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ERAWAST – a New Production Route for Exotic Long-lived Radionuclides

(Exotic Radionuclides from Accelerator WAste for Science and Technology)

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Overview

- The idea
- Description of PSI accelerator facilities
- Concept of ERAWAST
- Copper beam dump
- Graphite targets
- Lead targets
- Separation techniques
- Summary and Outlook

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The idea

- Accelerator waste with high beam dose available at PSI
- 590 MeV protons produce several spallation products in shieldings, beam dump and targets
- Accelerator waste contains considerable amounts of long-lived exotic radionuclides

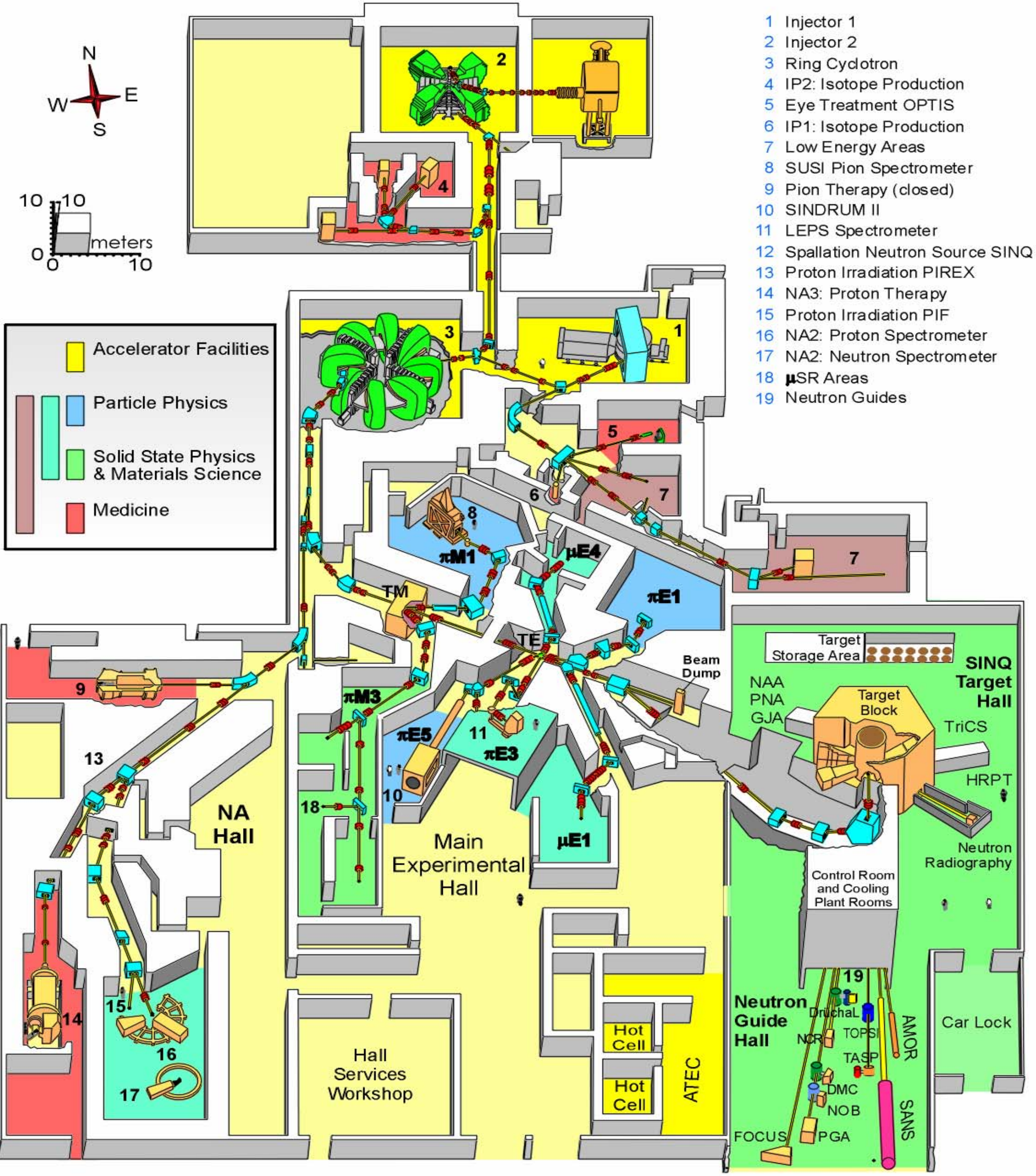
Application of exotic long-lived isotopes for several purposes

Collaboration between

- **Nuclide production facilities**
- **Basic physics research/Nuclear Structure**
- **Laser Spectrometry (RIMS)**
- **Nuclear Astrophysics**
- **Accelerator Mass Spectrometry (AMS)**
- **Pharmaceutical chemistry**

Workshop at PSI in Nov. 2006 (30 participants from 12 countries)

Chemical separation necessary!



Activated parts:

BX2-Target, Beam dump and shielding (Beam Control, 71 MeV protons)

BMA-Target, Beam dump and shielding (Pion therapy station, 590 MeV protons)

Target E, beam dump and shielding (590 MeV protons)

Lead and Zirkalloy from the SINQ facility

Materials:

Copper
Beryllium
Tungsten
Aluminium
Cast iron
Stainless steel
Graphite
Lead
Concrete

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Concept of ERAWAST

1. **Existing accelerator waste material**
   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 or others can be separated
   other irradiated materials like carbon (^{10}Be), stainless steel or concrete are also available
2. **Target material from the SINQ facility**
   Two irradiated lead targets from the spallation source are available. Heavier isotopes like ^{182}Hf or several rare earth elements (e.g. ^{146}Sm, several Dy isotopes) can be obtained. In principle, targets from the SINQ will be available every second year.
3. **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.

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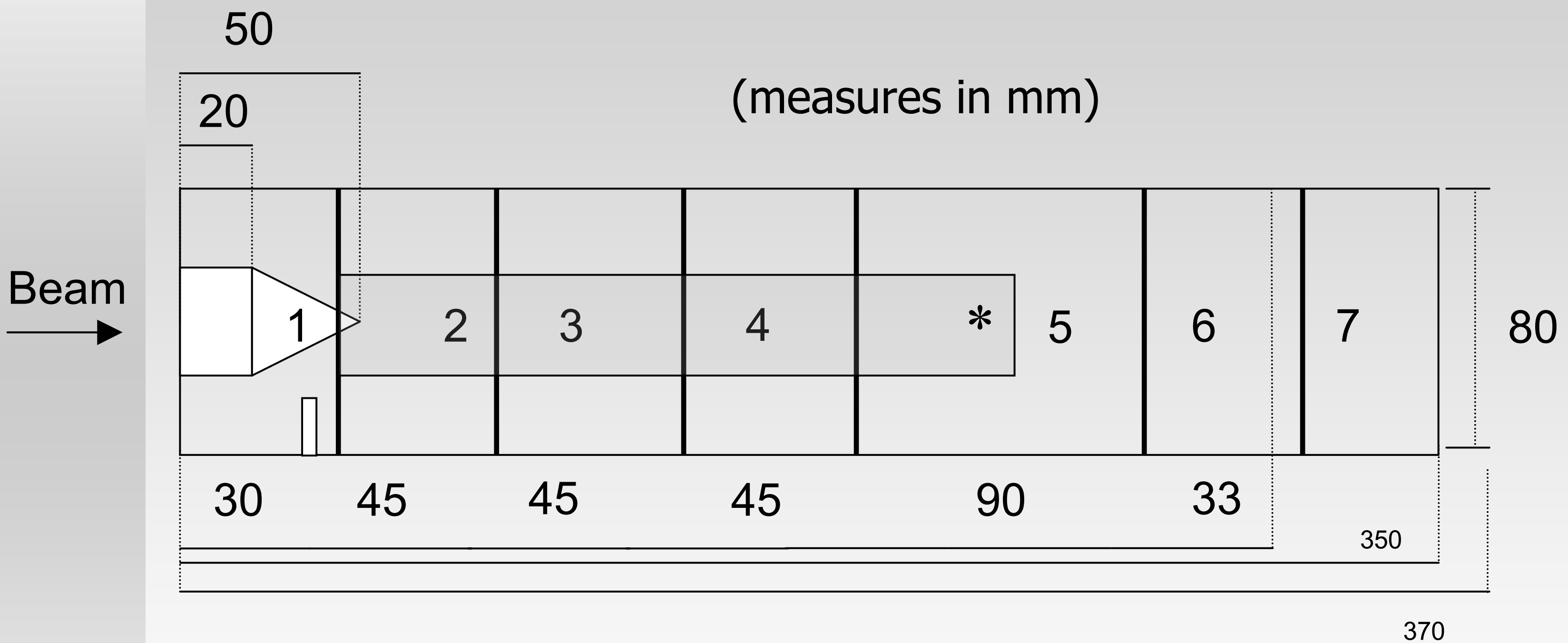
Characteristics of the copper beam dump

- Beam stop from the former BMA station
- Operated from 1980-1992, dismounted in 1993
- 0.1 Ah total beam dose (590 MeV protons)
- copper cylinder of ~ 10 kg; diameter 80mm
- Sample taking from several parts by drilling
- Characterization of the radionuclide inventory including radial and depth distribution

Sample	^{44}Ti [kBq/g]	^{36}Cl [Bq/g]	^{63}Ni [kBq/g]	^{55}Fe [kBq/g]	^{60}Fe [Bq/g]	^{26}Al [Bq/g]	^{110m}Ag [Bq/g]	^{108m}Ag [Bq/g]	^{59}Ni [Bq/g]	^{53}Mn [Bq/g]	**^{60}Co [kBq/g]**
C6.1.1	4.8	4.06	220.3	233.6		0.154	1.21	1.77			**224.0**
C6.1.2	0.54	0.49	133.7	37.3		0.016	1.98	2.52			**85.7**
C6.2.1	**1616.0**		34151.1	42450.3		**140**	3.89	62.58		**6900**	**49957.1**
C6.2.2	0.2	0.30	217.7	108.6			2.48	1.35	129	0.6	**111.6**
C6.3.1	**740.8**		36566.7	44136.3		**56**	2.3	37.27		**4310**	**37969.6**
C6.3.2	18.6		2006.0	2562.0	3.3	1	0.453	9.19	6620	112	**2691.8**
C6.3.3	1.5		1109.7	1552.4	1.9	0.2	1.32	9.22	2620	117	**1239.4**
C6.3.4	0.6		1841.6	257.0	0.5	0.03	0.92	1.69	759	6	**663.2**
C6.3.5	0.4	0.35	706.2	154.7			0.56	1.06	466	4.9	**438.8**
C6.4.1	**778.1**		16776.3	26590.4		**41**	1.64	27.70		**3600**	**47256.0**
C6.4.2		0.24	799.1	132.5			0.09	1.27		2.1	**505.5**
C6.5.1	**95.0**		5764.1	11520.4	**20.2**	**3**	11.70	13.64		**998**	**10091.9**
C6.5.2		0.27	545.6	157.8			0.59	1.80	422	2.0	**415.0**
C6.6.1	-	0.13	1005.7	287.6		0.012	3.75	3.93			**459.0**
C6.6.2	-	0.08	233.2	127.8		0.0019	0.49	0.94			**169.8**
C6.7.1	-	0.08	170.2	350.7			1.4	1.34			**148.6**
C6.7.2	-	0.04	118.8	233.6		0.0013	1.85	0.86			**91.1**
C6.7.3	-	0.04			0.1	0.0012	3.86	1.72	1		**56.1**
C6.7.4	-	0.04				0.0009	0.18	1.03		0.5	**6.1**

Schematic view of the beam dump



* - Area of drilling Ø 20mm

Drilling of appr. 500g of copper from the inner part containing about 80% of activity

Estimation of available radionuclides
(no separation)

^{44}Ti: 100 MBq
^{53}Mn: 500 kBq (10^{19} atoms)
^{26}Al: 7 kBq (10^{17} atoms)
^{60}Fe: (50 kBq – 10^{18} atoms)
^{59}Ni: ?
(^{60}Co: 5 GBq)

All these radionuclides can be provided without carrier, but some of them contain other long-lived isotopes (^{55}Fe/^{63}Ni)

Graphite targets

- Myon production station (target E)
- 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, $^{57/60}Co$

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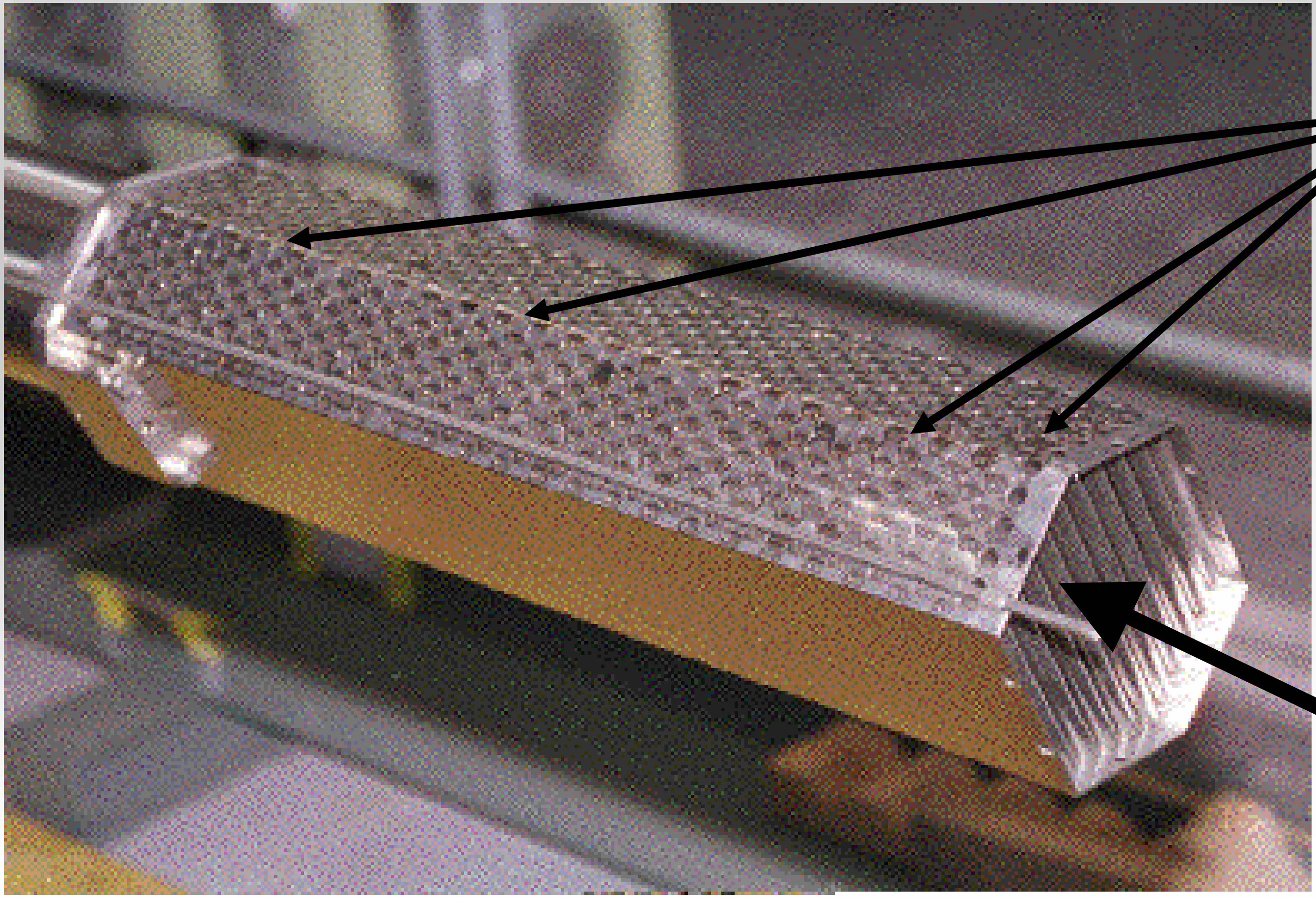
Results for $^{7/10}Be$

beam doses 4 – 11 Ah

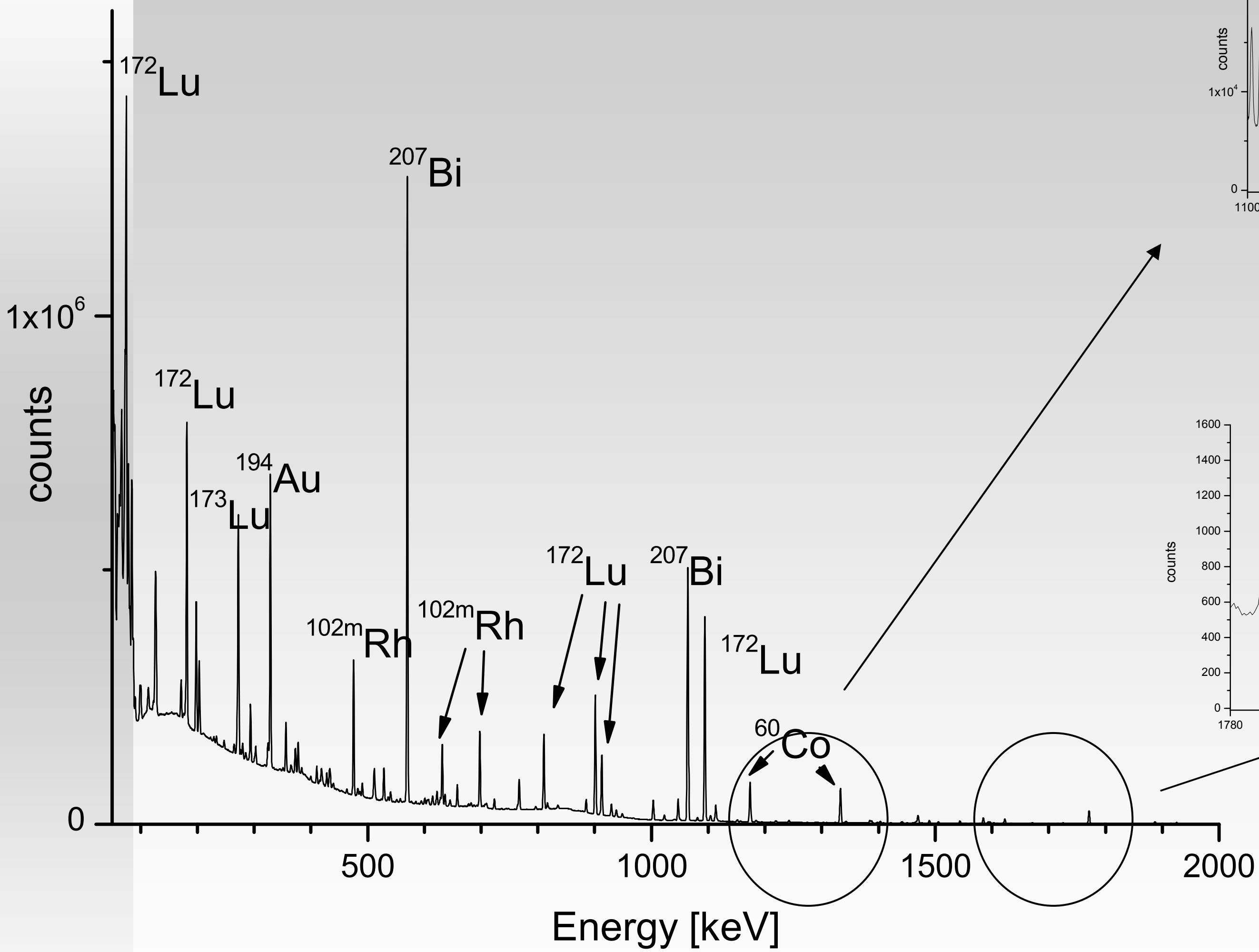
Sample	^{10}Be [Bq/g] ICP-MS	^{10}Be [Bq/g] AMS	Total amount of atoms	Total amount in µg	^{7}Be [Bq/g] EOB
1a	220		$6.7 \cdot 10^{16}$	1.6	-
1i		95			
2a	291	316	$8.4 \cdot 10^{16}$	1.4	$2.3 \cdot 10^{11}$
2i		7			
3a	506	495	$6.5 \cdot 10^{16}$	1.1	$1.5 \cdot 10^{11}$
4a	2049		$1.0 \cdot 10^{18}$	16.7	$8.4 \cdot 10^{10}$

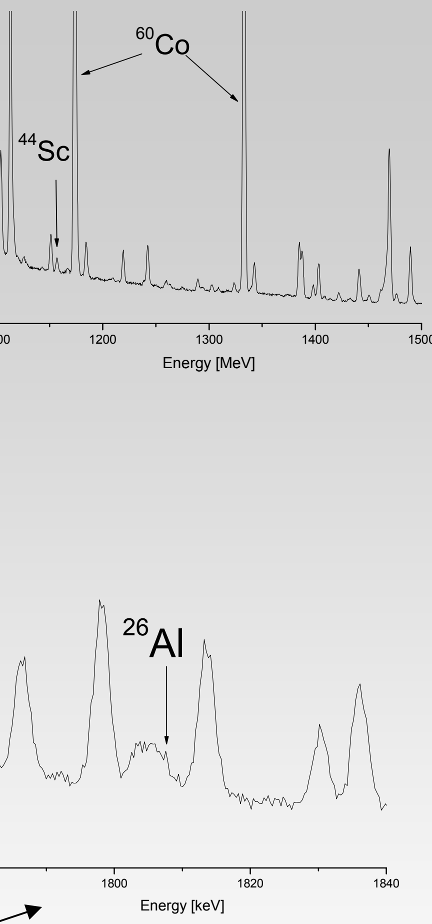
Lead targets from SINQ

2 Samples from target 4, 2 years operation; EOB 1999



Analytics





^{44}Ti (63y)	80kBq/g	^{194}Hg(520y)	20MBq/g
^{60}Co (5y)	4MBq/g	$^{172/3}$Lu/Hf (1.9/1.4y)	30MBq/g
^{207}Bi (31.55y)	30MBq/g		

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Examples for separation

- ^{60}Fe for determination of half-life, with carrier, 10^{15} atoms, collaboration with TUM)
- ^{60}Fe for neutron capture, carrier-free, 10^{16} atoms, collaboration with FZK
- ^{44}Ti for Ti/Sc generator (radiopharmaceutical use), carrier-free, 1 MBq, collaboration with University Mainz
- ^{44}Ti, probably for studies of core collapse supernovae, carrier-free, 3.5 MBq (collaboration with Uni Edinburgh)
- ^{26}Al, with carrier, standard material for AMS, 10 Bq; collaboration with ETH Zürich
- ^{26}Al, carrier-free, laser spectrometry (RIMS), 10^{13} atoms, collaboration with Uni Mainz
- ^{10}Be, carrier-free, radioactive ion beam, 5µg, collaboration with UCL
- ^{10}Be, carrier-free, laser spectrometry, 10^{13} atoms, collaboration with GSI

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Chemical separation of ^{60}Fe

Special Problem:

^{60}Fe (1.5·10^{6} y) $\xrightarrow{\beta^-}$ ^{60m}Co (10.5 min) $\xrightarrow{\gamma}$ ^{60}Co (5.3 y) $\xrightarrow{\gamma,\beta^-}$ ^{60}Ni (stable)

^{60}Fe: no γ radiation, low β–energy

Measurement of the increase of the Co-daughter → very good chemical separation from Co necessary

- Dissolution of 3.8g Cu (beam dump) in 7 M HNO_3
- Evaporation to dryness
- Dissolution in 7 M HCl
- + 5 mg Fe^{3+} and 5 mg Co^{2+} as carrier
- Extraction with Methylisobutylketone (MIBK)
- Aqueous phase: Ni, Co, Cu, organic phase: Fe
- Back Extraction with 0.1 M HCl, repetition of procedure
- Additional purification by precipitation of $Fe(OH)_3$
- Result: ~ 10^{15} ^{60}Fe atoms, decontamination factor (Co) < 10^{-7}

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Chemical separation of ^{44}Ti

- **Aliquot from the remaining solution of the Fe-separation (500 ml 7 M HCl)**
- **Contains Cu, ^{60}Co, ^{44}Ti and others**
- **Carrier free required**

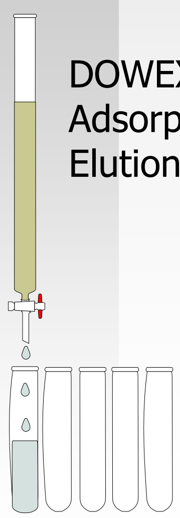
$Cu(NH_3)_4^{2+}/Co(NH_3)_4^{2+}$(95%)

$Fe(OH)_3+Ti(OH)_4+^{60}Co$

7 M HCl-solution

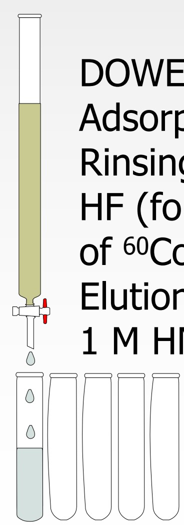
+Fe^{3+}, + NH_3

Dissolution
In 7 M HCl



DOWEX1x8
Adsorption of Fe
Elution of Ti

Evaporation to dryness
Fuming with conc. HF
Dissolution in 1 M HF



DOWEX1x8
Adsorption of Ti
Rinsing with 1 M HF (for removal of ^{60}Co)
Elution of Ti with 1 M HNO_3

Result:
~ 4.5 MBq ^{44}Ti
Decontamination Factor $^{60}Co<10^{-6}$

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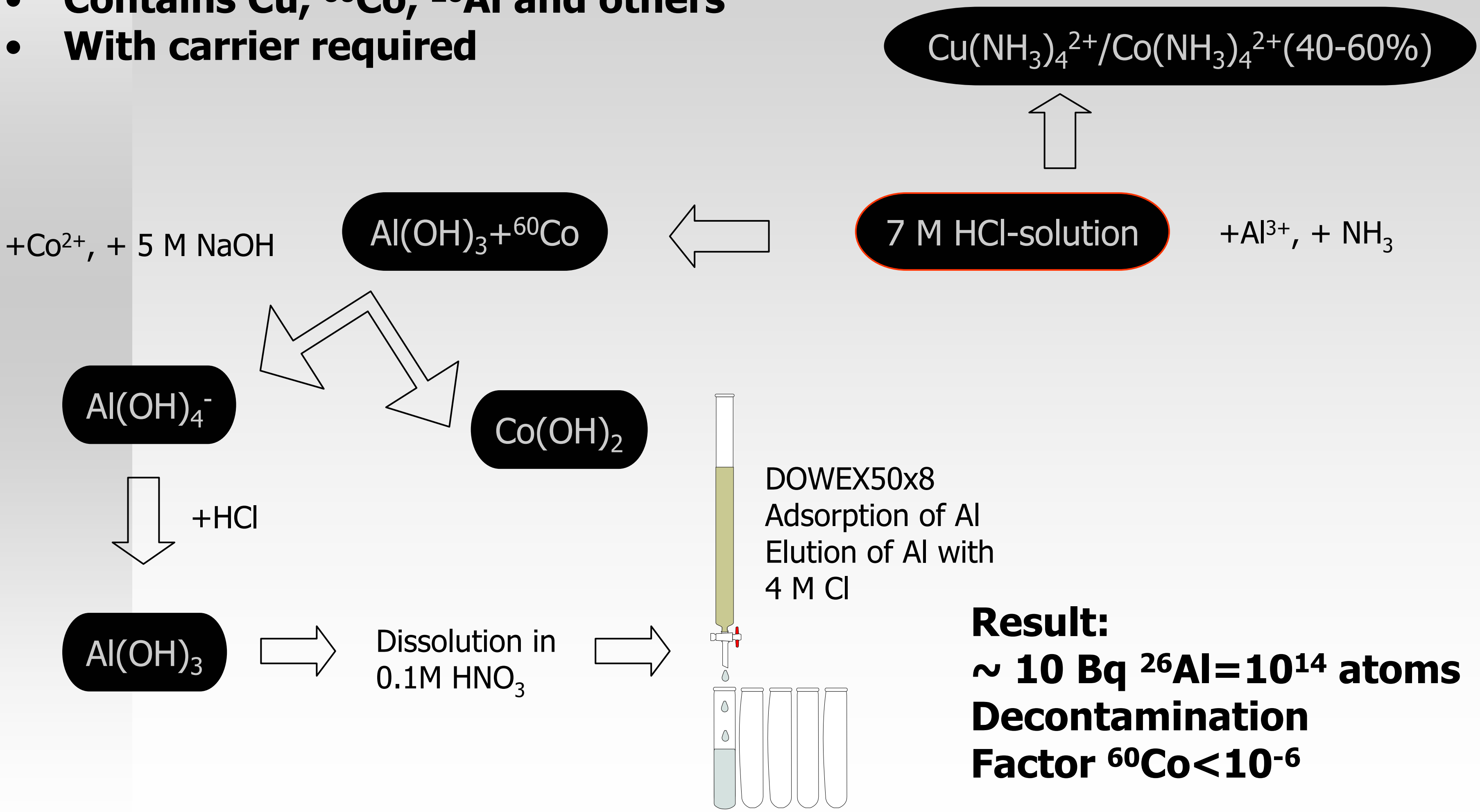
Chemical separation of ^{26}Al

- **Aliquot from the remaining solution of the Fe-separation (500 ml 7 M HCl)**
- **Contains Cu, ^{60}Co, ^{26}Al and others**
- **With carrier required**



Result:
~ 10 Bq $^{26}Al = 10^{14}$ atoms
Decontamination Factor $^{60}Co < 10^{-6}$

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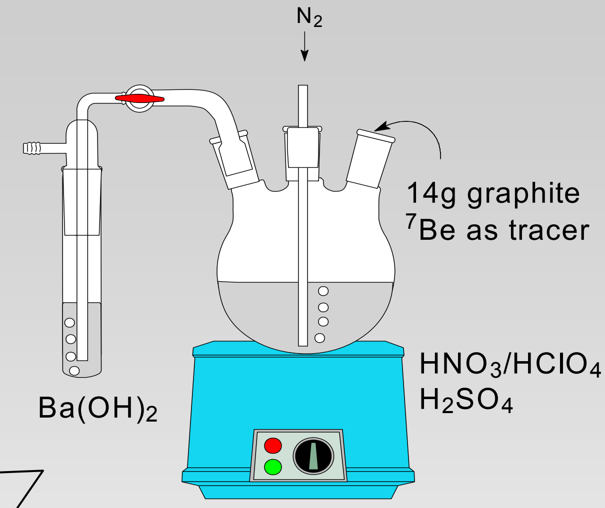
Chemical separation of ^{10}Be

- **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**
- **Carrier (stable ^{9}Be) not suitable for radioactive beam**

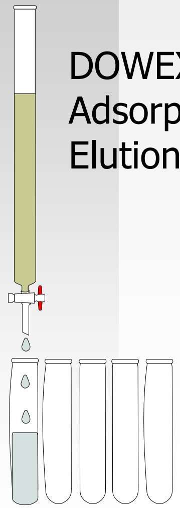


Filtration

$+Fe^{3+}$, $+NH_3$

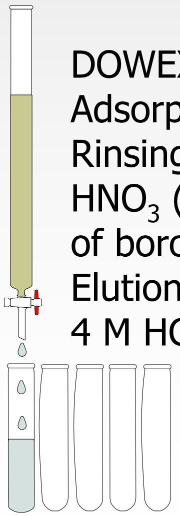
$Fe(OH)_3+Be(OH)_2$

Dissolution
In 7 M HCl



DOWEX1x8
Adsorption of Fe
Elution of Be

Evaporation to dryness
Dissolution in 0.1 M HNO_3



DOWEX50x8
Adsorption of Be
Rinsing with 0.1 M HNO_3 (for removal of boron)
Elution of Be with 4 M HCl

Result:
~ 16µg ^{10}Be =
$1 \cdot 10^{18}$ atoms

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Summary and Outlook

- Cu- and C-samples available
- Work on Pb-targets ongoing
- 10^{15-17} atoms of several radionuclides (^{26}Al, ^{60}Fe, ^{44}Ti) separated and available
- Up to 10^{18} atoms of ^{10}Be separated and available
- Possibilities for other irradiation positions (SINQ, beam dumps, collimators)
- ESF-funded Research-Network-Program launched
- Next step: Automated system for stepwise separation of big amounts of radionuclides from copper and carbon in a hotcell or glovebox
- Development of a similar system for the lead targets
- Routine production facility