DE LA RECHERCHE À L'INDUSTRIE

MEGAPIE-TEST

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# MEGAPIE: the world's first high-power liquid metal spallation neutron source.

**Ch. Latge[1]** (CEA) France ( christian.latge@cea.fr ), **M. Wohlmuther[2]** (PSI) Switzerland (michael.wohlmuther@psi.ch) ,

**Y. Dai[2], D. Gavillet[2], A. Gessi[3], A. Guertin[6], B. Hammer[2], S. Heinitz[2], J. Henry[1], M. Konstantinovic[4], R. Lindau[5], C. Fazio[5], S. Maloy[8], J. Neuhausen[2], S. Saito[7], K. Park[9], P. Roubin[1], K. Samec[2], D. Schumann[2], K. Thomsen[2], A. Türler[2], L. Zanini[2], W. Wagner[2]**

1 CEA Cadarache DEN-DTN 13108 Saint-Paul-lez-Durance France, 2 PSI Villigen; 3 ENEA Brasimone; 4 SCK-CEN Mol; 5 KIT ; 6 CNRS Subatech Nantes; 7 JAEA Tokai; 8 DOE-LANL, 9 KAERI

*Thorium Conference Geneva (Switzerland)*

*Oct 27th Oct 29th 2013*

# MEGAPIE EXPERIMENT

**A key experiment in the ADS roadmap:**

**MEGAwatt PIlot Experiment (MEGAPIE) (1 MW)** initiated in 1999 in order to design and build a liquid lead-bismuth spallation target, then to operate it into the Swiss spallation neutron facility SINQ at PSI .

It was to be equipped to provide the largest possible amount of

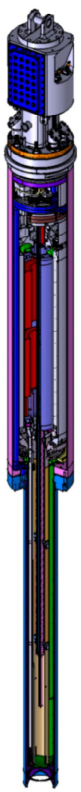
scientific & technical information without jeopardizing its safe operation.


SINQ @ PSI

**Several main challenges for the MEGAPIE project:**

- **to design a completely different concept of target in the same geometry of the current spallation targets used at PSI.**
- **to develop and integrate two main prototypical systems : a specific heat removal system and an electro magnetic pump system for the hot heavy liquid metal in a very limited volume.**
- **to design a 9Cr martensitic steel (T91) beam window able to reach the assigned life duration.**
- **to license a LBE in relevant conditions**
- **to operate a LBE target**
- **to develop the decommissioning strategy and waste management**
- **to characterize LBE and structural material (PIE)**

# MEGAPIE TARGET



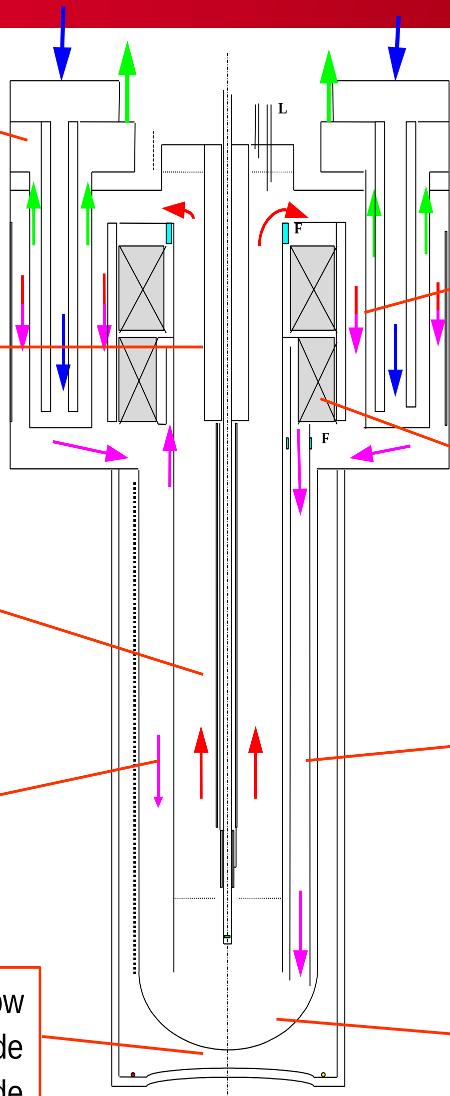
Heat Exchanger (Oil)
10 l/s, 5.5 m/s
140-175°C inside

Main Pump
4 l/s, 1.2 m/s
380°C

Guide Tube
4 l/s, 0.33 m/s
380°C

Downcomer
3.75 l/s, 0.33 m/s
230-240°C

Beam Window
380°C outside
330°C inside




Heat Exchanger
4 l/s, 0.33 l/s/pin
0.46 m/s
380-230°C inside

Bypass Pump
0.25 l/s, 0.2 m/s
230°C

Bypass Tube
0.25 l/s, 1m/s
230-240°C

Nozzle 1.2 m/s
Beam Window 1 m/s

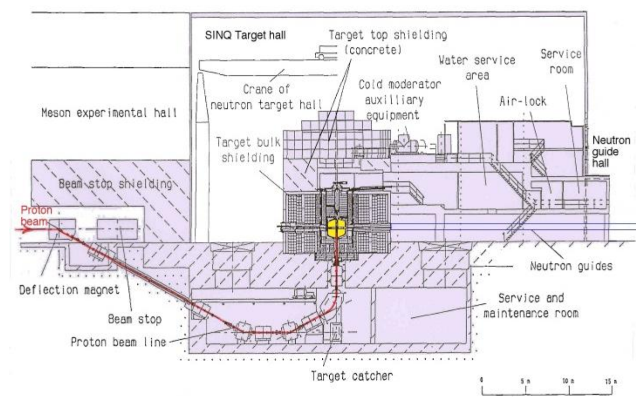
## Design parameters:

| | |
|---|---|
| p-beam energy: | 575 MeV |
| p-current: | 1.74 mA |
| Heat removal: | 650 kW |
| Design pressure: | 16/10 bar |
| Design temp.: | 400°C |
| Cover gas press: | 3.2 bar |
| Operation: | 1 year |

with max 6000 mAh
Radiation damage: 20-25 dpa

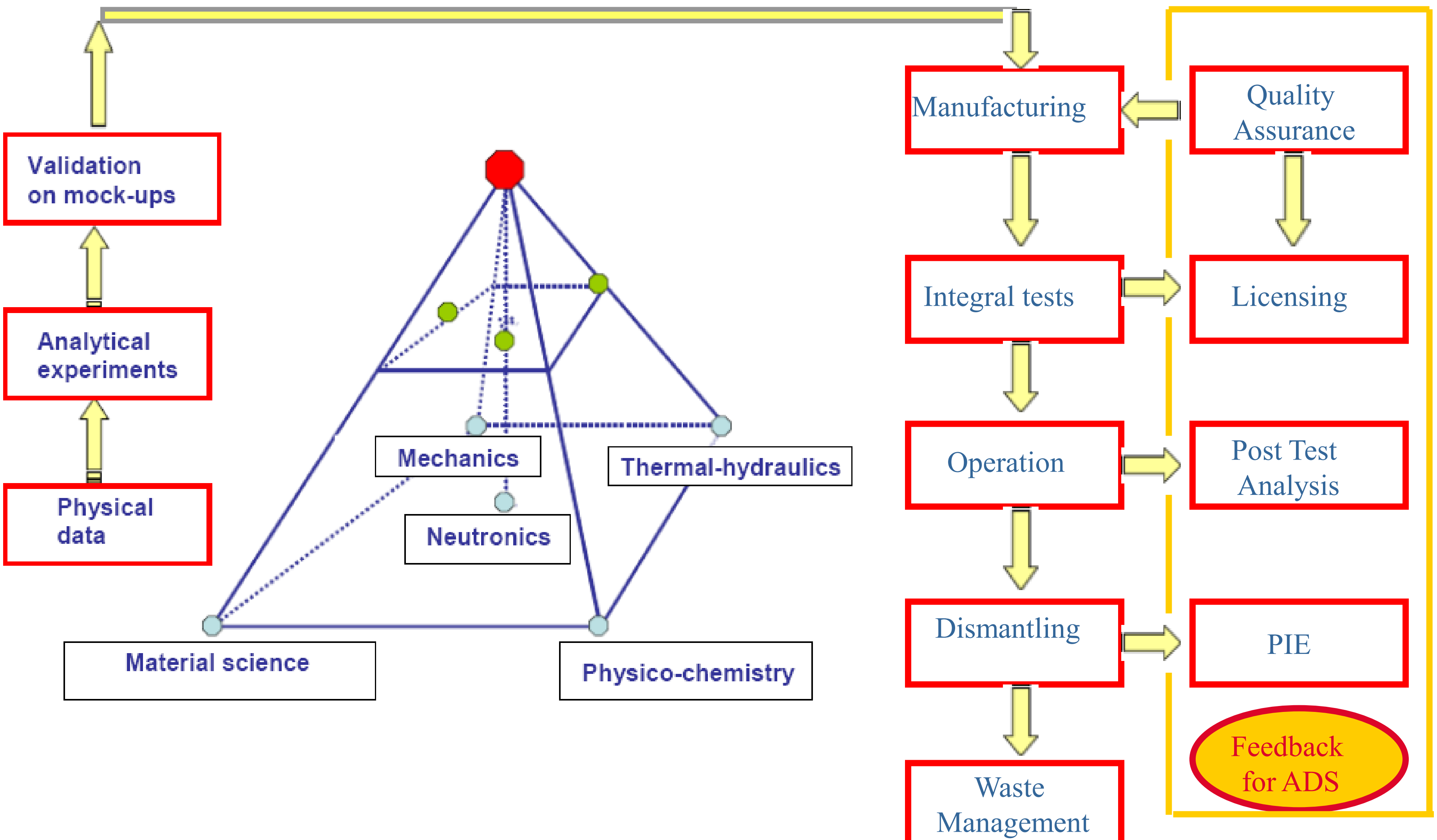
## Dimensions

| | |
|---|---|
| Length: | 5.35 m |
| Weight: | 1.5 t |
| LBE-Volume: | 89 l |



# MEGAPIE PROJECT: DEVELOPMENT STRATEGIE

- ➔ Numerical simulation + experiments : from basic science to engineering tools for design & operation
- ➔ Progressive validation of concept by basic studies, design calculations, integral tests
- ➔ Operation with Post Test analysis and Post Irradiation Examination
- ➔ Decommissioning and Waste management



# MEGAPIE PROJECT: MAIN STEPS

Requirements definition, organization: **1999-2000**

Feasibility studies : **2000**

Design studies: **2001-2004**

Design support: **2001-2005**

Manufacturing target & ancillary systems: **2004-2005**

Integral Test: **September to December 2005**

Transfer to SINQ: **January to Mai 2006**

Irradiation : **August 2006 to December 2006**

Post Test Analysis: **2007-2009,**
Decommissioning: **2009-2012**
Sampling for PIE: **2011-2012**
PIE: **2013 & 2014**
Waste management: **2011-2013**

# CHOICE OF LEAD-BISMUTH

**Lead bismuth eutectic (Pb44.5%-Bi55.5%) has been selected, due to its attractive neutronic and physical properties :** heat transfer coefficient, low melting point (125°C); nevertheless bismuth induces the production of activation products i.e. polonium,... under irradiation.

| Property | | Pb | Bi | LME * | LBE** | Hg |
|---|---|---|---|---|---|---|
| Composition | | elem. | elem. | Pb 97.5% Mg 2.5% | Pb 45% Bi 55% | elem. |
| Atomic mass A (g/mole) | | 207.2 | 209 | 202.6 | 208.2 | 200.6 |
| Density ($g/cm^3$) | 20°C | 11.35 | 9.75 | | | 10.5 |
| | liquid | 10.7 | 10.07 | 10.6 | 10.5 | 13.55 |
| Linear coefficient of thermal expansion ($10^{-5}$ $K^{-1}$) | solid | 2.91 | 1.75 | | | |
| | liquid (400°C) | 4 | | 4 | | 6.1 |
| Volume change upon solidification (%) | | 3.32 | -3.35 | 3.3 | 0 | |
| Melting point (°C) | | 327.5 | 271.3 | 250 | 125 | -38.87 |
| Boiling point at 1 atm (°C) | | 1740 | 1560 | | | 356.58 |
| Specific heat (J/gK) | | 0.14 | 0.15 | 0.15 | 0.15 | 0.12 |
| Thermal neutron absorption (barn) | | 0.17 | 0.034 | 0.17 | 0.11 | 389 |

* LME - lead/magnesium eutectic ** LBE - lead/bismuth eutectic

# FEEDBACK FROM BASIC STUDIES IN SUPPORT TO THE DESIGN

**The reasons for the choice of T91, compared to austenitic steel 316L, were the following ones:**
- higher strength
- much better resistance to heat deposit (due to a lower thermal expansion coefficient and a higher thermal conductivity).
- better corrosion resistance in Pb-Bi due to a low nickel content

**➔Many studies related to :**

**O control,**

**O measurement with ECOM**

**corrosion,**

**lead-bismuth freezing & consequences,**

**mechanical properties**

**lead-bismuth water interaction,…**

**Fundamental role of the natural surface oxides on the evolution of the corrosion of the two steels demonstrated,**

➔The oxide layer expected at the window surface delays the steel dissolution of the steel:

With $1 m.s^{-1}$ flow rate, 400°C

0.05m hydraulic diameter (without any specific turbulence) and

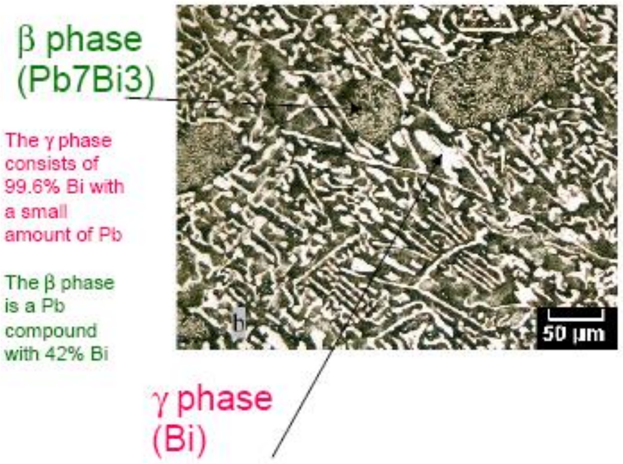
$10^{-10} < [O] < 10^{-11}$ wt%,

➔ expected mean T91 steel corrosion rate: between 45 and 130 µm yr-1 for a few thousand hours period.

# EXPANSION OF SOLID LBE AFTER RE-CRYSTALLIZATION

**LBE re-crystallization process was well understood:** when the solid alloy gets richer in γ phase, when temperature decreases at T lower than Tf (the melting temperature), the excess γ phase precipitates. The γ phase is richer in Bi than the β phase, and bismuth (unlike lead) expands on solidification. So the γ phase precipitation and the atoms' migration from grain to grain (i.e. re-crystallization) generate a volume increase.

**<u>Dedicated study:</u>** the expansion could be mitigated if the cooling rate was kept as low as 0.02 °C/min from solidification point to 60 °C: this specific procedure for Freezing the LBE in Lower Target Enclosure was validated by thermo-mechanical calculations using 2-D ANSYS Model.




**LBE recrystallization (Courtesy of ENEA)**

# FEEDBACK FROM TARGET MANUFACTURING

**Relevant issue: development and assessment of suitable joining and welding technologies of the selected materials for Target in different geometries e.g. window, tubes of heat exchanger...**

**Joining and welding procedures qualified for various configurations, including preparation, welding and control.**

**Target manufacturing by ATEA company in Nantes (France)**

# MEGAPIE – Target components (ATEA-IPUL-PSI)

Target window: T91, 1.5 mm

Target heat exchanger
12 pin LBE – Diphyl THT

Target head
Connector interface to ancillary systems

Main and by-pass tube
LBE flow guide

# FEEDBACK FROM INTEGRAL TESTS

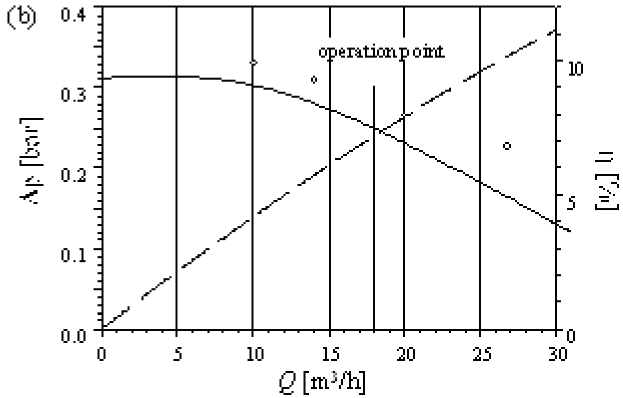
Integral Test:

- to integrate the target and the ancillary systems,
- to carry out LBE filling and draining operations
- to check the operability of the main components of the target,
- to check and calibrate the instrumentation (mainly flow-meters)
- to determine the thermal hydraulics characteristics of the system,
- to simulate heat deposition (with a heater)
- to characterize the heat transfer and hydraulic behavior at the window,
- to obtain the vital parameters for system control,
- to provide enough information for licensing the target for irradiation test,
- to gather technical and scientific data for model verification, in order to be able to extrapolate the Megapie experience for the future ADSs

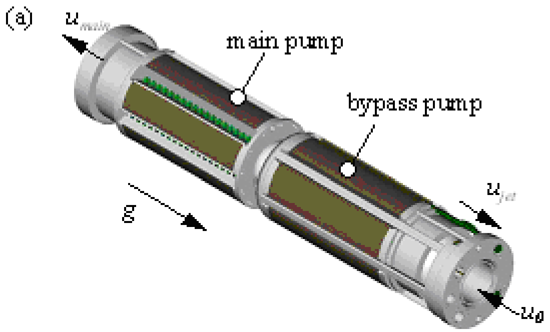
# FEEDBACK FROM ELECTRO-MAGNETIC PUMP SYSTEM (IPUL LATVIA)

**Circulation of the lead-bismuth alloy within the target:**

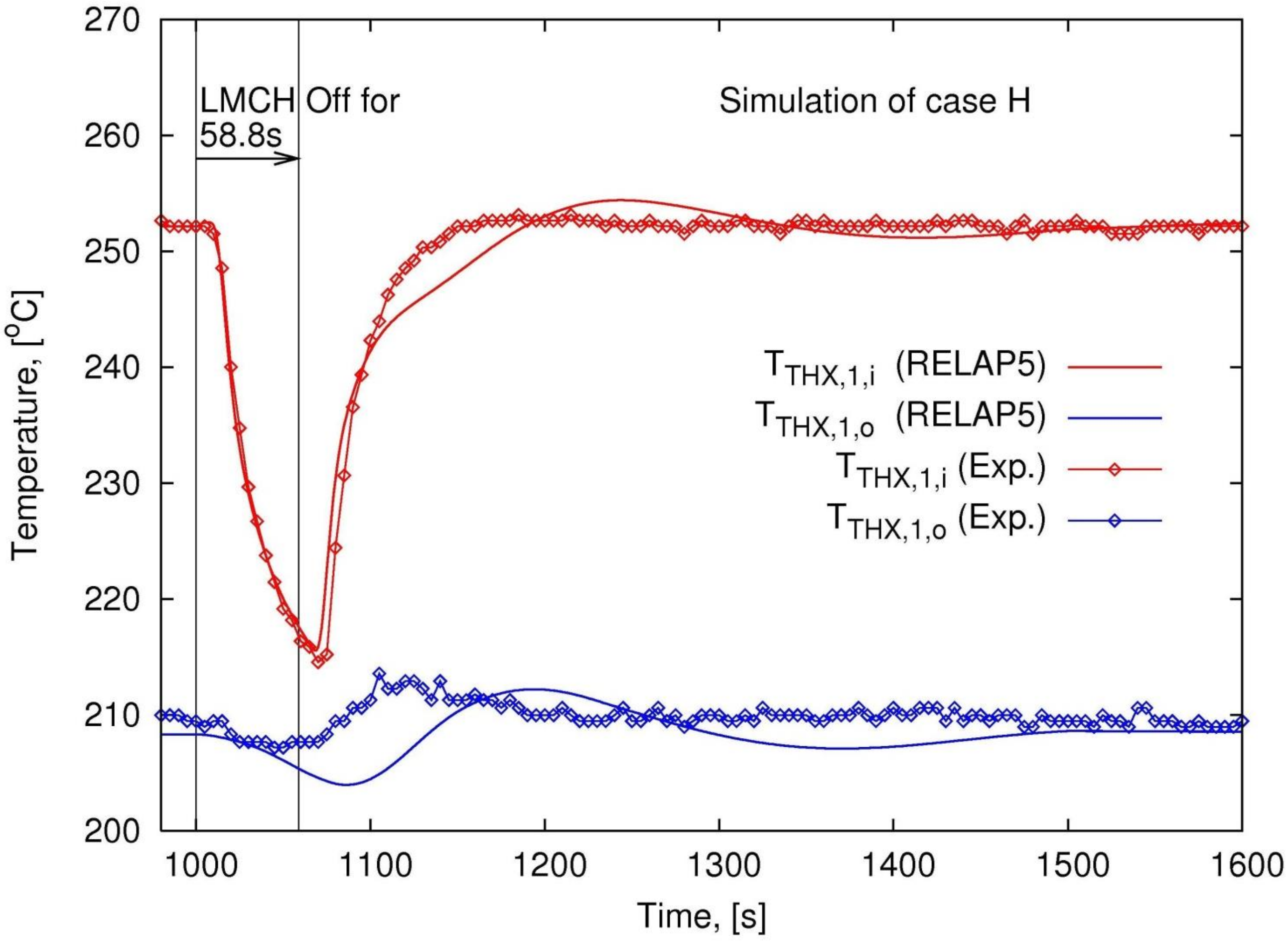
- **generated by two independently controllable annular linear induction pumps (ALIPs), immersed in the upwards directed riser flow (pumps delivered by Institute of Physics of Latvia (IPUL).**




*(b) Pressure head – flow-rate characteristics of the main flow electromagnetic pump for currents: 1 – 33A, 2 –30A, 3 – 25A, 4 – 20A, 5 – 15A, 6 – 10A, 7 – 5A; 8 – hydraulic characteristic of the target main channel.*



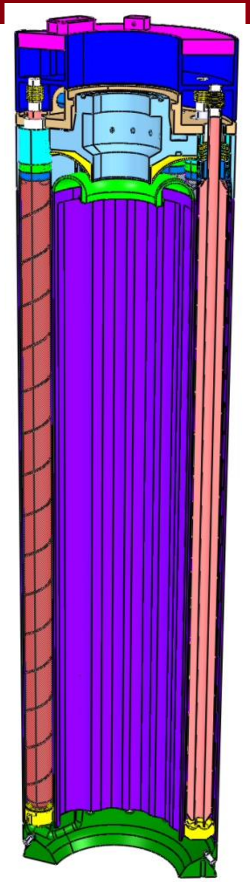
# EXPERIMENTAL / CALCULATED TEMPERATURES FOR AN UNPROTECTED TRIP.




RELAP model more or less verified but could be improved by a better estimation of :

- the thermal masses of LBE and structures (Cp,…)
- the heat transfers coefficients (using T of the main and bypass EMPs, ),

# INTEGRAL TESTS : THX DESIGN VALIDATION



**Sufficient capacity of the Heat Removal System to cope with about 600 kW of heating in the target and flexible to the changes, though the operating conditions might be differed from the predictions.**

**But : after the integral tests, bypass flow conditions still to be determined**

**Analysis of the experimental data of the LBE-oil thermal exchanger of the target ( with analytical heat exchanger calculations (Global model, e-NUT, and numerical model (1D), finite volumes) :**

**→For each of the 4 campaigns, computed values in agreement with the corresponding experimental results.(maximum variance between calculations and experiments very low, and below the accuracy of the model is about 20%).**

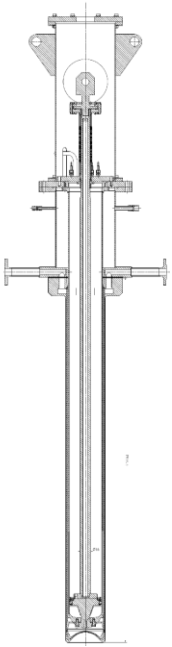
**→Thus, THX heat transfer model (and correlations) used to its design, validated, even if some uncertainties hang over flow rates assessments.**

**→Parametric study of sensitivity : large margins on the THX thermal exchange capacity.**

# FEEDBACK FROM FULL SCALE LEAK TEST

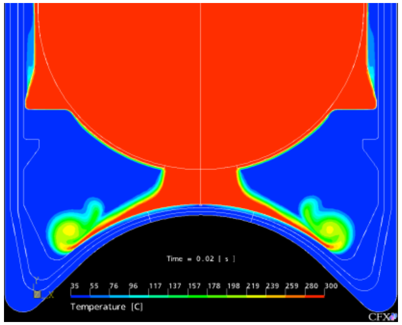
**Lower Target Enclosure able to contain Pb-Bi.**

**Leak detector system which had the following main requirements : detect a leaked LBE quantity below 0.5 liter within 1 second with a very high reliability, 100 % detection efficiency, a very low false alarm rate, and radiation and temperature resistant.**



**2 different sensors implemented:**

- **Thermocouples** (9 individual and independent sensors, 3 electrically preheated) as main leak sensor, and,
- **Stripe sensors type "AC impedance"** (3 separate units) (developped by PSI Switzerland)




➔ **Leak Detector system fully validated during Full Scale Leak Test**

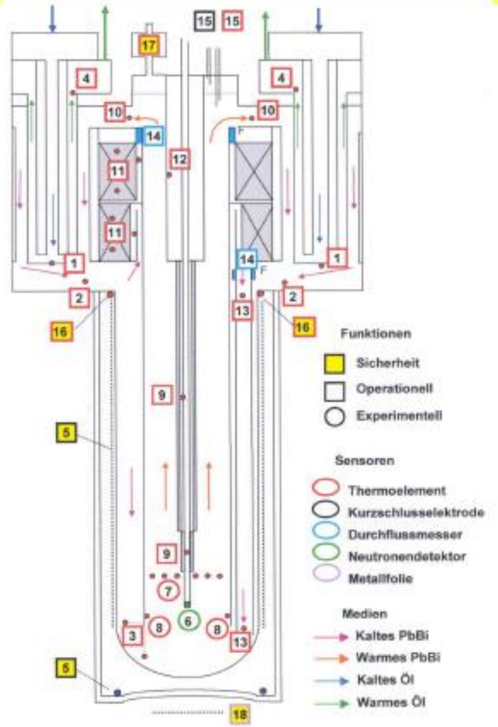
# IRRADIATION

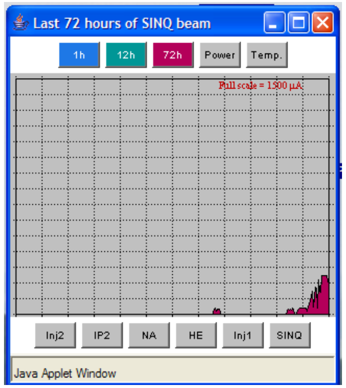
## Main goals :

- confirm the main operating parameters,
- obtain neutronic performances, in order to validate the neutronic codes and spallation models
- to confirm material performances evaluated during design support phase,

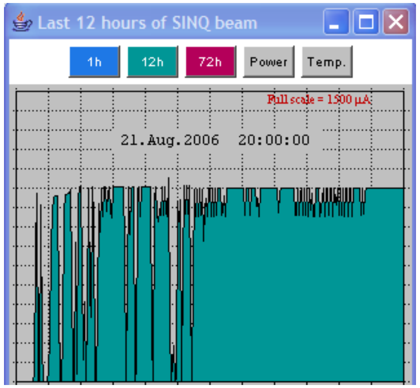
## Instrumentation

86 Thermocouples
2 Pressure transducers
5 Level Detectors
2 Flowmeters
19 Leak detectors




Aug 14th 2006




Aug 21st 2006

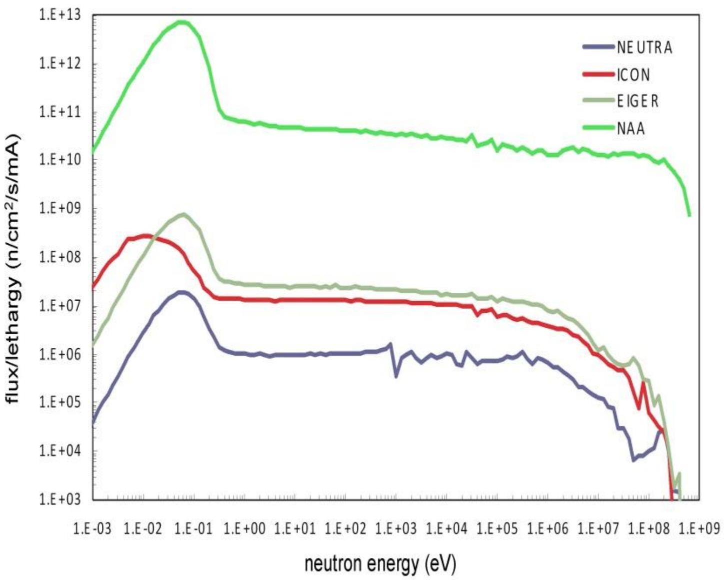
# MAIN RESULTS ON NEUTRONIC PERFORMANCE & GAS PRODUCTION

Mapping of the neutron flux in its different components (thermal, epithermal, fast) in the SINQ facility around the target.

Innovative measurements inside the MEGAPIE target

Measurements complemented by corresponding calculations: code validation

Comparison with solid targets



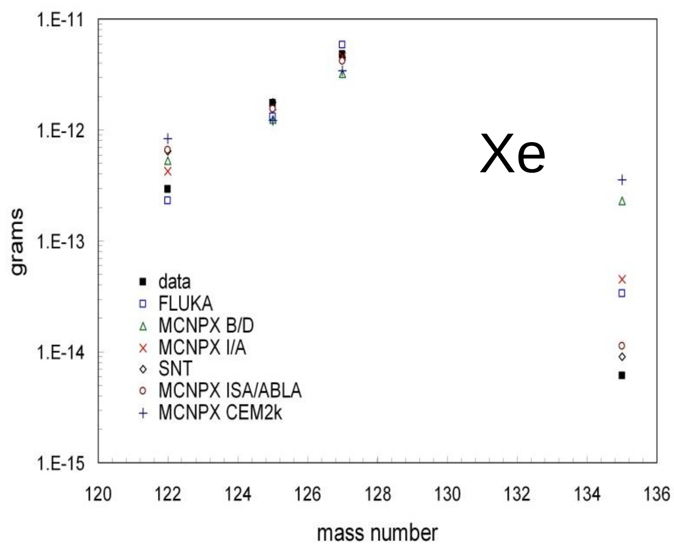

Successful measurements of radionuclides at different irradiation times

Estimation of the release rates at the beginning of irradiation and towards the end.

Good agreement with calculations

Confirmation of results on release of Hg and Po from the design phase

No mass measurement of stable isotopes, only indirect information from pressure measurements



# DECOMMISSIONING PHASE 1/2

➔ After the irradiation, target up to mid February 2007 in the operating position until the decay heat has decreased to about 300 W.

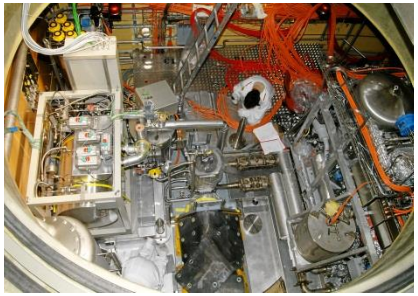
➔ Controlled freezing of LBE necessary due to expansion of solid LBE after re-crystallization:

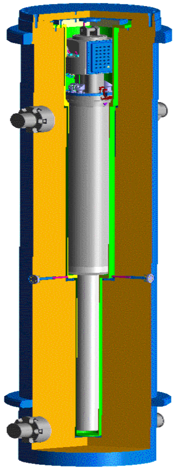
➔ Mitigation possible if cooling rate kept as low as 0.02 °C/min from solidification point to 60°C. (specific procedure for freezing the LBE in Lower Target Enclosure)

➔ Cooling circuits and gas volumes emptied, rinsed and dried,

➔ Target disconnected and sealed up with blind flanges, then stored for several months.

➔ Target transferred to ZWILAG hot laboratories, using a steel container.





Container for target transportation

# DECOMMISSIONING PHASE 2/2

➔ Target cut with band saw (provided by Behringer) in 19 slices.

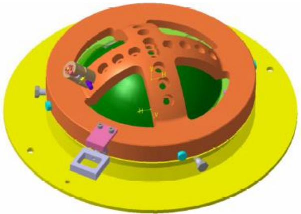
➔ About 8 (weight) % of the target transported to the PSI Hot labs

as samples material for PIE.

➔ Remaining target pieces (92%) in steel cylinder in a KC-T12 concrete container (TC2), for Storage and Disposal.

➔ Visual inspection of the T91 window done:

➔ some materials deposited in the central area of the window (mostly aluminum window of safety hull):
after removing the materials no cracks or other damages observed.
➔ SEM investigations: porous structure, C, O with little amount of Si for the black material and Si, O with little C for the white material.

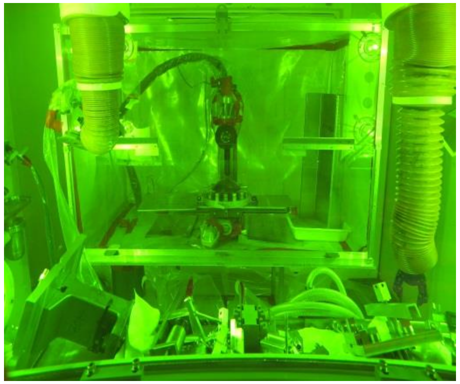


➔ US measurements on T91 window: no detectable change in the thickness of the window, (no evident dissolution/ corrosion effects). (validation of the choice not to control the oxygen activity in the LBE).

# POST IRRADIATION EXAMINATIONS

**Objectives of the PIE are to understand:**

- microstructural, mechanical and chemical changes in the structural materials in the target induced by irradiation and LBE corrosion,
- the production, distribution and release of the spallation and corrosion products in the LBE.



EDM cutting machine

➔ **Organized effort of 8 partners of the MEGAPIE initiative:** CEA, CNRS, ENEA, FZK, JAEA, LANL-DOE, PSI and SCK.

➔ **PIE program:** mechanical tests and microstructure analyses will be performed on different components, particularly on those in the lower part of the target with high irradiation doses.

➔ **Small specimens for the PIE** cut from the pieces with an EDM machine (Electrical Discharge Machining) and a diamond disc saw in PSI's hot cells.

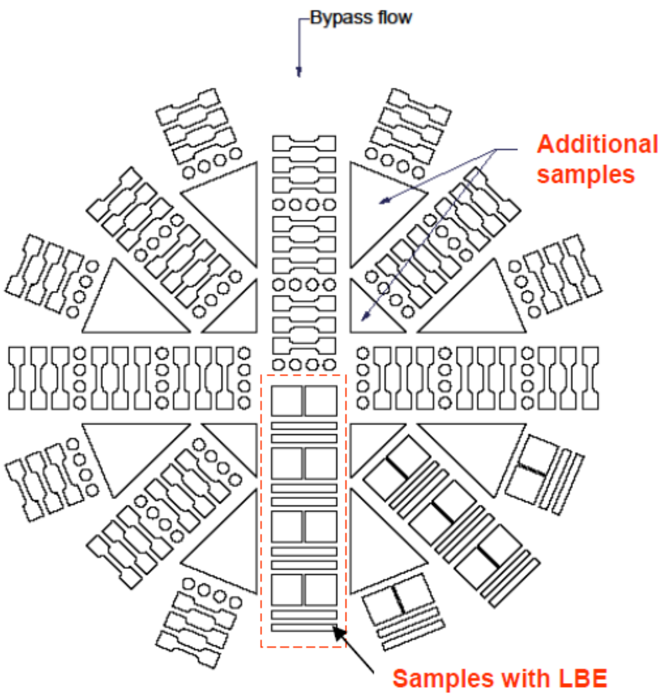
Samples produced by EDM machine

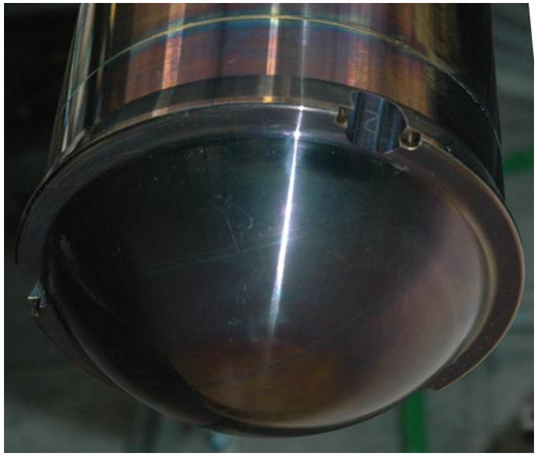
# PIE TESTS

**Specimens foreseen for mechanical tests and microstructure analyses of the target:**

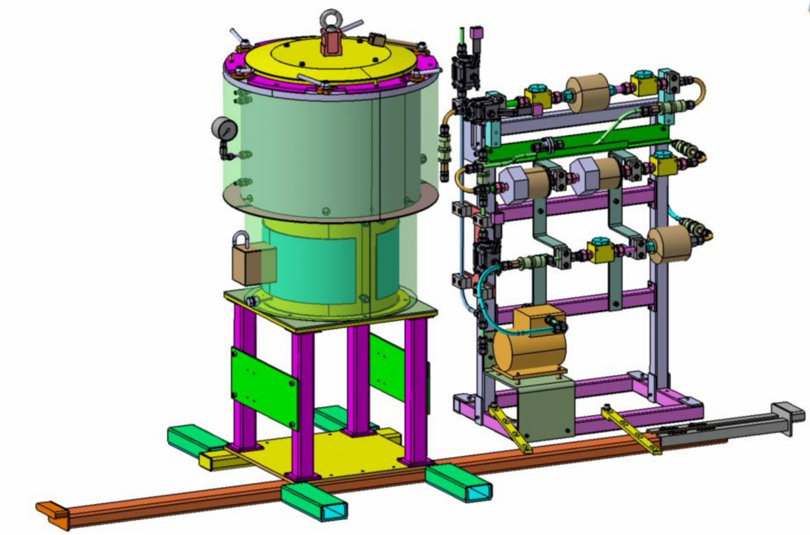
- **TEM (Transition Electron Microscopy),**
- **OM (Optical Microscopy),**
- **SP (Small Punch),**
- **Tensile and bending specimens.**

**➔ OM specimens also used for other surface analyses such as SEM (Scanning Electron Microscopy), EPMA (Electron Probe Micro-Analysis) etc**



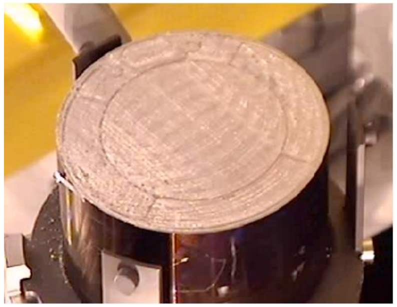



# SEGREGATION OF LBE



➔ To segregate the structural materials from the LBE **a special oven** has been designed and constructed by the engineering department of PSI.

➔ The oven is heated with 6 heaters (0.8 kW each) built into the intermediate floor on which the target sample pieces are positioned. The lower part of the oven serves as a collector of the LBE and can be separately heated (heater band of 1.5 kW).

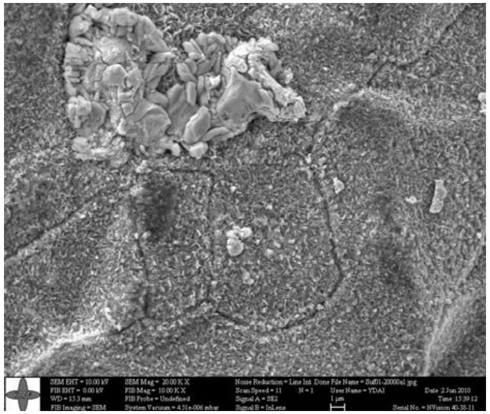


**Before melting**

**After melting**

# SAMPLE CLEANING & DECONTAMINATION

**Request done by some partners: specimens: contamination free.**



➔ **Necessity to clean the structures to remove the LBE, in a hot-cell after melting LBE and segmenting the large pieces into smaller ones.**

➔ **Process to clean LBE:**

**- to sweep LBE away with tissues after heating the specimens in oil up to about 150°C.**

**- to dissolve the remaining LBE on surface of samples, two different solutions evaluated:**

**-1 equimolar mixture of acetic acid, hydrogen peroxide and ethanol, (widely used for cleaning LBE on samples after corrosion tests)**

**-2 7M $HNO_3$.**

➔ **Option 2: could dissolve LBE much faster (at least 5 times) than 1.**

➔ **Option 1: gas and heat production looked higher than that in 2.**

➔ **Cooling needed during dissolving LBE in both cases.**

➔ **To solve the same amount of LBE, in 1 a much larger volume of the first mixture needed, producing more contaminated effluent.**

➔ **Decision to use nitric acid for the final cleaning of LBE.**

# TRANSPORT OF THE PIE SAMPLES

**Concerning the transports of the PIE samples to the partner laboratories (CEA, JAEA, KIT, LANL, SCK-Mol):**

**➔ investigations on the transportability made, based on calculations with detailed nuclide inventories for all samples.**

**➔ Transports performed as "UN2915, Radioactive Material, TYPE A" transports.**

**➔ All the transports done by end of April 2013:**

- **Belgium (SCK-Mol)**
- **France (Saclay),**
- **Germany (KIT)**
- **Japan (Tokai),**
- **USA (Los Alamos National Laboratory)**

**In addition to PSI Hot Labs**

# IRRADIATION PARAMETERS

➔ **Necessity to determine the irradiation parameters for all the specimens to be investigated, such as:**

**- irradiation dose (dpa),**

**- helium (He) and hydrogen (H) production,**

**- proton and neutron fluences,**

**- irradiation temperature etc**

➔ **Except for the irradiation temperature, the remaining parameters can be evaluated from the final (accumulated) proton fluence distribution in the target by neutronic calculations.**

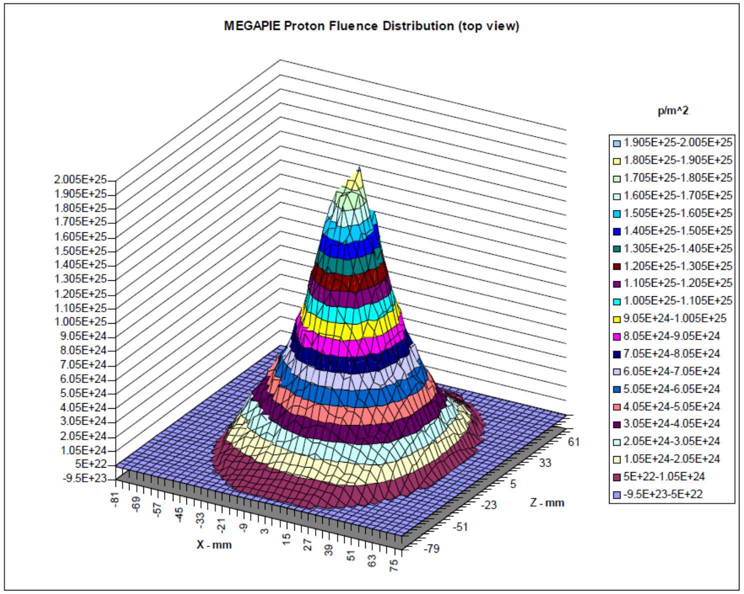
➔ **The irradiation temperature relies mainly on the instant proton beam current density which is known changing greatly but, unfortunately, could not be measured during the operation of the target.**

# PROTON FLUENCE DISTRIBUTION

➔ **To evaluate the distribution of the proton fluence accumulated after about four-month irradiation, gamma-mapping on the beam window of the aluminum safety-hull of the MEGAPIE target was conducted.**

- **Gamma-mapping performed in an area of 160x160 mm$^2$ in steps of 4 mm.**
- **At each measuring position a full gamma spectrum obtained.**
- **$^{22}Na$: the only radioactive isotope well detected, mainly produced by the incident high energy protons.**
- **To deduce the proton fluence distribution from the measured data, necessity to correct $^{22}Na$ counts because of differences in actual measuring volume and distance at different positions on the window.**

➔ **Distribution of proton fluence obtained and available for neutronic calculations of irradiation parameters.**




➔ ***Maximum proton fluence of the MEGAPIE target is 1.93E$^{+25}$ p/m$^2$***

# CFD CALCULATIONS FOR T MAPPING

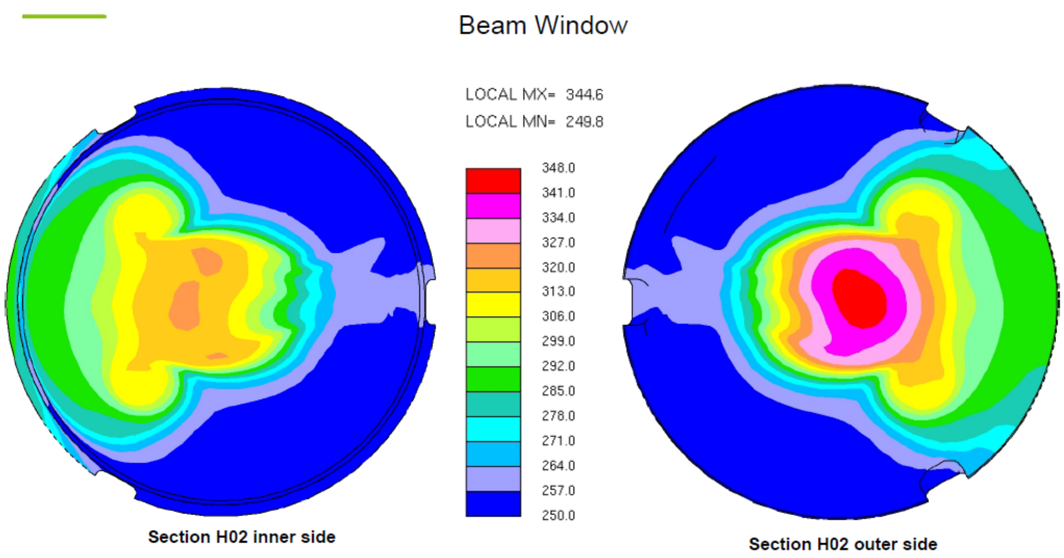
➔ Thermal hydraulics calculations carried out with CFD model to evaluate the temperature distribution in the lower part of the target,

➔ Comparison with temperatures measured with thermocouples.

➔ Some temperatures measured about 15°C higher than predicted.

➔ Some wrong attributed positions of the thermocouples could explain some of the calc/exp discrepancies.

➔ A significant difference of temperature calculated between inner side and outer side, up to 25 to 30°C at flow guide edge.



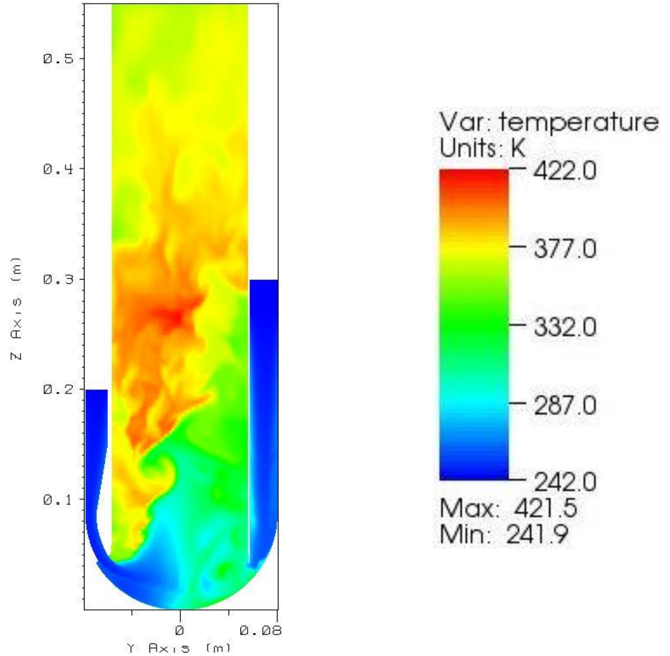
# T INSTABILITIES

**Instabilities, particularly due the effect of the by-pass pipe, and evidenced thanks to experiments with infra-red camera, during integral tests.**

**➔ Large Eddy Simulations (LES) carried out to analyse these instabilities, close to the window, with TRIO-U-VEF parallelized code, to assess near the window :**

- **level of temperature and**
- **velocity fluctuations**

**➔ to give realistic data for thermo-mechanical studies aiming to demonstrate the integrity of the T91 window.**




**➔ These data also show the difficulty to attribute an average temperature of LBE, close to the window.**

**➔ Nevertheless, average temperatures, taking into account the irradiation, need to be recalculated using the heat deposition data from the output of the neutronic calculations.**

# CHEMICAL & RADIOCHEMICAL ANALYSIS OF LBE

- **Decision to perform chemical and radiochemical analyses on spallation and corrosion products in LBE**, Ag-absorber, cold-trap (CGS).

- Samples taken from all slices before melting LBE (preparation of the samples for mechanical testing).

- Size of the LBE samples determined by the total activity that can be handled in the laboratories.

- **Distribution of the activity within the target material was not found homogeneous due to different phenomena such as deposition, transport during solidification and diffusion.**

➔ For an estimation of the maximum activity we have relied on the activation calculations: maximum activity that can be present in the LBE can be calculated assuming homogeneous distribution. ( assumption surely conservative, since the deposition of radioactive material lead to a decrease of the LBE-activity: such phenomena observed in a liquid LBE target irradiated at CERN ISOLDE).

➔ **at each individual sample position, pre-drilling/cutting necessary** to remove the contaminated surface, to avoid smearing of material during the sawing process and/or contamination with sawdust.

# RESULTS ON LBE SAMPLES 1/2

**The following results from chemical characterizations have been obtained:**

- **$\gamma$-spectrometry measurements of all specimens (without prior chemical separations) has been completed.**

**Results: $^{207}$Bi, $^{194}$Hg/Au and $^{173}$Lu distribution in the MEGAPIE target**

***The following radio-nuclides identified by $\gamma$-spectrometry: $^{60}$Co, $^{101}$Rh, $^{102}$Rh, $^{108m}$Ag, $^{110m}$Ag, $^{133}$Ba, $^{172}$Hf/Lu, $^{173}$Lu, $^{194}$Hg/Au, $^{195}$Au, $^{207}$Bi.***

- **For some of these nuclides the activities can be easily evaluated from $\gamma$-spectrometry results (e.g. $^{207}$Bi, $^{194}$Hg/Au), while other nuclides can only be determined after chemical separations (e.g. $^{108m}$Ag, $^{110m}$Ag, $^{195}$Au, $^{129}$I, $^{36}$Cl and $\alpha$-emitting $^{208-210}$Po).**

- **Bulk LBE contains only noble metals that have a significant solubility in LBE,**

# RESULTS ON LBE SAMPLES 2/2

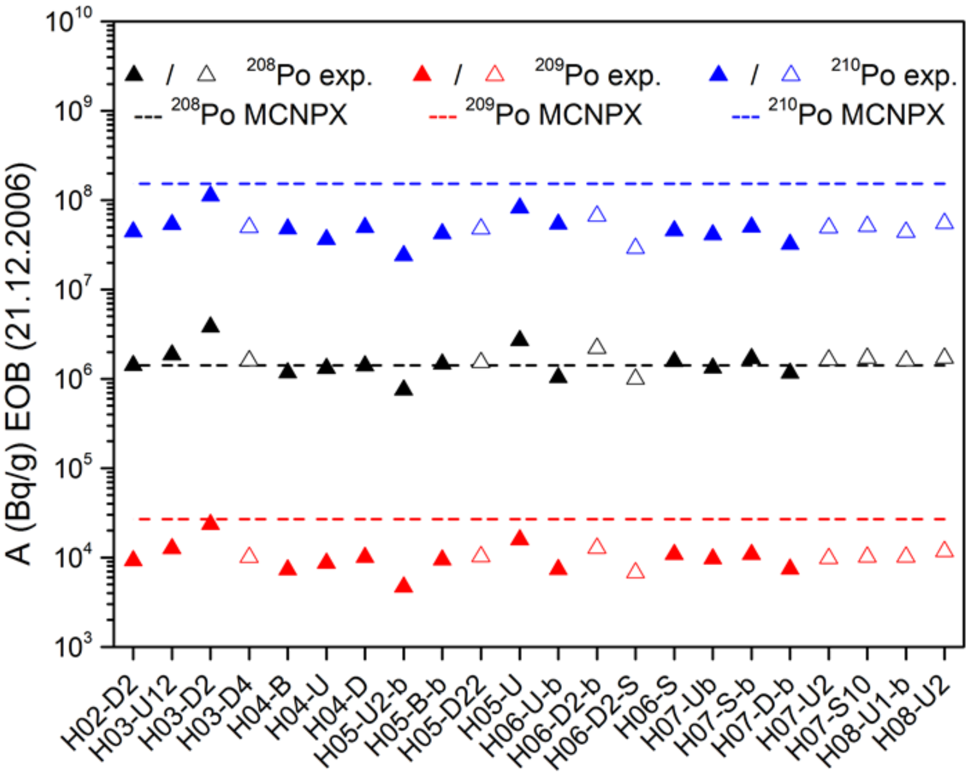
- **radionuclides of elements with low solubility in LBE or sensitive to oxidation:** **only detected in samples taken at the LBE/steel interface and the LBE/cover gas interface,**

**➔ accumulations on the walls increased dose rates and locally increased decay heat,**

- **$^{129}$I is lower than expected,**

- **$^{208-210}$Po homogeneously distributed**

**➔ α activity well predicted with FLUKA and MCNPX codes.**




Example of measured/modeled activity concentration of polonium;

* Filled triangles: bulk LBE samples and

* Non-filled triangle: samples from the LBE/steel and LBE/cover gas interfaces.

# ALKALINE EXTRACTION

**Complementary to these characterizations, studies, dedicated to the separation of spallation products from LBE, were performed at PSI (with University of BERN):**

**➔ Technique based on the alkaline extraction shown to be suitable for the extraction of Po from heavy liquid metals used in nuclear systems.**

**➔ For this purpose, irradiated LBE samples from CERN ISOLDE used for model experiments.**

**➔ Alkaline extraction proved to be a powerful technique for the extraction of numerous radionuclides present in the irradiated liquid metal.**

**➔ For the extraction of Po isotopes, maintaining reducing conditions and avoiding any ingress of moisture would guarantee their fast and reliable separation.**

**➔ Such purification would decrease radiological concerns during maintenance and shutdown of the facility.**

**➔ Nevertheless several crucial problems to be solved before licensing such a separation method on an industrial scale:**

- **high corrosivity of molten hydroxides,**
- **their hygroscopic property.**

# CONCLUSIONS

- Target was designed by Megapie Consortium, manufactured in France, Latvia & Switzerland (and Italy and Switzerland for ancillary systems),
- Target operability was demonstrated during integral tests in PSI,
- Irradiation was carried out end of 2006 (4 monthes) in SinQ PSI and contributed to the validation of the models used during design phase.
- Decommissioning, and waste management phases were defined and executed properly by PSI.
- Post Irradiation Examinations were prepared by Megapie consortium; cutting, sampling, cleaning, samples preparation and shipping were performed by PSI teams.
- The chemical analysis recently performed on spallation and corrosion products in the LBE are very relevant for further applications of LBE as a spallation media and more generally as a coolant.

- The Megapie project feedback provides to:

➔ ADS Community a unique relevant design and operational feedback which will be a decisive contribution to the development of this option for the transmutation of minor actinides (ie MIRRHA)

- ➔ Fast Reactor Community, Lead & LBE, a very large feedback on materials and coolant (also for Sodium Fast Reactors: innovative option for coolant of some ancillary cooling system, material behaviour,…)
- ➔ Neutron scattering Community, a significant feedback for future HLM targets.

# ACKNOWLEDGMENTS

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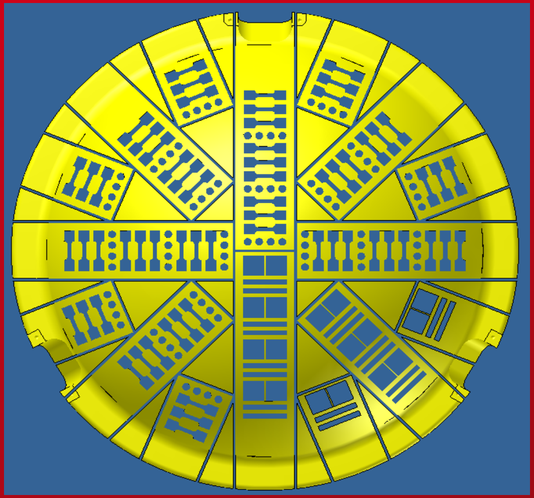
*- the PSI team for its constant dedication,*

*- the European Commission: Program MEGAPIE Test and currently GETMAT ( to support the European partners for the PIE of structural materials).*

*On behalf of MEGAPIE Consortium and Paul Scherrer Institute*

*Thank you for your kind attention !*



Commissariat à l’énergie atomique et aux énergies alternatives
Centre de Cadarache | DEN/CAD/DTN  Bâtiment 710
13108 Saint-Paul-Lez-Durance
T. +33 (0)4 42 25 44 71 | F. +33 (0)4 42 25 78 78

French Atomic & Alternative Energies Commission
Nuclear Energy Directorate
Nuclear Technolohy Department

Etablissement public à caractère industriel et commercial | RCS Paris B 775 685 019