Actinide and Fission Product Partitioning and Transmutation
Tenth Information Exchange Meeting
6-10 October 2008, Mito, Japan

Progress in Structural Materials for Transmutation Devices

Concetta Fazio

Outline

- Transmutation objectives
- Transmutation systems and material needs
- Past programs on materials selection and qualification
- Ongoing programs and selected experimental results
- Next steps on structural materials development and qualification
- Summary and Perspective

Transmutation objectives

- Generic objectives of P/T strategies:
  - reduce the burden on a geological storage in terms of waste mass minimization, reduction of the heat load and of the source of potential radiotoxicity.

- More specific objectives can be defined according to the specific policy adopted towards nuclear energy and according to specific strategies of reactor development.

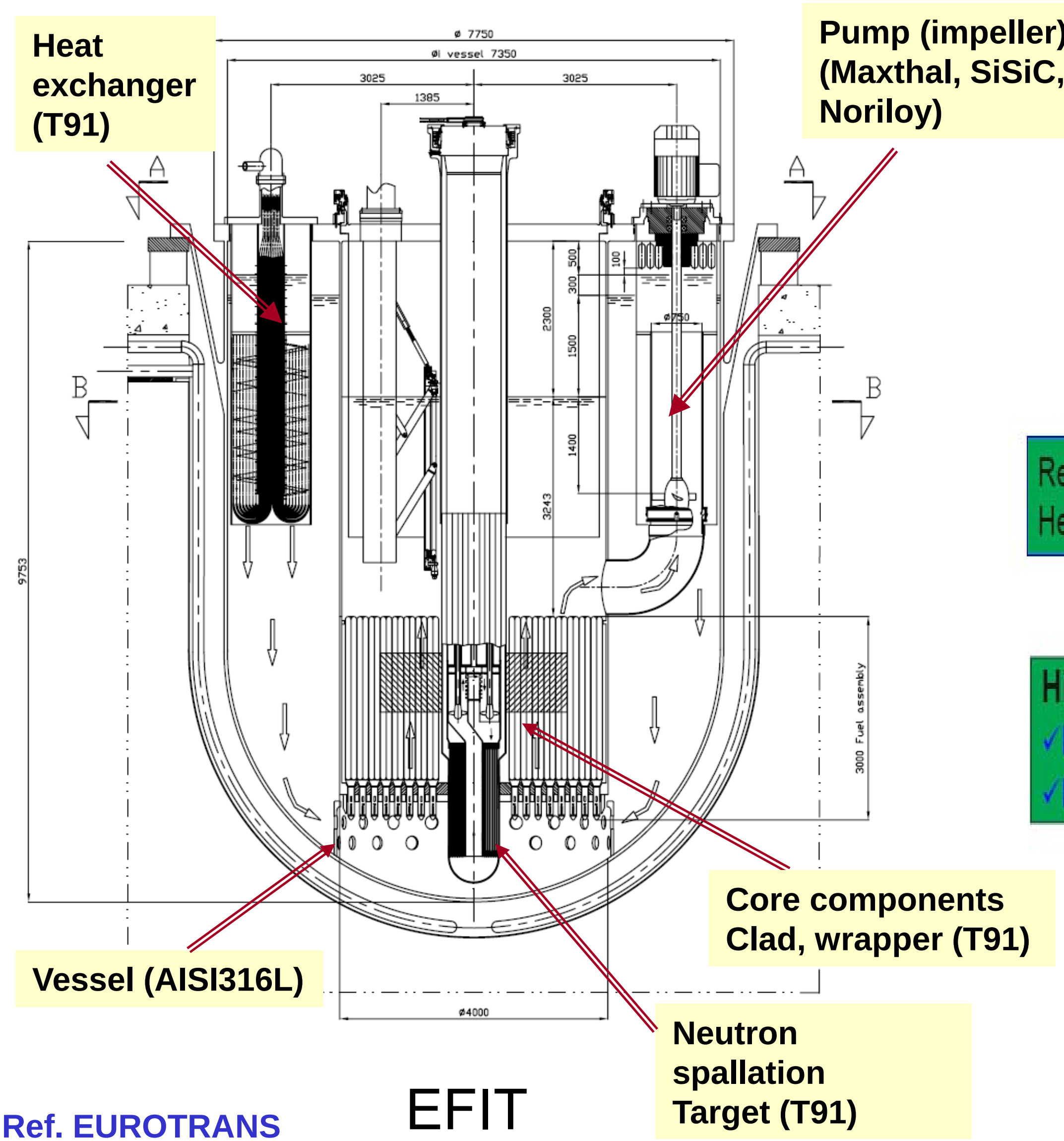
Three categories of specific objectives:

1. **Waste minimization and sustainable development** of nuclear energy and increased proliferation resistance of the fuel cycle. A transition from a LWR fleet to a **FR** fleet is foreseen.
2. **Reduction of MA inventory and use of Pu** as a resource in LWRs, in the hypothesis of a delayed deployment of fast reactors. Use of dedicated burners (**ADS or FR**)
3. **Reduction of TRU inventory** as unloaded from LWRs: Management of spent fuel inventories, as a legacy of previous operation of nuclear power plants in **ADS**.

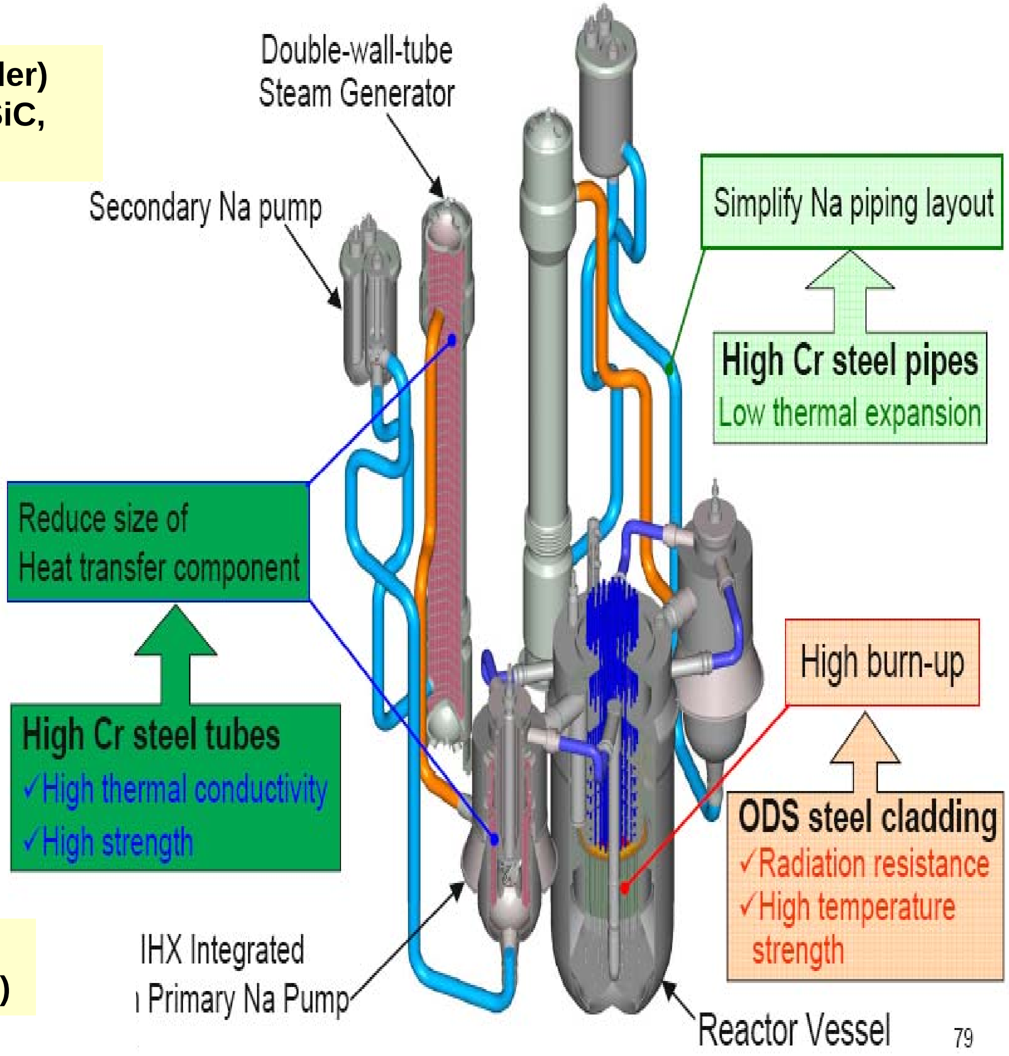
It is a generally agreed conclusion that fast neutron spectrum systems are more appropriate for transmutation of TRU

Ref. PATEROS

Transmutation systems: examples



Ref. EUROTRANS



Ref. SMINS, 2007

Transmutation systems: examples

- Innovative fast neutron reactors imply challenging issues for materials:
  - a range of different coolants (Na, HLM, Gas)
  - a range of different operating temperatures
  - High burn-up (high neutron doses)

- In what follows a summary of EU programs addressing material issues for innovative systems will be made

Past programs on materials (1/4)

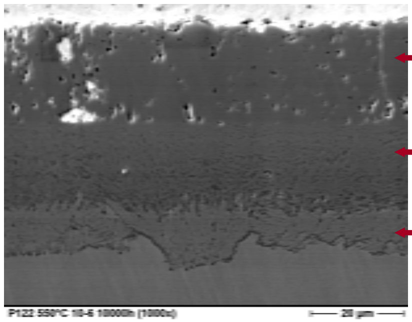
- At European level during the last ten years materials studies have been performed mainly for transmutation systems cooled with HLM
- FP5 projects TECLA, SPIRE, MEGAPIE-TEST:
  - Objectives:
    - screening tests on materials compatibility
    - assessment on materials irradiation behaviour in a spallation environment
    - Application of results on a real component: the MEGAPIE target

Past programs on materials (2/4)

TECLA: F/M and Austenitic Steels

Protection system	Oxide protection	Transition zone	FeAl based coatings protection
Corrosion mechanism	Oxide formation on martensite and austenite	Oxide formation on martensite Mixed corrosion mechanism : oxidation / dissolution on austenite	oxide layers unstable FeAl based coating stable

500 °C 550 °C



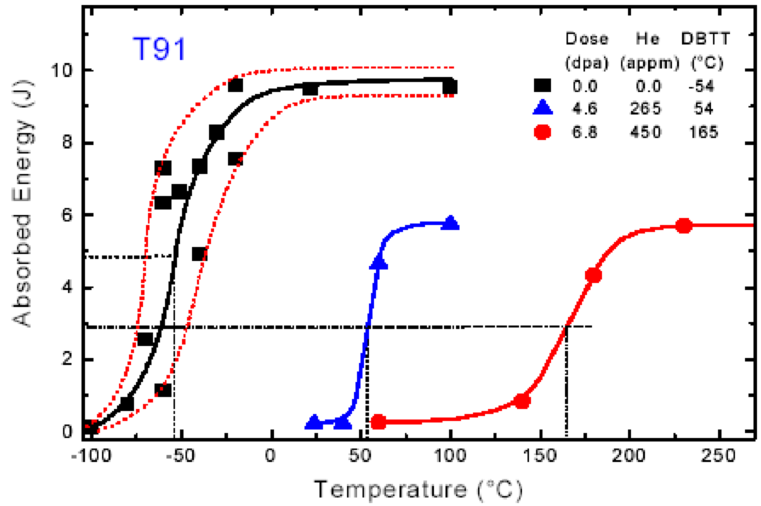
- Threshold limit of 550 to 600°C, above which the oxide layer becomes non-protective
- Aluminized layers protect austenitic and martensitic steels up to 550°C.
- Next step on coating: stability under irradiation

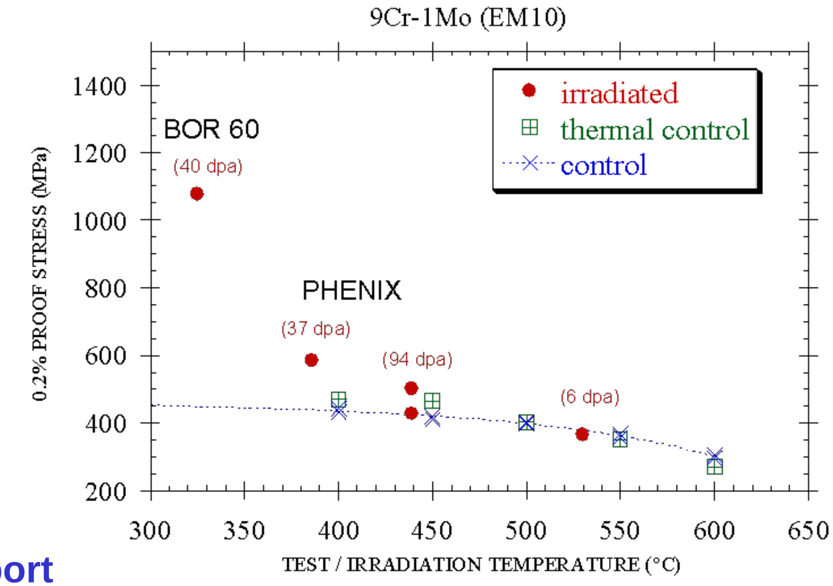
Ref. ADOPT final report

Past programs on materials (3/4)

SPIRE: 9Cr F/M steels

- p/n irradiation: increase of hardening with decreasing the irradiation temperature (T < 300°C),
- n irradiation: at T<350°C, hardening and embrittlement induced by the irradiation.
- At T > 500°C, besides irradiation effects, corrosion, thermal creep and creep-fatigue contribute to define the upper limit of in-service temperatures.
- For spallation target, the in-service temperature range would be: 350°C < T < 500-550°C,
- Conclusion: Combined experiments are needed to assess fuel cladding materials and window materials.





Ref. ADOPT final report

Past programs on materials (4/4)

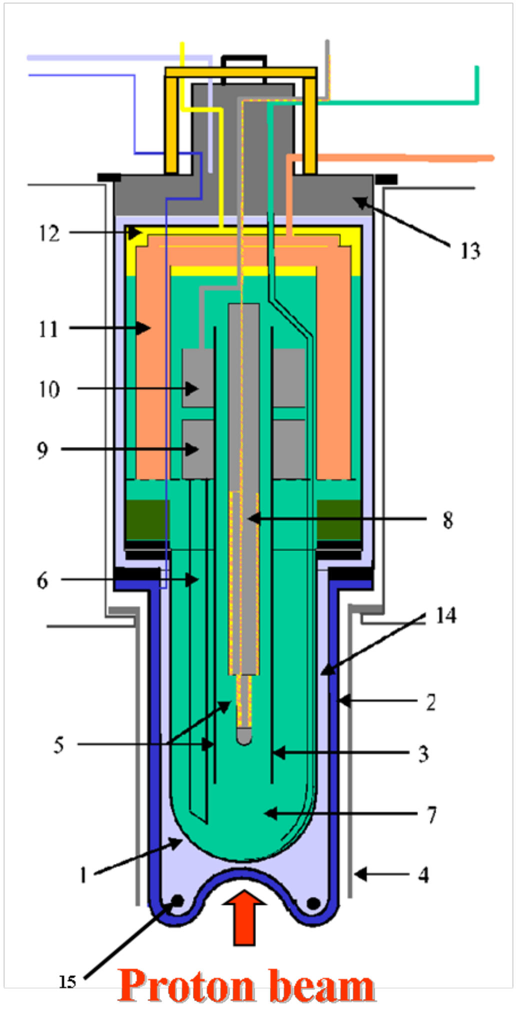
Materials assessment under MEGAPIE conditions

AISI 316L steel (pumps, heat exchanger, main/bypass flow guide tubes, central rod, fill and drain tubes)

- **Corrosion** ➡low impact
- **Irradiation damage** ➡ low impact
- **LME** ➡low impact

T91 steel (beam window, lower liquid metal container)

- **Corrosion** ➡ estimated < 60 μm in 5 months, low impact
- **Irradiation damage** ➡ DBTT shift limiting factor should remain below the minimum temperature, 230°C, i.e. max 3.4 Ah (8-9 dpa)
- **LME** ➡n/p-HLM combined effect assessed through Linear Elastic Fracture Mechanics (LEFM) analysis



Ref. Structural materials for the MEGAPIE target Summary report for MEGAPIE R&D tasks X7 and X10

Current Programs in the area of HLM (FP6)

- EUROTRANS
  - DM4: Development and assessment of structural materials and heavy liquid metal technologies for transmutation systems (DEMETRA)
- VELLA
  - JRA1 – Lead technology (tests in Pb)
  - JRA4 – Irradiation in the presence of LBE
- ELSY
  - WP6 – Lead technology

DEMETRA

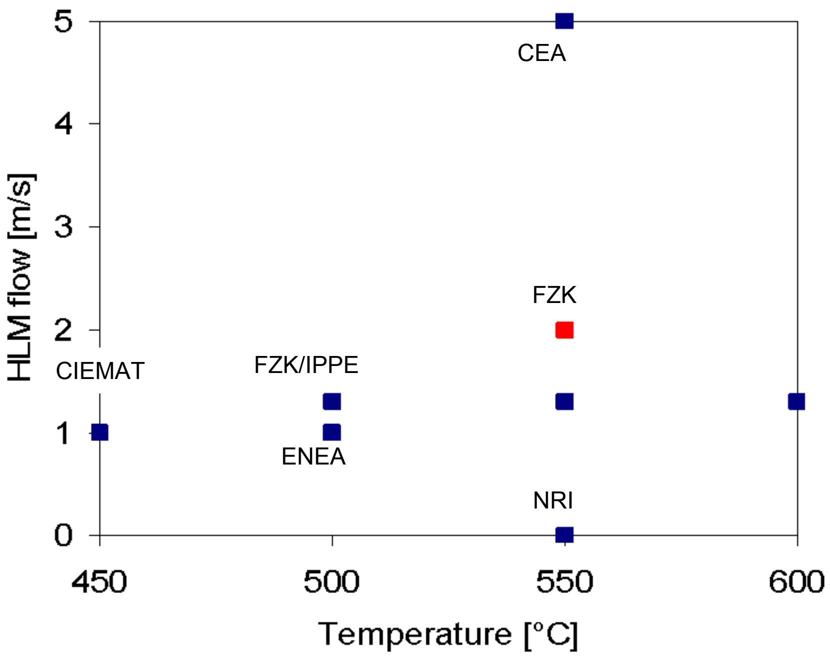
Needs for materials assessment

- HLM quality control and related corrosion phenomena
- Corrosion and corrosion prevention: metal loss and effect on mechanical performance
- Erosion phenomena
- Irradiation resistance
- Irradiation / corrosion combined effect
- Corrosion product effect on heat transfer capability (e.g. cladding and heat exchanger)

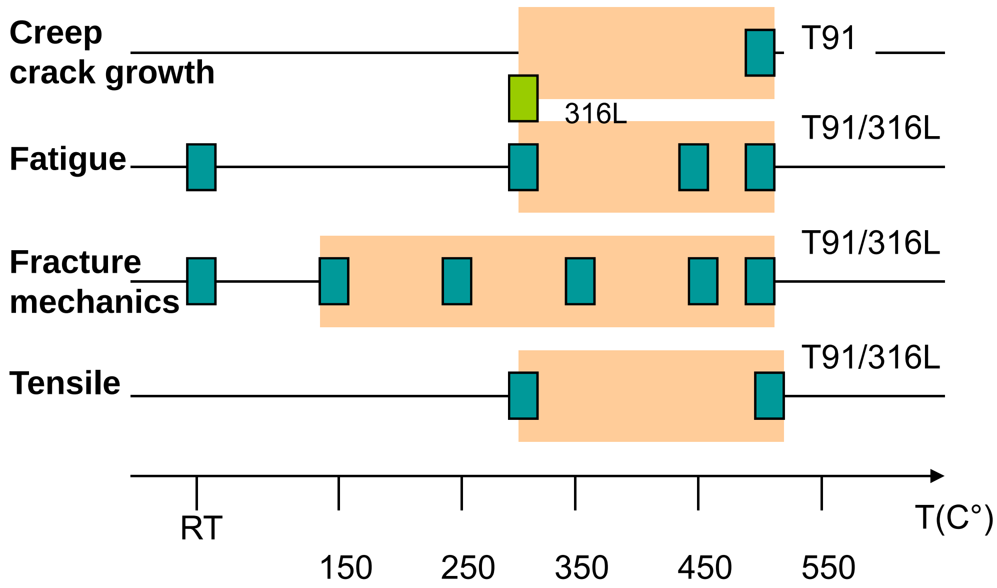
DEMETRA Experimental program (1/2)

Reference Structural Materials: T91, AISI316L
and GESA surface alluminisation

Corrosion

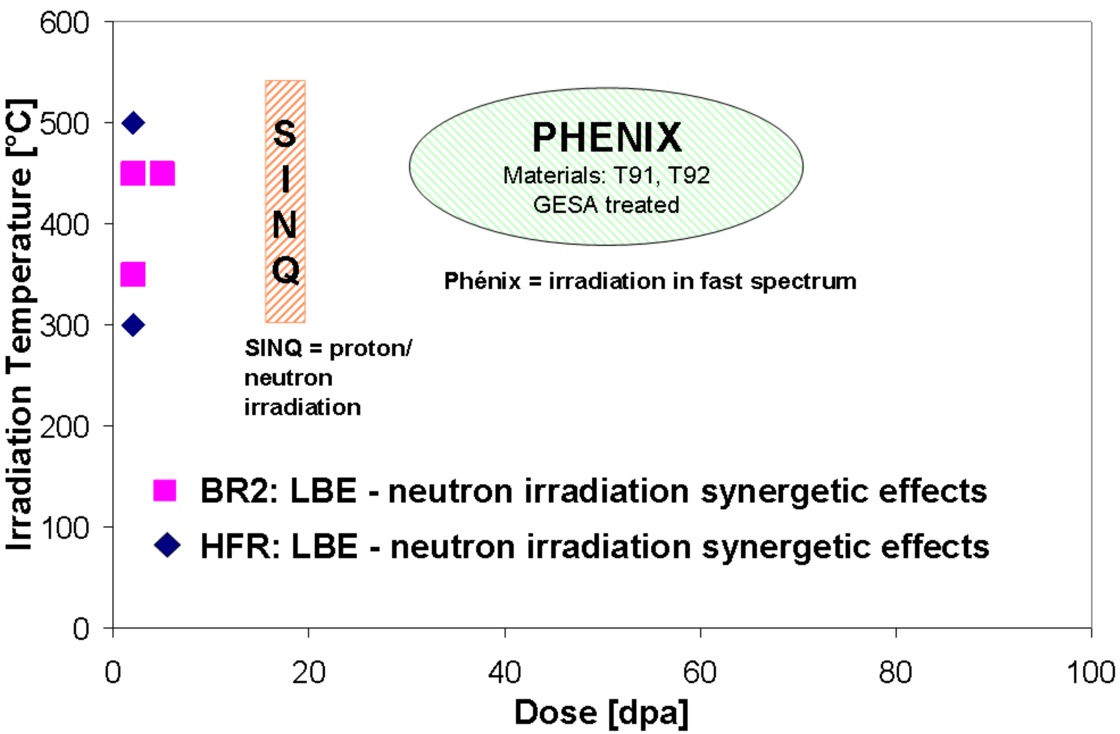


Mechanical properties in HLM



DEMETRA Experimental program (2/2)

Irradiation



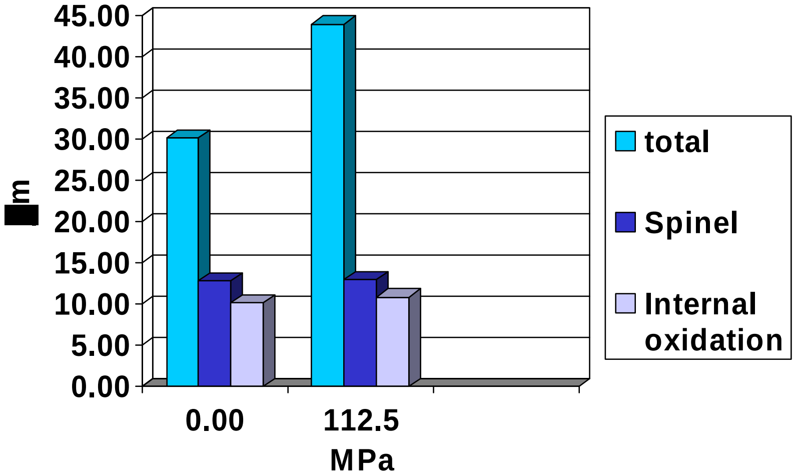
PIE Program

- Tensile properties
- Fracture toughness
- Creep properties (pressurised tubes)
- Compatibility and microstructure investigations
- Assessment of GESA alloyed samples

Results: Corrosion

Loop	Experimental conditions	2.000 h Oxide scale/LM attack		Up to 5.000 h Oxide scale/LM attack		Up to 10.000 h Oxide scale/LM attack	
		T91	316	T91	316	T91	316
CORRIDA (FZK) **LBE**	**550°C; 10^{-6} wt.-%O flow= 2 m/s**	**Oxide: 20-25 µm**	**Oxide : few**	**Oxide: 36 µm**		**Oxide: 55-85 µm**	**LM attack up to 350 µm**
CU2 (IPPE/FZK) **LBE**	**550°C; 10^{-6} wt.-%O flow= 1.3 m/s**	**Oxide: 39 µm**	—			**6600 h Oxide: 36-45 µm**	—
CHEOPE III (ENEA) **Pb**	**500°C; 10^{-6}wt%; flow = 1 m/s**	**20 µm (non homog.)**	**Oxide: few**	**Oxide: 25 µm**	**Oxide: thin**	**Oxide: 35 µm (dispersion)**	**Oxide: 4 µm**
LINCE (CIEMAT) LBE	**450 °C; 10^{-6}-10^{-8} wt.-%O (probl. on sensor)**	**T91: 4 µm (non homog. oxide)**	**Unaffected**	**LM attack up to 300 µm**	**LM attack up to 300 µm**		
LECOR (ENEA) LBE	**450°C; 10^{-10}-10^{-8} wt%**	**LM attack**	**LM attack**			—	—

Pressurised tube in HLM



A. Weisenburger, G. Müller, FZK

Parameters affecting corrosion of steels

- Oxygen activity: oxidation/dissolution
- Time, Temperature
- Flow rate: high flow rate ➡ erosion of Fe_3O_4
- Steel composition: high Cr content ➡ increased oxidation resistance.
- Stresses: hoop stress enhances Fe diffusion

Models to predict corrosion behaviour are under development

Results: Mechanical properties in HLM

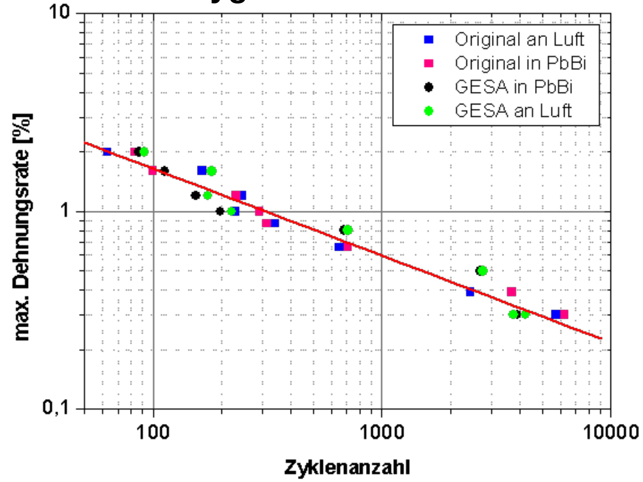
LCF Tests:
- *T91* (FZK)
  550°C no LCF reduction
- *AISI316L* (CNRS)
  450 °C no LCF reduction

Creep to rupture tests:
- *T91*(FZK)
  550°C significant reduction of creep resistance (140-220 MPa) with respect to air

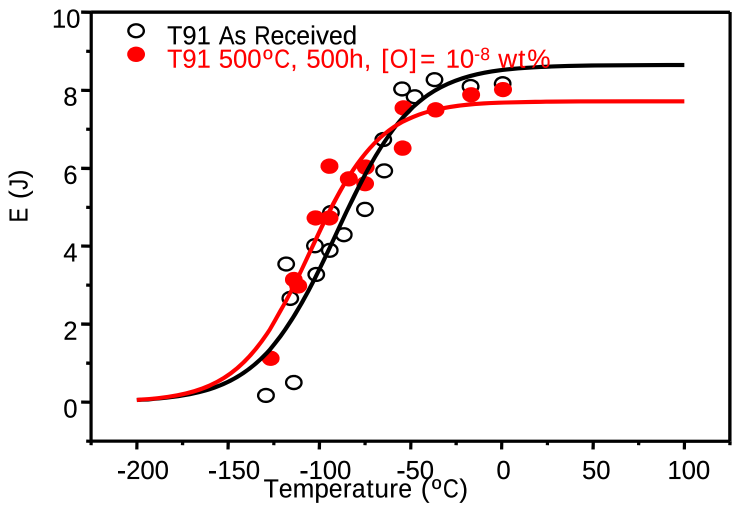
Impact tests:
- *Pre-wetted T91* (CIEMAT, NRI)
  No effect

Work to be completed. Preliminary results show
- *LME evidence not completely clarified.*
- *HLM effect on mechanical properties at e.g. high stress level and particular surface conditions.*

LCF on T91 (FZK): Samples where pre-oxidised in LBE at 550°C for 100h with oxygen content of 10-6wt. %

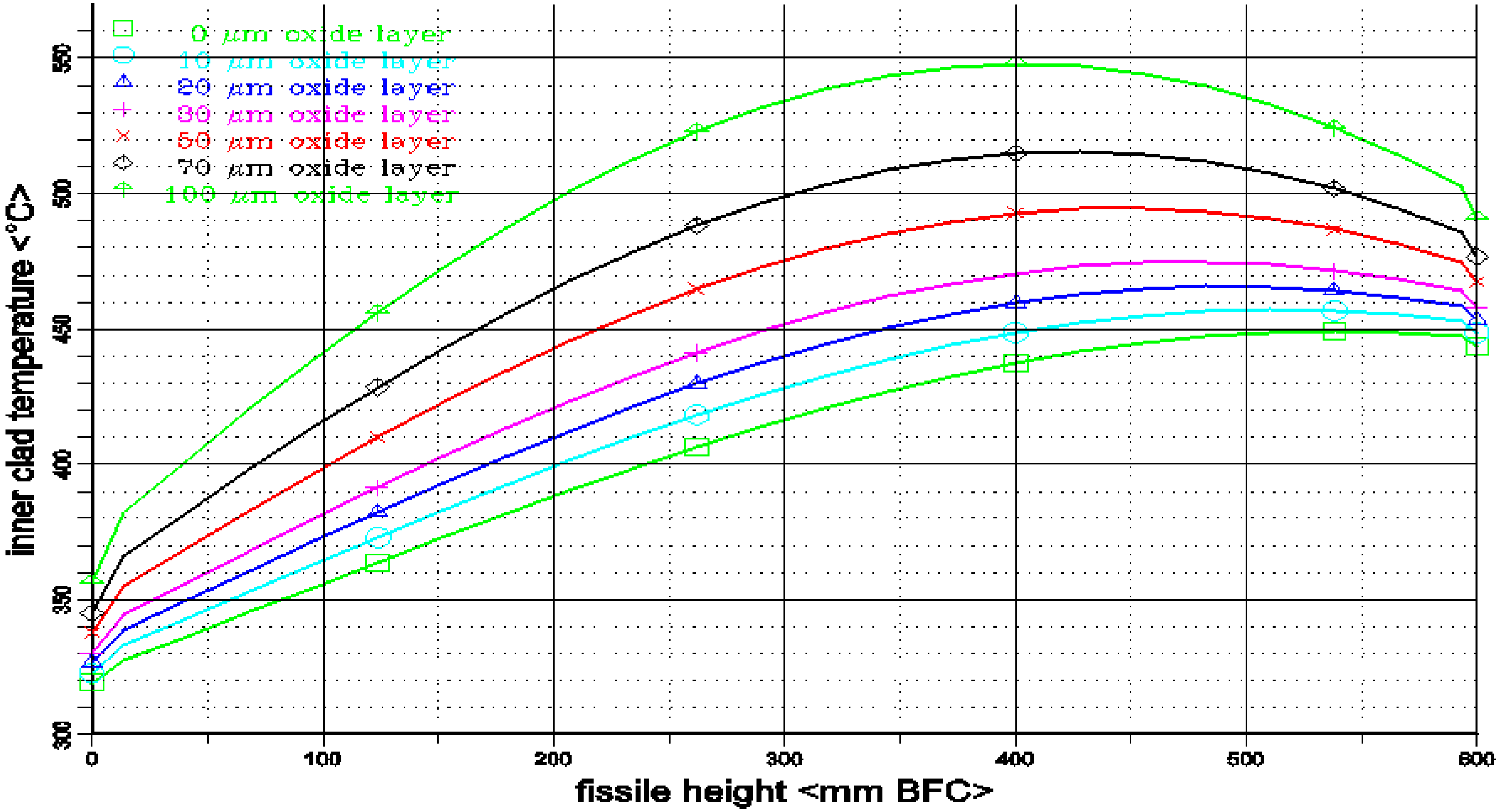


Impact tests (CIEMAT)



Example of application of DM4 results to Design

- Axial profiles of clad inner temperature modified calculation with different additional oxide layers



Oxide layer thickness should be limited to less than 20-30 µm in order to keep margin on the maximum allowable temperature for the T91 steel.

Control of oxidation process in a reactor system might not be applicable

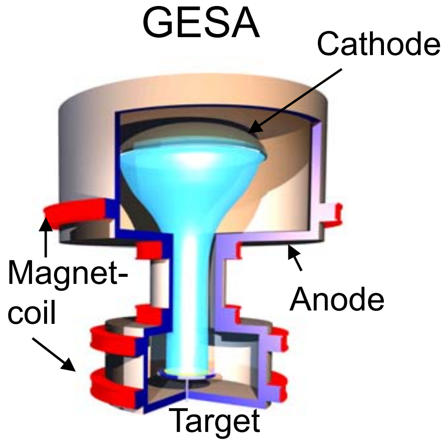
GESA surface alloyed steel can be seen as a solution

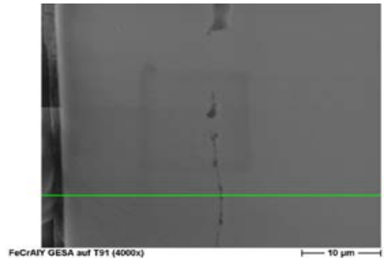
(D. Struwe, W. Pfrang, IRS/FZK)

Results: Surface alloying with GESA

Procedure:

1. LPPS of Fe, Al powders
2. Alloying of the LPPS coating with GESA



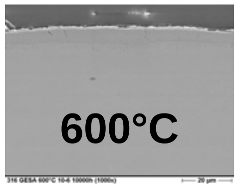


Cross section of GESA treated clad

Advantages:

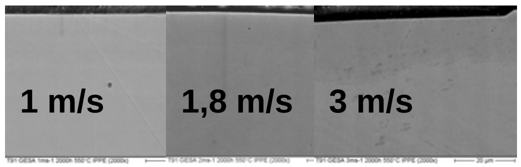
Enhance metallic bonding with substrate
Smoother surface
Controlled Al content

Corrosion resistance:
GESA samples tested for 10000 h in flowing LBE up to 600°C, flow rate 1 m/s and oxygen ptential equivalent to 10^{-6} wt%



No corrosion attack observed,
However control of Al content is relevant

High Flow rate resistance:
GESA samples tested for 2000 h in flowing LBE at 550°C, up to 3 m/s and oxygen ptential equivalent to 10^{-6} wt%



No flow velocity effect: no dissolution attack, no severe oxidation, no erosion.

GESA treated Pressurised tube test:
At 550°C with an internal pressure corresponding to a hoop stress up to 200 MPa

➔ 0,7 % Strain
➔ No change in the oxidation behaviour

GESA treated LCF Test in LBE and air
At 550 °C

No reduction of LCF

G. Müller, A. Weisenburger, FZK

Next steps on structural materials development and qualification

- FP7 Project GETMAT: Generation IV and Transmutation Materials

Kick-off meeting: February 11-13, 2008

GenIV and Transmutation Framework for Materials Cross-Cutting Activities

- As recalled previously, with respect to the current nuclear industry experience, demanding material-related operational conditions can be envisaged e.g.:
  - High in-service and off-normal temperatures
  - High burn-ups
  - Long service life-time (~ 60 years)
  - Compatibility with new coolants

Gen IV and transmutation systems

System	GFR	SFR –	LFR & ADS	VHTR Thermal neutrons	SCWR Thermal neutrons
Coolant	He, 70 bars 480-850°C	Na, few bars 390-600°C	Lead alloys (Pb, LBE)	He, 70 bars 600-1000°C	SC H_2O, 250bars, 280-500°C
Fuel	(UPu)C / O2 in plates of pins in hexagonal subassemblies	(UPu)O2 in pins in hexagonal subassemblies	various concepts	Coated particles (SiC or ZrC) in a graphite matrix	UO_2 enrich
Core structure	SiC-SiCf composite or (backup) **ODS**	Cladding: **ODS** Wrapper: **9Cr MS**	Cladding: 9Cr MS, **ODS** Wrapper: **9Cr MS**	Graphite Composites C/C, SiC/SiC for control rods	Cladd. Aust S (Ni alloys?)
Temp.	500-1200°C	390-750°C	350-480°C	600-1600°C	280-750°C
Dose	60-90dpa	up to 200dpa	100dpa	7-25dpa	
Out of core struct. and others	vessel & core struct: **9-12Cr MS** 350-500°C <<1dpa	prim/sec/steam circ.: **9-12Cr MS** 390-600°C	ADS target: **9Cr MS** 350-550°C 100dpa+He+H	-----	------

Ref. SMINS 2007

Overall Objectives of GETMAT

- Based on the analysis of the experimental programs in progress and of results already obtained the priorities and objectives for the GETMAT project can be defined as follows:
  - Improvement and extension of 9-12 Cr F/M steels qualification (PIE program of relevant ongoing irradiation experiments)
  - ODS alloys development and characterisation
  - Joining and welding procedures qualification (relevant for both ODS and F/M steels)
  - Development and definition of corrosion protection barriers
  - Improved modelling and experimental validation

Expected Results

- Manufacturing and assessment of 9Cr and 14Cr ODS alloys produced with the powder metallurgy technique and with innovative casting method.
- Demonstration of high T performance and compatibility with He, Pb, SCW of the three ODS alloys
- Development and qualification of weld processes of ODS and 9Cr steels
- Full manufacturing process of "smart coatings" with optimised chemical composition and standardised qualification process
- Critical compilation of the PIE results of MATRIX (Phénix), MEGAPIE/SINQ, ASTIR (BR2), IBIS/SUMO (HFR) and BOR60
- Completion of the model parameterisation and model development based on available experimental results

Summary and perspectives

Karlsruhe Institute of Technology

EU programs in FP5, FP6 and FP7 have allowed:

- **9Cr F/M steel assessment for HLM systems:**
  - an important database will be available by 2012.
  - a critical summary of these data will become mandatory for design purposes.

- **ODS steels:**
  - For high Temperature (> 500°C) and high burn-up (≥ 20%) ODS steels have been indicated as most promising materials.
  - Experimental program on ODS steels has been initiated, and a preliminary assessment can be done. However a more relevant program is needed to allow their nuclear application.

- **Corrosion protection barriers (e.g. GESA alloying):**
  - The experimental program on surface alloyed steels with the GESA technology will allow an in-depth assessment.
  - In parallel, studies to extend the GESA technology from laboratory scale to pre-industrial scale.

In summary the results of GETMAT, related international projects and, possibly, industry input will help to define research priorities in the field of materials beyond 2012

The MEGAPIE Experiment

- **A view on the MEGAPIE experiment beyond the materials assessment**

- **Co-authors**: C. Fazio from FZK, W. Wagner, L. Zanini, Y. Dai from PSI; M. Dierckx from SCK-CEN and C. Latgé from CEA and all team members involved in the MEGAPIE project from PSI, CEA, FZK, SCK-CEN, ENEA, CNRS, JAEA, KAERI, US-DOE

- **Outline**
  - Introduction
  - Summary of MEGAPIE operation
  - Summary of the Post Test Analysis
  - Lessons learned for ADS systems
  - Outlook: the PIE of MEGAPIE

Introduction

MEGAwatt Pilot Experiment:

- **Joint international initiative to design, build, licence, operate and explore a liquid metal*) spallation target for 1 MW beam power**

***) Lead-Bismuth-Eutectic (LBE)**

T_m=125°C

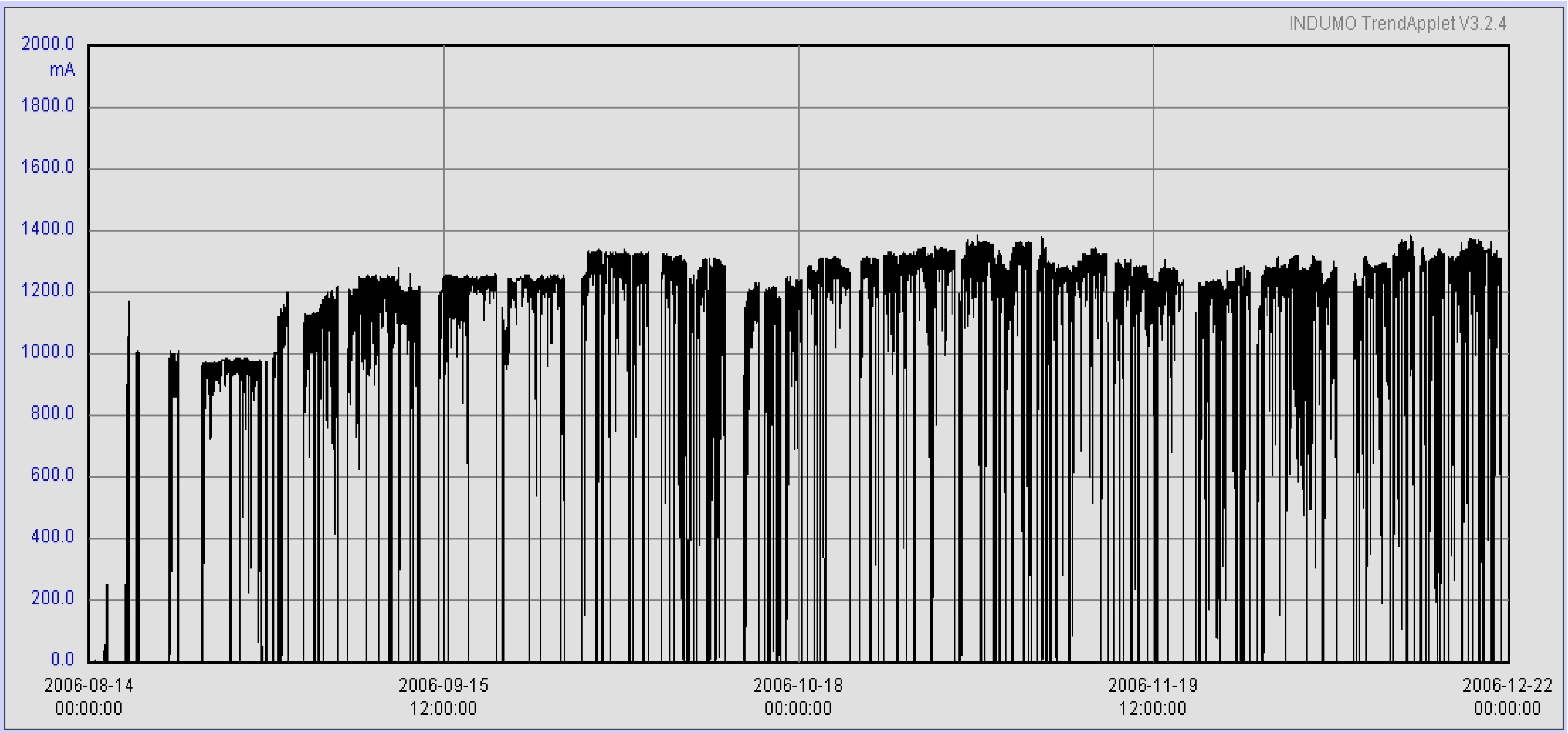
Goals of MEGAPIE:

- **Increase the neutron flux at SINQ**
- **Demonstrate the feasibility of a liquid metal target for high-power spallation and ADS applications**

MEGAPIE Operation: full history

On beam: August 14 – December 21, 2006

- Accumulated charge: 2.8 Ah
- Peak Current: 1400 μA
- Beam trips (< 1 min): 5500
- Interrupts (< 8 h): 570

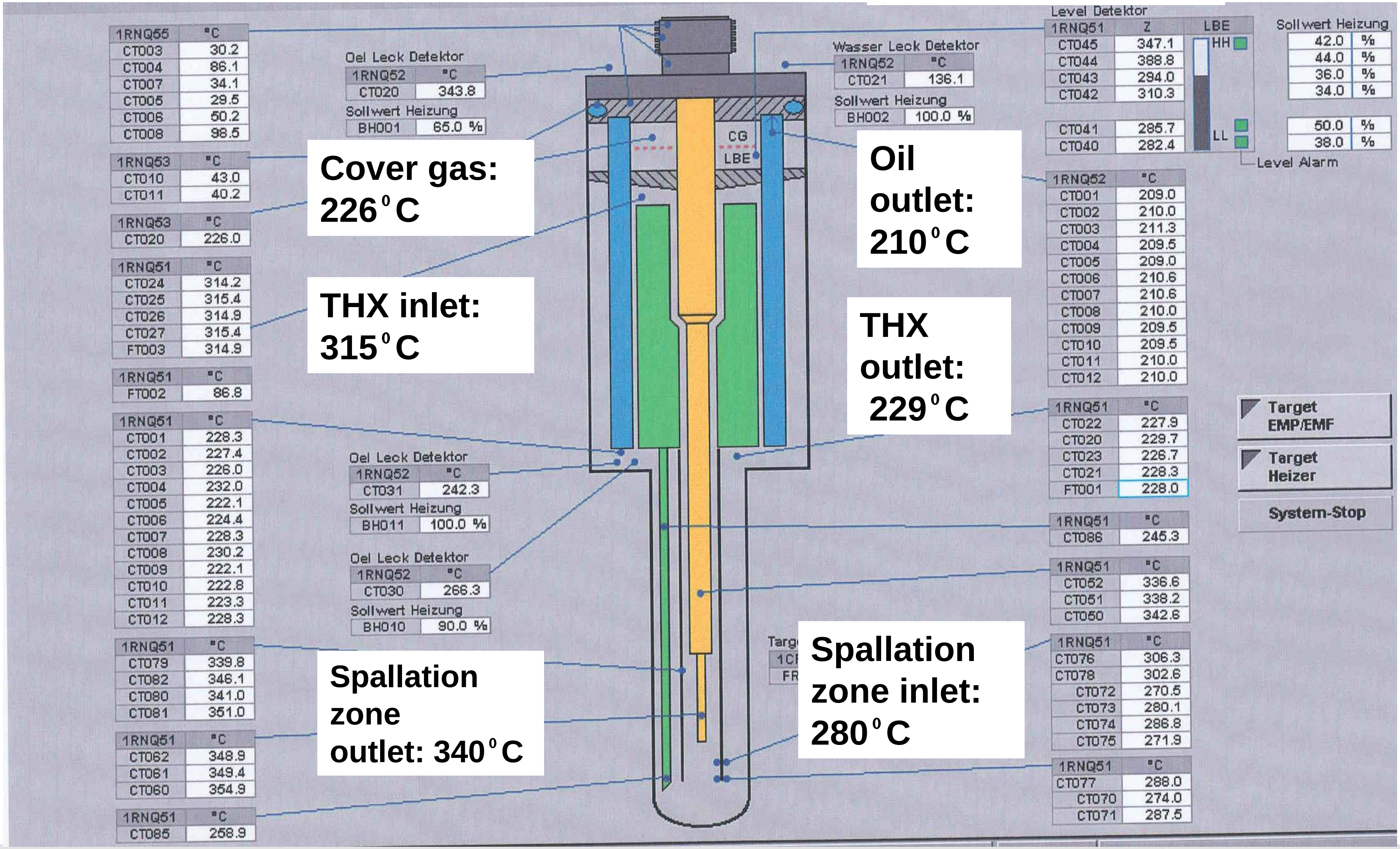


KIT – die Kooperation von Forschungszentrum Karlsruhe GmbH und Universität Karlsruhe (TH)

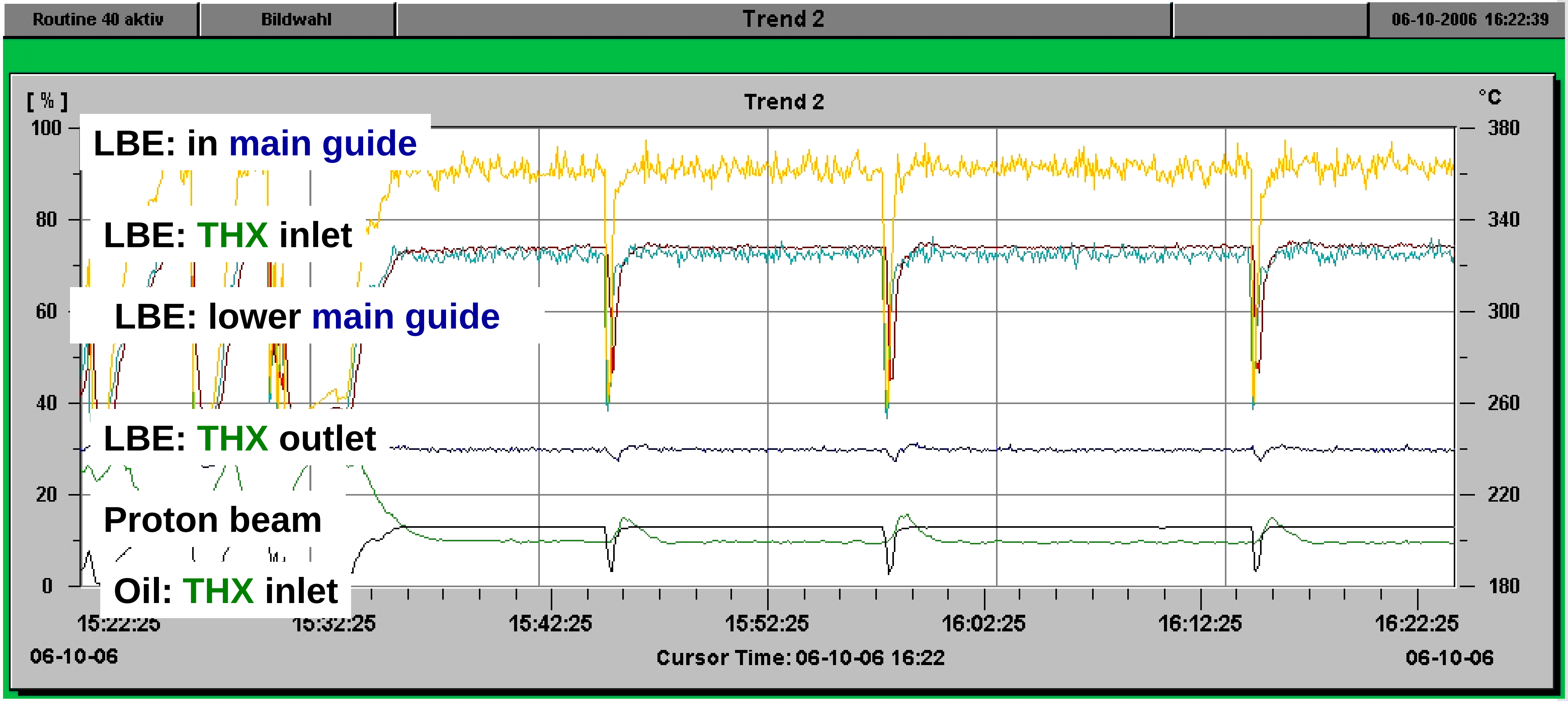
Forschungszentrum Karlsruhe in der Helmholtz-Gemeinschaft

MEGAPIE Operation: temperature at full beam

Beam current: 1274 mA



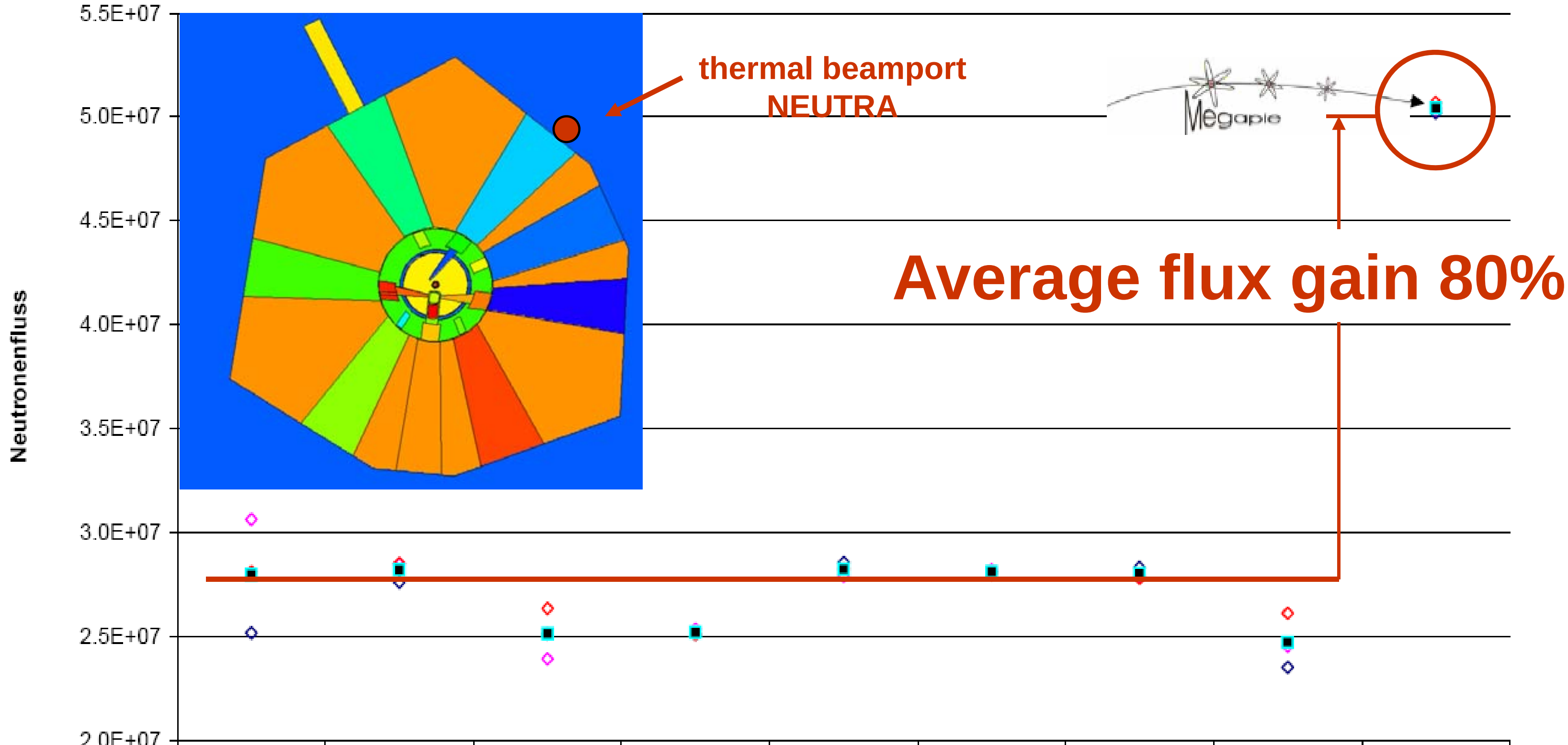
MEGAPIE Operation: temperature during beam trips



⇨ scurtinize / validate the thermo-hydraulic simulations

MEGAPIE Operation: Neutronic Performance

Neutronenfluss SINQ Strahlkanal 32 NEUTRA



	13.04.2000	18.05.2000	14.05.2001	29.04.2002	19.12.2003	15.06.2004	15.06.2005	05.07.2005	31.08.2006
◇ Pos. 1.1	2.51E+07	2.76E+07	2.51E+07	2.51E+07	2.85E+07	2.81E+07	2.83E+07	2.35E+07	5.02E+07
◇ Pos. Z	3.06E+07	2.85E+07	2.39E+07	2.53E+07	2.79E+07	2.82E+07	2.79E+07	2.45E+07	5.04E+07
◇ Pos. 5.1	2.81E+07	2.85E+07	2.63E+07	2.51E+07	2.82E+07	2.80E+07	2.78E+07	2.61E+07	5.07E+07
■ Mittelwert	2.80E+07	2.82E+07	2.51E+07	2.52E+07	2.82E+07	2.81E+07	2.80E+07	2.47E+07	5.04E+07

05.09.2006

Markus Lüthy
Koordination Betrieb Anlagen West 8830

Summary of MEGAPIE Post Test Analysis

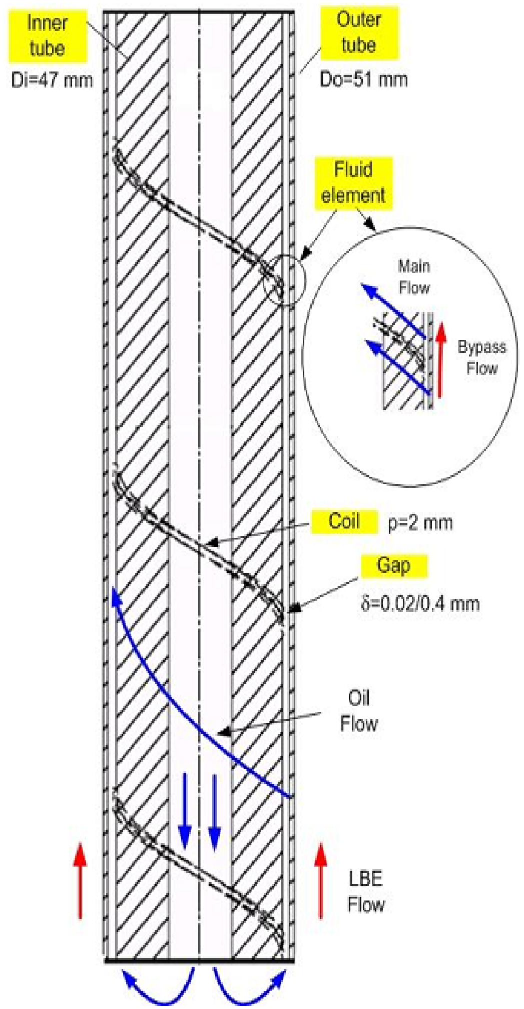
Post test Analysis: Thermal-hydraulics

1. Thermo hydraulic data collection (temperatures and flow rates) and pre-processing for further analysis.
2. Comparison temperatures calculated during design phase and measurements during experiments.
   - improvement of heat transfer correlations for better agreement between 1D system codes and measurements
   - detailed thermo hydraulic modelling via CFD of some components to explain the large discrepancy between measurements and calculations
3. Evaluation of target temperature control performance

Post test Analysis: Thermal-hydraulics

1. Thermo hydraulic data collection (temperatures and flow rates) and pre-processing for further analysis:
   - 1407 steady state regions out of which 113 regions of zero current
   - In total the target suffered 4500 beam trips → approximately 2 trips per hour
   - The accumulated charge on the target reached ~2800 mAh

Post test Analysis: Thermal-hydraulics



2. Comparison temperatures calculated during design phase and measurements during experiments:
   - An example: The measured THX heat transfer coefficient at the oil side is larger than design value
     - Initial calculations assumed a completely spiraling flow path for the oil
     - In reality the part of the oil flow is bypassing the spiral: CFD is used to confirm the effect of this parasitic bypass flow

Post test Analysis: Summary on Thermal-hydraulics

1. A huge amount of thermo hydraulic data has been statistically processed to render a manageable amount of data for further analysis.

2. Overall thermo hydraulic behaviour of the target and heat removal system was better than initially expected due to enhanced heat transfer at the target heat exchanger.

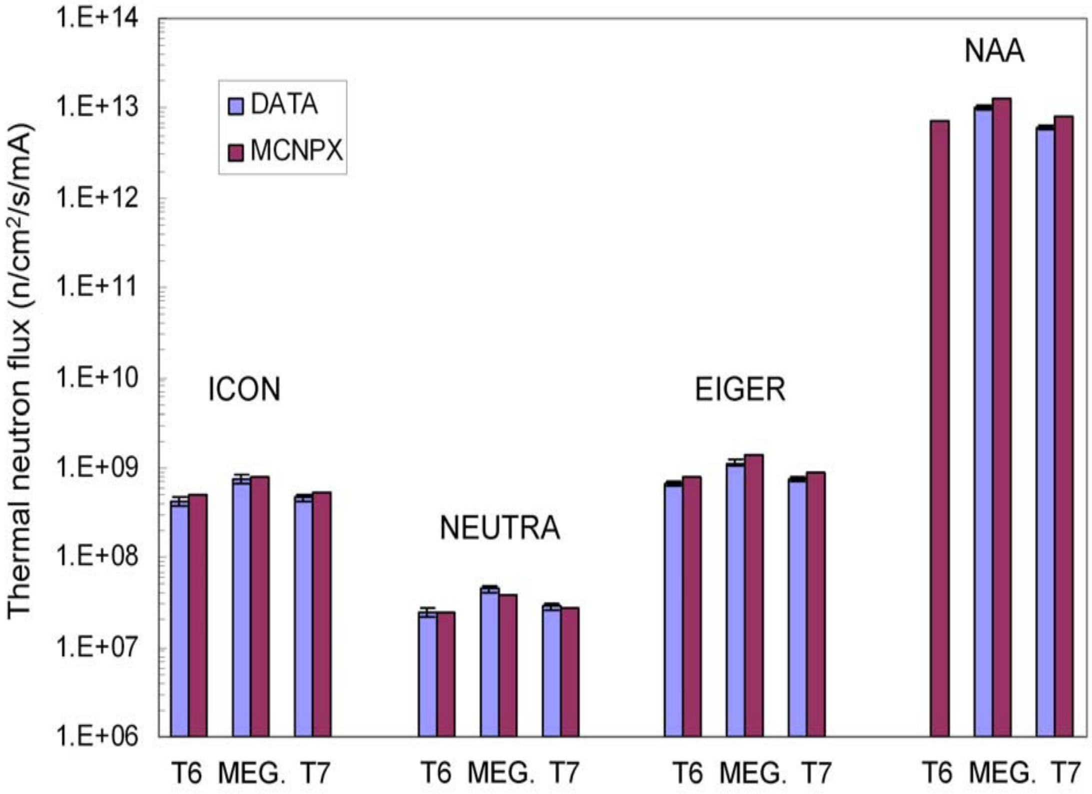
3. Most of the discrepancies between temperature measurements inside the target and predictions with CFD and 1D system code (HETRAF and RELAP5) have been explained (e.g. 5x more heat is transferred through the flow guide tube).

4. Target temperature control performed as adequately and valve fluctuation in low power regimes has been explained

Post test Analysis: Neutronics and nuclear data

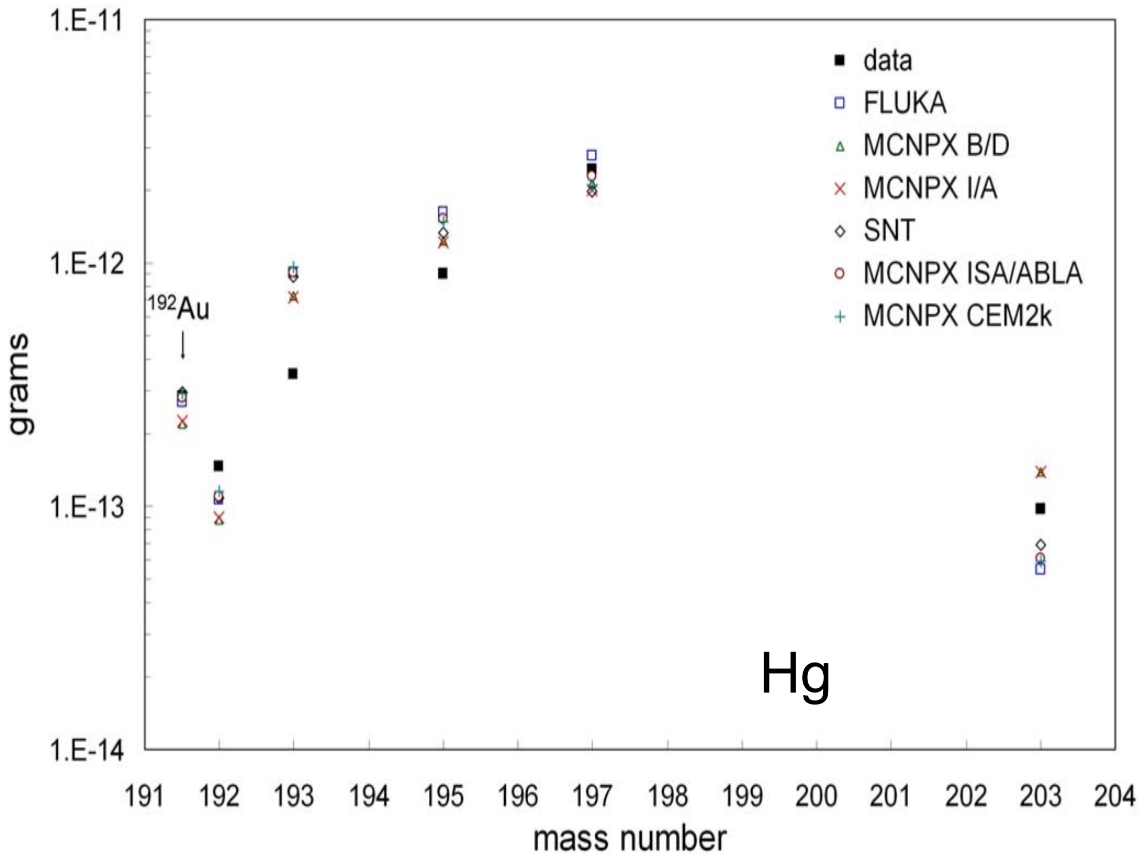
- **Main Tasks**
  - Neutronic performance
  - Delayed neutrons
  - Gas production
  - Activation

Post test analysis: Neutronic performance



- Mapping of the neutron flux in its different components (thermal, epithermal, fast) in the SINQ facility around the target. Most of the results within two standard deviations.
- Innovative measurements inside the MEGAPIE target
- Measurements complemented by corresponding calculations: code validation
- Comparison with solid targets

Post test analysis: Gas production



Recommendations:

- Feedback to operation issues:
  - Leak tightness
  - Pressure sensors (radiation damage?) reliability
  - Gas sampling operation to be improved
- Feedback to scientific/safety issues:
  - Need for reliable mass spectroscopy measurements, especially for H an He isotopes
  - Understanding the behaviour of Hg in the CGS

Lessons Learned for ADS systems

Feedback to ADS as studied in the EUROTRANS project

Importance of MEGAPIE for the roadmap towards XT-ADS

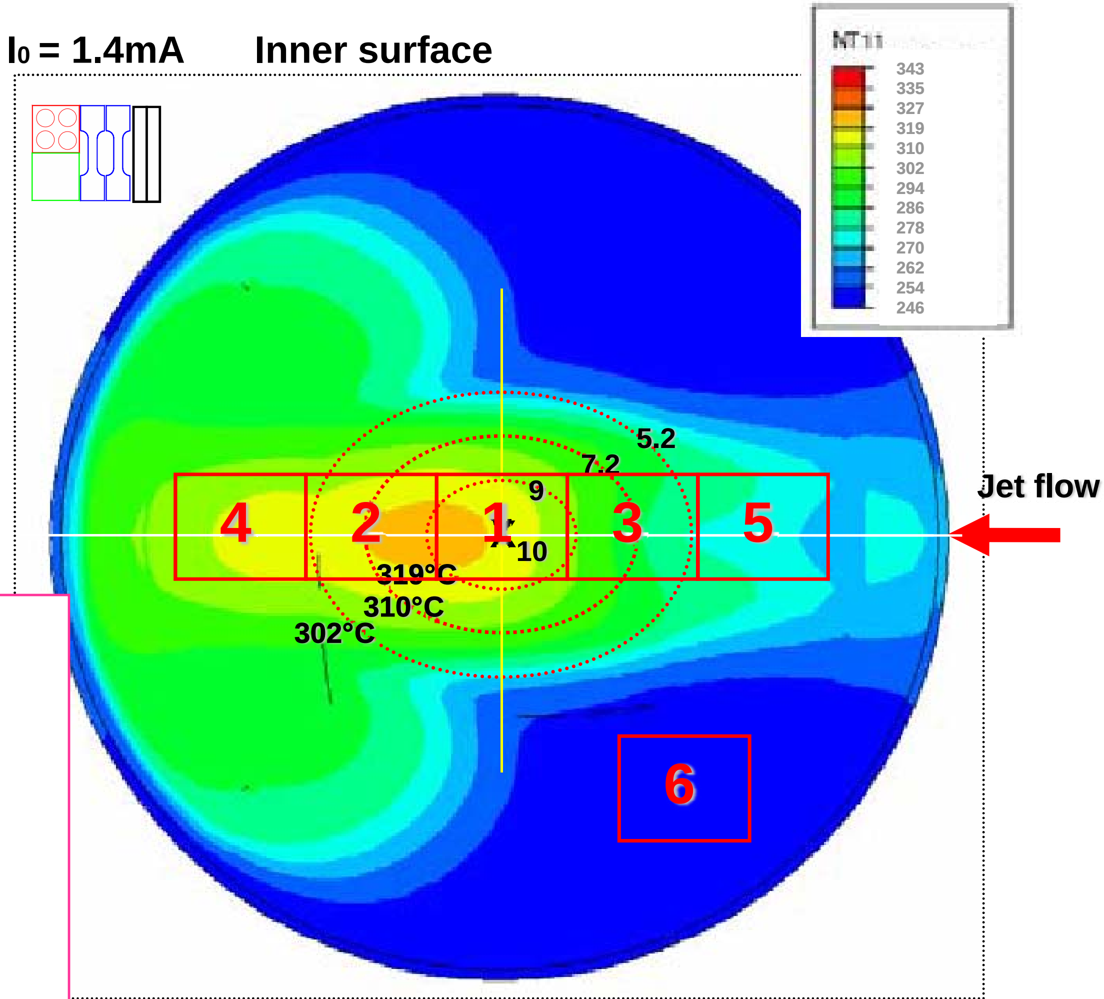
First LBE spallation target that

- has been experimentally demonstrated
- to be operated safely
- during an extended time period
- Some MEGAPIE results are very relevant for XT-ADS characteristics

Feedback to ADS

	MEGAPIE	XT-ADS target
Coolant / target	LBE	LBE
Beam energy	595MeV	600MeV
Beam current	1.4mA	2.5-3mA
Target diameter	Ø20cm	<Ø10cm
Lifetime	4 months	1 year
Accumulated charge on target	90 Ah/m²	2500 Ah/m²
Beam interface	window	windowless
Damage	7 dpa (window: T91)	30-40 dpa (target walls: T91)

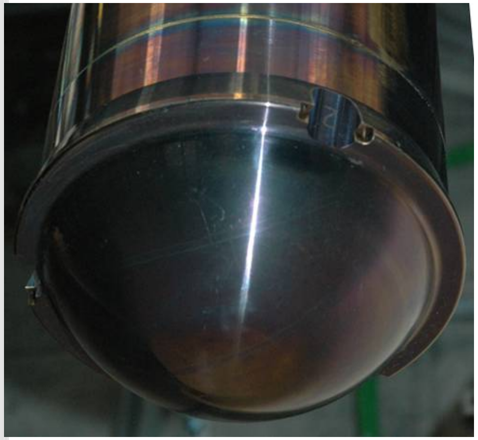
Feedback to ADS: The PIE of MEGAPIE



Conditions:

1: highest dpa & T
2: high T, medium dpa
3: medium dpa & T
4: low dpa, medium T
5: low dpa & T, high flow
6: low dpa, T & flow

Feedback to ADS: The PIE of MEGAPIE



	Corro-sion + erosion	Embrittle-ment (irr., He, LBE)	Mechanical change	Microstructural change	Chemical change	Association (number of conditions)
OM + μ-hardness + SEM/EPMA	X	X (μ-hard-ness)				CEA (2 cond) LANL (2 cond) PSI (2 cond) JAEA (2 cond), KAERI
TEM / FEGSTEM		X	X	X	X	CEA (2 cond) FZK (3 cond) PSI (2 cond) JAEA (2 cond), KAERI
H and He analyses		X				PSI (3 cond)
SIMS / XPS	X				X	PSI (SIMS:2cond) SCK (XPS:1cond)
XRD	X				X	CEA (2 cond)
Tensile test		X	X			CEA (2 cond) LANL (2 cond) PSI/ENEA(3cond)* KAERI
Bending test		X	X			LANL (3cond) PSI (3 cond)*
Small punch test **		X	X			CEA (2 cond) PSI (2 cond) CNRS (3 cond)*

* Tests to be done in both Ar (or Air) and LBE environments.
** CNRS will use 8.9 mm diameter discs and the others will use 3 mm diameter discs.

Conclusion: MEGAPIE feedback to ADS

- demonstration of the feasibility of a megawatt liquid Pb-Bi spallation target
- demonstration of safe operation for licensing
- development of design tools (thermalhydraulics and neutronics) and experience
- tests of components (electromagnetic pump, heat exchanger, measurement techniques) and procedures (start-up, shut-down, beam trips, etc.)
- Spallation Gas production and qualification
- structural materials test and qualification which will be completed with the PIE

Thank you for your attention