



## Outline

- What do we have?
- What can we do?
- What did we do?
- What do we want to do?

Dorothea Schumann, Rugard Dressler

# Radioactive isotope and target production for nuclear astrophysics

Nuclear Astrophysics at the Dresden Felsenkeller, 26.-28.06.2017

## **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**

(Exotic Radionuclides from Accelerator Waste for Science and Technology)

## 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

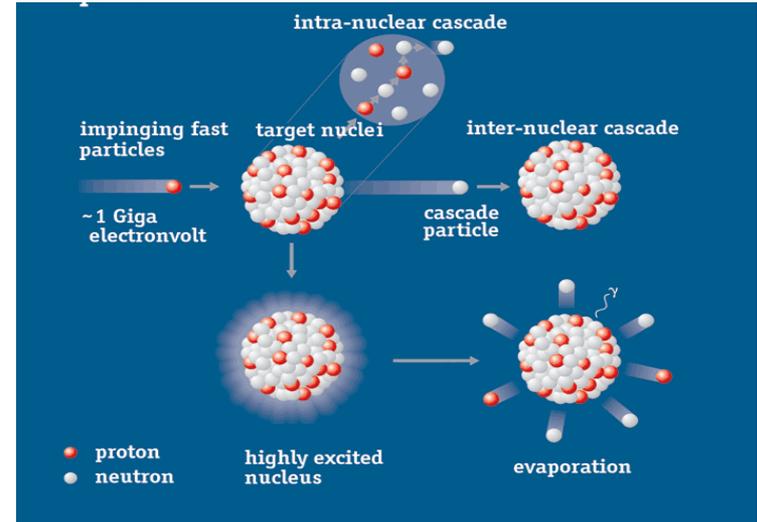
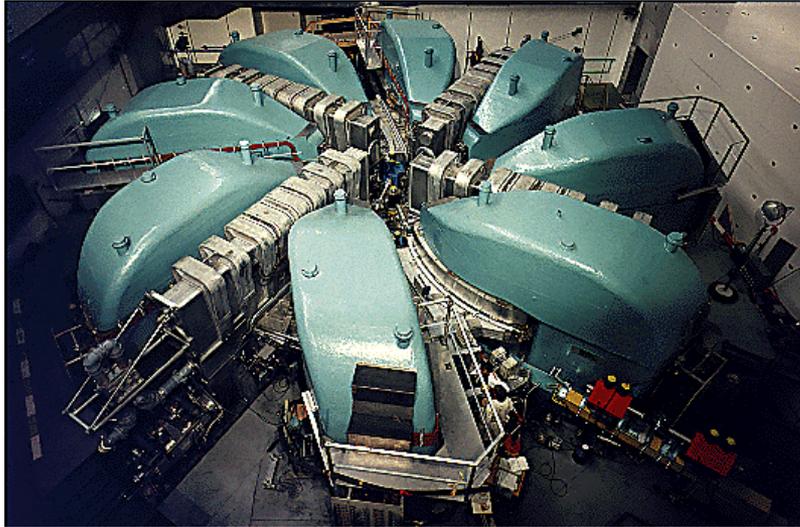
Basic nuclear physics research

Nuclear astrophysics

AMS measurement groups

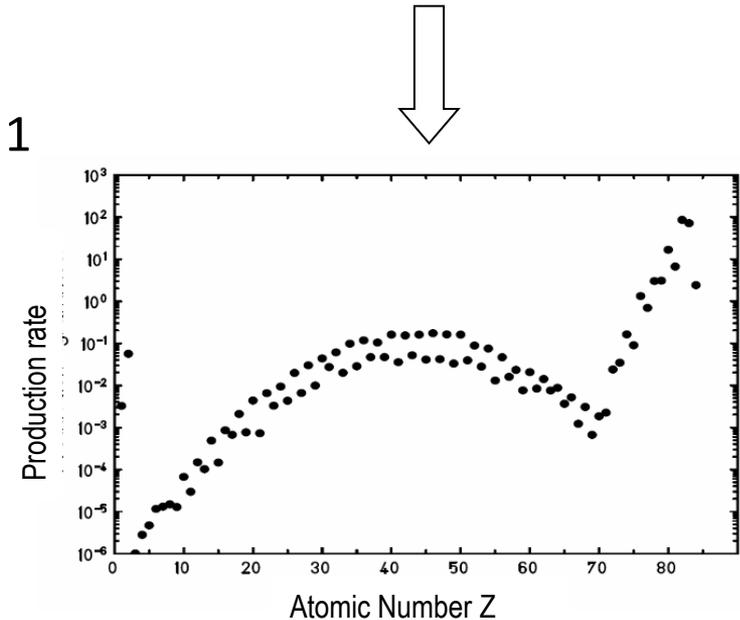
Environmental chemistry

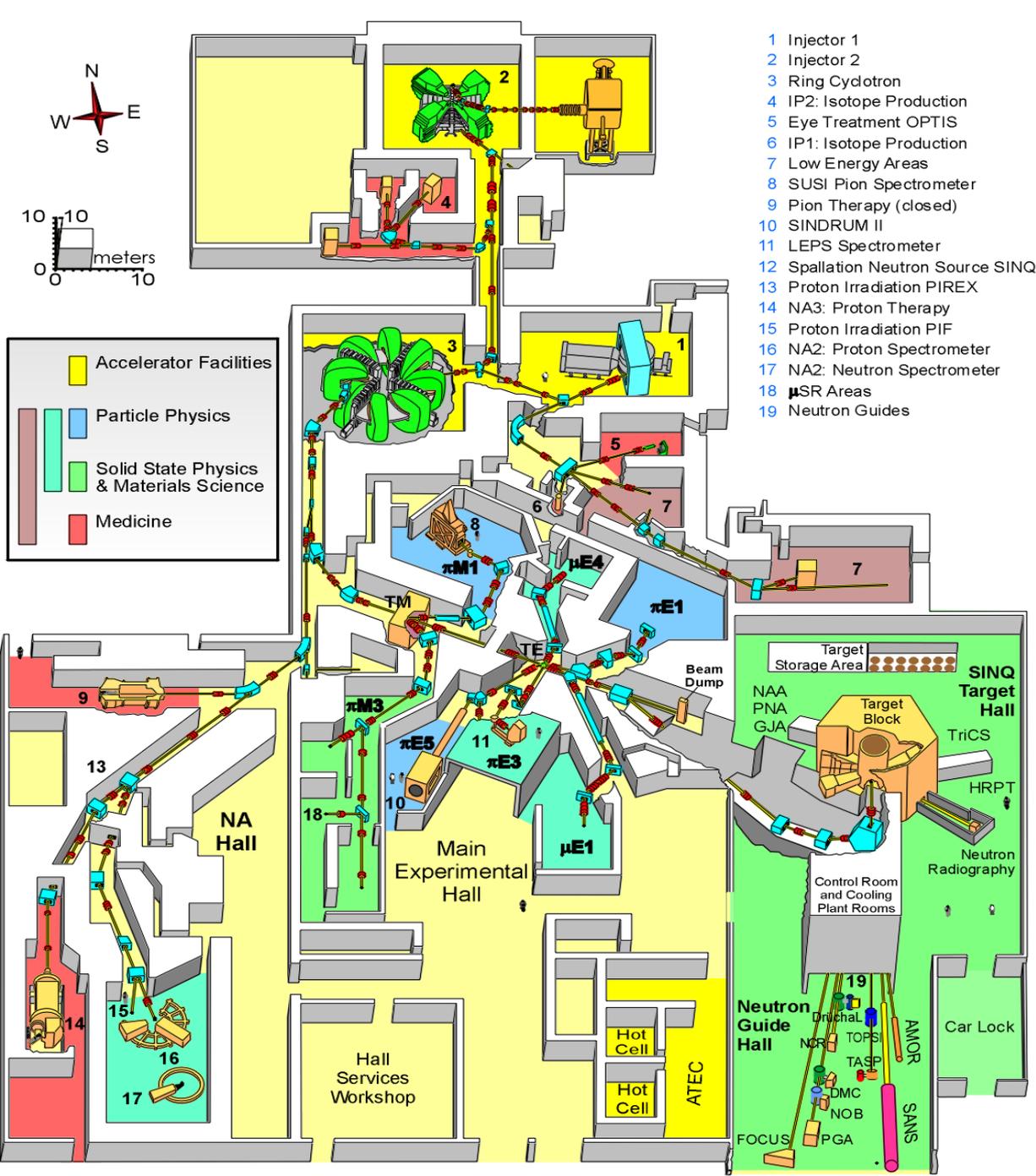
# Spallation Reactions and PSI facilities



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





- 1 Injector 1
- 2 Injector 2
- 3 Ring Cyclotron
- 4 IP2: Isotope Production
- 5 Eye Treatment OPTIS
- 6 IP1: Isotope Production
- 7 Low Energy Areas
- 8 SUSI Pion Spectrometer
- 9 Pion Therapy (closed)
- 10 SINDRUM II
- 11 LEPS Spectrometer
- 12 Spallation Neutron Source SINQ
- 13 Proton Irradiation PIREX
- 14 NA3: Proton Therapy
- 15 Proton Irradiation PIF
- 16 NA2: Proton Spectrometer
- 17 NA2: Neutron Spectrometer
- 18  $\mu$ SR Areas
- 19 Neutron Guides

# 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

# „Useful“ components

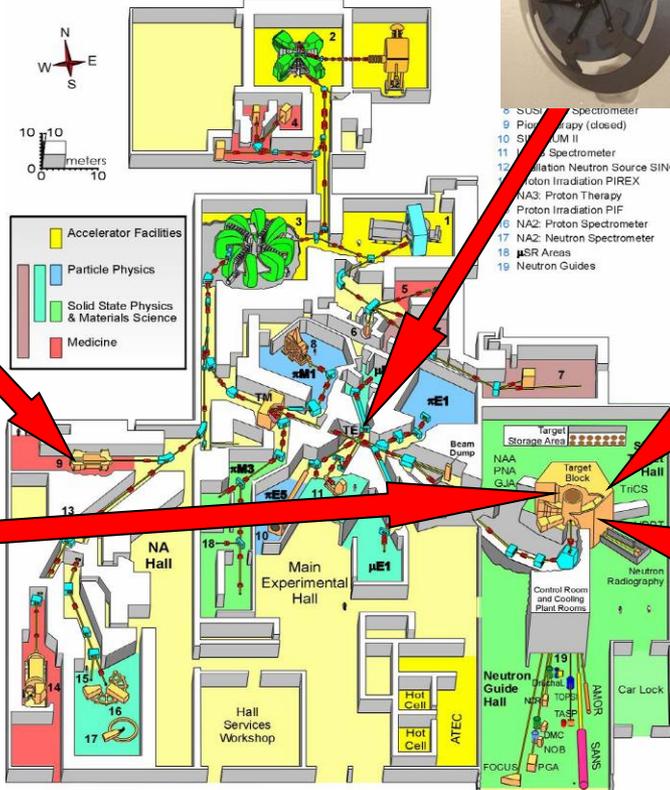
## Copper beam dump

- $^{44}\text{Ti}$ ,  $^{53}\text{Mn}$ ,  $^{26}\text{Al}$ ,  $^{60}\text{Fe}$
- $^{60}\text{Co}$  – 5 GBq



## Myon production station

- Operation 1-3 years
- Beam doses 4 – 11 Ah
- Source for  $^{10}\text{Be}$



## SINQ Target Irradiation Program-STIP

$^{44}\text{Ti}$ ,  $^{53}\text{Mn}$ ,  $^{26}\text{Al}$ , Lanthanides

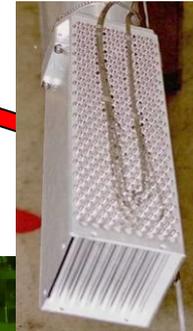


## SINQ cooling water

$^7\text{Be}$ ,  $^{54}\text{Mn}$ ,  $^{22}\text{Na}$ ,  $^{88}\text{Y}$

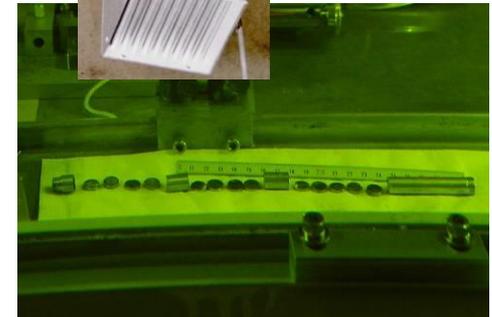
## SINQ target

- $^{207}\text{Bi}$ ,  $^{172}\text{Hf}$ ,
- $^{173}\text{Lu}$ ,  $^{194}\text{Hg}$ ,
- $^{202}\text{Pb}$ ,  $^{125}\text{Sb}$ ,
- $^{106}\text{Ru}$ ,  $^{44}\text{Ti}$



## Special irradiation positions with 590 MeV protons

V for  $^{32}\text{Si}$  production



## Target manufacturing and measurements finished

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

## Isotope separation performed, samples ready for use

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

## Isotope production planned

- |                           |   |                               |
|---------------------------|---|-------------------------------|
| • $^{79}\text{Se}$ @n_TOF | ? | neutron capture cross section |
|---------------------------|---|-------------------------------|

# Isotopes produced at PSI

## Isotope production and measurements finished

- $^{60}\text{Fe}$  1  $\mu\text{g}$  half-life and neutron capture cross sections, @several
- $^{44}\text{Ti}$  30  $\mu\text{g}$   $^{44}\text{Ti}(\alpha, p)$  reaction, @ISOLDE/Uni Edinburgh
- $^7\text{Be}$  15  $\mu\text{g}$   $^7\text{Be}(n, \alpha)$  and  $^7\text{Be}(n, p)$ , @n\_TOF
- $^{10}\text{Be}$  4 mg  $^{10}\text{Be}(n, \gamma)$  thermal, @Uni Mainz

## Isotope production performed, isotopes ready for use

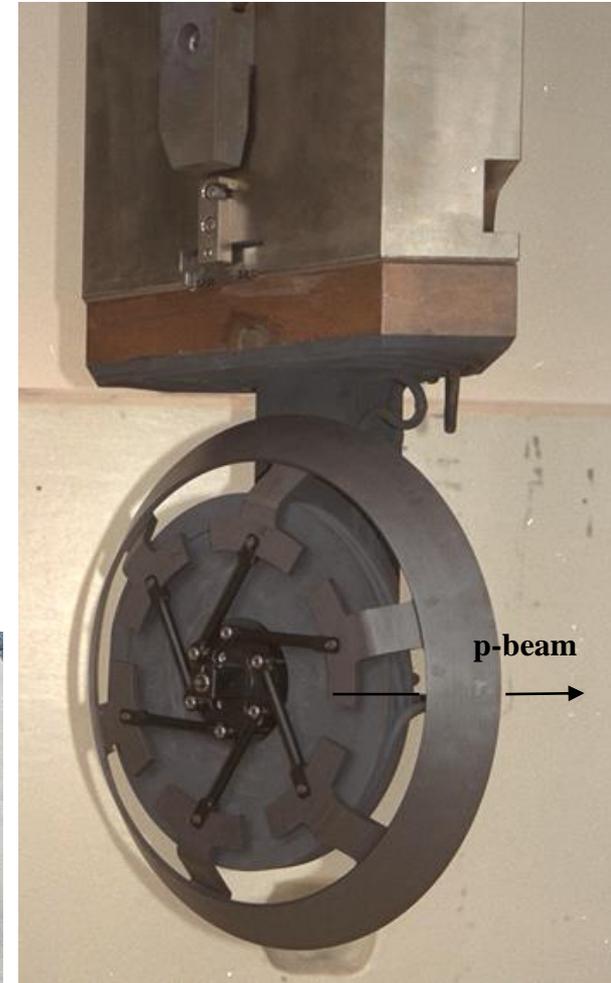
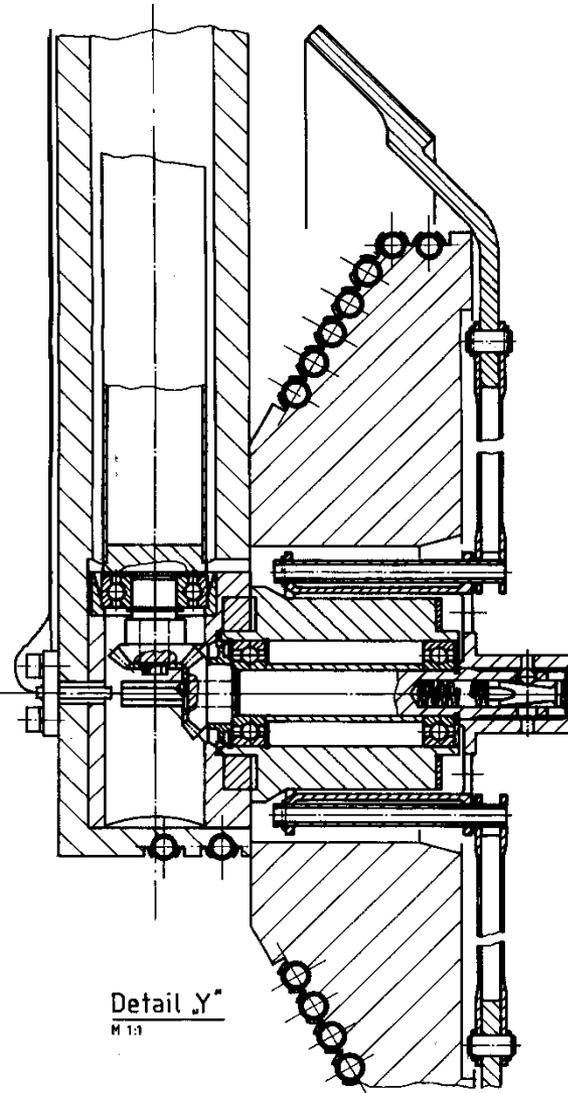
- $^{26}\text{Al}$  0.4  $\mu\text{g}$  half-life measurement
- $^{53}\text{Mn}$  3 mg half-life and neutron capture cross sections

## Isotope production planned

- $^{32}\text{Si}$  60  $\mu\text{g}$  half-life and cross section measurements
- $^{146}\text{Sm}$  100  $\mu\text{g}$  half-life measurements
- $^{148}\text{Gd}$  80  $\mu\text{g}$  half-life measurements
- $^{154}\text{Dy}$  25  $\mu\text{g}$  half-life measurements
- $^{209}\text{Po}$  ? half-life measurements
- Actinides and fission products from spent nuclear fuel solutions

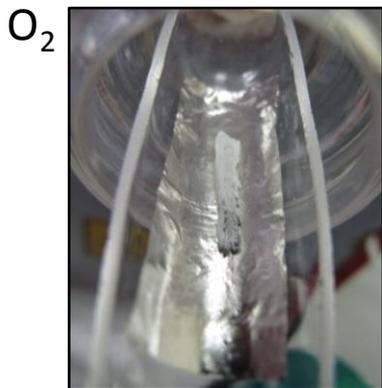
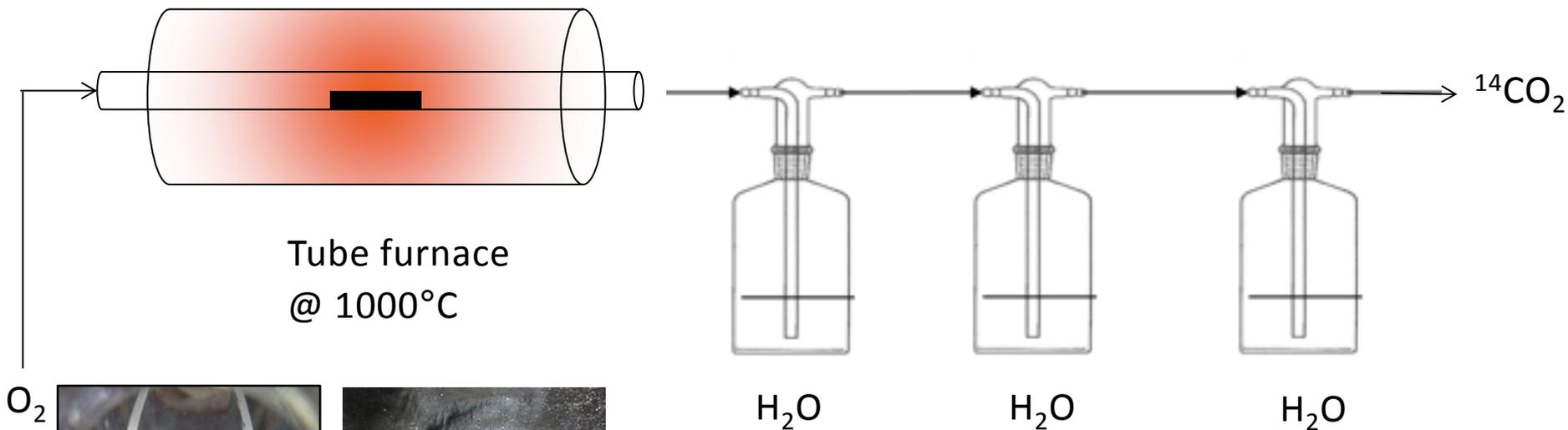
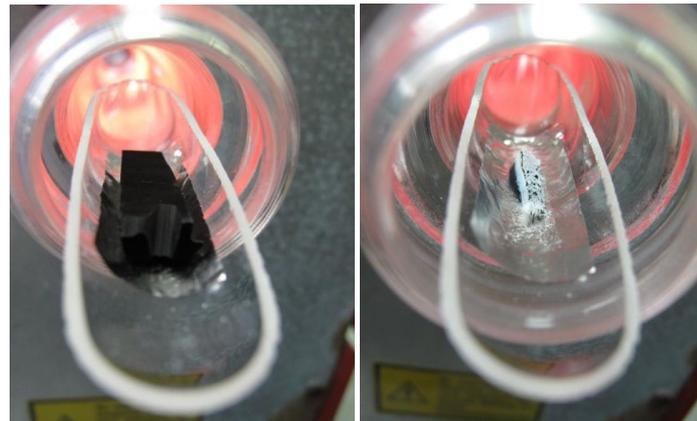
# $^{10}\text{Be}$ from target-E graphite targets

Myon production station  
consumes up to 20% of the  
proton beam  
Typical operation time: 1-3  
years  
Source for  $^7\text{Be}$  and  $^{10}\text{Be}$



# Chemical separation

Main radioactive components:  $^3\text{H}$ ,  $^{14}\text{C}$   
Combustion of graphite in oxygen stream



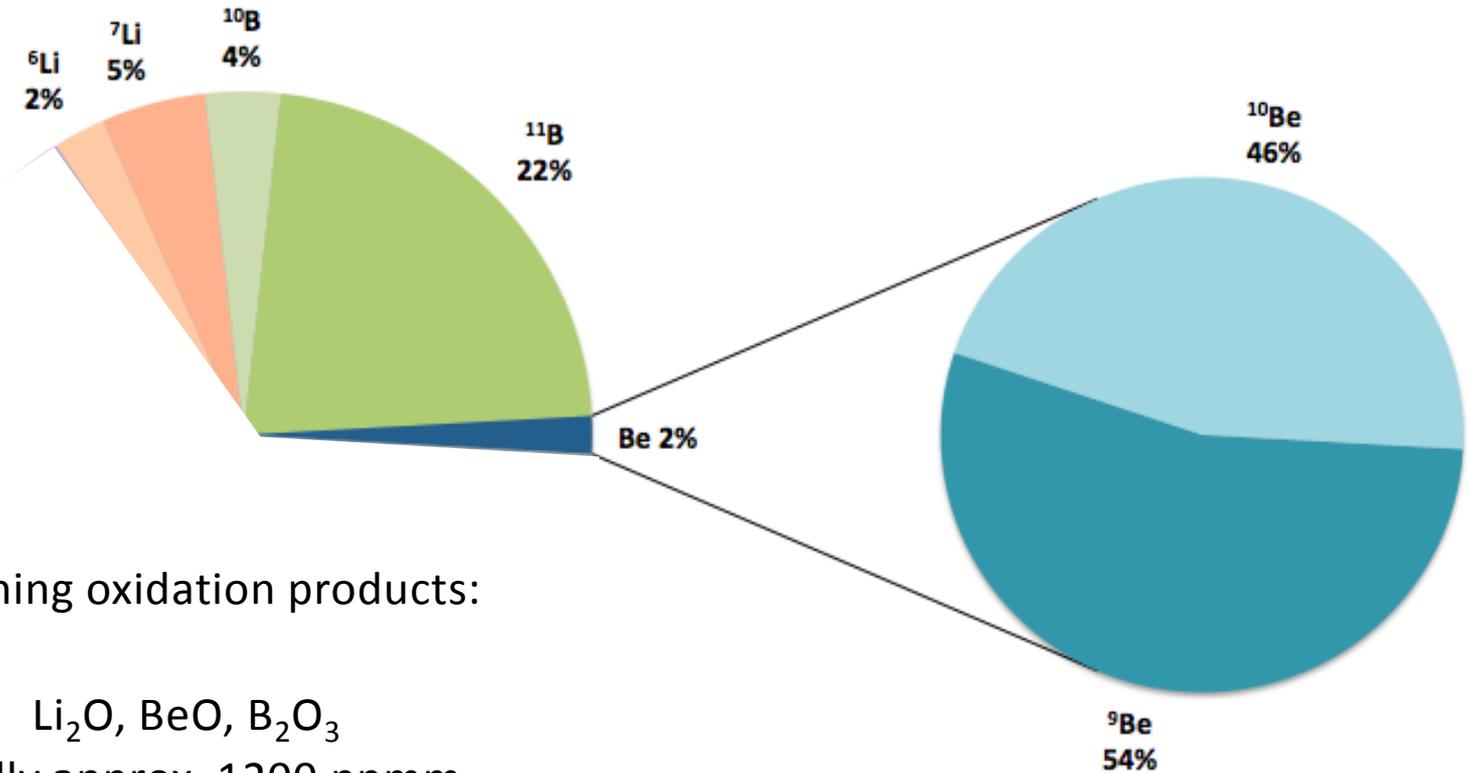
$^3\text{H}_2\text{O}$  capture efficiency > 99.99%

35 kBq  $^{10}\text{Be}$  on Carbon pellet



# MCNPX calculation for target E92

Spallation products in carbon by mass:



Remaining oxidation products:



totally approx. 1200 ppm

14  $\mu\text{g}$   ${}^{10}\text{Be}$  per g graphite

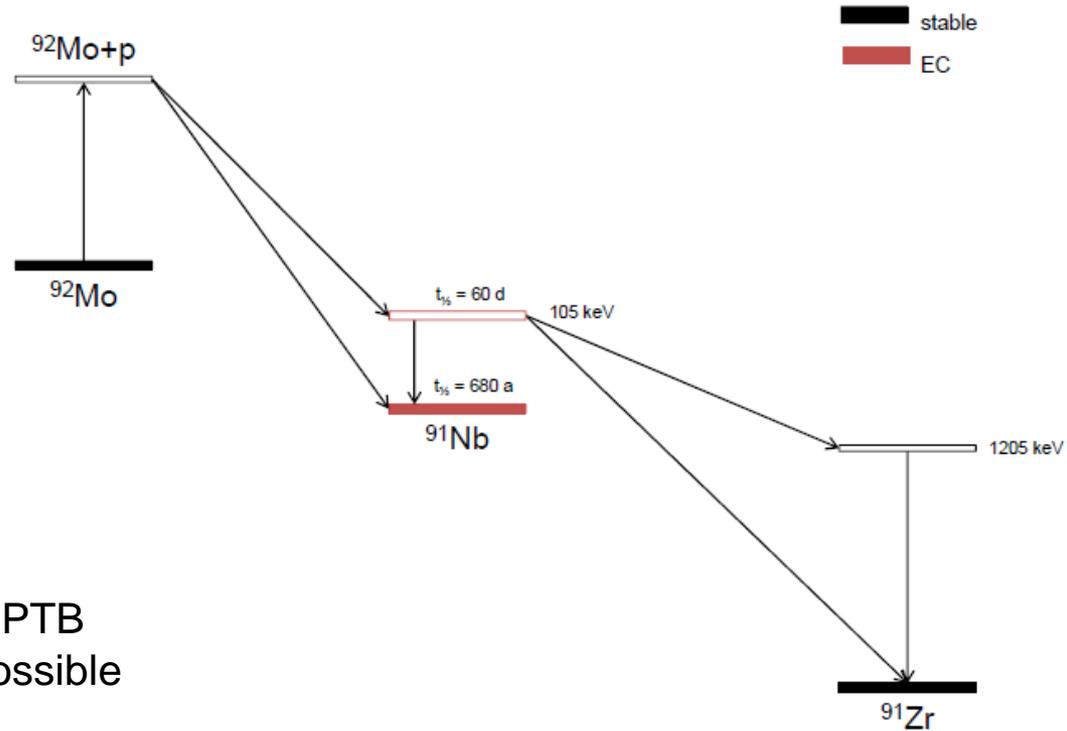
currently available: ca. 4 mg  ${}^{10}\text{Be}$

# <sup>91</sup>Nb – from proton-irradiated <sup>92</sup>Mo

<sup>91</sup>Nb(p,γ)<sup>92</sup>Mo reaction at 2 MeV proton energy

Relevant nuclear reaction for p-process

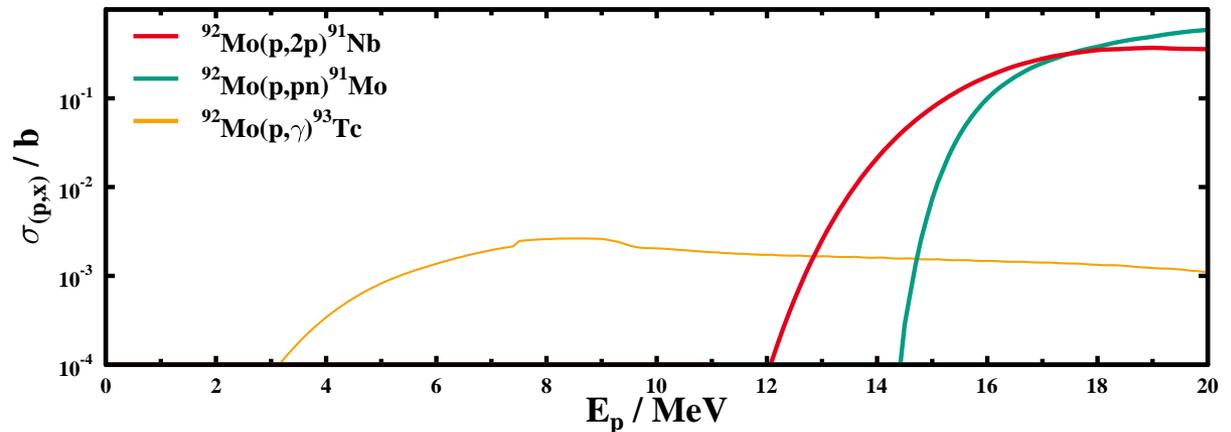
production of the most abundant p nucleus <sup>92</sup>Mo



Cross sections from TALYS

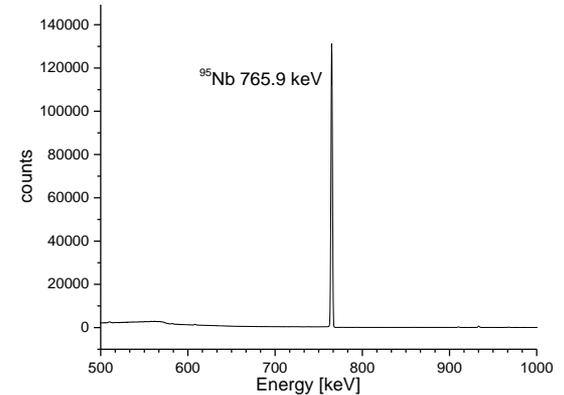
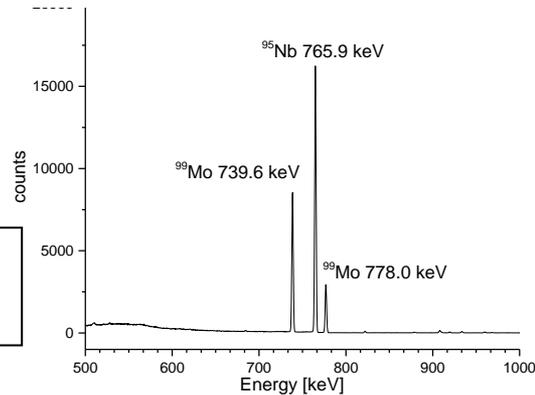
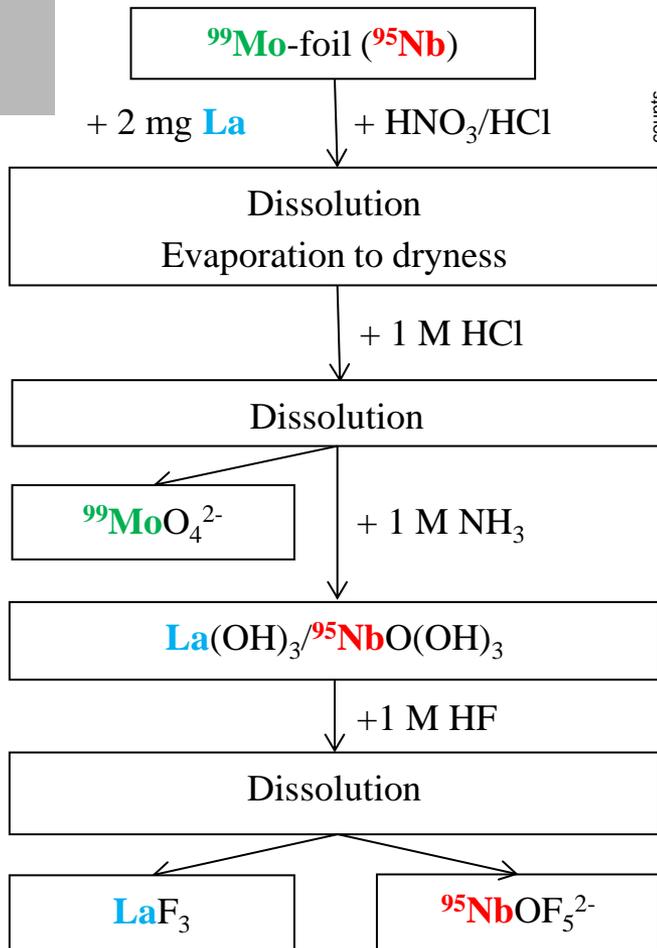
10<sup>16</sup> <sup>91</sup>Nb nuclei with cyclotron at PTB Braunschweig in about 7 days possible

<sup>91</sup> Tc 3 m	<sup>92</sup> Tc 4.4 m	<sup>93</sup> Tc 2.7 h	<sup>94</sup> Tc 4.9 h	<sup>95</sup> Tc 20 h
<sup>90</sup> Mo 5.7 h	<sup>91</sup> Mo 15.5 m	<sup>92</sup> Mo 14.84	<sup>93</sup> Mo 4*10 <sup>3</sup> a	<sup>94</sup> Mo 9.25
<sup>89</sup> Nb 2 h	<sup>90</sup> Nb 14.6 h	<sup>91</sup> Nb 680 a	<sup>92</sup> Nb 4*10 <sup>7</sup> a	<sup>93</sup> Nb 100
<sup>88</sup> Zr 83.4 d	<sup>89</sup> Zr 78.4 h	<sup>90</sup> Zr 51.45	<sup>91</sup> Zr 11.22	<sup>92</sup> Zr 17.15



# Radiochemical separation

Development of the chemical system using model tracers

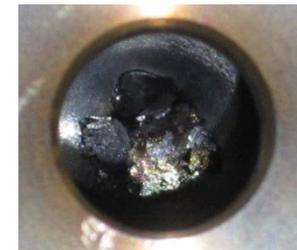


Sample preparation:

Irradiation of 971.3 mg  $^{92}\text{Mo}$  with 20 MeV protons

Determination of the decontamination factor

from  $^{92}\text{Mo}$  by ICP-MS:  $> 10^5$ ;  $\sim 1 \mu\text{g}$   $^{91}\text{Nb}$



RADIOACTIVE IRON RAIN: TRANSPORTING  $^{60}\text{Fe}$  IN SUPERNOVA DUST TO THE OCEAN FLOORBRIAN J. FRY<sup>1</sup>, BRIAN D. FIELDS<sup>1</sup>, AND JOHN R. ELLIS<sup>2</sup><sup>1</sup>Department of Astronomy, University of Illinois, Urbana, IL 61801, USA<sup>2</sup>Theoretical Physics and Cosmology Group, Department of Physics, King's College London, London WC2R 2LS, UK;  
Theory Department, CERN, CH-1211 Geneva 23, Switzerland

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**Nature 2016:**

LETTER

Transport calculations

doi:10.1038/nature17424

The locations of recent supernovae near the Sun  
from modelling  $^{60}\text{Fe}$  transportD. Breitschwerdt<sup>1</sup>, J. Feige<sup>1</sup>, M. M. Schulreich<sup>1</sup>, M. A. de Avillez<sup>1,2</sup>, C. Dettbarn<sup>3</sup> & B. Fuchs<sup>2</sup>

ASTROPARTICLE PHYSICS

Observation of the  $^{60}\text{Fe}$   
nucleosynthesis-clock isotope in  
galactic cosmic raysW. R. Binns,<sup>1\*</sup> M. H. Israel,<sup>1\*</sup> E. R. Christian,<sup>2</sup> A. C. Cummings,<sup>2</sup> G. A. de Nolfo,<sup>2</sup>  
K. A. Lave,<sup>1</sup> R. A. Leske,<sup>2</sup> R. A. Mewaldt,<sup>2</sup> E. C. Stone,<sup>2</sup>  
T. T. von Rosenvinge,<sup>2</sup> M. E. Wiedenbeck<sup>4</sup>**Science 2016:**Mass spectrometry  
with CRIS

PRL 116, 151104 (2016)

PHYSICAL REVIEW LETTERS

week ending  
15 APRIL 2016Interstellar  $^{60}\text{Fe}$  on the Surface of the MoonL. Fimiani,<sup>1</sup> D. L. Cook,<sup>2,\*</sup> T. Faestermann,<sup>1</sup> J. M. Gómez-Guzmán,<sup>1</sup> K. Hain,<sup>1</sup> G. Herzog,<sup>2</sup> K. Knie,<sup>1,†</sup>G. Korschinek,<sup>1,‡</sup> P. Ludwig,<sup>1</sup> J. Park,<sup>2</sup> R. C. Reedy,<sup>3</sup> and G. Rugel<sup>1,§</sup><sup>1</sup>Physik Department, Technische Universität München, D-85748 Garching, Germany<sup>2</sup>Department of Chemistry and Chemical Biology, Rutgers University, Piscataway, New Jersey 08854, USA<sup>3</sup>Planetary Science Institute, Los Alamos, New Mexico 87544-3826, USA

(Received 18 November 2015; published 13 April 2016)

samples from the Apollo missions  
measured by AMS**Nature 2016:**

LETTER

Investigation of ocean floor samples with AMS

doi:10.1038/nature17196

Recent near-Earth supernovae probed by global  
deposition of interstellar radioactive  $^{60}\text{Fe}$ A. Wallner<sup>1</sup>, J. Feige<sup>2†</sup>, N. Kinoshita<sup>2</sup>, M. Paul<sup>4</sup>, L. K. Fifield<sup>5</sup>, R. Golser<sup>2</sup>, M. Honda<sup>5</sup>, U. Linnemann<sup>6</sup>, H. Matsuzaki<sup>7</sup>, S. Merchel<sup>8</sup>,  
G. Rugel<sup>9</sup>, S. G. Timm<sup>1</sup>, P. Steier<sup>2</sup>, T. Yamagata<sup>9</sup> & S. R. Winkler<sup>7</sup>The discovery of  $^{60}\text{Fe}$  from a nearby  
supernova explosion  
~ 2.5 million years ago

# Separation and preparation

Dissolution of Cu chips (3 g) in 7 M  
 $\text{HNO}_3$

Evaporation to dryness

Dissolution in 7 M HCl + 5 mg  $\text{Co}^{2+}$  as  
carrier

Extraction with methylisobutylketone

Aqueous phase:

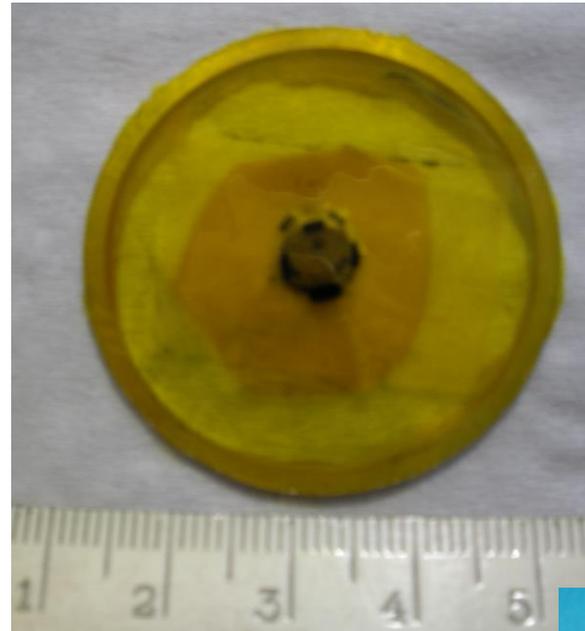
Ni, Co, Cu,

organic phase: Fe

Back extraction with 0.1 M HCl,  
repetition of procedure

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

Evaporation of the final solution onto a  
graphite backing



## $^{60}\text{Fe}$ - 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  $^{60}\text{Fe}$  world-wide within the last 20 years work with material produced at PSI

- New measurement of the  $^{60}\text{Fe}$  half-life, **G. Rugel** et.al. PRL 2009
- Determination of the neutron capture cross section at stellar energies, **E. Uberseder** et.al. PRL 2009
- Quantification of  $^{60}\text{Fe}$  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  $^{60}\text{Fe}$ , **T. Heftrich** et.al., PRC 2015
- Settling the half-life of  $^{60}\text{Fe}$  – fundamental for a versatile astrophysical chronometer, **A. Wallner** et.al., PRL 2015
- Activity measurement of  $^{60}\text{Fe}$  through the decay of  $^{60\text{m}}\text{Co}$  and confirmation of its half-life, **K. Ostiek** et.al. PRC 2017
- Nuclear properties of  $^{60}\text{Fe}$ , **R. Dressler** et.al., currently ongoing

# KARLSRUHER NUKLIDKARTE

8. Auflage 2012

CHART OF THE NUCLIDES, 8<sup>th</sup> Edition 2012 / CARTE DES NUCLEIDES, 8<sup>ème</sup> Edition 2012  
 CARTA DE NUCLEIDOS, 8ª Edición 2012 / ТАБЛИЦА НУКЛИДОВ, 8-е Издание 2012

核素图, 2012年第8版

J. Magill<sup>1</sup>, G. Pfennig<sup>2</sup>, R. Dreher<sup>1</sup>, Z. Sóti<sup>2</sup>

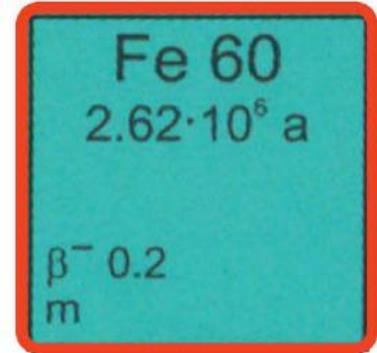
<sup>1</sup>Nucleonica GmbH, c/o European Commission, Hermann-von-Helmholtz-Platz 1, 76344 Eggenstein-Leopoldshafen, Germany, eMail: joseph.magill@nucleonica.com, http://www.nucleonica.com

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G. Rugel, et al.:  
 Phys. Rev. Lett. **103** (2009) 072502

										31																								
										Ga 69.723 α 2.9																								
										Zn 65.38	Zn 54 3.2 ms α 0.74 + 0.74 β 4.19	Zn 55 19.8 ms β 4.604	Zn 56 30.0 ms β 2.877, 2.082 1.147	Zn 57 47 ms β 1.87, 4.52 2.50 + 2701*	Zn 58 84 ms β 203.848	Zn 59 182 ms β 491.914 β 1.75, 2.09 1.62, 1.38	Zn 60 2.4 m β 2.5, 3.1 β 70.61, 273 334	Zn 61 1.5 m β 4.4 β 475.1660 970	Zn 62 9.13 h β 0.7 β 41.597, 548 428	Zn 63 38.1 m β 2.3 β 670.962 1412	Zn 64 49.17 h α 0.74 α <sub>min</sub> 1.1E-5 α <sub>max</sub> 1.2E-5	Zn 65 244.3 d α 0.3 α <sub>min</sub> 1.1E-5 α <sub>max</sub> 2.0	Zn 66 27.73 α 0.9 α <sub>min</sub> < 2E-5											
										Cu 63.546 α 3.8	Cu 53 <300 ns β 2.1	Cu 54 <75 ns β 7	Cu 55 27 ms β 2701, 1225 2507, 2780	Cu 56 53 ms β 7.7 β 1112	Cu 57 199 ms β 7.5 β 1424, 1448 338, 465	Cu 58 3.20 s β 3.8 β 1302, 878 338, 465	Cu 59 82 s β 2.0, 3.0 β 1332, 1792 1385	Cu 60 23 m β 1.2 β 283, 856, 87 1385	Cu 61 3.4 h β 2.8 β 1373	Cu 62 9.74 m α 4.5	Cu 63 69.15 α 4.5	Cu 64 12.7004 h β 0.6, β* 0.7 β 1246	Cu 65 30.85 α 2.17											
										Ni 58.6934	Ni 52 40.8 ms β 4.4 β 1037 205, 1057*	Ni 53 55.2 ms β 1.093, 2.354 2.072	Ni 54 104 ms β 937 β 948*	Ni 55 209 ms β 7.7 β 120.19, 2976 3003	Ni 56 6.075 d β 0.8 β 158, 812, 750 488, 278	Ni 57 36 h β 7.7 β 1278, 1920 127	Ni 58 68.077 α 4.6 α <sub>min</sub> < 3E-5	Ni 59 7.5 · 10 <sup>4</sup> a α 2.0 α <sub>min</sub> < 3E-5 α <sub>max</sub> 92	Ni 60 26.223 α 2.5 α <sub>min</sub> < 3E-5	Ni 61 1.1399 α 1.5	Ni 62 3.6346 α 1.5	Ni 63 100 s β 0.07 β 20	Ni 64 0.9255 α 1.6											
										Co 58.9332	Co 50 36.8 ms β 2.715 2.002 765, 462*	Co 51 68.8 ms β 2.1	Co 52 163 ms β 1.4 β 849, 1038 1042	Co 53 247 ms β 1.4 β 1.4, 1.3 1136, 1130 407	Co 54 148 ms β 4.3 β 1211, 373 1969	Co 55 17.54 h β 1.5 β 931, 477 1400	Co 56 77, 236 d β 1.5 β 847, 1238 2588, 1771 1035	Co 57 271.80 d β 1.5 β 122, 136, 14	Co 58 894 h β 1.5 β 2.8, 1.3 16000, 1960	Co 59 100 β 0.6 β 1009, 1290 α < 10	Co 60 16.5 m β 2.8 β 1373, 1173 58	Co 61 1.65 h β 1.2 β 67, 908	Co 62 13 m β 2.8, 2.6 β 1205, 1027 298	Co 63 27.5 s β 3.6 β 87, 982										
										Fe 55.845	Fe 49 64.7 ms β 1.307 β 752, 702, 707 482	Fe 50 150 ms β 0.51	Fe 51 305 ms β 7.0 β 237	Fe 52 827 s β 1.4 β 895, 801 208, 180 240	Fe 53 2.5 m β 4.3 β 1211, 373 1969	Fe 54 8.845 β 0.3 β 1031, 101 1000, 1E-5	Fe 55 2.73 a β 0.1 β 1031, 101 1000, 1E-5	Fe 56 91.754 β 2.8 β 122, 136, 14	Fe 57 2.119 α 1.4	Fe 58 0.282 α 1.3	Fe 59 44.494 d β 0.5, 1.6 β 1009, 1290 α < 10	Fe 60 2.62 · 10 <sup>6</sup> a β 0.2	Fe 61 6.0 m β 2.6, 2.6 β 1205, 1027 298	Fe 62 68 s β 2.5 β 906										
										Mn 54.938045	Mn 48 158 ms β 752, 1106 3678	Mn 49 382 ms β 0.7 β 273, 2505	Mn 50 175 ms β 1.3 β 1399, 1399 1451	Mn 51 46.2 m β 2.2 β 1224, 1224 1339, 1339 1748	Mn 52 21 m β 5.2 β 1224, 1224 1339, 1339 1748	Mn 53 3.7 · 10 <sup>4</sup> a β 70 β 835 α < 10	Mn 54 312.2 d β 2.8 β 847, 1611 2113	Mn 55 100 β 2.8 β 847, 1611 2113	Mn 56 2.58 h β 2.8 β 847, 1611 2113	Mn 57 1.5 m β 2.8 β 847, 1611 2113	Mn 58 63 s β 5.7 β 847, 1611 2113	Mn 59 4.5 s β 4.4, 4.2 β 728, 473 1965, 1170 973, 183	Mn 60 6.0 m β 5.7 β 847, 1611 2113	Mn 61 0.71 s β 9.4 β 929, 207										
										Cr 51.9404	Cr 47 472 ms β 8.4 β 87	Cr 48 21.8 h β 0.7 β 308, 112	Cr 49 42 m β 1.4, 1.5 β 91, 153, 62	Cr 50 4.345 α 15	Cr 51 27, 7010 d α 3209 α < 10	Cr 52 83.789 α 0.8	Cr 53 9.501 α 18	Cr 54 2.365 α 0.36	Cr 55 21.1 s β 2.6 β 1828	Cr 56 5.94 m β 1.5 β 83, 28	Cr 57 7.0 s β 5.1 β 83, 850, 1752 1535	Cr 58 1.05 s β 883, 128, 290 520 β 1238, 1900 112, 663	Cr 59 0.49 s β 8.7 β 949, 410, 758 g	Cr 60 0.49 s β 8.7 β 949, 410, 758 g										



# 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 ( ${}^{7/10}\text{Be}$ ,  ${}^{60}\text{Fe}$ ,  ${}^{53}\text{Mn}$ ,  ${}^{44}\text{Ti}$  and others)**
- **Examples for front-end experiments using targets made by PSI**
  - ${}^{60}\text{Fe}$  half-life and neutron capture cross section measurements
- **Examples for ongoing experiments ( ${}^{10}\text{Be}$ ,  ${}^{91}\text{Nb}$ )**
- **We need a network on target preparation**
- **We need a dedicated mass separation device for exotic radionuclides!**

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