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

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Source?

- Large volume of liquid Hg or Pb-Bi 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/Pb-Bi when bombarded by a few GeV high current proton beam
- Continuous source of radionulcides

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

Aim of the project

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 steel container (and from the trace elements present in steel container)
- ✓ α and β emitting radionuclides are shielded by Hg or Pb-Bi target
- ✓ Isobaric interferences for detection of stable elements.

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. 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. 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

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.**

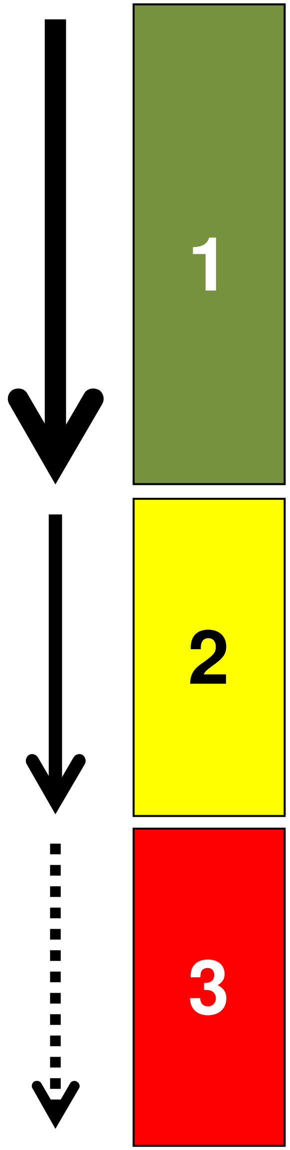
  (B) Physicochemical techniques

  - **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 ($\leq$ 1 d) radionuclides

Place: CERN/Near the source of irradiation

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

ork 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

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

(Irradiation : 21st April, 2006 with 1.5×10^{15} protons of 1.4 GeV for 7-8 hours)

- Samples are CERN1 and CERN2

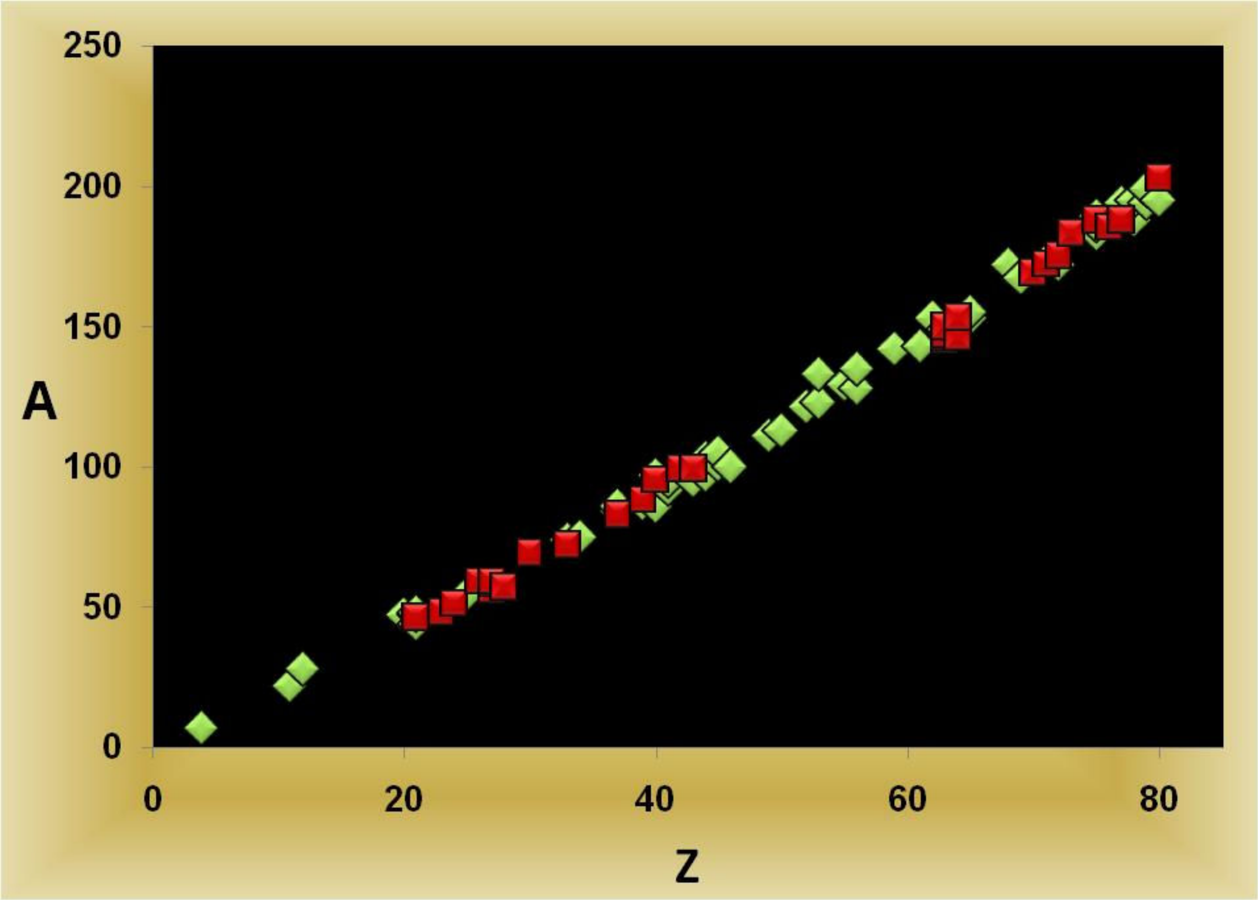
- We were able to identify some of the radionuclides produced in CERN 2 sample

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)

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

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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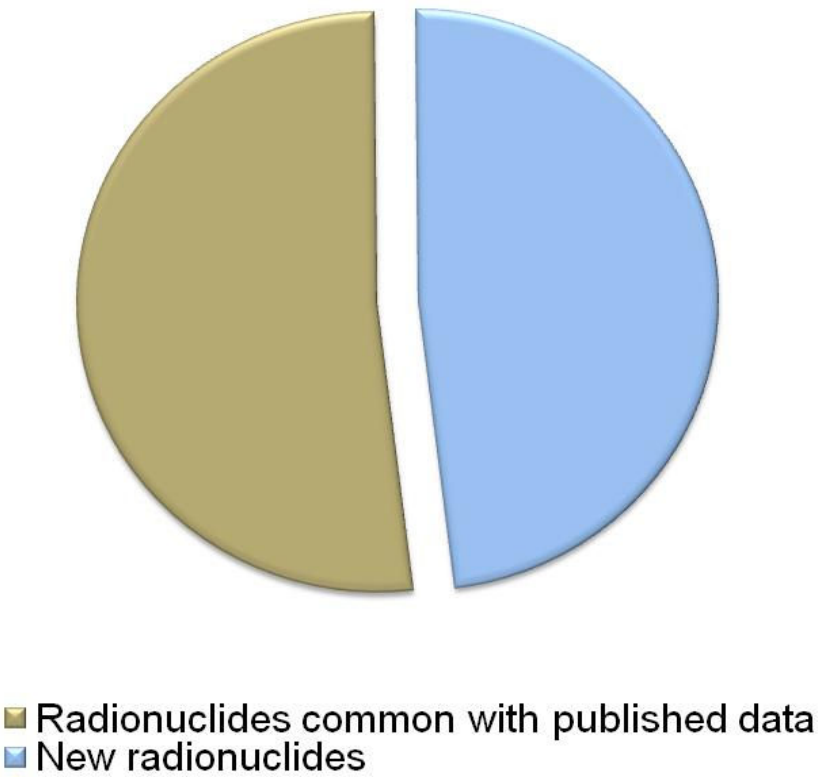

Radionuclides common with published data
New radionuclides

- **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?**

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

indings……

- **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%

acilities in ……

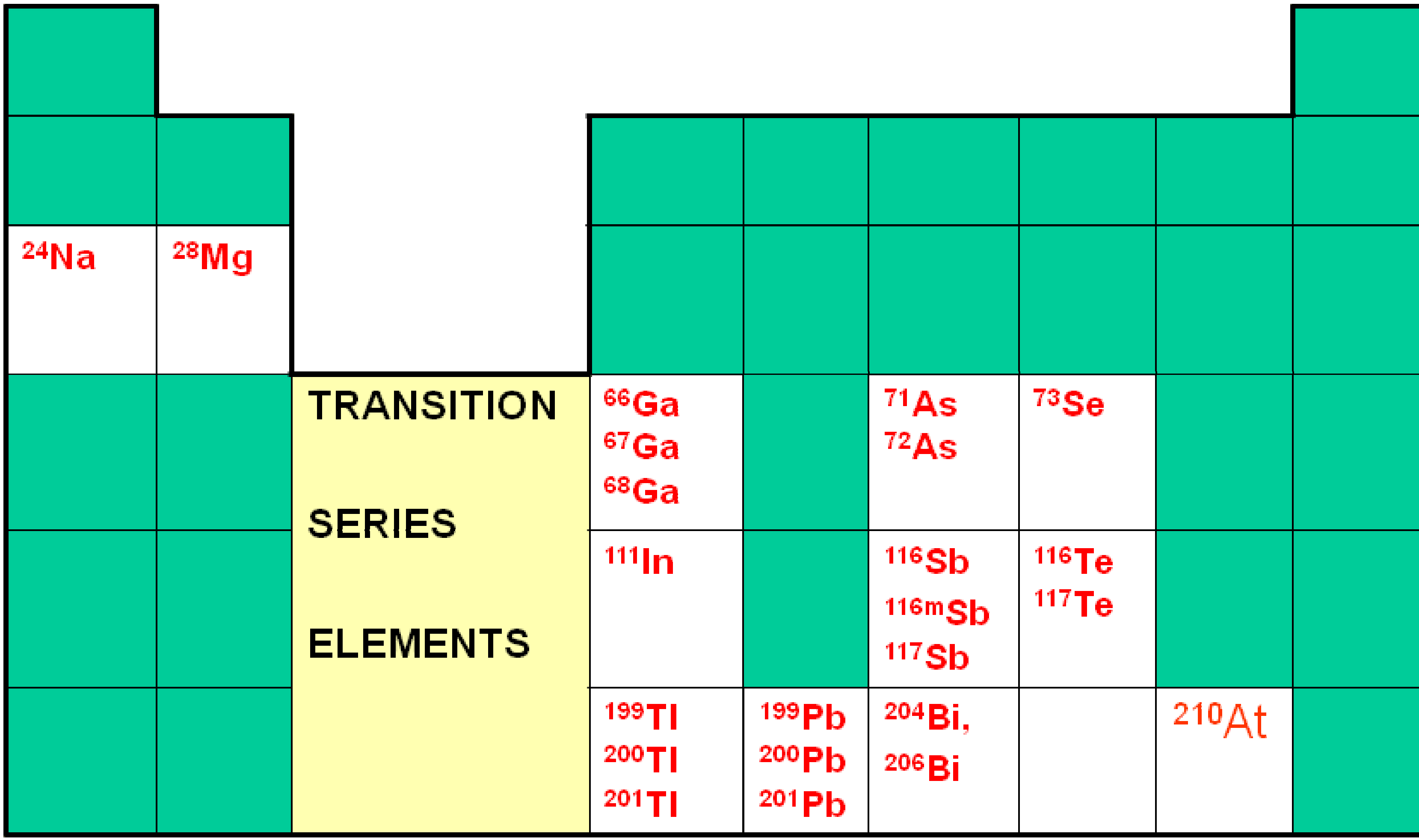
- HPGe detectors
- NaI(Tl) detector
- Compton suppression system
- α-spectrometer
- Approved radioanalytical laboratory

Laser ablation

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



^{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

^{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

for better understanding of arth's

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

- **Production of ^{163}Ho : interest of basic science**

- **Online thermochromatographic studies**

Thank you….