Outline

What do we have?
What can we do?
What did we do?
What do we want to do?
The ERAWAST project

Sources for isotopes at PSI
Processing material from outside

List of available isotopes and performed measurements

Examples for isotope and target preparation

$^{10}\text{Be}$ from graphite wheels
$^{91}\text{Nb}$ from irradiated $^{92}\text{Mo}$
$^{60}\text{Fe}$ from copper

Requirements for isotope and target preparation

Summary
Objective:
Exploitation of accelerator waste for isolating rare exotic radionuclides

History:
- Radiochemical analytics of activated components for disposal
- Results showed high content of several rare isotopes
- Looking for potential users of these isotopes: I. ERAWAST workshop 2006 (PSI), funded by ESF
- Five-years working program
- II. ERAWAST workshop 2011 at PSI: first results and future program
- CHANDA-workshop in 2015
- ~ 20 Partners
- Member of n_TOF

Collaboration between
Nuclide production facilities
Basic nuclear physics research
Nuclear astrophysics
AMS measurement groups
Environmental chemistry
All elements of periodic table with $Z \leq Z_{\text{target}} + 1$

- 590 MeV protons
- 2.4 mA beam current
- High activation of shieldings, targets, structure material
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
Copper beam dump
- $^{44}$Ti, $^{53}$Mn, $^{26}$Al, $^{60}$Fe
- $^{60}$Co – 5 GBq

Myon production station
- Operation 1-3 years
- Beam doses 4 – 11 Ah
- Source for $^{10}$Be

SINQ Target Irradiation Program-STIP
- $^{44}$Ti, $^{53}$Mn, $^{26}$Al, Lanthanides

SINQ cooling water
- $^7$Be, $^{54}$Mn, $^{22}$Na, $^{88}$Y

Special irradiation positions with 590 MeV protons
V for $^{32}$Si production

Useful components
## Target manufacturing and measurements finished

- $^{63}\text{Ni} \ @ \ n\text{-TOF}$  
  10 mg  
  neutron capture cross section
- $^{171}\text{Tm} \ @ \ n\text{-TOF}, \ SARAF, \ Mainz$  
  3 mg  
  neutron capture cross section
- $^{147}\text{Pm} \ @ \ n\text{-TOF}$  
  72 $\mu$g  
  neutron capture cross section

## Isotope separation performed, samples ready for use

- $^{163}\text{Ho}@\text{HOLMES}$  
  1.5 mg  
  neutrino mass measurement
- $^{91}\text{Nb}@\text{FRANZ}$  
  1 $\mu$g  
  neutron capture cross section

## Isotope production planned

- $^{79}\text{Se}@n\text{-TOF}$  
  ?  
  neutron capture cross section
### Isotopes produced at PSI

#### Isotope production and measurements finished

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Mass (μg or mg)</th>
<th>Measurements</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{60}$Fe</td>
<td>1</td>
<td>half-life and neutron capture cross sections, @several</td>
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<tr>
<td>$^{44}$Ti</td>
<td>30</td>
<td>$^{44}$Ti($\alpha$,p) reaction, @ISOLDE/Uni Edinburgh</td>
</tr>
<tr>
<td>$^{7}$Be</td>
<td>15</td>
<td>$^{7}$Be(n,$\alpha$) and $^{7}$Be(n,p), @n_TOF</td>
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<td>$^{10}$Be</td>
<td>4</td>
<td>$^{10}$Be(n,γ) thermal, @Uni Mainz</td>
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</table>

#### Isotope production performed, isotopes ready for use

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Mass (μg or mg)</th>
<th>Measurements</th>
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<tr>
<td>$^{26}$Al</td>
<td>0.4</td>
<td>half-life measurement</td>
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<tr>
<td>$^{53}$Mn</td>
<td>3</td>
<td>half-life and neutron capture cross sections</td>
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#### Isotope production planned

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Mass (μg or mg)</th>
<th>Measurements</th>
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<tbody>
<tr>
<td>$^{32}$Si</td>
<td>60</td>
<td>half-life and cross section measurements</td>
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<tr>
<td>$^{146}$Sm</td>
<td>100</td>
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<tr>
<td>$^{148}$Gd</td>
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<tr>
<td>$^{154}$Dy</td>
<td>25</td>
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<tr>
<td>$^{209}$Po</td>
<td>?</td>
<td>half-life measurements</td>
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<tr>
<td>Actinides and fission products from spent nuclear fuel solutions</td>
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</table>
$^{10}$Be from target-E graphite targets

Myon production station consumes up to 20% of the proton beam

Typical operation time: 1-3 years

Source for $^7$Be and $^{10}$Be
Chemical separation

Main radioactive components: $^3$H, $^{14}$C

Combustion of graphite in oxygen stream

Tube furnace @ 1000°C

$^3$H$_2$O capture efficiency > 99.99%

35 kBq $^{10}$Be on Carbon pellet
Spallation products in carbon by mass:

Remaining oxidation products:

Li$_2$O, BeO, B$_2$O$_3$

totally approx. 1200 ppmm

14 µg $^{10}$Be per g graphite

currently available: ca. 4 mg $^{10}$Be

data by courtesy of Dr. D. Kiselev
\[ ^{91}\text{Nb} \] – from proton-irradiated \[ ^{92}\text{Mo} \]

\[ ^{91}\text{Nb}(p,\gamma)^{92}\text{Mo} \] reaction at 2 MeV proton energy

Relevant nuclear reaction for p-process

production of the most abundant p nucleus \[ ^{92}\text{Mo} \]

Cross sections from TALYS

\[ 10^{16} \] \[ ^{91}\text{Nb} \] nuclei with cyclotron at PTB
Braunschweig in about 7 days possible
Radiochemical separation

Development of the chemical system using model tracers

\[
\begin{align*}
\text{99Mo-foil (95Nb)} & \quad + 2 \text{ mg La} \quad + \text{HNO}_3/\text{HCl} \\
& \quad \text{Dissolution} \quad \text{Evaporation to dryness} \\
& \quad \downarrow \quad + 1 \text{ M HCl} \\
\text{99MoO}_4^{2-} & \quad + 1 \text{ M NH}_3 \\
& \quad \text{La(OH)$_3$
95NbO(OH)$_3$} \quad + 1 \text{ M HF} \\
& \quad \downarrow \quad \text{Dissolution} \\
\text{LaF}_3 & \quad \text{95NbOF}_5^{2-}
\end{align*}
\]

Sample preparation:
Irradiation of 971.3 mg 92Mo with 20 MeV protons
Determination of the decontamination factor from 92Mo by ICP-MS: > 10^5; ~ 1 \mu g 91Nb
The discovery of $^{60}$Fe from a nearby supernova explosion ~ 2.5 million years ago

Nature 2016:
- Investigation of ocean floor samples with AMS

Science 2016:
- Mass spectrometry with CRIS

LETTER
- Transport calculations

ASTROPHYSICS
- Observation of the $^{60}$Fe nucleosynthesis-clock isotope in galactic cosmic rays

PRL 116, 151104 (2016)

The locations of recent supernovae near the Sun from modelling $^{60}$Fe transport

LETTER
- Samples from the Apollo missions measured by AMS

Nature 2016:
- Radioactive iron rain: transporting $^{60}$Fe in supernova dust to the ocean floor
Separation and preparation

Dissolution of Cu chips (3 g) in 7 M HNO₃

Evaporation to dryness

Dissolution in 7 M HCl + 5 mg Co²⁺ as carrier

Extraction with methylisobutylketone

Aqueous phase:
Ni, Co, Cu,
organic phase: Fe

Back extraction with 0.1 M HCl,
repetition of procedure

Result: 7.8 \times 10^{15} \text{ or } 777 \text{ ng } ^{60}\text{Fe atoms,}
decontamination factor (Co) > 10^8
(0.3 \text{ Bq})

Evaporation of the final solution onto a graphite backing
60Fe - summary

- Sample preparation for 4 half-life determinations
- Target preparation for 2 neutron capture cross section experiments (thermal energy and 25 keV)
- Preparation of standard material for AMS measurement

All sample-requiring experiments on 60Fe world-wide within the last 20 years work with material produced at PSI

- New measurement of the 60Fe half-life, G. Rugel et.al. PRL 2009
- Determination of the neutron capture cross section at stellar energies, E. Ueberseder et.al. PRL 2009
- Quantification of 60Fe atoms by MC-ICP-MS for the redetermination of the half-life, N. Kivel et.al. ABC 2013
- The thermal neutron capture cross section of the radioactive isotope 60Fe, T. Heftrich et.al., PRC 2015
- Settling the half-life of 60Fe – fundamental for a versatile astrophysical chronometer, A. Wallner et.al., PRL 2015
- Activity measurement of 60Fe through the decay of 60mCo and confirmation of its half-life, K. Ostiek et.al. PRC 2017
- Nuclear properties of 60Fe, R. Dressler et.al., currently ongoing
G. Rugel, et al.: 
Requirements for sample and target preparation

**Isotope production and separation**
- 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?

**Target preparation**
- Self-supporting or with backing?
- Which backing?
- Thickness of the backing to be known?
- Isotope composition required?
- Distribution to be known?
- Thickness measurement required?
- Single or multi-use?
- Radioprotection issues
- Transport issues

Collaboration on the basis of a material transfer agreement (MTA)
Summary and conclusions

• Exotic radionuclides are produced in components of the 590 MeV proton accelerator at PSI

• After chemical separation, these isotopes are available for scientific applications
  – Nuclear astrophysics
  – Geoscience
  – Basic nuclear physics
  – AMS standards

• PSI owns a store house of several very rare isotopes, some of them being unique world-wide in quality and quantity ($^7$/$^{10}$Be, $^{60}$Fe, $^{53}$Mn, $^{44}$Ti and others)

• Examples for front-end experiments using targets made by PSI
  – $^{60}$Fe half-life and neutron capture cross section measurements

• Examples for ongoing experiments ($^{10}$Be, $^{91}$Nb)

• We need a network on target preparation

• We need a dedicated mass separation device for exotic radionuclides!
My thanks go to

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  – n_TOF
  – Uni Frankfurt
  – and many others
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