

PAUL SCHERRER INSTITUT

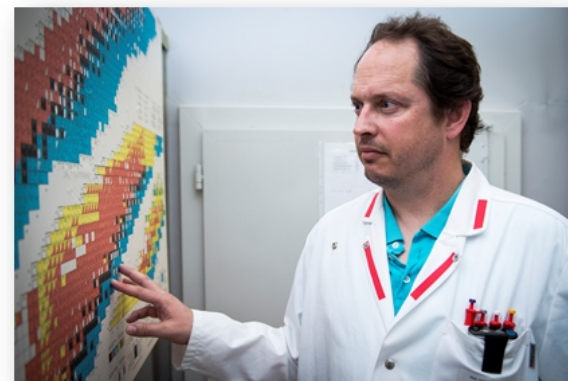


Robert Eichler

Paul Scherrer Institute

# Laboratory for Radiochemistry (**LRC**)

„NES präsentiert: Kompetenzen und Highlights“, Villigen, 24.10.2017



### **HEAVY ELEMENTS**

Dr. Robert Eichler

Dr. Rugard Dressler

Alexander Vögele

Dominik Hermann

Dr. Zeynep Talip (SNF)

Benjamin Kraus (SNF)

Yves Wittwer (SNF)

Jiri Ulrich (CROSS/NUM)

Paul Ionescu

### **ISOTOPE AND TARGET CHEMISTRY**

Dr. Dorothea Schumann

Dr. Jörg Neuhausen

Dr. Emilio Maugeri

Dr. Ivan Kajan (COFUND/sn)

Dr. Stephan Heinitz (EU<sub>finishing</sub>)

Mu Lin (swissnuclear (sn))

Erik Karlson (EU)

Ivan Danilov

### **RADIONUCLIDE DEVELOPMENT**

Dr. Nicholas van der Meulen

NN @ CRS

Katharina Domnanich

Nadezda Gracheva @ CRS

Sarah Jordi

Postdoc

Graduate Students

1/3 of all elements have only radioactive isotopes

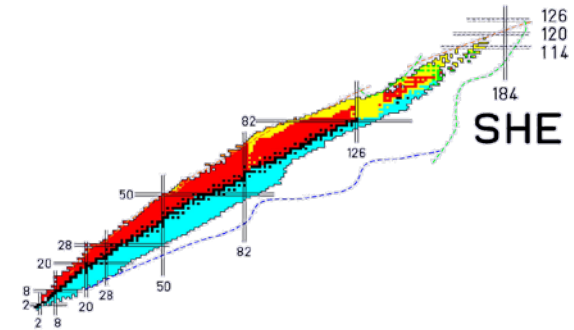
>90% of all isotopes are radioactive current status

1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18
H																	He
Li	Be											B	C	N	O	F	Ne
Na	Mg											Al	Si	P	S	Cl	Ar
K	Ca	Sc	Ti	V	Cr	Mn	Fe	Co	Ni	Cu	Zn	Ga	Ge	As	Se	Br	Kr
Rb	Sr	Y	Zr	Nb	Mo	Tc	Ru	Rh	Pd	Ag	Cd	In	Sn	Sb	Te	I	Xe
Cs	Ba	La	Hf	Ta	W	Re	Os	Ir	Pt	Au	Hg	Tl	Pb	Bi	Po	At	Rn
Fr	Ra	Ac	Rf	Db	Sg	Bh	Hs	Mt	Ds	Rg	Cn	Nh	Fl	Mc	Lv	Ts	Og

119 120

\* Ce Pr Nd Pm Sm Eu Gd Tb Dy Ho Er Tm Yb Lu

\*\* Th Pa U Np Pu Am Cm Bk Cf Es Fm Md No Lr



## ➤ Industry:

- industrial tracers chemistry, biology
- inspection (radiation sources)
- gauges (e.g. thickness measurements)
- Nuclear energy:
  - nuclear fuel / waste

## ➤ Medicine:

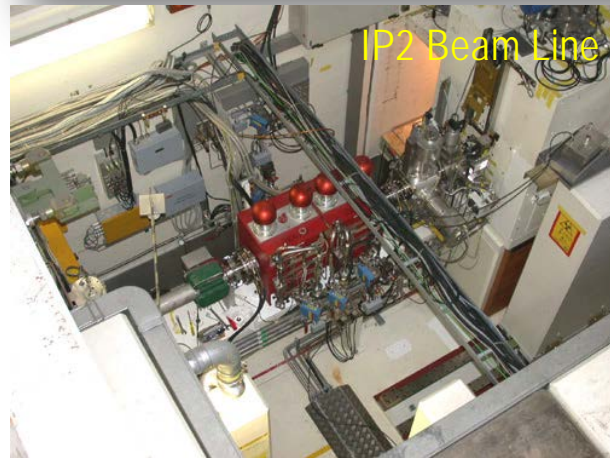
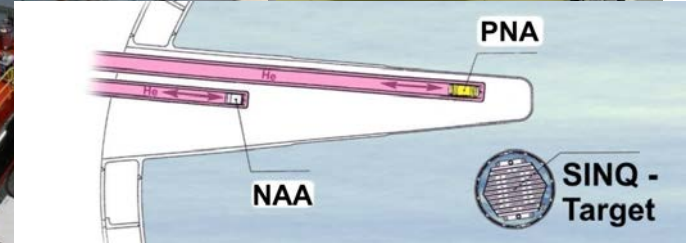
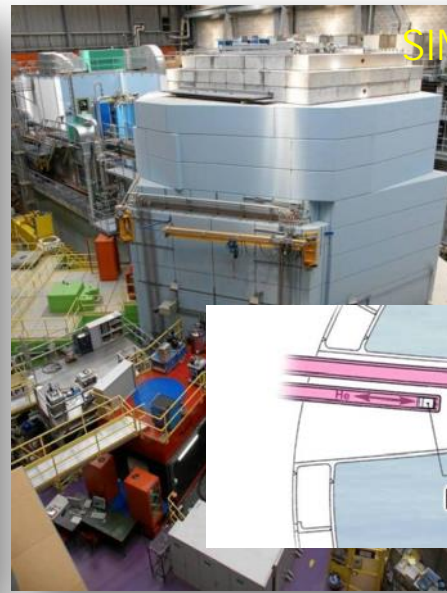
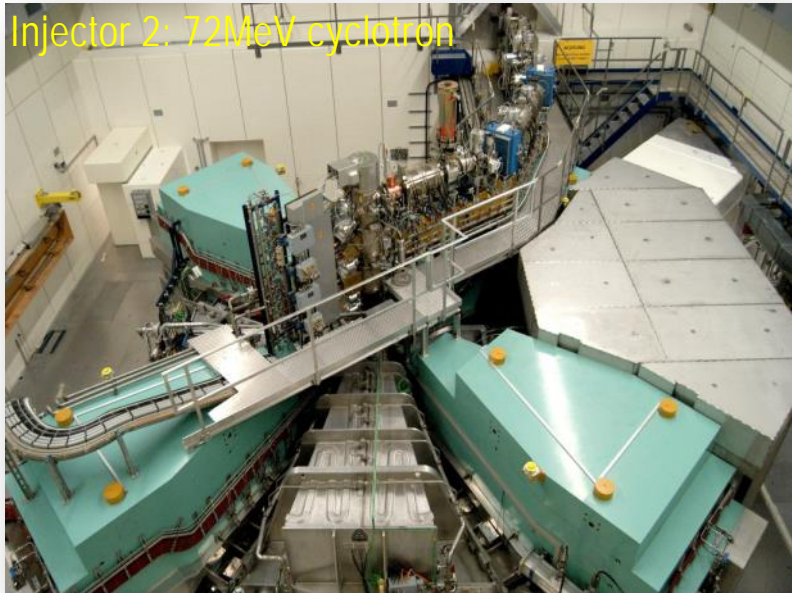
- radiopharmaceuticals
- radiation therapy

### *Radiochemical science @ PSI*

- fundamental aspects: nucleosynthesis, astrophysics, chemistry (kinetics, thermodynamics -> catalysis) of new elements
- isotope production, separation and handling
- nuclear data

- We maintain theoretical and experimental academic expertise in radiochemistry for Switzerland:
  - We provide exciting and challenging high level international science and research;
  - We educate the next generation scientists;
- We make use of the PSI-Large Scale Facilities and other PSI-Infrastructure:
  - We make use of valuable radioactivity produced at PSI, even if (or despite) it is declared “waste”;
  - We make use of production facilities for producing radionuclides as tracers for fundamental research and radiopharmacy (SINQ-NIS, SINQ-Stip, IP2, PROTRAC);
  - We make use of radioanalytics and 0-, A-, B-, and C- laboratories at PSI
- We find and use synergies with the other NES Laboratories and other PSI departments;

# LRC's Use of Large Scale Facilities at PSI



# The Research of **LRC** within PSI

- Fundamental research for education and maintenance of radiochemical expertise in Switzerland;
  - Chemistry in spallation targets (ADS (Myrrha), ESS, SINQ);
  - (Super-) Heavy elements chemistry (material sciences, fast separations)
    - High temperature materials (PSI-NES-LNM/LRS/LTH: Gen III+IV, ESS),
    - Solid state diffusion and thermal release (PSI-NES-LNM/LRS: Gen IV, ESS);
    - Volatilization and gas phase chemistry (PSI-NES-LRS/LTH: Gen III+IV);
- Disposal strategy for radioactive PSI accelerator-generated waste and Hotlab waste
  - Chemical separation of valuable isotopes and their use in astrophysical experiments (NUM-UCN/NIAG) and radiopharmacy (BIO-CRS),
  - Nuclear Data (Astrophysics, PSI-NES-LRS/LTH);
- Production and separation of radionuclides (PSI-IP, SINQ):
  - New isotopes for radiopharmacy (BIO-CRS) and tracers for radiochemistry (NES);
  - Nuclear Data (NES-LRS, BIO-CRS);

# Isotope and Target Chemistry

## Copper beam dump

- $^{44}\text{Ti}$ ,  $^{53}\text{Mn}$ ,  $^{26}\text{Al}$ ,  $^{60}\text{Fe}$ ,  $^{59}\text{Ni}$ ,  $^{32}\text{Si}$
- $^{60}\text{Co}$  – 5 GBq



## Myon production station

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



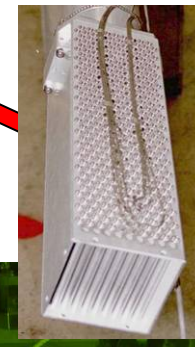
## SINO Target Irradiation Program-STIP

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



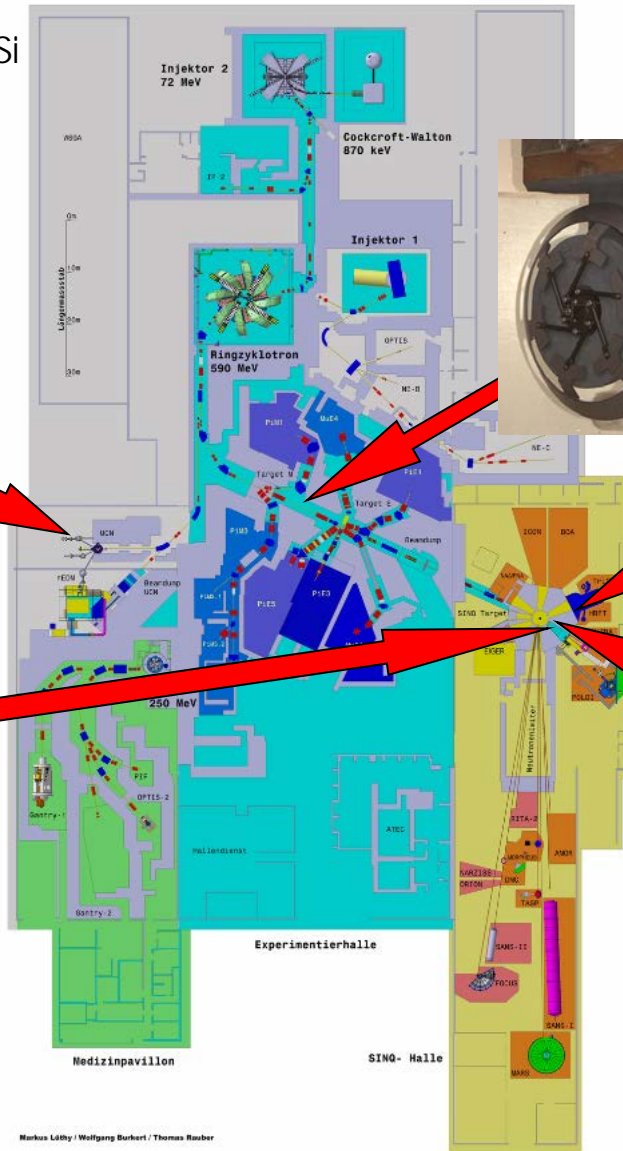
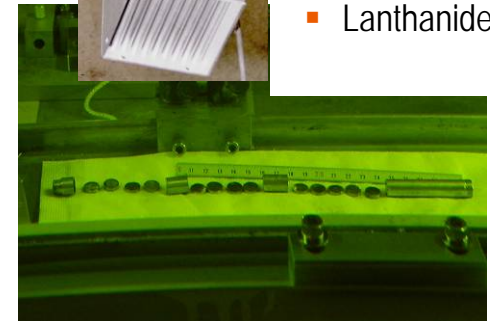
## SINO cooling water

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



## SINO target

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



Markus Lüthy / Wolfgang Burkert / Thomas Rosler



# Isotope and Target Chemistry achievements

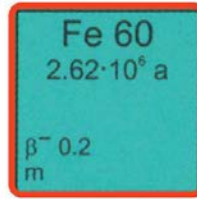
## KARLSRUHER NUKLIDKARTE 8. Auflage 2012

CHART OF THE NUCLIDES, 8<sup>th</sup> Edition 2012 / CARTE DES NUCLÉIDES, 8<sup>ème</sup> Edition 2012  
 CARTA DE NUCLEIDOS, 8<sup>ª</sup> 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. Solti<sup>3</sup>

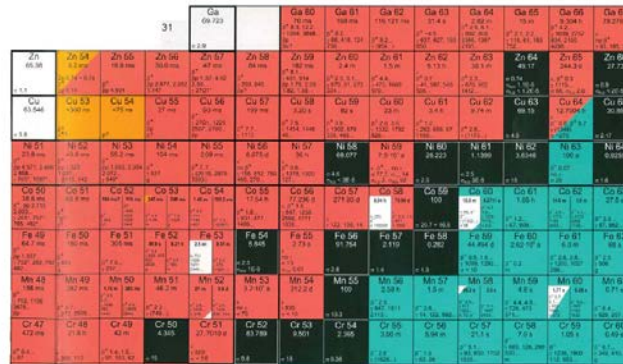
<sup>1</sup>Nucleonica GmbH, c/o European Commission, Hermann-von-Helmholtz-Platz, 1, 70344 Eggenstein-Leopoldsdafen, Germany, eMail: joseph.magill@nucleonica.com, http://www.nucleonica.com  
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 P.O. Box 2345, 70300 Karlsruhe, Germany  
<sup>3</sup>Nucleonica GmbH 2012, developed under a License of the European Atomic Energy Community

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➤ <sup>60</sup>Fe important waiting point for astrophysical nucleosynthesis

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



Great collaboration with Hotlab (M. Martin, Niko Kivel)

### Editors' Suggestion

## <sup>7</sup>Be(n, α)<sup>4</sup>He Reaction and the Cosmological Lithium Problem: Measurement of the Cross Section in a Wide Energy Range at n\_TOF at CERN

M. Barbagallo *et al.* (n\_TOF Collaboration)

Phys. Rev. Lett. **117**, 152701 (2016) – Published 3 October 2016



The neutron-<sup>7</sup>Be cross section—an important ingredient in Big Bang nucleosynthesis—is measured at a wide range of neutron energies.

[Show Abstract +](#)

# Isotope and Target Chemistry achievements

## Metal chemistry (J. Neuhausen)

- The thermal release of mercury, iodine, cesium, polonium from LBE is investigated by transpiration methods;
- The indirect identification of released species is tried using various reactive gas phases via thermochromatography;
- The use of various stationary gas chromatographic phases allows for investigation of deposition properties relevant for filtering and safety absorbers;
- The possibility of the tracer production at PSI is absolutely crucial for this work (e.g.  $^{210}\text{Po}$ , I,  $^{134}\text{Cs}$ ,  $^{131}\text{I}$ );
- This work is supported by the EU
  - the projects SEARCH and MYRTE under EURATOM HORIZON2020
  - >10 publications already published

### MYRRHA: an innovative research installation

SCK•CEN is actively working on designing and building a new multifunctional research installation: MYRRHA as in *Multi-purpose hybrid Research Reactor for High-tech Applications*.



## The Chemistry of Transactinide Elements

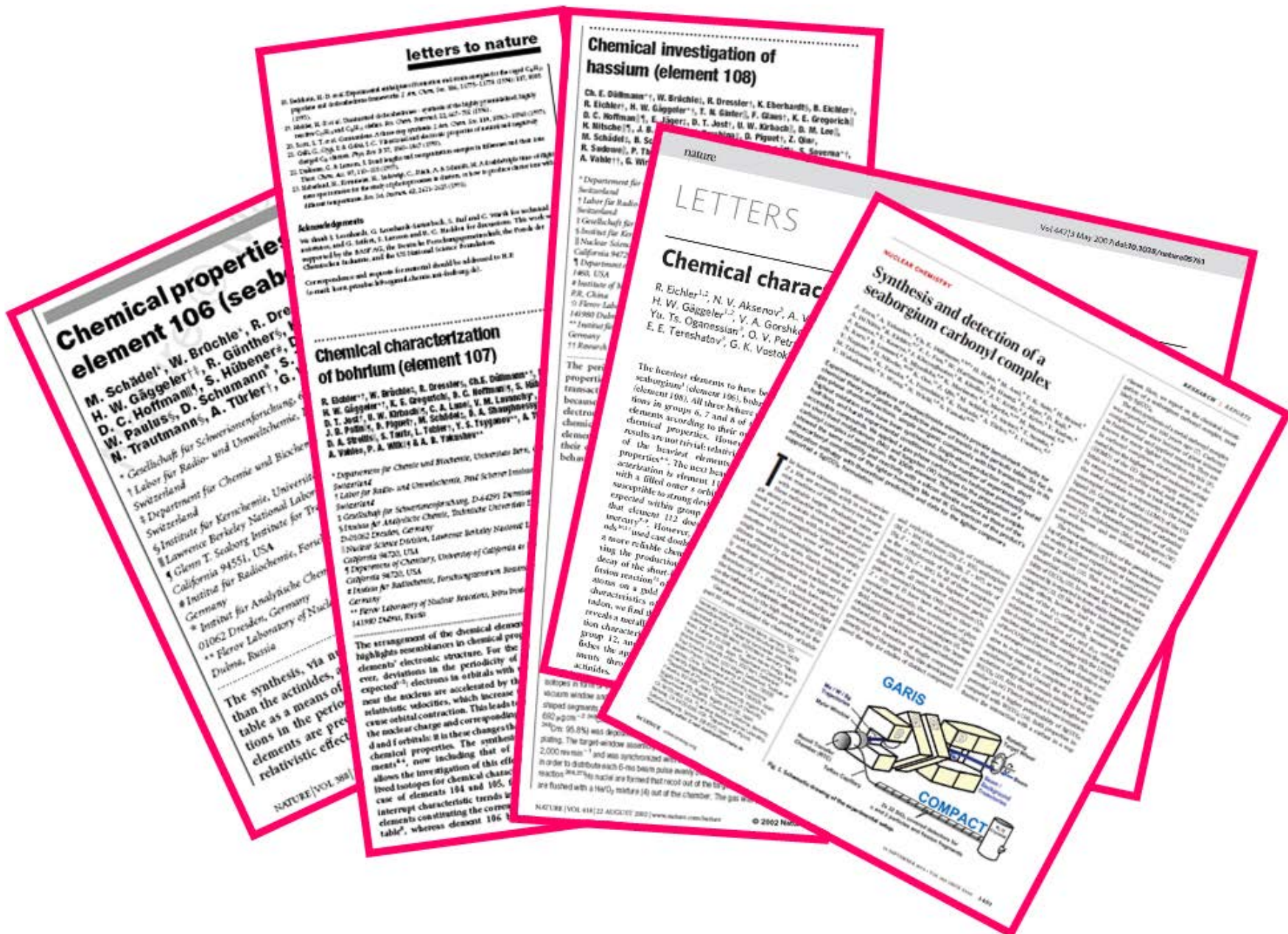
1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18
H																	He
Li	Be											B	C	N	O	F	Ne
Na	Mg											Al	Si	P	S	Cl	Ar
K	Ca	Sc	Ti	V	Cr	Mn	Fe	Co	Ni	Cu	Zn	Ga	Ge	As	Se	Br	Kr
Rb	Sr	Y	Zr	Nb	Mo	Tc	Ru	Rh	Pd	Ag	Cd	In	Sn	Sb	Te	I	Xe
Cs	Ba	La*	Hf	Ta	W	Re	Os	Ir	Pt	Au	Hg	Tl	Pb	Bi	Po	At	Rn
Fr	Ra	Ac**	Rf	Db	Sg	Bh	Hs	Mt	Ds	Rg	Cn	Nh	Fl	Mc	Lv	Ts	Og

*119 120*

\* Ce Pr Nd Pm Sm Eu Gd Tb Dy Ho Er Tm Yb Lu

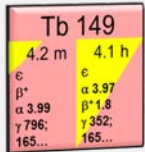
\*\* Th Pa U Np Pu Am Cm Bk Cf Es Fm Md No Lr

# The Chemistry of Transactinide Elements

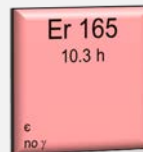


# Radionuclide Development

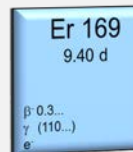
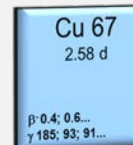
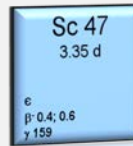
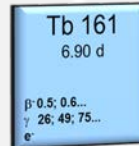
## $\alpha$ -Therapy



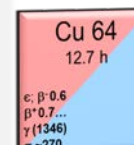
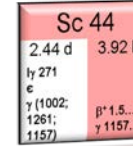
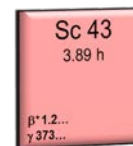
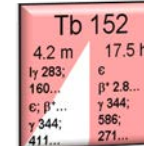
## Auger-e<sup>-</sup> Therapy



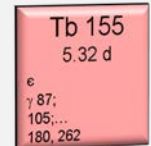
## $\beta$ -Therapy



## PET ( $\beta^-$ )



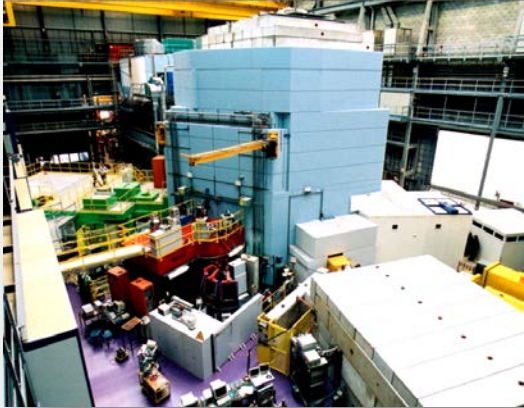
## SPECT ( $\gamma$ )



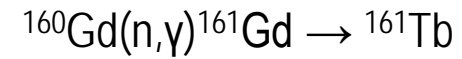
➤ Theranostic principle of *matched pairs*

# $^{161}\text{Tb}$ : A $\beta^-$ -Emitter for Therapy

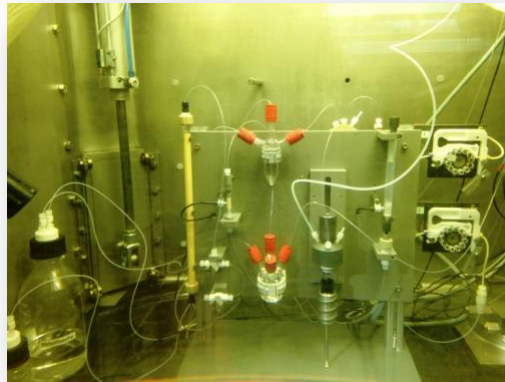
## Neutron Irradiation of Target Material



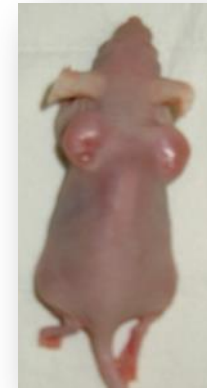
Tb 159 100 # 23.2	Tb 160 72.3 d $\beta^-$ 0.6; 1.7... $\gamma$ 879; 299; 966... # 570	Tb 161 6.90 d $\beta^-$ 0.5; 0.8... $\gamma$ 26; 49; 75; 87	Gd 161 3.66 m $\beta^-$ 4.7... $\gamma$ 361; 315; 102... # 20000
Gd 158 24.84 # 2.3	Gd 159 18.48 h $\beta^-$ 1.0... $\gamma$ 364; 58...	Gd 160 21.86 # 1.5	



## Chemical Separation and Processing



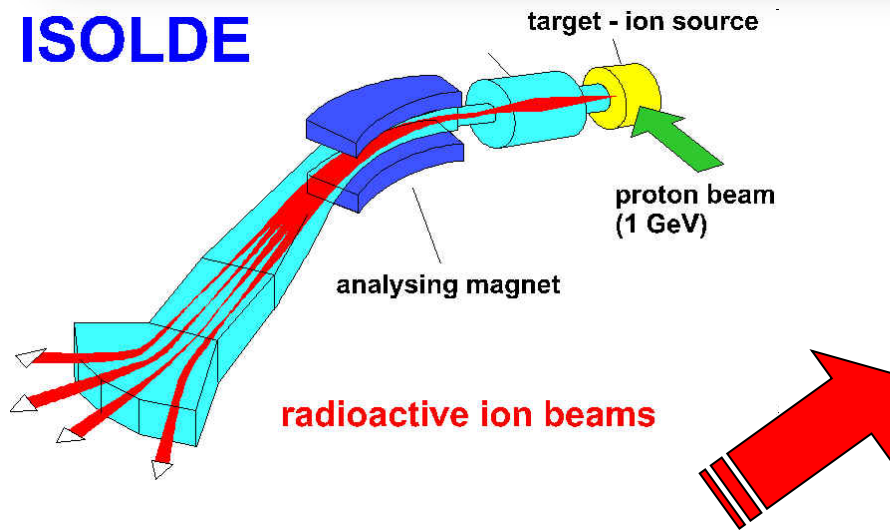
## $^{161}\text{Tb}$ therapy with low-energy $\beta$ -particles



# Collaboration: ISOLDE (CERN) & PSI

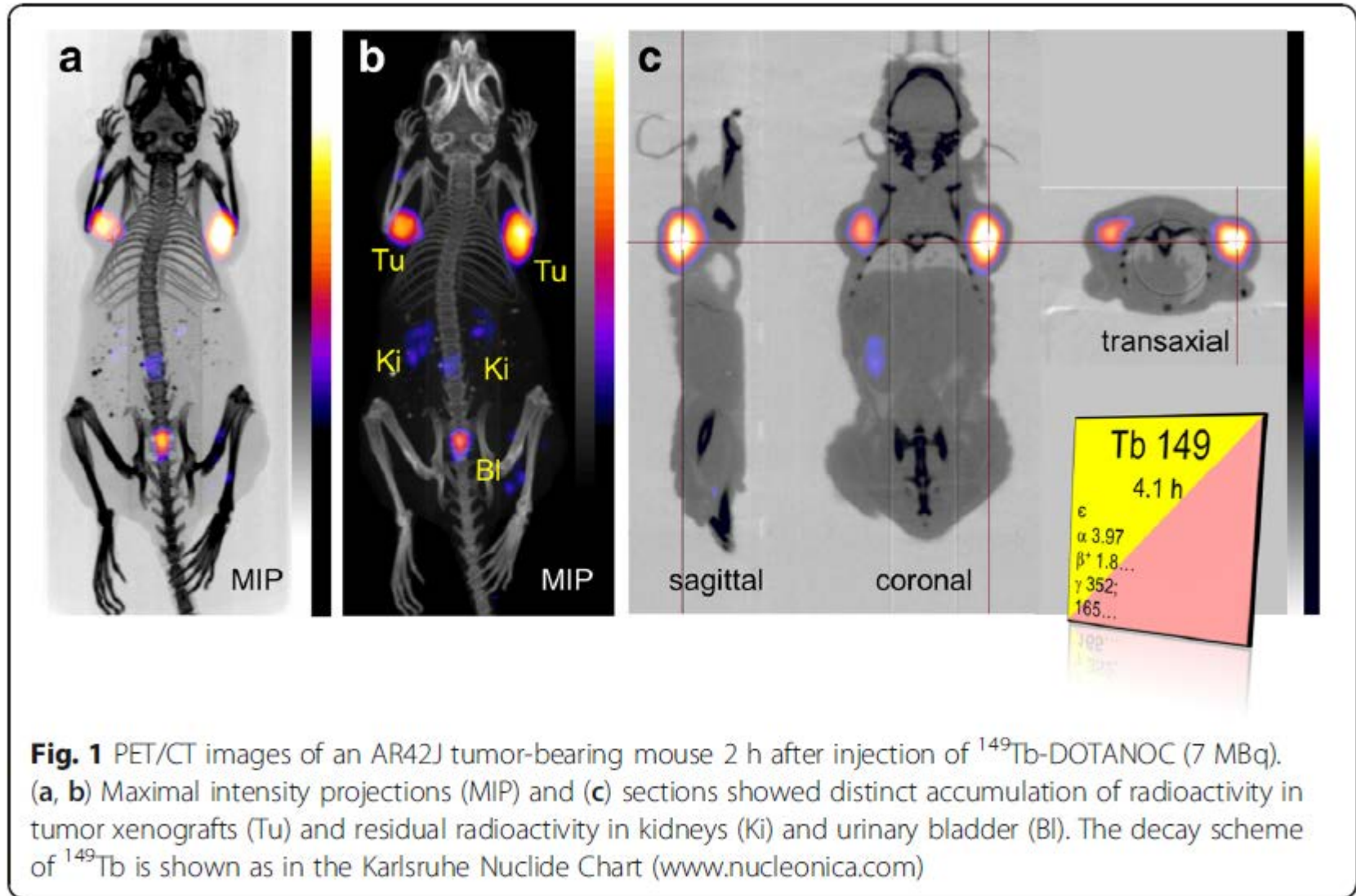
Dy 150 7.2 m ε; β <sup>+</sup> ... α 4.23 γ 397	Dy 151 17 m ε; α 4.07 γ 386; 49; 546; 176... m	Dy 152 2.4 h ε 3.63 γ 257	Dy 153 6.29 h ε; β <sup>+</sup> ... α 3.46... γ 81; 214; 103; 254	Dy 154 3.0 · 10 <sup>6</sup> a α 2.87	Dy 155 10.0 h ε <sup>+</sup> β <sup>+</sup> 0.9; 1.1... γ 227...	Dy 156 0.056 σ 33 σ <sub>n, α</sub> < 0.009	Dy 157 8.1 h ε γ 326...	Dy 158 0.095 σ 33 σ <sub>n, α</sub> < 0.006	Dy 159 144.4 d ε γ 58; β <sup>-</sup> σ 8000	Dy 160 2.329 σ 60 σ <sub>n, α</sub> < 0.0003	Dy 161 18.889 σ 600 σ <sub>n, α</sub> < 1E-6	Dy 162 25.475 σ 170	Dy 163 24.896 σ 120 σ <sub>n, α</sub> < 2E-5
Tb 149 4.2 m β <sup>+</sup> α 3.99 γ 796; 165...	Tb 150 4.1 h 3.8 m 3.67 h ε 4.07 γ 386; 49; 546; 176... m	Tb 151 25 s 17.6 h ε 3.63 γ 257	Tb 152 4.2 m 17.5 h ε; β <sup>+</sup> ... α 3.46... γ 81; 214; 103; 254	Tb 153 2.34 d ε <sup>+</sup> β <sup>+</sup> 0.9; 1.1... γ 227...	Tb 154 23 h 9.0 h 21 h ε 4.17 γ 248; 160; 180; 347; 123; 1420; 246; 1274 540	Tb 155 5.32 d ε γ 87; 105; 180; 262...	Tb 156 24 h 5.4 h 5.4 d ε 4.17 γ 248; 160; 180; 347; 123; 1420; 246; 1274 540	Tb 157 99 a ε γ (54)	Tb 158 10.5 s 180 a ε β <sup>-</sup> 0.9 γ 944; 962, 80	Tb 159 100 σ 23.2	Tb 160 72.3 d β <sup>-</sup> 0.6; 1.7... γ 879; 299; 966... σ 570	Tb 161 6.90 d β <sup>-</sup> 0.5; 0.6... γ 26; 49; 75... ε <sup>-</sup>	Tb 162 7.76 m β <sup>-</sup> 1.4; 2.4... γ 260; 808; 888...
Gd 148 74.6 a α 3.183 σ 14000	Gd 149 9.28 d ε; α 3.016 γ 150; 299; 347...	Gd 150 1.8 · 10 <sup>6</sup> a α 2.72	Gd 151 120 d ε; α 2.60 γ 154; 243; 175...	Gd 152 0.20 1.1 · 10 <sup>14</sup> a α 2.14; σ 700 σ <sub>n, α</sub> < 0.007	Gd 153 239.47 d ε γ 97; 103; 70... σ 20000 σ <sub>n, α</sub> 0.03	Gd 154 2.18 σ 60	Gd 155 14.80 σ 61000 σ <sub>n, α</sub> 0.00008	Gd 156 20.47 σ ~ 2.0	Gd 157 15.65 σ 254000 σ <sub>n, α</sub> < 0.05	Gd 158 24.84 σ 2.3	Gd 159 18.48 h β <sup>-</sup> 1.0... γ 364; 58...	Gd 160 21.86 σ 1.5	Gd 161 3.66 m β <sup>-</sup> 1.6; 1.7... γ 381; 315; 102... σ 20000

**ISOLDE**



Spallation of Ta with high-energy protons, followed by online mass separation







Collaboration between PSI (CRS-BIO and LRC-NES) and ISOLDE/CERN Geneva

Müller *et al.* *EJNMMI Radiopharmacy and Chemistry* (2016) 1:5  
DOI 10.1186/s41181-016-0008-2

 EJNMMI Radiopharmacy and Chemistry  
a SpringerOpen Journal

LETTER TO THE EDITOR

Open Access

## Alpha-PET with terbium-149: evidence and perspectives for radiotheragnostics



Cristina Müller<sup>1\*</sup>, Christiaan Vermeulen<sup>1</sup>, Ulli Köster<sup>2</sup>, Karl Johnston<sup>3</sup>, Andreas Türlér<sup>4,5</sup>, Roger Schibli<sup>1,6</sup> and Nicholas P. van der Meulen<sup>1,4\*</sup>

Tb 149	
4.1 h	
e	
α 3.97	
β <sup>+</sup> 1.8...	
γ 352;	
165...	

- Radiolanthanide for α-therapy (easy chelation using DOTA)
- Ideal half-life of 4.1 h
- Low α-energy of 3.9 MeV
- No α-emitting daughters
- Is it also suitable for PET imaging?

# Where do we want to go @ LRC ?

## ➤ *Isotope and Target Chemistry + Radioanalytics:*

Chemical Separation (tracers), Mass separation;

Targets for astrophysical experiments ( $^{53}\text{Mn}$ ,  $^{10}\text{Be}$ ,  $^{26}\text{Al}$ ,  $^{59}\text{Ni}$ ,  $^{93}\text{Mo}$ ,  $^{163}\text{Ho}$ );

Nuclear data;

## ➤ *Heavy Elements + Metal chemistry:*

Fast separation (tracers, diffusion and release) ;

Reaction chromatography (Hg, Pb & Cn, Fl);

Inorganic radiochemistry (Tl, Po, Bi, At & Nh, Lv, Mc, Ts);

Organometallics (Mo/W, Tc/Re, Ru/Os & Sg, Bh, Hs);

Stern-Gerlach for SHE (diffusion and release, single atomic beam, Lr);

High power target development;

Single molecular speciation, mass measurement;

## ➤ *Radionuclide development + Isotope production:*

>350 Isotopes with scientific potential can be produced at SINQ & IP

Relevant for radiopharmacy e.g.:  $^{43,44,47}\text{Sc}$ ,  $^{64,67}\text{Cu}$  &  $^{165,169}\text{Er}$

Chemical Separation / Mass separation (targets and tracers)

Nuclear data

# Mass separation @ LRC Examples

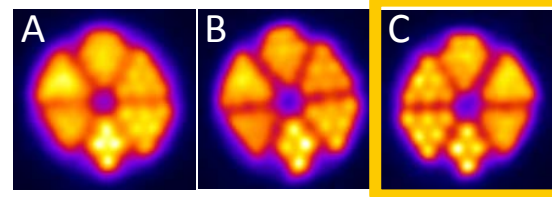
## ➤ Measurement of the $^{53}\text{Mn}(n,\gamma)^{54}\text{Mn}$ cross section

Fe 54 5.845 $\sigma$ 2.3 $\sigma_{n,\alpha}$ 1E-5	Fe 55 2.73 a $\epsilon$ no $\gamma$ $\sigma$ 13 $\sigma_{n,\alpha}$ 0.01	Fe 56 91.754 $\sigma$ 2.8	Fe 57 2.119 $\sigma$ 1.4
Mn 53 $3.7 \cdot 10^6$ a $\epsilon$ no $\gamma$ $\sigma$ 70	Mn 54 312.2 d $\epsilon$ $\gamma$ 835 $\sigma$ < 10	Mn 55 100 $\sigma$ 13.3	Mn 56 2.58 h $\beta^-$ 2.9... $\gamma$ 847, 1811 2113...
Cr 52 83.789 $\sigma$ 0.8	Cr 53 9.501 $\sigma$ 18	Cr 54 2.365 $\sigma$ 0.36	Cr 55 3.50 m $\beta^-$ 2.6 $\gamma$ (1528...)

### Relevance:

- $^{53}\text{Mn}$  used for dating (e.g. meteorites);
- neutron capture  $\rightarrow$  additional loss;
- PSI accelerator waste  $\rightarrow$  world's largest  $^{53}\text{Mn}$  source

## ➤ Scandium isotopes for radiopharmacy



### PET phantom images of

$^{44}\text{Sc}$  (A),

$^{43}\text{Sc}$  produced from  $^{43}\text{Ca}$  (B) 25%  $^{44}\text{Sc}$

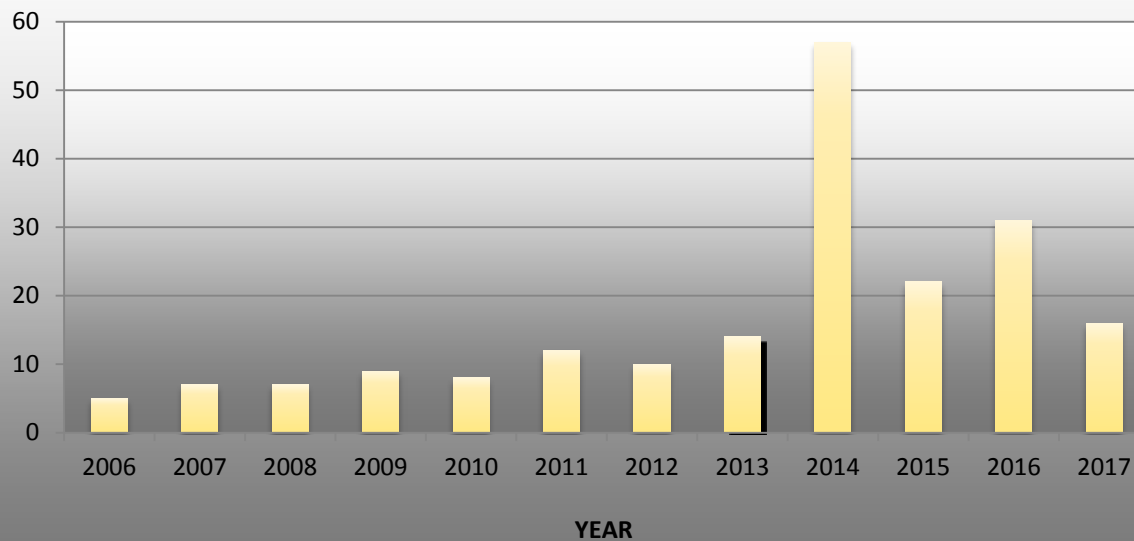
$^{43}\text{Sc}$  produced from  $^{46}\text{Ti}$  (C) 1.5%  $^{44}\text{Sc}$

# Nuclear- and Radiochemistry Research World with LRC-collaborations



# PSI-based LRC is a Driver for Radiochemistry in Switzerland

PEER-REVIEWED PUBLICATIONS (LRC-PSI-based groups)  
2006 - 08/2017



## Take Home Message **LRC**

...LRC

→ belongs to a modern division in an institute with visions and tasks for radiochemistry in the future Switzerland from nuclear energy and beyond;

...LRC

→ is at the frontiers of fundamental chemical and physical research and stays involved in isotope production for life science and research;

... LRC

→ does exciting research and solves important tasks for PSI in multidisciplinary approaches and solid international collaborations;

...LRC

→ Offers unique expertise and exciting educational environment and possibilities

