EUROPEAN ORGANISATION FOR NUCLEAR RESEARCH

Proposal to the ISOLDE and Neutron Time-of-Flight Committee

^{206}Po sources for production and release studies relevant for high power spallation targets

25th September 2013

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CERN-INTC-2013-048 / INTC-P-401
25/09/2013

Abstract: The knowledge of the evaporation behaviour of Po is of essential importance for several scientific and technological applications, like accelerator driven systems (ADS) or the LIEBE project at CERN-ISOLDE. Fundamental investigations on the experimental conditions for the formation of volatile Po species as well as on the chemical composition of the volatile compounds are necessary for a safe operation of such facilities. ^{206}Po, a mainly γ-ray-emitting Po isotope with a half-life of 8.8 d, is best suited for model studies, due to the lower radiation hazard compared to the longer-lived α-emitting isotopes $^{208-210}$Po as well as the easy-to-measure γ-ray emission. We propose the production of ^{206}Po samples in several matrices via the implantation of its precursor ^{210}Fr into selected metal foils at CERN-ISOLDE. Using these samples, experiments will be carried out at PSI studying the volatilization of Po from different matrices under varying chemical conditions.

Requested shifts: 12 shifts, (split into 4 runs over 2 years)

Motivation

Polonium is one of the most hazardous radioelements produced during irradiation of targets made of lead or heavier elements with high-energetic protons or secondary particles, as for instance already applied in the neutron spallation source SINQ at PSI, or foreseen for Accelerator Driven Systems like MYRRHA as well as for the development of radioactive beams in the frame of the LIEBE project at CERN-ISOLDE. The hazard caused by this element is due to its isotopes ^{208,209,210}Po, all α-emitters, partially with comparably long half-lives (^{209}Po being the longest-lived with a half-life of 102 a), combined with a high volatility of the chemical element and/or its compounds.

Our research group is currently involved in both the key projects MYRRHA and LIEBE, with our expertise lying mainly in the study of the release properties of volatile elements from spallation targets. While MYRRHA is foreseen as a prototype for an ADS design, being developed at SCK-CEN Mol, Belgium, with a liquid metal target consisting of Lead-Bismuth-Eutectic (LBE), LIEBE is a CERN-initiated project aimed on the development of a radioactive beam out of an irradiated LBE loop, with a particular focus on Po beams. Depending on the special objective of the project, Po release has to be avoided (ADS) or fostered (LIEBE). In both cases, a better understanding of the fundamental chemical processes involved in the formation of volatile Po species is mandatory.

State of the Art in the research field

The temperature dependent saturation vapor pressure of elemental Po was determined by Brooks [1] and Abakumov [2]. Although purified Po was used for the determination, Abakumov already in these studies proposed the formation of a compound - PbPo, the lead resulting from accumulation of small amounts of radiogenic Pb – being the volatile species. Vapor pressure data for the evaporation of Po from LBE of Ohno [3] and Buongiorno [4], were found to be in fair agreement with Abakumov's data when one assumes an ideal solution of PbPo in LBE, leading to the conclusion that Po evaporates from LBE predominantly as PbPo [4]. In general, most of these studies cover a temperature range above 400°C. Hence, there is still a comparable big lack of precise and reliable data on the volatility properties especially under conditions relevant to the operation of facilities like MYRRHA or LIEBE.

First own investigations [5] on the determination of the Henry constant of Po in dilute solution in LBE in the temperature range below 400°C in an inert (He) atmosphere showed a significant deviation from the behaviour that was expected based on literature data and their exptrapolation to lower temperatures (Fig.1).

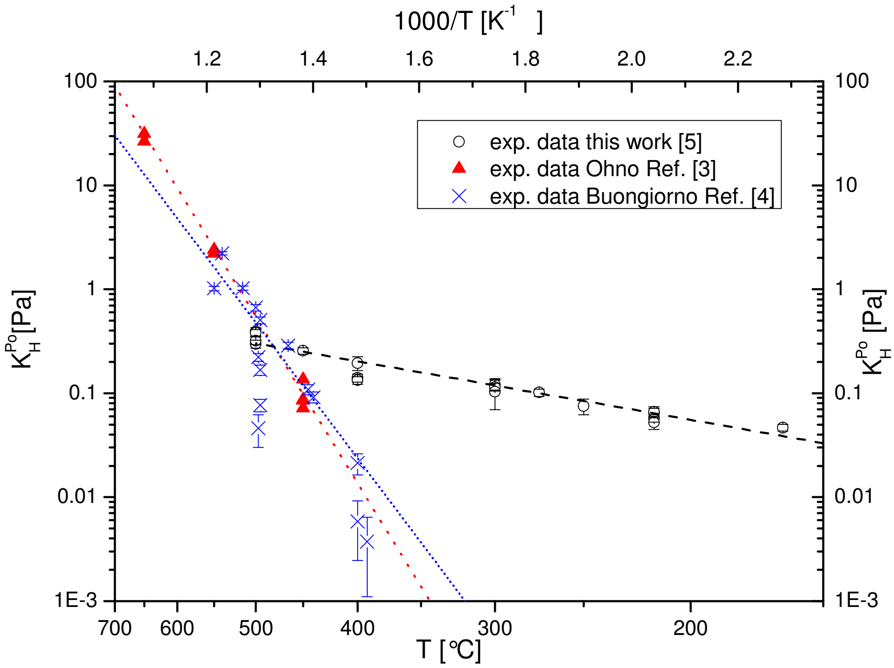

Figure 1: Comparison of Henry constant data of Po over liquid LBE under inert gas atmosphere and corresponding functions according to [3] (red), [4] (blue) and [5] (black)

In a similar series of experiments in hydrogen atmosphere, an even more volatile Po species was found that was transported at room temperature to the exhaust filter of the apparatus. However, this very volatile species was only found when a thin oxide layer was present on the surface of the LBE samples. Similar observations are reported also in [6], explaining the phenomenon by assuming the formation of volatile H_2Po in the presence of moisture and/or H_2. On the other hand, results of our thermochromatography experiments [7] indicate that in a pure hydrogen atmosphere no enhanced volatilization of Po is observed. In conclusion, the chemical composition of the volatile species and the conditions for their formation remain unclear. Thus, additional systematic experiments on Po volatilization under different chemical conditions are necessary.

The performance of such experiments is nowadays additionally complicated by the poor availability of Po and the radiation protection requirements necessary for the handling of highly active α-samples. The use of the short-lived ^{206}Po would both simplify the handling of the activity during the experiment concerning the radiation protection measures and the measurement itself, because γ-rays in a suitable energy range can be measured without any additional sample preparation treatment. Furthermore, the conventional production methods for ^{206}Po (p-irradiation of Bi or α-irradiation of Pb) always result in samples containing a large excess of Bi or Pb. ^{206}Po production by implantation of precursors into different matrices enables us to study the volatilization of Po in the absence of Bi or Pb, ruling out the possibility of formation of diatomic BiPo or PbPo molecules and thus providing more fundamental data for the element itself.

Scientific objectives

The work is aimed to perform systematic studies of the experimental conditions under which Po species of different volatility are formed and how their release from several matrix materials may be enhanced or reduced. The experimental parameters to be varied are in particular the composition of the gas plenum, e.g. moisture as well as hydrogen and/or oxygen content, oxide layer formation on the matrix surface, temperature dependence as well as the possible formation of radicals. These fundamental studies will also cover the influence of the matrix material (Pb, Bi, LBE, impurities) on the volatilization. The determinations will be carried out using the well-known "transpiration method", which had been recently adapted at PSI for first test experiments, with the experimental apparatus set up in an inert gas box, providing pure gas atmospheres with minimum concentrations of unwanted impurities.

For investigating the nature of the chemical species, thermo-chromatographic studies are foreseen, enabling us to determine adsorption enthalpies on various materials. Apart from the extension of the basic knowledge on Po chemistry, our final aims of the project are to establish which are the experimental conditions suitable for

A) minimizing possible release of Po – regardless of the chemical form - from LBE in MYRRHA

B) maximizing evaporation of the most volatile Po species for the development of a Po beam in the frame of LIEBE

Summary of requested shifts

We estimate a total of 12 shifts of beam time distributed within two years. ^{206}Po can be obtained by implanting a precursor isotope ^{210}Fr ($T_{1/2}$=3.18 m) from an UC_x target with a tungsten surface ionizer. The implantations will be made in the solid state collection chamber in the GLM beam line. Other physics experiments using higher masses can proceed in parallel in the central beam line.

References

[1] L. S. Brooks, J. Am. Chem. Soc., 77 (1955) 3211.

[2] A.S. Abakumov, Z.V. Ershova, Sov. Radiochim., 16 (1974) 397–401.

[3] S. Ohno, Y. Kurata, S. Miyahara, S. Yoshida, J. Nucl. Sci. Tech., 43 (2006) 1359–1369.

[4] J. Buongiorno, C. Larson, K.R. Czerwinski, Radiochim. Acta, 91 (2003) 153–158.

[5] M. Rizzi et.al. unpublished results

[6] P.E. MacDonald, J. Buongiorno (eds.), Design of an Actinide Burning, Lead or Lead-Bismuth Cooled Reactor that Produces Low Cost Electricity, INEEL report INEEL/EXT-02-01249 (2002) 211 p.

[7] E. Maugeri et.al., JNM 2013, submitted

Appendix

DESCRIPTION OF THE PROPOSED EXPERIMENT

The experimental setup comprises: *(name the fixed-ISOLDE installations, as well as flexible elements of the experiment)*

Part of the Choose an item.	Availability	Design and manufacturing
SSP-GLM chamber	☒ Existing	☒ To be used without any modification
	☐ Existing	☐ To be used without any modification ☐ To be modified
	☐ New	☐ Standard equipment supplied by a manufacturer ☐ CERN/collaboration responsible for the design and/or manufacturing
	☐ Existing	☐ To be used without any modification ☐ To be modified
	☐ New	☐ Standard equipment supplied by a manufacturer ☐ CERN/collaboration responsible for the design and/or manufacturing
[insert lines if needed]		

HAZARDS GENERATED BY THE EXPERIMENT

(if using fixed installation) Hazards named in the document relevant for the fixed [COLLAPS, CRIS, ISOLTRAP, MINIBALL + only CD, MINIBALL + T-REX, NICOLE, SSP-GLM chamber, SSP-GHM chamber, or WITCH] installation.

Additional hazards:

Hazards	*[Part 1 of the experiment/equipment]*	*[Part 2 of the experiment/equipment]*	*[Part 3 of the experiment/equipment]*
Thermodynamic and fluidic			
Pressure	10^{-6} mbar		
Vacuum			
Temperature	Room Temperature		
Heat transfer			
Thermal properties of materials			
Cryogenic fluid			
Electrical and electromagnetic			
Electricity			
Static electricity			
Magnetic field			
Batteries	☐		
Capacitors	☐		
Ionizing radiation			
Target material	PbBi samples		
Beam particle type (e, p, ions, etc)	Ions		
Beam intensity	1×10^{8} ions/s		
Beam energy	30-60 kV (from the separator)		
Cooling liquids			
Gases			

Calibration sources:	☐		
• Open source	☐		
• Sealed source	☐ [ISO standard]		
• Isotope			
• Activity			
Use of activated material:			
• Description	☐		
• Dose rate on contact and in 10 cm distance	100 µSv/hr at 10 cm		
• Isotope	^{206}Po		
• Activity	< 1 MBq		
Non-ionizing radiation			
Laser			
UV light			
Microwaves (300MHz-30 GHz)			
Radiofrequency (1-300MHz)			
Chemical			
Toxic	PbBi		
Harmful			
CMR (carcinogens, mutagens and substances toxic to reproduction)			
Corrosive			
Irritant			
Flammable			
Oxidizing			
Explosiveness			
Asphyxiant			
Dangerous for the environment			
Mechanical			
Physical impact or mechanical energy (moving parts)			
Mechanical properties (Sharp, rough, slippery)			
Vibration			
Vehicles and Means of Transport			
Noise			
Frequency			
Intensity			
Physical			
Confined spaces			
High workplaces			
Access to high workplaces			
Obstructions in passageways			
Manual handling			

Poor ergonomics			

0.1 Hazard identification

3.2 Average electrical power requirements (excluding fixed ISOLDE-installation mentioned above):
Standard electrical usage at ISOLDE hall