Chemistry at the lead bismuth loop for EURISOL prototype

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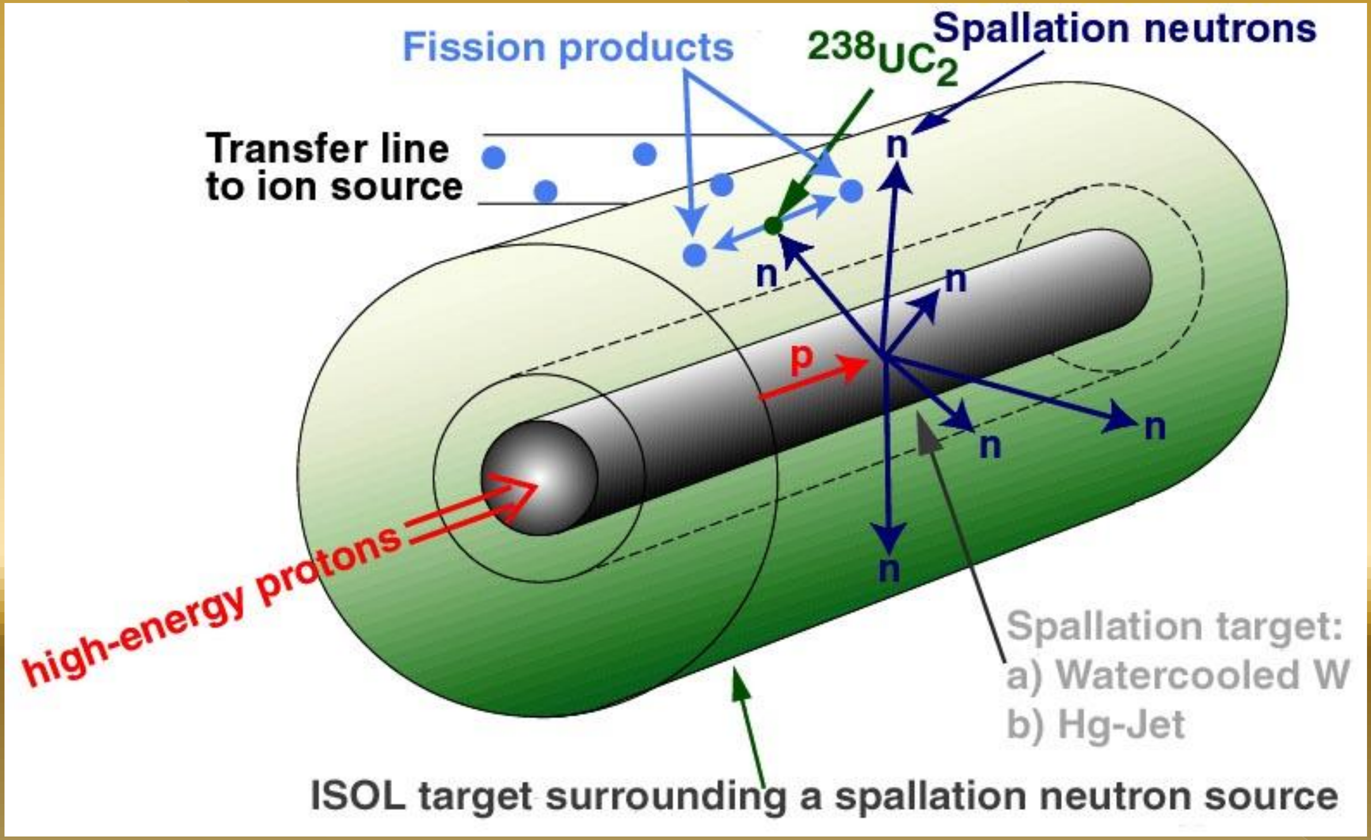
Liquid lead-bismuth eutectic : prototype converter target in proposed EURISOL project.

Composition:	**44.5% lead and 55.5% bismuth**
Melting temperature	**123.5 °C**
Boiling temperature	**1670 °C.**

It has good heat transfer properties.

The EURISOL Project
(The ultimate ISOL facility)



Source : EURISOL facility

- Large volume of liquid Hg or Pb-Bi target will be used as 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

This multi-MW converter target of the proposed "*next generation*" European Isotope Separation On-Line (EURISOL) facility may be able to serve as a potential alternative source of several pronounced radionuclides

- **Identification of these radionuclides are important**
  - They can be enormous source of useful radionuclides in all branches of sciences **especially for clinical applications**.

Why large facilities became so important?

1

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

How to identify?

The high shielding from high Z targets is the main constrain for identification of these radionuclides.

Quantification of each radionuclide is important

- To make exact inventory of each radionuclide is important for commercial purpose.
- The successful commercial application may even be helpful to share the cost of such facilities for basic sciences.

2

How to Quantify?

Constrains:

- *High shielding by high Z target*
- *Irregular geometry*
- *Large number of radionuclides with overlapping peaks.*

PROBLEM + SOLUTION

RADIOCHEMICAL SEPARATION

We would like to develop chemical separation technique for each radionuclides with high radiochemical and radio-isotopical purity --- therefore fulfilling the primary requirement for clinical radionuclides

3

Our proposal is to build *Radionuclide Bank* from proton irradiated Pb-Bi targets

Techniques

Identification & quantification of radionuclides

- γ-spectrometry by HPGe detectors
- ICP-OES and ICP-MS

Chemical separation

- Radioanalytical techniques
  - (i) LLX
  - (ii) Ion-exchange chromatography
  - (iii) Adsorption
  - (iv) Amalgamation …… etc.
- Thermochromatography

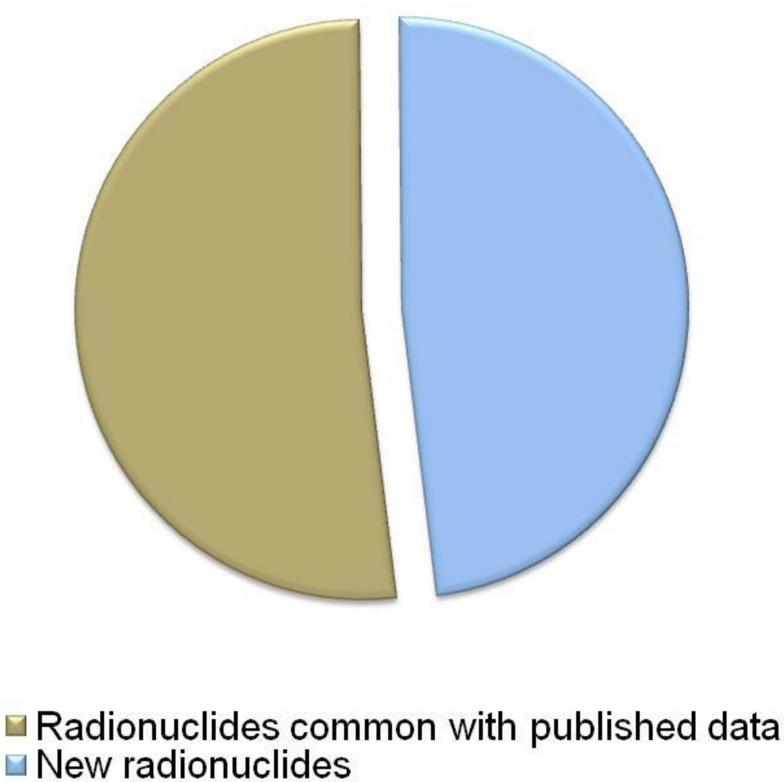
Identification of radionuclides of proton irradiated Hg target

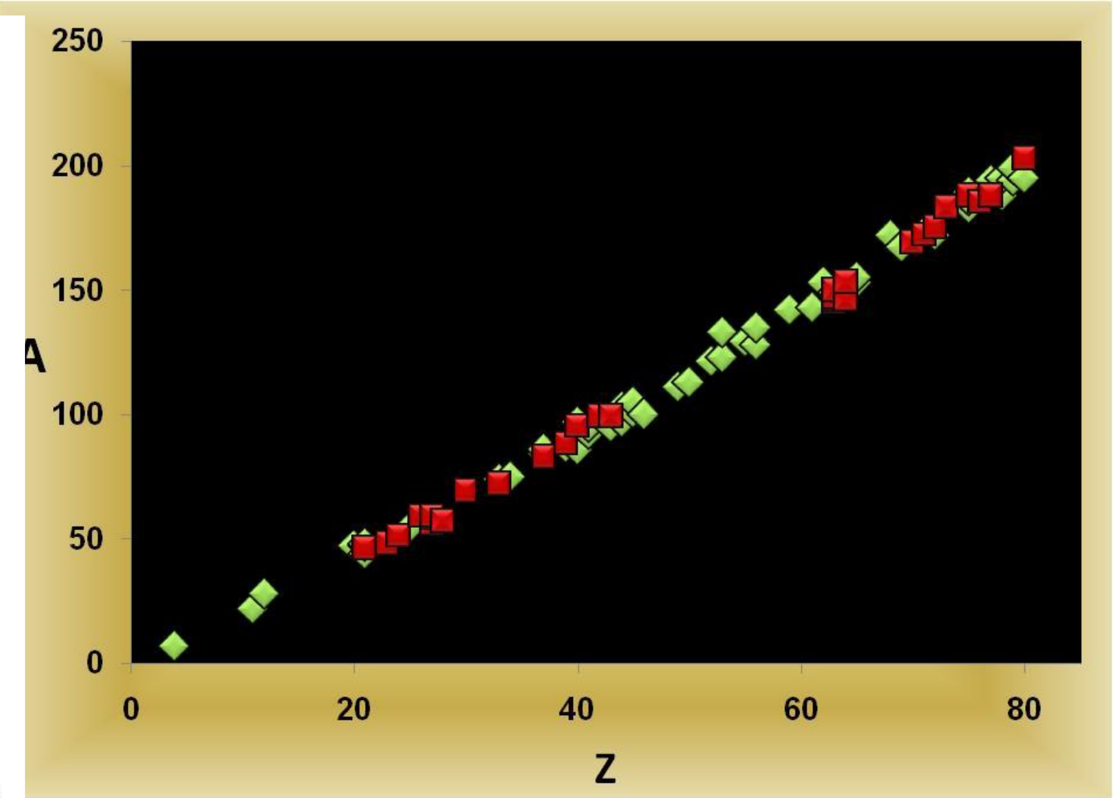
Work report available in this direction

- ☛ Report of Neuhausen et al. from PSI
- ☛ Large number of radionuclides were identified
- ☛ Isolation of radionuclides from liquid Hg target

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)





Identified probable radionuclides by our group

Behavior of ^{99}Mo-^{99m}Tc activity in bulk Hg environment

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. HNO_3 (pH =1)
- Dil HCl (pH = 2)
- Hot (~65°C) and Cold (~26°C) water

Findings……

- **Mercury shows high self-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.**
- **As per expectation, bulk mercury partially dissolved in dil HNO_3 removing the clarity of the transparent HNO_3 phase during distillation of ^{99}Mo-^{99m}Tc activity from bulk mercury.**

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

Other important properties to be investigated:

- Long term proton irradiation will introduce enough activity in the LBE assembly, which needs to be separated at a regular frequency in order to recycle the target matrix.
- Emphasis will be given to build a data bank of various thermodynamic parameters like adsorption enthalpy, sublimation enthalpy for these radionuclides on Pb-Bi surface.
- The study will be extended to the migration behavior of these radionuclides in various solid materials like stainless steel, glass, graphite, etc.

Thank you…..