

# (Subcritical) system experiments

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*International Training Course (ITC-8)  
3-6 February 2009  
Politecnico di Torino*

# Spallation target experiments

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# Introduction

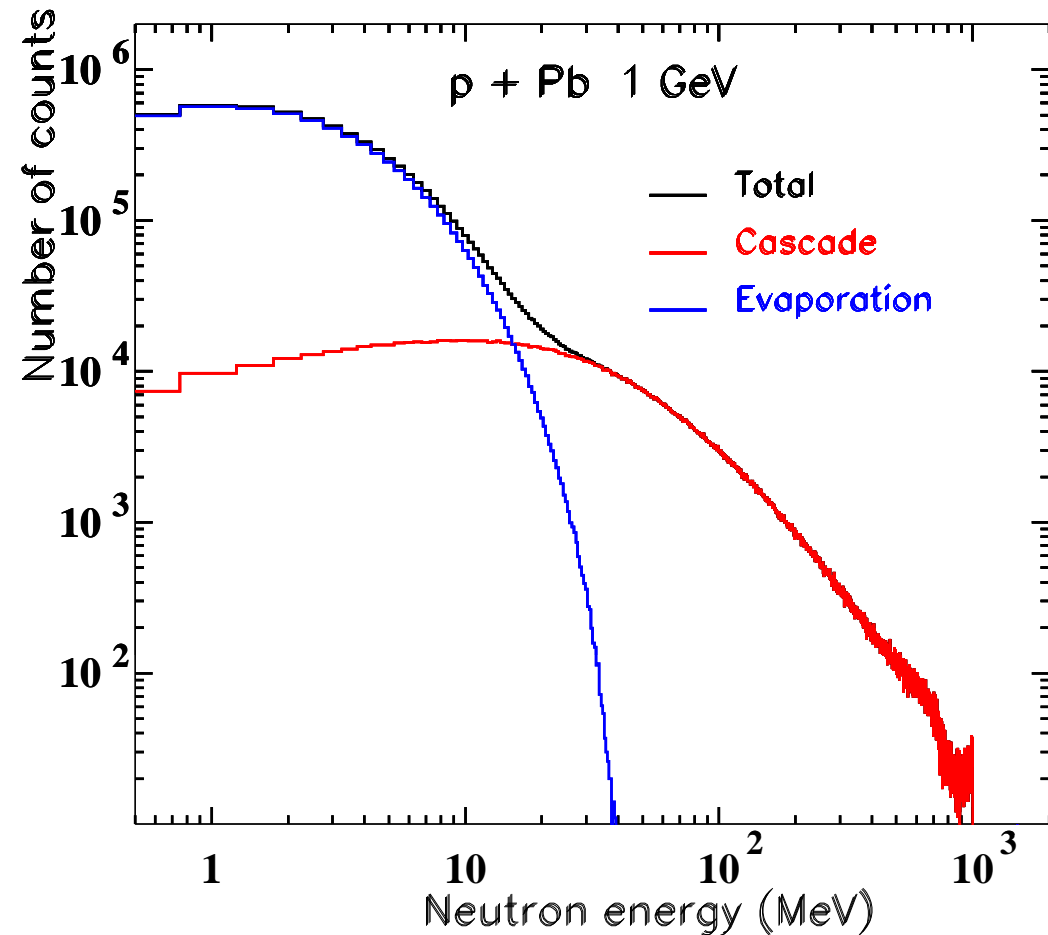
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- Difference between thin and thick target
- A lot of target systems around the world... for many different uses...!
- Some examples to identify common features and problems
- The example I know the best: MEGAPIE
  - Nuclear and neutronic assessment
  - Measurements campaign
  - Modeling
  - Lessons learnt

# Interaction in a thin target

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- ~2 neutrons with  $E > 20$  MeV (intra-nuclear cascade)
- ~15 neutrons with  $E < 20$  MeV (evaporation)
- Energy carried out by cascade neutrons is 85% (95% for protons)



# Interaction in a thick target

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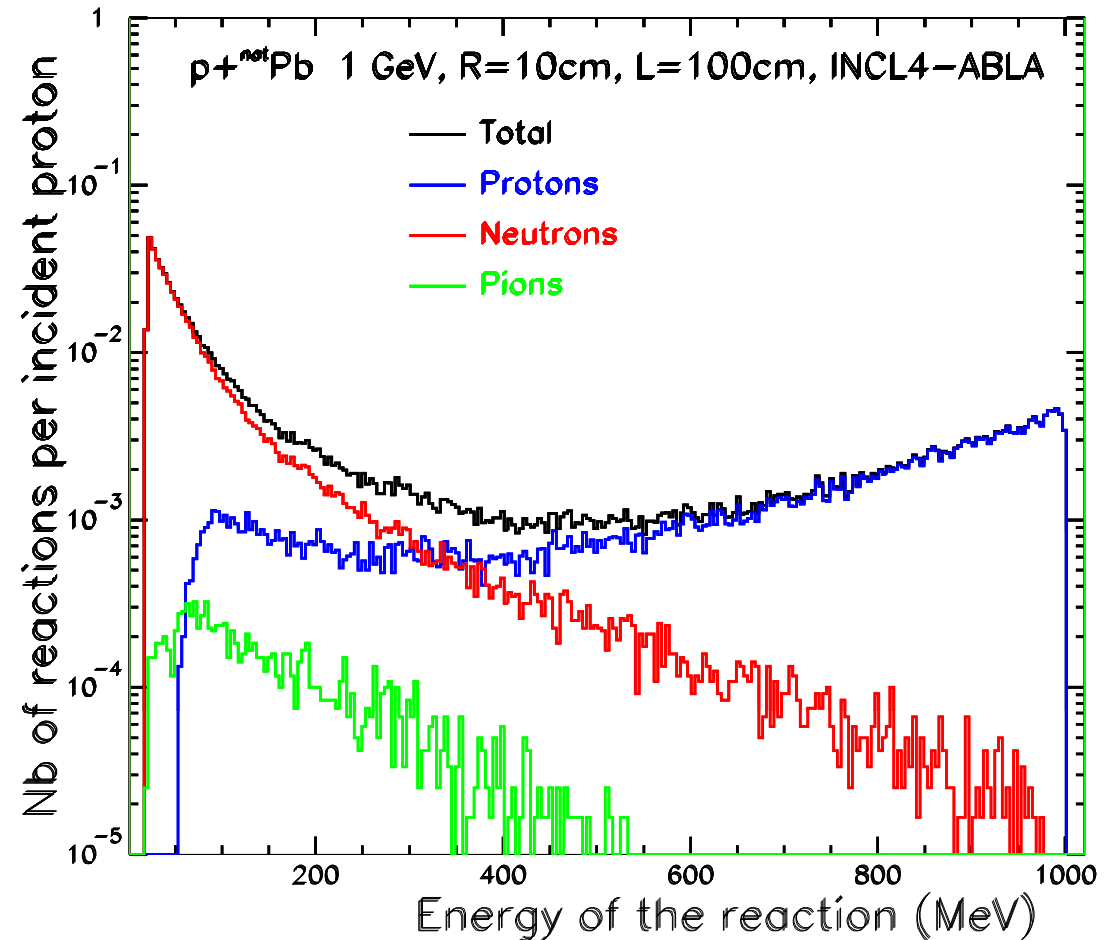
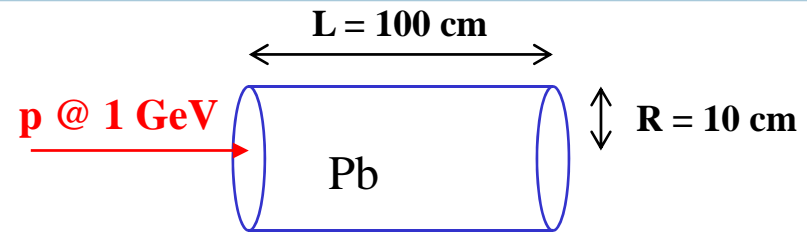


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- Protons interact before slowing down

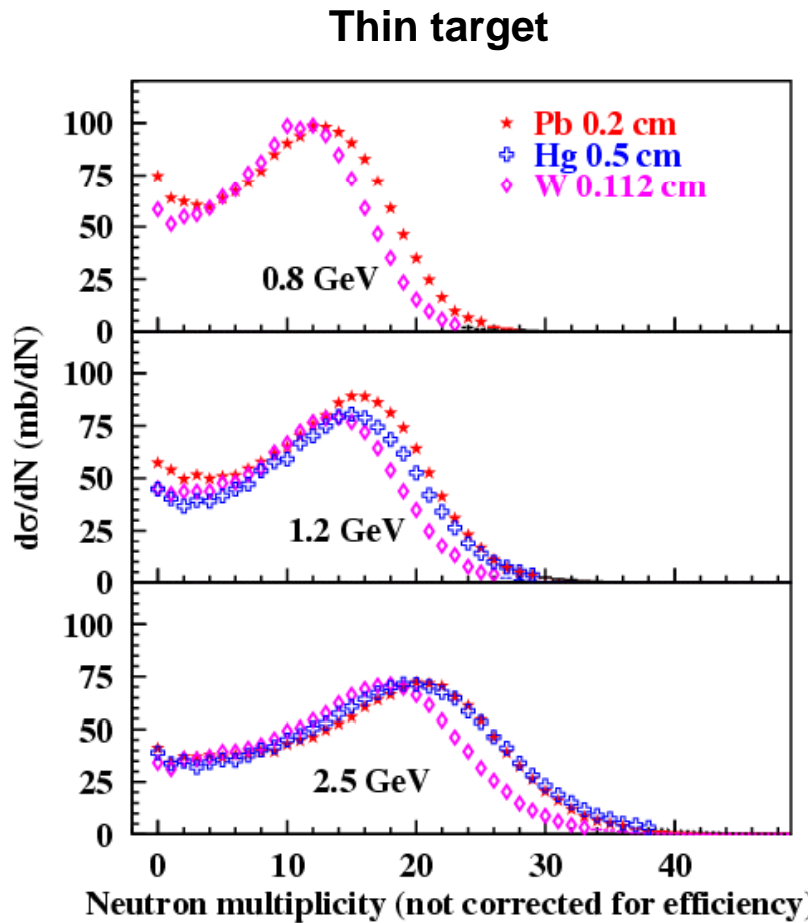
- 3.5 reactions per incident proton in average

- Large number of secondary neutron interactions

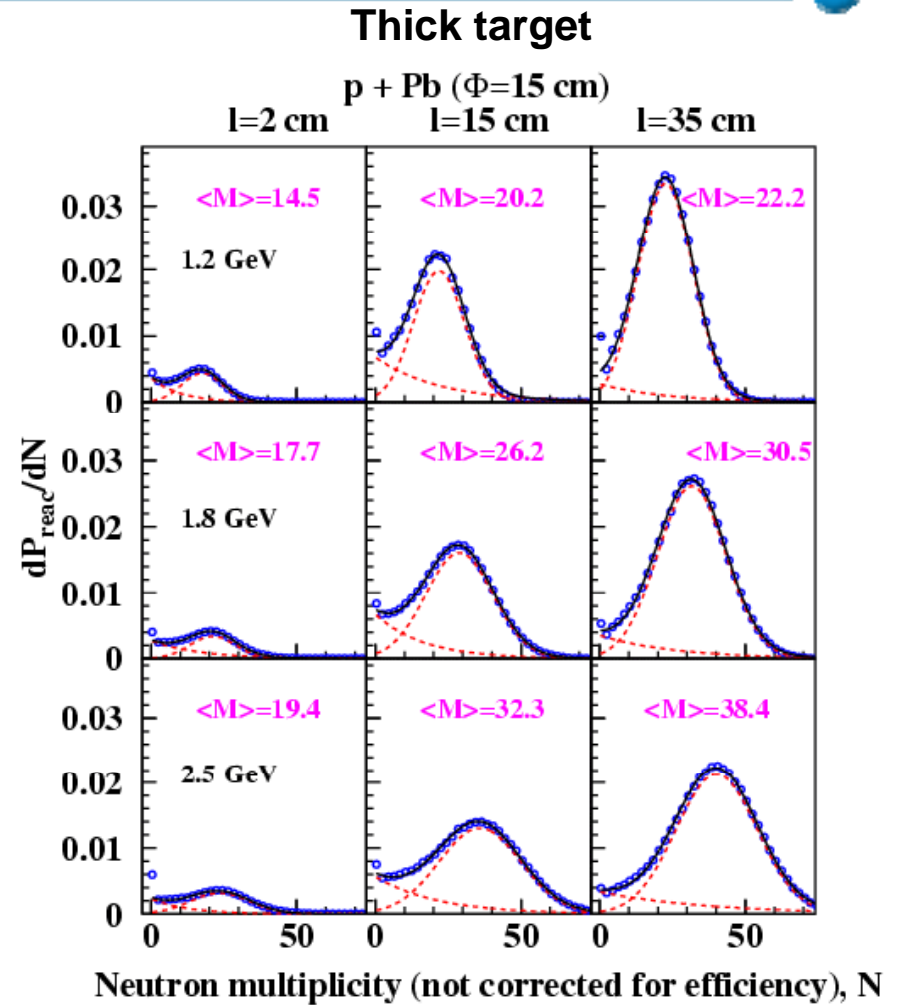


# Neutron multiplicity: thin vs thick target

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A. Letourneau et al., Nucl. Instr. Meth. B170 (2000) 299

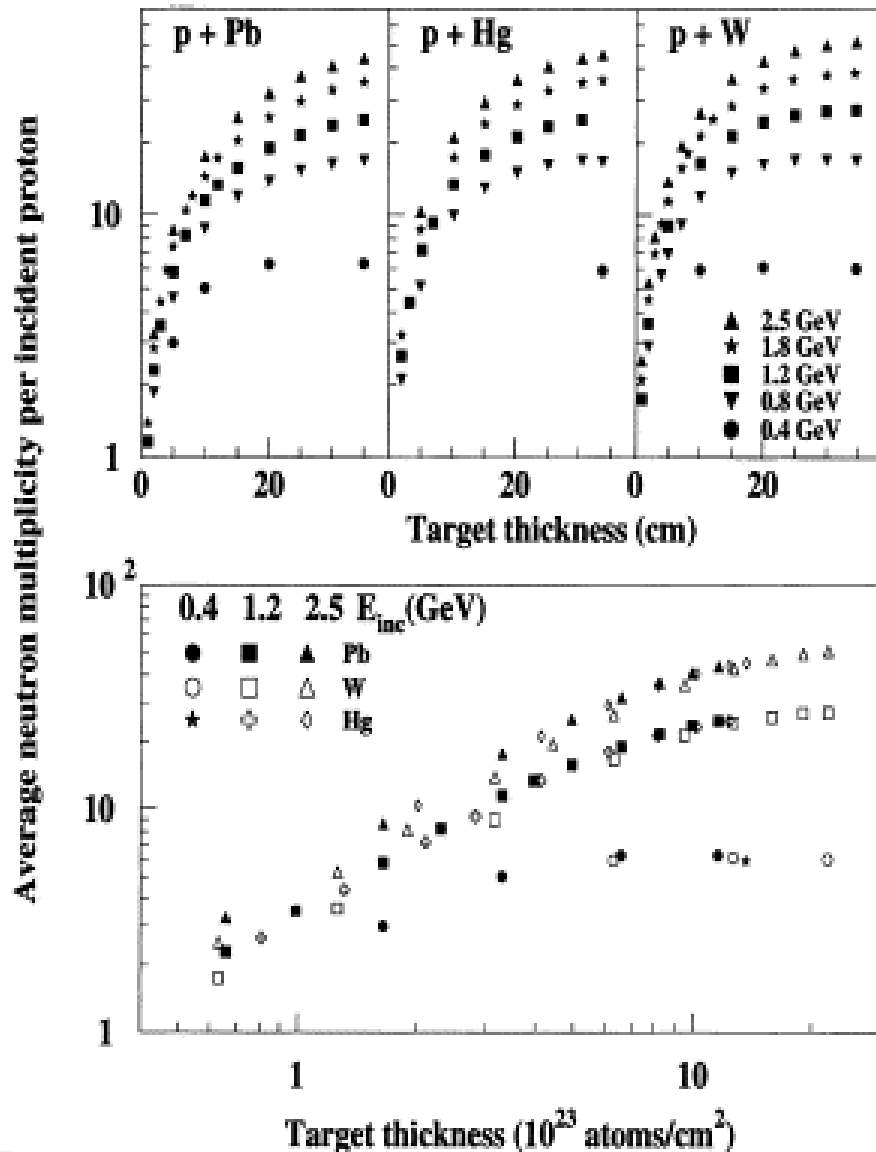


**Increase of  $M_n$  with target thickness due to intra-nuclear cascade**

# Choice of the target material

A. Letourneau et al., Nucl. Instr. Meth. B170 (2000) 299

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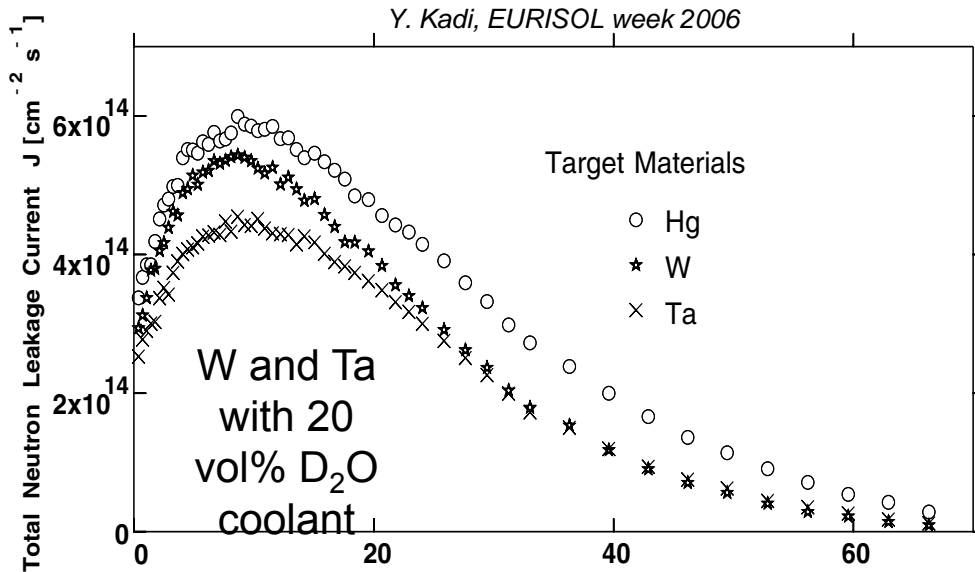


## Neutron production

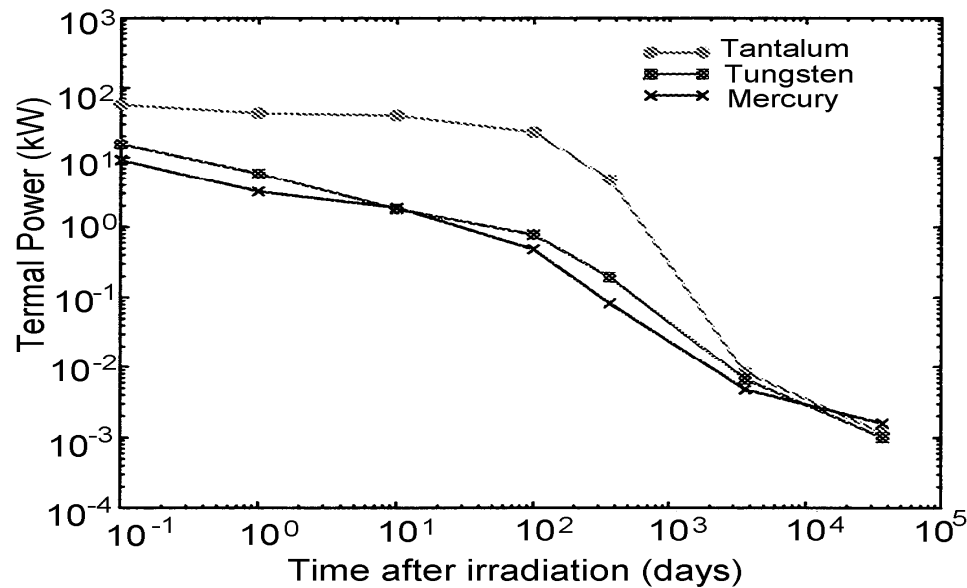
- increases with target thickness and saturates above 30 cm
- is similar for W to Pb for the same atom density

→ choice of material from technological criteria

# Choice of the target material: the Hg case



Calculated neutron leakage for different target materials



Calculated decay power for 200 days of operation in a 5 MW beam



# Choice of the target material

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## Solid targets :

### W, Ta, depleted U

- Spallation products confined
- No container
- Simplicity
- Limited power evacuation
- Necessity of a coolant
  - reduced density of spallation material
- Irradiation damage (dpa, gas production... ) can lead to embrittlement of the target

## Liquid targets :

### Hg, Pb, Pb-Bi eutectic

- Target = coolant
  - high compactness
- Volatile spallation products can be removed during operation
- No structural damages in target
- Damages in window and container
- Corrosion due to liquid metal
- Highly activated circulating fluid
- Ancillary heater(s) necessary to keep Pb or LBE liquid

# Choice of the incident energy

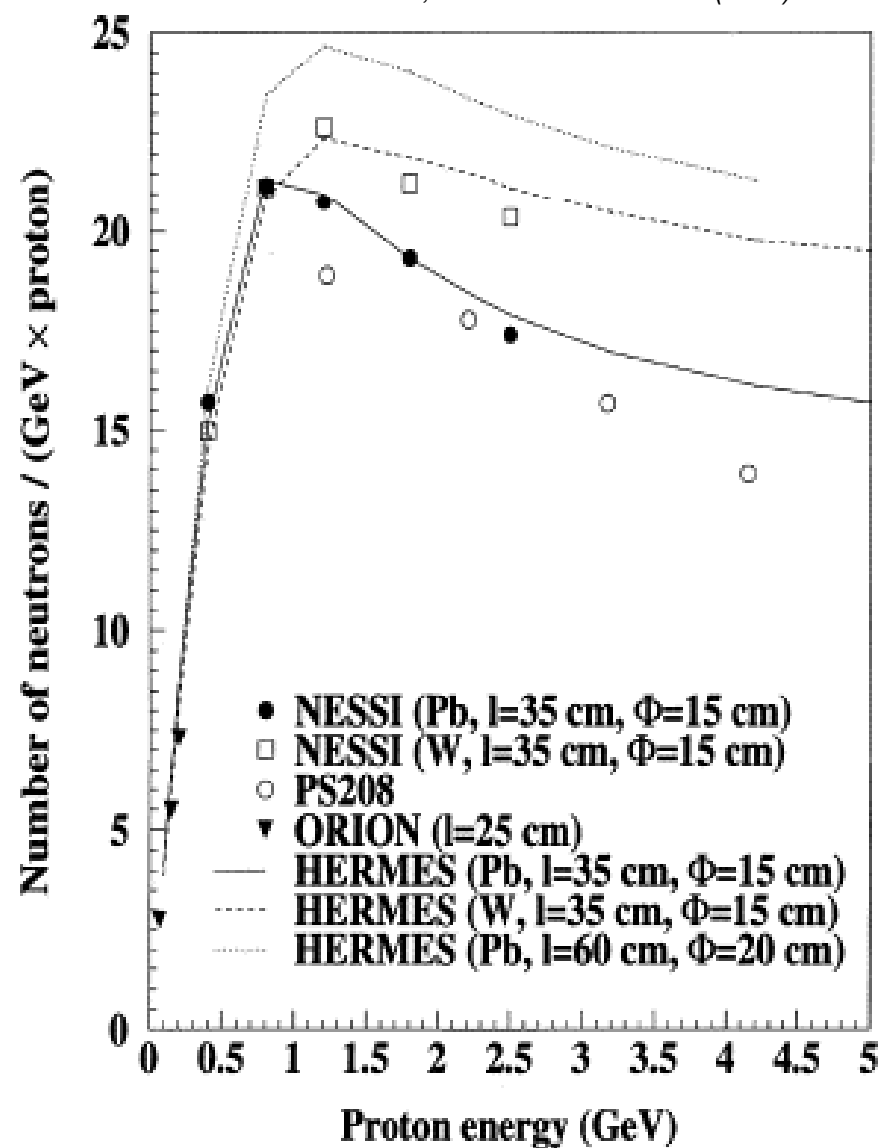
A. Letourneau et al., Nucl. Instr. Meth. B170 (2000) 299

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- In heavy metal targets (Pb, W, Ta, Pb-Bi) around 20 neutrons per incident proton and GeV
- Maximum efficiency around 1 GeV



# Choice of the incident energy

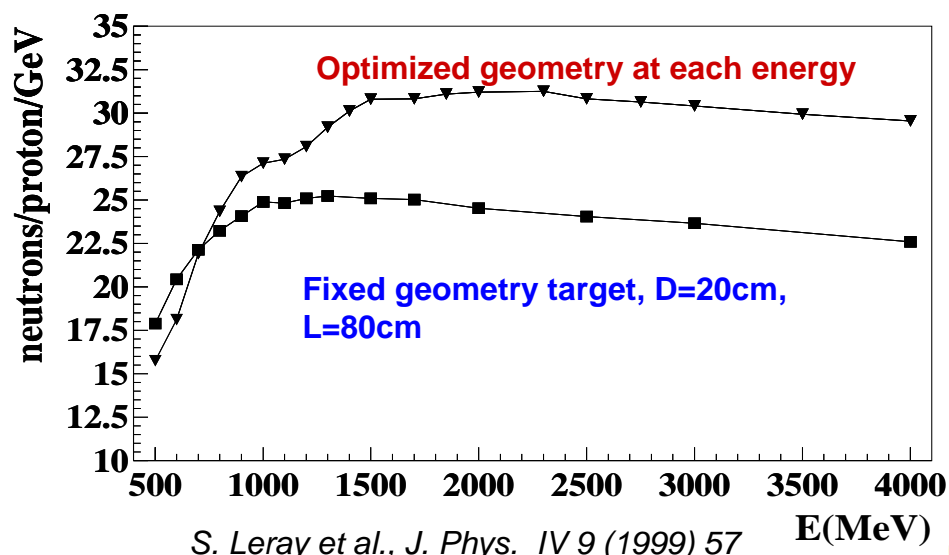
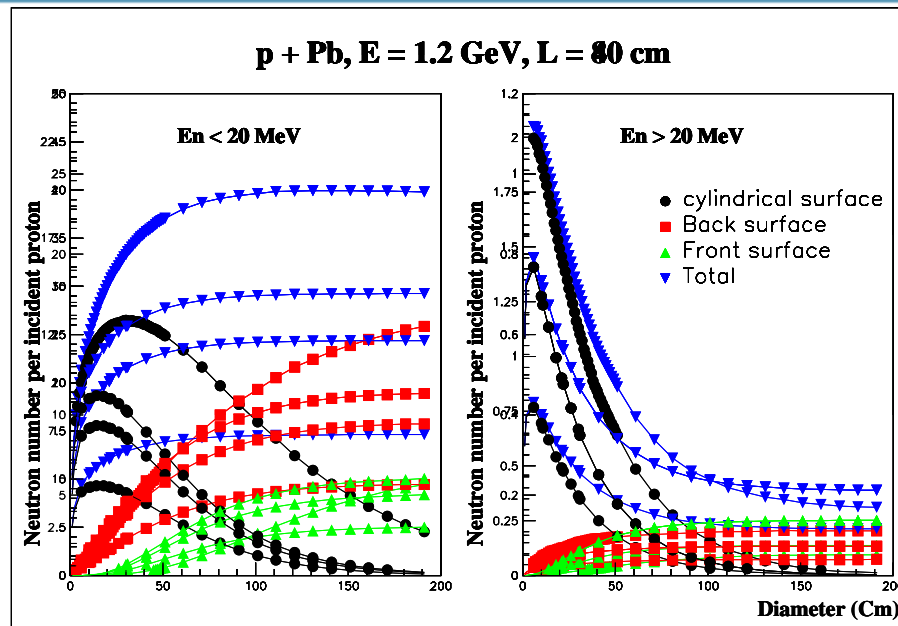
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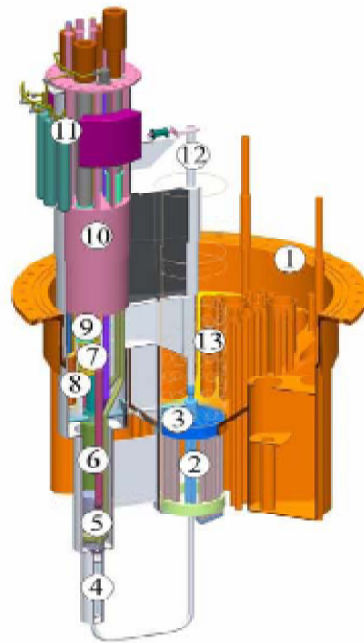
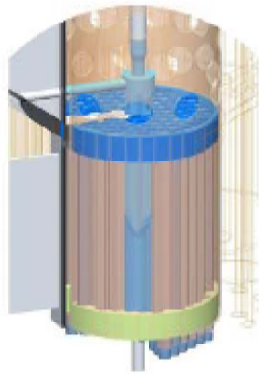
## Target length and diameter optimization

- maximize lateral low energy n
- minimize high-energy leaks



# Some examples: MYRRHA

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1. diaphragm
2. spallation target
3. core support plate slot
4. heat exchanger
5. turbine & pump
6. electromagnetic pump
7. hydraulic drive
8. Pb-Bi conditioning system
9. vacuum system with cryopumps
10. shielding bloc
11. regeneration circuit with absorber pumps
12. proton beam line
13. core barrel



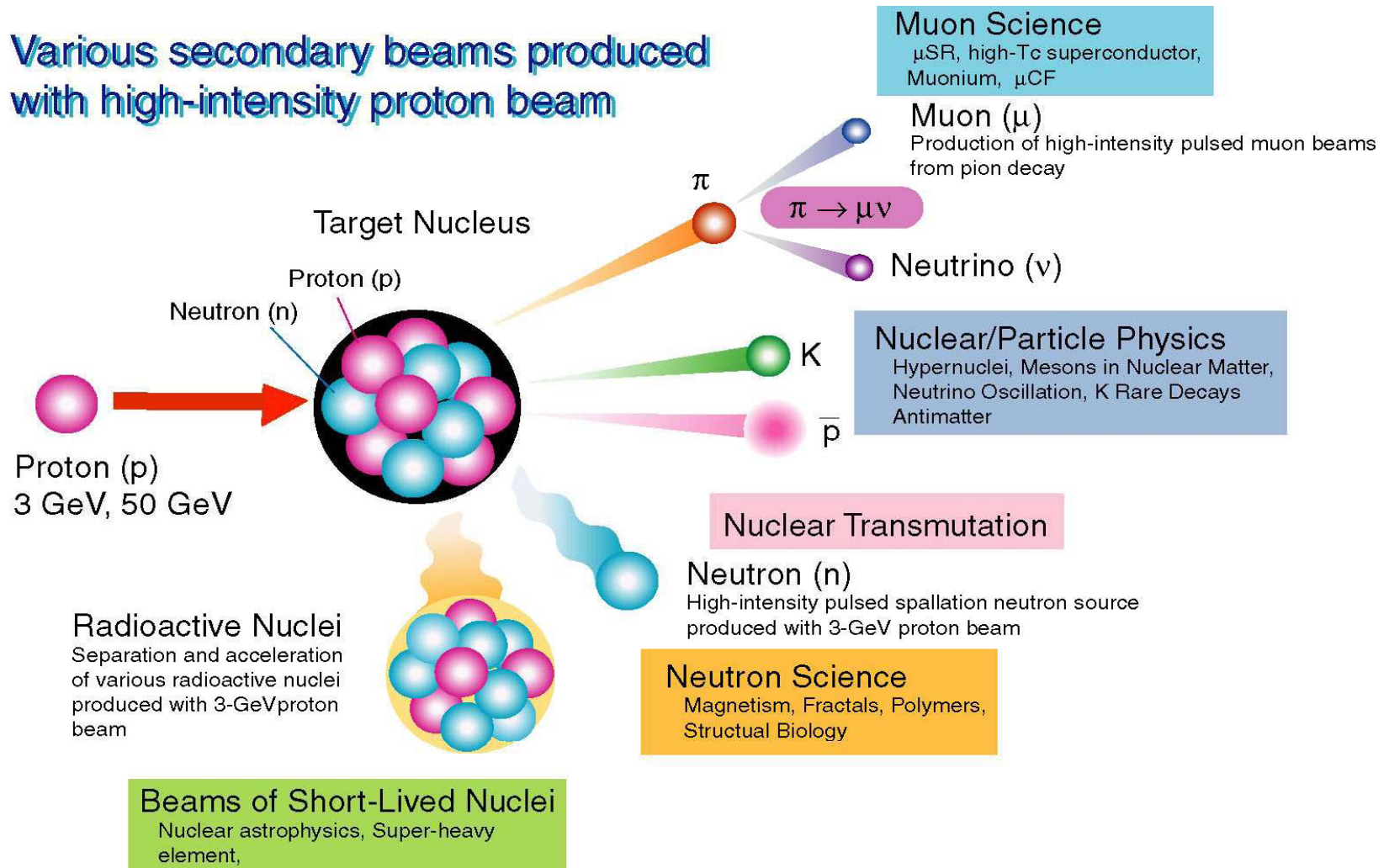
- Target boundary conditions

- Produce  $\sim 10^{17}$  neutrons/s to feed subcritical core @  $k_{\text{eff}} \approx 0.95$
- Accept 600 MeV x 2.5 mA or 350 MeV, 5 mA proton beam
- Evacuate deposited heat 1-1.43 MW depending on beam choice
- Erosion limit:  $v < 2.5$  m/s
- Lifetime:  $> 1$  y

# Some examples: J-PARC

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Various secondary beams produced with high-intensity proton beam



From Sawada, EXA05

# Some examples: J-PARC

## ADS Target Test Facility (TEF-T)

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- **Liquid lead-bismuth**
- **600 MeV-200 kW** proton beam
- **$1.5 \cdot 10^{14}$  n/cm<sup>2</sup>/s**

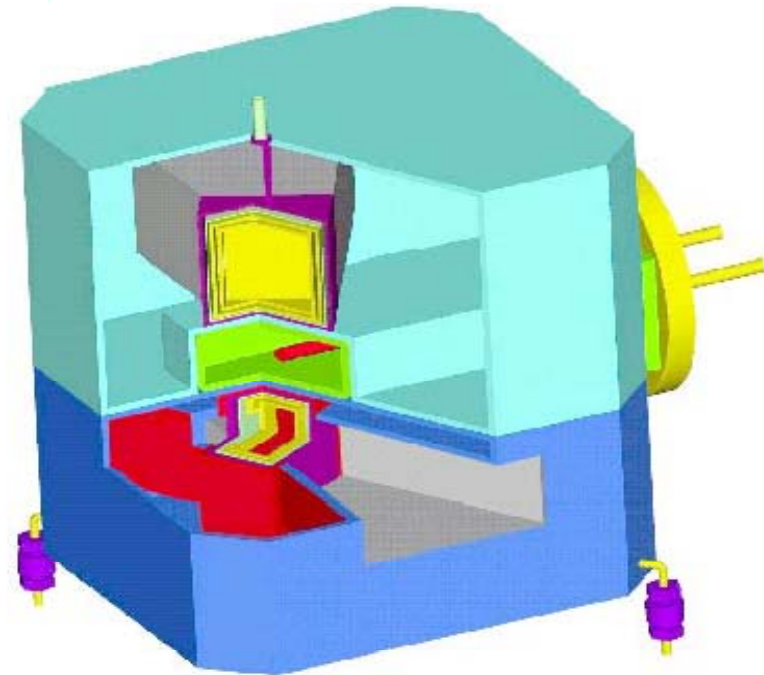
# Some examples: ISIS

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- Proton energy : 800 MeV
- Proton current: 60  $\mu$ Amps
- Average power : 48 kW
- Pulse rate : 10 Hz
- Proton beam size : 1.5 cm FWHM
- Target : Tungsten (W) clad in tantalum (Ta)
- Target coolant : D2O
- Reflector : Beryllium
- Reflector coolant : D2O



# Some examples: SNS

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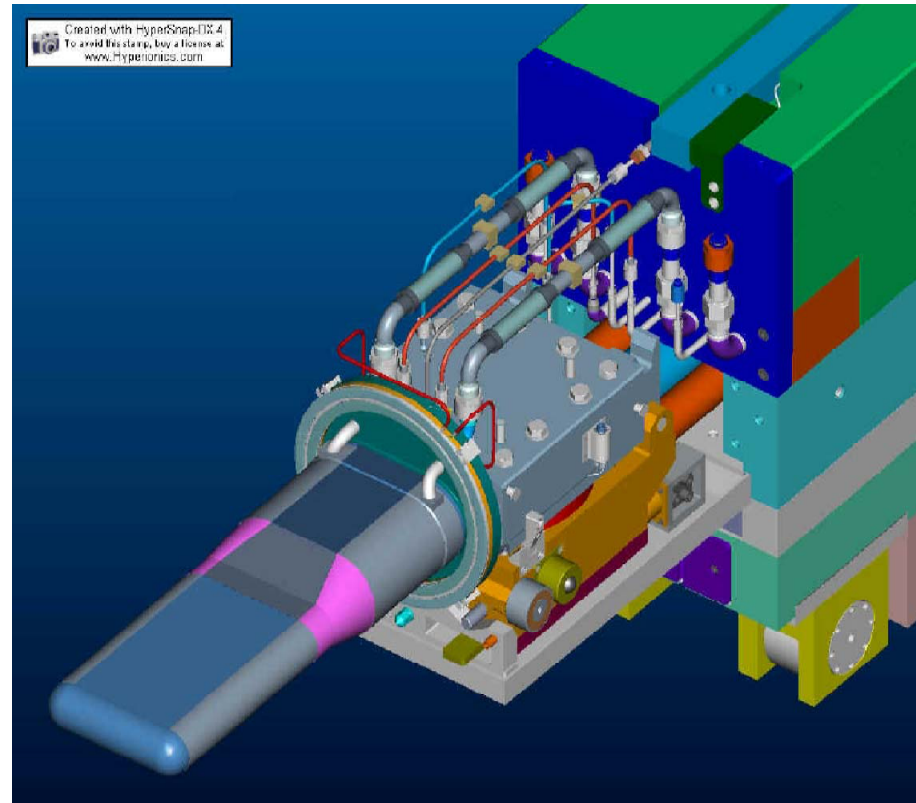
- Nominal Operating Pressure : 0.3 MPa

- Flow Rate : 340 kg/s

- Vmax (In Window) : 3.5 m/s

- Total Hg Inventory : 1.4 m<sup>3</sup>

- Pump Power : 30 kW



- Power absorbed in Hg : 1.1 MW

- Temperature

  - Inlet to target : 60°C

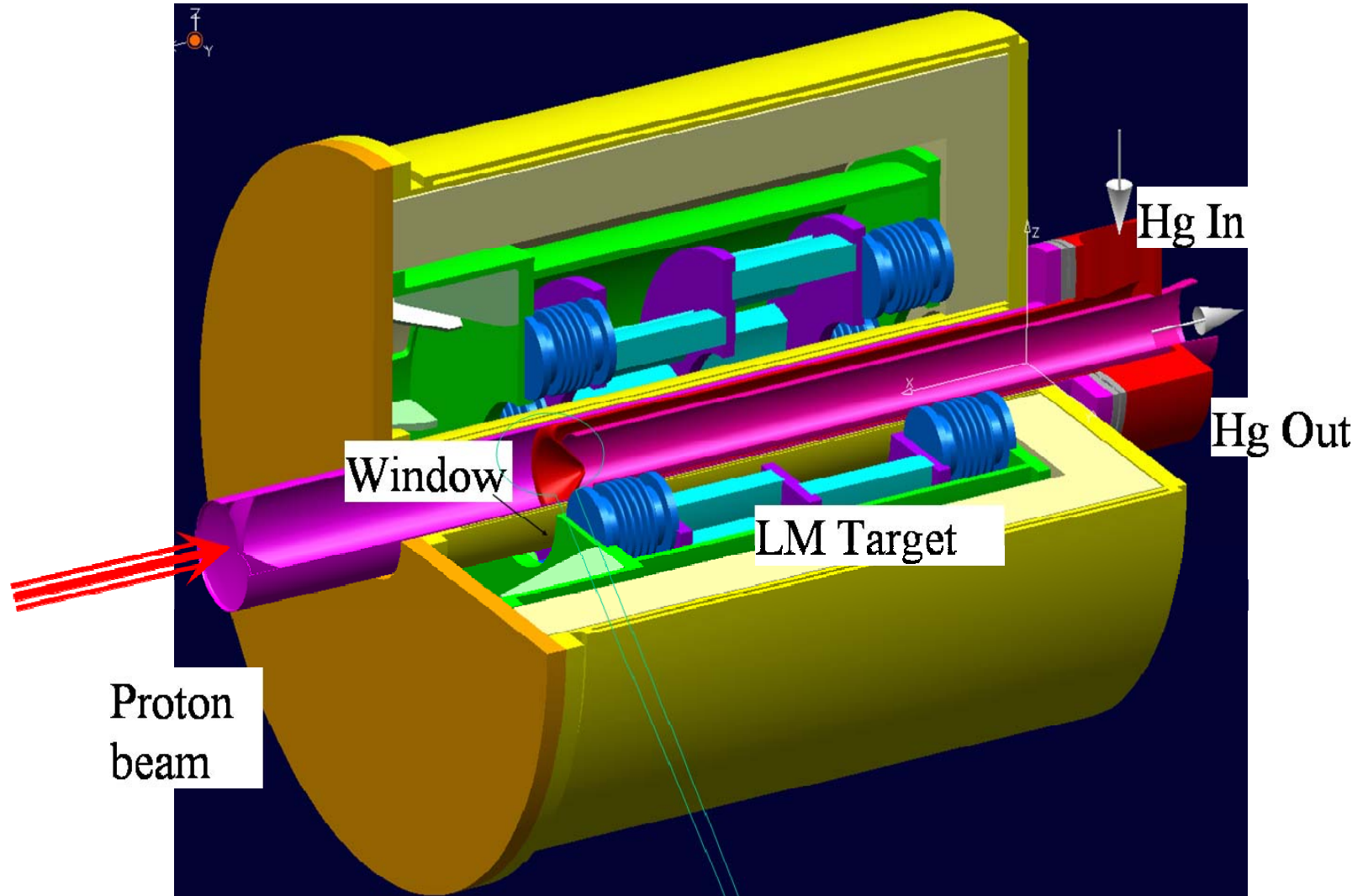
  - Exit from target : 90°C



# Some examples: EURISOL

## Hg converter and secondary fission targets

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# The MEGAPIE project

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- A key experiment in the ADS roadmap
- **MEGAwatt Pilot Experiment (MEGAPIE) (1 MW)** initiated in 1999 in order to design, build and operate a liquid **lead-bismuth spallation target**
- Operate it into the Swiss spallation neutron facility SINQ at PSI
- Minimum design service life fixed at 1 year (6000 mAh).



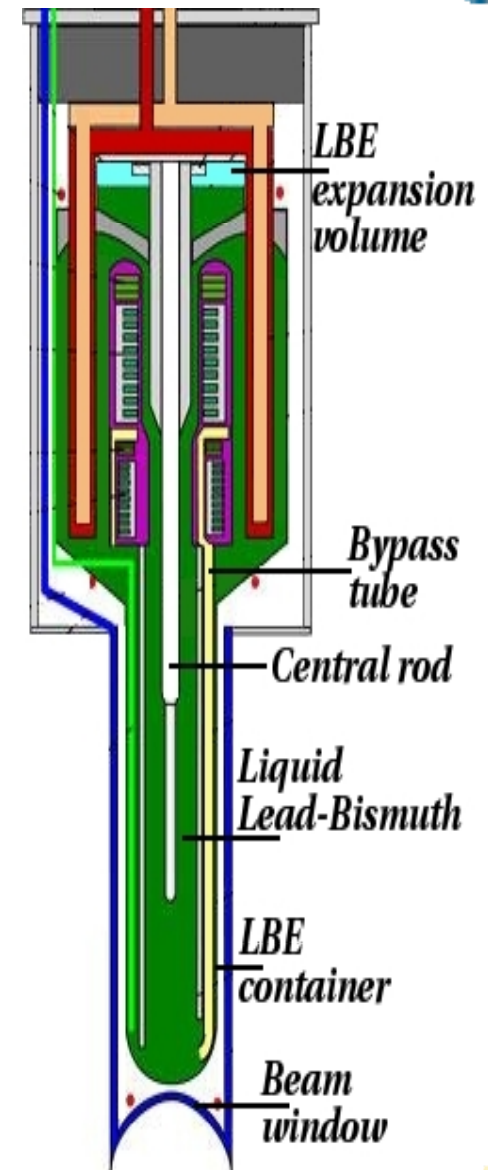
# The MEGAPIE challenges

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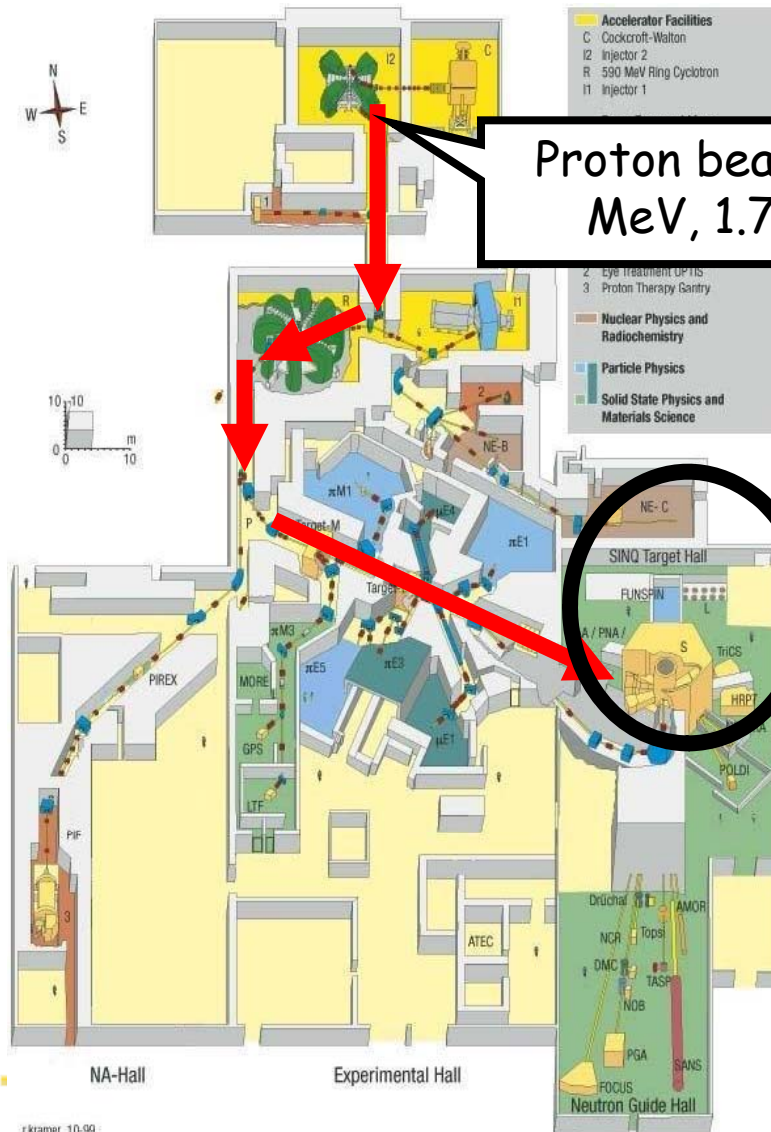
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- **Design** a completely different concept of target in the same geometry of the current spallation targets used at PSI
- Develop and integrate two main **prototypical systems** : a specific heat removal system and an electro magnetic pump system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

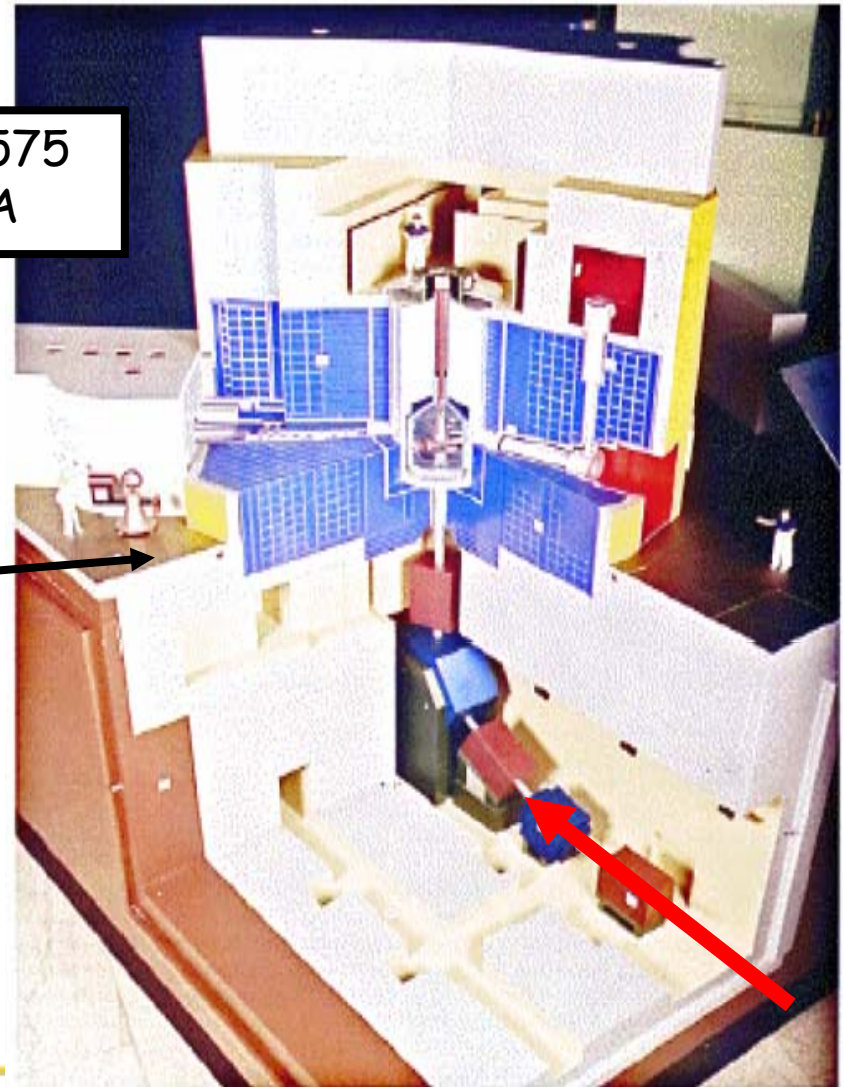


# The MEGAPIE installation at PSI

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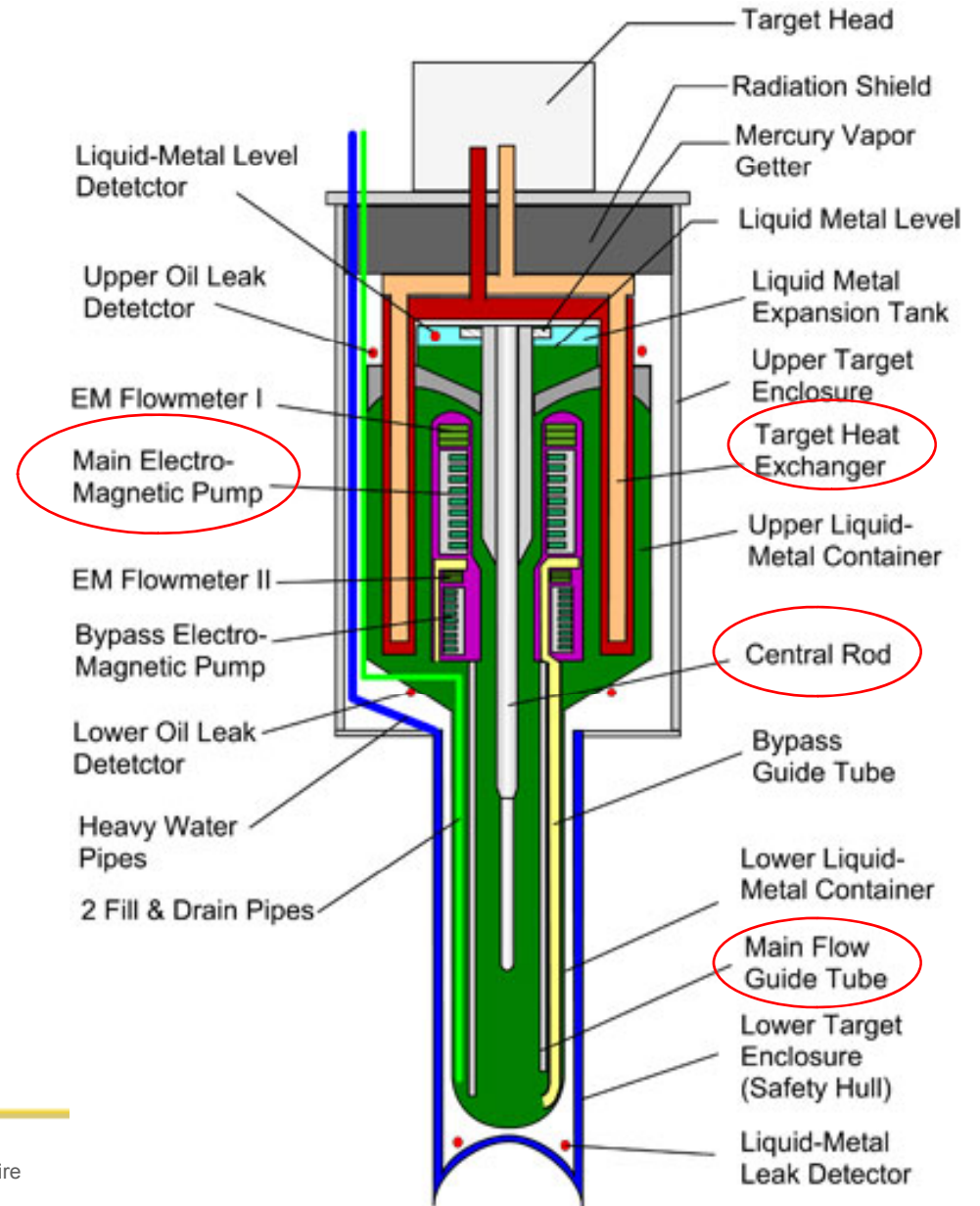
Proton beam 575 MeV, 1.7 mA



# The MEGAPIE target

## CHARACTERISTICS

- Irradiation time: 5 months
- Proton energy: 575 MeV
- Beam intensity: 1,4 mA
- Volume: 80 l of liquid Pb-Bi



# The MEGAPIE target

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# The choice of LBE

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 to the production of activation products (i.e. polonium,...)

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 <sup>3</sup> )	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 <sup>-5</sup> K <sup>-1</sup> )	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

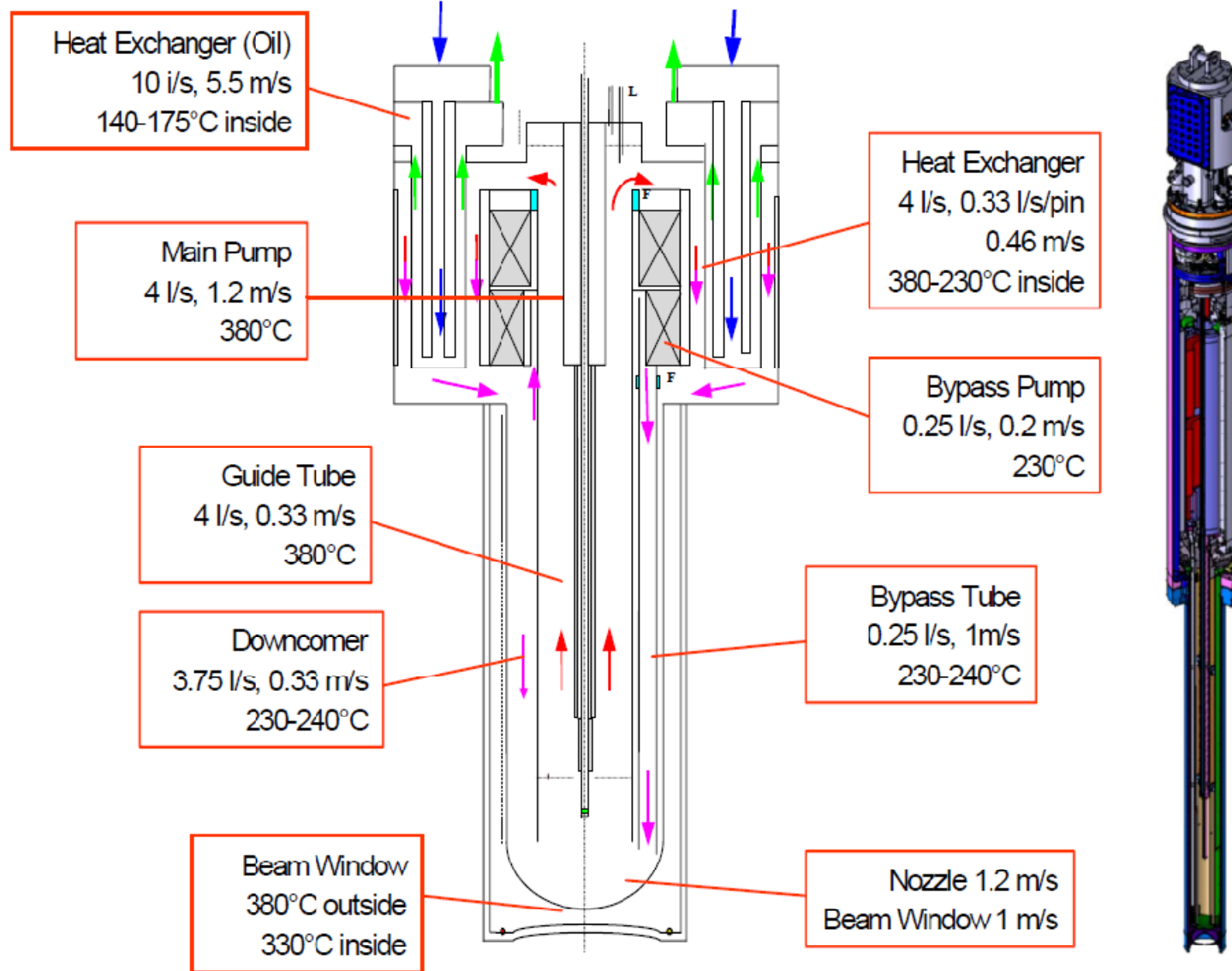
# The choice of T91 beam window

- Compared to austenitic steel 316L, T91 has:
  - higher strength
  - much better resistance to heat deposit (due to a lower thermal expansion coefficient and a higher thermal conductivity).
    - As a result, thermal stresses are about twice as high in 316 as in T91 for a given geometry and heat deposit
  - better corrosion resistance in Pb-Bi due to a low nickel content
- For applications under irradiation up to high doses at temperatures higher than about 400°C, T91 has additional advantages over 316 :
  - Much lower swelling
  - better resistance to the “high temperature helium embrittlement” phenomenon
- The main weakness of martensitic steels is the existence of the Ductile-to-Brittle Transition temperature (DBTT) which is shifted as a result of irradiation.
  - This shift is small for 9Cr martensitic steels up to high doses at irradiation temperatures higher than 400°C.
  - At lower irradiation temperature, a significant DBTT shift occurs. (**T91 : (0.1C, 0.32Si, 0.43Mn, 8.73Cr, <0.01W, 0.99Mo, 0.19V, 0.031Nb, 0.029N, 0.24Ni)**)



# MEGAPIE: main characteristics

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# The MEGAPIE neutronics: motivation

- MEGAPIE is the **first high power liquid metal target** built
- It needed a **full characterization** of the facility from the neutronics point of view (inner and outer flux)
- Gain **experience** and give **recommendations** for
  - HLM target technology
  - ADS development
- Calculation of **important quantities** and their **influence** on the target design:
  - neutron fluxes,
  - power deposition in the target,
  - shielding,
  - dose rates,
  - activation,
  - gas production,
  - radiation damage.
- The main tools were Monte Carlo codes; benchmarking experiments also provided useful data.

General reference: X9 summary report PSI Bericht Nr. 05-12, December 2005, ISSN 1019-0643 and references therein

# The MEGAPIE neutronics: tools

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*From the design, through the operation and to the post-test analysis we needed:*

- **Modeling:**
  - Benchmark and validate MC tools (FLUKA, MCNPX),
  - Integrate a VERY complex geometry (target & facility)
  - Calculate fundamental neutronic quantities (p, n, g fluxes, activation, dose rates, power deposition, ...)
  - Provide a “mapping” of the neutron flux
  - Study the impact of a large spectrum of variables (physics models, materials, geometry, beam, ...)
- **Measurement:**
  - Overall characterization of the facility (matrix of observables)
  - Compare with and validate calculations
  - Span over a large energy range (from thermal to 575 MeV)
  - Span over a large fluency range
  - Need to use different techniques
  - Take benefit of startup phase vs nominal irradiation conditions

# Neutronic benchmark

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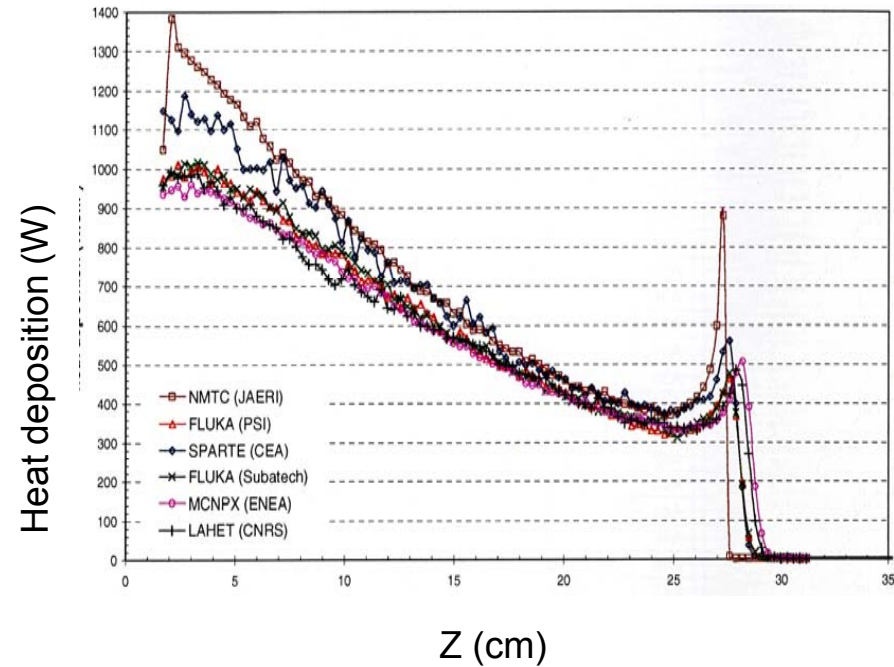
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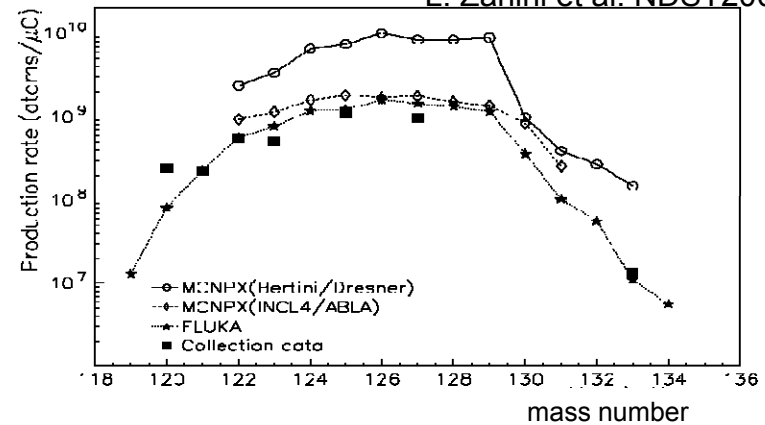
- This initial benchmark exercise was a very important code inter-comparison
- The choice led to FLUKA and MCNPX codes for the prosecution of the work for the following reasons:
  - Consistently good results in the benchmark
  - Code expertise
  - Strong development work for both codes, with constant upgrades of models/cross section sets.

## Lesson learned:

Code validation is of fundamental importance at the beginning of a large project



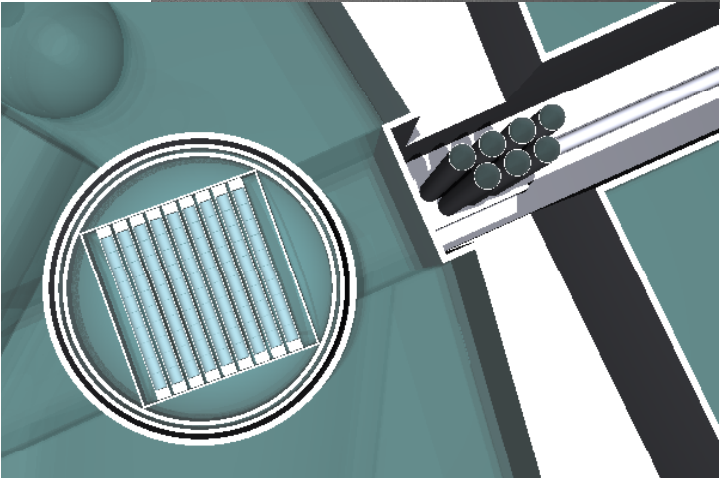
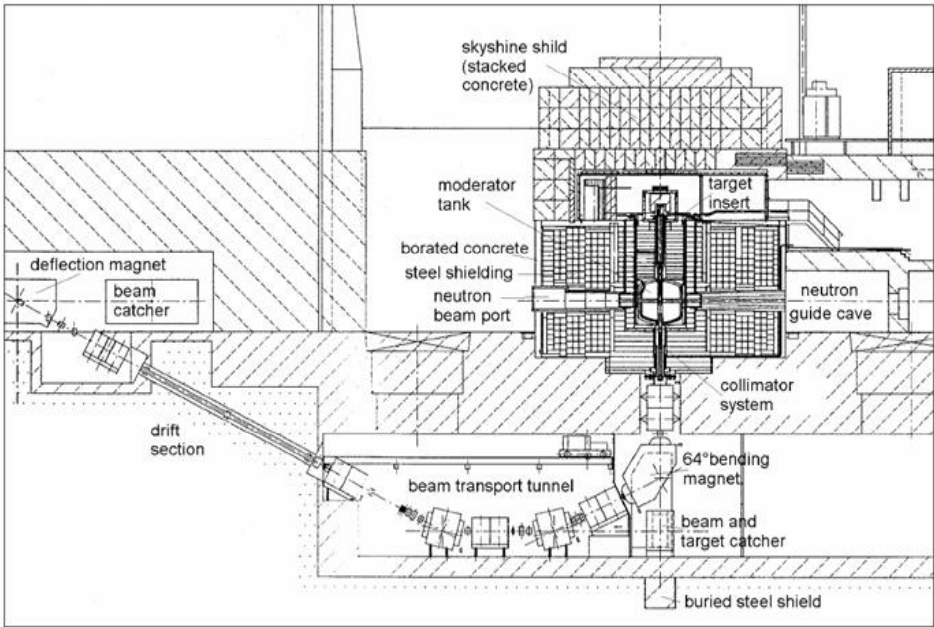
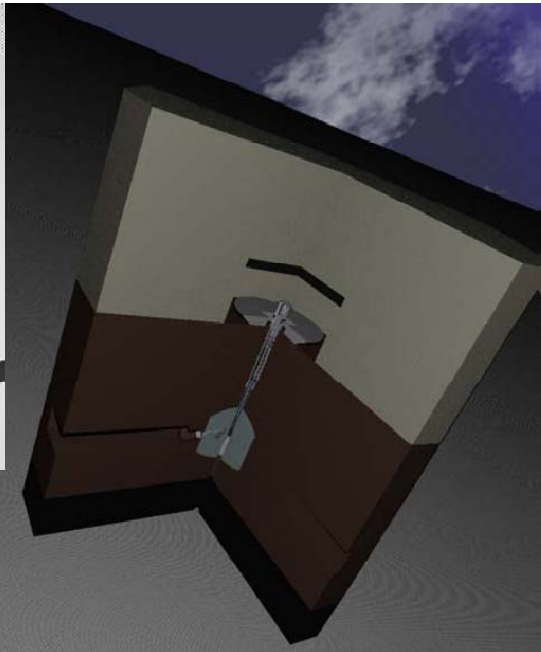
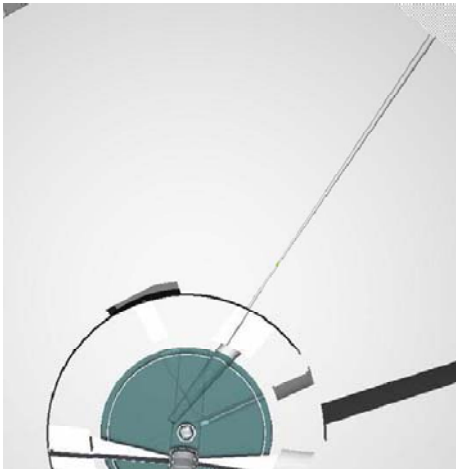
L. Zanini et al. NDST2004



# The MEGAPIE modeling

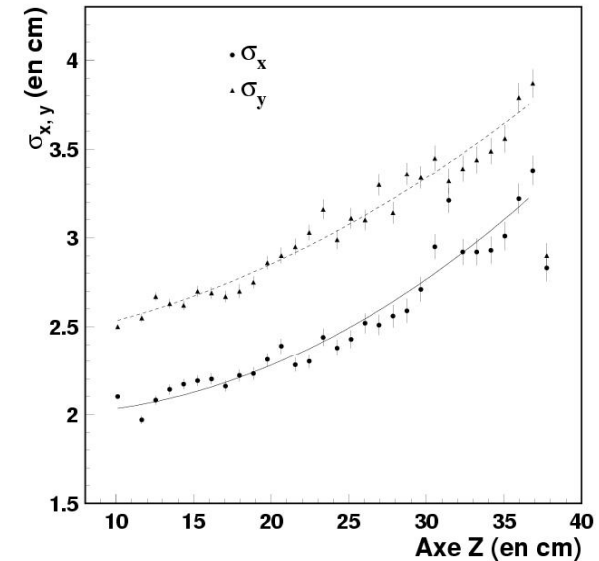
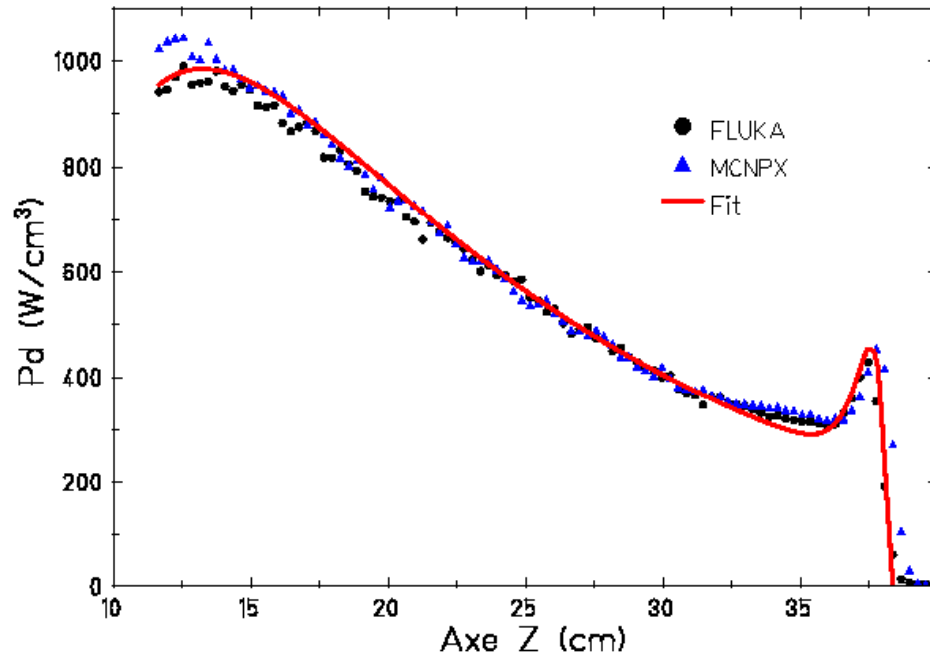
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- Geometry
- Materials (including impurities)
- Beam profile
- Irradiation history
- Physics models



# Power deposition in the LBE

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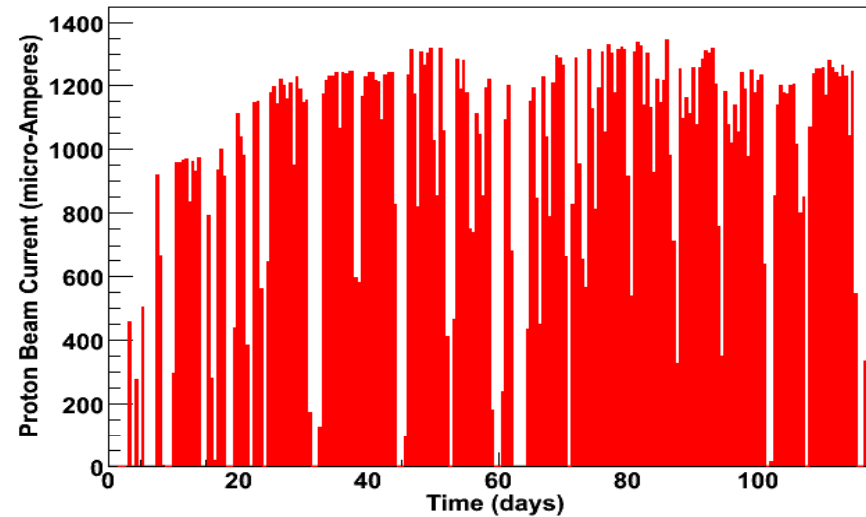
- Determine the power deposition in the Pb-Bi
  - dissipated power in the central part of the target
  - evolution of the axial components of the power deposition
- These results have been fitted with an analytical function and used as **input for thermo-hydraulics calculations.**

# The MEGAPIE operation



Operated successfully as the neutron source for SINQ hall during 2006 :

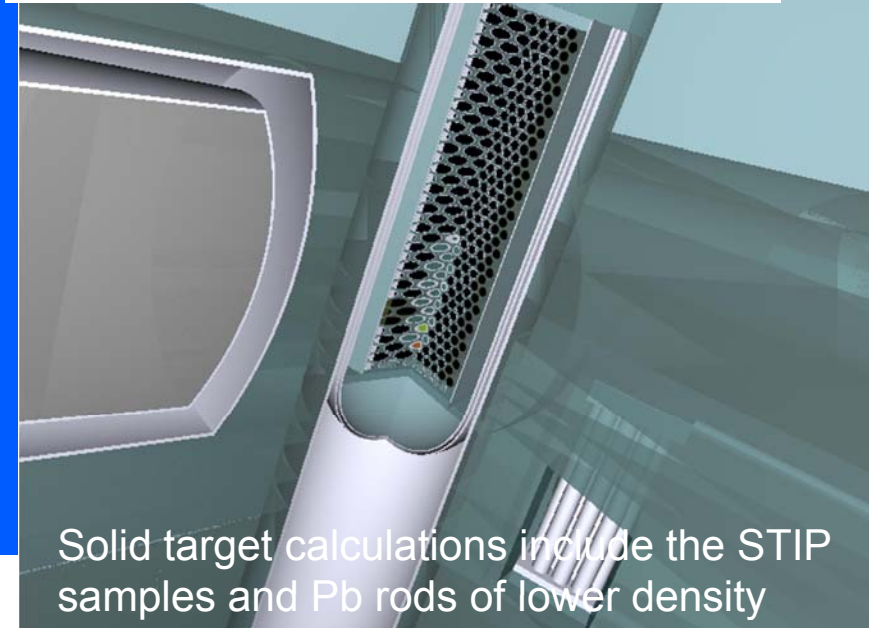
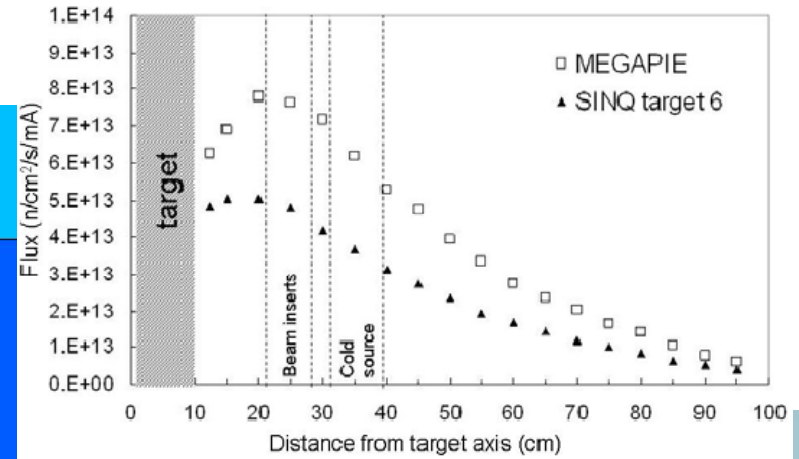
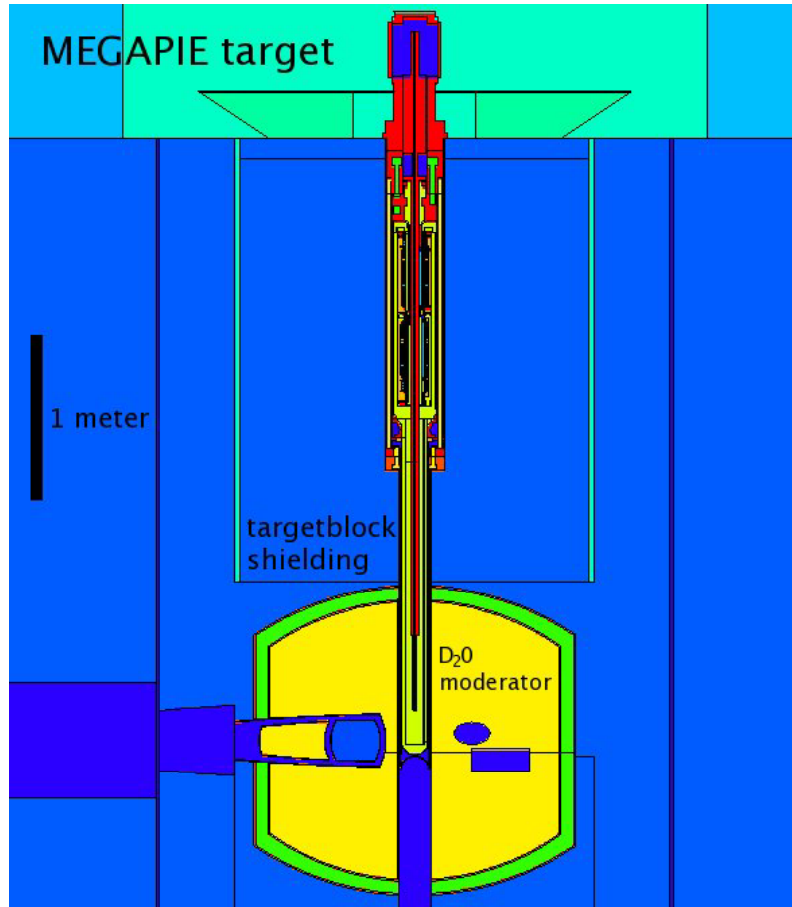
- 4 month beam on, average 960  $\mu\text{A}$
- 80 % more neutron production than current SINQ solid targets



# The target modeling: solid vs MEGAPIE

Validation of simulation codes and re-assessment of the target performances at the measured positions of the fluxes

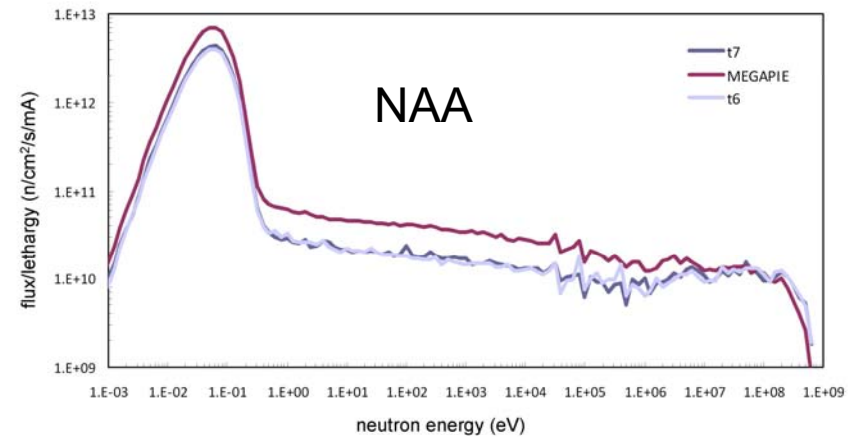
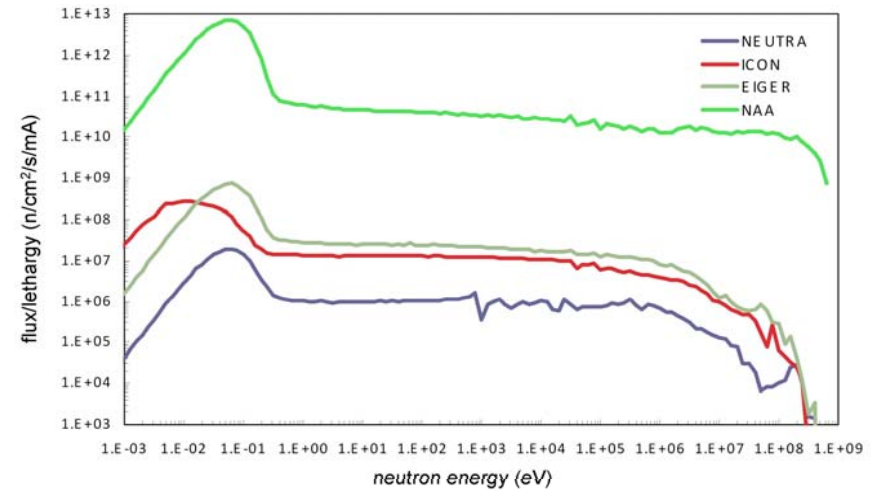
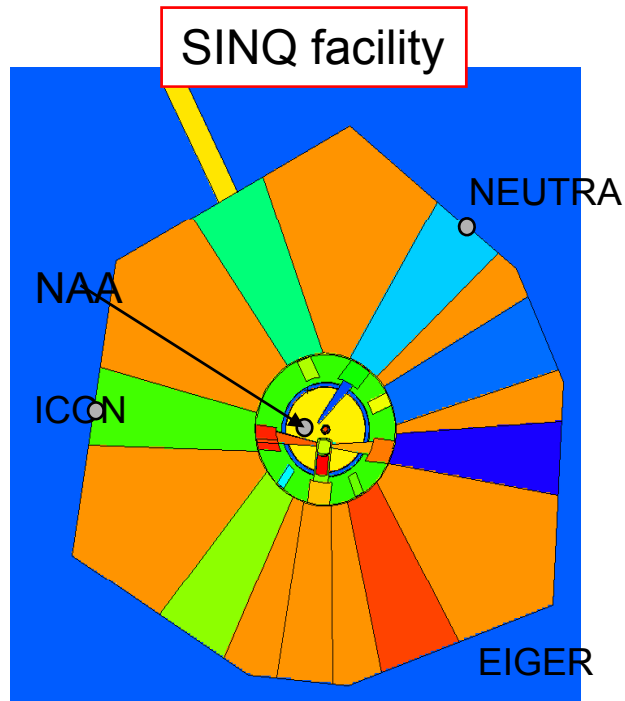
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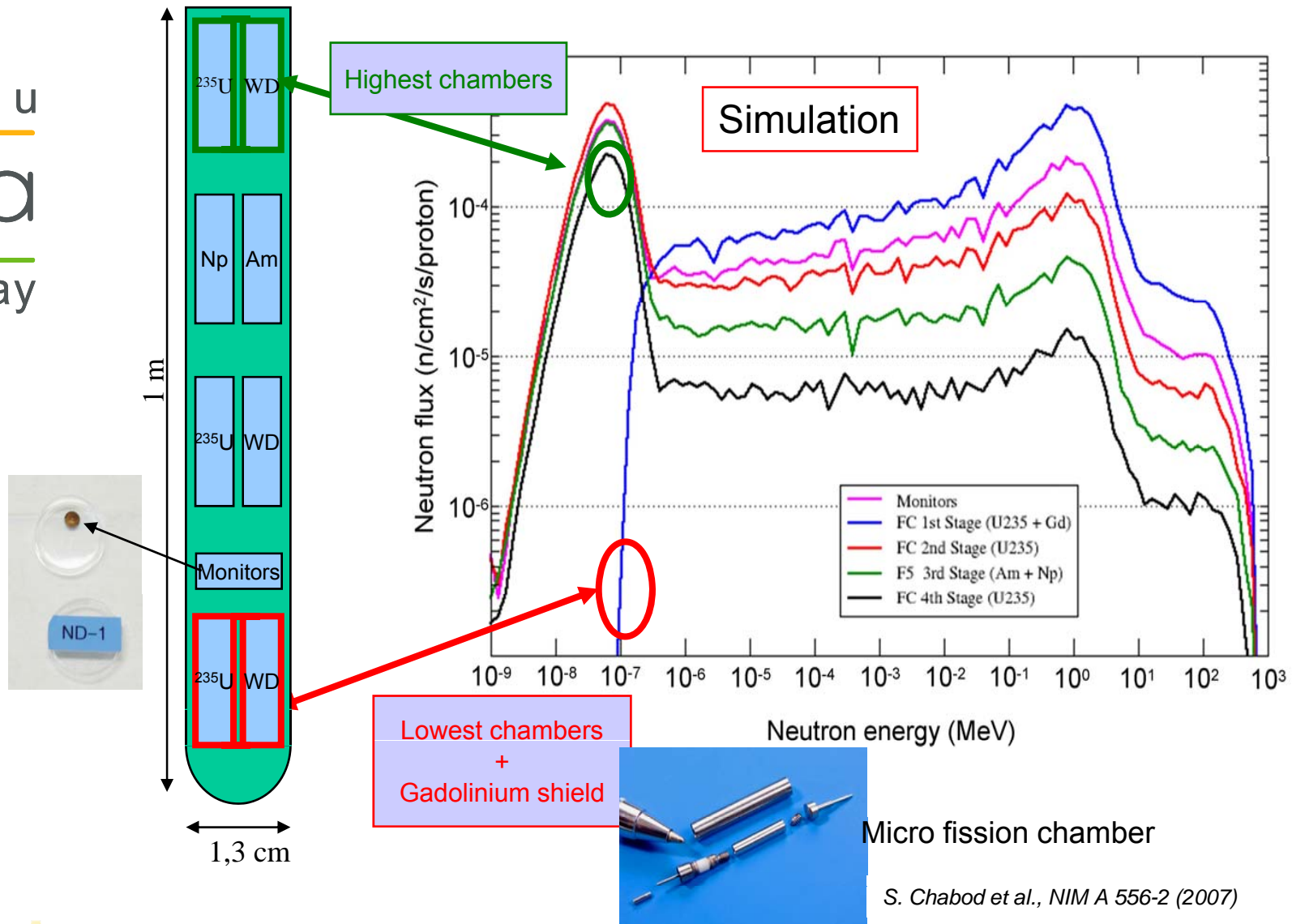
# The modeling: outer flux

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# Inner neutron flux

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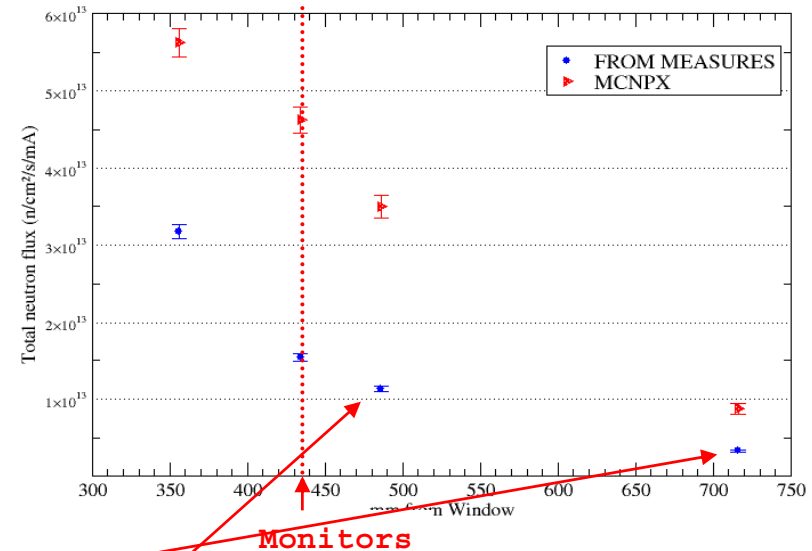
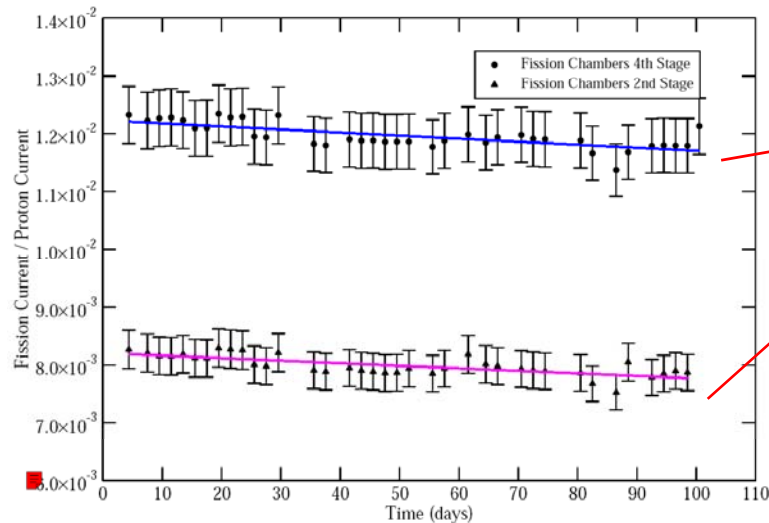
# Inner neutron flux

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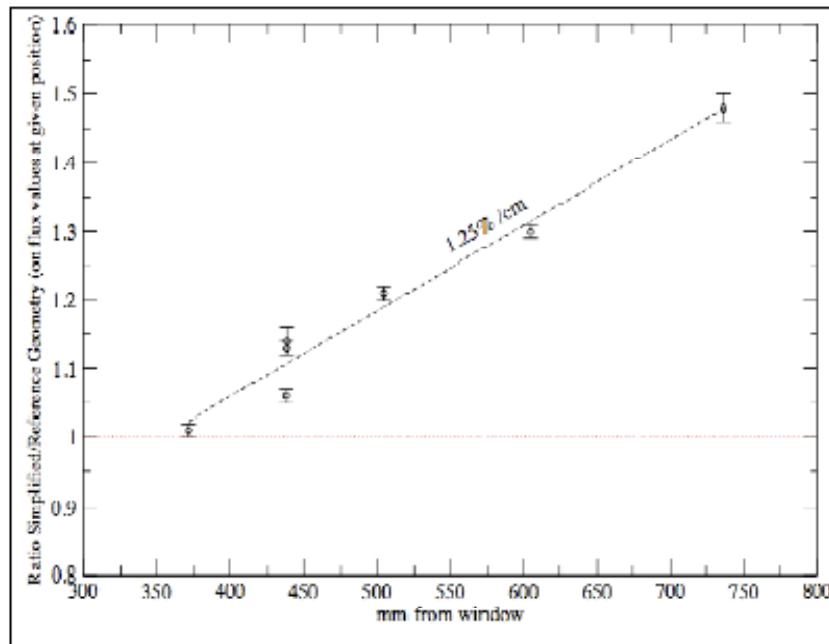
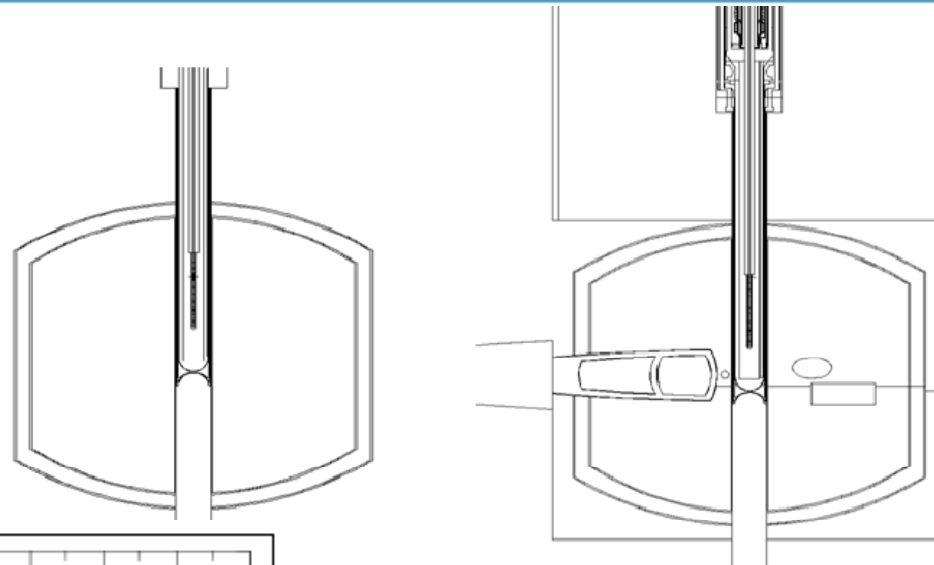
- Online monitoring of neutron flux
- Very tough experimental conditions
- Small  $^{235}\text{U}$  burn-up (6%)
- Extraction of neutron flux from fission current (given a spectrum from MC)



- Discrepancy of a factor  $\sim 2$  between measures (FC & monitors agree!)
- Deep sensitivity analysis on MC parameters and experimental systematics  
*S. Panebianco et al., proc. AccApp07*  
*F. Michel-Sendis et al., proc. ICRS08*
- Beam divergency effect under study

# Effect of structures

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# Effect of boron impurities

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Simulated fission rates (fissions  $s^{-1} mA^{-1}$ ) for different boron concentrations in LBE. The Bertini spallation model was used is MCNPX. Std boron= 2 weight ppm.

Position (isotope)	1 ( $^{235}U + Gd$ )	3 ( $^{235}U$ )	4 ( $^{241}Am$ )	5 ( $^{235}U$ )
No boron	$6.74 \times 10^{-10}$	$8.63 \times 10^{-9}$	$3.92 \times 10^{-11}$	$3.08 \times 10^{-9}$
Std boron	$6.63 \times 10^{-10}$	$8.48 \times 10^{-9}$	$3.99 \times 10^{-11}$	$3.07 \times 10^{-9}$
Std x 10	$6.43 \times 10^{-10}$	$7.28 \times 10^{-9}$	$3.66 \times 10^{-11}$	$2.83 \times 10^{-9}$
Std x 100	$5.44 \times 10^{-10}$	$2.93 \times 10^{-9}$	$1.63 \times 10^{-11}$	$1.02 \times 10^{-9}$

# Effect of beam shape

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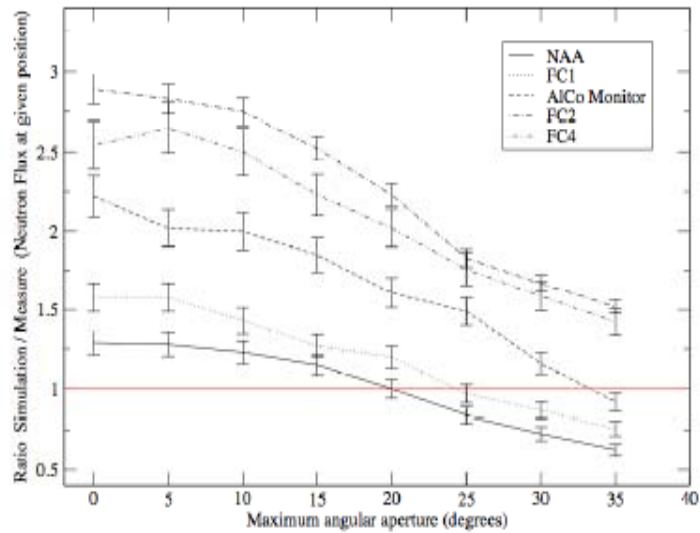
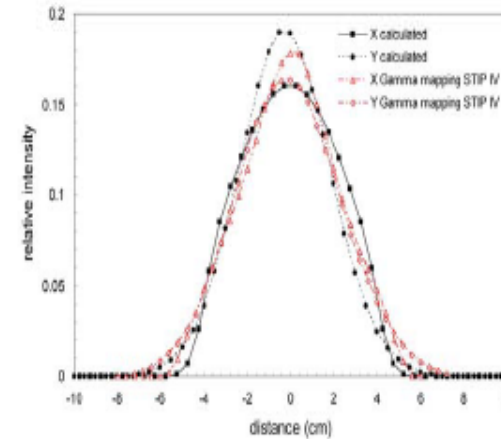
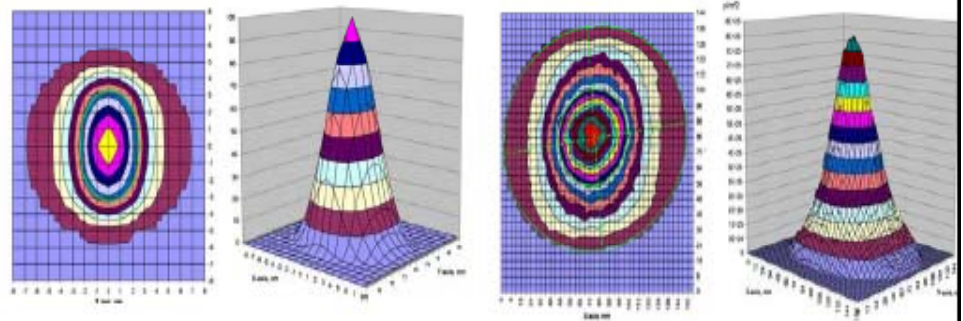


Figure 3.14. Ratios between calculated and measured neutron flux as a function of the the proton source angular aperture.

calculated

Gamma mapping target 4



10-15%  
effect

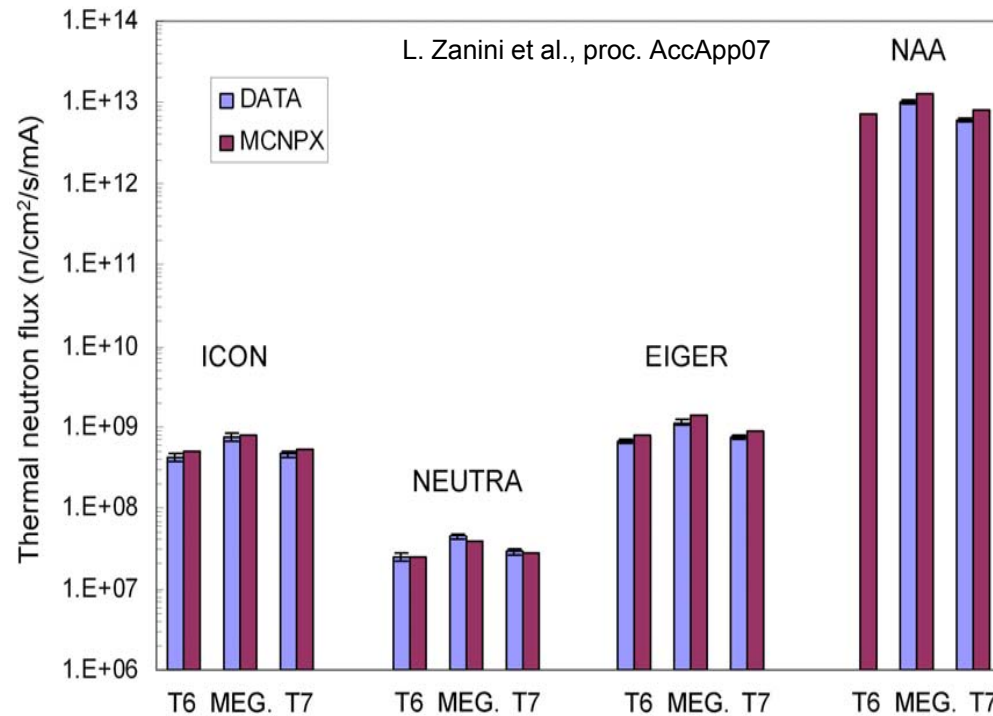
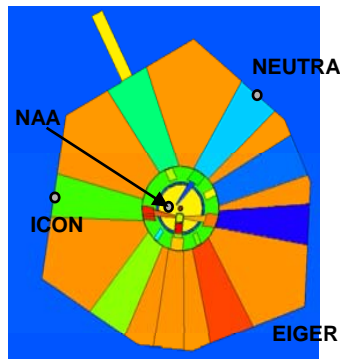
# Thermal neutrons measurement

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**Experimental technique: activation method (thin Au foils w/wo Cd)  
self-shielding correction applied**



**Increase of neutron flux at the thermal beam line MEGAPIE/solid target of about 1.7  
Good agreement with simulated flux**

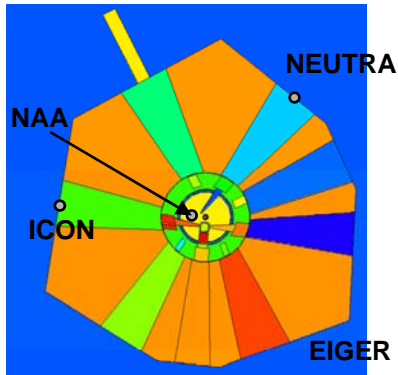
# Epithermal neutrons measurement

Experimental technique: activation method (thin Au foils w/wo Cd)  
self-shielding correction applied

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L. Zanini et al., proc. AccApp07

	$\Phi(\text{epi}) / (\Phi_{\text{epi}} + \Phi_{\text{th}})$ experimental	$\Phi(\text{epi}) / (\Phi_{\text{epi}} + \Phi_{\text{th}})$ calculated
ICON		
T6	0.13	0.13
MEGAPIE	0.19	0.15
T7	0.10	0.12
NAA		
MEGAPIE	0.045	0.038
T7	0.034	0.034

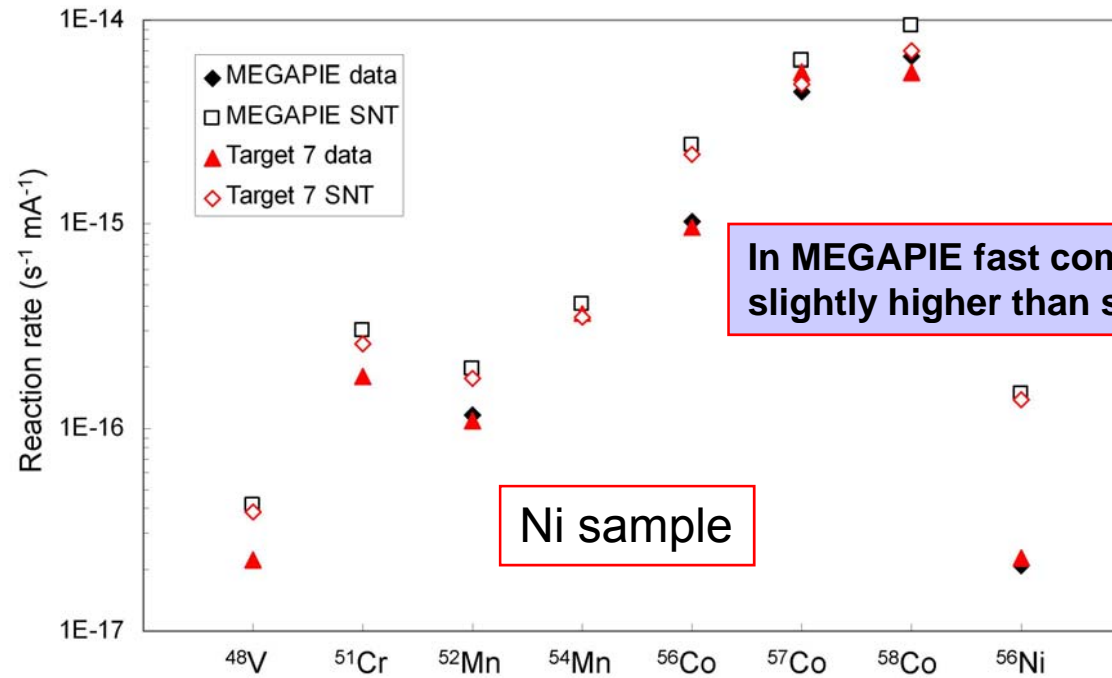
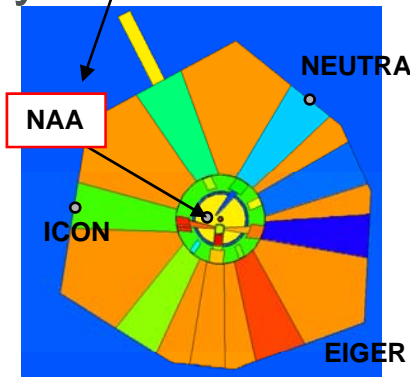
The **epithermal component** with MEGAPIE is **larger** than with the solid targets  
The experimental value are 20-25% higher than the predicted one



# Fast neutrons measurement

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**Experimental technique: reaction rates measurements from a set of activation foils sensitive to threshold reactions (above 1 MeV)**



Calculations performed with **SNT code** (A. Konobeyev et al., *subm. NIM A*)  
Main contribution comes from neutron from 20 to 150 MeV: **(n,xnp) reactions**  
Agreement between calculations and measurement depends on the nuclide  
→ **improvement of evaluated data (JEFF, IEF, ...)**

# Neutronics: recommendations

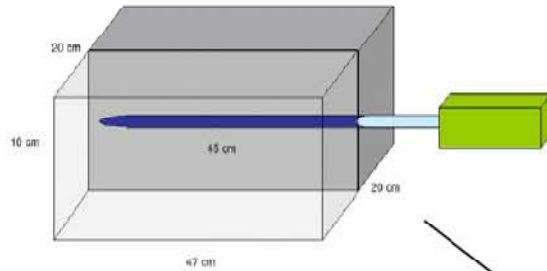
- We stress the importance of correct simulations (geometry, materials, beam profile, models)
- More sensitivity studies
  - Importance of correct beam profile
  - Importance of structural materials
- Better measurements: activation foils should be  $< 5\%$
- More spectral measurements at different parts of the facility

# Delayed neutron measurement

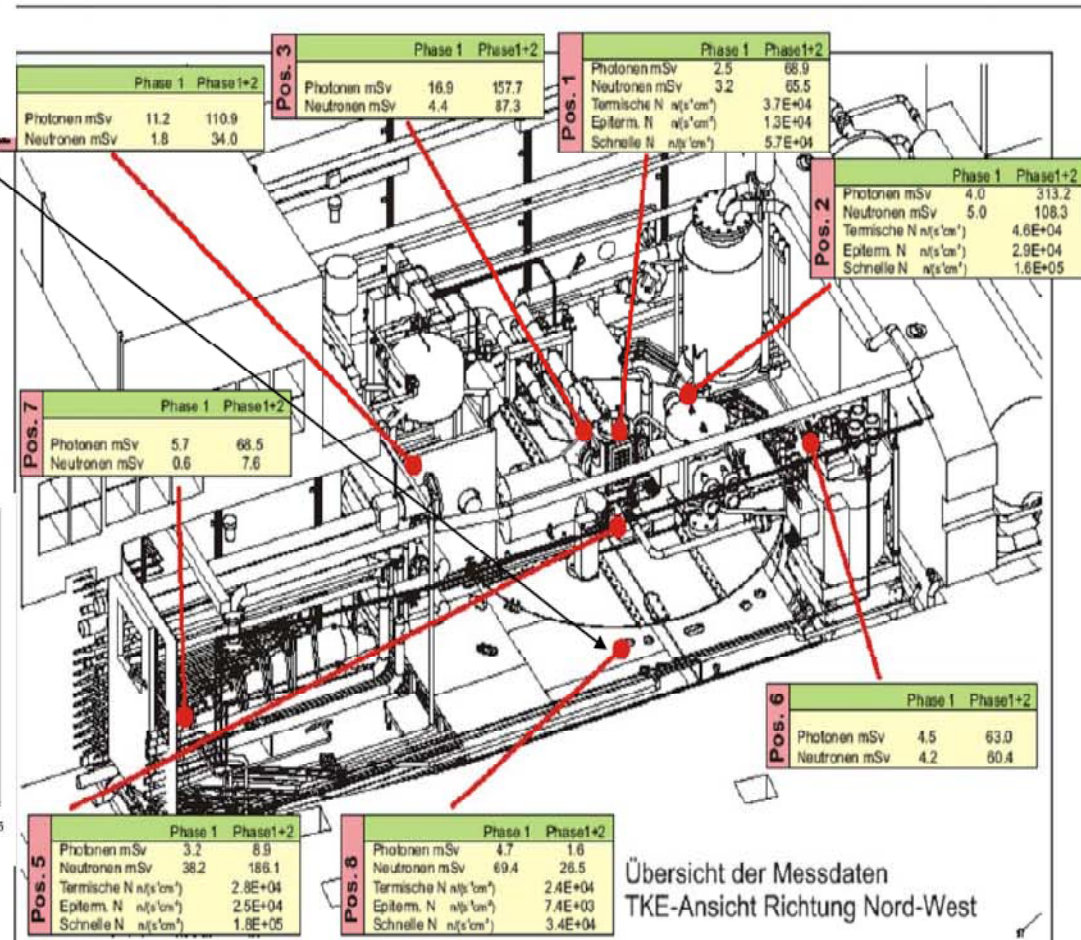
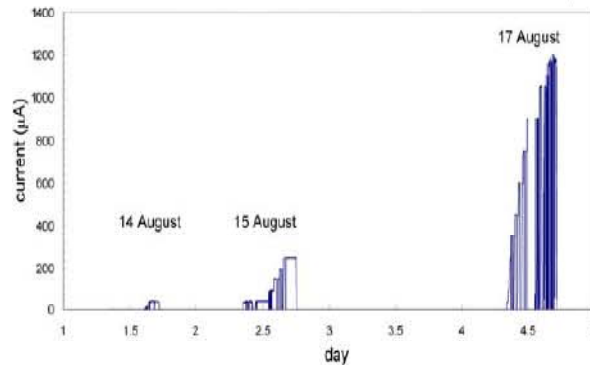
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$^3\text{He}$  tube inside  $\text{CH}_2$  box  
placed above the MEGAPIE  
target



# Delayed neutron measurement

- Fit of the data
- Precursors identified in solid target experiment\*

\* D. RIDIKAS et al., *Europ. Phys. Journ. A*, **32**, 1-4 (2007)

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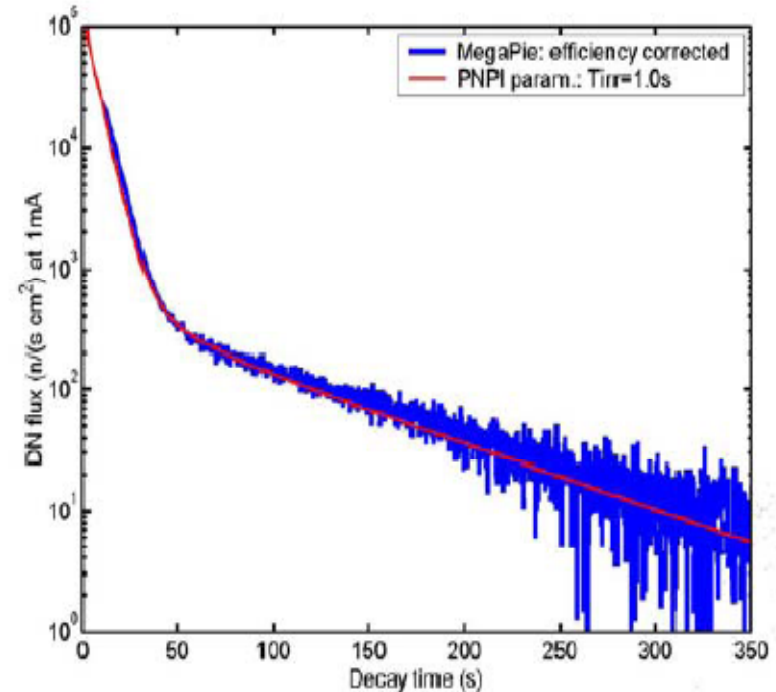
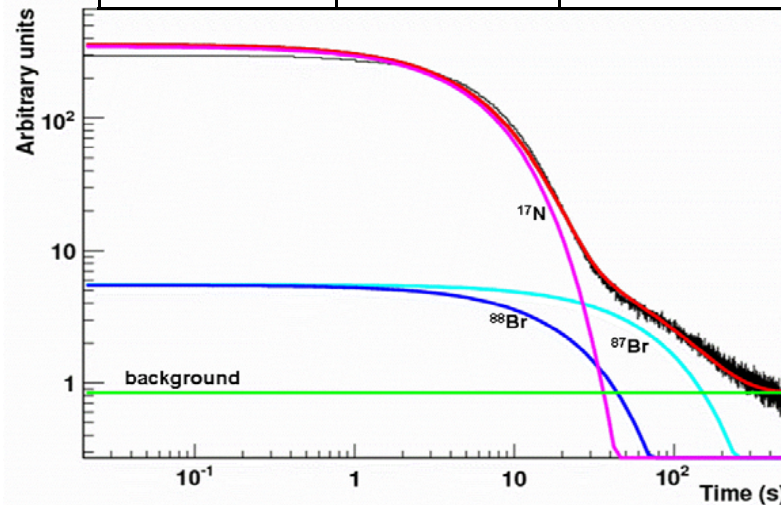


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$$a(x) = \sum_{i=1}^n a_i(x) = \sum_{i=1}^n a_i^a \frac{1 - \exp(-\lambda_i \tau_a)}{1 - \exp(-\lambda_i T)} \exp(-\lambda_i \tau_d(x))$$

$\tau_a \sim 0.5$  s irradiation  
 $T \sim 20$  s relaxation  
 $\tau_d \sim 10$  s heat exchanger

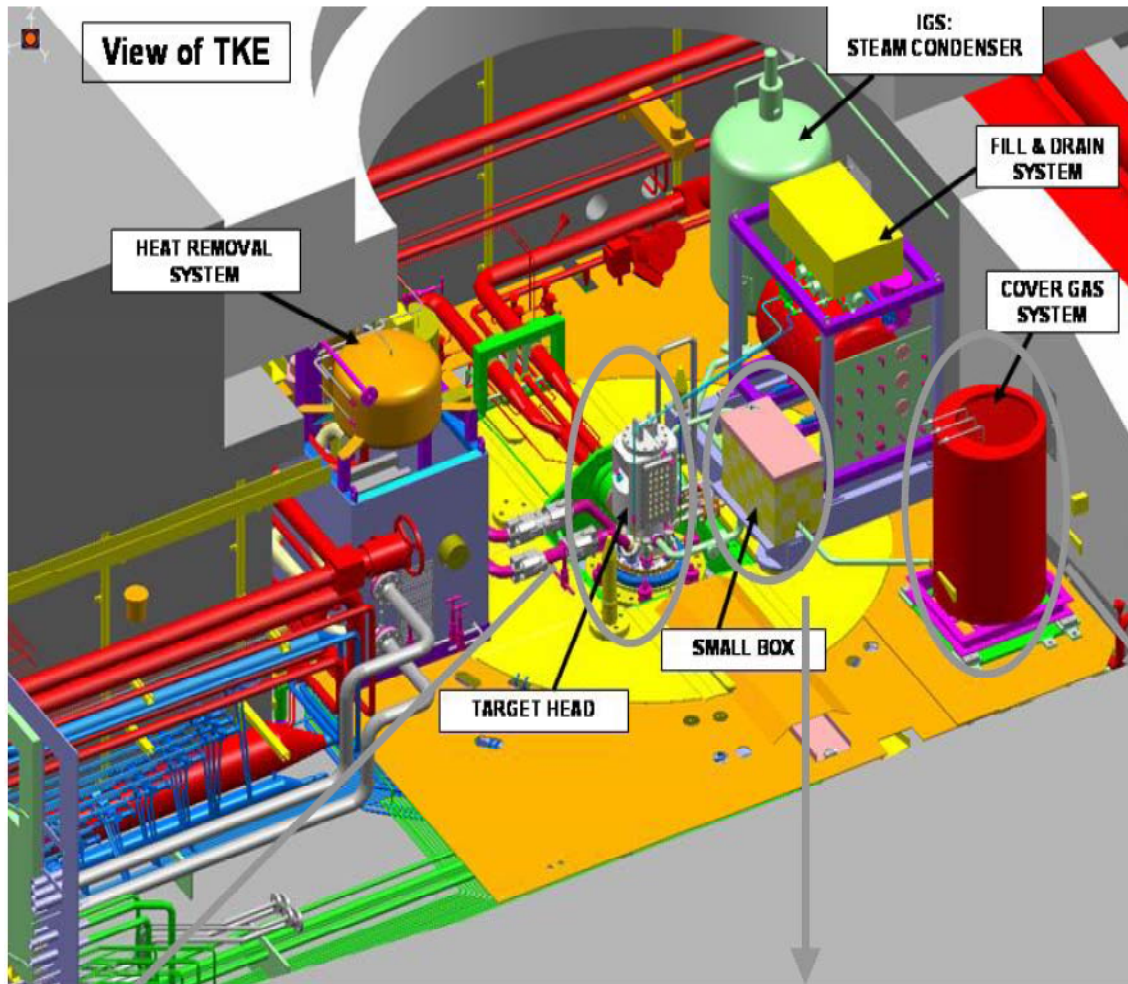
Precursor	Half-life, s	$a_i$ (%)
<sup>87</sup> Br	55.60	4.3
<sup>88</sup> Br	16.29	3.3
<sup>17</sup> N	4.173	92.4



DN densities in agreement with solid target experiment

# Gas measurement: setup

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## Gas operation goal:

- handling and control radioactive gases
- handling and control the pressure
- analysis of the gas production

~13 liters shielded decay tank and small sampling unit

LBE expansion volume (~2 liters)

Pb shielded box containing valves for operation and pressure transducers

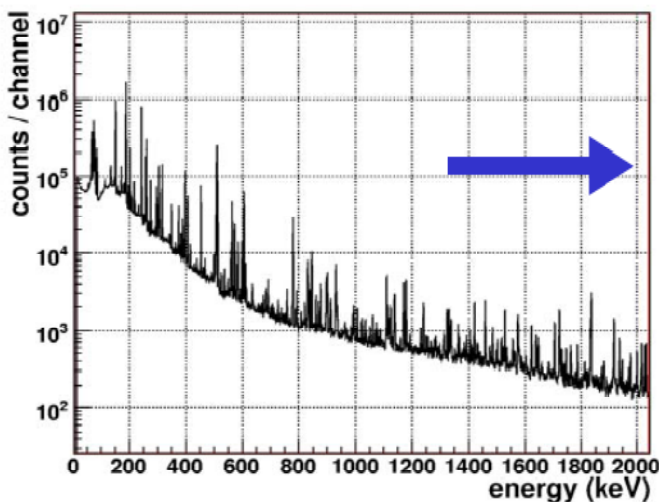
# Gas measurement: procedure

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A gas sampling is performed two days after the beginning of irradiation (1mA·h of accumulated charge) in order to access short half-life isotopes.



Au → Hg decay ←

Isotope	$t_{1/2}$	Activity (Bq)	Isotope	$t_{1/2}$	Activity (Bq)
$^{41}\text{Ar}$	1.8 h	$3.2 \cdot 10^2$	$^{192}\text{Au}$	5.0 h	$3.4 \cdot 10^4$
$^{79}\text{Kr}$	34.9 h	$4.5 \cdot 10^4$	$^{193}\text{Au}$	17.7 h	$1.2 \cdot 10^4$
$^{85\text{m}}\text{Kr}$	4.5 h	$1.5 \cdot 10^5$	$^{195}\text{Au}$	186 d	$1.2 \cdot 10^2$
$^{88}\text{Kr}$	2.8 h	$2.7 \cdot 10^4$	$^{192}\text{Hg}$	4.9 h	$1.8 \cdot 10^4$
$^{122}\text{Xe}$	20.1 h	$1.4 \cdot 10^4$	$^{193\text{m}}\text{Hg}$	11.1 h	$1.2 \cdot 10^4$
$^{125}\text{Xe}$	16.9 h	$9.5 \cdot 10^4$	$^{195\text{m}}\text{Hg}$	41.6 h	$2.9 \cdot 10^3$
$^{127}\text{Xe}$	36.4 d	$5.0 \cdot 10^3$	$^{197}\text{Hg}$	64.1 h	$2.1 \cdot 10^4$
$^{129\text{m}}\text{Xe}$	8.9 d	$7.6 \cdot 10^3$	$^{197\text{m}}\text{Hg}$	23.8 h	$3.6 \cdot 10^3$
$^{135}\text{Xe}$	9.1 h	$5.7 \cdot 10^2$	$^{203}\text{Hg}$	46.6 d	$5.0 \cdot 10^1$

Two different gas sample have been measured and have revealed :

$$A_{\text{nb-s1}} \approx 1.4 A_{\text{nb-s2}}$$

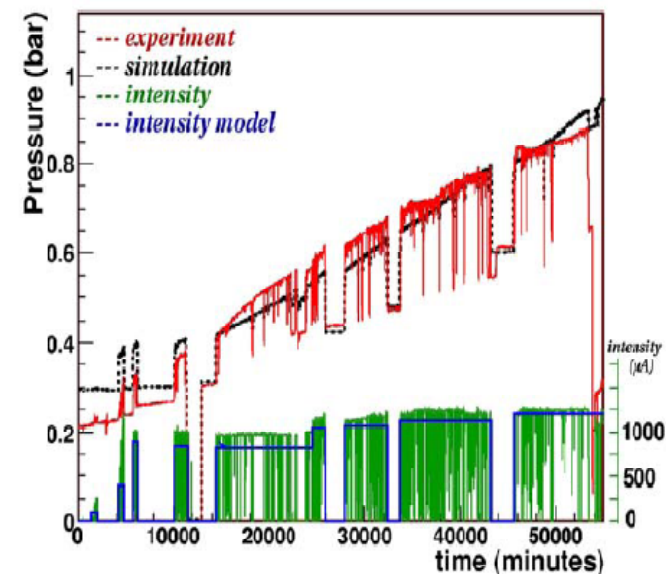
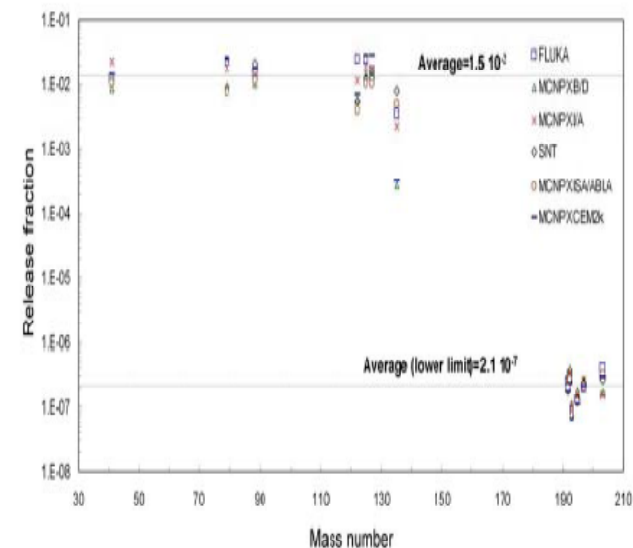
$$A_{\text{Hg-s2}} = A_{\text{Au-s2}} = 0 \rightarrow$$

→ indication on uncertainty (~40%)

doubt in mercury behavior...

# Gas measurement: results

- Successful measurements of radionuclides at different irradiation times.
- Estimation of the release fractions at the beginning of irradiation and towards the end.
- Good agreement with calculations. Mass distributions allow selecting best models.
- Agreement of results with estimations on release of Hg and Po from the design phase (but measurements not precise).
- No mass measurement of stable isotopes, but indirect information from pressure measurements (in fair agreement with calculations).



# Gas measurement: issues

Measurement has revealed traces of  $^{208}\text{Po}$  which is supposed to be weakly released. Further investigations (Y. Tall et al., IS419 experiment, 2008) show that Po is not expected to be directly released at MEGAPIE operated conditions.

→ It is reasonable to assume that measured  $^{208}\text{Po}$  comes from  $^{208}\text{At}$  released and decay.

→ experimental and simulated data give a release factor for At close to  $10^{-6}$ .

Measured amounts of mercury in the expansion tank have a high uncertainty. This may be due to possible isotopes sticking on the CGS pipes and walls.

→ The precise Hg during sampling procedure should be understood.

→ Analysis of the cold trap will be done during the Post Irradiation Experiment.

Leaks of radioactive Xe isotopes (~1% of the total) were detected in the insulation gas system and a high amount of migrated tritium were detected in Dyphil oil ( $10^6$  time higher than expected value calculated from reaction induced by spallation neutron only).

→ Radioactive gas leaks should be inspected in order to be prevented.



# Gas measurement: recommendations

- Feedback to operation issues:
  - Leak tightness is difficult to achieve
  - Radiation damage in pressure sensors to be investigated
  - Gas sampling operation to be improved
- Feedback to scientific/safety issues:
  - Need for reliable mass spectroscopy measurements, especially for H and He isotopes
  - Understanding the behaviour of Hg in the CGS

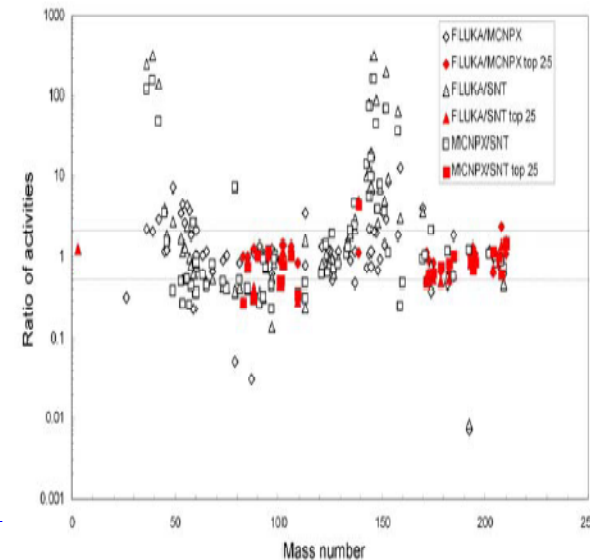
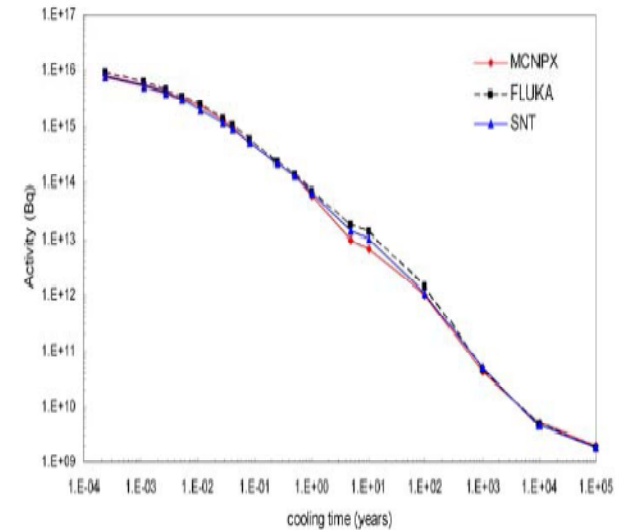
# Target activation

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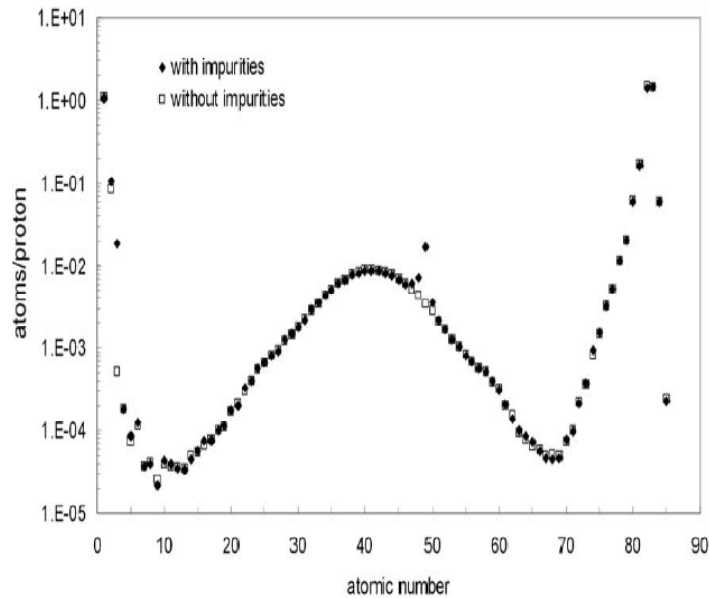


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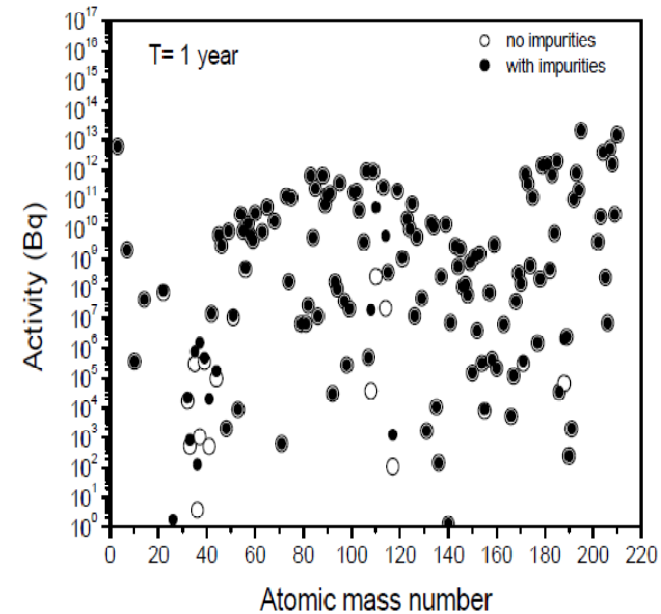
- Activation of LBE and target structure.
- Large effort with different code systems:
  - MCNPX/CINDER'90
    - Different spallation/evaporation models
  - MCNPX/FISPACT
  - MCNPX/SNT
  - FLUKA/ORIHET
- Origin of nuclides produced (contributions from protons and neutrons).
- Effect of impurities.



# Target activation: effect of impurities



The only significant differences are for lithium ( $Z=3$ ), cadmium and indium ( $Z=48, 49$ ). The isotopes showing larger production rate in the LBE with impurities are  ${}^7\text{Li}$  (from  $(n, \alpha)$  reaction on the  ${}^{10}\text{B}$  impurity),  ${}^{116}\text{In}$  (capture in  ${}^{115}\text{In}$ ) and  ${}^{114}\text{Cd}$  (inelastic reactions in In and Sn impurities).



After one year of cooling, there are only a few isotopes with activity greater than  $10^9$  Bq, which show a significantly higher activity with the impurities:  ${}^{114}\text{In}$  and  ${}^{114\text{m}}\text{In}$  (factor 270 increase) and  ${}^{110\text{m}}\text{Ag}$  (factor 210 increase). These isotopes come from thermal neutron capture in the In and Ag impurities.

# Conclusions

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- Neutronic performance of an ADS target with window has been studied in great detail
- System of „tools“ (measurement techniques, Monte Carlo strategies) developed, essential for the design and construction of future ADS
- Importance of good calculation tools and of proper use of them
  - Materials definition
  - Beam profile definition
  - Geometry definition
  - Physics models and cross section libraries
- Importance of neutronic measurements inside the target, where the fast component is dominant
- Importance of gas measurements as gas production is one of the key safety issues in an ADS
  - Improve and control leak tightness
- Delayed neutrons must be taken into account

