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
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
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.
Characteristics of the copper beam dump

- Beam stop from the former BMA station
- 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
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<th>$^{36}\text{Cl}$ [kBq/g]</th>
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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)

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

All these radionuclides can be provided without carrier, but some of them contain other long-lived isotopes ($^{55}\text{Fe}/^{63}\text{Ni}$)
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
# Results for $^7/^{10}$Be

beam doses 4 – 11 Ah

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<tr>
<th>Sample</th>
<th>$^{10}$Be [Bq/g] ICP-MS</th>
<th>$^{10}$Be [Bq/g] AMS</th>
<th>Total amount of atoms</th>
<th>Total amount in μg</th>
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Lead targets from SINQ

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

- $^{172}_{77}$Lu (606.5 keV) 80 kBq/g
- $^{173}_{77}$Lu (202.2 keV) 310 MBq/g
- $^{207}_{83}$Bi (207.7 keV) 310 MBq/g
- $^{194}_{79}$Au (194 keV) 80 kBq/g
- $^{102}_{49}$Rh (205 keV) 102 MBq/g

- $^{60}_{28}$Co (1332.5 keV) 60 MBq/g
- $^{172/3}_{77}$Lu/Hf (1.9/1.4 y) 30 MBq/g
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, 5 μg, collaboration with UCL
- $^{10}$Be, carrier-free, laser spectrometry, $10^{13}$ atoms, collaboration with GSI
Special Problem:

\[ ^{60}\text{Fe} \ (1.5 \cdot 10^6 \text{ y}) \xrightarrow{\beta^-} ^{60}\text{mCo} \ (10.5 \text{ min}) \xrightarrow{\gamma} ^{60}\text{Co} \ (5.3 \text{ y}) \xrightarrow{\gamma, \beta^-} ^{60}\text{Ni} \ \text{(stable)} \]

\(^{60}\text{Fe}: \text{no } \gamma \text{ radiation, low } \beta^- \text{ energy}\)

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: \(\sim 10^{15}\) \(^{60}\text{Fe}\) atoms, decontamination factor (Co) < 10\(^{-7}\)
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

Dissolution in 7 M HCl

DOWEX1x8 Adsorption of Fe
DOWEX1x8 Adsorption of Ti
Evaporation to dryness
Fuming with conc. HF
Dissolution in 1 M HF

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

Cu$(\text{NH}_3)_4^{2+}$/Co$(\text{NH}_3)_4^{2+}$ (40-60%)

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

Dissolution
In 7 M HCl

$\text{Fe(OH)}_3 + \text{Be(OH)}_2 \xrightarrow{\text{Filtration}} + \text{Fe}^{3+}, +\text{NH}_3$

DOWEX1x8
Adsorption of Fe
Elution of Be

DOWEX50x8
Adsorption of Be
Rinsing with 0.1 M HNO$_3$ (for removal of boron)
Elution of Be with 4 M HCl

Evaporation to dryness
Dissolution in 0.1 M HNO$_3$

Result:
$\sim 16 \mu g \ 10^{18}$ atoms
Summary and Outlook

- Cu- and C-samples available
- Work on Pb-targets ongoing
- $10^{15-17}$ atoms of several radionuclides ($^{26}\text{Al}$, $^{60}\text{Fe}$, $^{44}\text{Ti}$) separated and available
- Up to $10^{18}$ atoms of $^{10}\text{Be}$ 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