Wir schaffen Wissen – heute für morgen

Paul Scherrer Institut

Matthias Rizzi, Jörg Neuhausen

Polonium Evaporation Studies from Liquid Metal Spallation Targets

INDEX

- **INTRODUCTION**
- **POLONIUM RELEASE**
- **EXPERIMENTAL TECHNIQUE**

  transportation method - theory & experimental
- **RESULTS & DISCUSSION**
- **SUMMARY & OUTLOOK**

Radiological issues of spallation targets

- due to the high beam intensity and the large variety of nuclear interactions the target material becomes highly radioactive
- for a target material with a proton number N, every nuclide from 1 to N +1 can be formed
- production of highly radiotoxic polonium 210 due to neutron capture of bismuth
- lack of thermodynamical data for polonium (vapor pressure, gaseous species)

INTRODUCTION

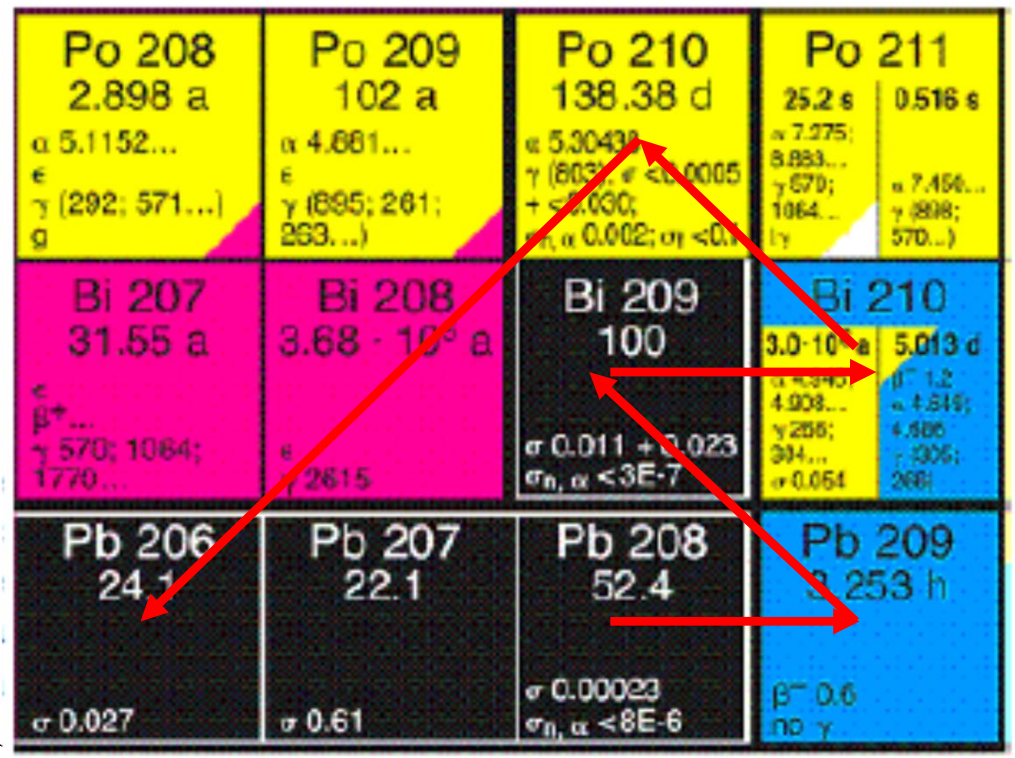
^{210}Po is produced by neutron capture of ^{209}Bi and its following β^- decay

$$^{209}Bi + {}^{1}_{0}n \rightarrow {}^{210}Bi \xrightarrow{5.3d;\beta^-} {}^{210}Po$$

$$^{210}Po \xrightarrow{138.4d;\alpha} {}^{206}Pb$$

$$^{208}Pb + n^0 \rightarrow {}^{209}Pb$$

$$^{209}Pb \xrightarrow{3.3h,\beta^-} {}^{209}Bi$$



Polonium-210

- biological half-life of 30-50 days
- high radiotoxicity in case of incorporation (LD_{50} 2-15 MBq)
- is believed to form a volatile hydrogen species in presence of hydrogen and/or moisture
- for liquid spallation targets risk estimations and dispersion calculations in case of leakage are necessary for licensing

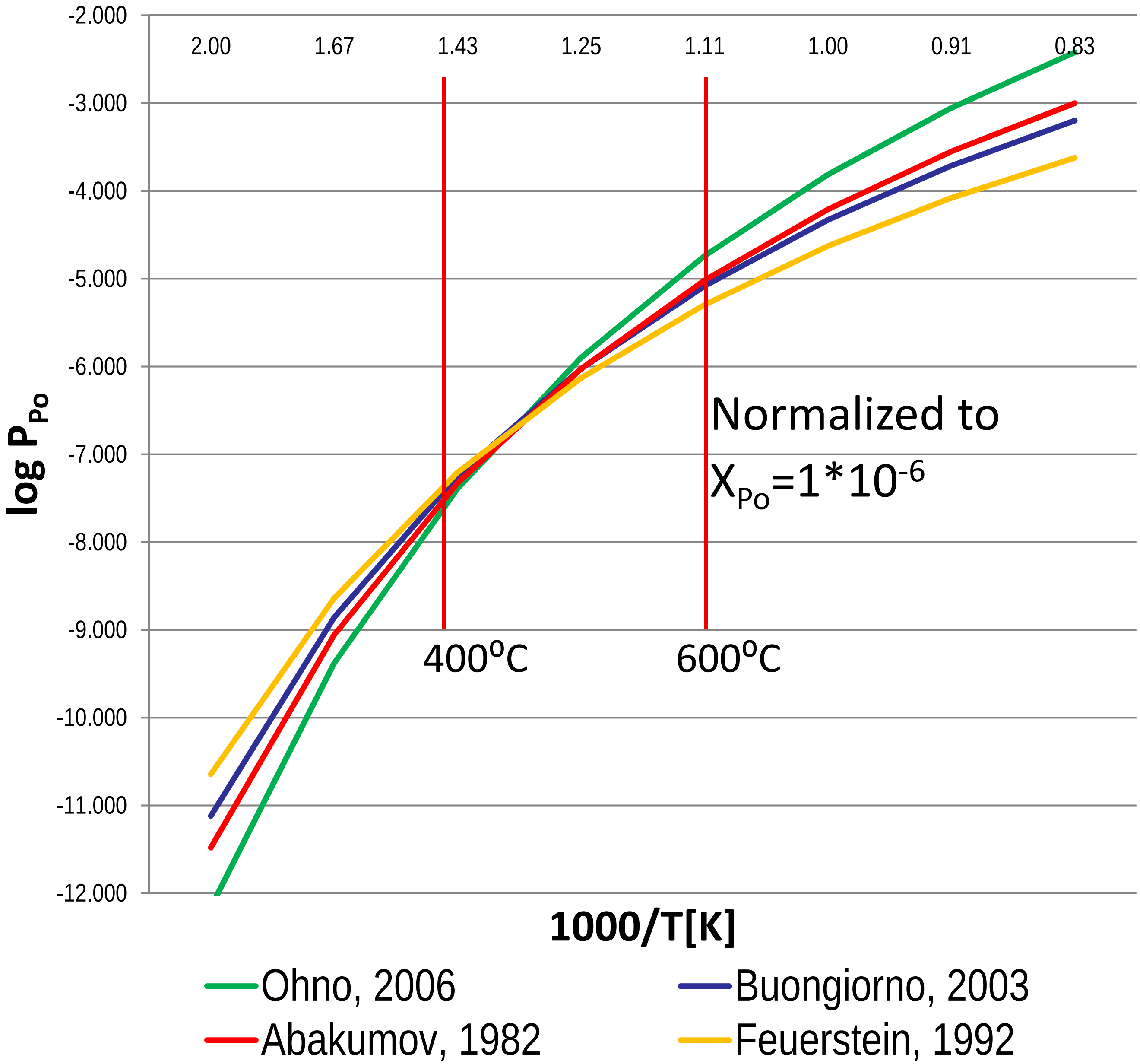
Polonium

- the apparent vapor pressure of polonium is of main interest for thermodynamic calculations
- the volatile species of polonium and conditions for formation
- influence of cross effects like sputtering and aerosol formation
- stability and possible extraction/absorption of volatile species for filter design in nuclear facilities

→ **release experiments**

POLONIUM RELEASE

Selected vapor pressure data for ^{210}Po



Abakumov [1982]: release experiments from lead, assumed $\gamma_{PbPo}=1$, radiometric and direct vapor pressure measurement, T:368-745°C

Feuerstein [1992]: release experiments from $Li_{0.17}Pb_{0.83}$, $X_{Po}=1.5*10^{-13}$, α-counting, T:350-700°C

Buongiorno [2003]: release experiments from LBE, $X_{Po}=2.5*10^{-8}$, Liquid Scintillation Counting (LSC), T:400-550°C

Ohno [2006]: release experiments from LBE, $X_{Po}=2.2*10^{-10}$, LSC, T:450-750°C

A dynamic method for vapor pressure measurements

- a solid or liquid sample gets heated in a constant stream of gas
- the gas phase gets saturated with the gaseous sample species
- quantification of the evaporated material
- calculation of the apparent vapor pressure

⟶ vapor pressure function *f*(T)

- in case of pure substances, direct calculation of the saturation vapor pressure

⟶ apparent vapor pressure functions in our case!

POLONIUM RELEASE

$$N_{release} = \frac{\Delta A}{\lambda}$$

$$n_{Po} = \frac{N_{release}}{N_A}$$

$$P_{Po}^{sat} = \frac{n_{Po} \cdot R \cdot T}{V_{sat}}$$

$$K_{Po}^{Henry} = \frac{P_{Po}^{sat}}{X_{Po}}$$

ΔA is measured at following energies: 286, 338, 522, 807 and 1032keV

detector efficiency , specific γ-Intensity and a sample-geometry correction factor are included in ΔA

EXPERIMENTAL SETUP

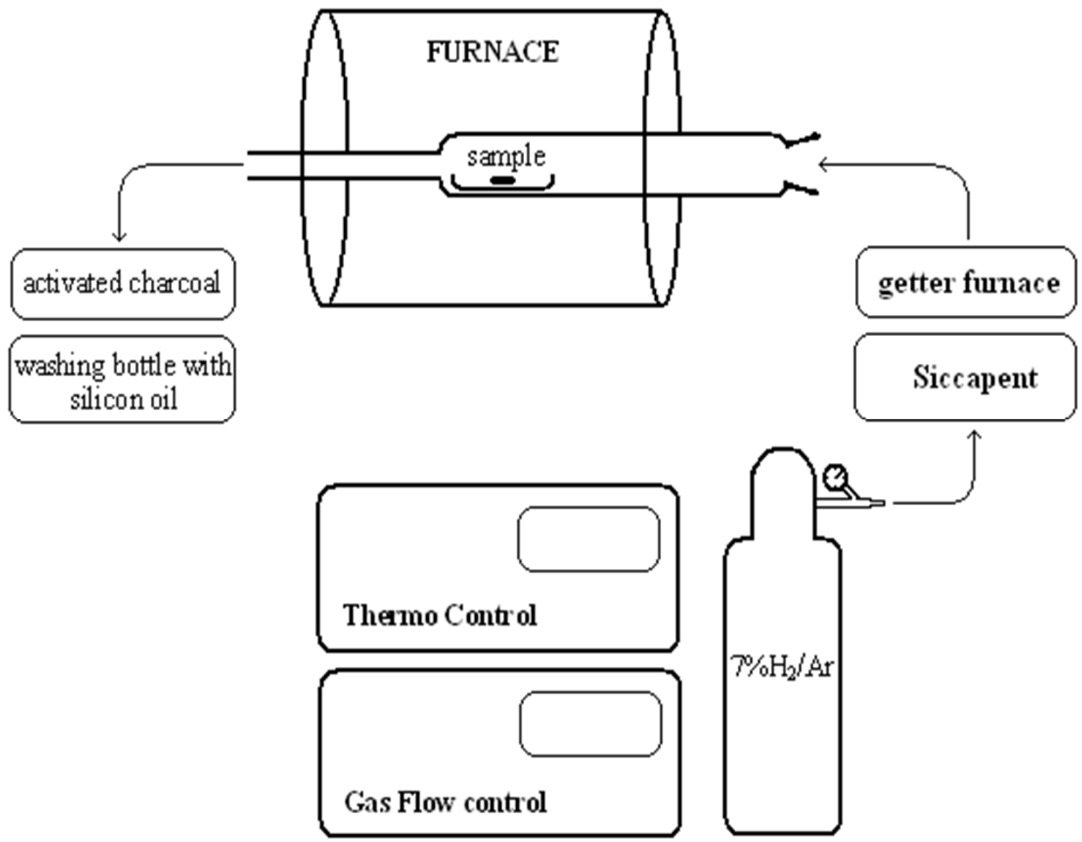
•Closed system to prevent moisture and oxygen entering the rection tube

•Argon with 7% hydrogen in order to reduce the lead surface (PbO)

•Applied atmosphere gets directed through a drying cartridge and a getter furnace in order to remove moisture and oxygen

•quartz reaction tube

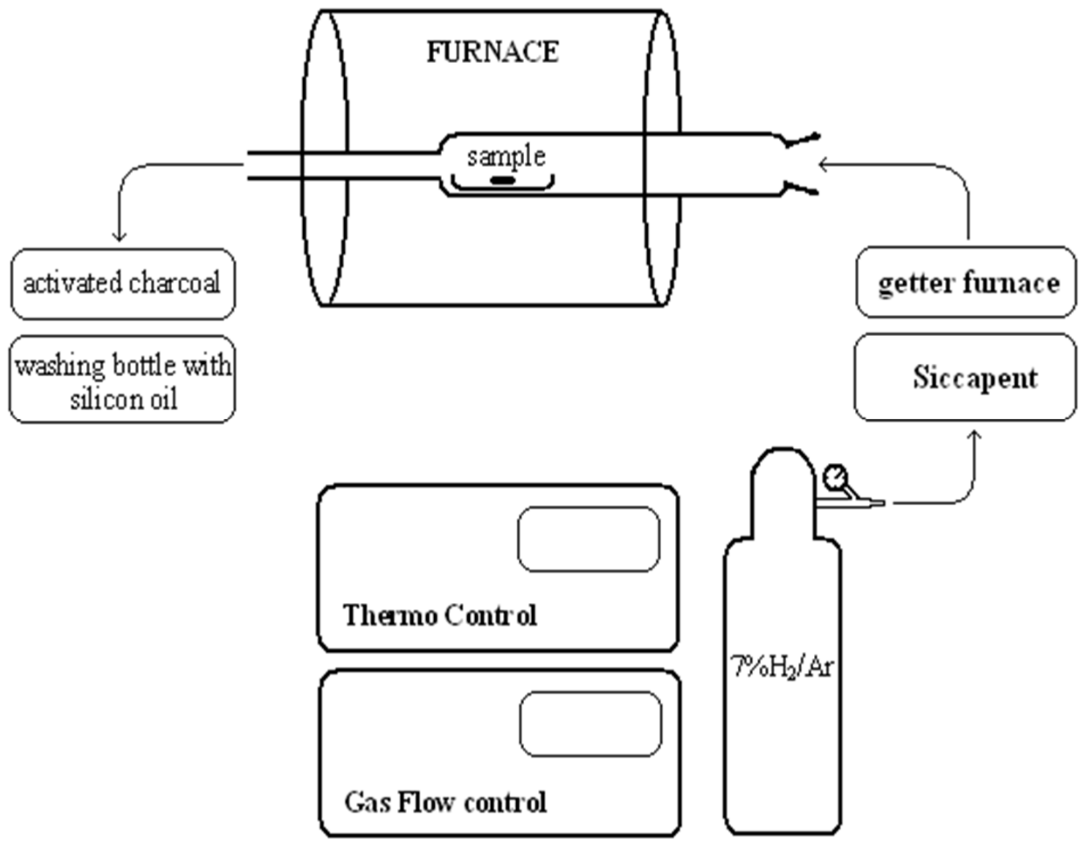
•sliding arrangement to apply temperature



experimental setup scheme

EXPERIMENTAL SETUP

- irradiated with α-particles
- lead-samples of ~0.7g mass
- sample on a quartz filter within a quartz boat
- 53 ml/min of 7%H_2/Ar atmosphere applied
- temperatures from 600^{o}C up to 1100 ^{o}C reaction-time 60min



experimental setup scheme

EXPERIMENTAL TECHNIQUE

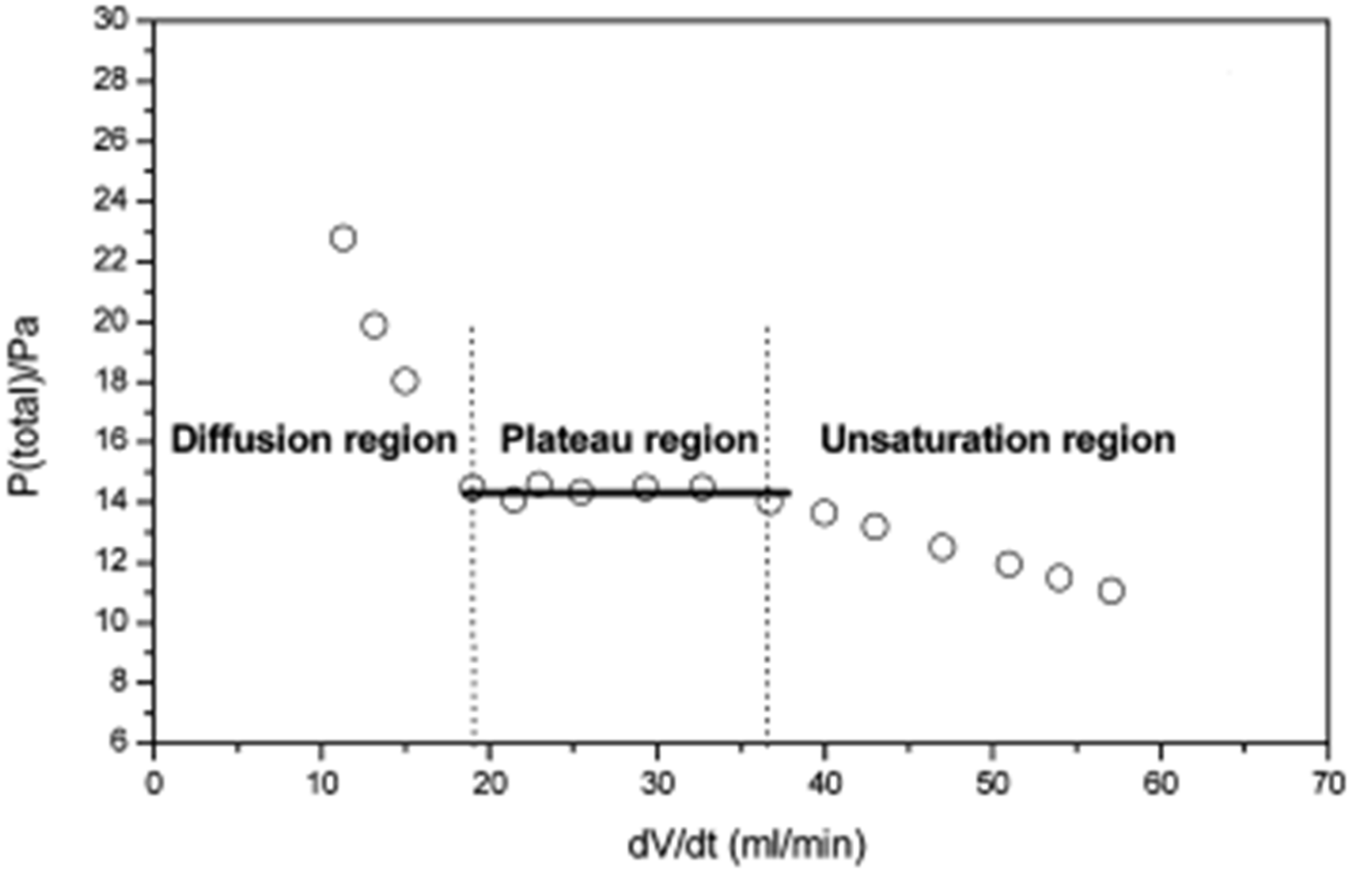
PROCEDURE

- sample gets weighed and measured by γ-spectroscopy (detection of ^{206}Po!)
- placement in a quartz-boat (with quartz filter) within the quartz heating gadget
- closing of the gadget, flush with atmosphere, heating of the furnace
- start of the experiment by sliding the furnace into the right postion
- stop after 60min, cooling down, γ-detection

EXPERIMENTAL TECHNIQUE

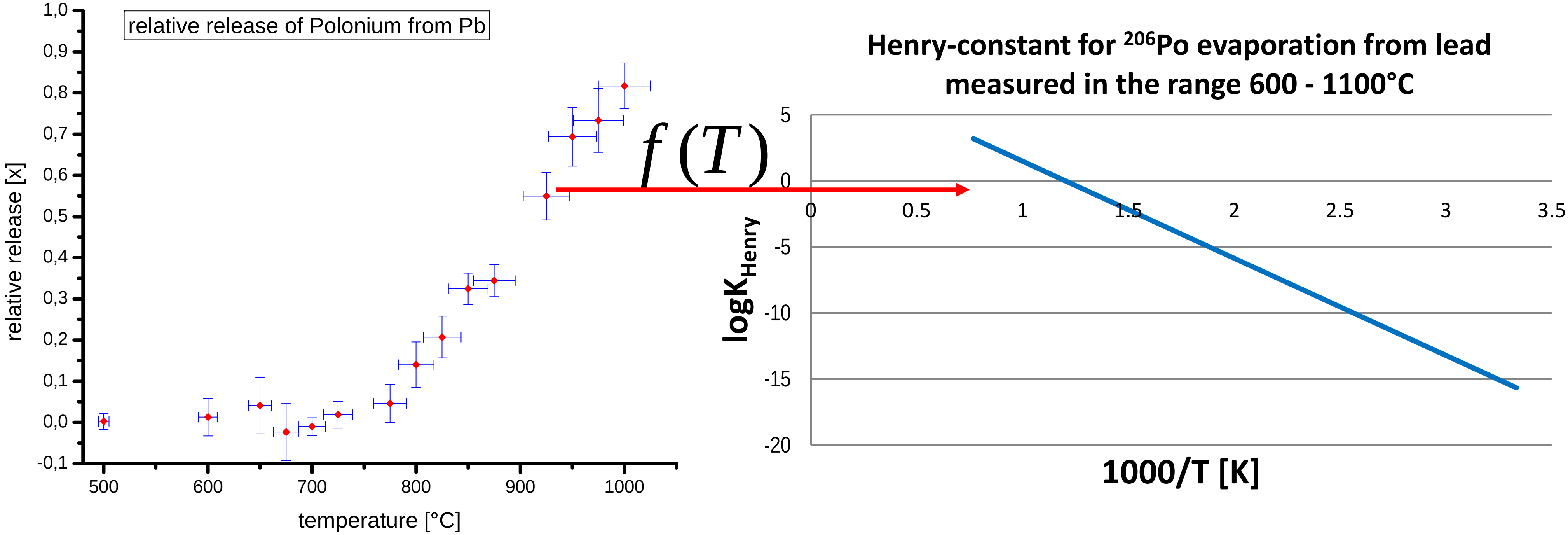
saturation of the gas phase with the sample species is crucial!

– Selection of the right atmosphere flux, sample and gadget geometry has to be considered



variation of apparent vapor pressure with volumetric flow rate
(Viswanathan et al., J. Phys. Chem. B, Vol. 113, No. 24, 2009)

RESULTS & DISCUSSION



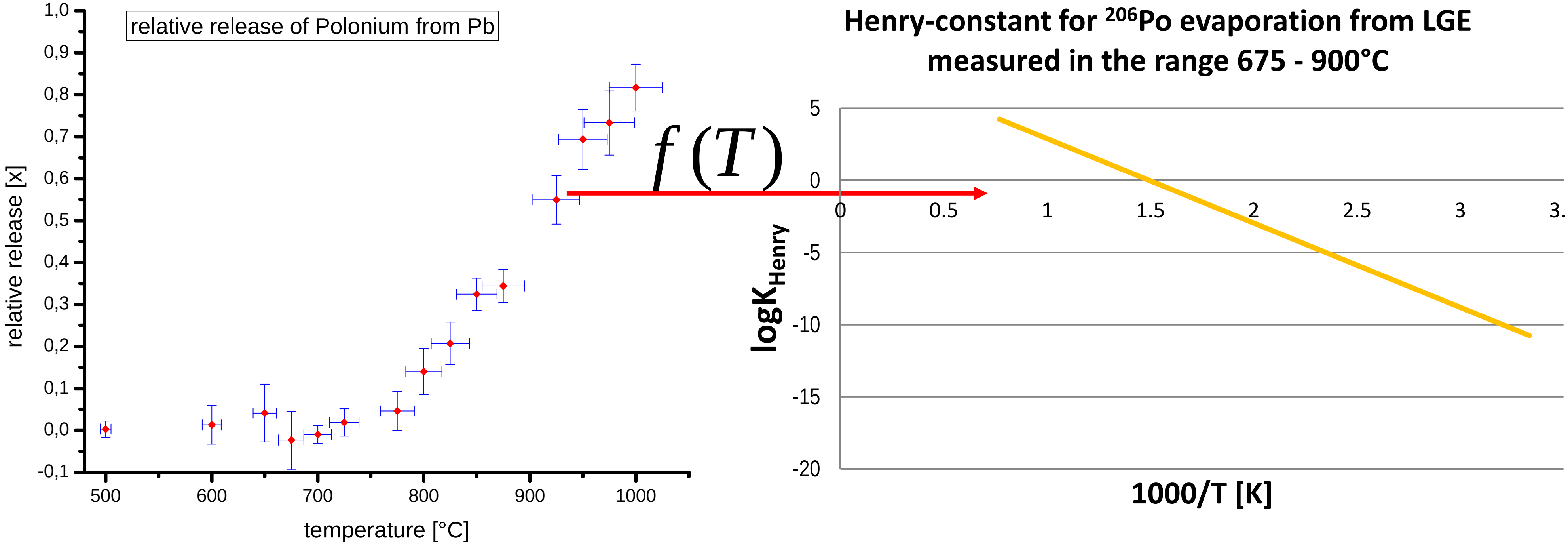
with the given flux and reaction time

⟶ calculation of the apparent vapor pressure

$$K_{Po}^{Henry} = \frac{P_{Po}^{sat}}{X_{Po}}$$

$$\log K_{Po-Pb}^{Henry} = 8.85 \pm 1.09 - \frac{7352.6 \pm 1012}{T}$$

RESULTS & DISCUSSION

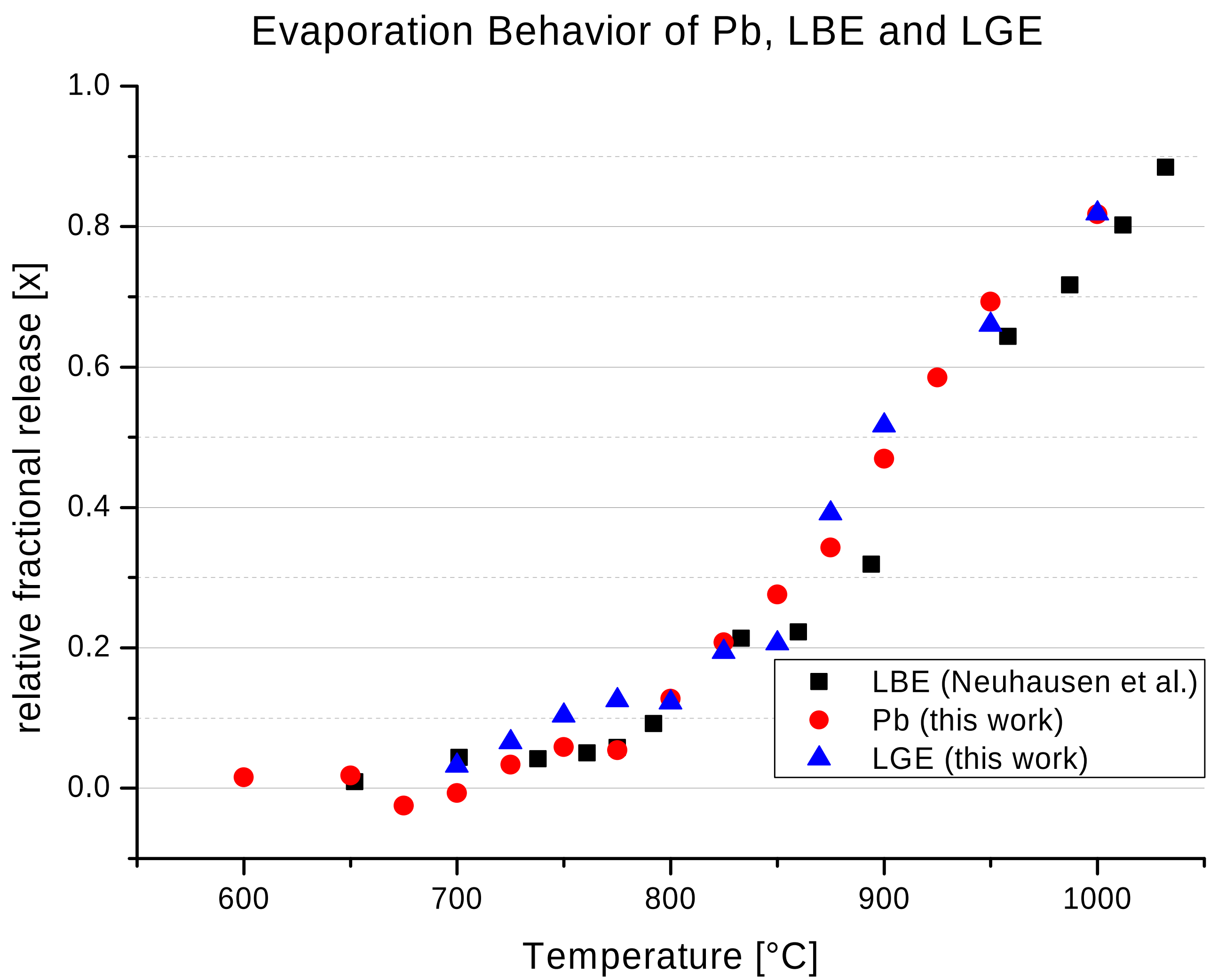


systematically higher apparent vapor pressure for ^{206}Po over a LGE than compared to LBE or lead

$$K_{Po}^{Henry} = \frac{P_{Po}^{sat}}{X_{Po}}$$

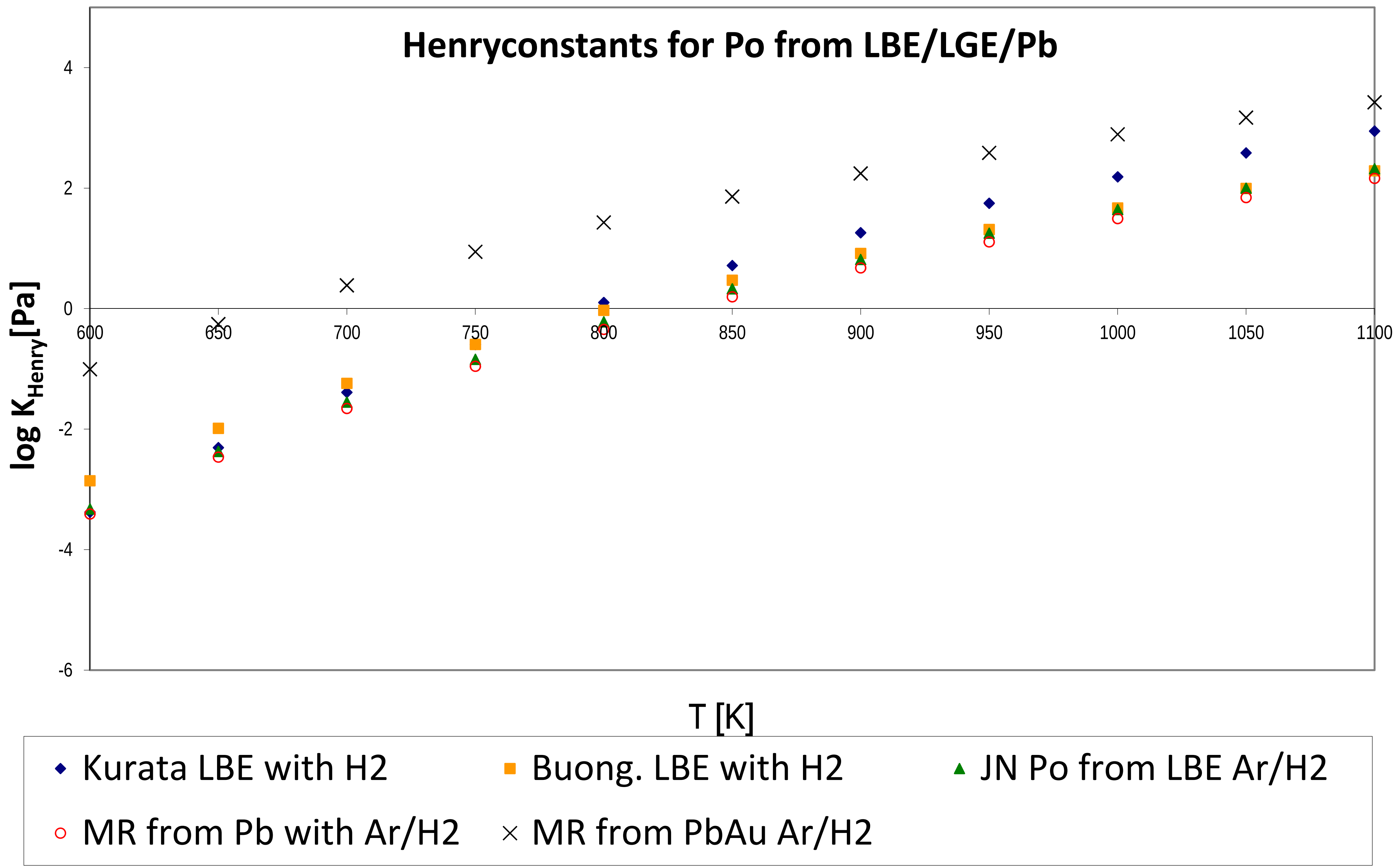
$$\log K_{Po-LGE}^{Henry} = 8.74 \pm 1.05 - \frac{5848.2 \pm 996}{T}$$

RESULTS & DISCUSSION



evaporation of Po from lead is similar to that from LBE and LGE

RESULTS & DISCUSSION



SUMMARY & OUTLOOK

use of ^{206}Po instead of ^{210}Po

the calculated saturation vapor pressures are "apparent" vapor pressures – the composition of the gas phase is unknown (PbPo, H_2Po,…)

large errors due to experimental uncertainities

fraction of polonium in the sample is not constant, continuous loss of signal

saturation of the vapor is crucial!

SUMMARY & OUTLOOK

error reduction in our experimental technique

proof of gas phase saturation

- *f(T,j)* for given flux j at different temperatures
- influence of sample and gadget geometry

performing experiments under an oxygen-controlled atmosphere (glove box)

investigation of aerosol formation during transportation method experiments

verification of the volatile polonium species (?)

thermochromatographic studies of polonium

THANKS TO

R. Eichler , D. Schumann, J. Neuhausen

S. Heinitz and S. Lüthi

THANK YOU FOR YOUR ATTENTION