Achievements and Perspectives of ERAWAST

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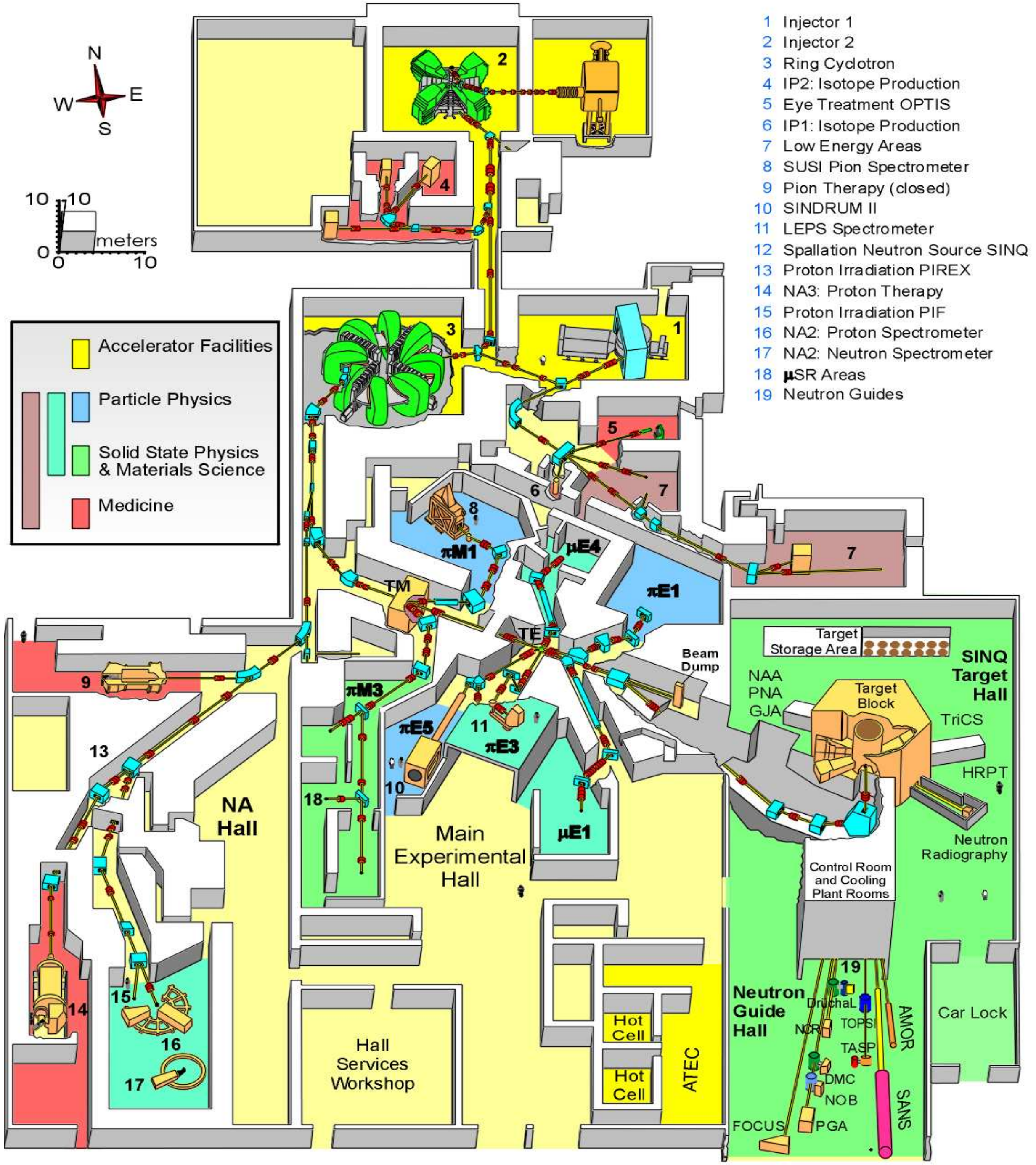
Outline

- The ERAWAST project
- Sources of radionuclides and their analytics
- Achievements
  - Re-measurement of the half-life of ^{60}Fe
  - Determination of the neutron capture cross section of ^{60}Fe
  - Remote-controlled system for copper separation
  - Separation of ^{7}Be from SINQ cooling water
- Future of ERAWAST
  - Future experiments with ^{60}Fe
  - Experiments with ^{44}Ti, $^{7/10}Be$
  - Cross sections and half-life of ^{53}Mn



Cyclotron:
590 MeV protons
Up to 2.5 mA

Activated parts:

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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Exotic Radionuclides from Accelerator WAste for Science and Technology

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

1. **Existing accelerator waste material**
   Copper beam dump irradiated at the 590-MeV proton beam station at PSI, dismounted about 15 years ago for ^{26}Al, ^{59}Ni, ^{53}Mn, ^{60}Fe, ^{44}Ti
   Carbon from target E for ^{10}Be
   SINQ cooling water for ^{7}Be
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. 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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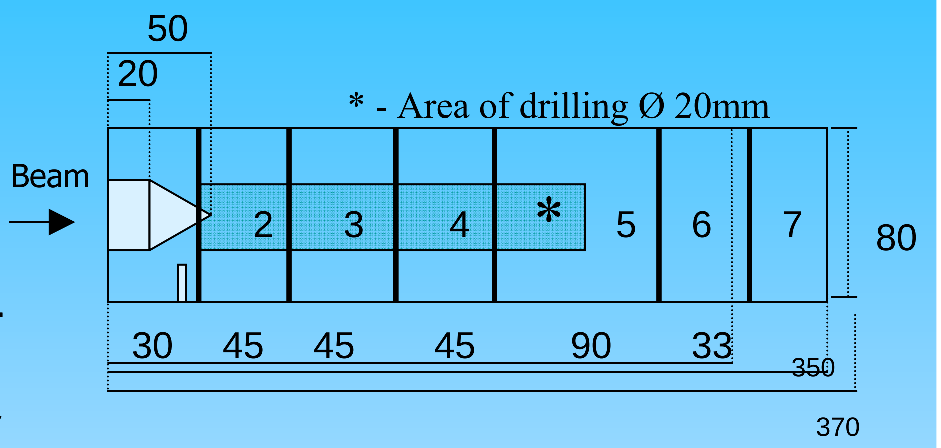
Sources and Analytics

Copper beam dump
Graphite targets
SINQ cooling water
Lead targets

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



^{44}Ti: 100 MBq (10^{18} atoms)
^{53}Mn: 500 kBq (10^{19} atoms)
^{26}Al: 7 kBq (10^{17} atoms)
^{60}Fe: 5 kBq (10^{17} atoms)
^{59}Ni: 8 MBq (10^{19} atoms)
(^{60}Co: 5 GBq)

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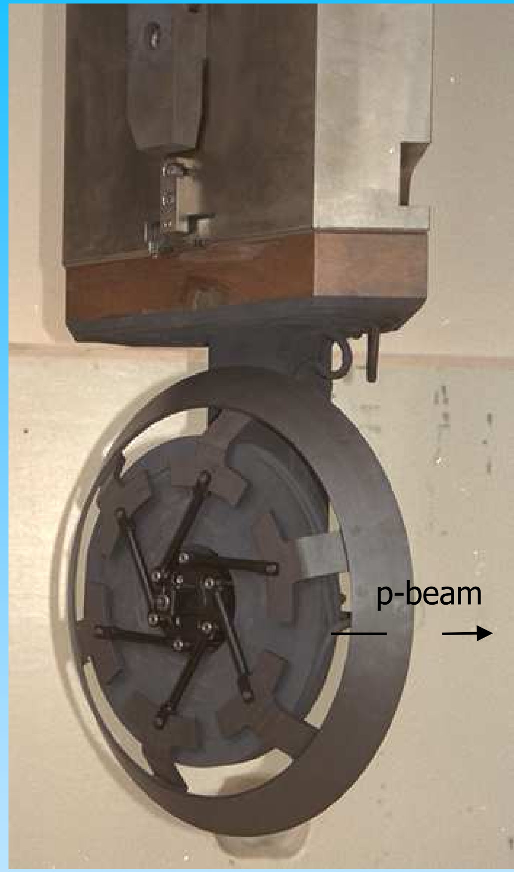
Graphite targets

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		$6.7 \cdot 10^{16}$	1.1	-
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	$8.4 \cdot 10^{10}$



- 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

now available: around 80µg; more can be prepared

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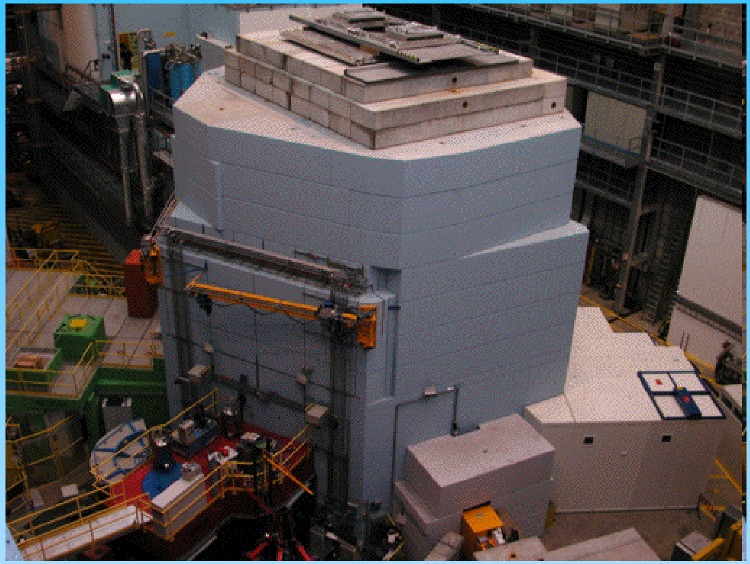
SINQ CoolingWater



- ^{7}Be (477.6 keV, 53.3 days) Produced by Spallation Reactions on ^{16}O in the Cooling Water (D_2O) of SINQ
- No by-products - high specific activity of ^{7}Be
- Filter Device with Mixed-bed Ion Exchanger LEWATIT
- Estimated Activity ~ TBq range (5.10^{18} atoms)
- Dose Rate ~ 1 Sv/h
- Impurities – ^{22}Na (2.6 y), ^{75}Zn (244 d), ^{83}Rb (86 d), ^{88}Y (107 d), ^{54}Mn (313 d), ^{75}Se (120 d)

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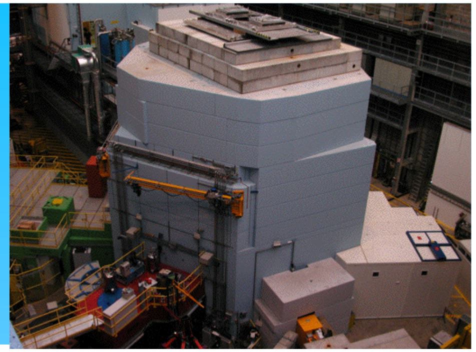
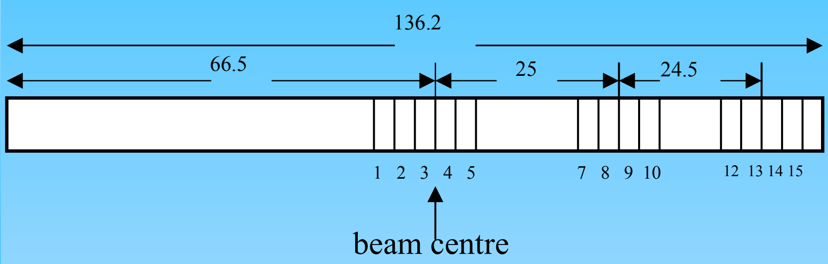
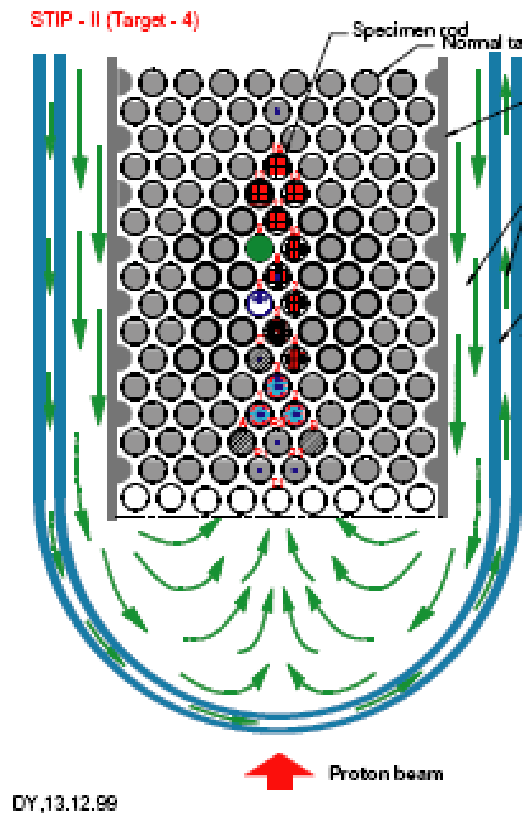
Lead targets from SINQ

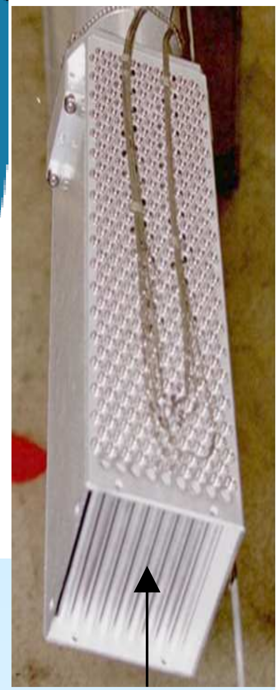


750 kW, solid target (lead)
2 Samples from target 4, 2 years operation; EOB 1999





	D9 [Bq/g]	D14 [Bq/g]		D9 [Bq/g]	D14 [Bq/g]
^{207}Bi	$3.00\cdot10^{7}$	$1.01\cdot10^{7}$	^{106}Ru	$4.83\cdot10^{6}$	$3.91\cdot10^{6}$
^{172}Lu/^{172}Hf	$2.00\cdot10^{7}$	$5.41\cdot10^{7}$	^{110m}Ag	$1.29\cdot10^{6}$	$3.93\cdot10^{5}$
^{173}Lu	$2.76\cdot10^{7}$	$4.30\cdot10^{7}$	^{125}Sb	$1.32\cdot10^{6}$	-
^{194}Au/^{194}Hg	$1.86\cdot10^{7}$	$3.13\cdot10^{6}$	^{133}Ba	$2.8\cdot10^{6}$	$7.94\cdot10^{5}$
^{102}Rh	$5.53\cdot10^{6}$	$1.44\cdot10^{5}$	^{44}Sc/^{44}Ti	$8.00\cdot10^{4}$	$2.84\cdot10^{4}$
^{202}Tl/^{202}Pb	$4.80\cdot10^{5}$	$1.87\cdot10^{5}$	^{108m}Ag	$3.75\cdot10^{5}$	$1.56\cdot10^{4}$
^{60}Co	$3.67\cdot10^{6}$	$1.40\cdot10^{6}$	^{194}Os/^{194m}Ir	$2.61\cdot10^{4}$	-
^{54}Mn	$2.29\cdot10^{5}$	$7.01\cdot10^{4}$	^{26}Al	0.5	0.2
^{58}Co	$1.55\cdot10^{6}$	$9.47\cdot10^{5}$	^{36}Cl	$9.5\cdot10^{1}$	$4.8\cdot10^{1}$
^{55}Fe	$8.73\cdot10^{7}$	$5.99\cdot10^{7}$	^{63}Ni	$6.30\cdot10^{8}$	$4.52\cdot10^{8}$



Proton beam

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Achievements

Re-measurement of the half-life of ^{60}Fe
Determination of the neutron capture cross section of ^{60}Fe
Development of a remote-controlled separation system for copper
Separation of ^{7}Be from the SINQ cooling water

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Re-measurement of the ^{60}Fe half-life

PSI – TU Munich collaboration

Literature:

~ $3 \cdot 10^{5}$ yrs (uncertain by a factor 3)
Roy and Kohman 1957

$1.49 \pm 0.27 \cdot 10^{6}$ yrs
Kutschera et.al. 1984

new value
G. Rugel et.al. PRL 103, 072502 (2009)

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Experimental requirements for the separation

$$T_{1/2\,^{60}Fe} = \frac{N_{^{60}Fe}}{A_{^{60}Fe}} \ln 2$$

A: ingrowth of ^{60}Co; N: ICP-MS

^{60}Fe ($1.5 \cdot 10^6$ 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

→ very good chemical separation from Co necessary

- ICP-MS can measure isotope ratios for the iron isotopes
- ICP-MS in principle possible, but interference with ^{60}Ni
- Correction with other Ni-isotopes not possible, because no natural isotope ratios (production of stable isotopes via spallation in the beam dump)

→ addition of stable Fe carrier necessary
→ addition of stable Ni carrier necessary

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Results

- $A(t=0)_{60Co}$ = 0.207±0.006 Bq (starting with ~ 50 MBq in the Cu-chips)
- A_{60Fe} = 49.19±0.11 Bq
- $m(Fe_{stable})$ = 2.6662±0.0009 mg (M=55.9020±0.0033 g/mol because of non-natural abundance)
- $I(N_{60Fe}/N_{Fe}) = 2.0483 \pm 0.0035 \cdot 10^{-4}$

$T_{1/2} = 2.62 \pm 0.04 \cdot 10^{6}$ years (1σ)

First determination of the neutron capture cross section at stellar energies

$^{60}Fe(n,\gamma)^{61}Fe$

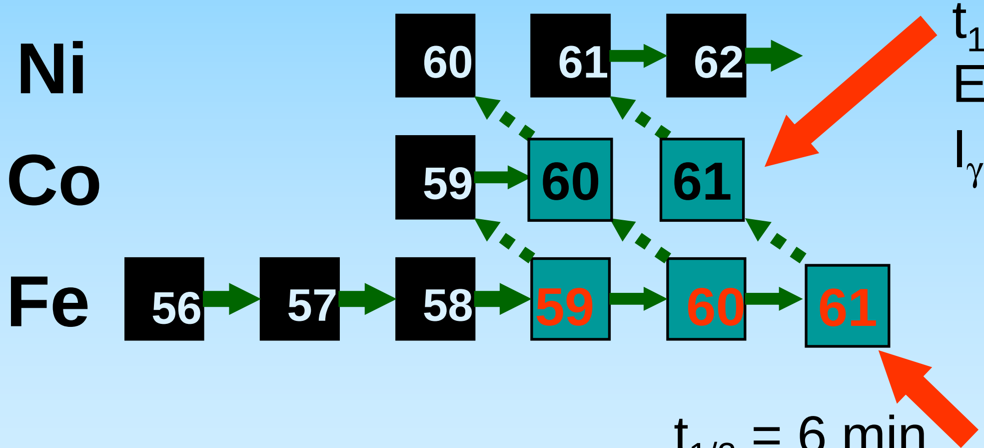
PSI – GSI – University of Notre Dame – FZK – collaboration

E. Uberseder et.al. PRL 102, 151101 (2009)

Experimental requirements for the separation

- **No stable carrier due to ^{59}Fe (44.5d) production (1099, 1292 keV)**
- **Measurement of the increase of the Co-daughter (determination of the number of ^{60}Fe-atoms)**
- ^{60}Fe (2.62·10^{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 ^{61}Fe production (1027/1205 keV)**



$t_{1/2}$ = 1.65 h
E_γ = 67, 909 keV
I_γ = 85, 4 %

$t_{1/2}$ = 6 min
E_γ = 298, 1027, 1205 keV
I_γ = 22, 43, 44(5)%

Slight contamination with ^{44}Ti (68, 78, 1157 keV)

→ very good chemical separation from Co and Ti necessary

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Result

- total number of ^{60}Fe atoms = $7.8 \cdot 10^{15}$ or 777ng (1.37 μg)
- total number of capture events = 118 (single) and 17 (coinc)
- time-integrated neutron flux = $1.7 \cdot 10^{14}$

$^{60}Fe(n, \gamma)^{61}Fe$ cross section @ kT = 25 keV

$$\langle\sigma\rangle = 10.2 \pm 2.9_{syst} \pm 1.4_{stat} \text{ mbarn}$$

5.8 mbarn (corrected with the new half-life)

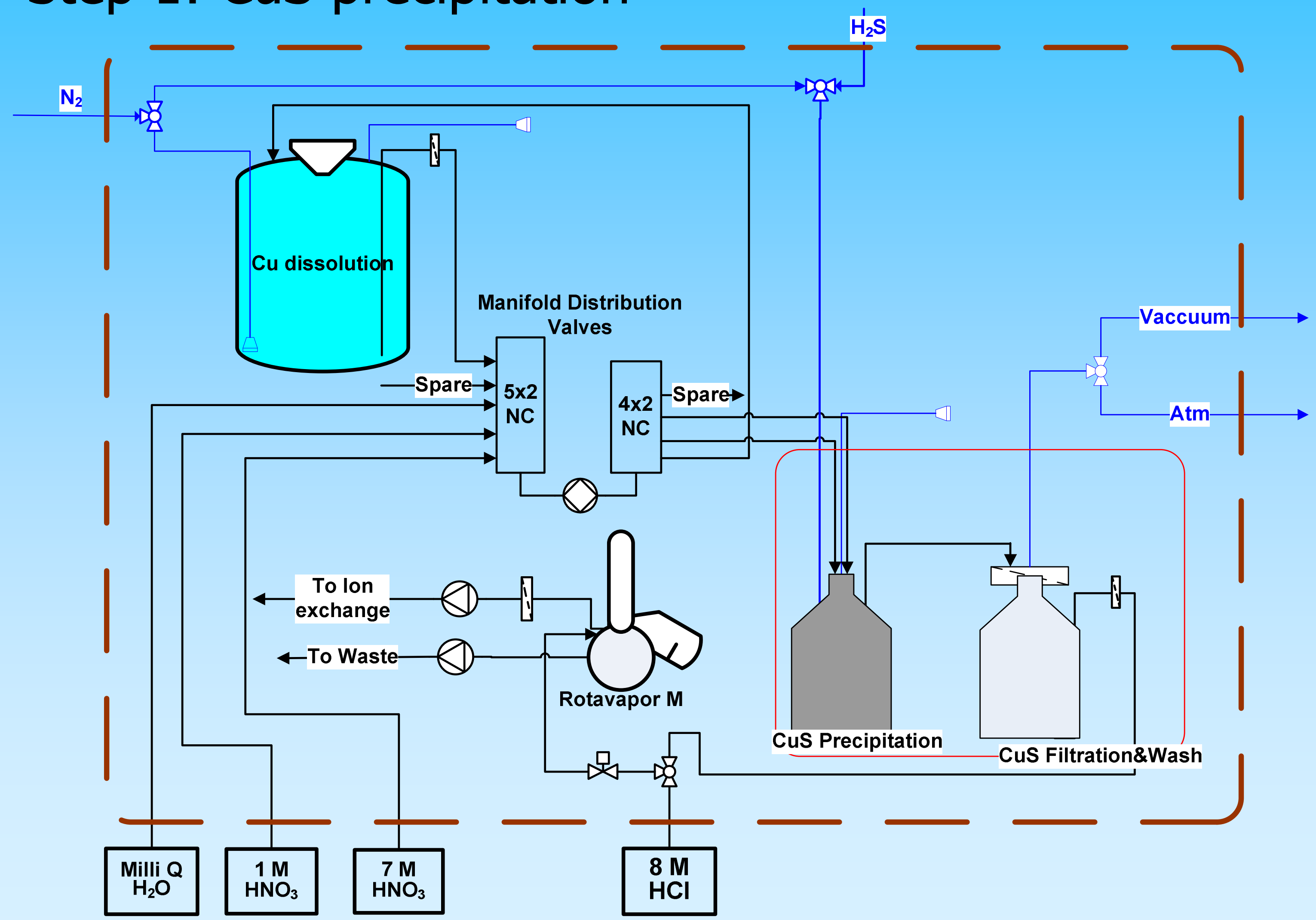
theoretical predictions between 1 and 20 mbarn

Development of a remote-controlled separation system for the copper beamdump

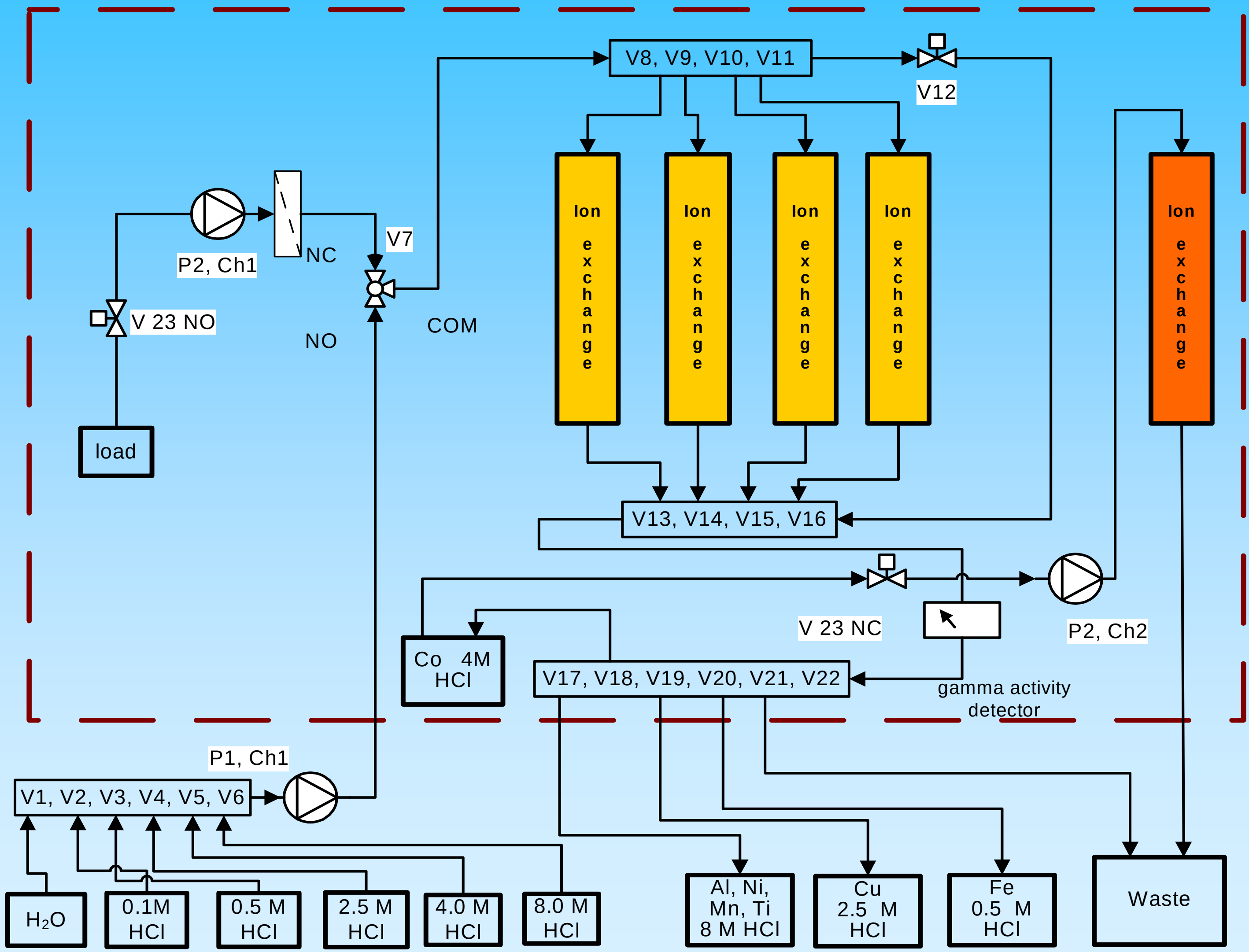
- high dose rate coming from ^{60}Co
- stepwise separation for all interesting isotopes
- applicable for Hotcell

Remote controlled system for copper separation

Step 1: CuS precipitation



Step 2: Ion exchange



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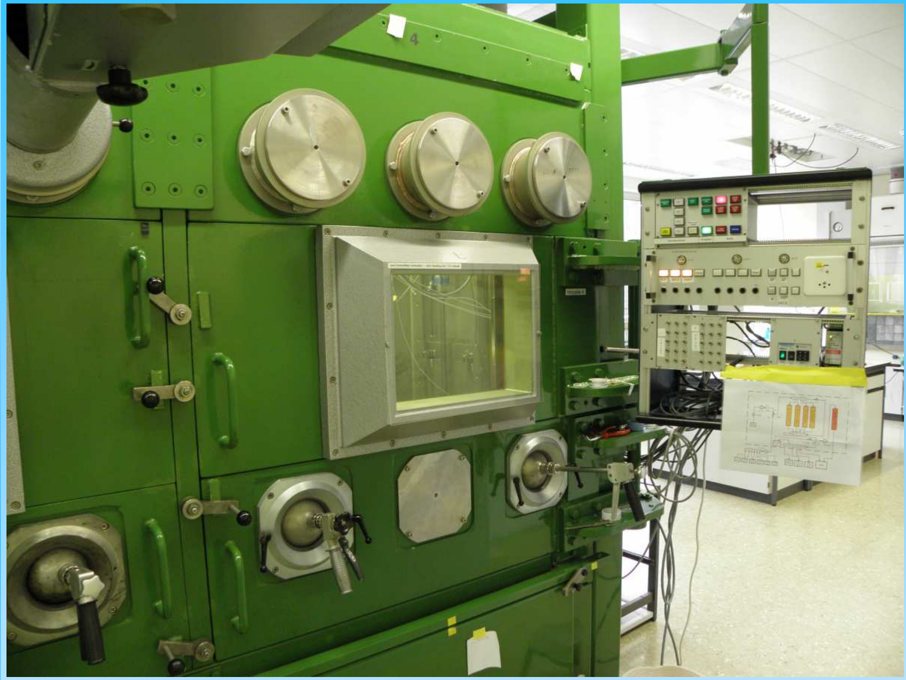
Hot-Cell Separation System



Overall Radiochemical Yield, %				
Al	Ni	Mn	Ti	Fe
45	53	65	70	72

results from model experiments
real separation soon

Separation of ^{7}Be from the SINQ cooling water

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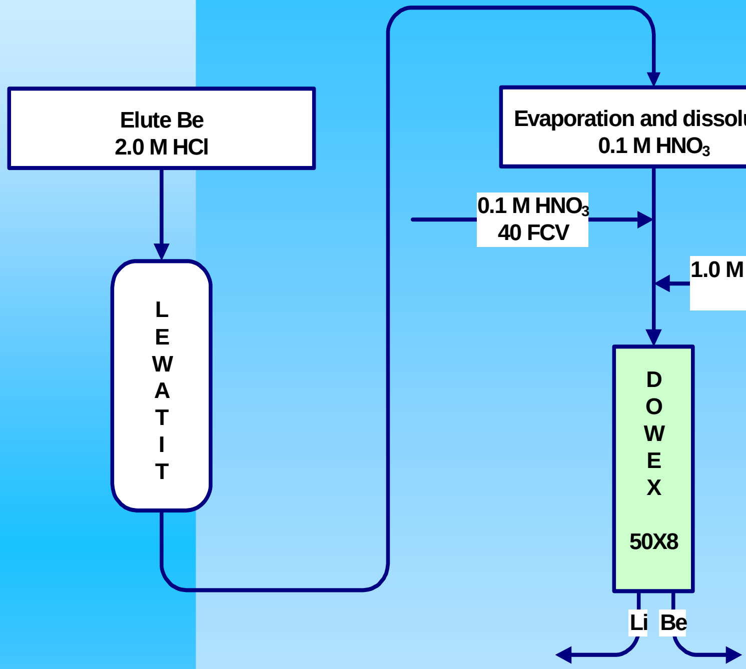
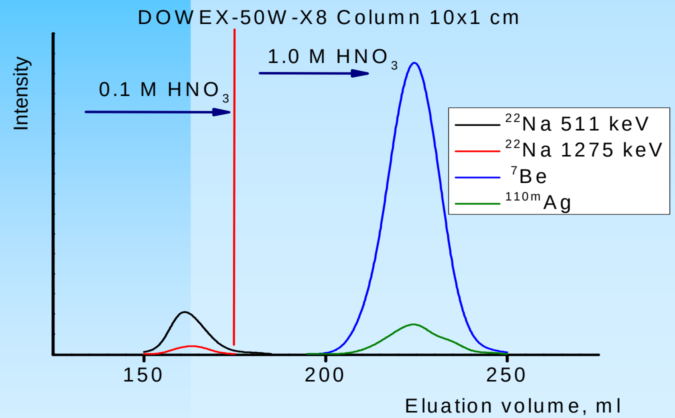
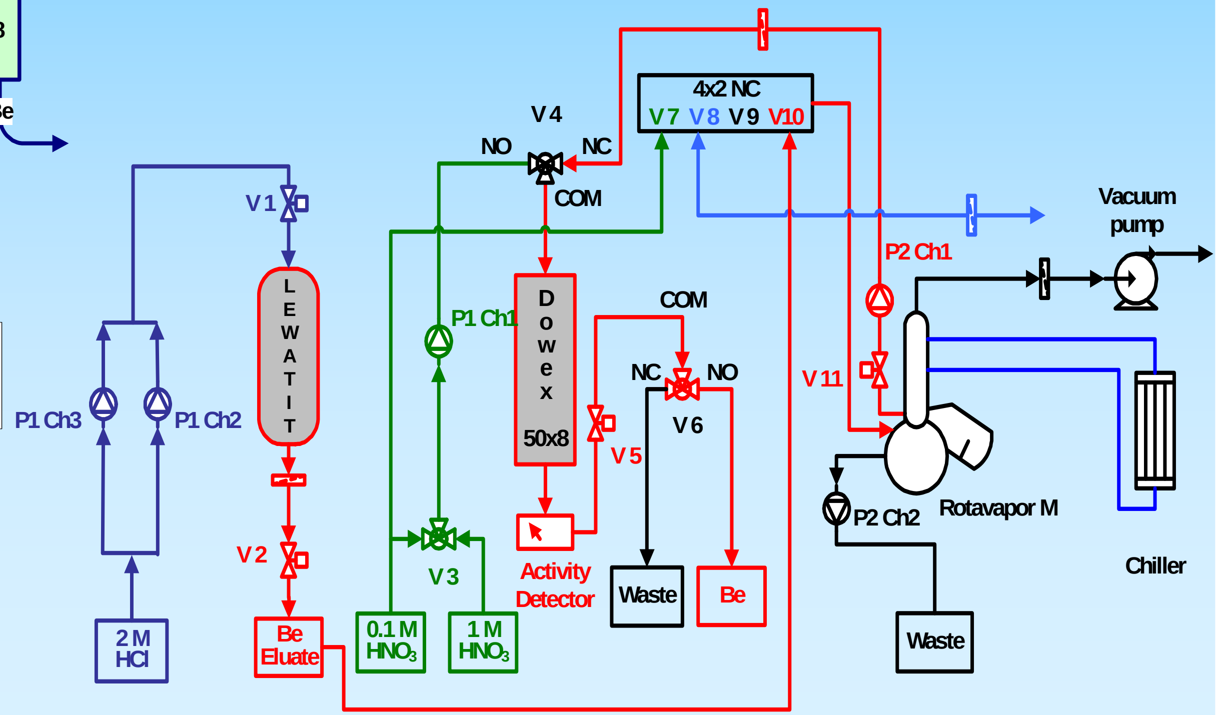
^{7}Be separation from the SINQ cooling water



^{7}Be - 5 GBq
^{22}Na - 400 MBq
^{54}Mn - 8 MBq
^{75}Se, ^{83}Rb, ^{75}Zn, ^{88}Y





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Future experiments

- Measurement of the $^{59}Fe(n,\gamma)^{60}Fe$ reaction:
  - Irradiation of enriched ^{58}Fe at ILL
  - Sample preparation ready
  - AMS-measurement of $^{60}Fe/^{58}Fe$ ratio at TUM ongoing
  - Second experiment at FRANZ (Frankfurt) at 25 keV
- Measurement of the low-energetic $^{60}Fe(n,\gamma)^{61}Fe$ cross section
  - New ^{60}Fe target prepared
  - Experiment at the reactor of TU Vienna ongoing
- Re-measurement of the half-life
  - Two identical samples
  - Similar measurement like the previous one at PSI ongoing
  - Re-measurement with AMS at University Vienna
- ^{10}Be from graphite targets/ ^{7}Be from SINQ cooling water
  - Half-life measurement of ^{10}Be
  - Study of halo nuclei properties (^{10}Be)
  - Studies of key reactions for evaluation of the solar neutrino flux (^{7}Be)
- Separation of ^{44}Ti from Cu and Fe targets
  - Development of a $^{44}Ti/^{44}Sc$ generator system for PET
  - For astrophysical studies: target and beam
- ^{53}Mn from Cu and Fe targets
  - Re-measurement of the half-life
  - Determination of neutron capture cross sections