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

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EURISOL facility

- Large volume of liquid Hg will be used as neutron converter target as well as coolant
- Large number and huge amount of radionuclides will be produced in the converter targets: Hg when bombarded by a few GeV high current proton beam
- Continuous source of radionulcides

Why radionuclide bank?

- Some of 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

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 $(Parent)_{T1/2} < (Daughter)_{T1/2}$**
- ✓ **Large number of radionuclides are produced from the steel 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 (at least over a time span of 1 year or more)
2. An advanced software is required to deconvolute γ peaks.
3. Hg targets should be irradiated in high heat sustaining carbon container in addition to a SS container to exclude the radionuclides produced from steel container
4. Series of chemical separation is required to separate the radionuclides in a lexicon way so that each separated fraction contains less number of radionuclides
5. **Compton suppressed γ-spectrum** will be highly helpful
6. Chemical separation is must to identify α-emitting radionuclides
7. For stable elements both **ICP-OES** and **ICP-MS** measurements will be done along with NAA.

   (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 Hg/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.
- **The handling of bulk mercury is a big problem with respect to researchers health and safety.**
- **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.**

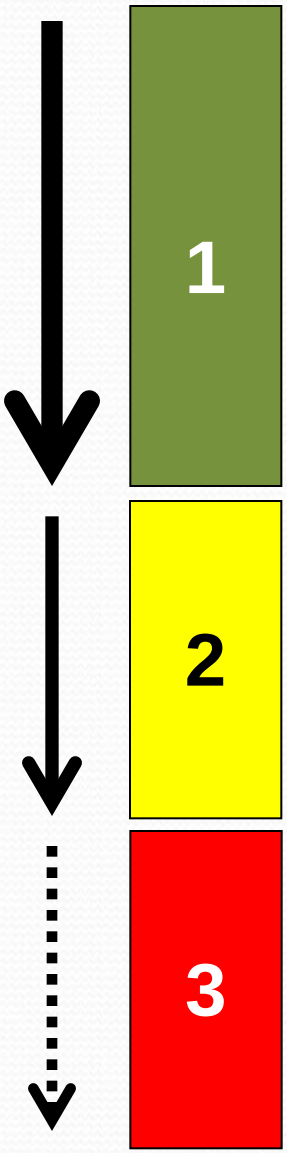
(B) Physicochemical techniques

- **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. 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

Place: **Radiochemistry laboratory**
Saha Institute of Nuclear physics, INDIA

Identification of α-emitting radionuclides

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

Place: CERN/Near the source of irradiation

Separation and detection of exotic ($T_{1/2}$ ~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

Our experience towards the project

- Analysis of γ-spectra of CERN irradiated two Hg samples collected at PSI

  (Irradiation : 21st April, 2006 with 1.5x10^{15} protons of 1.4 GeV for 7-8 hours)

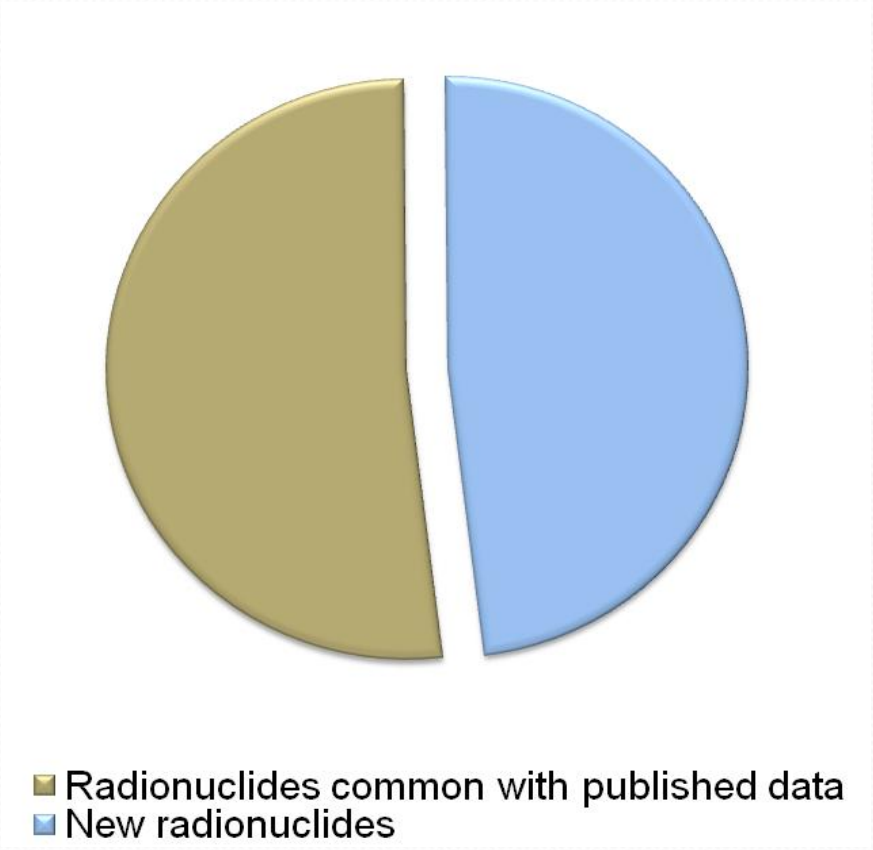
- Samples are CERN1 and CERN2

- We were able to identify some of the radionuclides produced in CERN 2 sample

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)

A brief comparison



Important to look

- To know the actual source of radionuclides

- p + steel container = production of $^{57,60}Co$?

or

- p + Hg = production of $^{57,60}Co$?

or

- both?

- needs irradiation of Hg in another container (preferably C) and comparison between the spectrum?

Facilities in SINP……

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

- ICP-OES
- ICP-MS

} Laser ablation

- HPLC
- GC

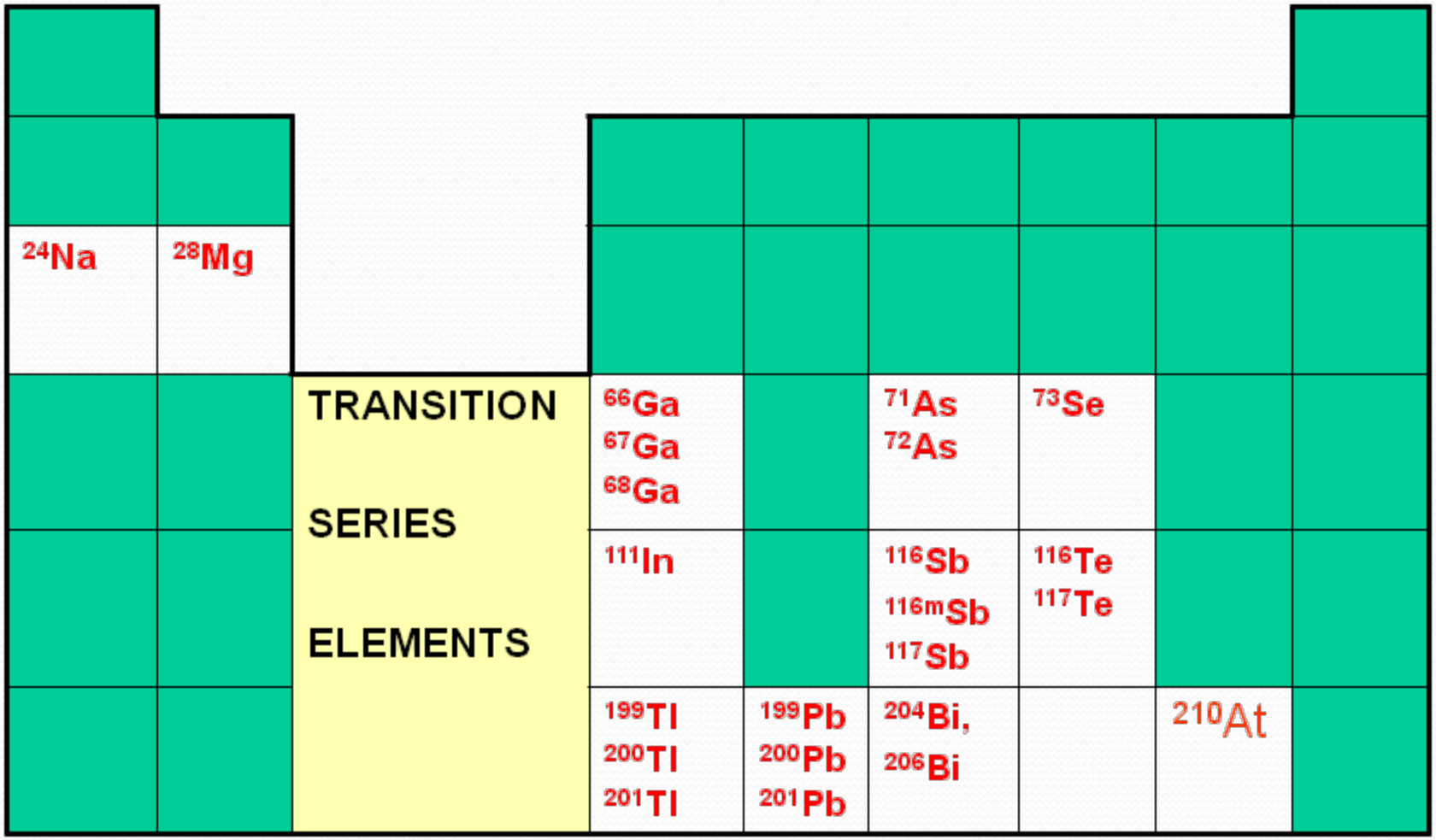
Our experience

Light and heavy ion induced production and separation of no-carrier-added radionuclides

- ^{199}Tl
- ^{111}In
- ^{211}At
- $^{204,206}Bi$
- ^{61}Cu, $^{62,63}Zn$, $^{66,67,68}Ga$
- $^{71,72}As$, ^{73}Se,
- $^{116,117}Te$, $^{16,116m,117}Sb$
- ^{95}Tc
- ^{48}V and $^{48,49}Cr$

- ^{166}Ho

NCA radionuclides produced and separated



Transition series elements

^{46}Sc		^{48}V	^{48}Cr ^{49}Cr ^{51}Cr	^{52}Mn, ^{56}Mn		^{55}Co, ^{56}Co, ^{58}Co	^{56}Ni, ^{57}Ni	^{61}Cu	^{62}Zn ^{63}Zn
^{90}Y	^{89}Zr	^{90}Nb, ^{91m}Nb	^{93m}Mo	^{93}Tc, ^{94}Tc, ^{99m}Tc	^{94}Ru, ^{95}Ru, ^{97}Ru	^{101}Rh, ^{105}Rh, ^{106}Rh		^{103}Ag, ^{104}Ag, ^{105}Ag	^{104}Cd, ^{105}Cd, ^{107}Cd
	^{170}Hf ^{171}Hf	^{176}Ta ^{177}Ta	^{176}W ^{177}W	^{181}Re		^{187}Ir ^{187}Ir	^{187}Pt ^{188}Pt	^{192}Au ^{193}Au ^{199}Au	^{192}Hg ^{193}Hg

Lanthanide series elements

^{133}Ce ^{135}Ce					^{145}Eu ^{146}Eu ^{147}Eu	^{147}Gd ^{149}Gd
^{150}Tb ^{151}Tb ^{152}Tb	^{150}Dy ^{151}Dy ^{152}Dy	^{158}Ho ^{159}Ho ^{160}Ho		^{163}Tm ^{165}Tm ^{166}Tm	^{166}Yb ^{167}Yb	^{167}Lu ^{170}Lu ^{171}Lu

Separation of isobaric pairs

1. Separation of ^{53}Mn from ^{53}Cr--- for better understanding of Earth's surface processes

   Analytical Chemistry, 78 (2006) 7517

2. Separation of ^{146}Sm from ^{146}Nd--- a prerequisite for getting signals from nuclear synthesis

   The Analyst, 131 (2006) 1332

3. Separation of ^{182}Hf and ^{182}W--- a step toward to solve astronomical puzzle

   Analytical Chemistry, 78 (2006) 2302

Technical support from CERN

- Proton irradiated samples in TWO capsules (SS & C)
  - (i) Liquid Hg
  - (ii) molted Pb-Bi
- Each sample will contain ~ 5mCi when they will be dispatched from CERN
- Specific design of packing is required for the necessary permission from the Government of India for shipping of the active sample
- Technical support to develop thermochromatographic method
- Annual technical meeting to evaluate the progress of the project
- Financial support

Future scope

- **Once the standard 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….