PAUL SCHERRER INSTITUT

PSI

2nd workshop on
Exotic Radionuclides from Accelerator Waste for Science and Technology (ERAWAST II)

ERAWAST – nuclear chemistry for nuclear science

Dorothea Schumann for the ERAWAST - collaboration

Paul Scherrer Institute

Overview

ERAWAST: objectives

Outcome of the previous workshop

Examples for chemical separations

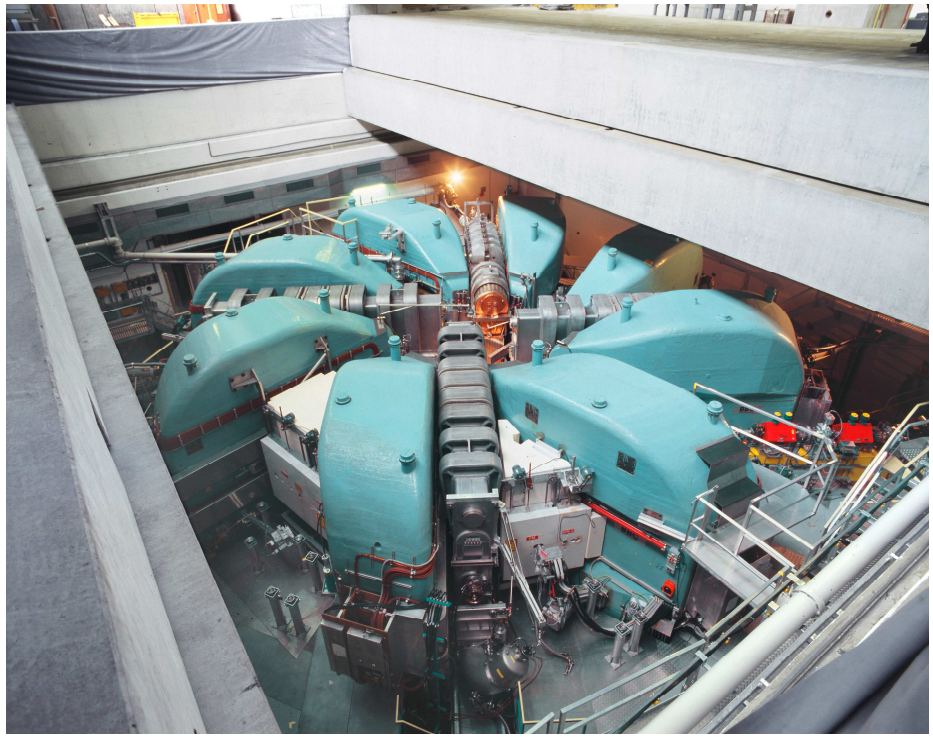
- Graphite wheels: Analytics and separation of ^{10}Be and others
- Lead from SINQ targets: Analytics and separation of ^{207}Bi
- ^{63}Ni-^{63}Cu separation

Potential for future experiments

PSI accelerator facilities and the ERAWAST-project

Exotic Radionuclides from Accelerator WAste for Science and Technology

Background: High-energetic protons and secondary particles produce in spallation reactions exotic isotopes with $A \leq 1+A_{Target}$



Collaboration between

Nuclide production facilities

Basic nuclear physics research
Nuclear astrophysics
AMS measurement groups
Environmental chemistry

1st ERAWAST workshop

15.-17.11.2006 at PSI

Exploratory workshop, supported by ESF (European Science Foundation)

25 talks, 30 participants from 12 countries

Objective: identify possibilities for isotope production on the one side and potential users on the other side

Topics:

- Accelerator waste analysis
- Radionuclide production possibilities
- Separation techniques
- Nuclear astrophysics
- Basic nuclear physics
- Medical applications
- Applied research

Outcome of the 1st ERAWAST workshop (1)

Bi- or multilateral collaborations which can be started in the near future (2007-2008)

- Laser based investigations and analytics of ultra rare trace isotopes (University of Mainz, Germany in collaboration with Nagoya University, Japan and University of Jyväskylä, Finland) + -
- ^{60}Fe (n,γ)^{61}Fe at stellar energies: Astrophysical quests and experimental challenges (Forschungszentrum Karlsruhe, Germany) + +
- Spectroscopy of ^{10}Be on the search for the beryllium halo nuclei charge radii (GSI Darmstadt, Germany) + -
- ^{7}Be for measuring the cross section of the ^{7}Be(p,γ)^{8}B reaction (Weizmann Institute Rehovot, Israel, ISOLDE/CERN Switzerland) + -
- ^{7}Be for ion-implantation at ISOLDE/CERN to study the half-life in various media and to perform ^{7}Be(n,p) emission channeling experiments at ILL Grenoble + + -
- A new ^{10}Be beam at CRC/UCL (Université catholique Louvain, Louvain-la Neuve, Belgium) + -
- ^{44}Ti abundance as a probe of nucleosynthesis in core collapse supernovae (University of Edinburgh, UK; UCL, Belgium) - -
- Model studies with a ^{44}Ti/^{44}Sc radionuclide generator (University of Mainz, Germany) + -
- Half-life measurement of ^{60}Fe and ^{53}Mn (Technical University of Munich, Germany) + + + -
- Proton-neutron interaction at the proton dripline near ^{44}V (ATOMKI, Debrecen, Hungary, in collaboration with RCNP, Osaka, Japan) - -
- AMS measurements with an ^{26}Al standard (ETH Zürich, Switzerland, University of Vienna, Austria) ++ -

Outcome of the 1st ERAWAST workshop (2)

Plans for collaboration in a longer time scale (start 2009 and later)

- Branchings in the s-process path (GSI Darmstadt, Germany) ?
- Neutron capture on radioactive isotopes for astrophysics (Los Alamos National Laboratory, USA) ?
- Perspectives for measurements of neutron reaction cross sections of rare radioactive isotopes at CERN n_TOF (CERN, Switzerland) +
- Half-life measurements of long-lived rare earth isotopes (Technical University Munich, Germany) ongoing
- Construction of a ^{44}Ti/^{44}Sc-generator for animal and clinical application (Uni Mainz), transferred to ZRW (Radiopharmaceutical department of PSI)
- Development of a ^{26}Al beam for nuclear astrophysics (Université catholique Louvain, Louvain-la Neuve, Belgium; TUM Munich, Germany) cancelled
- Irradiation of special samples in the SINQ target - Bi for the production of ^{205}Pb (Los Alamos National Laboratory, USA), enriched ^{34}S for the production of ^{32}Si (University of Vienna, Austria). + -
- Long-lived radionuclides produced in the irradiation of samples at the instruments LOHENGRIN and GAMS of the Institute Laue Langevin (Grenoble, France). +
- Mass separation needs further discussion - in particular for the study of ^{60}Fe production at stellar energies. partially solved

Outcome of the 1st ERAWAST workshop (3)

Generell agreements

- The Saha Institute of Nuclear Physics (Kolkata, India), the University of Mainz (Institute for Nuclear Chemistry, Mainz, Germany) and PSI (Villigen, Switzerland) will discuss possible collaboration concerning chemical separation techniques. not successful
- Other facilities shall be contacted for a possible collaboration (TRIUMF, Vancouver, Canada; Rutherford Appleton Laboratory, UK; SNS, Oak-Ridge, USA). +
- Other institutes and universities acting as users shall be included +
- A dedicated ERAWAST-webpage will be installed +
- Applications for joint projects (7th framework of EC, national funding and others). + -
- An announcement on ERAWAST will be made in Nuclear Physics News, the magazine of NuPECC +
- A proposal for an ESF Research Networking Program named ERIMAST (Exotic Radionuclides from Irradiated MAterials for Science and Technology) was launched to support the network of collaboration from 2008 + -

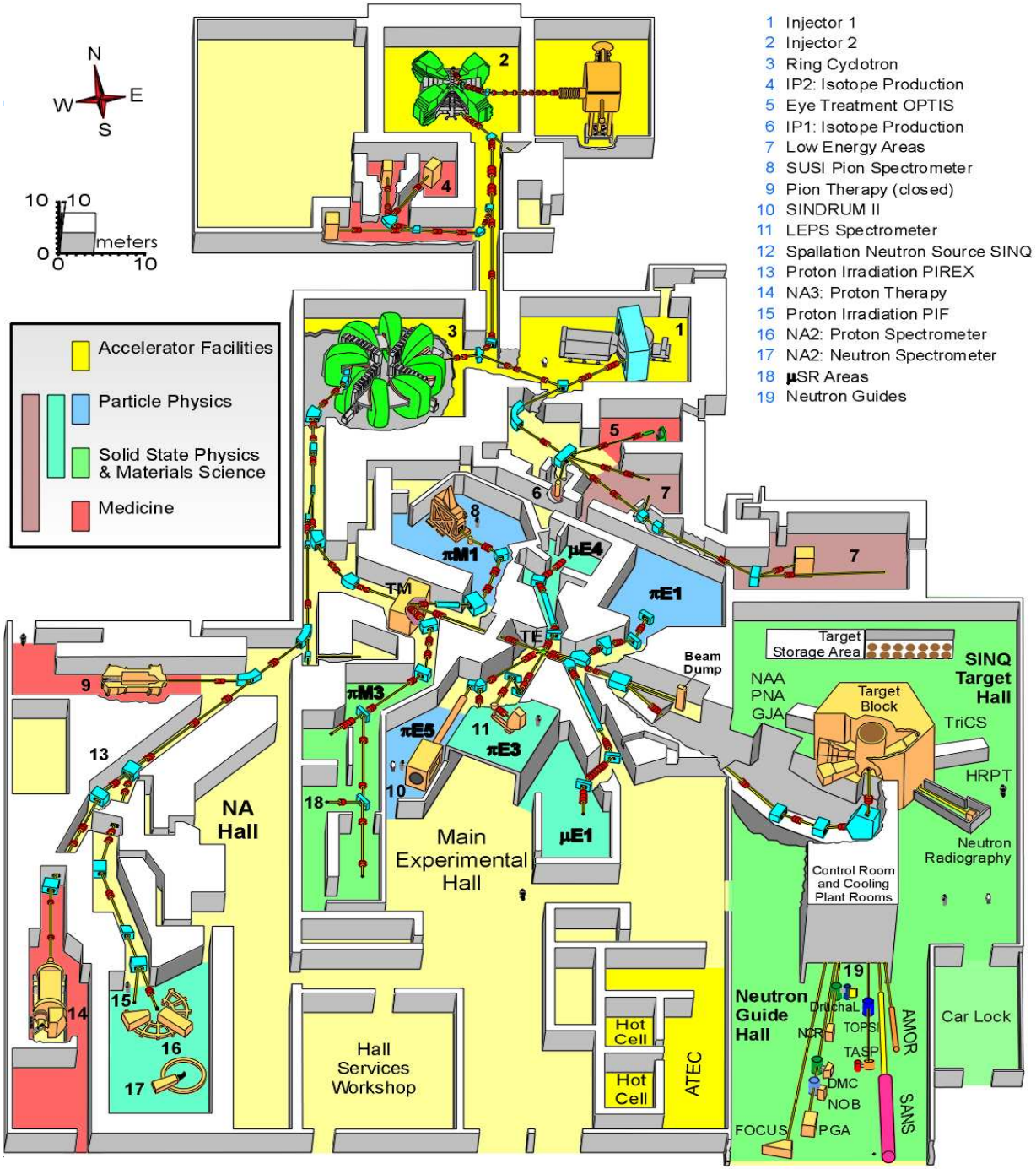
Objectives of the present workshop

Celebrate achievements

Continue with successful collaborations

Establish new collaborations

PSI accelerator facilities



Injector cyclotron (72 MeV protons)

590 MeV Ring Cyclotron
Up to 2.4 mA proton beam current

SINQ – spallation neutron source

COMET (cyclotron 250 MeV) for medical use

Ultra Cold Neutrons

SLS Swiss Light Source

Isotope production possibilities at PSI

Accelerator waste

Shielding, construction material, targets, beam dumps, cooling
intensely exposed by high-energetic protons and secondary particles
dismounted, cooled
ready or foreseen for disposal

Waste components:

Copper beam dump irradiated at the 590-MeV proton beam station at PSI, dismounted about 15 years ago
^{26}Al, ^{59}Ni, ^{53}Mn, ^{60}Fe, ^{44}Ti Ayranov

Proton-irradiated carbon from target E
^{10}Be, ^{7}Be ^{14}C, ^{3}H

Material from the SINQ facility

Lead targets
^{207}Bi, ^{182}Hf, rare earth elements (e.g. ^{146}Sm, several Dy isotopes) and lighter isotopes

STIP program (material research program)
Stainless steel for ^{44}Ti, ^{26}Al, ^{53}Mn production Bunka

SINQ cooling water Ayranov

^{7}Be, long-lived isotopes from irradiated structure material (^{22}Na, ^{88}Y and many others)

Special irradiations

The SINQ facility offers the possibility to irradiate materials with 590 MeV protons at special positions.
Tended experiments for isotope production can be offered
V for ^{44}Ti production
Bi for ^{205}Pb production

Irradiation with 71 MeV protons (injector 2) and up to 590 MeV neutrons (NAA, PNA)

Chemical separations with other material

Requirements for sample preparation

Important questions

Separation techniques depend strongly on the corresponding sample requirements

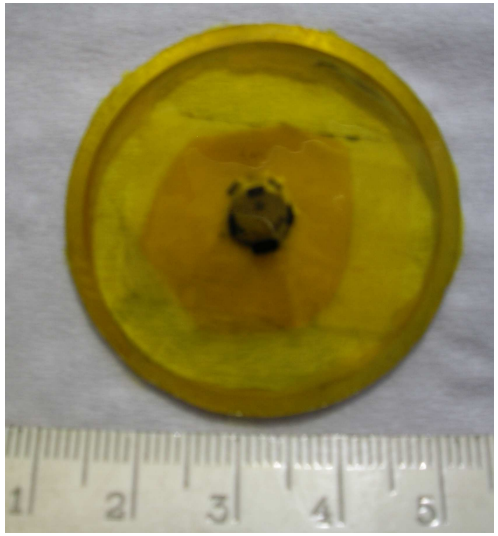
- Total amount of activity?
- Which chemical form?
- With carrier or non-carrier-added?
- Disturbing isotopes?
- Magnitude of decontamination factors?
- Matrix of the final sample?
- Single or multiple separation?
- Shielding equipment (hotcell) necessary?

The long way from a source to a sample

How do we get from a beam dump

to a sample solution

or a target?



Analytics

Separation

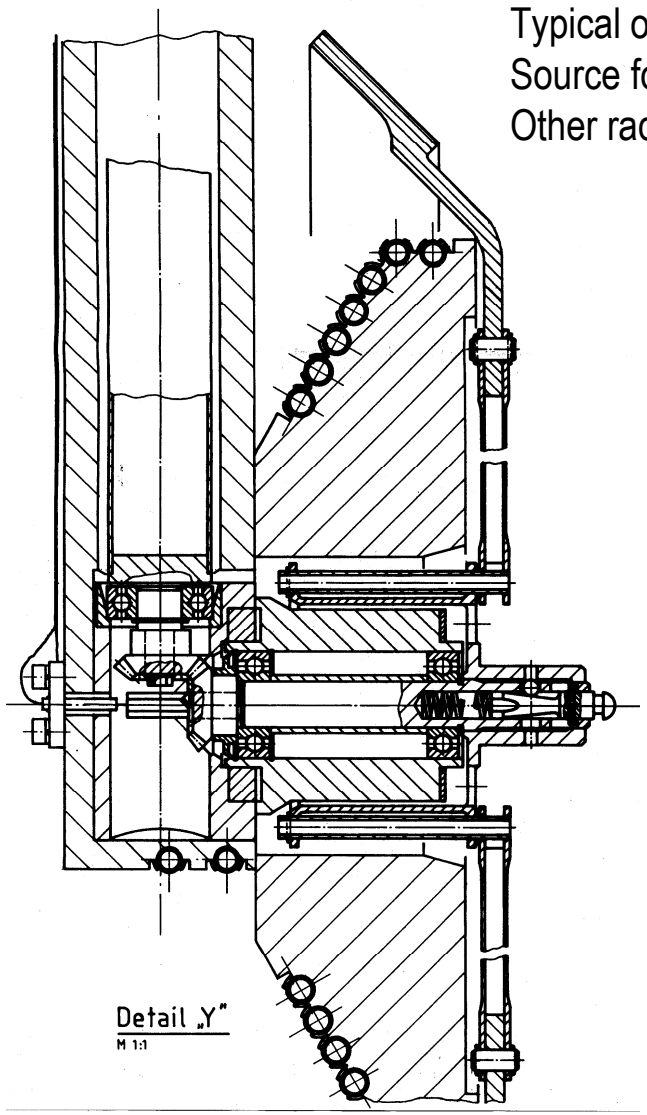
Preparation

Application

Analytics of accelerator waste at PSI

- Activation of components no more negligible due to the high beam current (mA range)
- Residue nuclide production in spallation targets is not as well-known as for Nuclear Power Plants – especially poor knowledge for long-lived products
- Complex nuclear reactions (protons, neutrons, other secondary particles) make theoretical predictions difficult
- Reactions with impurities and reaction products make theoretical predictions much more difficult
- Product spectrum covers all elements up to 1 unit higher than the target mass → complex chemistry
- Target material (solid or liquid heavy metals) requires knowledge on intermetallic compounds and interactions, in many cases not available
- Volatiles require special attention
- Theoretical calculations for predictions of the radionuclide inventory exist at PSI, but experimental determinations for benchmarking are mandatory
- Radiochemistry is an essential issue for evaluation processes, licensing procedures, decommissioning and waste management connected with the operation of large accelerator facilities
- Gives information on promising material for the production of valuable exotic isotopes

Graphite targets from Target-E



Myon production station, consumes up to 20% of the proton beam
Typical operation time: 1-3 years
Source for ^{7}Be and ^{10}Be
Other radionuclides: ^{14}C, ^{3}H, impurities of ^{22}Na, ^{54}Mn, $^{57/60}$Co

TARGET CONE

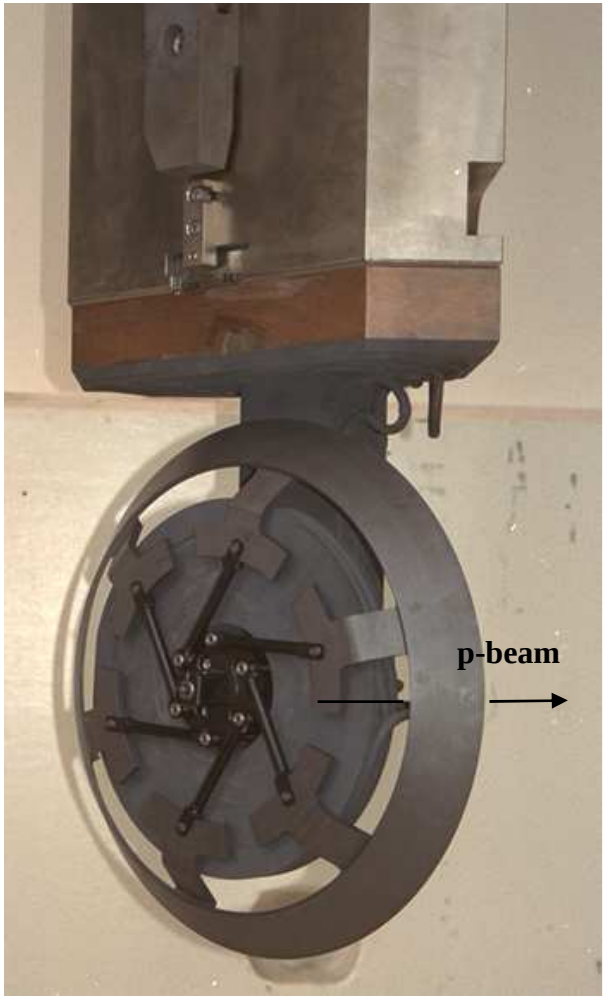
Mean diameter:	450 mm
Graphite density:	1.8 g/cm^3
Operating Temperature:	1700 K
Irradiation damage rate:	0.1 dpa/Ah
Rotational Speed:	1 Turn/s
Target thickness:	60/40 mm
	10/ 7 g/cm^2
Beam loss:	18/12 %
Power deposition:	30/20 kW/mA

SPOKES

To enable the thermal expansion of the target cone

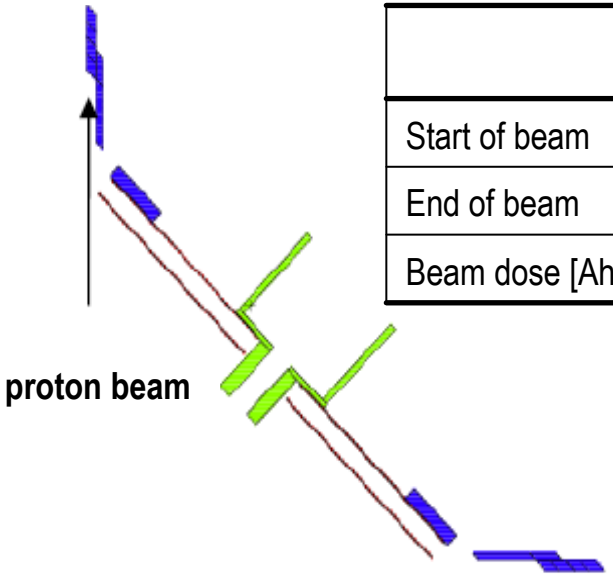
BALL BEARINGS *)

Silicon nitride balls
Rings and cage silver coated
Lifetime 2 y
*) GMN, Nürnberg, Germany

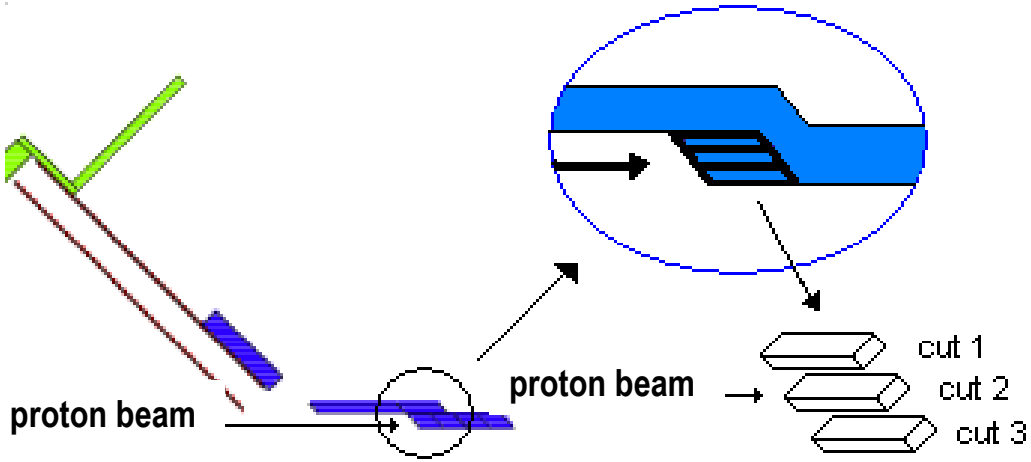


A new design of graphite wheel

Sample taking



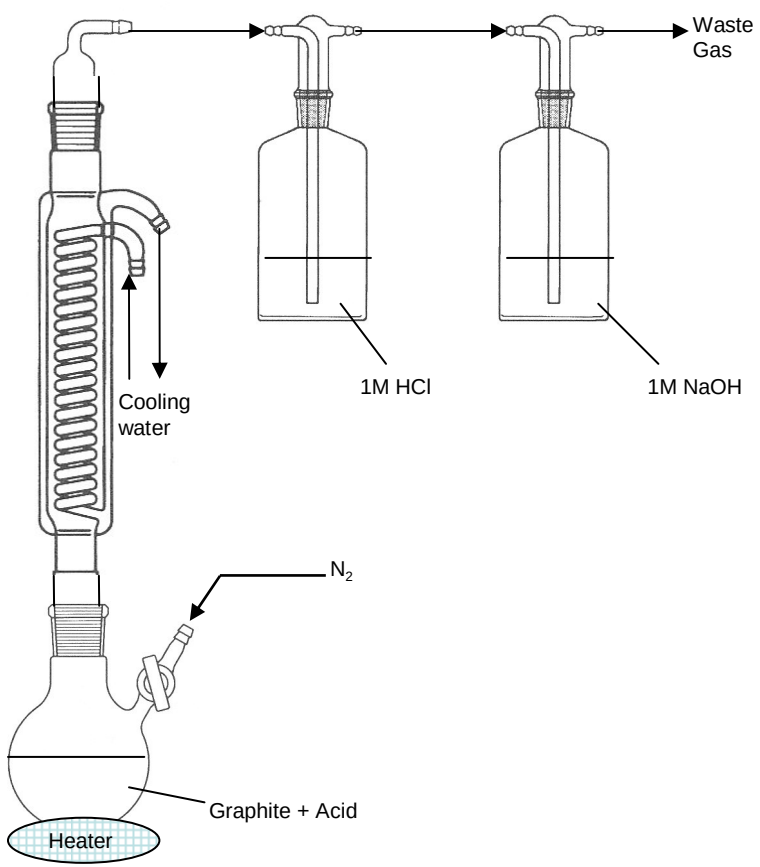
	E70	E71	E72	E78	E79	E83	E92
Start of beam	24.6.91	07.6.94	11.3.96	12.2.97	12.3.99	18.4.02	19.6.02
End of beam	07.6.94	11.3.96	20.9.96	18.12.98	28.10.99	19.6.02	23.12.05
Beam dose [Ah]	3.98	5.14	3.89	11.35	6.64	1.84	28.79



Graphite pieces from several targets

Sewing of the pieces 90 to beam direction to get the radionuclide distribution along the beam profile

Chemistry



Determination of ^{3}H and ^{14}C:

- Dissolving the graphite in $H_2SO_4/HNO_3/HClO_4$ 2:1:1
- Evaporation of CO_2 and adsorption in 1M NaOH for determination of ^{14}C
- ^{3}H remains in the solution (aliquot for LSC measurement)

Determination of ^{10}Be

- Adding stable Be as carrier
- Dissolving the graphite in
- $H_2SO_4/HNO_3/HClO_4$ 2:1:1
- Precipitation of $Be(OH)_2$ with NH_3
- Purification of the Be fraction by ion exchange on DOWEX50x8
- Measurement by AMS and ICP-MS

Results (1)

Measured data for all targets: ^{3}H [MBq/g]; ^{14}C, ^{10}Be [Bq/g]; beam dose in Ah

sample	Beam dose	^{10}Be meas. ICP-MS AMS	^{10}Be calc.	^{3}H meas.	^{3}H calc.	^{14}C meas.	^{14}C calc.
E70	3.98	220	998	$7{,}7\cdot10^{8}$	$1.1\cdot10^{9}$	$1\cdot10^{3}$	8.8
E71	5.14	291 316	1290	$4.8\cdot10^{8}$	$1.6\cdot10^{9}$	$1.1\cdot10^{3}$	11.4
E72	3.89	506 495	1140	$1.4\cdot10^{9}$	$1.3\cdot10^{9}$	$2.4\cdot10^{3}$	8.6
E78	11.35	2049	2850	$1.5\cdot10^{6}$	-	< 500	-
E79	6.64	3971		$6.9\cdot10^{8}$	$2.7\cdot10^{9}$	2041	11
E83	1.84	541		$3.2\cdot10^{8}$	$8.8\cdot10^{8}$	<500	3
E92	28.79	13456		$3.6\cdot10^{8}$	$1.6\cdot10^{10}$	9401	53

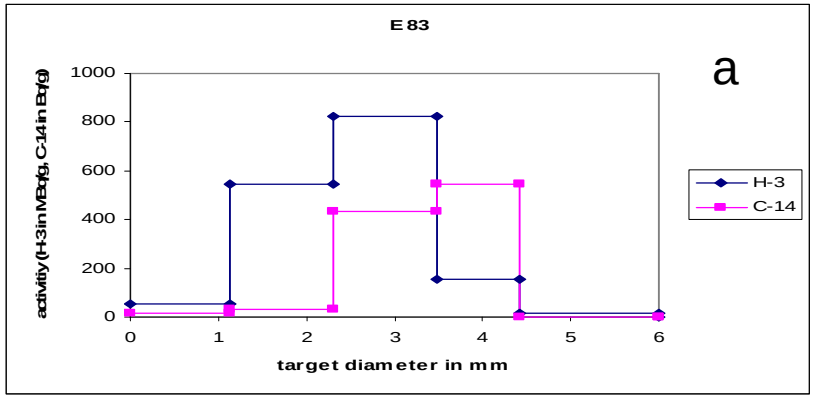
Calculations do not agree with measurements

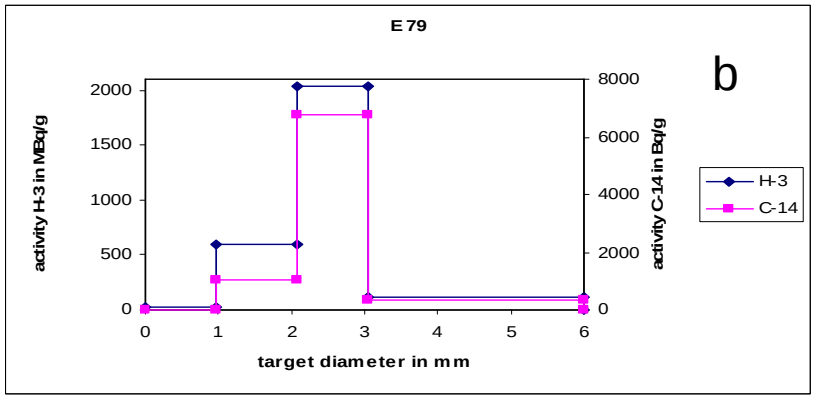
Measurements do not in all cases correspond to the beam dose (evaporation of tritium, secondary particle production dependent on beam dose)

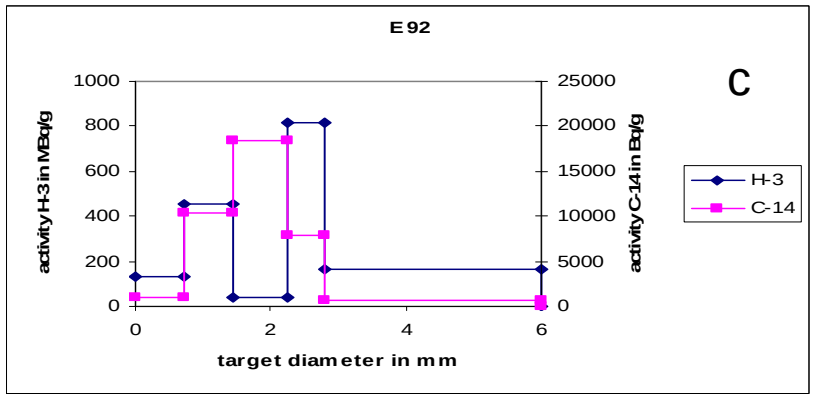
Sample taking position important (see next slide)

Results (2)

- Dependence of the measured ^{14}C and ^{3}H concentration on the sample taking position:
  - Figure a-c: Radial distribution of ^{3}H and ^{14}C in the graphite target wheels No. E 79, 83 and 92
  - ^{10}Be distribution is expected to be similar
- Important result: Distribution not homogenious; follows the beam profile; sample taking positions for analytics have to be selected carefully
- For isotope production positions with highest specific activity shall be used

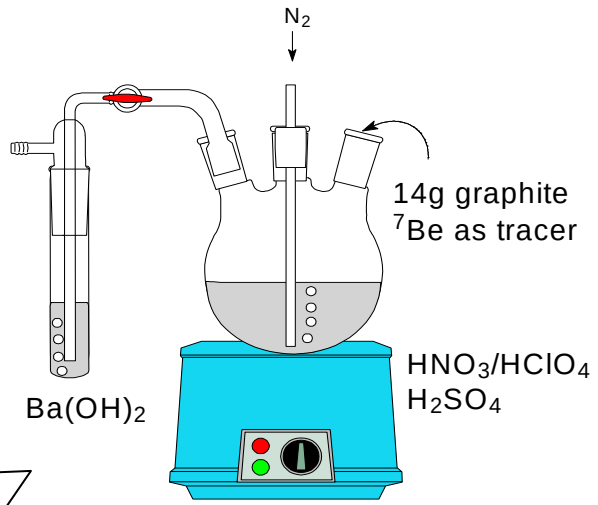


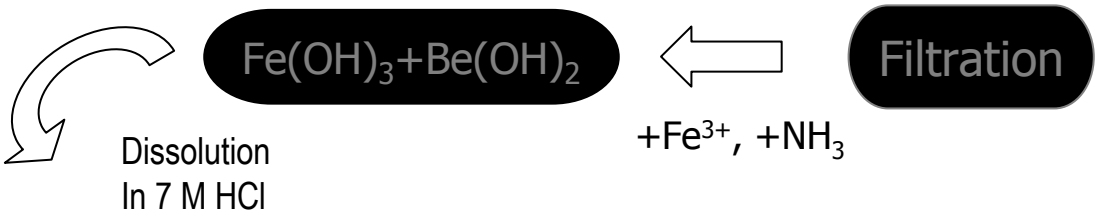


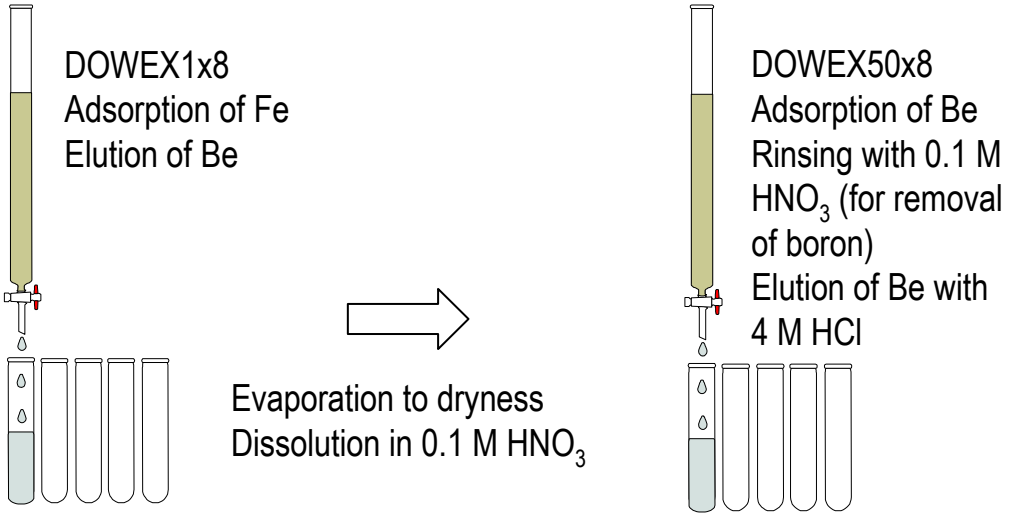


Chemical separation of ^{10}Be

- **Graphite of Target E contains mainly: ^{14}C, ^{3}H, ^{10}Be (^{7}Be - decayed)**
- **^{10}B as the stable isobaric isotope of ^{10}Be has to be separated nearly completely**

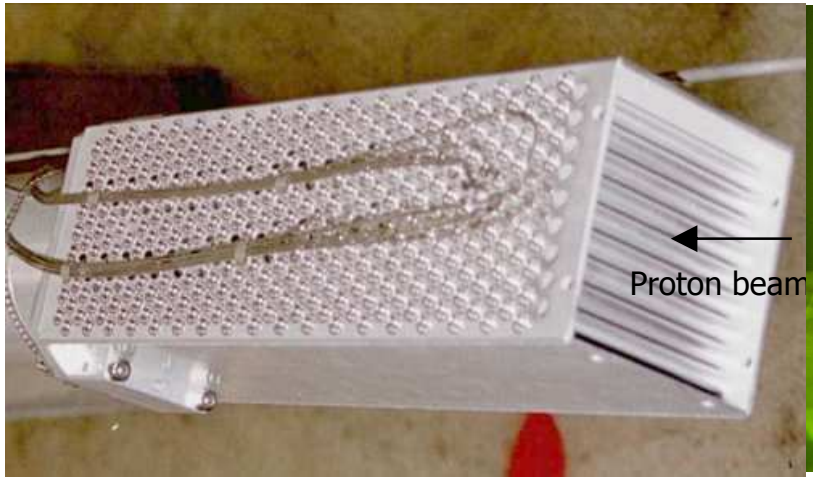


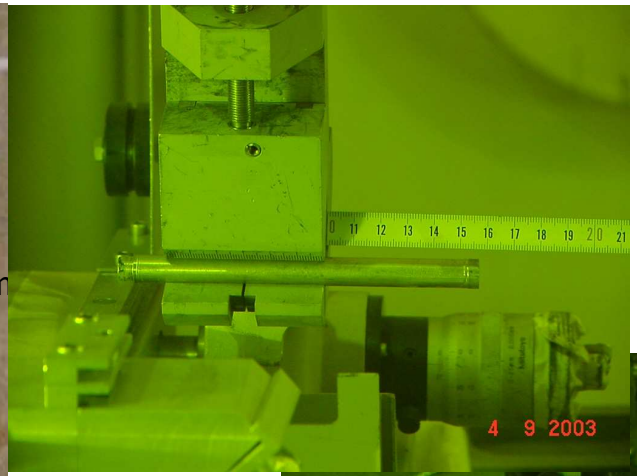




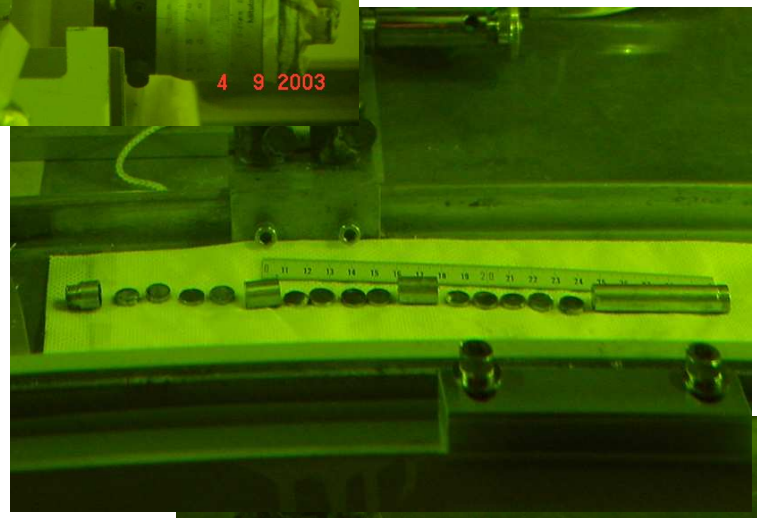
Result:
~ 80 µg ^{10}Be = $5 \cdot 10^{18}$ atoms

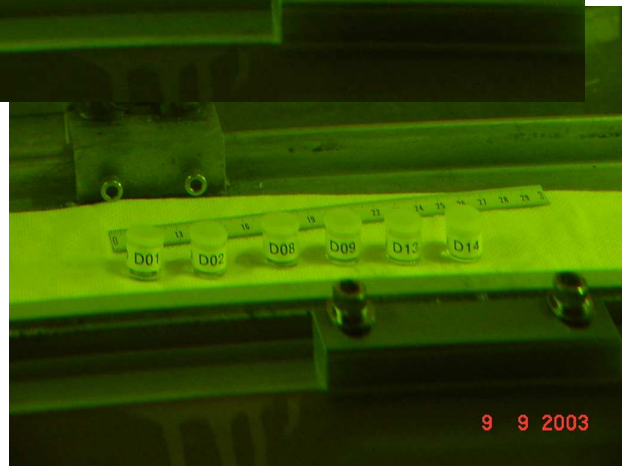
Lead targets from SINQ

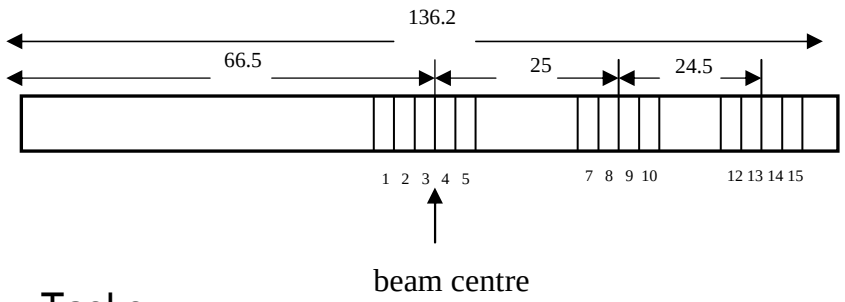




Samples from
target 4
2 years operation
EOB 1999







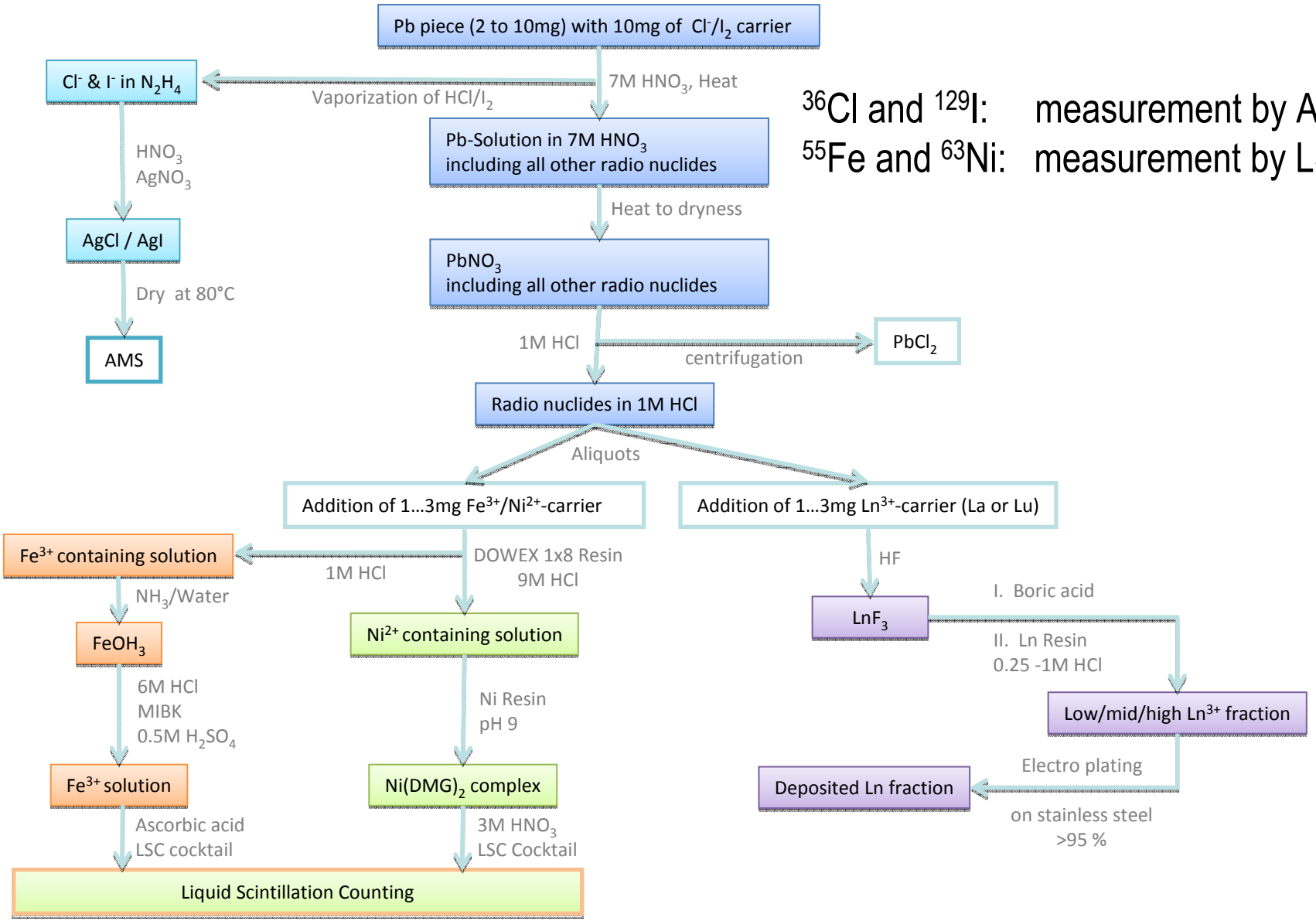
Tasks:

- Determination of the radionuclide distribution within the targets by analysing several rods (top ten radionuclides)
- Selection of material for radionuclide extraction (^{182}Hf, lanthanides and others)

First preliminary analytical results

γ-measurements (3/2011)	D9 [Bq/g]	D14 [Bq/g]	AMS-, LSC-, measurement	D9 [Bq/g]	D14 [Bq/g]
^{207}Bi	$2.52 \cdot 10^{7}$	$1.18 \cdot 10^{7}$	^{55}Fe		$1.05 \cdot 10^{5}$
^{172}Lu/^{172}Hf	$4.99 \cdot 10^{6}$	$2.12 \cdot 10^{6}$	^{26}Al	0.5	0.2
^{173}Lu	$2.82 \cdot 10^{6}$	$9.76 \cdot 10^{5}$	^{36}Cl	95	48
^{194}Au/^{194}Hg	$1.19 \cdot 10^{7}$	$5.08 \cdot 10^{6}$	^{63}Ni		$1.70 \cdot 10^{5}$
^{102}Rh	$2.00 \cdot 10^{6}$	$7.24 \cdot 10^{5}$			
^{202}Tl/^{202}Pb	$4.80 \cdot 10^{5}$	$1.30 \cdot 10^{5}$			
^{60}Co	$2.10 \cdot 10^{6}$	$8.73 \cdot 10^{5}$			
^{125}Sb	$5.17 \cdot 10^{5}$	$4.13 \cdot 10^{5}$	LSC-measurement: 4/2011		
^{133}Ba	$1.65 \cdot 10^{6}$	$6.93 \cdot 10^{5}$	AMS: 7/2006		
^{108m}Ag	$4.44 \cdot 10^{5}$	$1.50 \cdot 10^{4}$			

Radionuclides requiring chemical separations



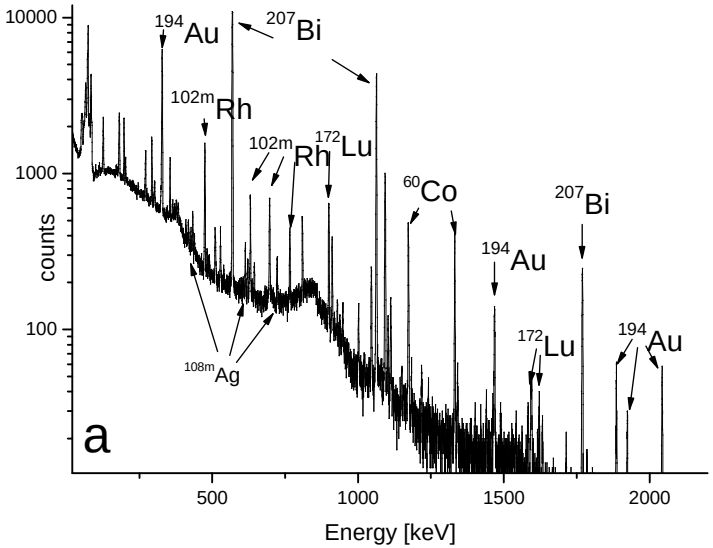
^{36}Cl and ^{129}I: measurement by AMS

^{55}Fe and ^{63}Ni: measurement by LSC

^{207}Bi - Calibration source for PTB

Application as γ-spectroscopic calibration source (240 kBq)

- relatively long half-life of 31.5 ys
- three γ-lines up to 1800 keV

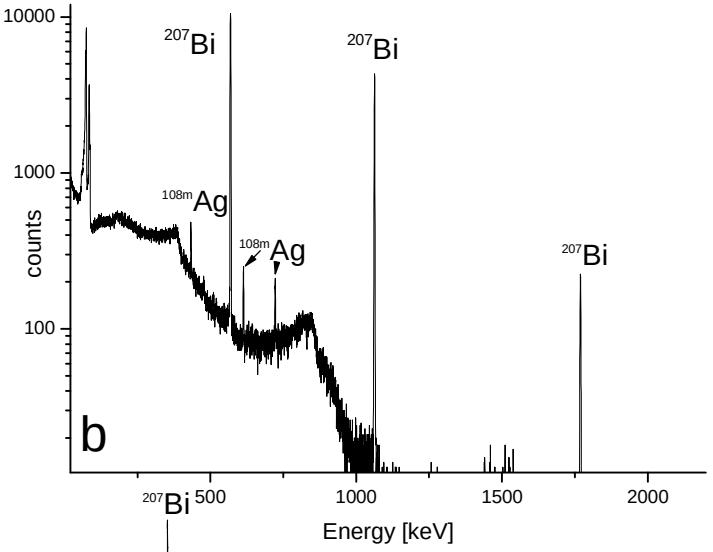


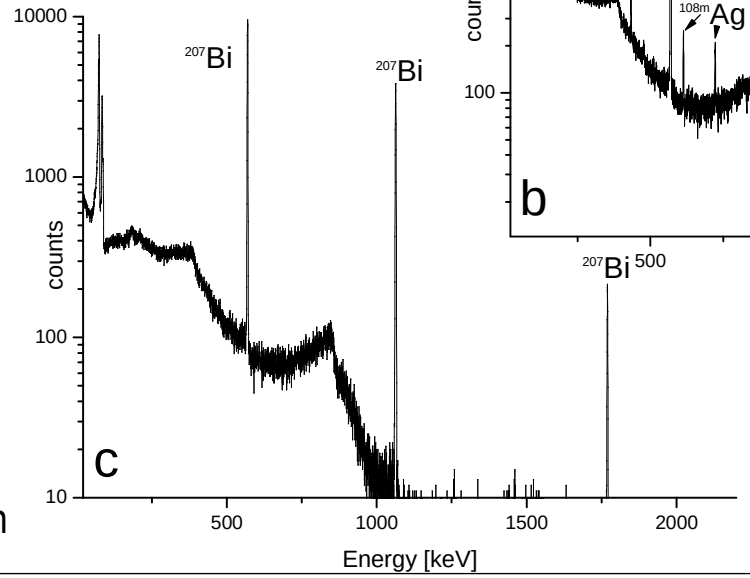
Purification from ^{108m}Ag

1mg La added
precipitation of the bismuth hydroxide with NH_3 solution
silver completely in solution
precipitate dissolved in 1M HNO_3
removal of La using cation exchange again

Chemical separation of ^{207}Bi

10 mg of irradiated Pb dissolved in 10 ml 7M HNO_3 (spectrum a)
solution evaporated
dissolved in 3M HNO_3 and passed through a lead-specific ion exchange column (Eichrom)
evaporated and dissolved in 1 M HNO_3
Cation exchange on a DOWEX50x8 column
Column washed with 10 ml 1M HNO_3
Bi-fraction eluted with 6 ml 0.4 M HCl (spectrum b)



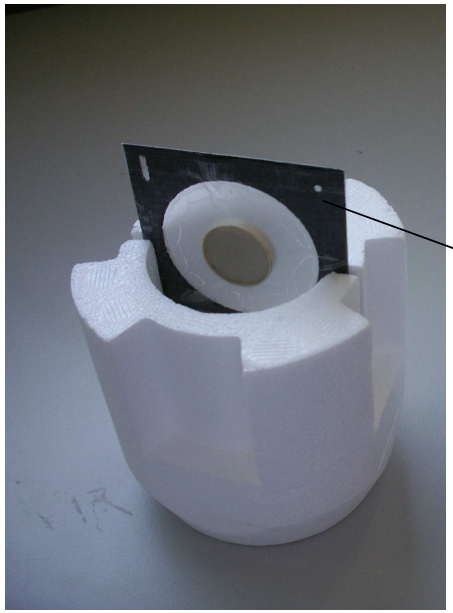


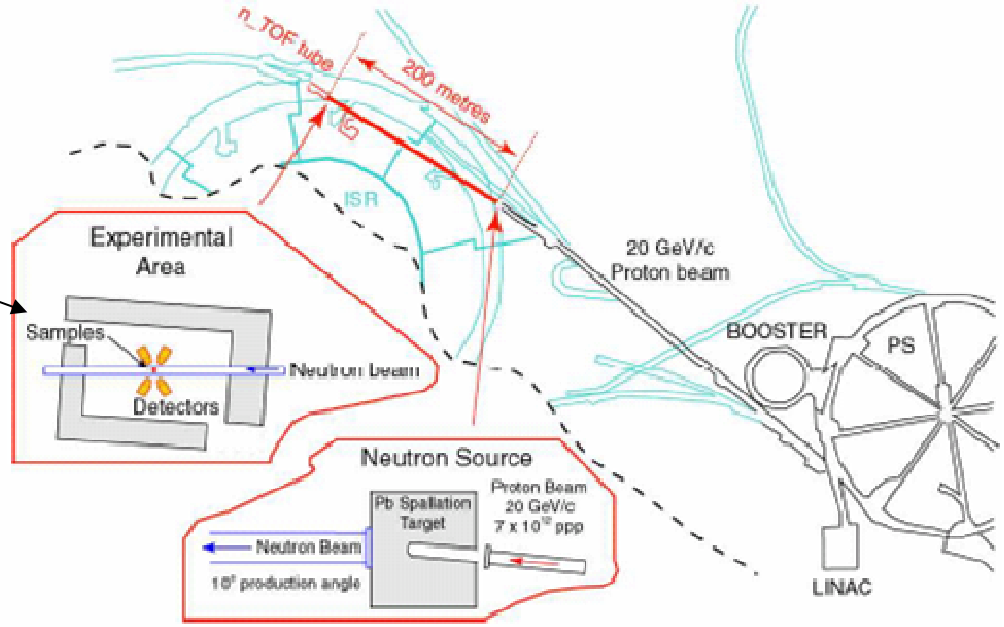
^{63}Ni-^{63}Cu separation – tasks for chemistry

Study of the neutron capture cross section of ^{63}Ni at n_TOF (s-process)

Tasks

- Separation of the in-growed ^{63}Cu because produces high resonances in the n_TOF experiment
- Re-measurement of the ^{63}Ni half-life

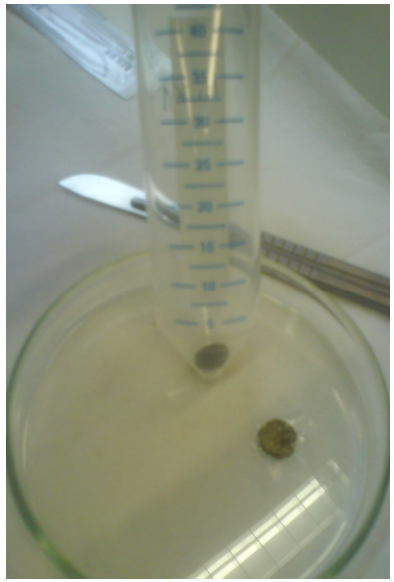


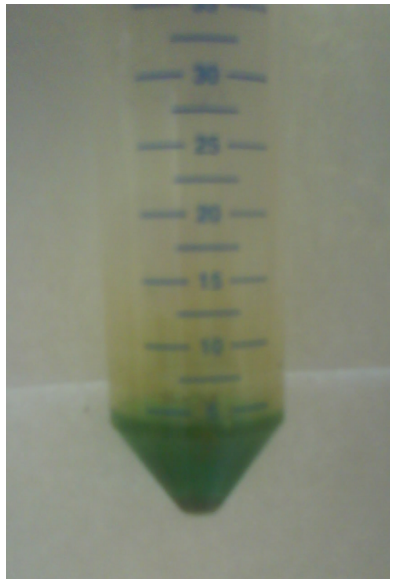


Sample description and chemical purification

Sample description

2 targets: LANL 347 mg, Karlsruhe 661 mg
(Originally from TU Munich)
Enriched ^{62}Ni (98%), irradiated in high flux reactors about 30 years ago
~11 % ^{63}Ni; total activity about 200 GBq
Expected amounts of isotopes after 30 years:
879 mg ^{62}Ni, 109 mg ^{63}Ni, 20 mg ^{63}Cu, 20 mg others
Total: 988 mg Ni, corresponds to 1255 mg NiO
Separation factor for Cu 2000 (model experiments, ICP-OES)



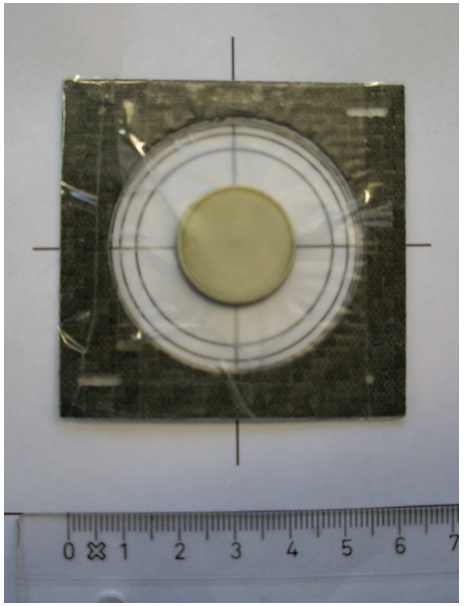


Chemical separation of ^{63}Cu

- Dissolution of the Ni-foils in 7 M HNO_3
- Evaporation
- Dissolution in 1M HCl
- Precipitation of CuS using H_2S
- Precipitation of $Ni(OH)_2$ with NaOH

Target preparation

- Drying at 80°C and calcination at 800°C to produce NiO
- Total amount NiO: 1147 mg (91% chemical yield)
- Packing into PEEK capsule



Summary and outlook

Achievements

^{207}Bi from lead	calibration source PTB
^{44}Ti from copper	calibration source HZDR
^{7}Be from graphite	precise measurement of the half-life
^{60}Fe from copper	target for neutron capture cross section
^{60}Fe from copper	determination of the half-life
^{63}Ni from LANL/KIT/ TUM	neutron capture cross section at n_TOF
^{7}Be from SINQ water	environmental studies
^{68}Ge activation in SINQ	AMS measurements
^{26}Al from STIP	300 Bq ready
^{10}Be from Target E	80 μg ready

Ongoing or upcoming experiments

^{44}Ti	50 MBq for target HZDR
^{44}Ti	100 MBq for radioactive beam (CERN, TRIUMF)
^{53}Mn	~ 10^{18} atoms for the half-life measurement (PSI) and neutron capture cross section
^{7}Be	1 GBq for ^{7}Be(n,α)α (SOREQ Nuclear Center)
^{60}Fe	~ 10^{15} atoms for the re- measurement of the half-life (PSI and Uni Vienna)
^{60}Fe	~ 10^{15} atoms for a target for thermal neutron capture cross section (Uni Vienna)
^{63}Ni	several MBq for the re-measurement of the half-life

^{59}Fe(n,γ)^{60}Fe reaction: irradiation of enriched ^{58}Fe at ILL, sample preparation ready, AMS-measurement of ^{60}Fe/^{58}Fe ratio at TUM ongoing

Future

Remote controlled separation of the copper beam dump

- ^{60}Fe n.c.a.
- ^{44}Ti n.c.a.
- ^{26}Al n.c.a.
- ^{53}Mn n.c.a.

Complete analysis of the SINQ-target

- Production of ^{207}Bi in the MBq range
- Separation of Lanthanides (^{146}Sm)
- Separation of ^{182}Hf
- Other radionuclides

New collaborations

ERAWAST III workshop?