ***Thus sharing experimental efforts in a systematic way is highly desirable, MEGAPIE being a good precursor for such an international collaboration.*** *"**

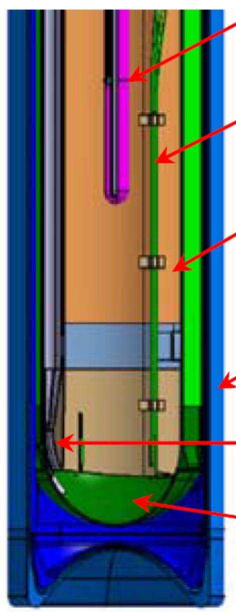


*"Research and Test Facilities Required in Nuclear Science and Technology", Nuclear Energy Agency (NEA) Report, NEA No. 6293, OECD 2009, ISBN 978-92-64-99070-8

PIE of MEGAPIE

PIE of MEGAPIE Target can be subdivided in 3 PIE sub-projects

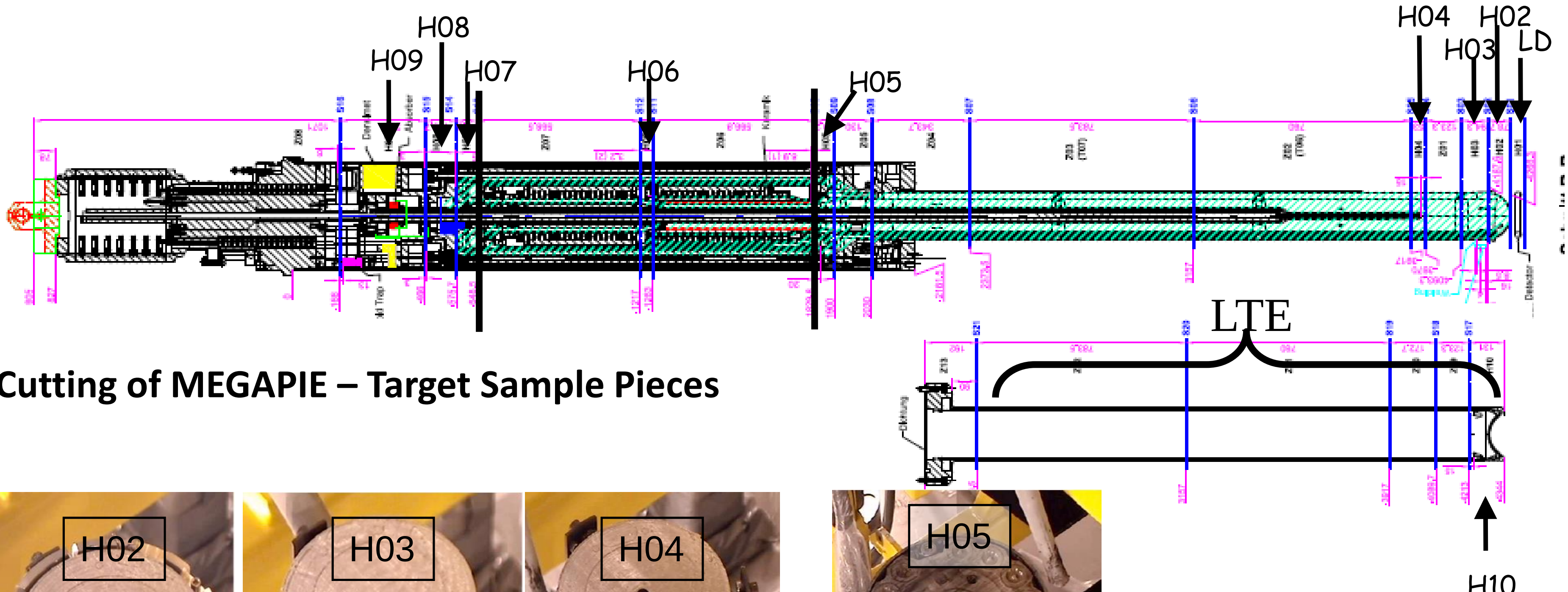
1. Non Destructive Investigations
2. Investigation on LBE
3. Investigation on structural materials
    3.1. Lower Liquid Metal Container (LLMC) from T91
    3.2. Flow Guide Tube (FGT) from SS316L



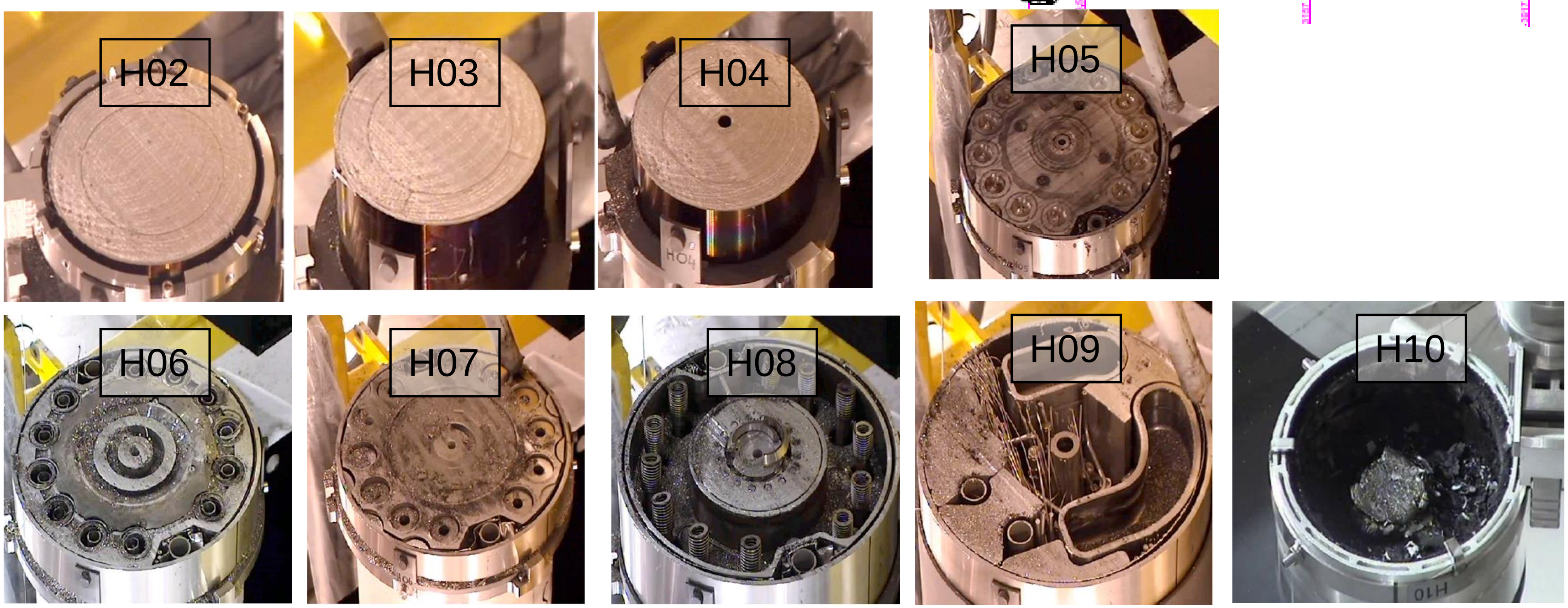
Production of PIE samples 2 step process

1. Cutting of Target Pieces to be investigated in ZWILAG
2. Production of LBE and structural material samples in the Hot Laboratory at PSI

Cutting & Disposal of MEGAPIE at ZWILAG

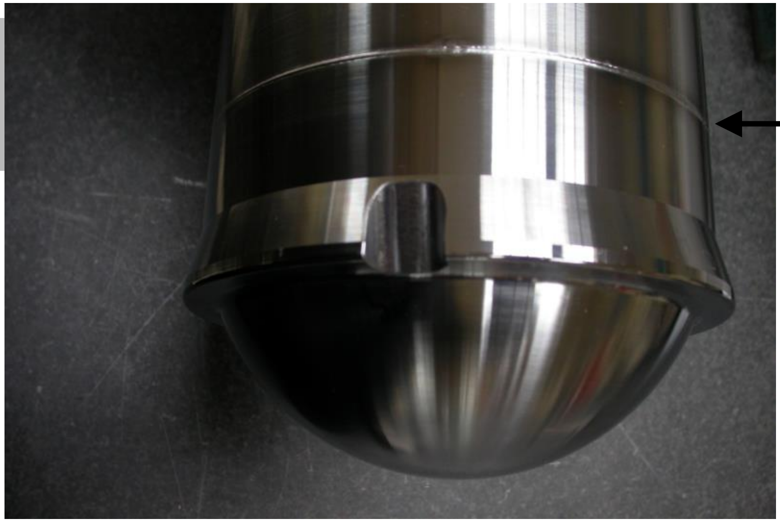


Cutting of MEGAPIE – Target Sample Pieces

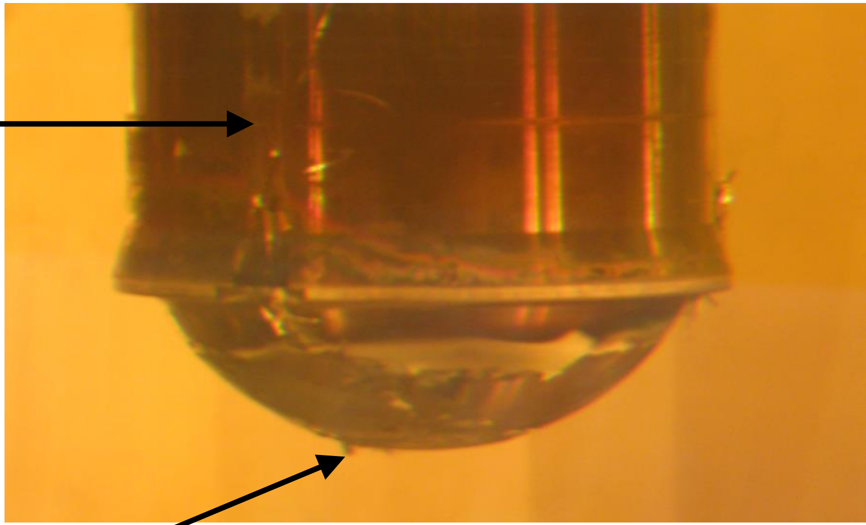


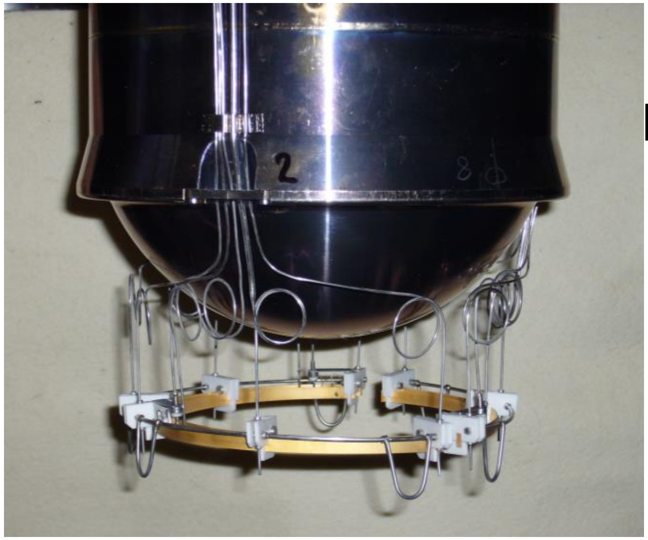
Before & After Irradiation

T91 Lower Liquid Metal Container (Calotte + Leak Detector)

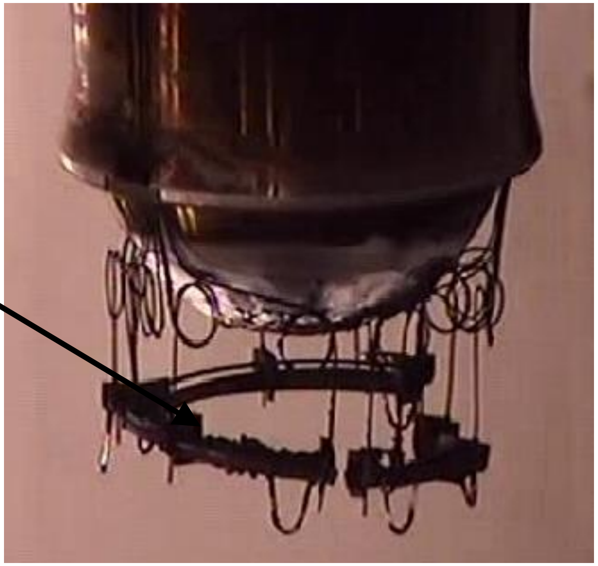


EBW



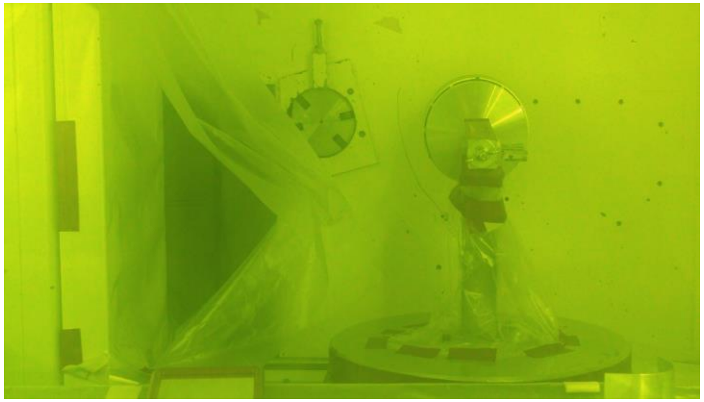


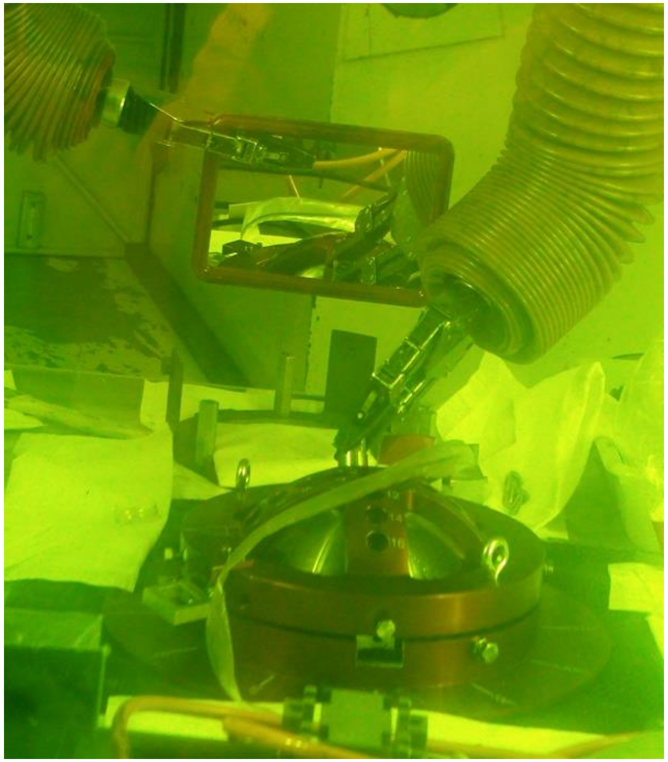
Material Deposit

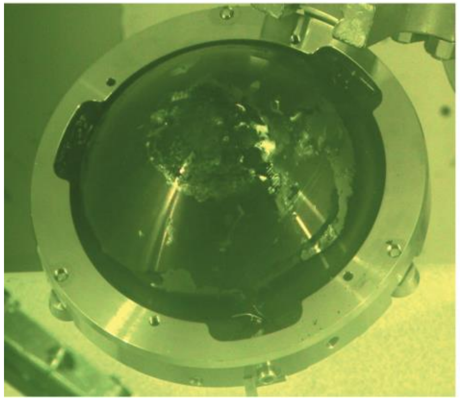


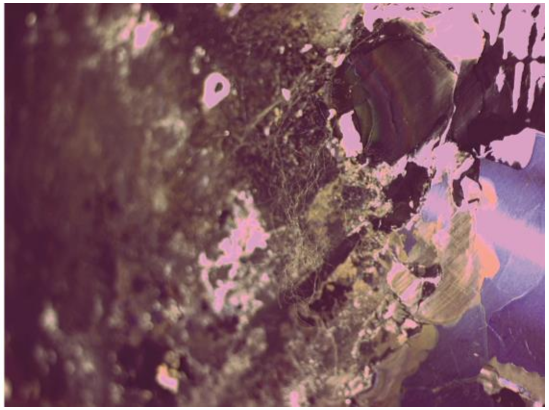
Non Destructive Testing

- Gamma Mapping of the AlMg3 safety shroud (LTE)
- Investigation of material sticking on the BEW (OM, SEM, EDX)
- Ultrasonic thickness measurement of the BEW

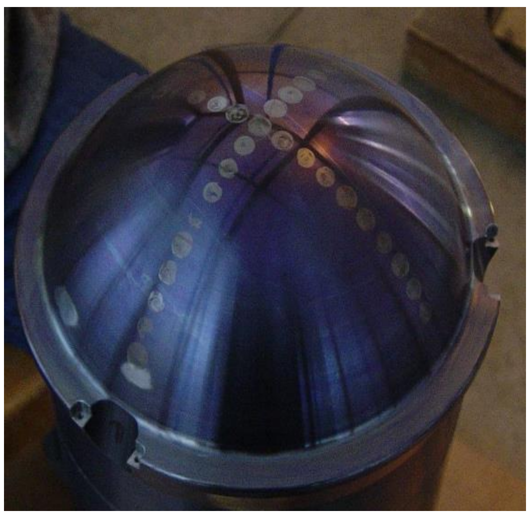


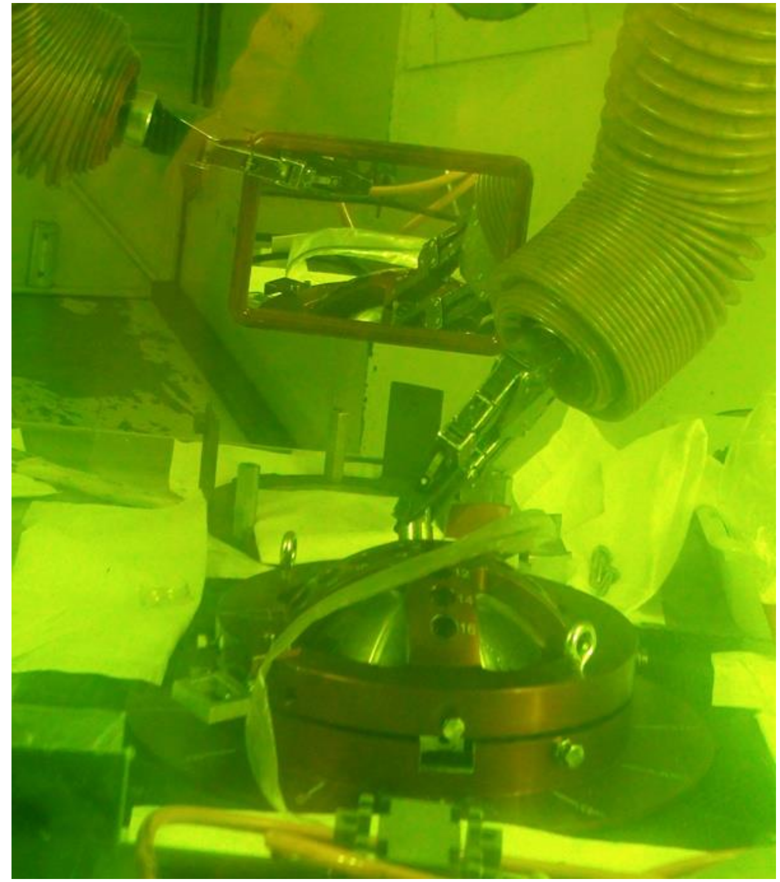






NDT – Thickness measurement of T91 BEW





Constant 20 µm difference between measurements before and after irradiation ➔ no evident dissolution corrosion effects considering very different LBE flow velocity and temperature etc at different positions.

20 µm difference due to measurements of T91 BEW filled with LBE after irradiation

➔ NO THICKNESS CHANGE

LBE Sample Taking & Analysis

Core drilling tool

Sample breaking device

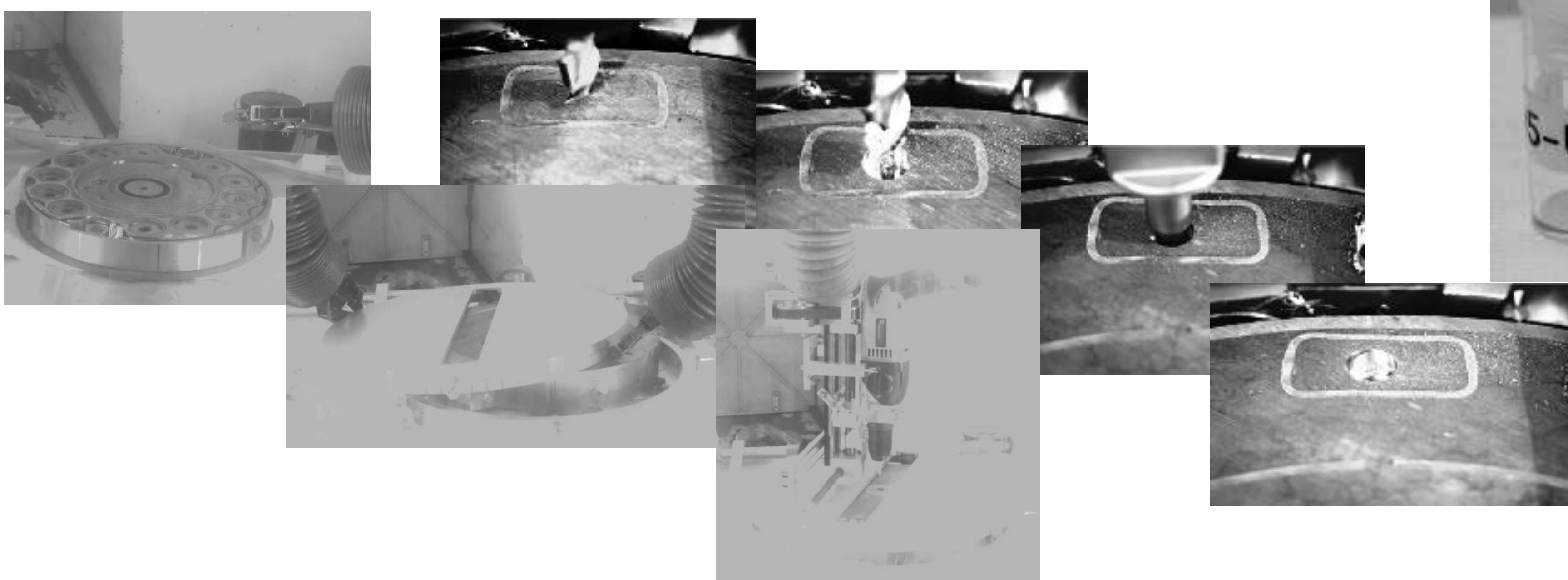
LBE Sample Sizes

Length 5 mm

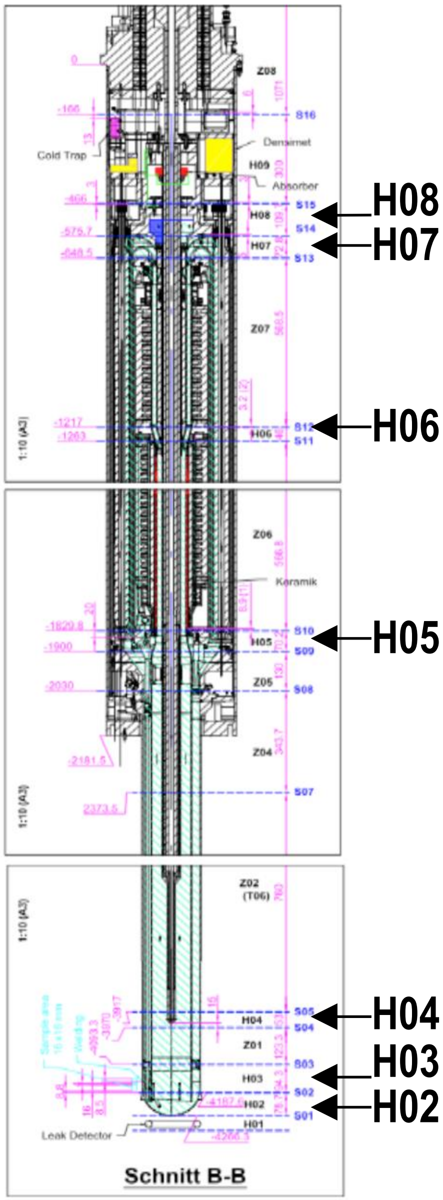
Diameter 1.5 – 2.0 mm

Dose rates 5 – 10 µSv/h in 10 cm distance

Cold test (above) + Sample taking in Hotcell (below)



LBE sample Analysis

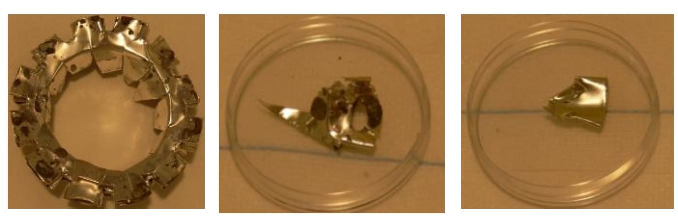


- **In total 76 samples were taken from the bulk (43 samples), from steel-LBE and from LBE-gas interfaces (33 samples)**
- **Absorber foils from the gas expansion tank extracted**
- **Goal: Investigate the spatial distribution of radionuclides in the target and validate theoretical predictions of the nuclide inventory**

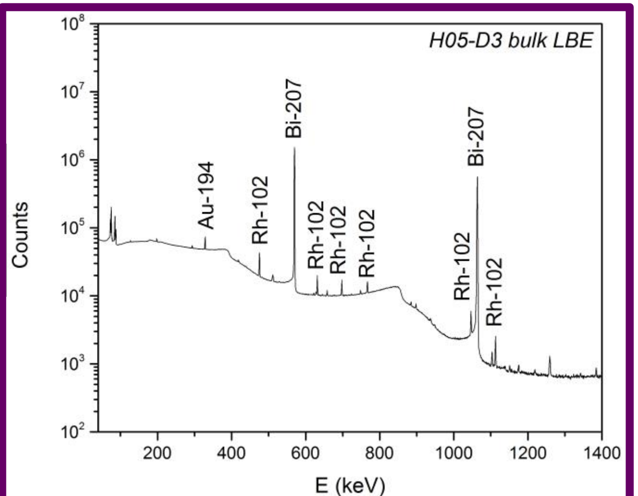
1. **Gamma Spectroscopy**
2. **Chemical separation of radionuclides most relevant for disposal and operation**

Absorber foils from Expansion volume

Pd and Ag foils

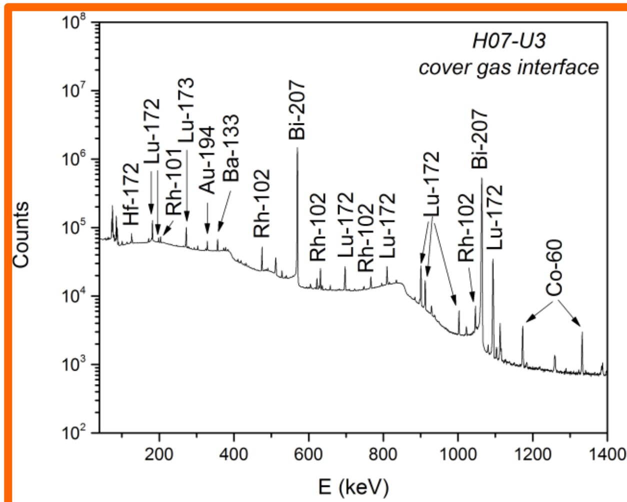


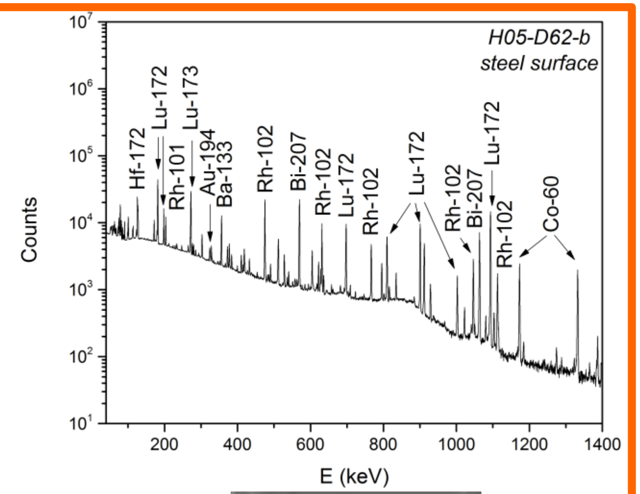
Qualitative evaluation of γ-measurements



Bulk:

^{207}Bi, ^{101}Rh, ^{102}Rh, ^{108m}Ag, ^{110m}Ag, ^{194}Hg/Au, ^{195}Au, ^{202}Pb/Tl





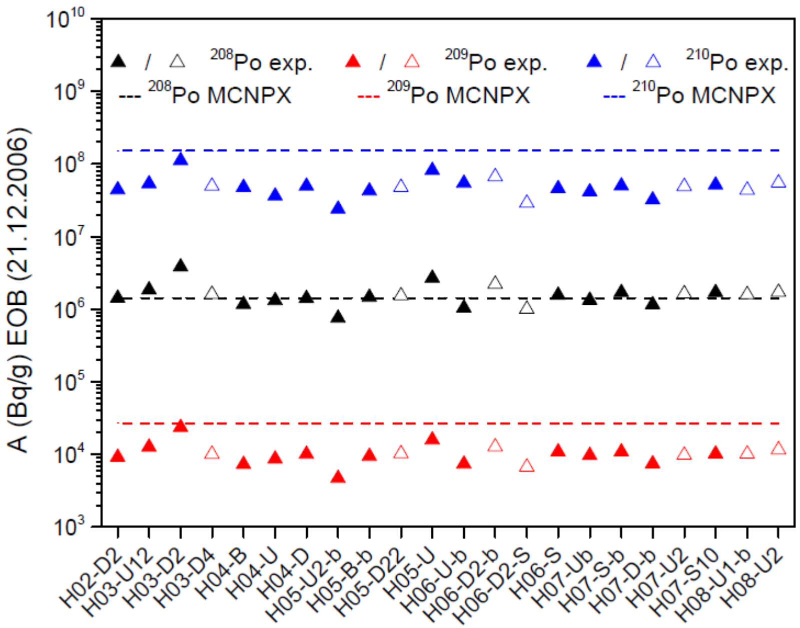
Surfaces:

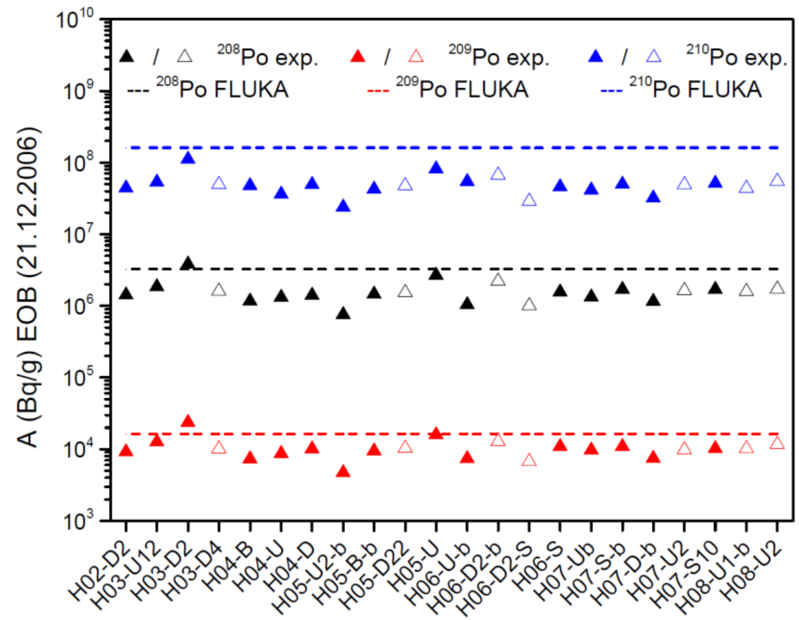
Additionally, deposition of ^{172}Hf/Lu, ^{173}Lu, ^{133}Ba, ^{60}Co

Results:

- Bulk LBE contains only **noble** metals that have a significant solubility in LBE
- Radionuclides of elements that have only low solubility in LBE or are sensitive to oxidation are only detected in samples taken at the LBE/steel interface and the LBE/cover gas interface
- These findings are consistently observed in all sections/samples, with the exception of a stagnant Zone in H07.

Distribution of $^{208-210}Po$





	^{208}Po (Bq/g)	^{209}Po (Bq/g)	^{210}Po (Bq/g)
chem. anal.	$1.63 \pm 0.14 \times 10^6$	$1.04 \pm 0.08 \times 10^4$	$5.04 \pm 0.39 \times 10^7$
FLUKA	3.28×10^6	1.63×10^4	1.61×10^8
MCNPX	1.42×10^6	2.68×10^4	1.53×10^8

^{209}Bi $(p/n,\gamma)$ ^{210}Po

^{209}Bi (p,n) ^{209}Po

^{209}Bi $(p,2n)$ ^{208}Po

- Po is mainly homogeneously distributed (not in agreement with previous findings)
- Very thin Po layer on top of surfaces
- greement with theoretical predictions

Production pathways of Polonium

Results from investigation of Pb samples from “solid” SINQ Target

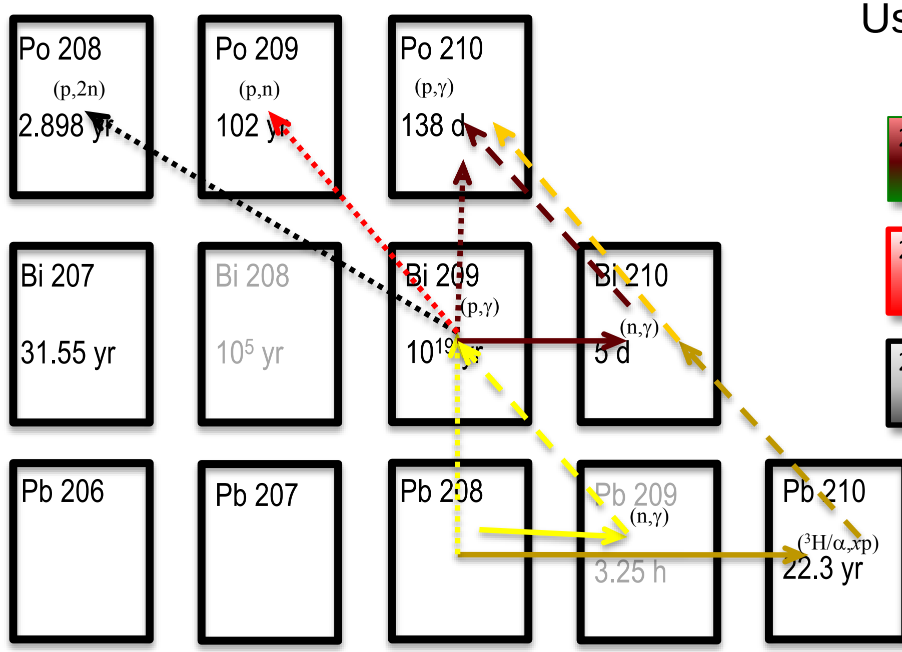
^{209}Bi content in

SINQ lead:

220 ppm

(ICP OES)

$=7 \cdot 10^{17}$ atoms/g lead



Usual pathways via ^{209}Bi

^{209}Bi $(p/n,\gamma)$ ^{210}Po

^{209}Bi (p,n) ^{209}Po

^{209}Bi $(p,2n)$ ^{208}Po

^{208}Pb $(p/n,\gamma)$ ^{209}Bi

^{208}Pb $(2n,\gamma)$ ^{210}Pb

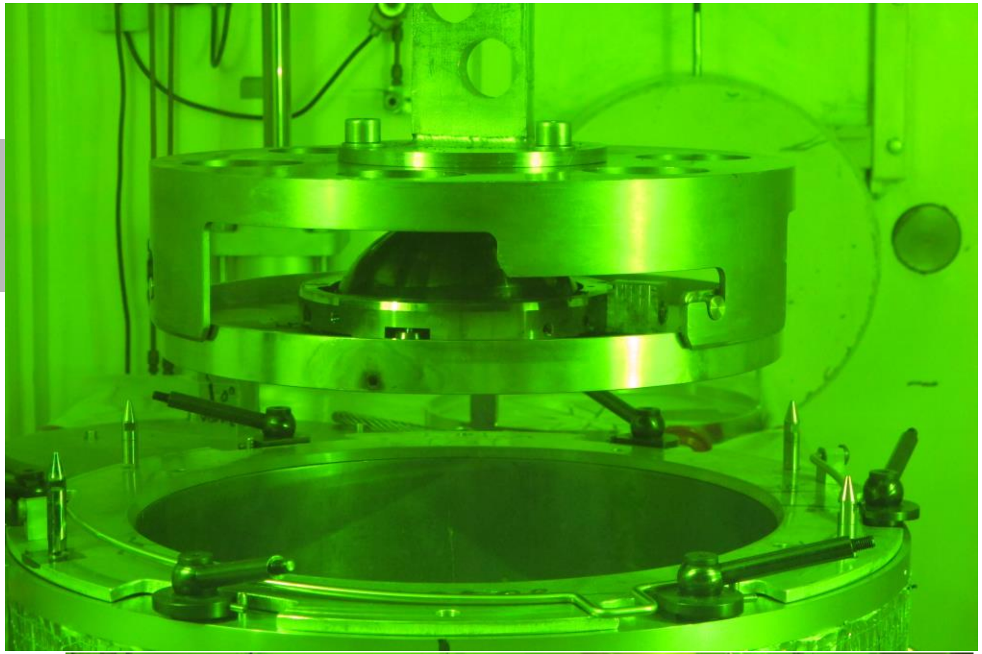
^{208}Pb $(^{3}H/\alpha,2p)$ ^{210}Pb

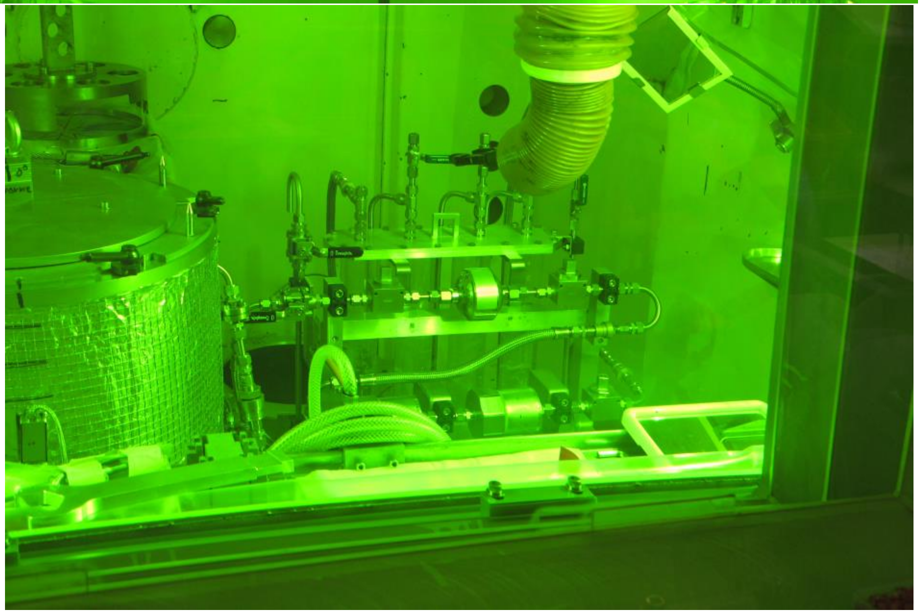
Long lived mother ^{210}Pb leads to equilibrium concentration of ^{210}Po

Main Results of LBE investigations

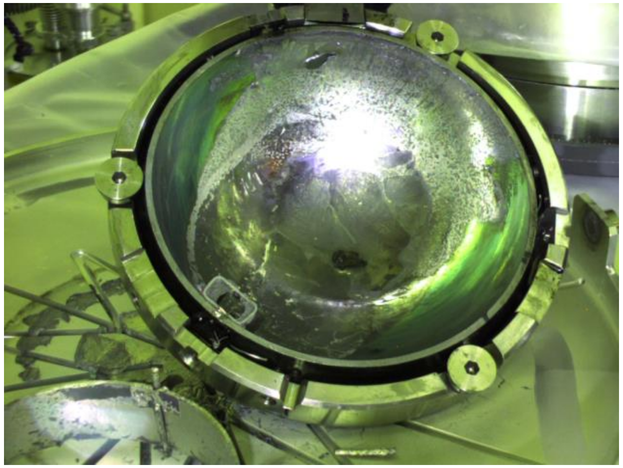
- In total: 20 radionuclides were analysed
- Bulk LBE contains mainly noble metals that have a significant solubility in LBE (Gold, Mercury, Silver, Rhodium)
- Radionuclides of elements that are sensitive to oxidation/reduction are only detected in samples taken at the LBE/steel interface and the LBE/cover gas interface (Lanthanides, Iodine, Chlorine)
- Most of the determined radionuclides show good or fair agreement with theoretical predictions
- $^{208\text{-}210}Po$ is homogeneously distributed
- ^{210}Po is produced not only from Bi impurities, but also from the precursor ^{210}Pb.
- For end of beam, the experimental values for Po agree with the predicted ones.
- For long-term storage or disposal, the amount of ^{210}Po, produced by the precursor, is dominating.

LBE melting – example H02

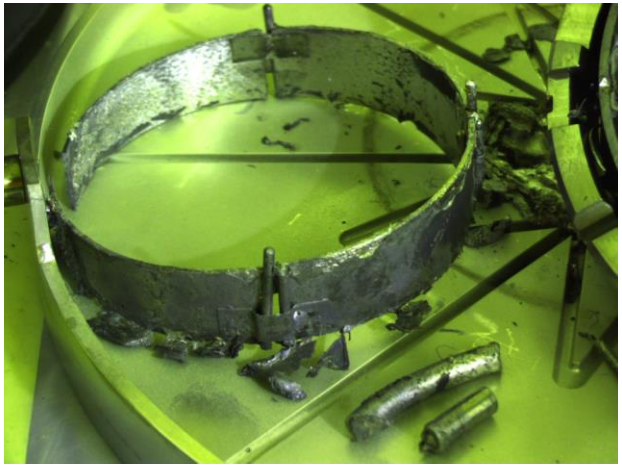




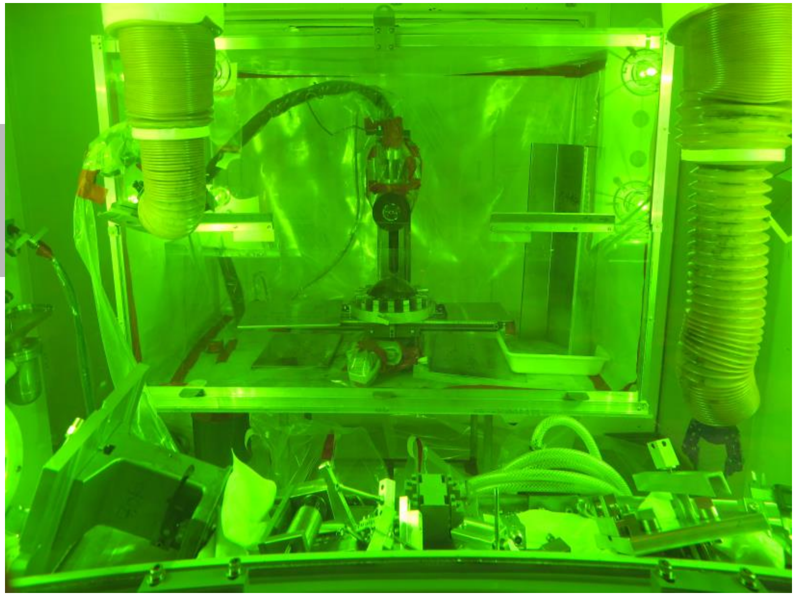
H02 BEW (T91)



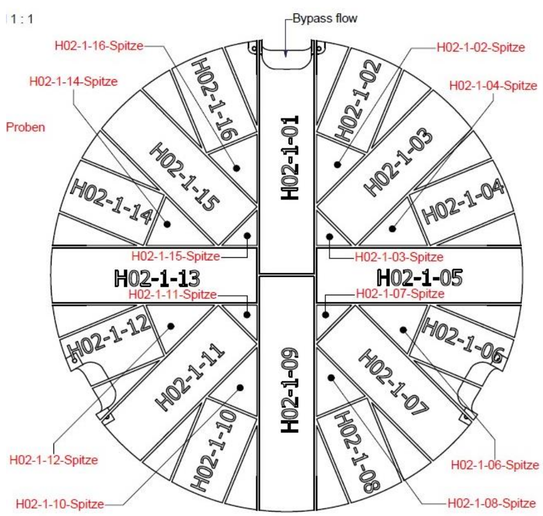
H02 FGT (316L)



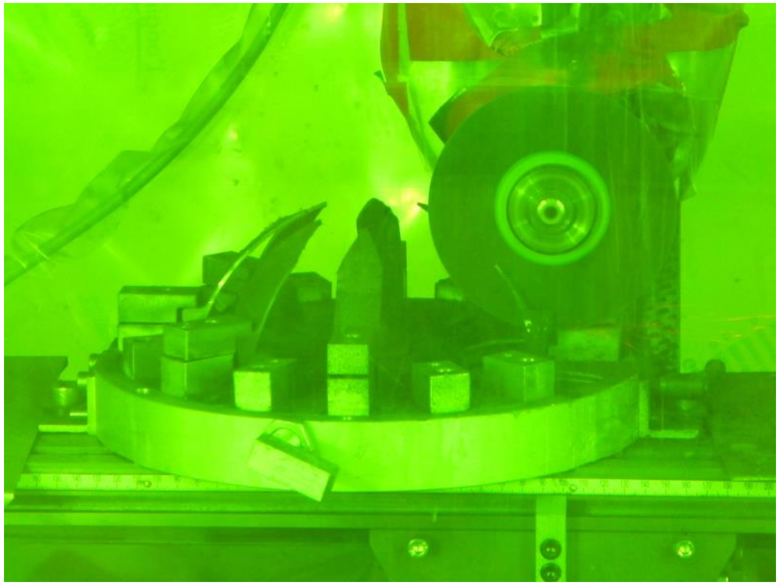
Raw cutting of PIE samples



- EDM cutting & cleaning not easily feasible with the (large) target sample pieces.
- Cut H02 (beam entrance window,BEW), H03-1 (lower liquid metal container, LLMC), H03-2 (flow guide tube, FGT), H04-1 (LLMC) and H04-2 (FGT) with grinder disc.
- Cutting done in substeps to minimize temperature rise. Grinder discs exchanged after cutting 1 sample piece.

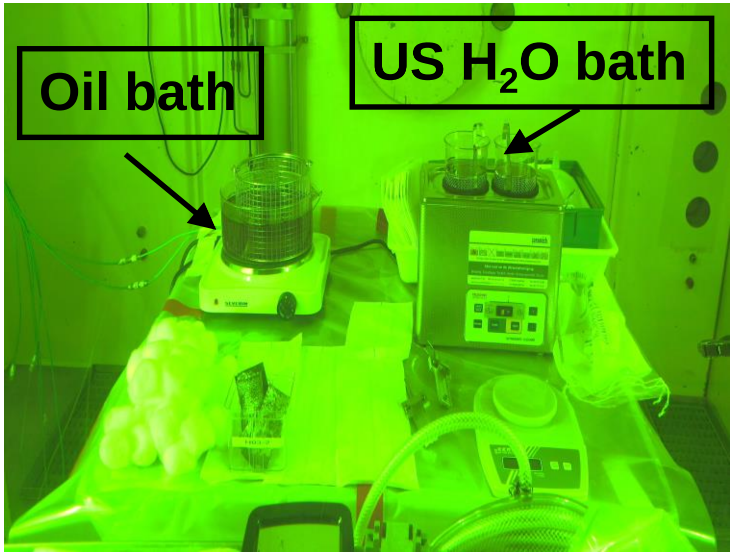


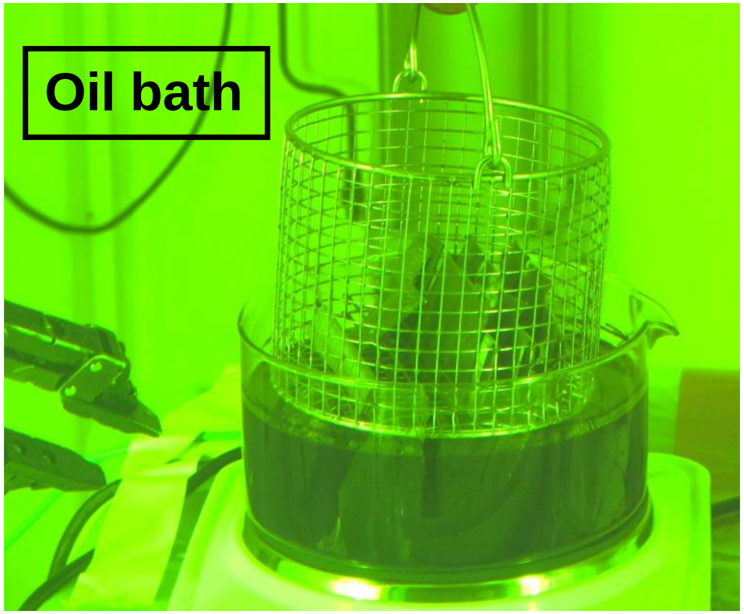
Raw cutting scheme of the BEW into 28 pieces



Cleaning raw cut sample pieces

- The original cleaning scheme of raw cut sample pieces is a 4-step process proposed by Yong Dai
  - ‚Mechanical cleaning‘ with heated oil (UCON-HTF @ 190°C max.)
  - Ultrasonic bath in de-ionized H_2O
  - ‚Chemical cleaning‘ in 5 molare HNO_3
  - Ultrasonic bath in de-ionized H_2O
- Measurements after step 4 showed still ‚high‘ removable (α-) activity
- Tests to clean with EDTA (**E**thylen**d**iamin-**t**etra-**a**cetic di-sodium-salt) instead of H_2O. Not significantly better, but disposal more complicated → cleaning continued with H_2O
- 2 – 4 additional cleaning steps in ultrasonic H_2O bath per target sample piece

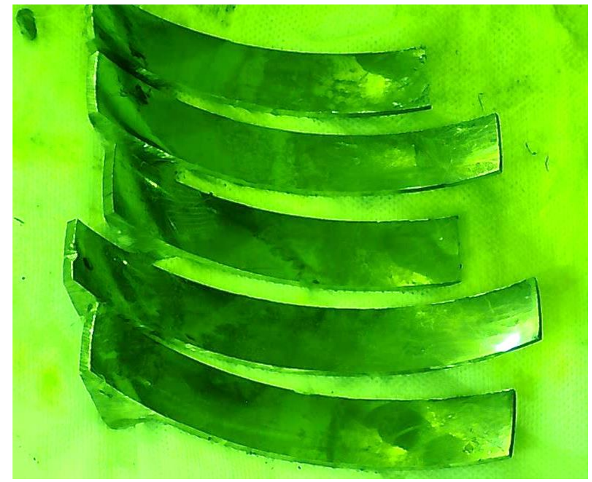




Cleaning raw cut sample pieces

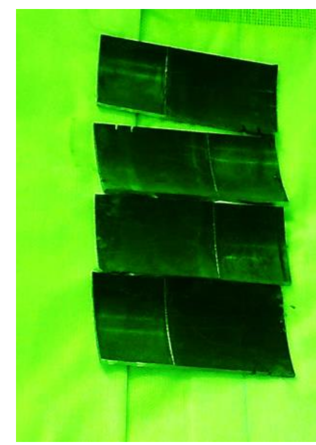
H02 raw cut samples before oil bath

H02 raw cut samples after oil bath



- ❑Each raw cut sample piece at least put twice (for 5 – 10 minutes) into the oil bath (UCON-HTF, Poly-ethylene glycol).
- ❑Sticking LBE wiped off with tissue.

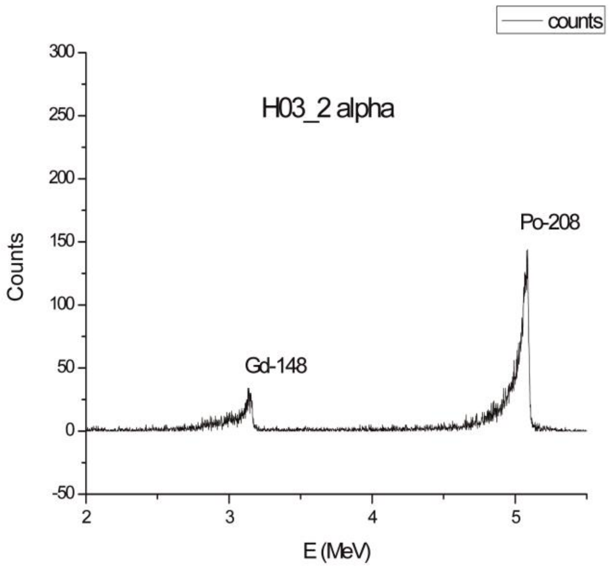
H03-1 before oil bath

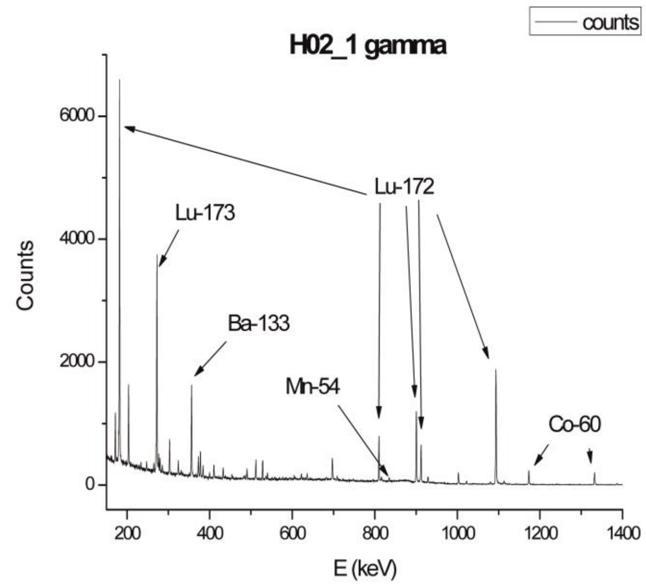


H03-1 after oil bath

Cleaning raw cut sample pieces

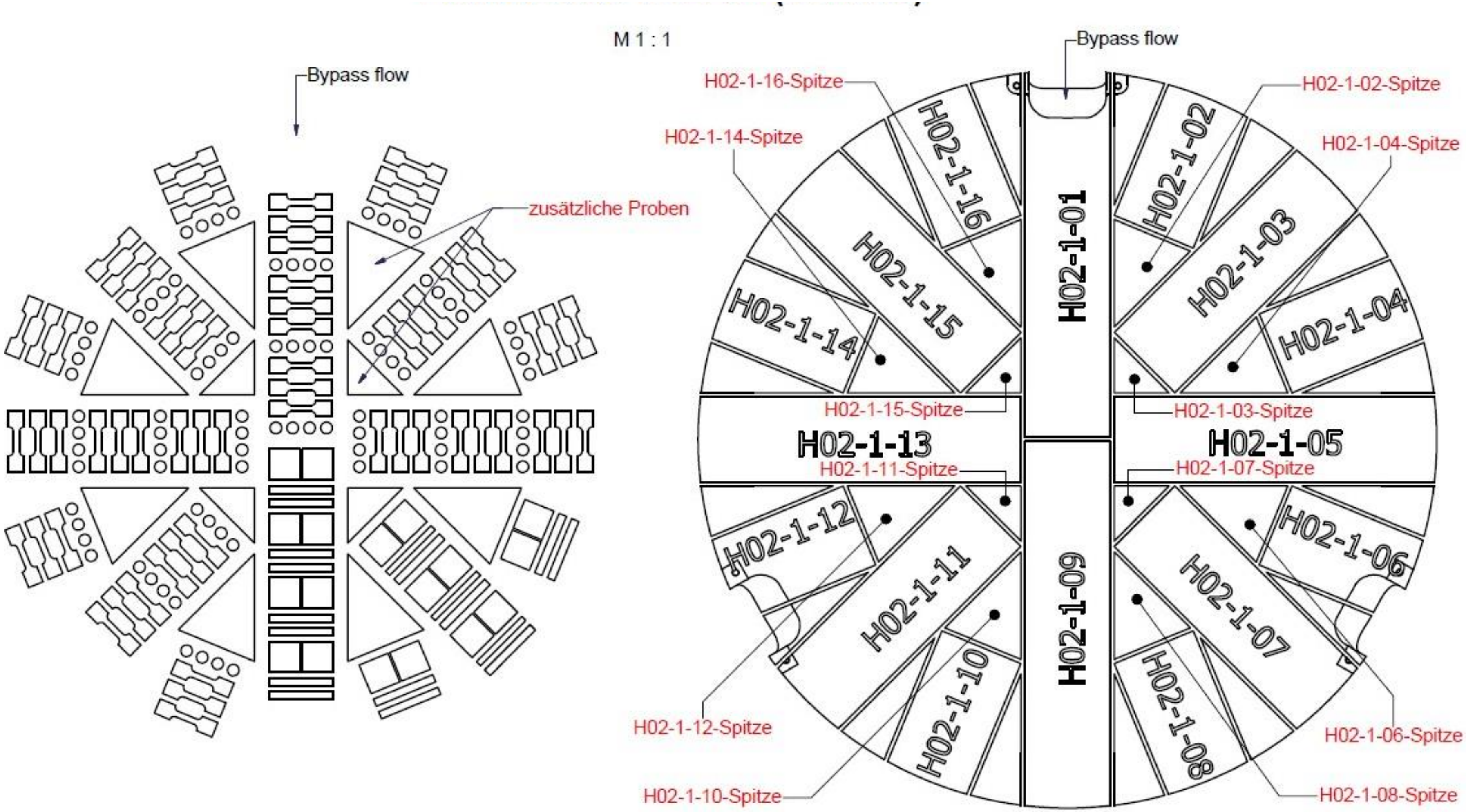
α- and γ-spectra of wipe test taken from H02/03 raw cut sample



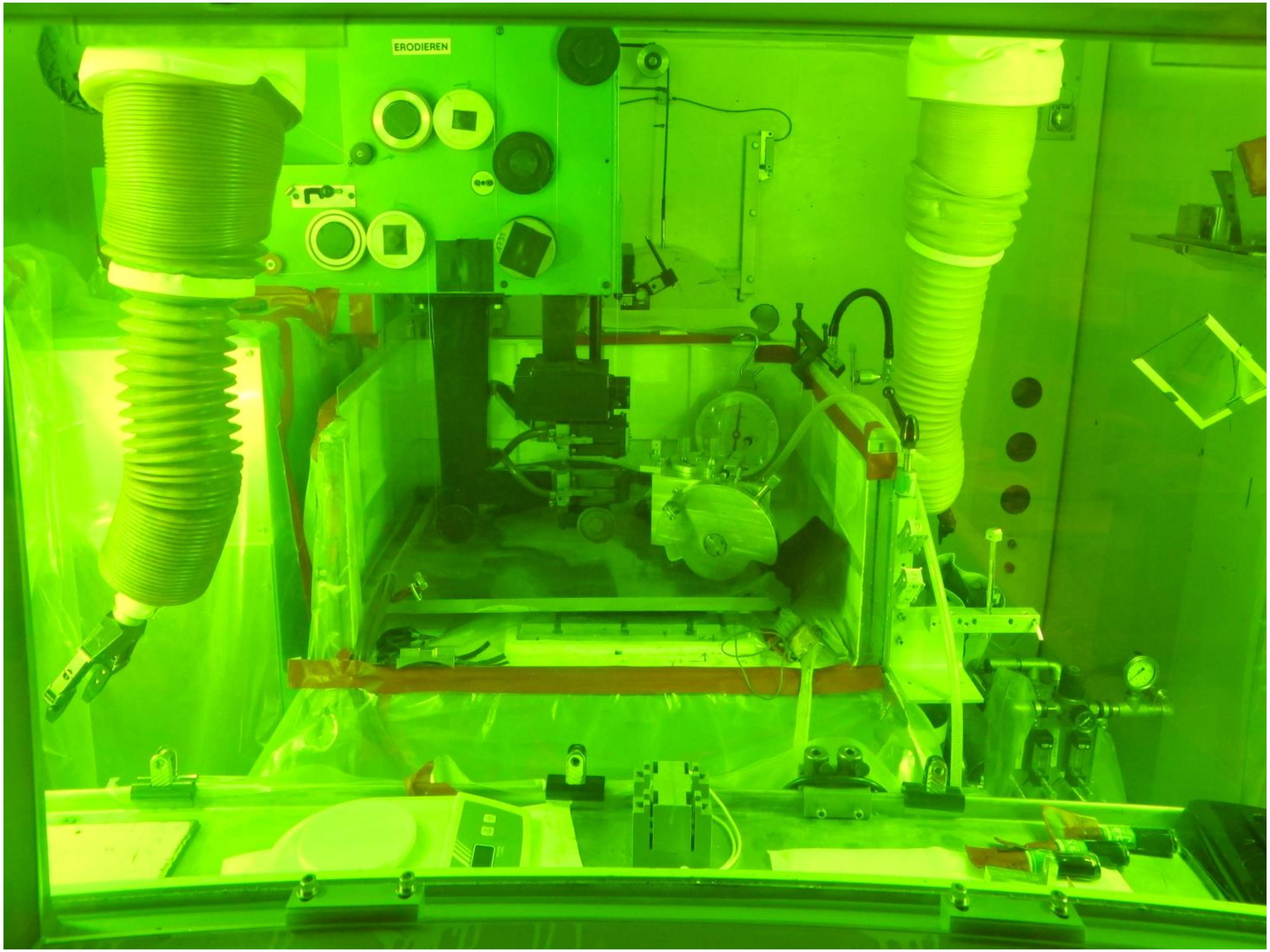


- ❑After cleaning procedure (mechanical cleaning, US bath, chemical cleaning, US bath) α-activity was still too high to be accepted by some of partner laboratories (especially LANL).
- ❑More cleaning (➔ trial with EDTA ➔ same efficiency as H_2O) ➔ further cleaning with H_2O
- ❑2 – 4 additional cleaning sessions with de-ionized H_2O

EDM cutting plan for the T91 BEW

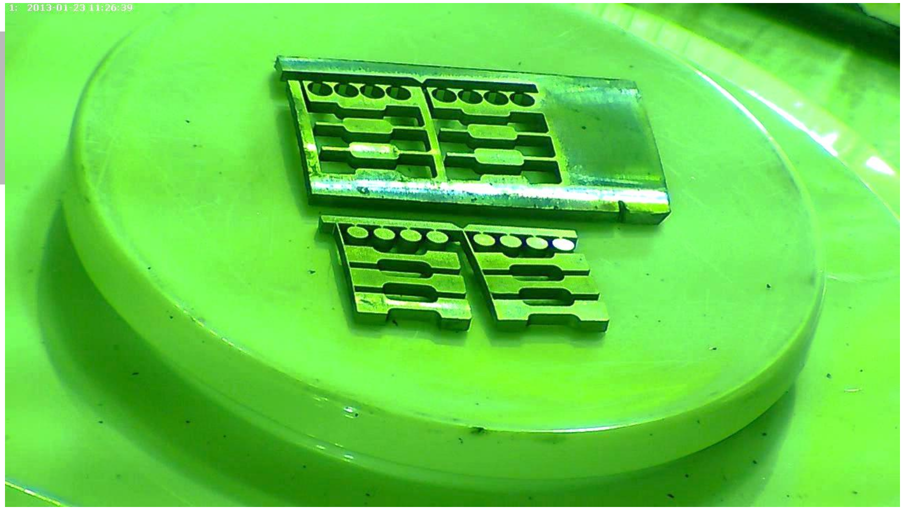


EDM Cutting

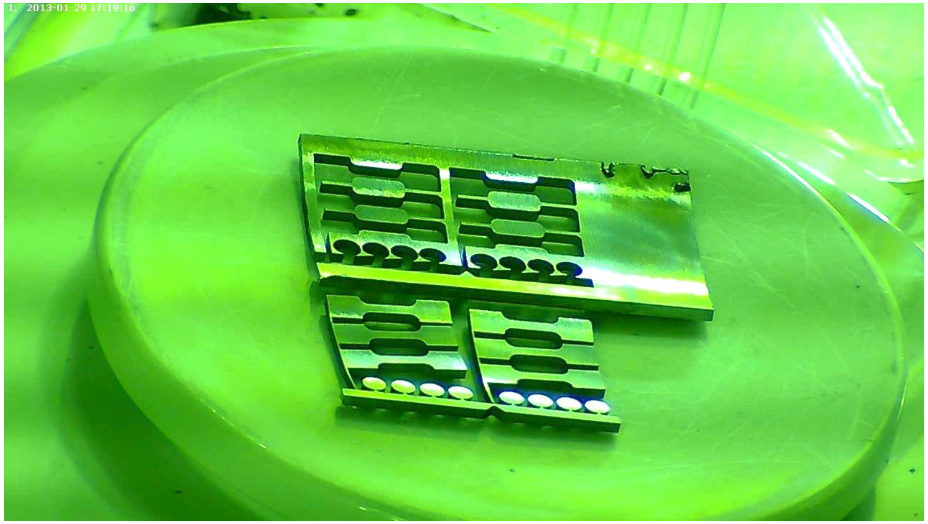


PIE sample production

H04-1-A



H04-2-A



- 82 sample groups, in total 793 samples, without LBE on the surface have been fabricated.
- CEA (71 samples), JAEA (67 samples), KIT (83 samples), LANL (71 samples) and SCK·CEN (80 samples) received 47% of all (LBE cleaned) samples in spring 2013.

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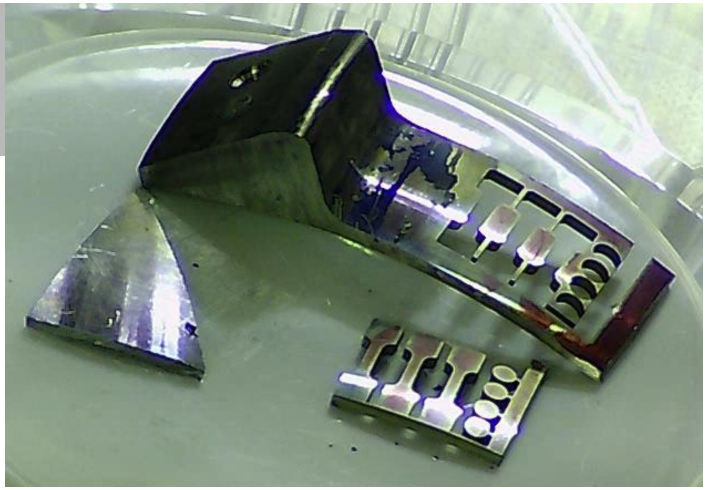
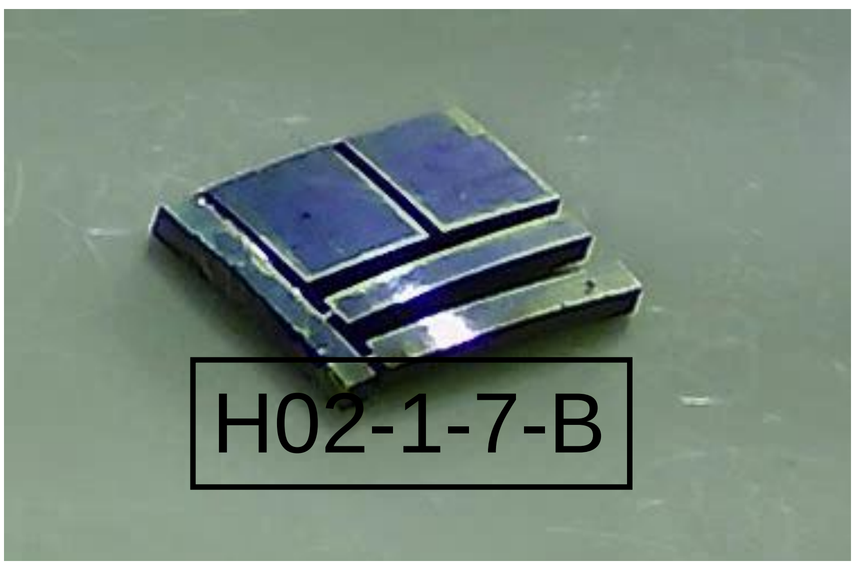
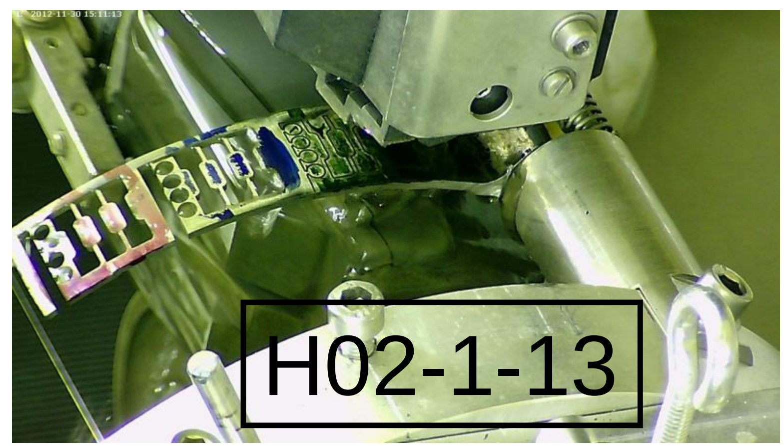
EDM Cutting

H02-1-02

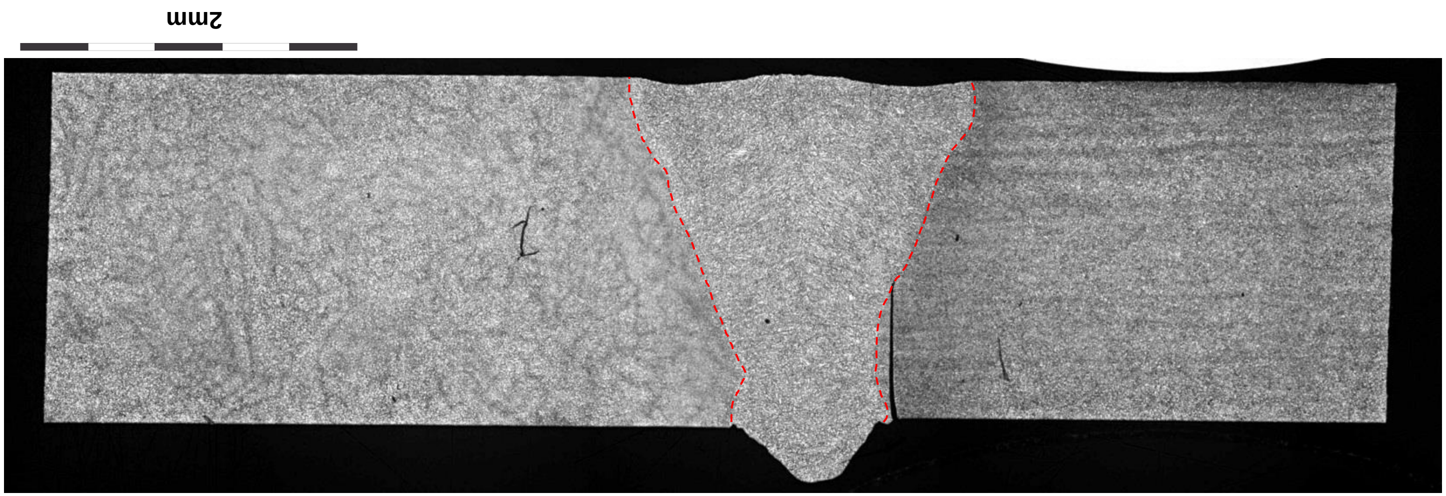


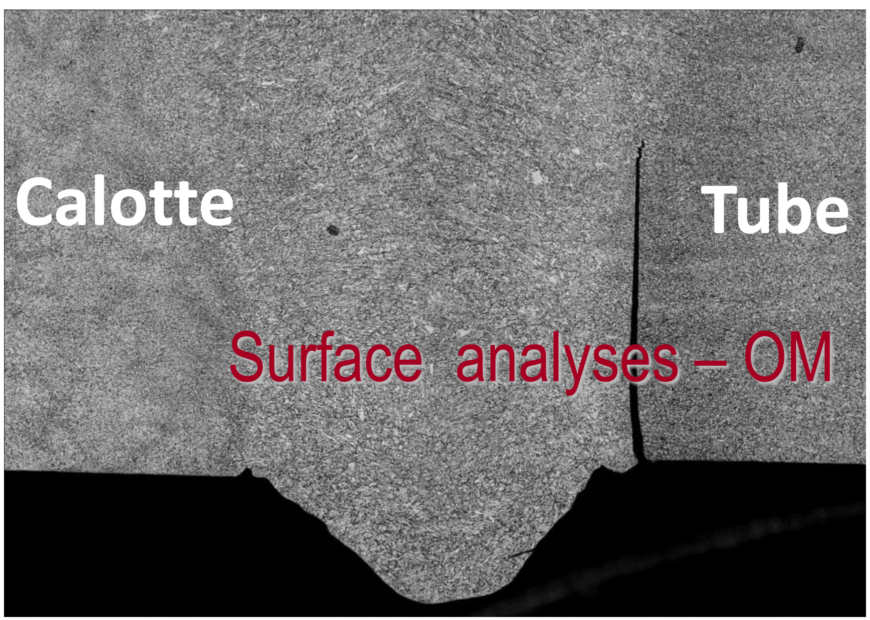
H02-1-05





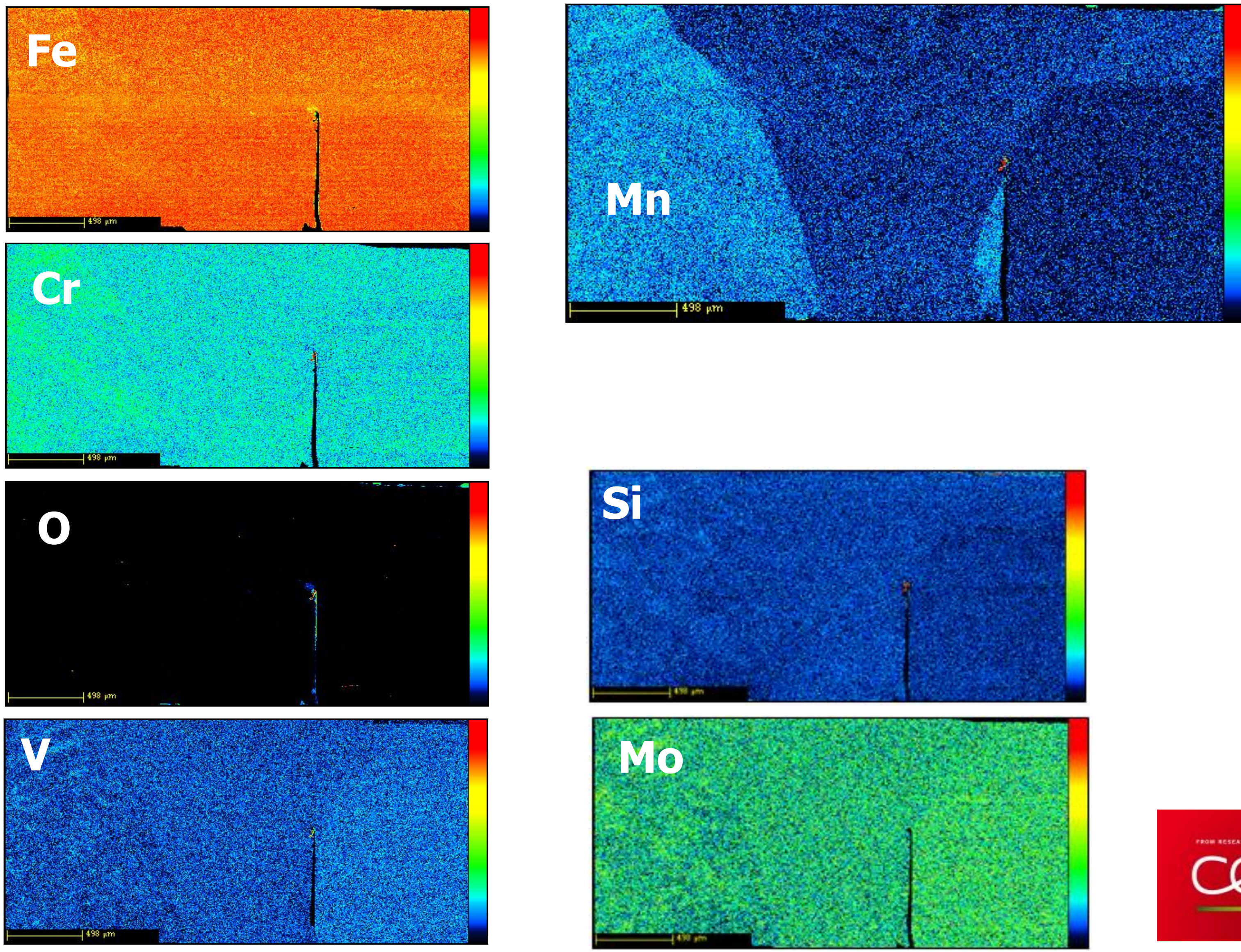
PIE Analysis – LLMC (T91) - OM





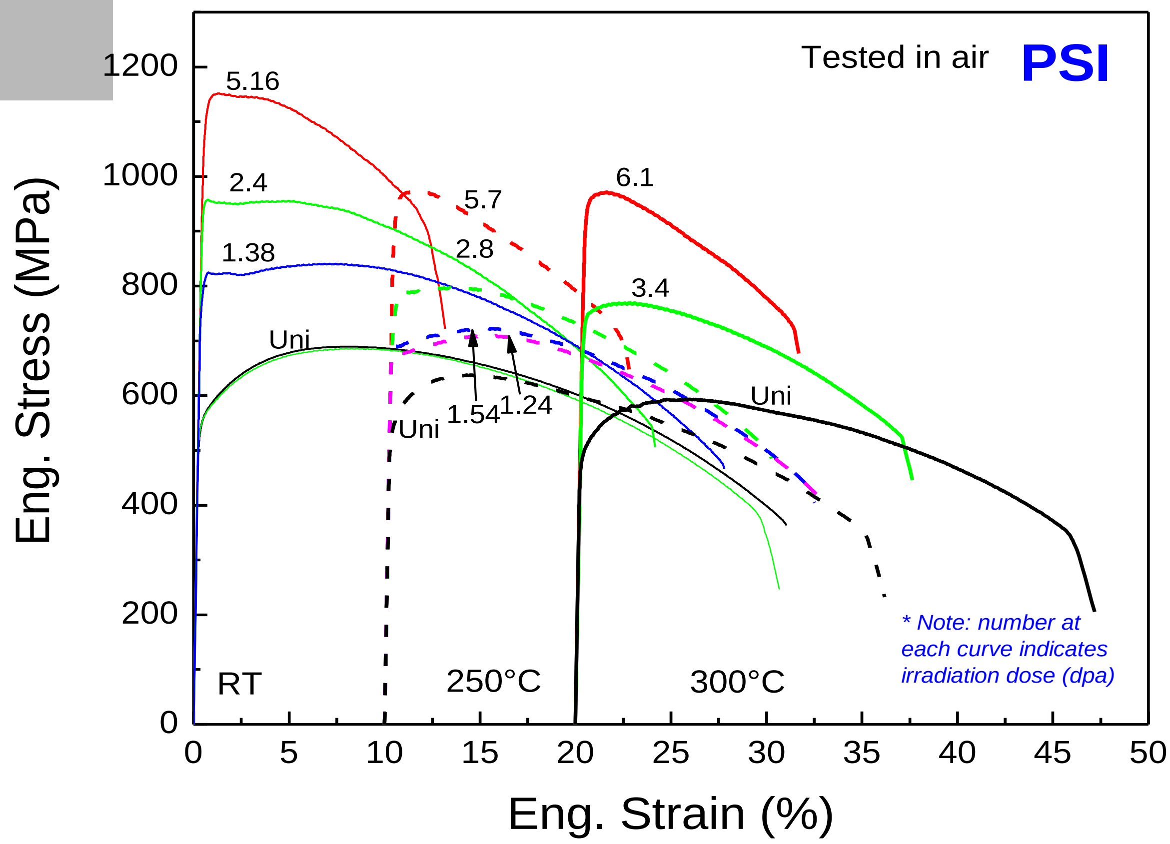
- **Crack most likely due to imperfect alignment during EB welding**
- **No evidence of propagation during operation**

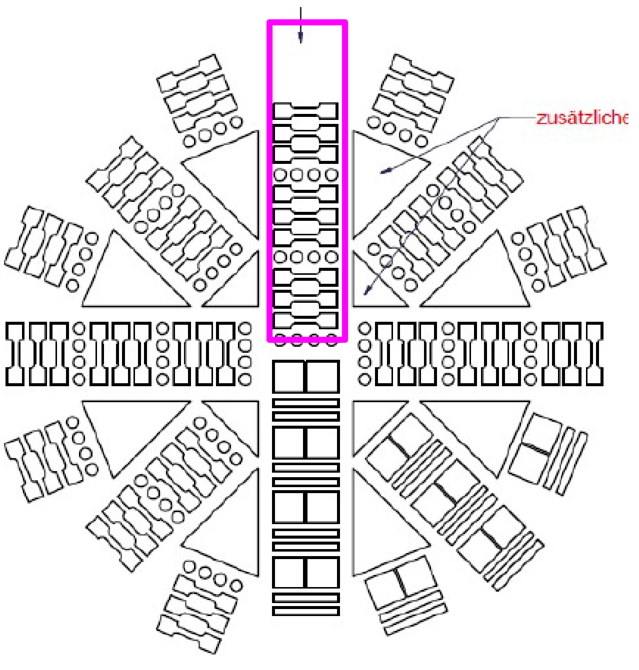
PIE Analysis – LLMC (T91) - EPMA



PIE Analysis – LLMC (T91) – Tensile Test

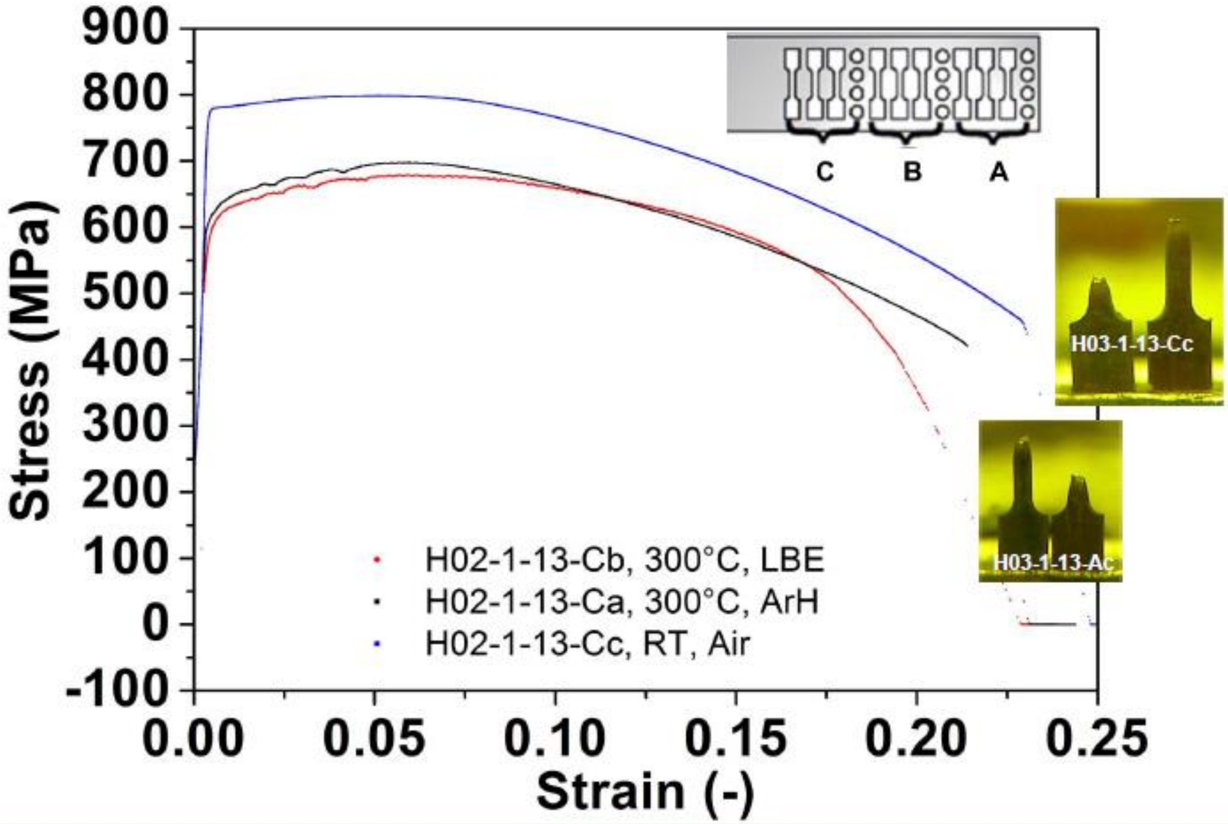
Eng. strain-stress tensile curves of specimens of MEGAPIE T91 Calotte

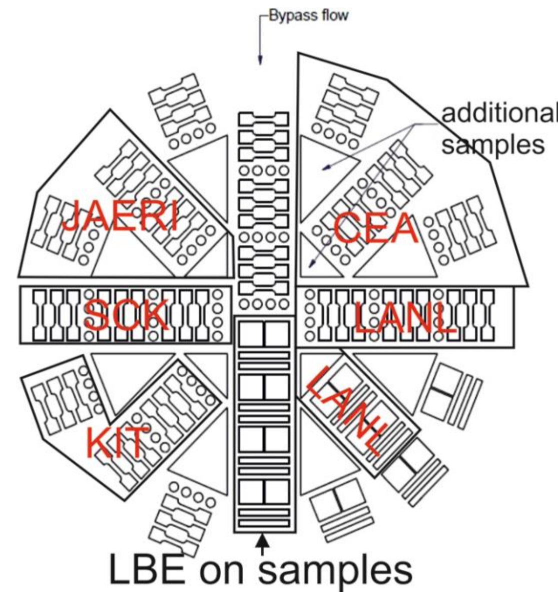




Results of tensile test show the T91 steel in the beam window area persists significant ductility after irradiation.

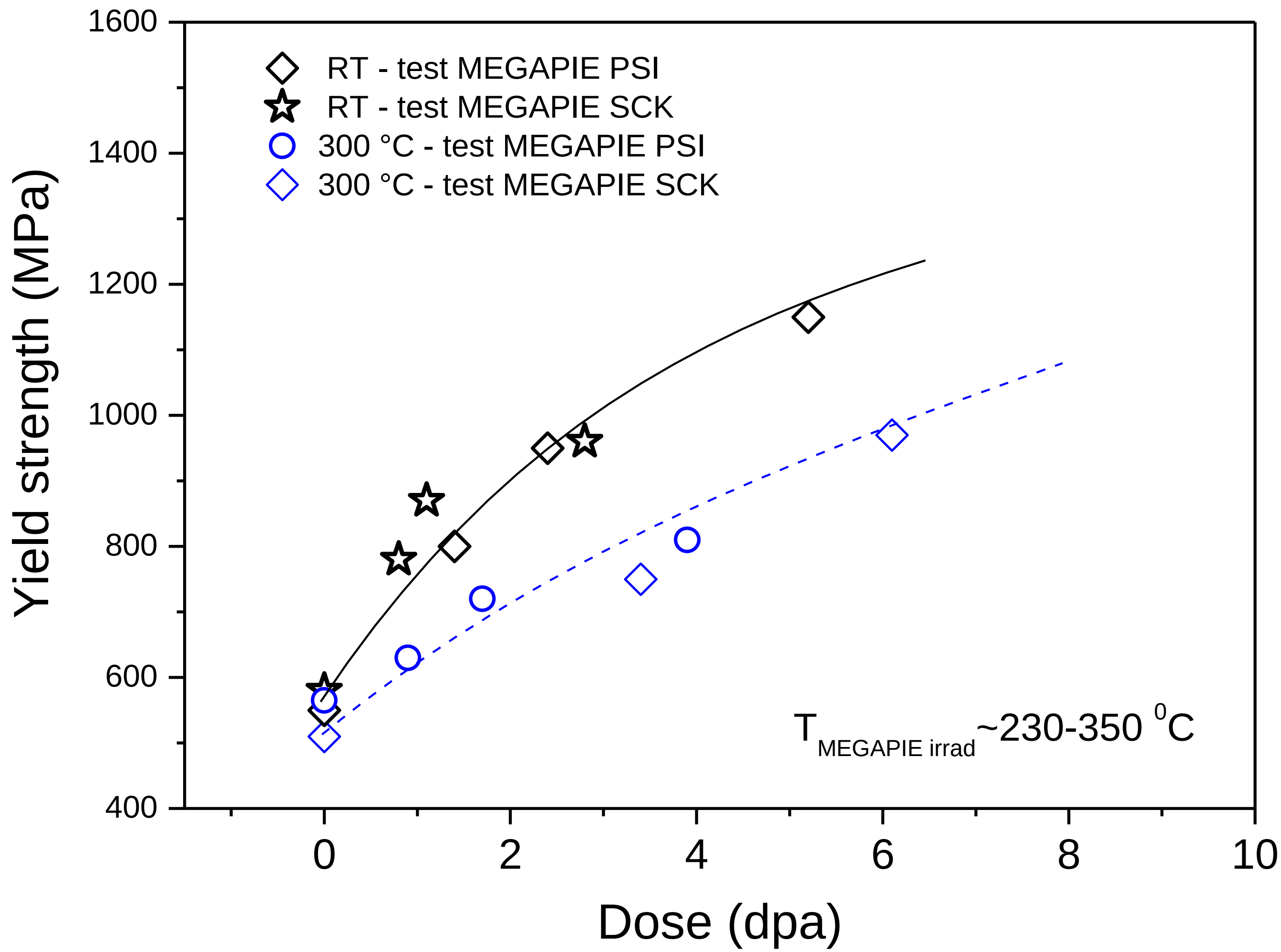
PIE Analysis – LLMC (T91) – Tensile Test





SCK•CEN
STUDIECENTRUM VOOR KERNENERGIE
CENTRE D'ETUDE DE L'ENERGIE NUCLEAIRE

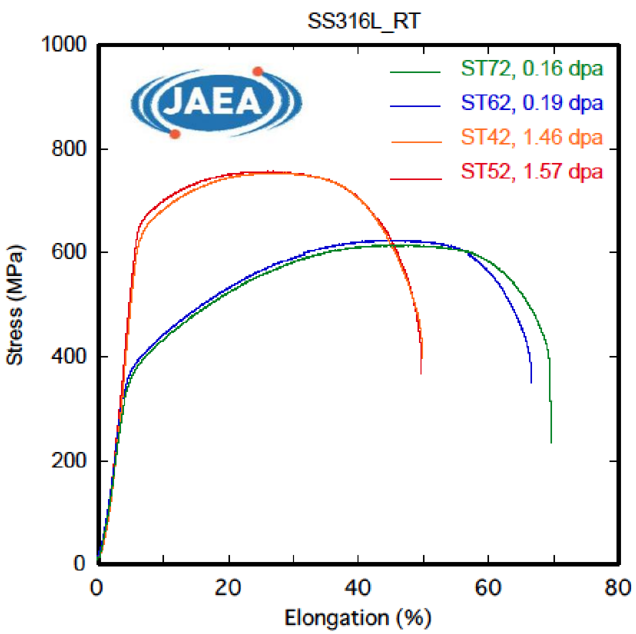
PIE Analysis – LLMC (T91) – Tensile Test

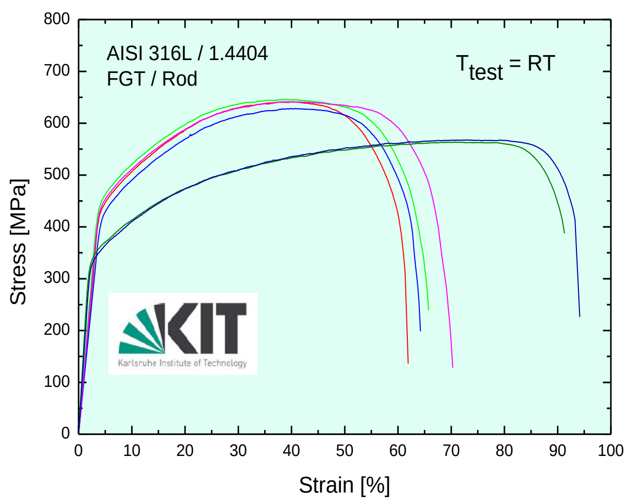


Tensile test results obtained at SCK·CEN and PSI consistent

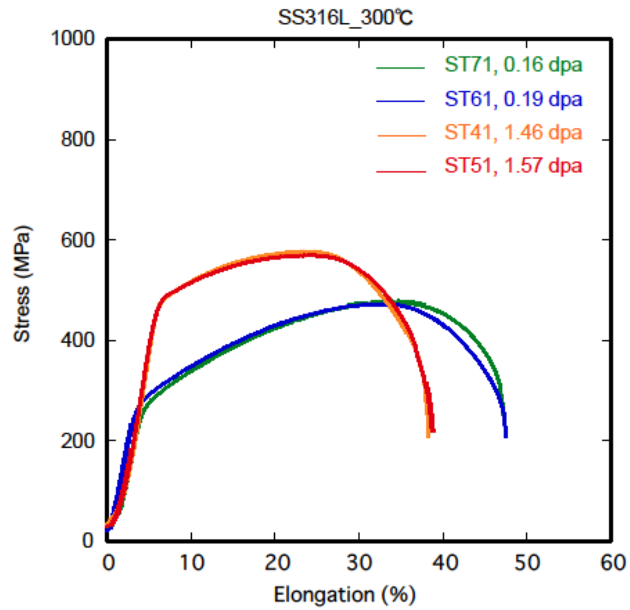
PIE Analysis – FGT (SS316L) – Tensile Test

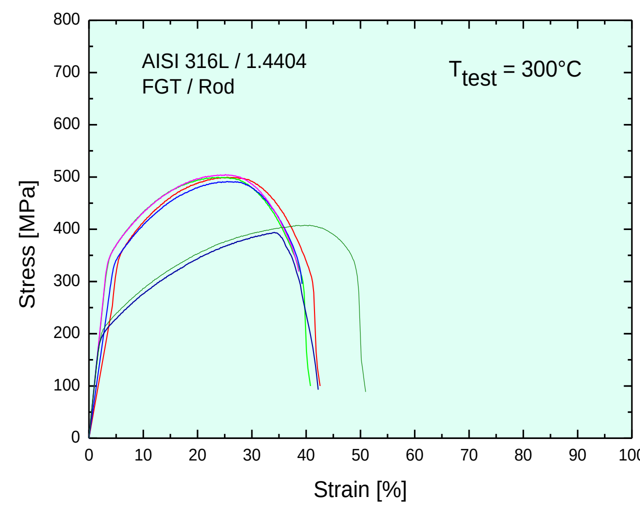
RT tests





300℃ tests





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PIE investigation – Summary

The PIE has been performed on the structural materials, T91 and SS 316L steels, irradiated in the lower part of the MEGAPIE target.

The main conclusions are:

•No evident failure and detectable change in thickness in the beam window area were observed.

•Slight corrosion damage was detected on the inner surface of the beam window.

•A slight misalignment was observed at the EBW of the lower liquid metal container.

•Results of tensile test show the T91 steel in the beam window area persists significant ductility after irradiation.

A MEGAPIE project meeting is planned to be held in the second half of 2017.

PIE results will be reviewed and subsequently published.

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Publications of PIE of MEGAPIE

Y. Dai et al., *Non-destructive testing of the MEGAPIE target ,* Journal of Nuclear Materials, 468, pp. 221-227, 2016.

M.J. Konstantinović et al., *Comparison of the mechanical properties of T91 steel from the MEGAPIE, and TWIN-ASTIR irradiation programs,* Journal of Nuclear Materials, 468, pp. 228-231, 2016.

B. Hammer-Rotzler, et al*. Radiochemical determination of 129I and 36Cl in MEGAPIE, a proton irradiated lead-bismuth eutectic spallation target,* Radiochimica Acta, 103 (11), pp. 745-758, 2015.

B. Hammer, et al. *Analysis of the 207Bi, 194Hg/Au and 173Lu distribution in the irradiated MEGAPIE target,* Journal of Nuclear Materials, 450 (1-3), pp. 278-286, 2014.

Ch. Latge et al. *Decommissioning and PIE of the MEGAPIE spallation target,* International Nuclear Fuel Cycle Conference, GLOBAL 2013: Nuclear Energy at a Crossroads, 1, pp. 651-657, 2013.

E. Donegani, et al. *Calculation of the radiation damage of the beam entrance window and other structural components of the Megapie target,* 10th International Topical Meeting on Nuclear Applications of Accelerators 2011, AccApp 2011, pp. 242-248, 2011.

Thank you for your attention.