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MEGAPIE final TRM, Bregenz (A) 23.-24.10.2014

MEGAPIE – The Impact of Radiochemical Investigations

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GFA

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NES

Outline

Motivation   Why do you want to do this that way?

- Requirements from BAG and ENSI
- Theoretical predictions versus experiments
- Model studies of Po behaviour in LBE
- Results from the ISOLDE target

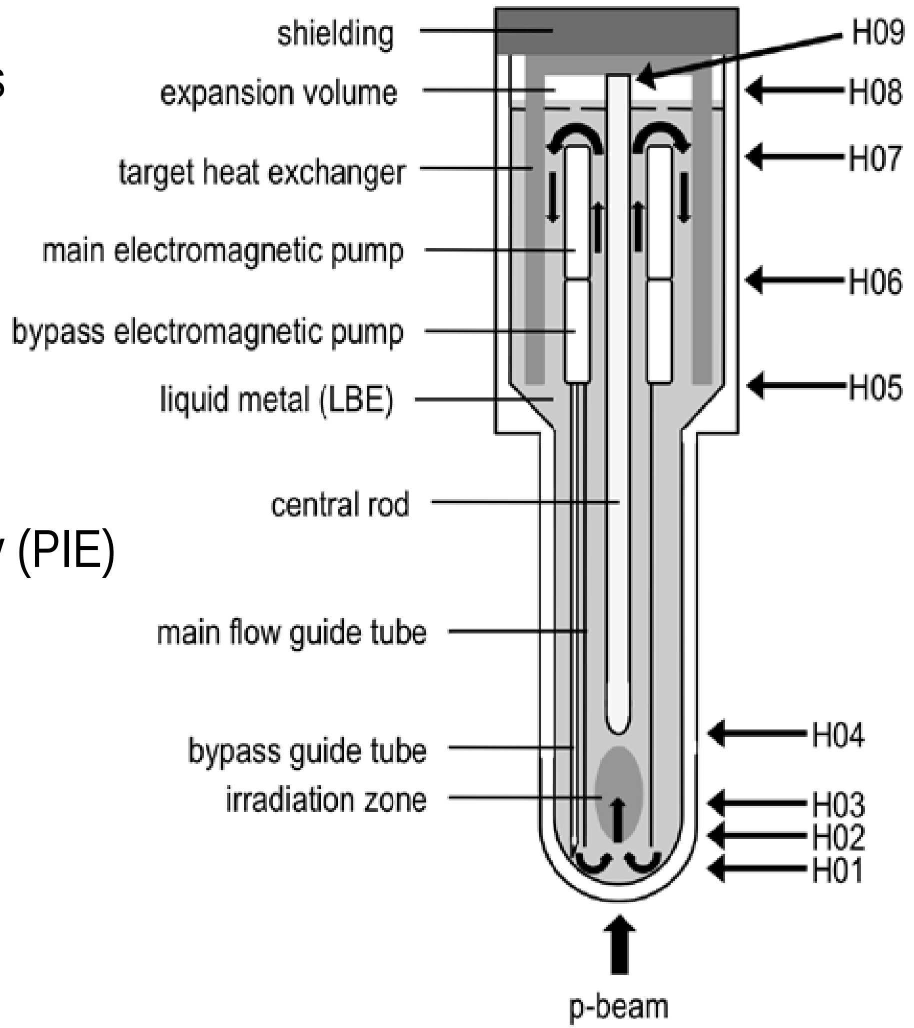
Radiochemical investigations

- Release studies
- Determination of the radionuclide inventory (PIE)

MEGAPIE sample taking

PIE results

Impact



BAG and ENSI requirements

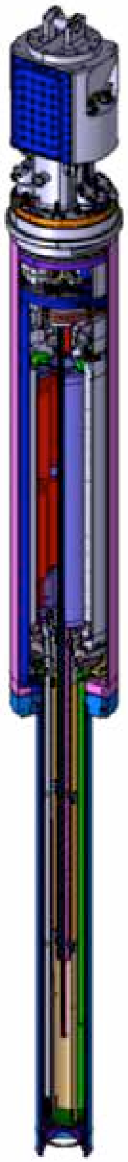
MEGAPIE is:
Joint initiative by six European research institutions + Japan to design, build, operate and explore a liquid metal LBE spallation target for 1 MW beam power

Main professions involved: physists and technicians

Radionuclides are produced in the target
Radiochemistry takes place

BAG: estimation of the radionuclide inventory (predictions)
investigation of the release behaviour of hazardous radionuclides (experiments) licensing and operation

ENSI: experimental determination of the radionuclide inventory (top ten) disposal



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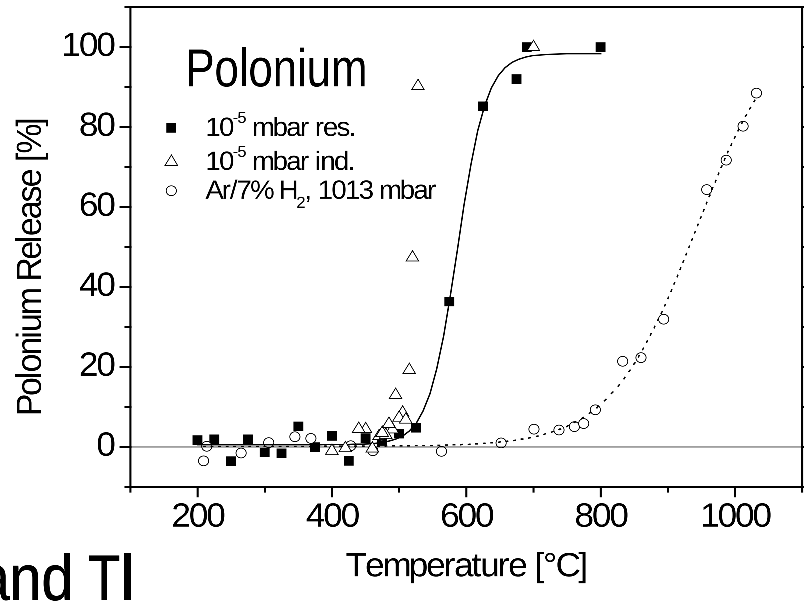
Licensing and operation (BAG)

Theoretical predictions of the radionuclide inventory:

Codes: FLUKA; MCNPX, SNT

Pitcher, 2002; Foucher, 2002; Zanini, 2002, 2008

Release behaviour:



Release of Po from LBE under vacuum (»1´ 10^{-5} mbar) and Ar/7%-H_2 (ambient pressure); Neuhausen et.al.

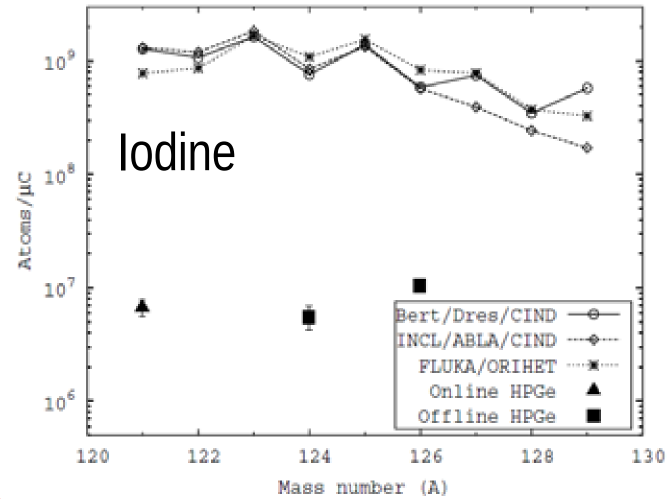
Model studies with Po, I, Hg and Tl

Neuhausen et.al. 2005

ISOLDE experiment – online release

Tall, Zanini et.al. 2005

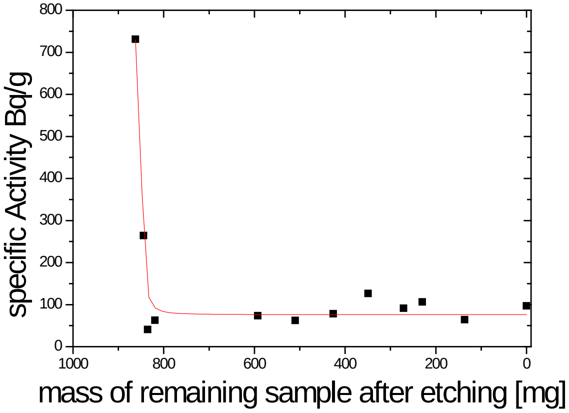
Y. Tall et.al.: Release of I isotopes from an irradiated LBE target at ISOLDE

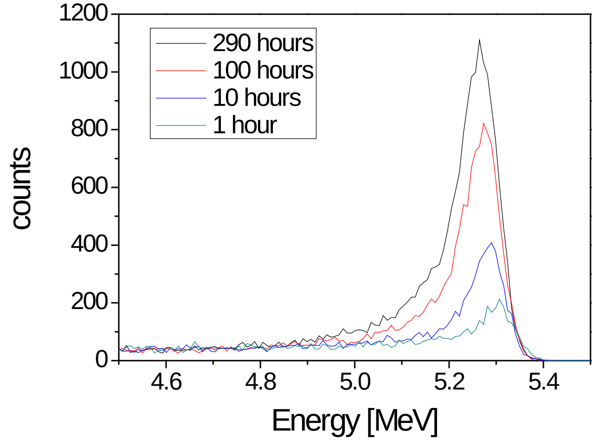


Result: low release up to 500 °C - MEGAPIE licensing possible

Disposal (ENSI) – preparative studies

Polonium migration and segregation in solidified LBE

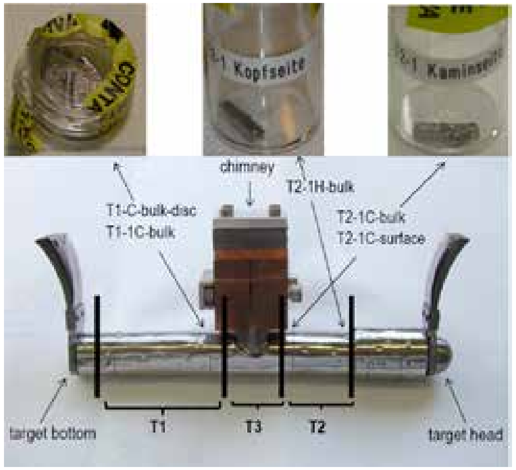


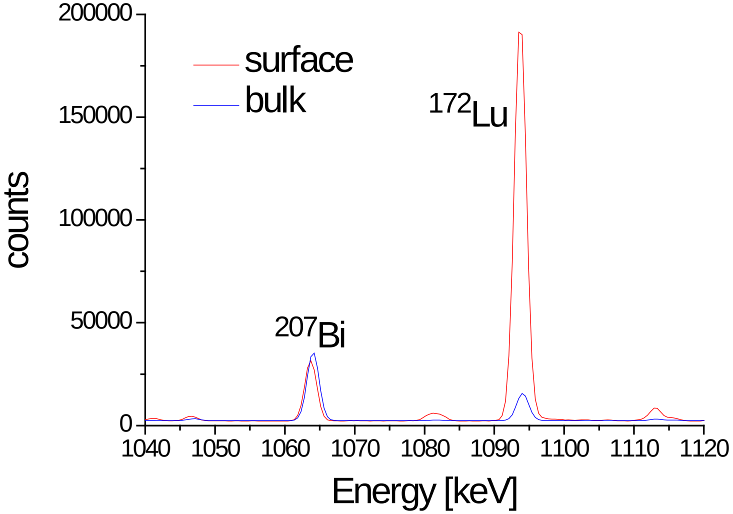


Result:

Enrichment of radionuclides on LBE surface

Analytics of the LBE from the ISOLDE experiment





Questions:

Which radionuclides enrich?

Where are they deposited?

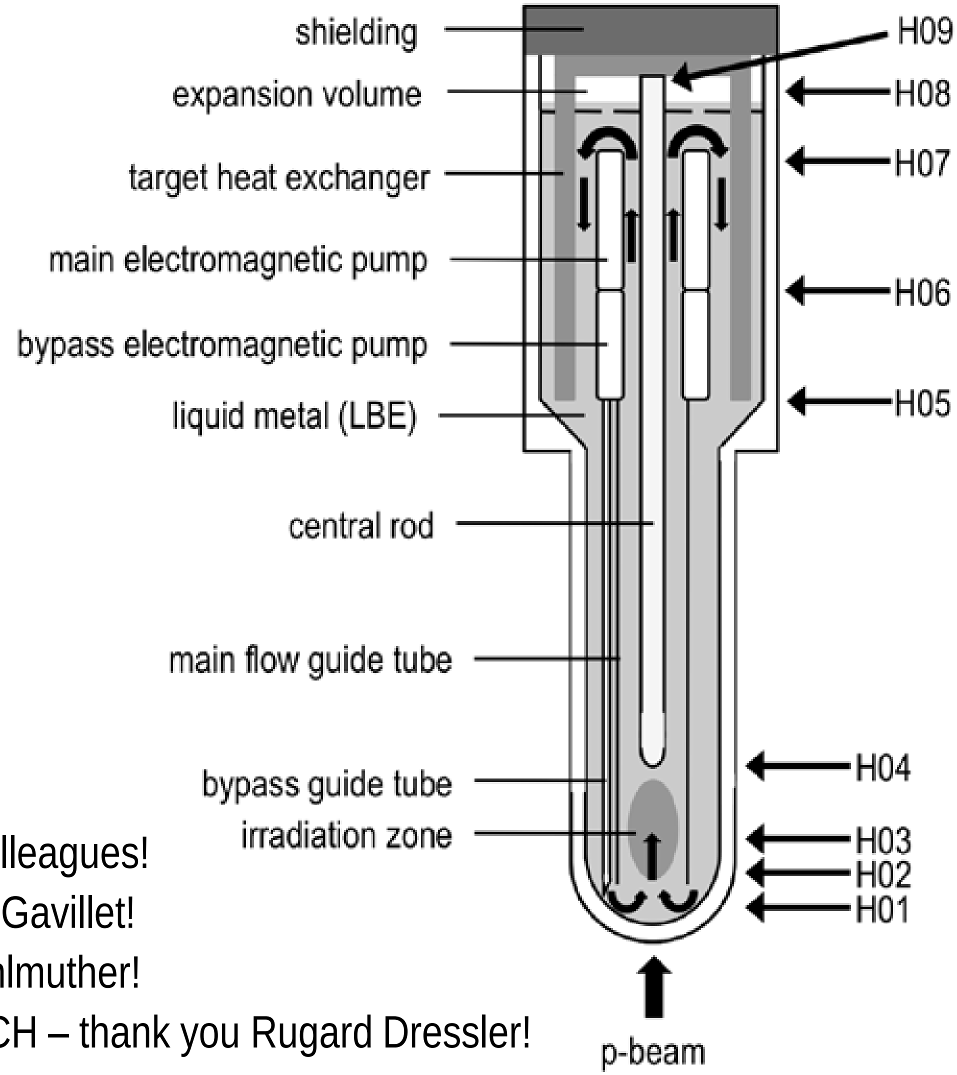
Consequence:

Homogenious distribution of radionuclides in LBE cannot be assumed

What did we need?

- More than 70 LBE samples (from nearly every cut)
  - Bulk
  - LBE-steel interface
  - LBE covergas interface

- Lab with permission for a-activity
- Measurement technique
- Lab equipment
- Man power



How did we get this?

Samples: Hotlab – thank you Viktor Boutellier and colleagues!
Lab: Hotlab – thank you Manuel Pouchon and Didier Gavillet!
Lab equipment: MEGAPIE – thank you Michael Wohlmuther!
Measurement technique: SNF project ASPIT and LCH – thank you Rugard Dressler!
Man power: EC funded projects GETMAT and ANDES
AMS: thank you Christof Vockenhuber from ETHZ!

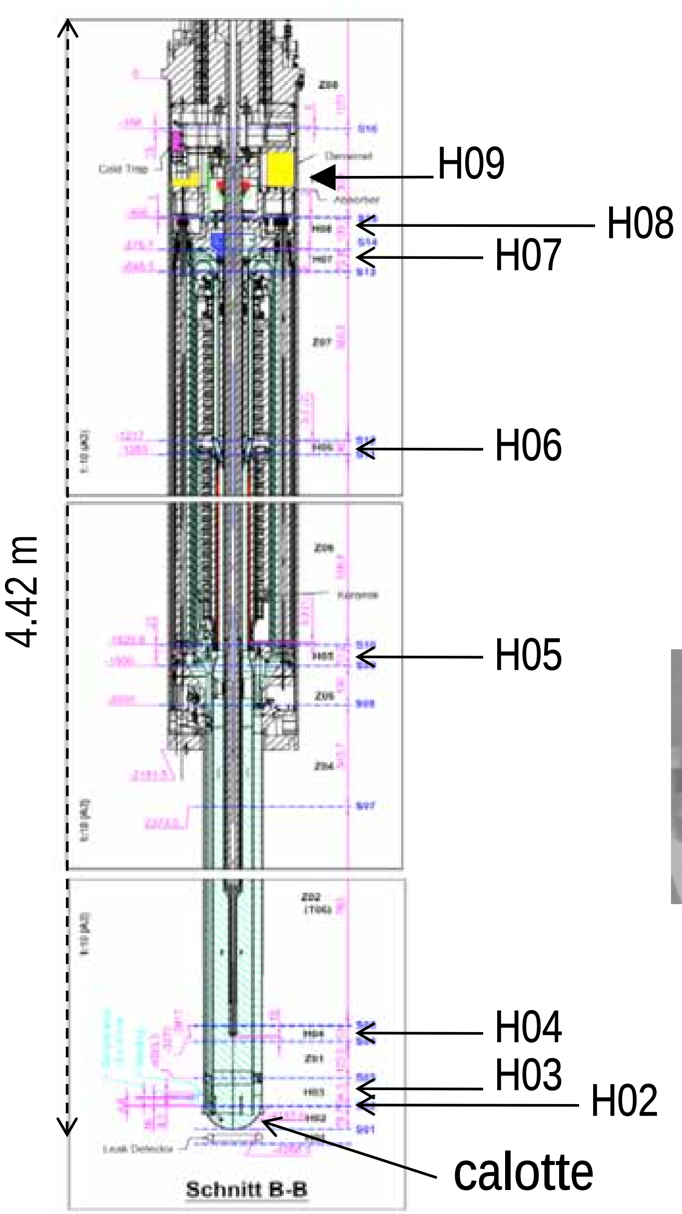
New HotLab for wet chemistry (Summer 2011)

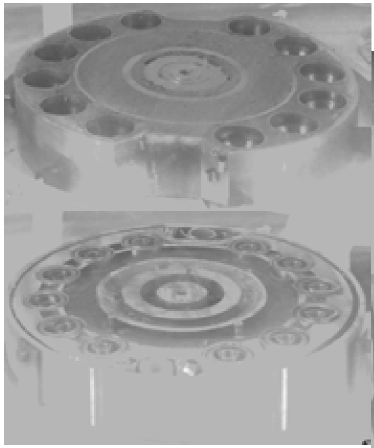
Still missing:

- Electricity
- Equipment
- Mesurement technique (a and g)

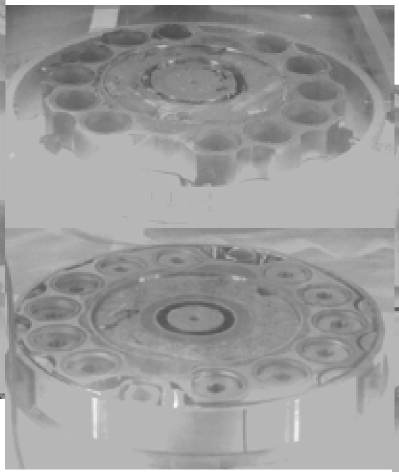
New HotLab for wet chemistry (October 2014)

Sample taking MEGAPIE

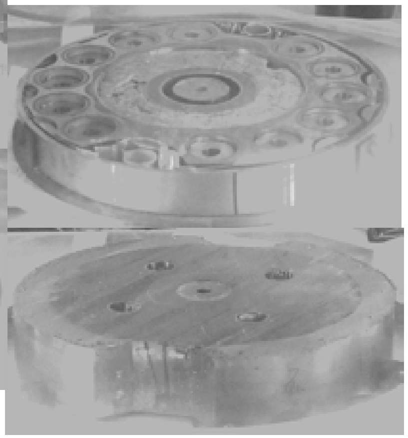




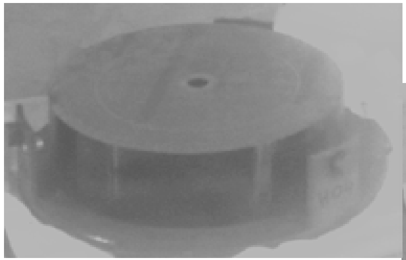
H07
(Ø 37.1cm x 7.28cm)



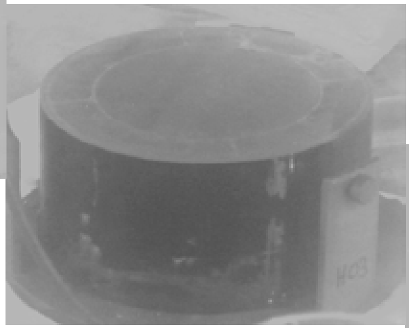
H06
(Ø 37.1cm x 4.8cm)



H05
(Ø 37.1cm x 7.02cm)



H04
(Ø 17cm x 5.3cm)



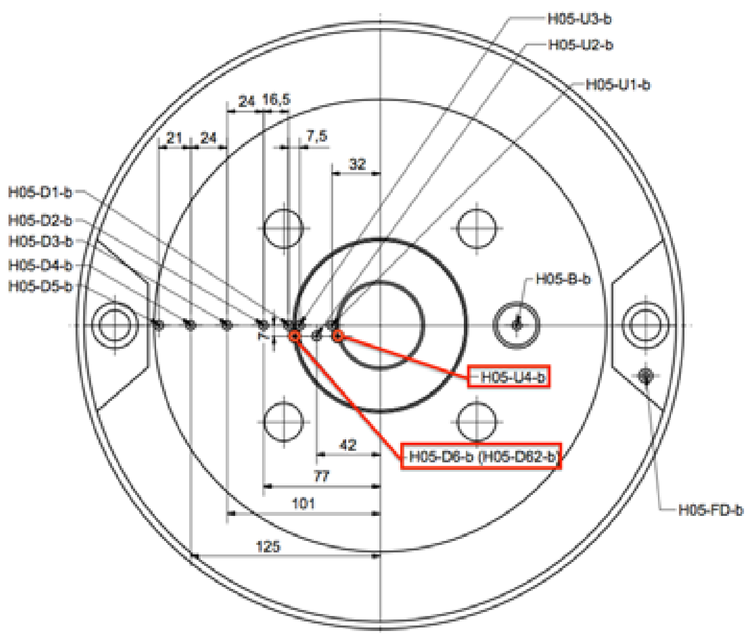
H03
(Ø 17cm x 9.43cm)

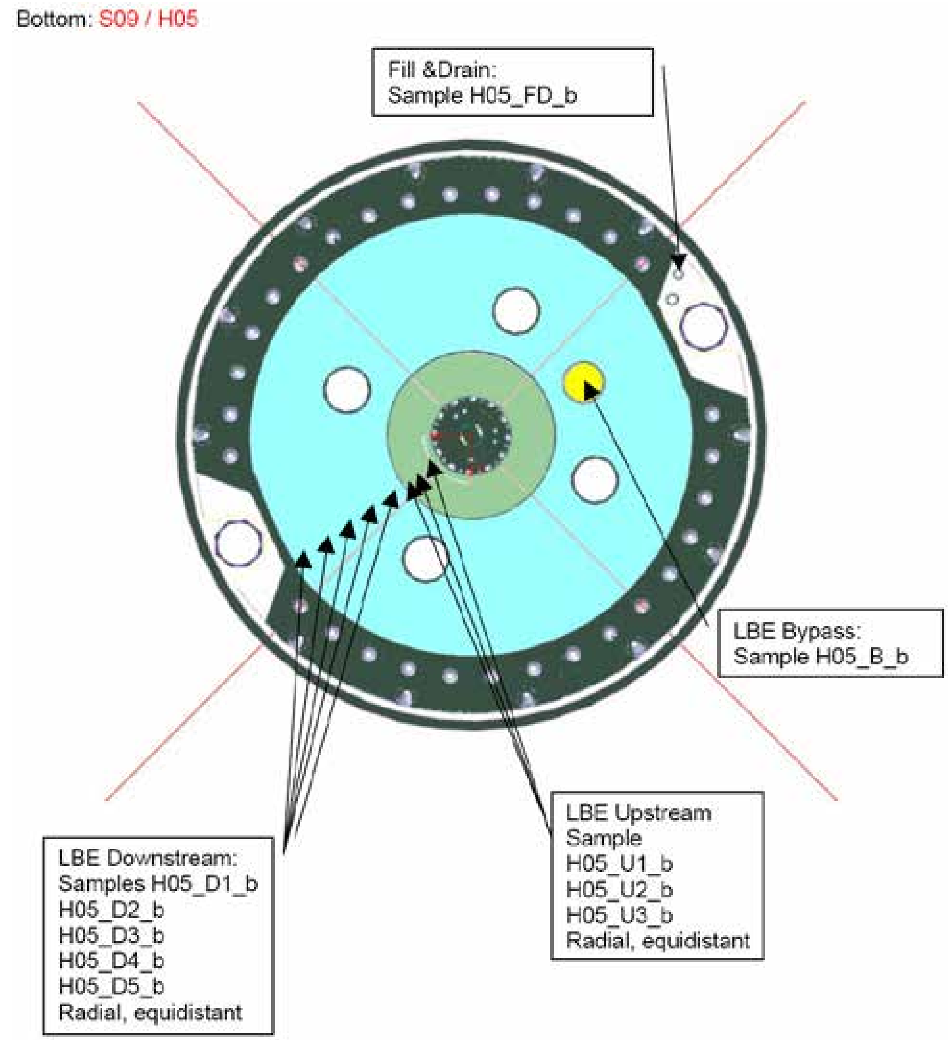
H02
(Ø 17cm x 7.87cm)

Example : Cut H05

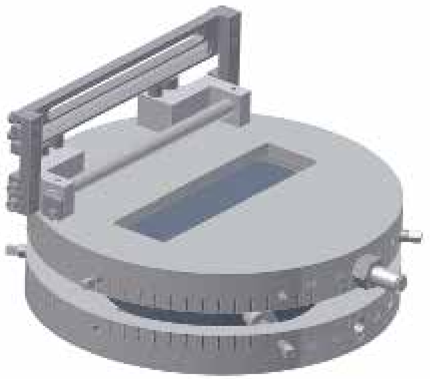
LBE samples from H05 bottom (10 samples) and top surface (5 samples)

- LBE samples were taken from upstream and downstream area as well as the bypass tube.
- Fill and Drain tubes were empty in H05.
- Radial samples have been taken, three in the upstream (inner) and five in the downstream (outer) part to enable the study of inhomogeneities that may occur.
- Additional samples containing the interface of LBE and the steel wall to obtain information on depositions on the wall (e.g. H05-U4-b, H05-D6-b, H05-D62-b).





Tools + Sampling procedure

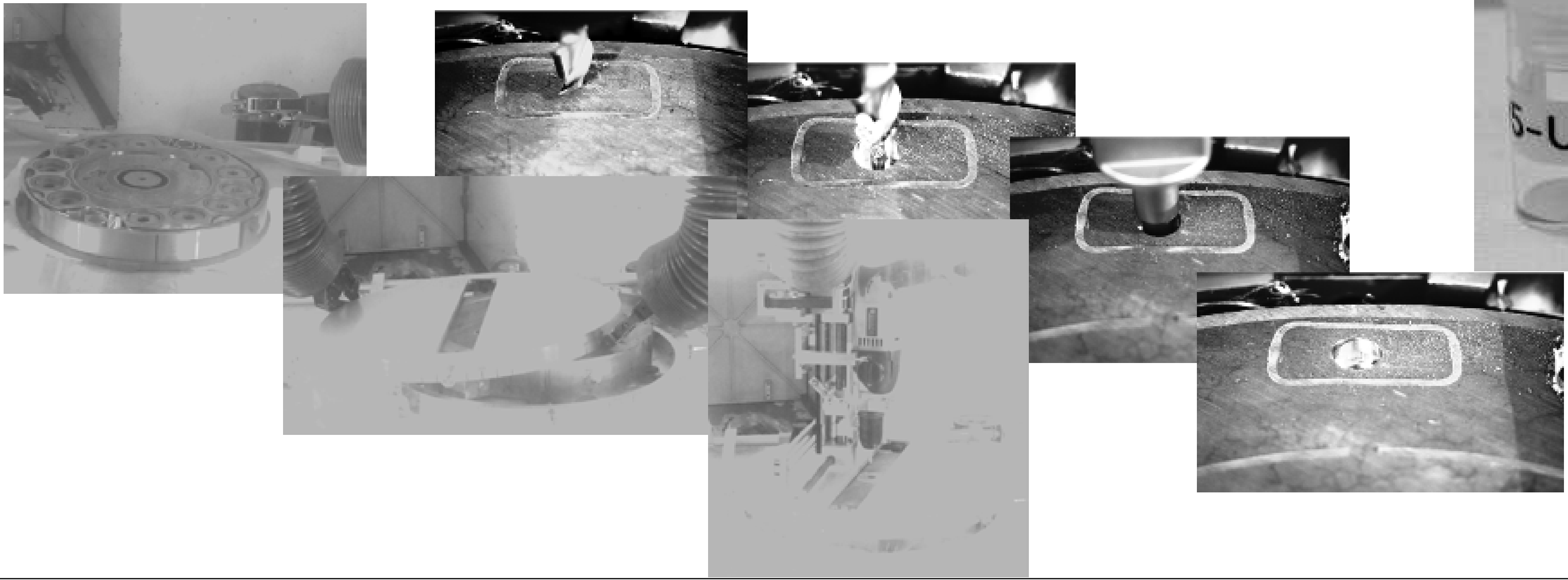


Turning device

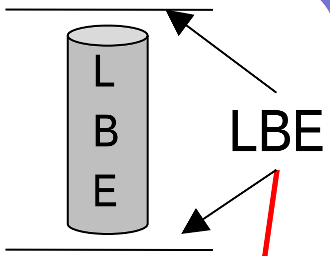
Core drilling tool

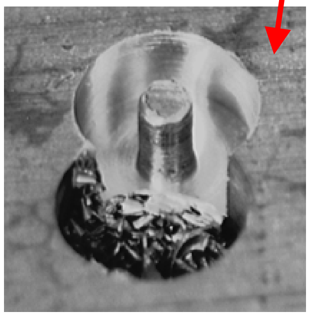
Sample breaking device

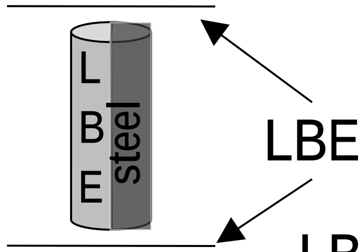
Cold test (above) + Sample taking in Hotcell (below)



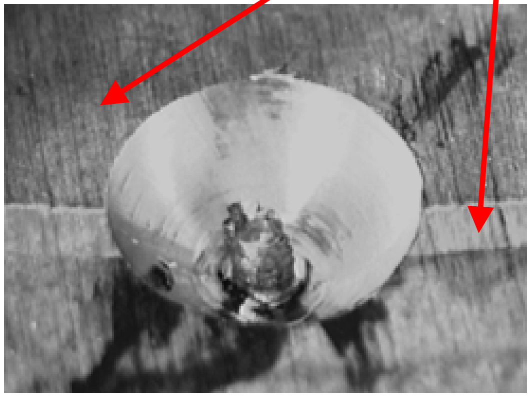
Sample description 1

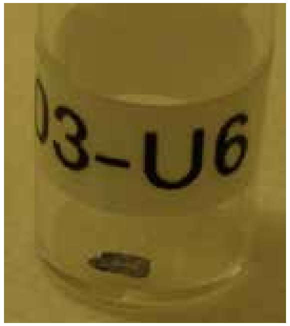


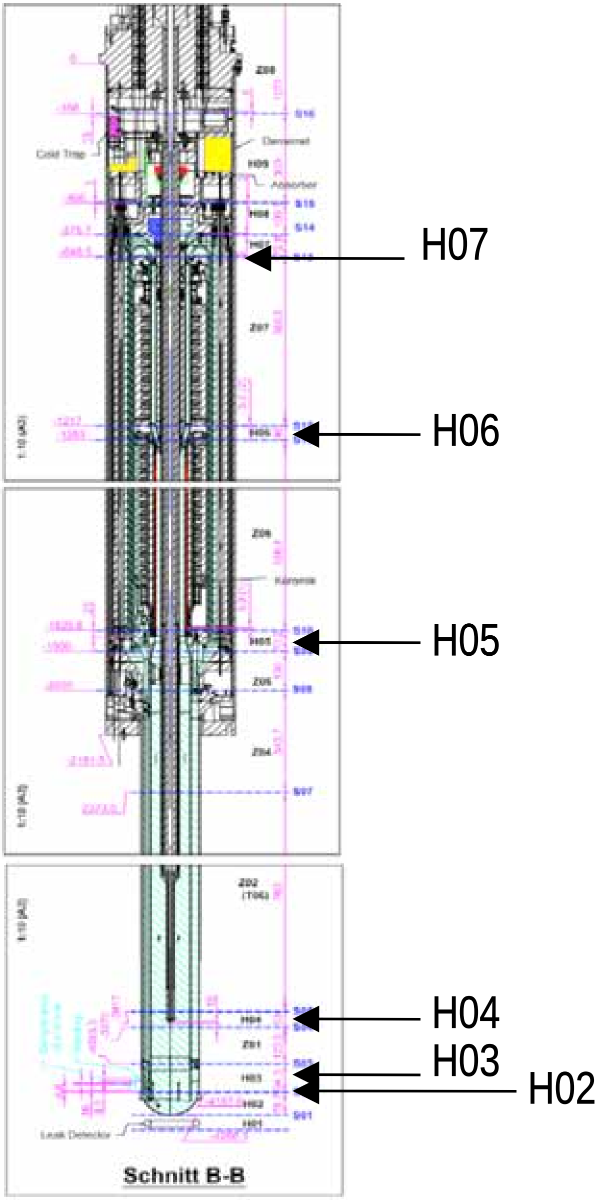




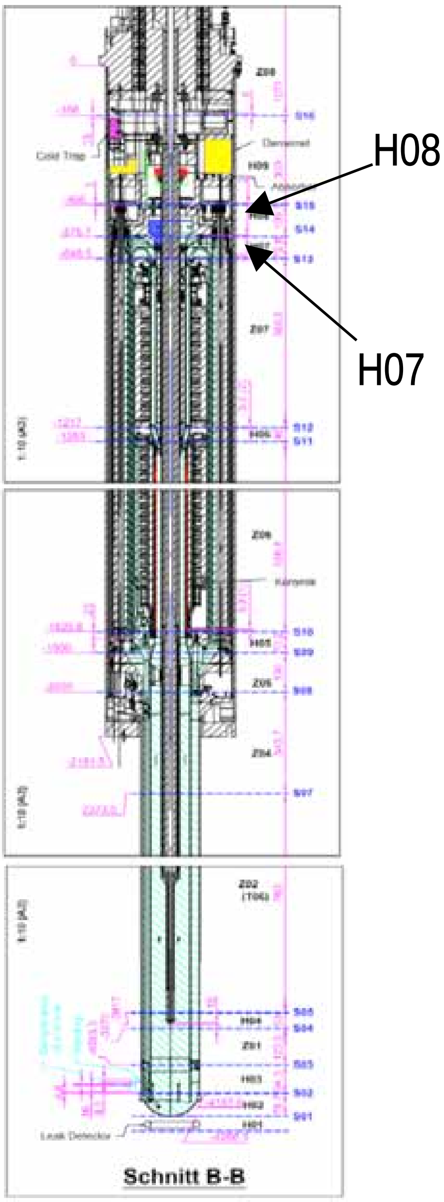
LBE steel

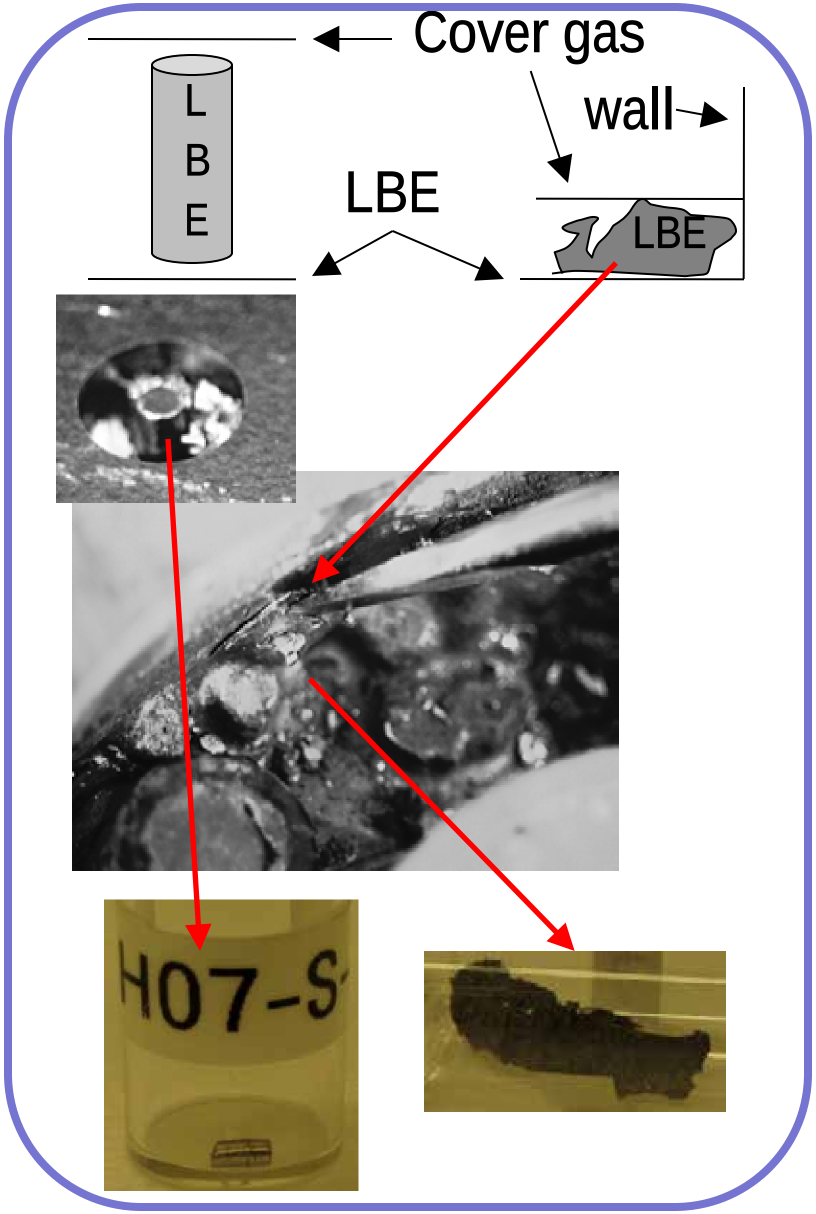


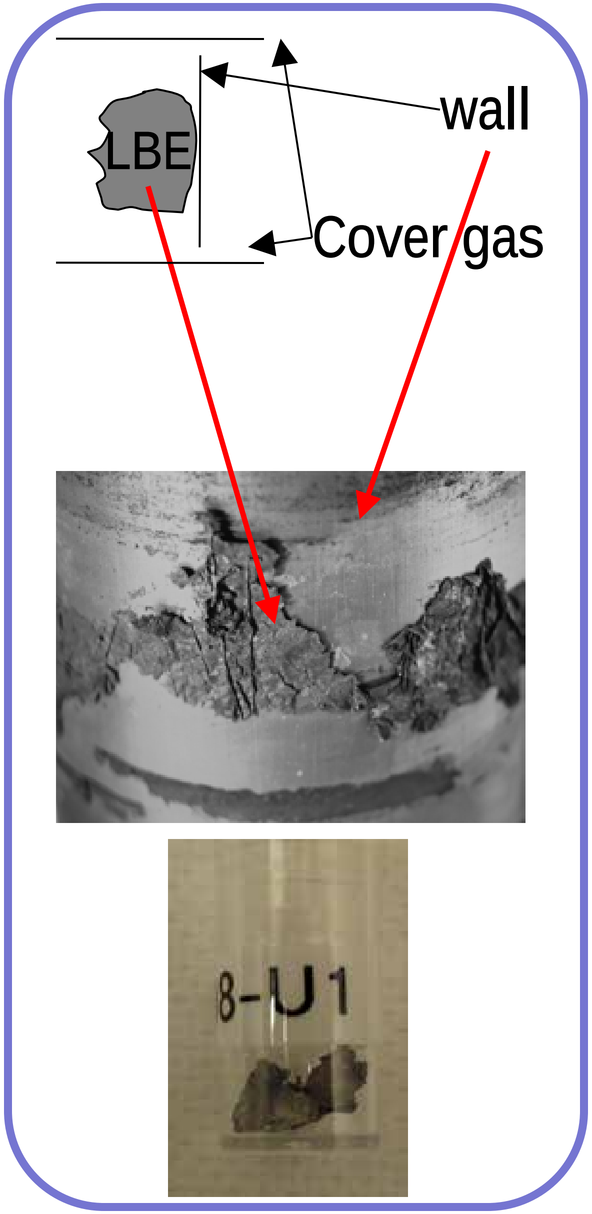




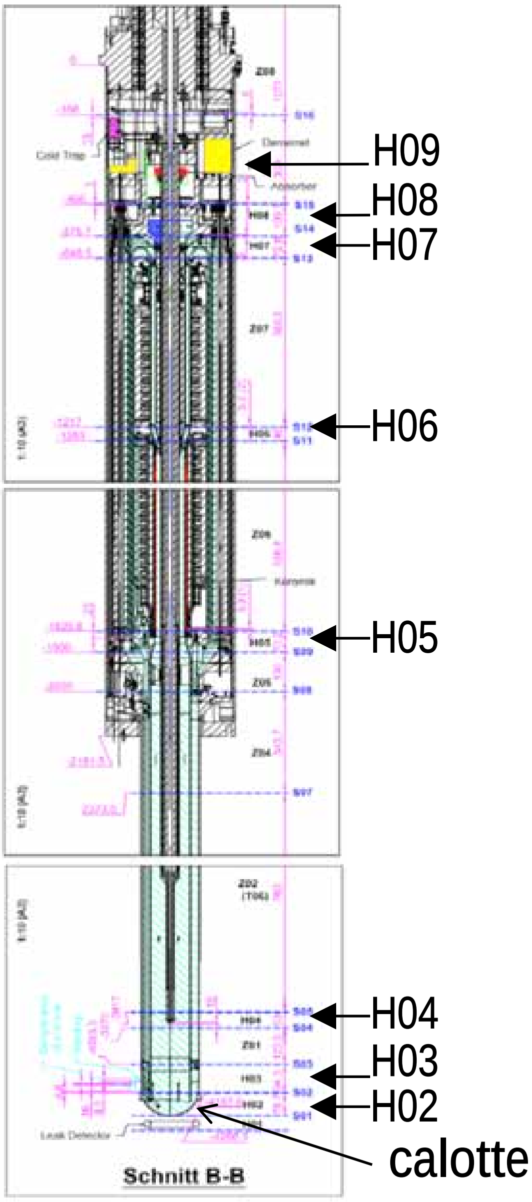
Sample description 2







Summary on obtained samples



Core drillings from H02 – H07

Size: cylindrical, nominally Ø 2mm x 5mm and Ø 1,5mm x 5mm

Mass: 50 – 100 mg

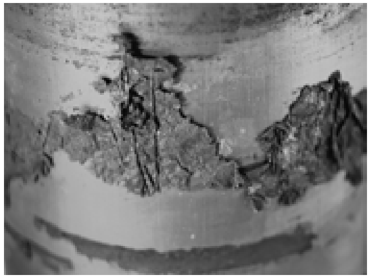
Dose rates of 5 – 10 mSv/h at 10 cm distance

H07 and H08 samples:

Mass: 200 – 900 mg

Material sticking to the wall

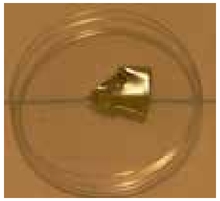
Dose rates of 7 – 150 mSv/h at 10 cm distance



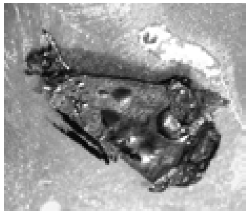
Totally 76 samples, 33 Samples from the LBE/steel and LBE/cover gas interface.
43 samples represent bulk LBE.

Samples from Expansion volume:

Gas absorbers (Ag and Pd foils separated)



Sample from calotte:



Radiochemical procedures

LBE Sample

g-Analysis

HPGe-Detector

Dissolving

7M HNO_3

Carrier I_2 & Cl^-

Vessel with N_2H_2

Separation

Precipitation,

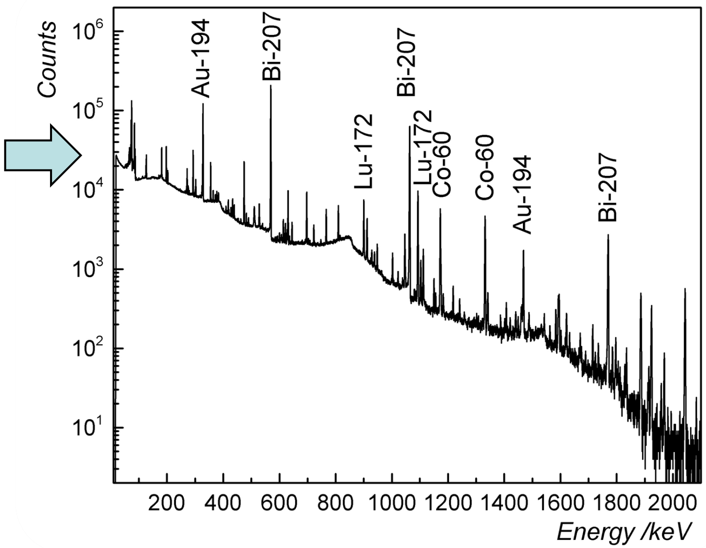
Extraction,

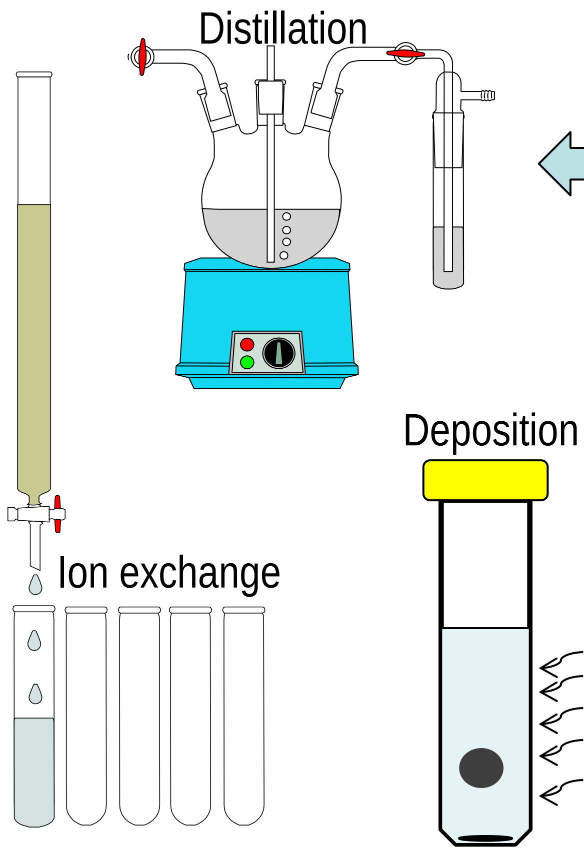
Chromatography,

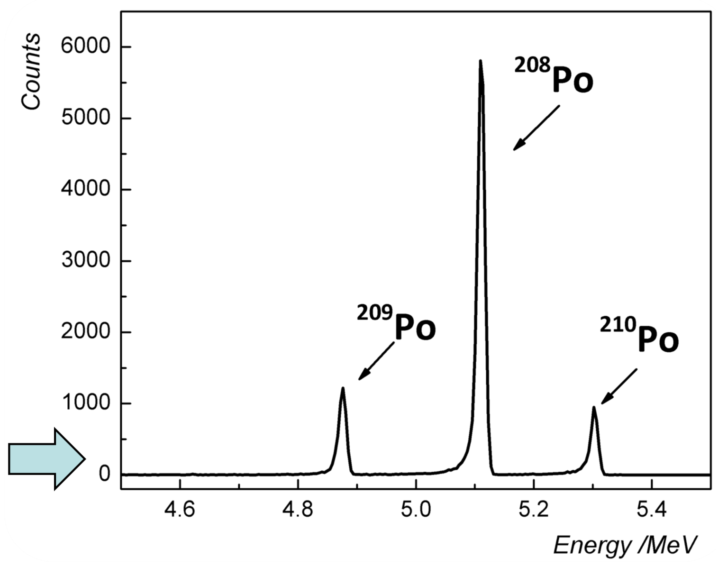
Deposition

Measurement

a-Counting, AMS

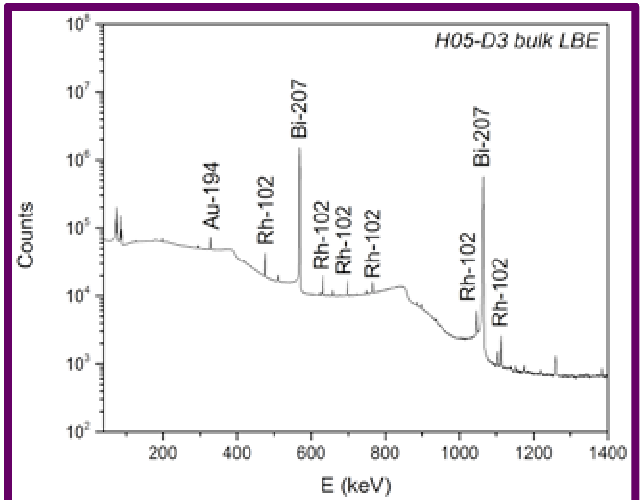






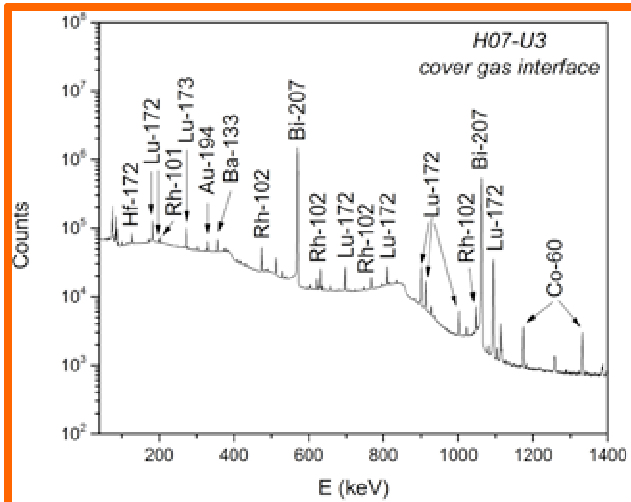
Results

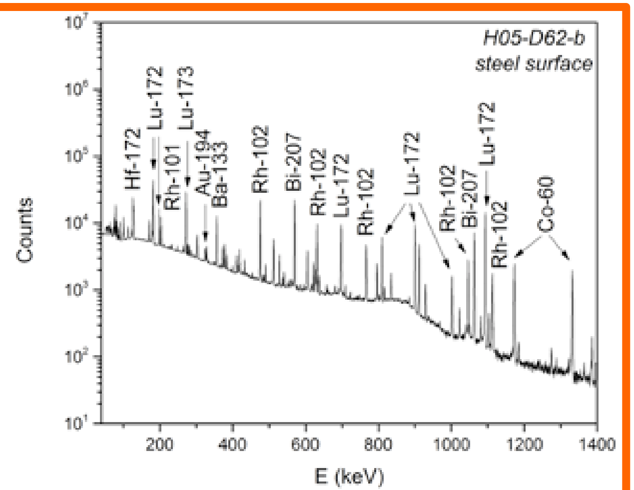
Qualitative evaluation of g-measurements



Bulk:

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





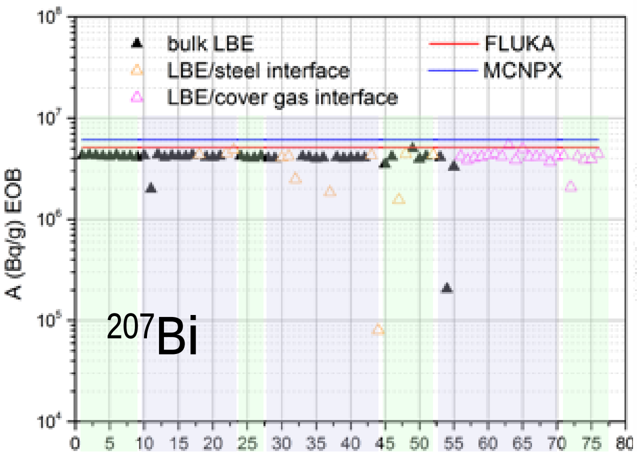
Surfaces:

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

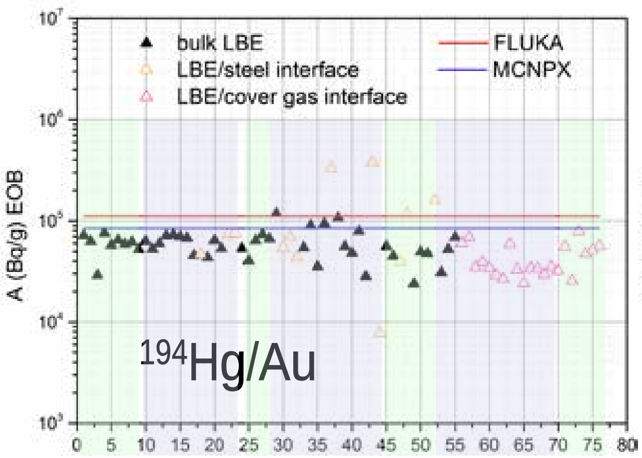
Results:

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

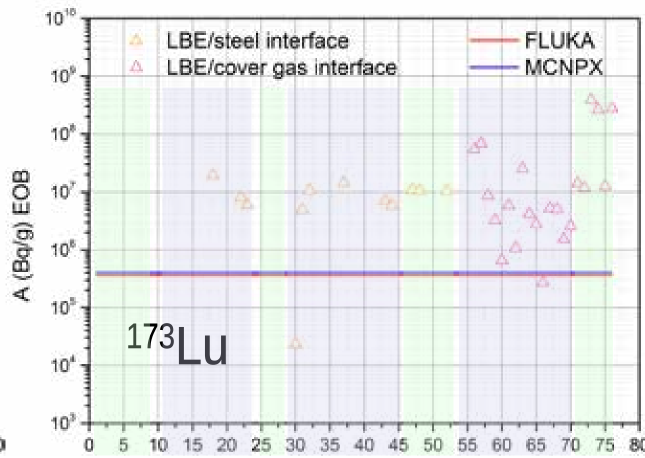
Distribution of ^{207}Bi, ^{194}Hg/Au and ^{173}Lu



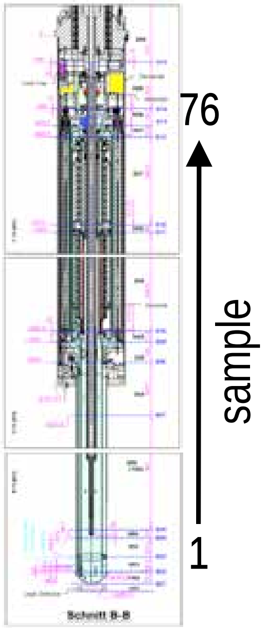
sample



sample



sample



	^{207}Bi (Bq/g)	^{194}Hg/Au (Bq/g)	^{173}Lu (Bq/g)
bulk LBE		2.36×10^4 – 1.17×10^5	-
LBE/steel interface	$3.49 \pm 0.09 \times 10^6$	7.69×10^3 – 3.76×10^5	1.34×10^3 – 1.08×10^6
LBE/cover gas interface		2.53×10^4 – 7.80×10^4	1.51×10^4 – 2.17×10^7

Corresponds to results from ISOLDE

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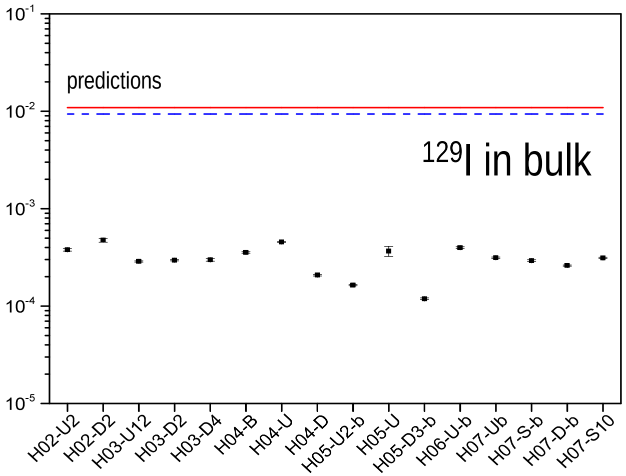
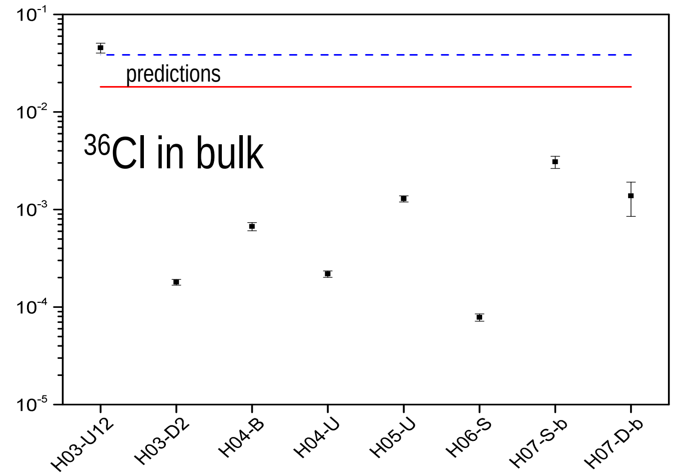
Distribution of ^{129}I and ^{36}Cl





Total ^{129}I activity, [1]free surface corresponds to approx. 290cm^2; [2]steel surfaces relates to 16m^2; [3]LBE surfaces relates to 16m^2.

	measured total [kBq]	predicted averaged [kBq]	% of predicted
bulk	0.26		3
free surface[1]	0.009		<1
steel surface[2]	6.38	8.56	75
LBE surface[3]	9.03		105
absorber	0.0004		<1

Total ^{36}Cl activity , [1]free surface corresponds to approx. 290cm^2; [2]steel surfaces relates to 16m^2; [3]LBE surfaces relates to 16m^2.

	measured total [kBq]	predicted averaged [kBq]	% of predicted
bulk	0.833		3
free surface[1]	0.04		<1
steel surface[2]	6.13	23.9	26
LBE surface[3]	0.29		1
absorber	0.01		<1

Conclusions:

- Iodine depletion in bulk
- ~ 180% of predicted ^{129}I found in target
- no evaporation (according to release studies)

Conclusions:

- Chlorine depletion in bulk
- ~ 30% of predicted ^{36}Cl found in target
- reasons unknown

Distribution of lanthanides in MEGAPIE

Total ^{148}Gd activity; [1]free surface corresponds to approx. 300cm^2; [2]steel surfaces relates to 16m^2; [3]LBE surfaces relates to 16m^2.

	measured total [MBq]	predicted [MBq]	% of predicted amount
free surface[1]	0.02		<1
steel surface[2]	20.7	153	14
LBE surface[3]	48.6		32

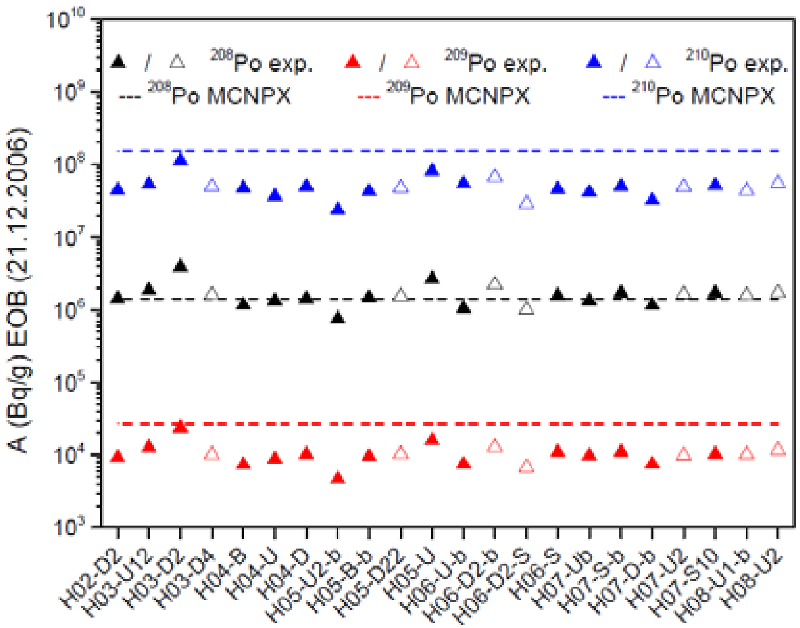
Total ^{173}Lu activity; [1]free surface corresponds to approx. 300cm^2; [2]steel surfaces relates to 16m^2; [3]LBE surfaces relates to 16m^2.

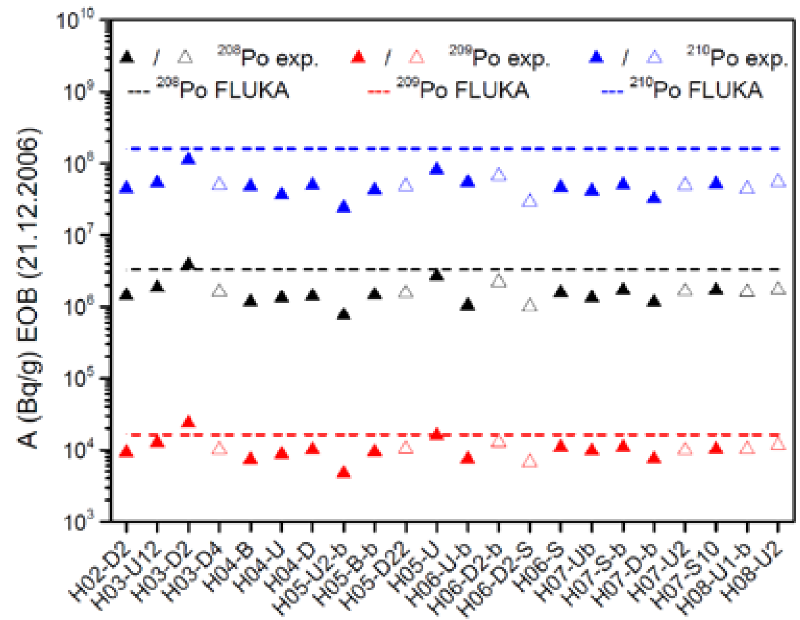
	measured total [GBq]	predicted averaged [GBq]	% of predicted amount
bulk	22.4		7
free surface[1]	0.1		<1
steel surface[2]	186	325	57
LBE surface[3]	323		99

Conclusions:

- Depletion of lanthanides in bulk (according to ISOLDE experiment)
- ~ 50-160% of predicted lanthanides found in target
- no evaporation
- Accumulation of a-emitters on walls of the structure material

Distribution of $^{208-210}$Po





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

^{209}Bi (*p*/*n*,g) ^{210}Po

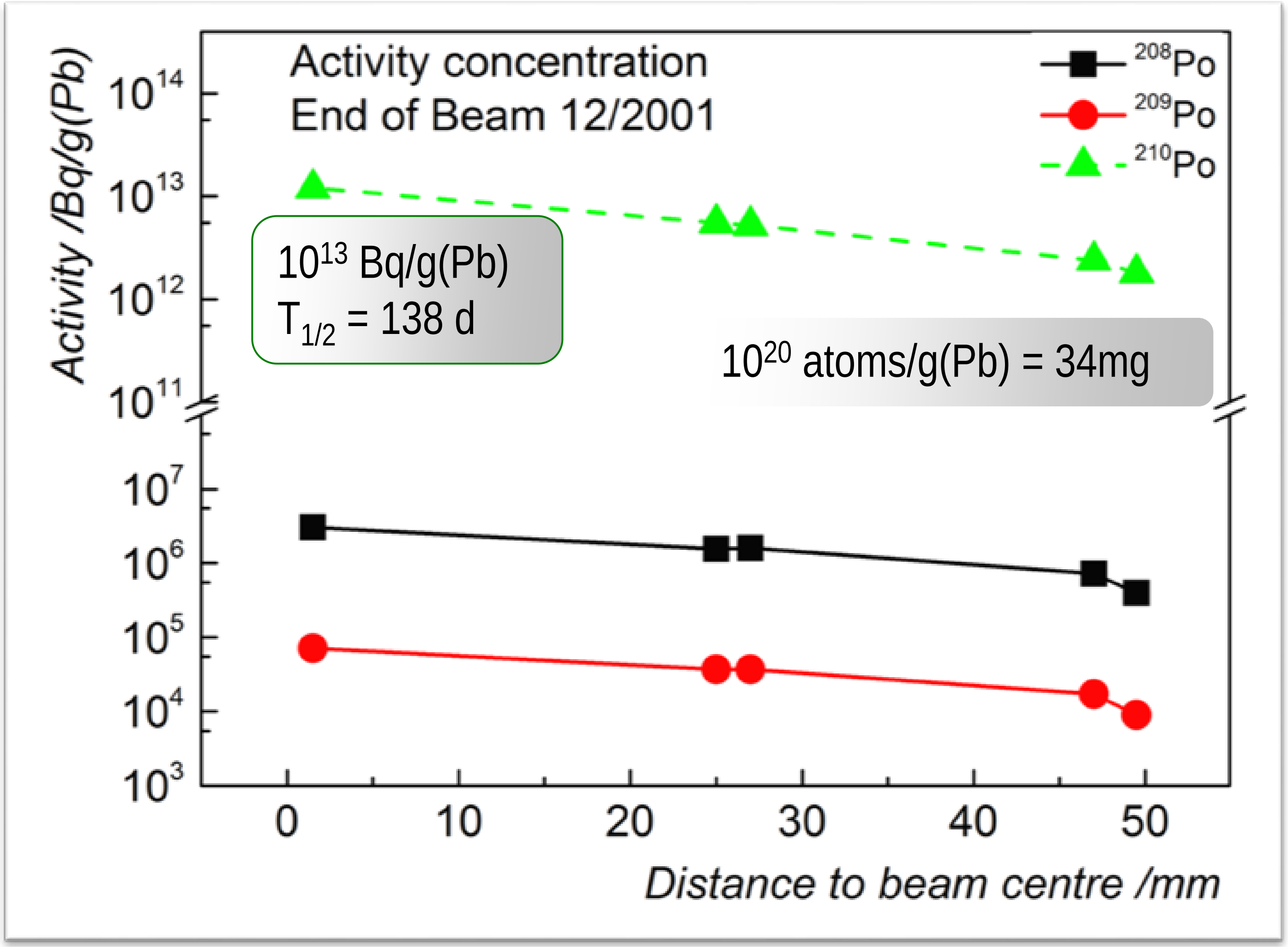
^{209}Bi (*p*,*n*) ^{209}Po

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

Conclusions: Po is mainly homogeneously distributed (not in agreement with previous findings)
(only very thin Po layer on top of surfaces)
in agreement with theoretical predictions

Po determination in SINQ lead target 4

Activity concentration at EoB (12/2001):



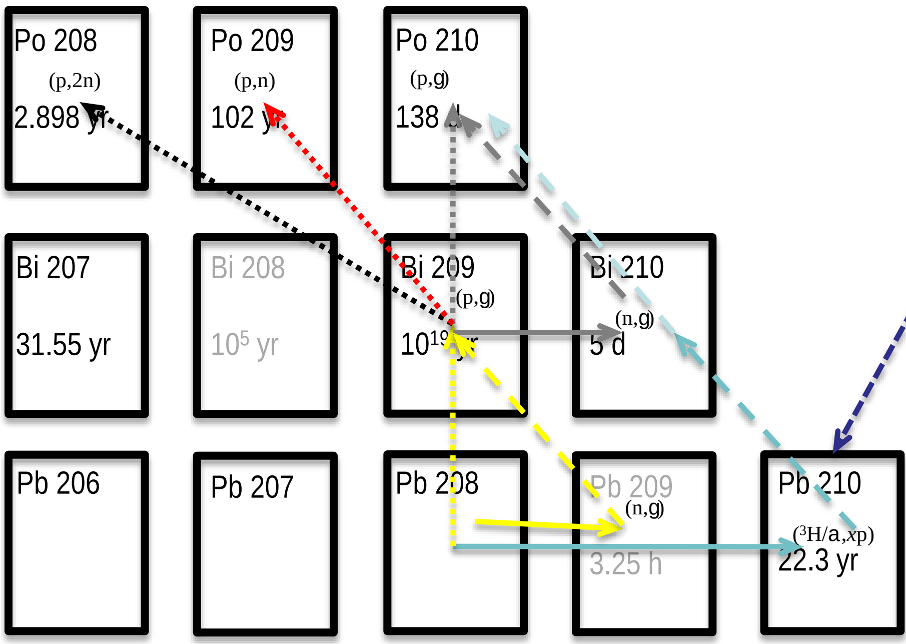
Production pathways of Polonium

^{209}Bi content in SINQ lead:

220 ppm

(ICP OES)

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



^{nat}U/Th content in SINQ lead:

no values

(ICP OES)

^{208}Pb (*p*/*n*,*g*) ^{209}Bi

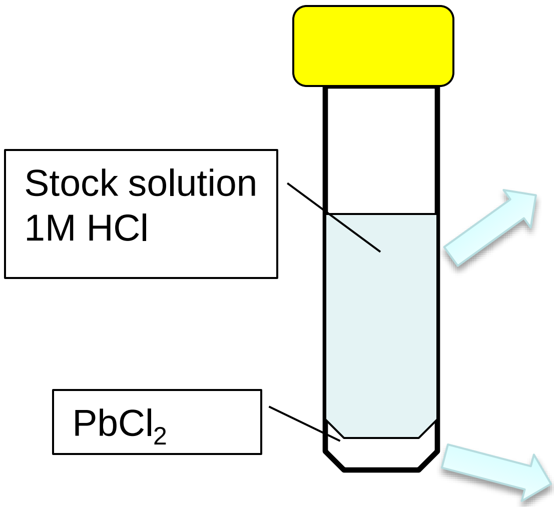
^{209}Bi (*p*/*n*,g) ^{210}Po

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

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

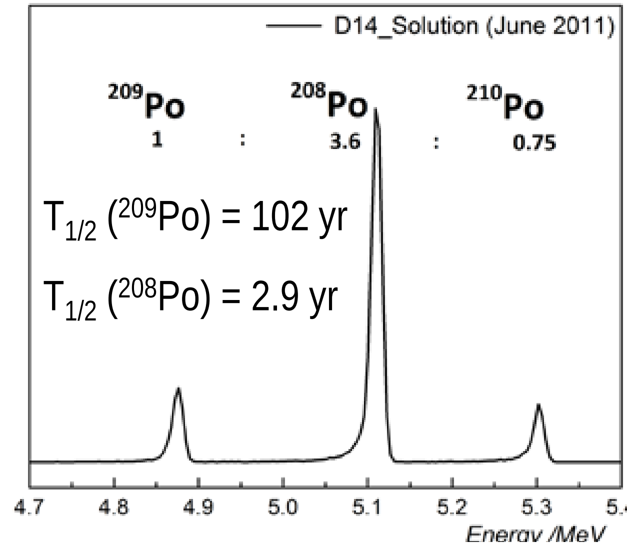
^{208}Pb (2*n*,g) ^{210}Pb

^{208}Pb (^{3}*H*/*a*,2*p*) ^{210}Pb



<u>Po in solution</u>

Deposition of equilibrated Po in 2011



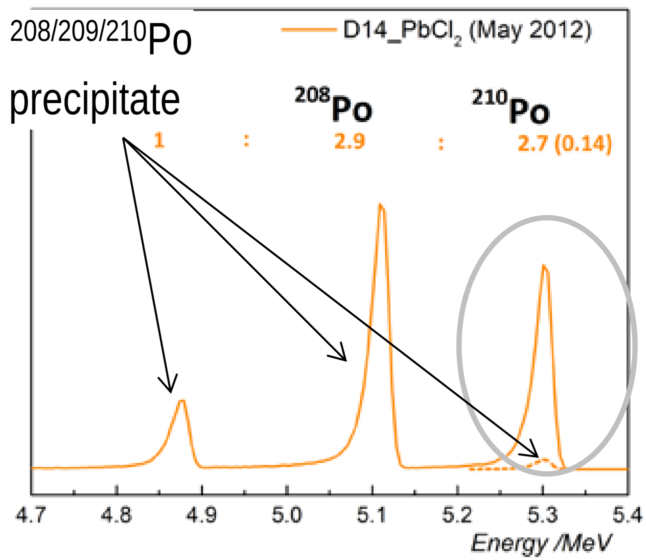
<u>^{210}Pb in $PbCl_2$</u>

Residues of $^{208/209/210}Po$ in the $PbCl_2$ precipitate

One year of ^{210}Po ingrow

↓

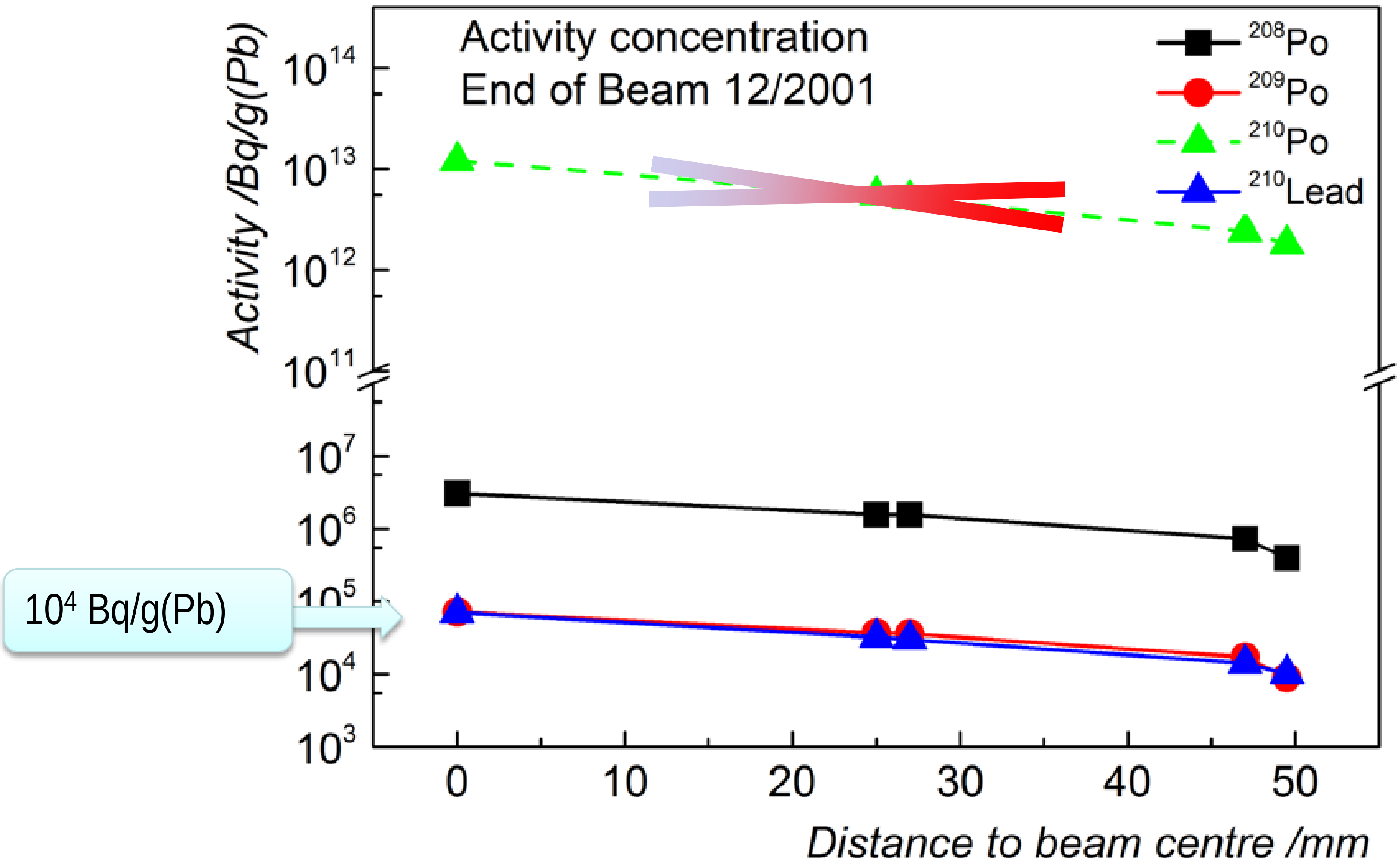
Deposition of Po in 2012



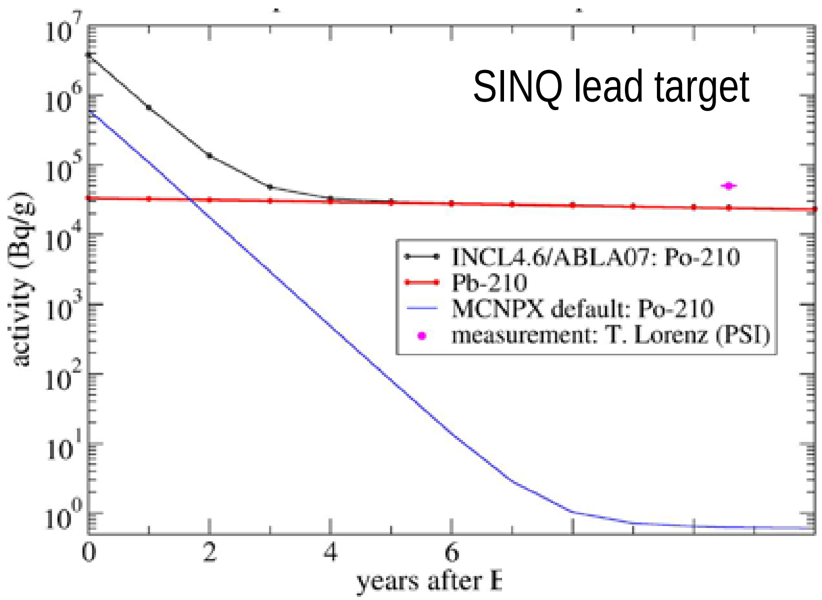
$$^{210}Pb\ (22.3\ a) \longrightarrow\ ^{210}Bi\ (5\ d) \longrightarrow\ ^{210}Po\ (138\ d)$$

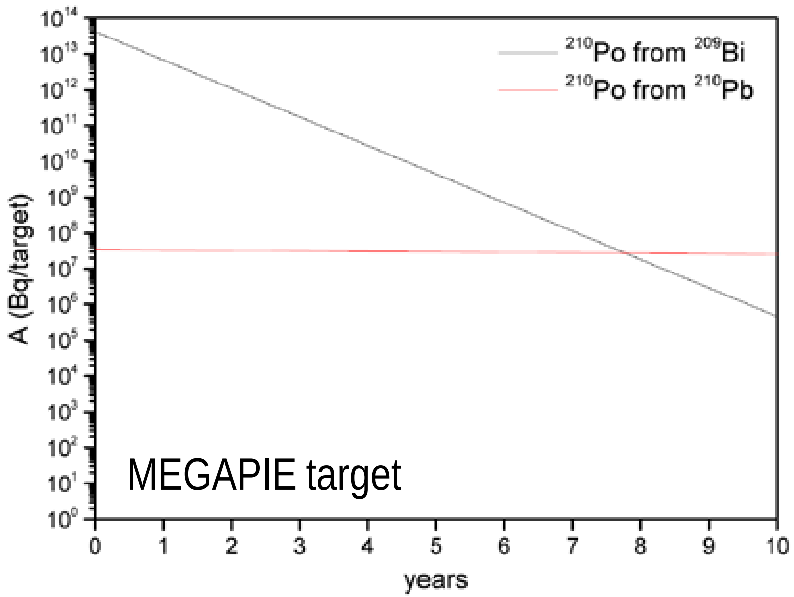
Po determination in SINQ lead target 4

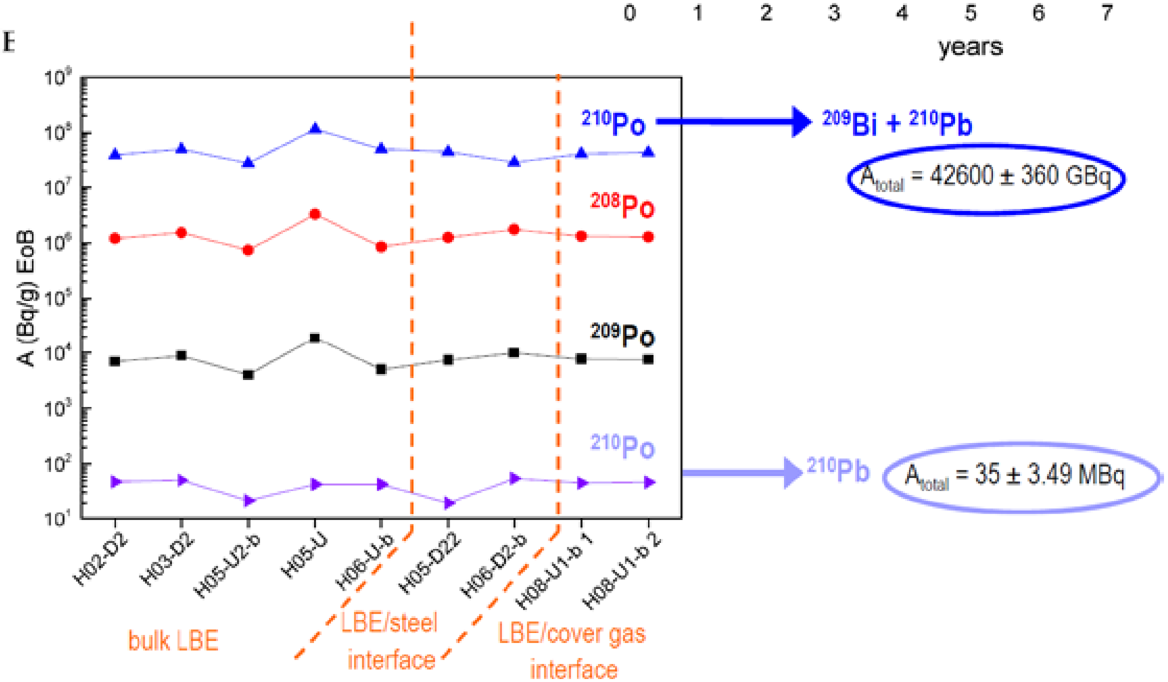
Activity concentration at EoB (12/2001):



Po evaluation in MEGAPIE







Summary of the results

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

Impact

Existing calculation codes

Code development/
New target designs

Validation of calculation codes

Knowledge of the radionuclide inventory

Analysis

Risk assessment
Decommissioning
Disposal
Licensing new facilities

Knowledge of the chemical behaviour

Impact of MEGAPIE

- Deposition on surfaces have a radiological impact, especially a-emitters (^{148}Gd)
- Release studies are safety-relevant
- Inventory determination improves the calculation codes (^{210}Pb-^{210}Po case)

Acknowledgement

«Thank you very much for ….. important information which tell reality in the spallation target business.
I believe everyone who are involved in the spallation neutron source technology, should be interested in.»

Yujiro Ikeda, Director of J-PARC, Japan, 2013 in a mail to the project leader Michael Wohlmuther

Thank you for your attention!