Chemistry at the lead bismuth loop for EURISOL prototype

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Our proposal given on 2009 December ISOLDE meeting therefore is still relevant

Towards building a *Radionuclide Bank* from proton irradiated Pb-Bi targets

The 1-2 GeV proton induced spallation reaction on the Pb-Bi eutectic converter target will produce large variety of radionuclides along with strong flux of fast neutrons.

We would like to develop methods for separate confinement of each radionuclides with high radiochemical and radioisotopical purity.

Why?

- The radionuclids will have potential applications in medical science as well as in the industry

  Diagnostic : ^{99m}Tc, ^{111}In, ^{123}I, ^{201}Tl, etc.
  Therapeutic : ^{153}Sm, ^{188}Re, ^{186}Re, ^{166}Ho, ^{90}Y, ^{117m}Sn, ^{89}Sr, ^{149}Tb etc.
  Industrial : ^{192}Ir, ^{55}Fe, ^{109}Cd, ^{35}S, ^{63}Ni, ^{85}Kr, ^{204}Tl etc.

- Radionuclides having demand in basic science

- Separation of radionuclides will help to recycle the converter target

Why large facilities became so important

- Reduced reactor facilities/shutdown of reactors
- Limitations of low energy medical accelerators

Example from ^{149}Tb

^{149}Tb is among the few promising α-emitters, which are presently projected for human clinical use.

Half life : 4.118 h
Decay modes: EC (82.3%) and α (17.7 %)
α-particle energy: 3.97 MeV

Production routes

❶Light charged particle (p, α, ^{3}He) induced reactions

^{152}Gd(p

^{152}Gd(α

^{152}Gd(^{3}

❷ proton induced spallation on heavy targets, like Ta, W etc.

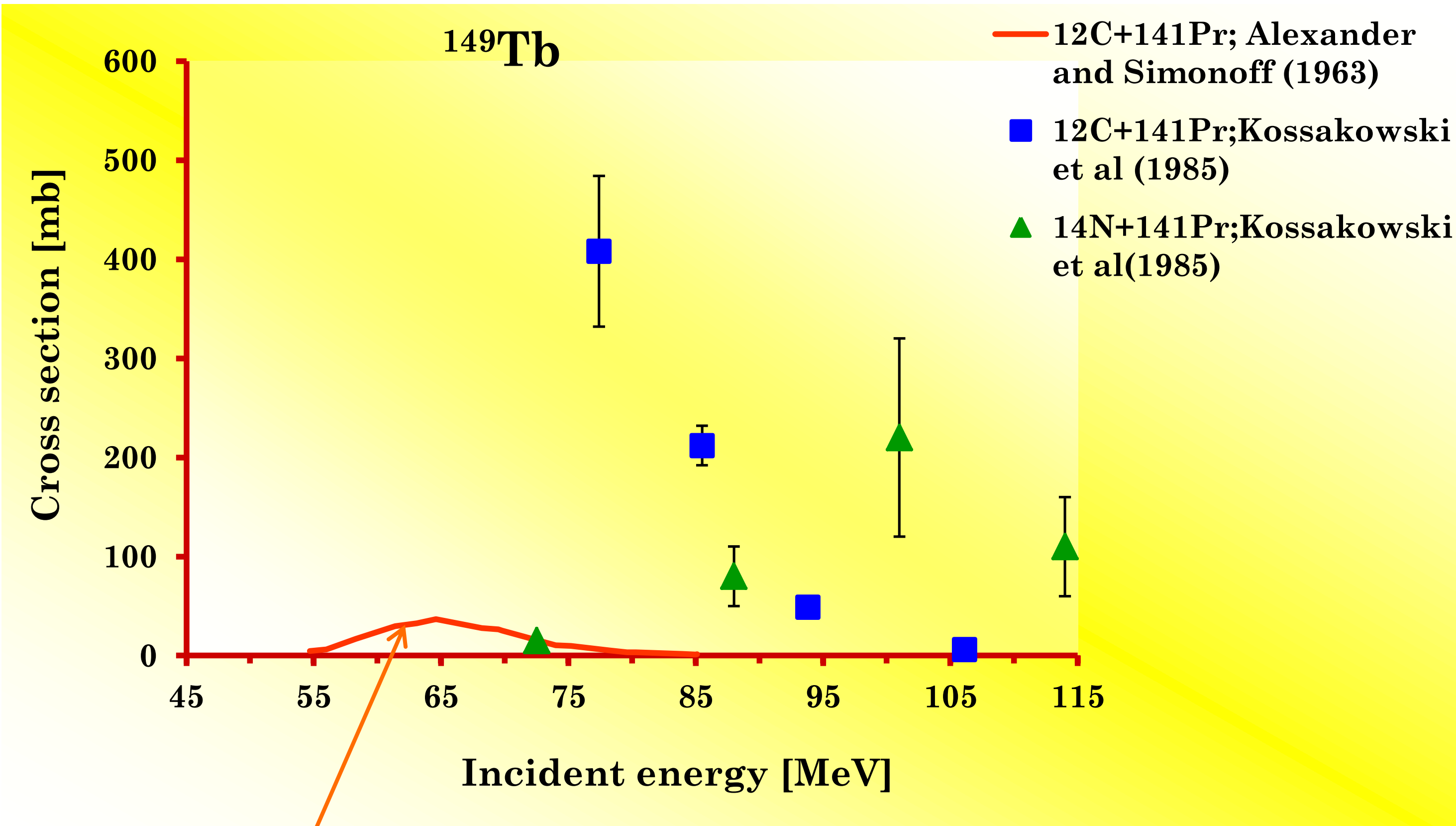
❸ heavy ion induced reactions

^{141}Pr(^{12}C, 4n)^{149}Tb

^{141}Pr(^{14}N, p5n)^{149}Tb

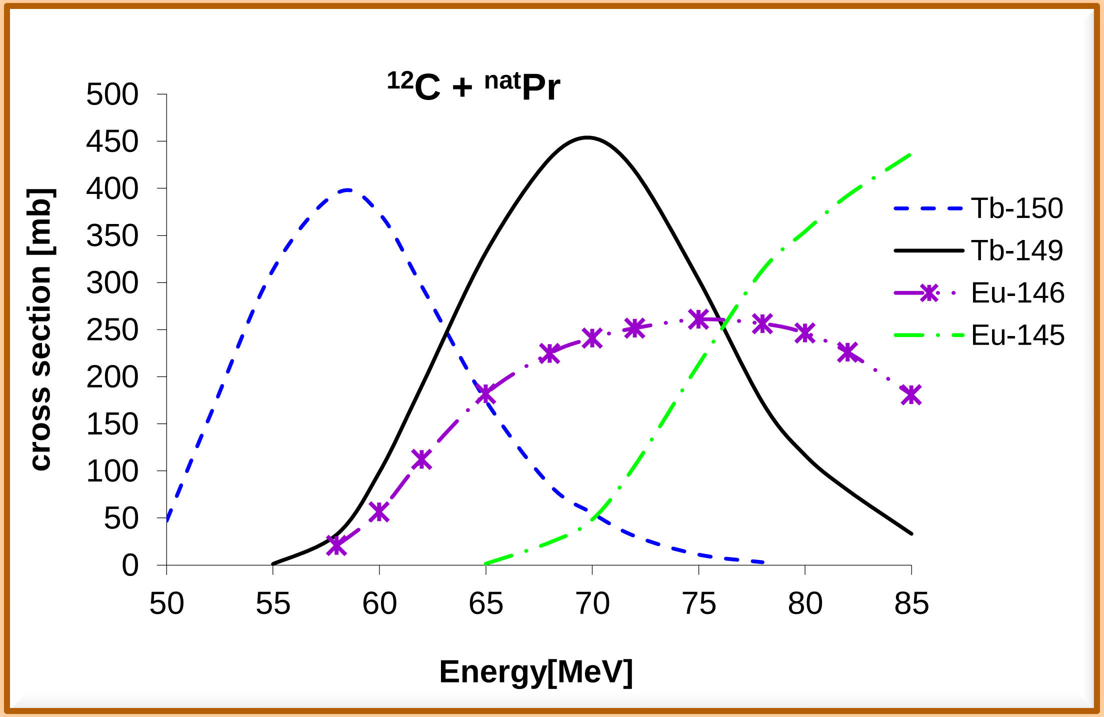
^{144}Sm(^{9}Be, 4n)^{149}Dy (EC) ^{149}Tb $^{142/143}$Nd(^{12}C, 5/6n)^{149}Dy(EC)^{149}Tb

Existing reports…



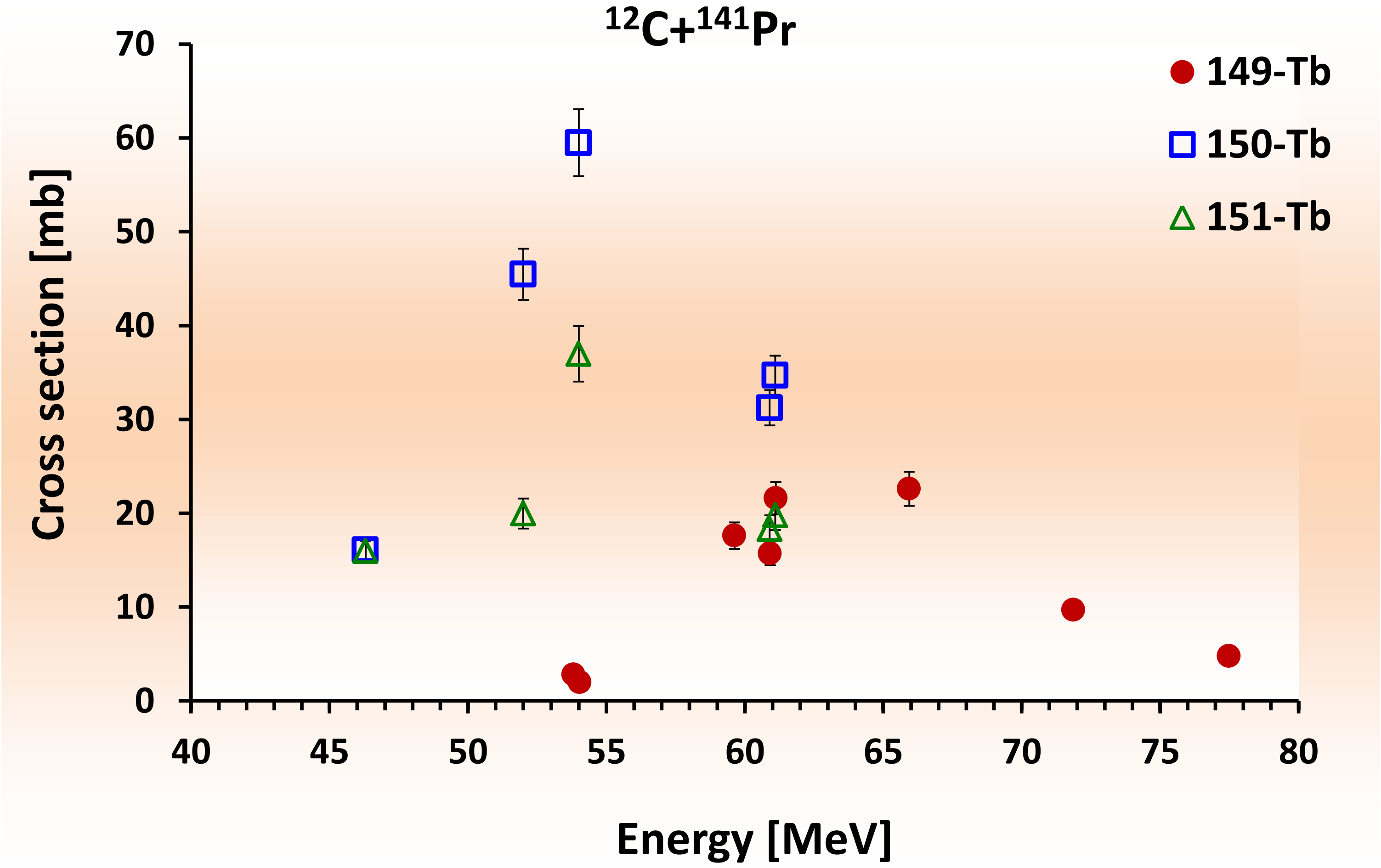
$^{12}C+^{141}Pr$ (1963)

Theory…

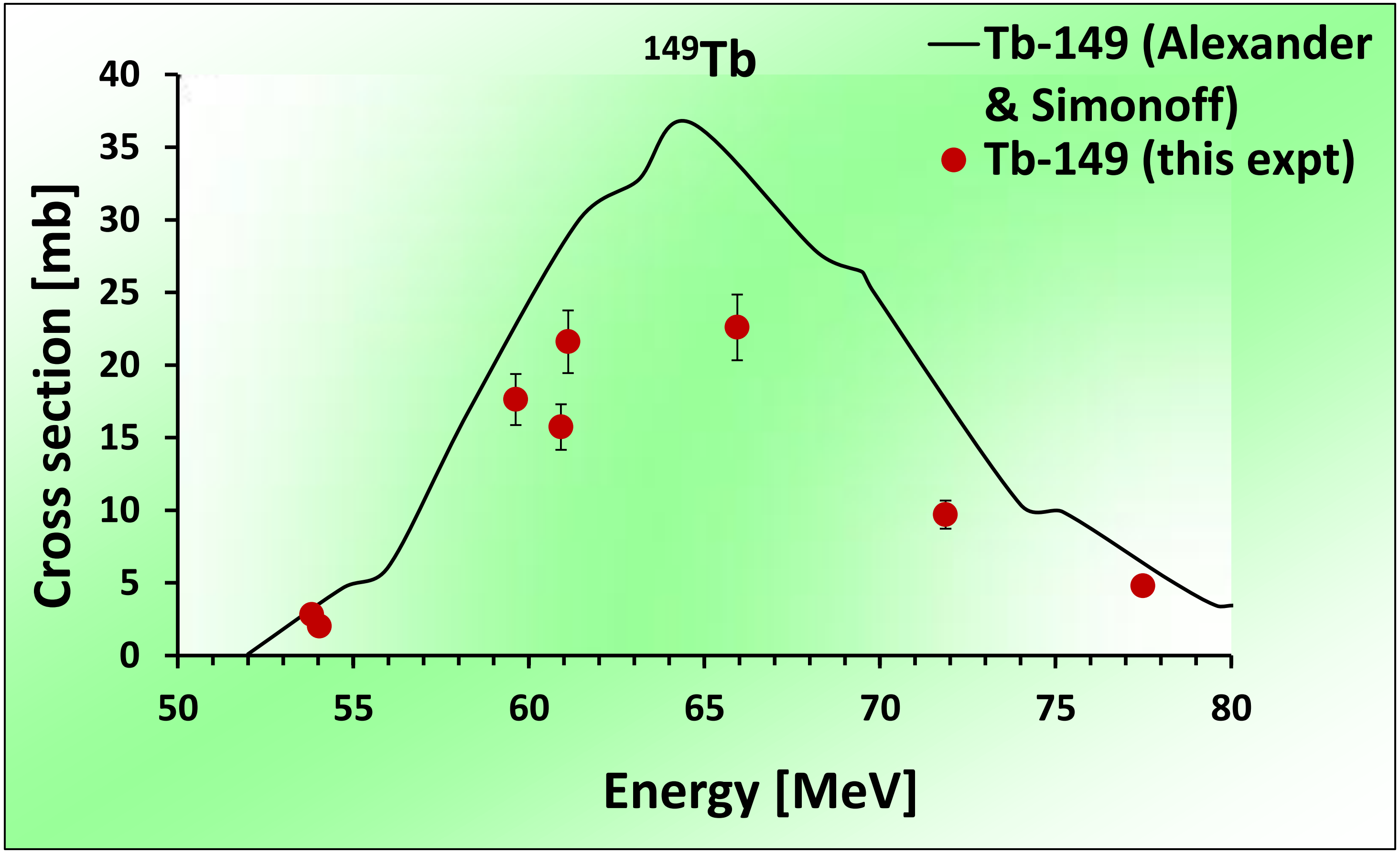


In order to understand discrepancies in the production of ^{149}Tb from the natural praseodymium target we made an attempt to measure production cross sections in the same reaction.

Our measurement…



Comparison…



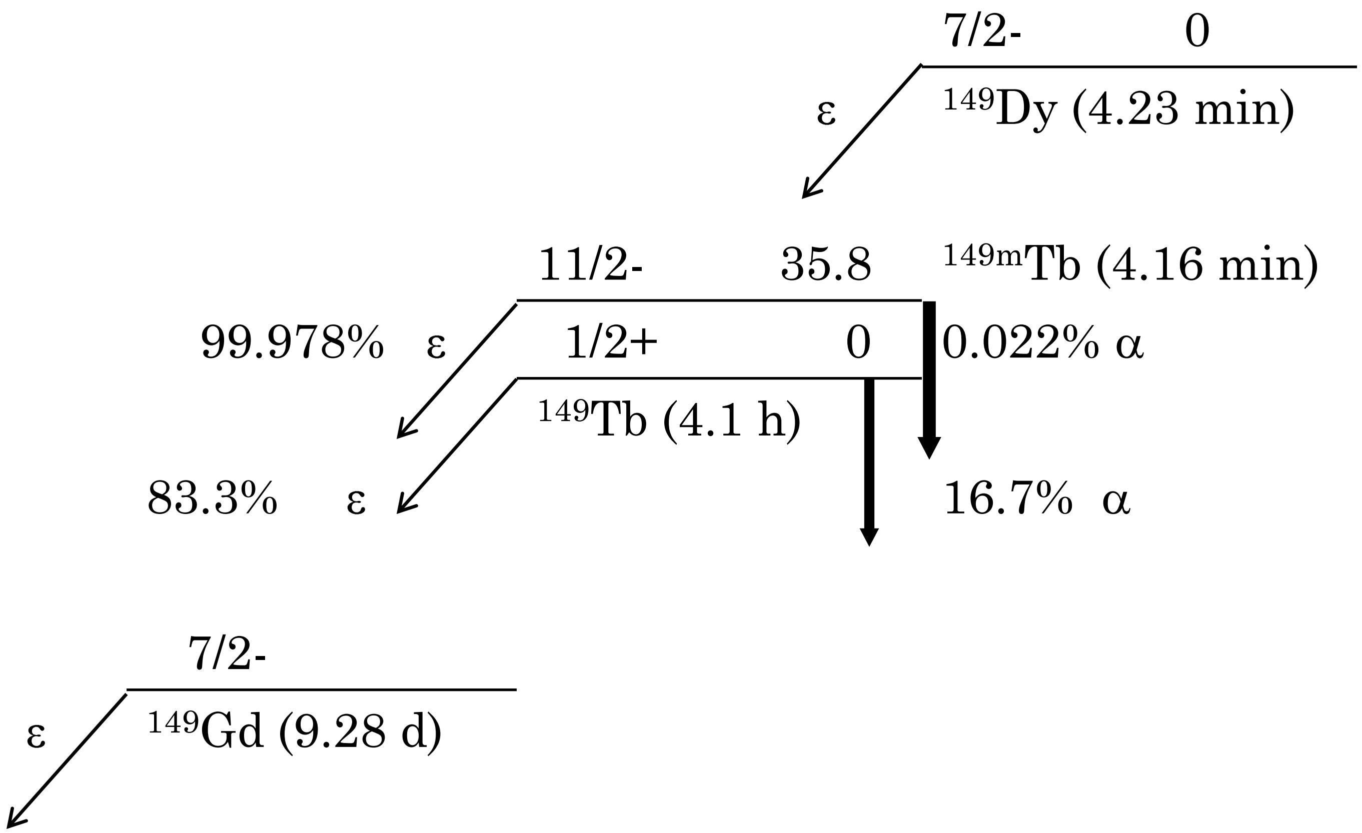
^{149}Tb
Tb-149 (Alexander & Simonoff)
Tb-149 (this expt)
Cross section [mb]
0
5
10
15
20
25
30
35
40
50
55
60
65
70
75
80
Energy [MeV]

Production of ^{149}Tb was not satisfactory

Possible reasons

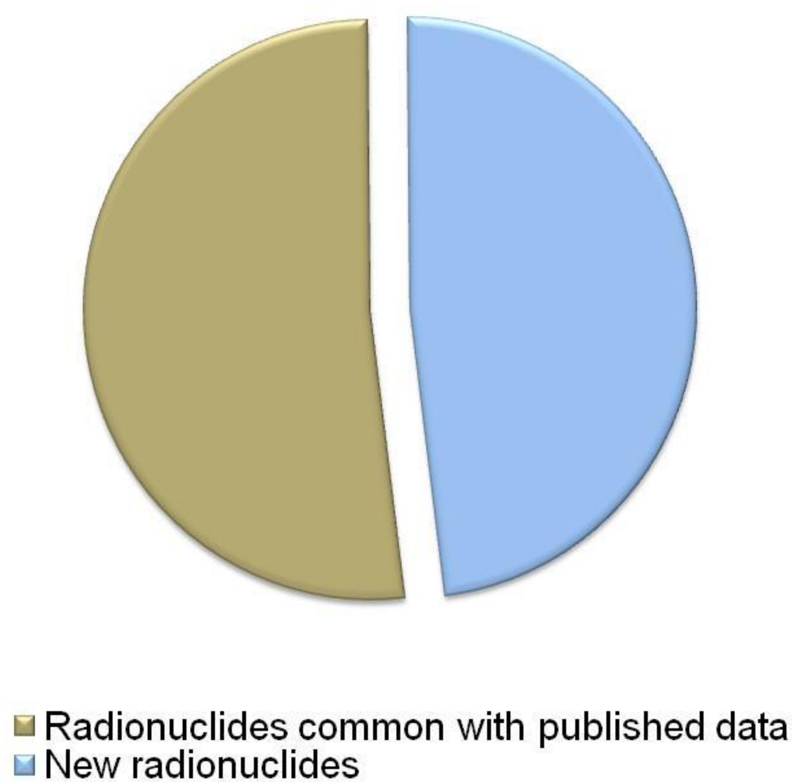
- ^{149m}Tb shows high shielding towards the production of ^{149}Tb
- The high initial angular momentum of the compound nucleus formed in $^{12}C+^{141}Pr$ reaction likely populates the high spin (11/2-) isomer (^{149m}Tb) which directly decays to ^{149}Gd.
- ^{149}Tb (spin 1/2+) is produced only from the decay of low spin states of the compound nucleus.

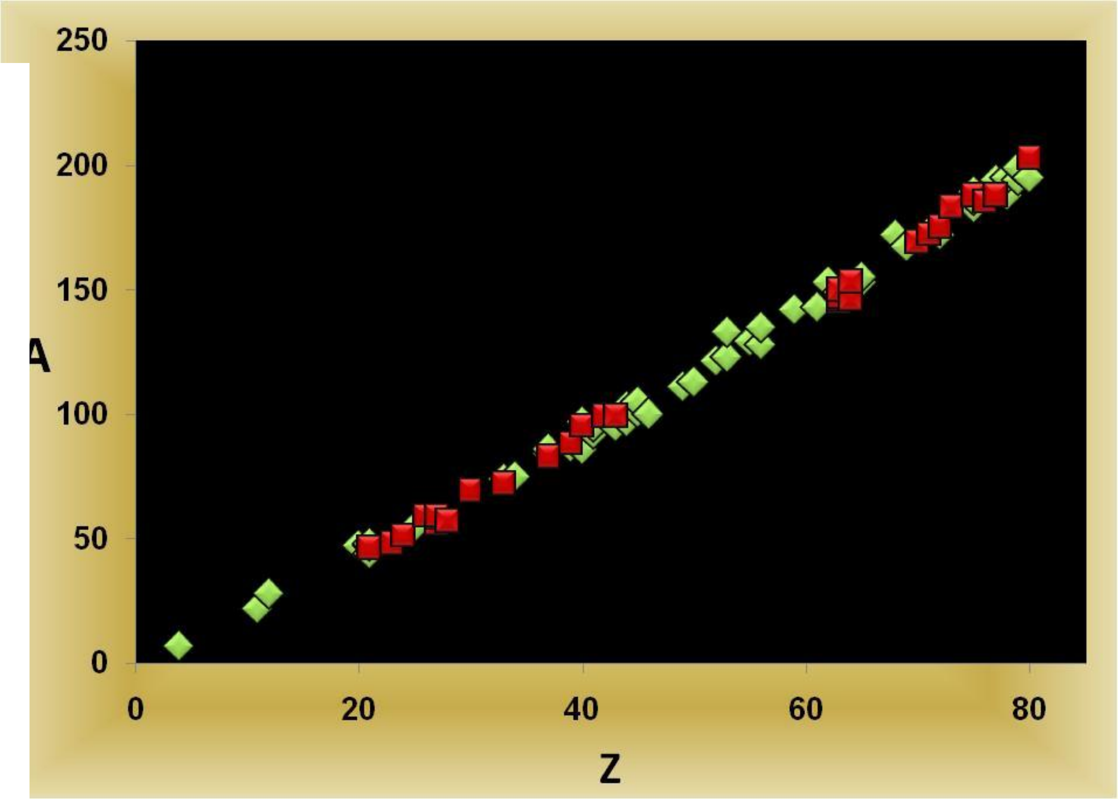
Why low cross section?



Our aim

- Identification of radionuclides
- Quantification of each radionuclide
- Development of chemical separation techniques



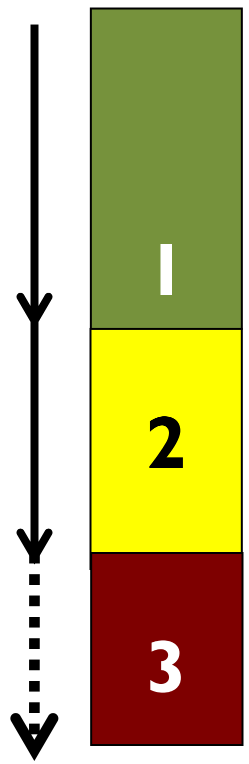


Identified probable radionuclides by our group

Results we found

Radioisotope present	Radioisotopes to be confirmed	Radioisotopes to be confirmed
As-72 (26.0 h)	As-74 (17.77 d)	Pr-142 (19.12 h)
Co-56 (77.27 d)	Au-194 (38.02 h)	Pt-188 (10.2 d)
Co-58 (70.86 d)	Au-199 (3.139 d)	Pt-195m (4.01 d)
Co-60 (1925.28 d)	Ba-128 (2.43 d)	Rb-84 (33.1 d)
Cr-51 (27.7025 d)	Ba-135m (28.7 h)	Rb-86 (18.642 d)
Eu-145 (5.93 d)	Be-7 (53.22 d)	Re-183 (70.0 d)
Eu-146 (4.61 d)	Ca-47 (4.536 d)	Re-186 (3.7186 d)
Eu-147 (24.1 d)	Co-57 (271.74 d)	Re-189 (24.3 h)
Eu-150m (12.8 h)	Cs-129 (32.06 h)	Rh-101 (3.3 y)
Fe-59 (44.495 d)	Er-172 (49.3 h)	Rh-101m (4.34 d)
Gd-146 (48.27 d)	Eu-148 (54.5 d)	Rh-105 (35.36 h)
Gd-153 (240.4 d)	Eu-149 (93.1 d)	Ru-103 (39.26 d)
Hf-175 (70 d)	Hf-172 (1.87 y)	Ru-97 (2.791 d)
Hg-203 (46.595 d)	Hg-195m (41.6 h)	Sc-44m (58.61 h)
Ir-188 (41.5 h)	I-123 (13.232 h)	Sc-47 (3.3492 d)
Lu-172 (6.7 d)	I-133 (20.8 h)	Sc-48 (43.67 h)
Mo-99 (2.7489 d)	In-111 (2.8047 d)	Se-75 (119.779 d)
Os-185 (93.6 d)	Ir-192 (73.827 d)	Sm-153 (46.284 h)
Rb-83 (86.2 d)	Ir-194 (19.28 h)	Sn-113 (115.09 d)
Re-188 (17.003 h)	Lu-173 (1.37 y)	Tb-153 (2.34 d)
Sc-46 (83.79 d)	Mg-28 (20.915 h)	Tb-155 (5.32 d)
Ta-183 (5.1 d)	Mn-54 (312.12 d)	Tc-95 (20.0 h)
Tc-99m (6.0058 h)	Na-22 (2.6027 y)	Te-121m (154 d)
V-48 (15.9735 d)	Nb-92m (10.15 d)	Tm-167 (9.25 d)
Y-88 (106.616 d)	Nb-95 (34.991 d)	Y-87m (13.37 h)
Yb-169 (32.018 d)	Ni-57 (35.6 h)	Zn-69m (13.76 h)
Zr-95 (64.032 d)	Pd-100 (3.63 d)	Zr-86 (16.5 h)
	Pm-143 (265 d)	Zr-97 (16.744 h)

Work plan



C1 Irradiation and Production of radionuclides
C2/S1 Identification of γ-emitting radionuclides
S2 Chemical separation
S3 Development of sequential separation technique of clinical radionuclides ($T_{1/2} > 7d$)

C3/S4 Identification of α-emitting radionuclides

C4/E 1 Development of chemical separation technique for short lived (≤ 1 d) radionuclides

S5 Separation and detection of long-lived ($T_{1/2}$ ~100 yr-few Myr) radionuclides

Constrains ?

Safety clearance….

Transport

Finance

Outlook…

- **It becomes important to study production of ^{149}Tb by spallation in targets like LBE.**

Aim of the project

- Identification
- Quantification
- Separation

To develop methods for separate confinement of each radionuclides with high radiochemical and radioisotopical purity. Special attention to be paid for the quantitative decontamination of bulk Hg.

Identification

Problems

- ✓ Presence of large numbers of radionuclides in the sample
- ✓ Highly complex and convoluted γ-spectra
- ✓ Presence of large numbers of parent-daughter pair, especially where (Parent) $T_{1/2}$ < (Daughter) $T_{1/2}$
- ✓ Large number of radionuclides are produced from the container (and from the trace elements present in the container)
- ✓ α and β emitting radionuclides are shielded by Hg or Pb-Bi target
- ✓ Isobaric interferences for detection of stable elements.

Approach

1. A large number of **time resolved γ-spectra** is necessary to correctly identify all radionuclides produced (at least over a time span of 1 year or more)
2. An advanced software is required to deconvolute γ peaks.
3. Series of chemical separation is required to separate the radionuclides in a lexicon way so that each separated fraction contains less number of radionuclides
4. Chemical separation is must to identify α-emitting radionuclides
5. For stable elements both **ICP-OES** and **ICP-MS** measurements will be done. ICP-OES will give information on the elements and ICPMS can give information on mass. However, sensitivity of these two techniques vary by two order of magnitudes.

Quantification

Problems

- High shielding by Pb-Bi target
- The distribution of radionuclides in both surface and bulk material make the quantification more complicated
- Convoluted peaks
- α and β emitting radionuclides are shielded by Hg or Pb-Bi target

Approach

- Chemical separation of each radionuclide
- Comparison with standard calibrated source
- Calculation of chemical yield (separation efficiency) for each radionuclides.
- Simulation studies
- For stable elements (or long-lived radionuclides) ICPMS data will be compared with the standard

Separation

Problems

- **Scale of separation:** **Huge amount of Hg is present while the products are present in trace quantity.**
- **Traditional difficulties of separation of chemically similar elemental pair (For example, Zr-Hf, Mo-W, lanthanides, etc).**

Approach

(A) Chemical techniques

- **Liquid liquid extraction (LLX)**
- **Aqueous biphasic extraction**
- **Ion exchange and other chromatographic techniques**
- **Precipitation etc.**

(B) Physicochemical techniques

- **Adsorption of radionuclides on hot and cold metal surfaces**
- **Thermochromatography**

Effort should be given to develop greener technologies, i.e., not to generate additional hazards

Work plan

Time scale : 5 years

1

1. Identification of γ-emitting radionuclides ($T_{1/2} \geq 1$ d)
2. Chemical separation
3. Development of sequential separation technique of clinical radionuclides
4. Study on the distribution of reaction products

2

Identification of α-emitting radionuclides

Development of chemical separation technique for short lived (≤ 1 d) radionuclides

Place: CERN/Near the source of irradiation

3

Separation and detection of long-lived ($T_{1/2}$ ~100 y-few My) radionuclides which has high demand in basic science

Work report available in this direction…

- EURISOL-DS/Task2 Report of Neuhausen et al. from PSI
- Large number of radionuclides were identified
- Isolation of some radionuclides from liquid Hg target

Simulation experiment in our lab

Liquid Hg in capped stainless steel vial

^{99}Mo-^{99m}Tc activity injected in liquid Hg and homogeneous mixed in Hg by sonication

Radiochemical extraction of ^{99}Mo-^{99m}Tc activity from bulk Hg by liquid-liquid extraction using

- Dil HCl (pH = 2)
- Hot (~65°C) and Cold (~26°C) water

Findings......

- **Mercury shows high shielding of ~90%**
- **A major part of the ^{99}Mo-^{99m}Tc (~80%) goes to Hg**
- **Rest amount is almost evenly distributed between the surface of the steel capsule and in the tiny amount of aqueous solution, which carried the total activity to the mercury.**

Extracting phase	Dil HCl (pH = 2)	Cold (~26°C) water	Hot (~65°C) water
Extraction of activity	~20%	~50%	~20%

Facilities in SINP……

- HPGe detectors
- NaI(Tl) detector
- Compton suppression system
- α-spectrometer
- Approved radioanalytical laboratory

- ICP-OES
- ICP-MS
- HPLC
- GC

(ICP-OES, ICP-MS: Laser ablation)

Future scope

Once the separation protocol of the radionuclide bank is established, application of radionuclides in various fields will be easy to many research groups.

On behalf of Radiochemistry group of SINP

Thank you….