Technology and Components of Accelerator Driven Systems (TCADS-2)

The MEGAPIE PIE sample preparation

Motivation

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 ($T_m=125^\circ\text{C}$)

"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."

Timeline of MEGAPIE

- Design Phase (Target + Ancillary systems) 1999 – 2004/5
- Construction Phase 2004/5 – 2006
- Cold Test of Target Operation 2005/6
- Preparation of 1st Dismantling/Disposal Phase in ZWILAG 2006 – 2008/9
- 1st Dismantling/Disposal Phase in ZWILAG August – December 2009
- Preparation of PIE sample production in the PSI Hot Laboratory 2010 – April 2011
- PIE Sample Production April 2011 – March 2013
- Final Disposal of Remaining Waste
Before & After Irradiation

T91 Lower Liquid Metal Container (Calotte + Leak Detector)

EBW

Material Deposition
Cutting & Disposal of MEGAPIE at ZWILAG

Cutting Plan of MEGAPIE – Target Sample Pieces
MEGAPIE PIE Sample Preparation

- Safety Analysis of each Handling Step and Clearance from the Safety Authorities (mainly Swiss Nuclear Safety Inspectorate, ENSI)
- Non Destructive Tests (NDT)
  - Determination of the Time-Averaged Proton Beam Profile
  - Visual Inspection (LBE Leak?)
  - Analysis of Black/Whitish Material in the BEW Region
  - Measurement of Thickness of T91 BEW (Corrosion Effects ?)
- LBE (and Absorber Foils) Sample Taking & Analysis
- Segregation of LBE and Structural Material (Steel) Parts
- First Conditioning of Waste in the Hot Laboratory
- Raw Cutting of Structural Material Sample Pieces
- Cleaning of Raw Cut Structural Material Sample Pieces
  - Mechanical Cleaning
  - Chemical Cleaning
- Electrical Discharge Machining (EDM) of PIE Sample Groups
- Packing & Transport of PIE Sample Groups to Partner Laboratories
- Preparation of Structural Material Samples with LBE Layer
- Final Waste Disposal and Cleaning of the Hot Cell(s)
Gamma-Mapping (Time-Averaged Proton Beam Profile)
- Spatial Measurement of Na-22 Activity ($2 \times 2 \text{ mm}^2$)
- Input for Damage Calculations (dpa) of MC models
NDT – Analysis of Material in BEW region

Visual Inspection: No LBE Leak detected

Optical Microscopy

Layer 1

Layer 2

Layer 3

Layer 2

Layer 3

Layer 1
NDT – Analysis of Material in BEW region

Optical Microscopy

Analysis via Energy-Dispersive X-ray Spectroscopy (EDX):

Material mainly Carbon

Possible Oil Leak ?!
US Thickness Measurements before (no LBE inside LLMC) and after Operation (LBE inside LLMC)

Systematic difference of 20 µm due to presence of LBE.

After correction for systematics: **NO DIFFERENCE IN THICKNESS** within measurement accuracy

No (disolution) corrosion effects for T91 detected.
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 Taking & Analysis
Results:

- The distribution of $^{207}$Bi is **homogeneous** within the LBE, with the exception of one sample from H07.
- Bulk LBE contains only **noble** metals that have a significant solubility in LBE.
- Elements with **low solubility in LBE** or **sensitive to oxidation** (rare earth elements) are only detected in samples taken at LBE/steel and LBE/cover gas interfaces (e.g. Lu, Hf, Ba).
- For $^{207}$Bi and $^{194}$Hg/Au good agreement between **theoretical** and **experimental results** are found.
Chemical Separation of Polonium

LBE sample (~12mg) dissolved in 7 M HNO₃

Distillation of Iodine and Chlorine

Spontaneous Deposition of Polonium onto Silver

Accelerator Mass Spectroscopy at ETH Zürich

α-spectroscopy

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

$^{209}\text{Bi}(p,xn)^{208-210}\text{Po}$

Part of the nuclide chart, www.nucleonica.com
α-Spectroscopy for Polonium

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

Polonium is homogeneously distributed in LBE (no enrichment)!
LBE melting – example H02

H02 BEW (T91)

H02 FGT (316L)
1st Waste conditioning - Cementation of oven

- Cement filling from top of the cell
- Control and conduct from front window of the cell
- Filling the LBE container
- Filling up the oven with cement
- Adding highly active waste pieces
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
The original cleaning scheme of raw cut sample pieces is a 4-step process proposed by Yong Dai:

1. "Mechanical cleaning" with heated oil (UCON-HTF @ 190°C max.)
2. Ultrasonic bath in de-ionized H₂O
3. "Chemical cleaning" in 5 molare HNO₃
4. Ultrasonic bath in de-ionized H₂O

Measurements after step 4 showed still 'too high' removable (α-) activity.

Tests to clean with EDTA (Ethylenediamin-tetra-acetic di-sodium-salt) instead of H₂O. Not significantly better, but disposal more complicated → cleaning continued with H₂O.

2 – 4 additional cleaning steps in ultrasonic H₂O bath per target sample piece.
Cleaning raw cut sample pieces

- 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.
Cleaning raw cut sample pieces

- Raw cut samples remained in HNO₃ bath for 3 minutes 30 seconds.
- US H₂O baths followed.
Time evolution in HNO$_3$ acid

After 1 minute in HNO$_3$

Gravity

After 2 minutes in HNO$_3$

LBE dissolved in the 5 molare HNO$_3$ acid and moves down the raw cut samples.
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 H₂O) further cleaning with H₂O

2 – 4 additional cleaning sessions with de-ionized H₂O
EDM cutting plan for the T91 BEW

M 1:1

Bypass flow

H02-1-16-Spitze
H02-1-14-Spitze
H02-1-15-Spitze
H02-1-13-Spitze
H02-1-12-Spitze
H02-1-11-Spitze
H02-1-10-Spitze
H02-1-09-Spitze
H02-1-08-Spitze
H02-1-07-Spitze
H02-1-06-Spitze
H02-1-05-Spitze
H02-1-04-Spitze
H02-1-03-Spitze
H02-1-02-Spitze
H02-1-01-Spitze

H02-1-01
H02-1-02
H02-1-03
H02-1-04
H02-1-05
H02-1-06
H02-1-07
H02-1-08
H02-1-09
H02-1-10
H02-1-11
H02-1-12
H02-1-13
H02-1-14
H02-1-15
H02-1-16
EDM cutting

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**EDM type: AGIE 250**

**Technical data:**
Dimensions: 2000x1290x1300mm
Weight: 1350kg

Cutting samples without LBE

- Set-up of the EDM machine in hot cell → 5 weeks
- Inactive cutting tests performed in hot cell, before cutting active raw cut sample pieces.
- Only then active cutting.
H02-1-7 has been successfully cut on 5th of November 2012.

While cutting the sample group H02-1-7-B the wire of the EDM broke. Reason unclear.

Cutting speed: 2 - 3 groups per day.
EDM cutting – H03-1/2

H03-1-01-A

H03-2-B2
All sample groups without LBE have been cut successfully.

No major failures. Wire broke several times.

Exchange of wire rather frequently.
PIE sample Packing and Transport

Sample holders put into sample containers

KIT transport left PSI on April 4th 2013.
All PIE sample groups have successfully been produced. In total more than 750 structural material samples and roughly 80 LBE samples have been produced.

All sample transports have left PSI (370 samples). Besides JAEA all Partner Institutes have received their samples.

The final waste disposal has started and is planned to be finished by end of June 2013. By then the MEGAPIE PIE sample preparation phase (and the full disposal of the MEGAPIE Target) will officially be finished.

All Partners prepare for PIE of structural material samples.

LBE sample and absorber foil analysis ongoing.

Results of all Partner Laboratories will finally be jointly published.
We would like to express our gratitude to our colleagues from all partner laboratories which have been working on MEGAPIE in the last 14 years.

Moreover, we would like to thank the team of ZWILAG for their dedication and the excellent team work during the dismantling phase of MEGAPIE in their hot cell.

Thank you for your attention.
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