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Polonium Behavior in eutectic Lead Bismuth Alloy

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Outline

1. Importance of Po for spallation targets
2. A short look on MEGAPIE licensing
3. New results on polonium behavior in LBE
4. Things remaining to be done

Spallation Products

Nuclides formed by nuclear reactions

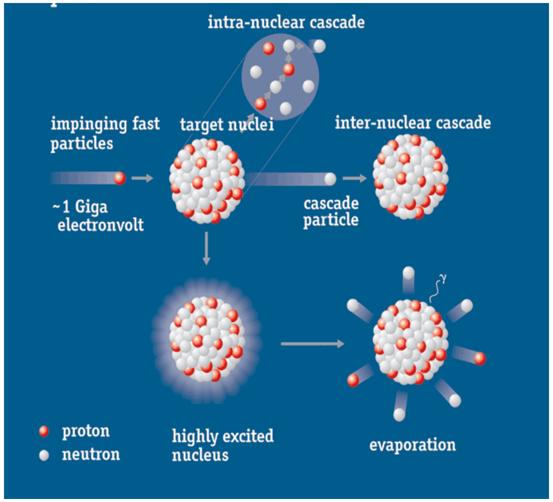
Spallation, Fission, Neutron capture

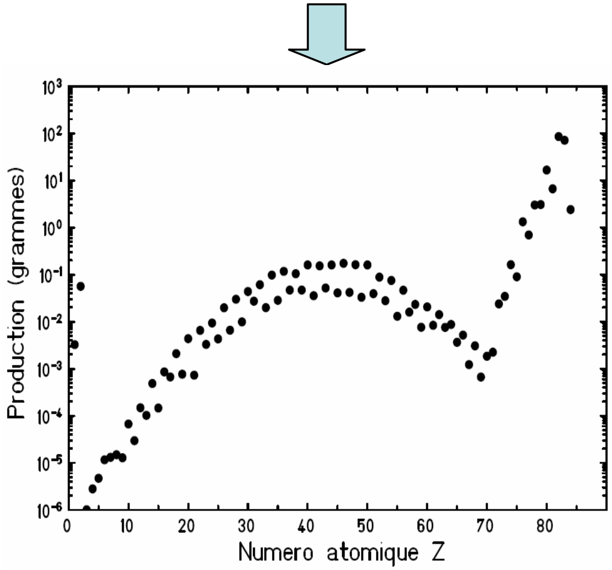
All elements of periodic table with $Z \leq Z_{target} + 1$

For LBE: Significant amounts of highly radiotoxic Po-isotopes are formed (MEGAPIE: gram amounts, future ADS systems: some orders of magnitudes more)

Evaluation of consequences for the behavior of the system

- Normal operation conditions
- Accident scenarios





MEGAPIE Licensing: Accident Scenarios

Compared to a solid target: faster release of volatiles

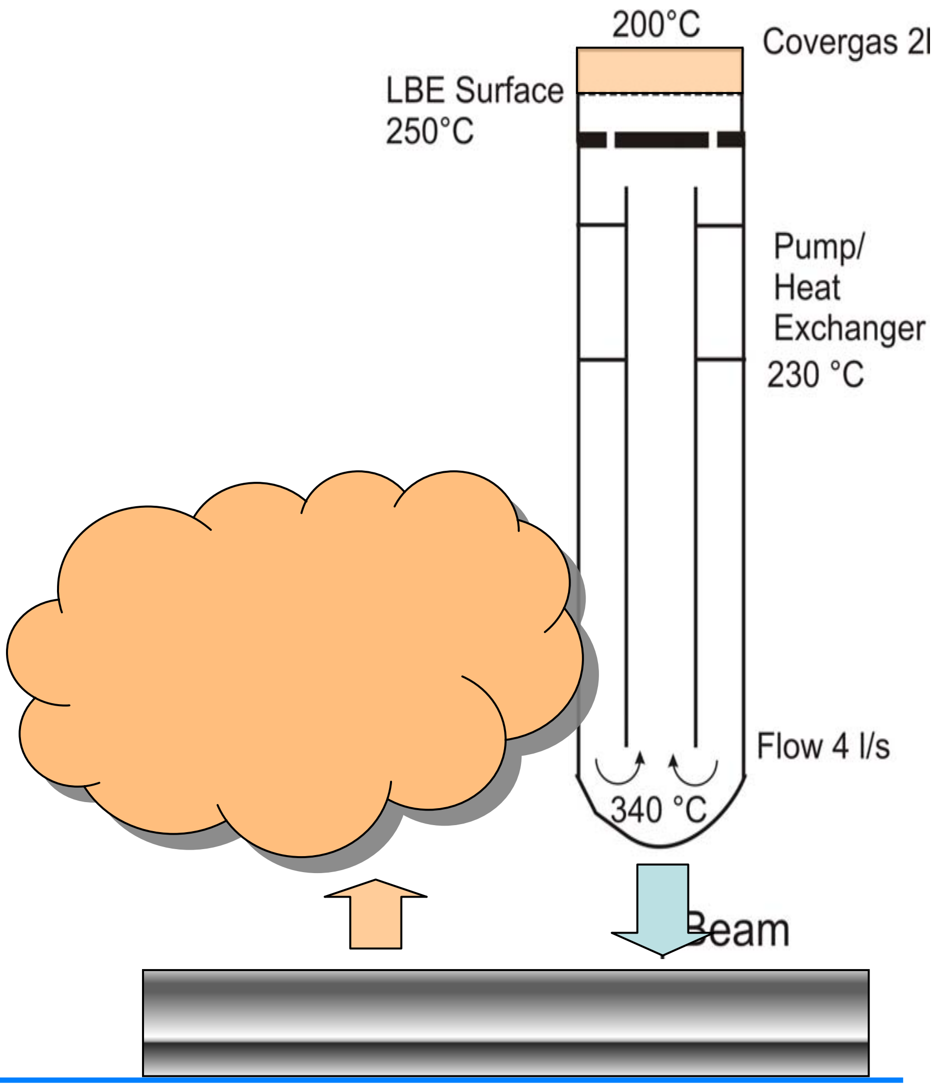
3 Source terms in case of break:

- Expansion volume
- Spilled LBE
- LBE adhering to the target walls

Dominant for accident case:

LBE adhering to the target walls,

Effected by slow cooling of target



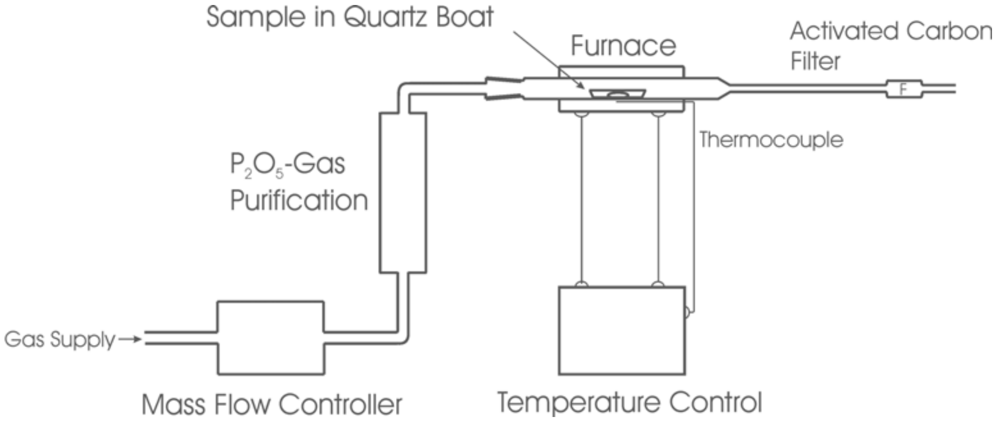
Expansion Volume: Determination of vapour pressure

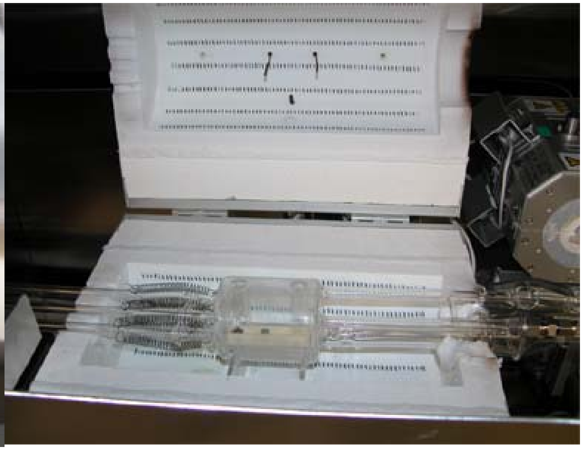
Po data:

only unreliable vapor pressure data due to experimental problems arising from α-decay

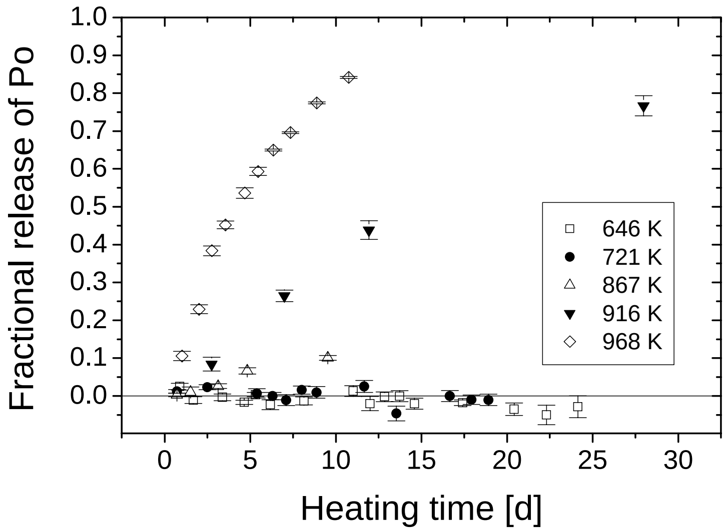
Licensing authorities demanded a reevaluation of data

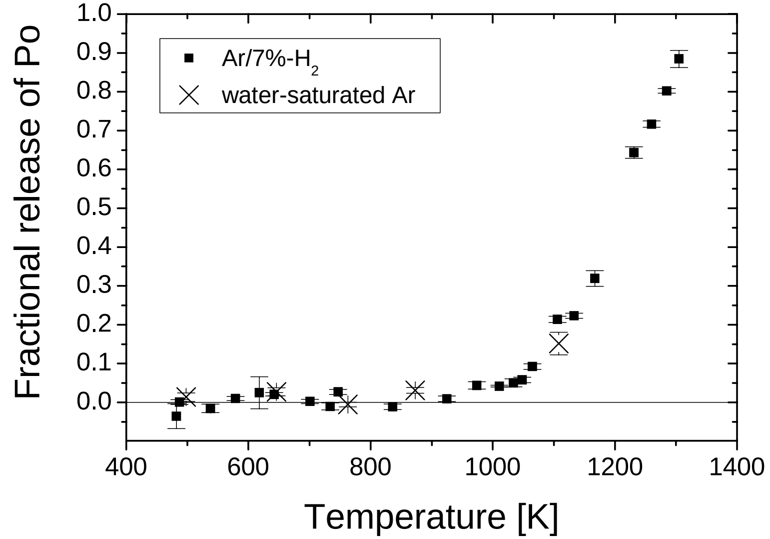
Transpiration method

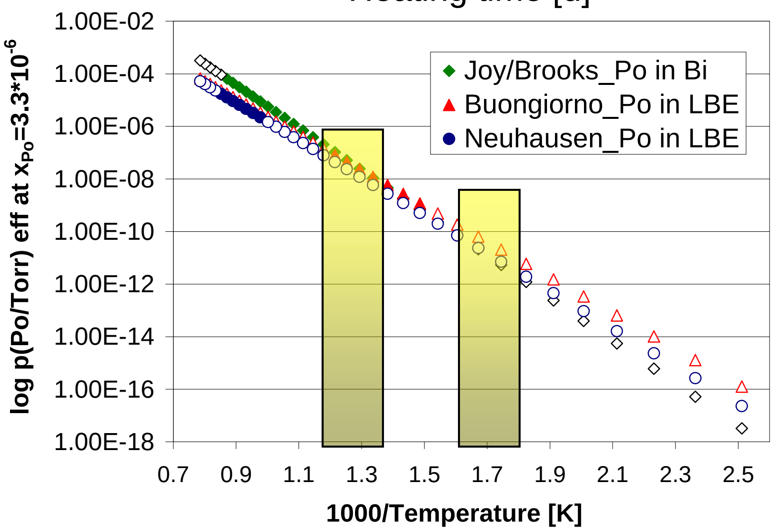




Time and temperature dependent Po-release





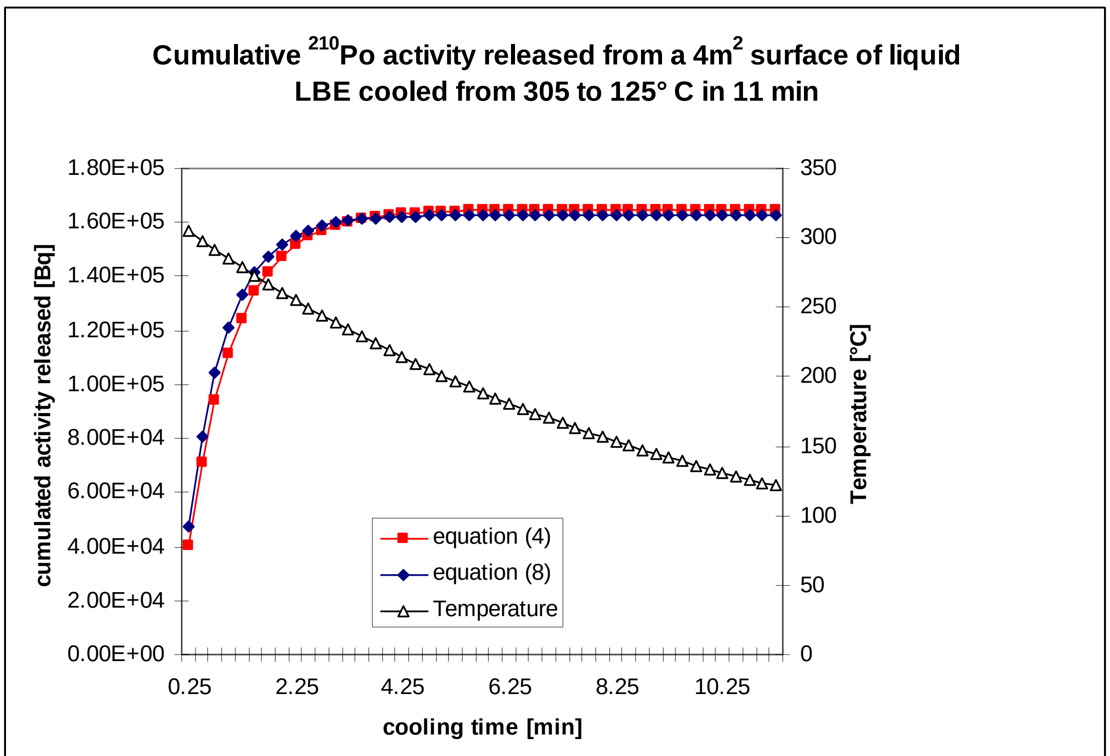


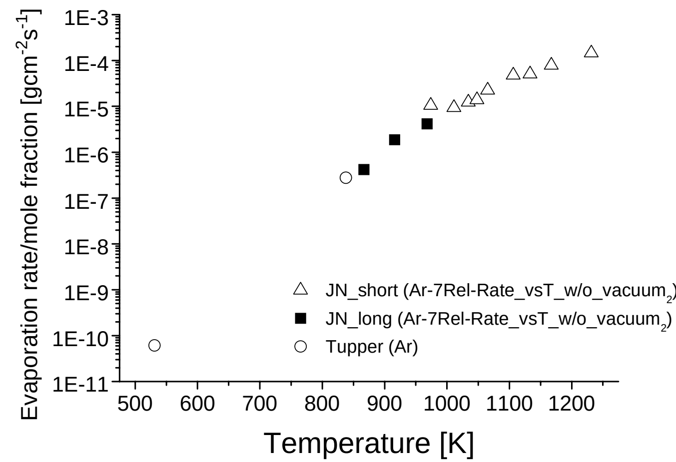
Reasonable aggreement with earlier studies

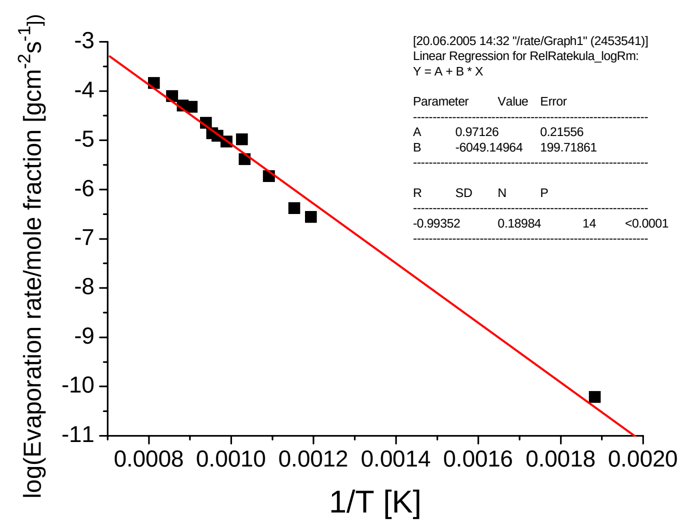
Only a few Bq Po are expected to be released from expansion tank based on equilibrium vapor pressure

Spilled and adhering LBE: Evaporation rate

BAG demanded a reevaluation of evaporation rate stated in PSAR





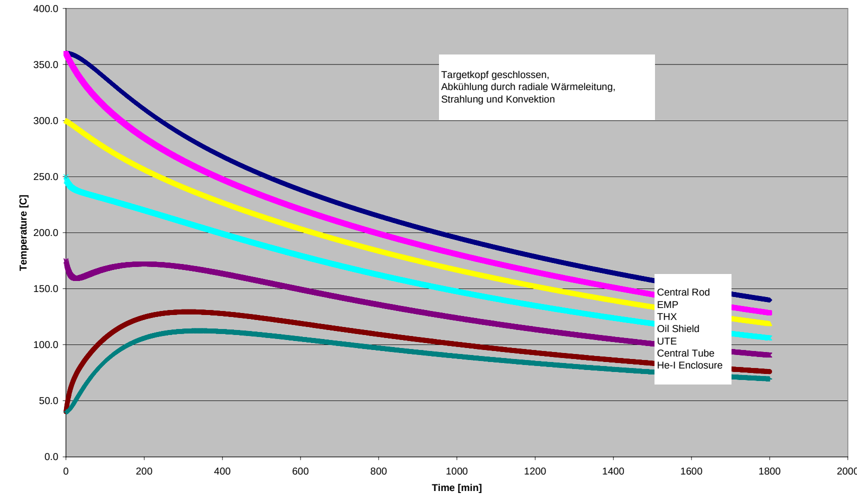


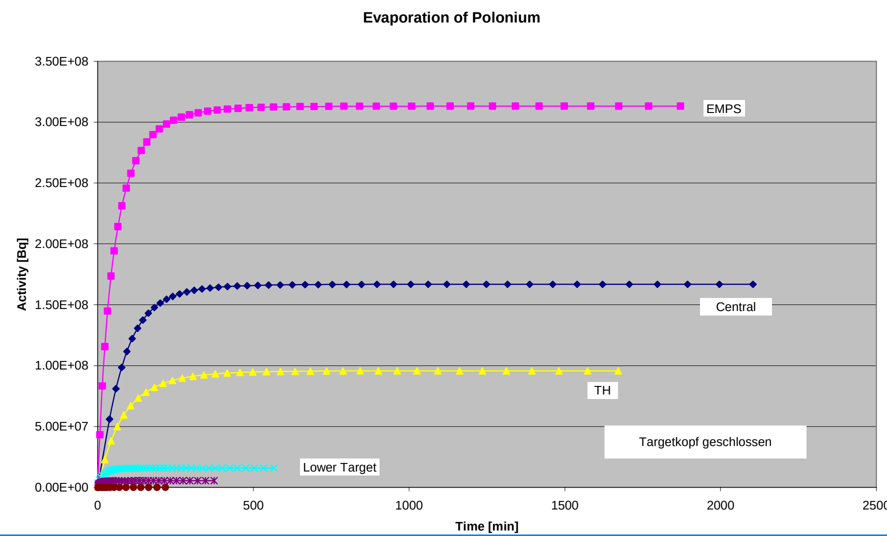
Cooling of inner surfaces and radionuclide release

- Cooling curves for many parts of the target inner surface have been calculated for different accident scenarios

- For each of these surfaces the evaporation of Po and Hg were calculated using temperature dependent evaporation rates

- Acceptable Doses are obtained by ESS41 calculations in case of <u>filtering</u> of the gas can be assured. Additional filters including a mobile filter unit was installed





Recent investigations

Investigation of older SINQ-irradiated LBE samples containing ^{210}Po revealed that polonium is enriched at the surface of the solid samples.

Consequences:

a) For release measurements, the samples have to be homogenized.

b) Suitable analytics have to be developed to determine the α-activity.

c) The process of segregation may be of importance for the handling of LBE after shutdown of the system and for the interim storage of decommissioned target and core material

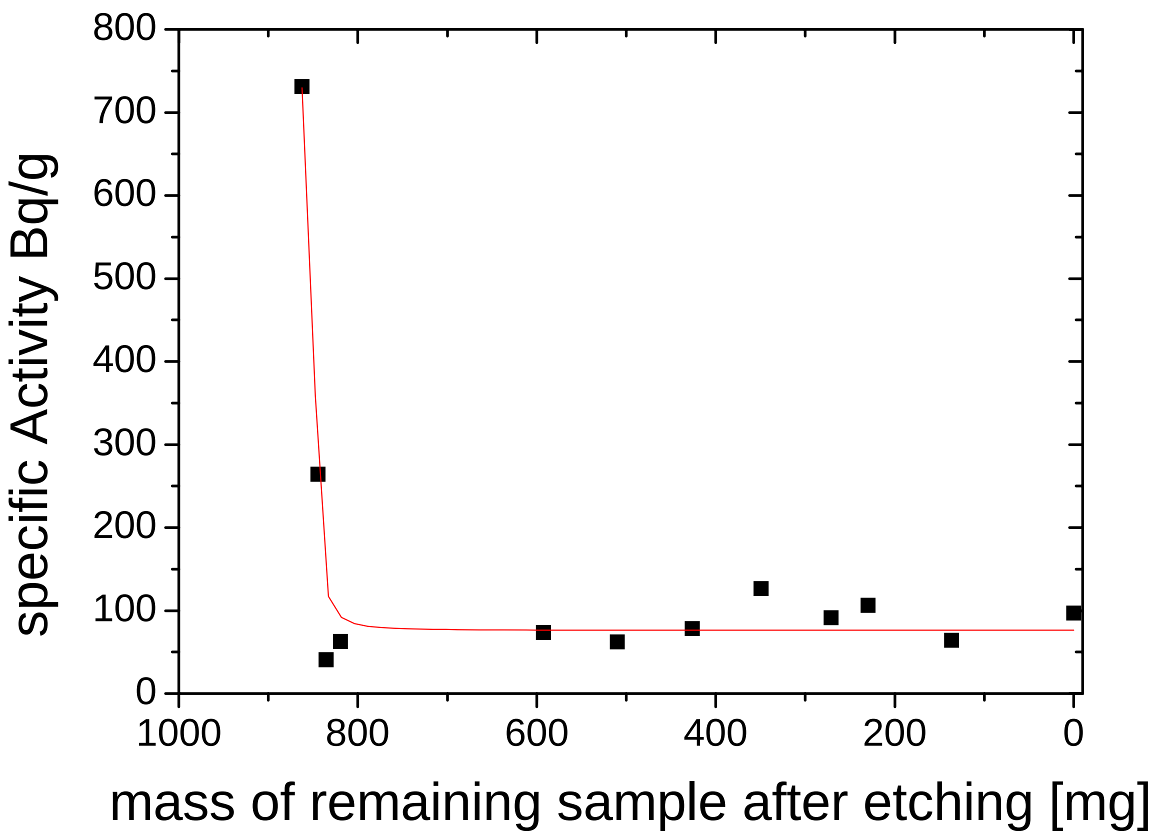
Segregation of Polonium in LBE

A study of the phenomenon of polonium segregation was started.

LBE-samples containing ^{210}Po were etched and the etching solutions analyzed by liquid scintillation counting. As shown in the figure, polonium is highly enriched in the surface.

Surface layer appr. 20 µm

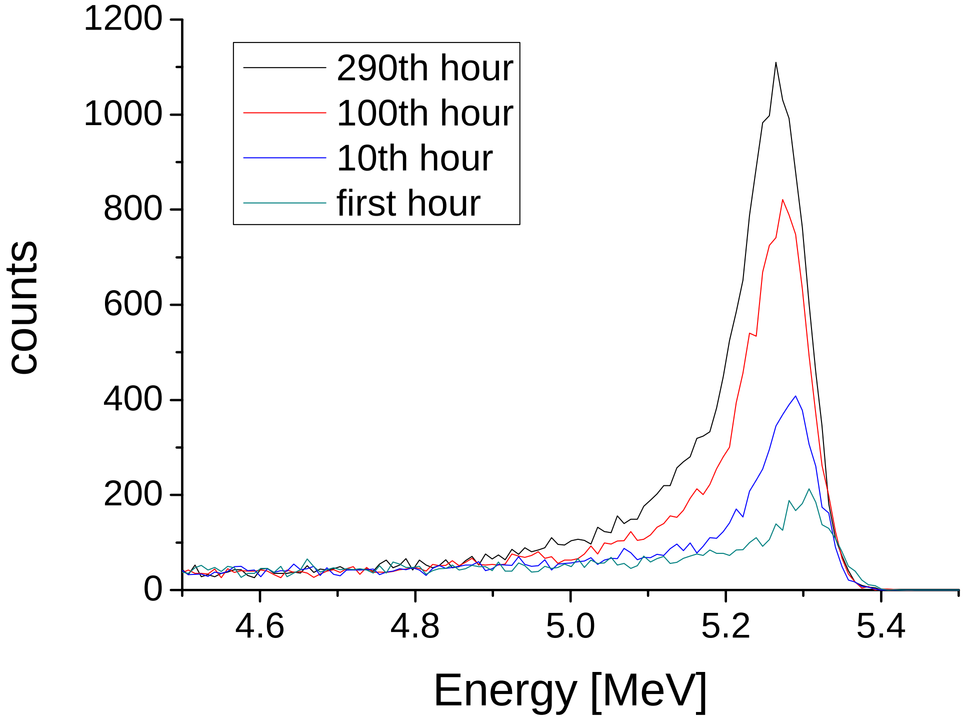
Can be homogenized by melting



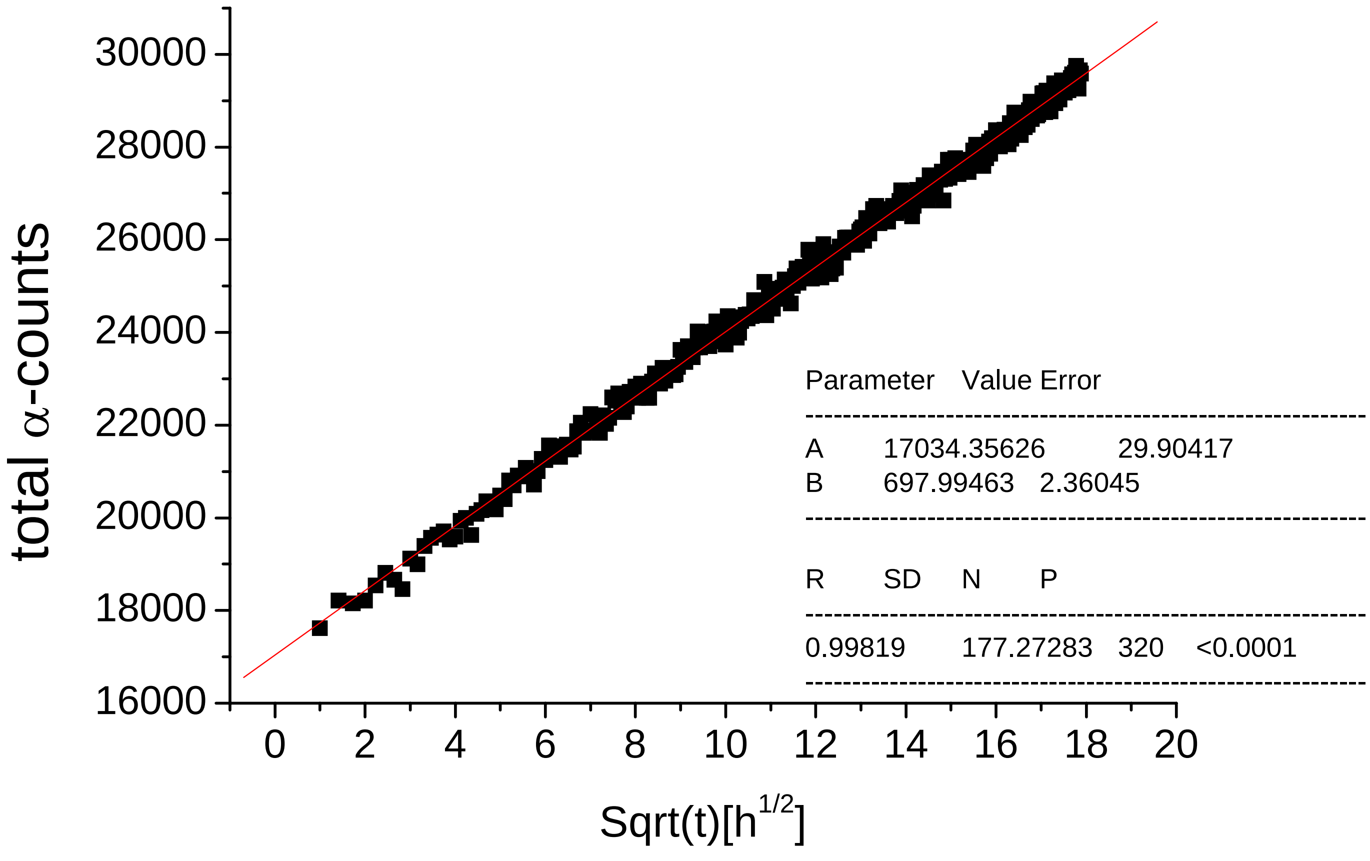
Segregation of Polonium in LBE

α-spectra successively taken from a freshly irradiated and homogenized sample shows that substantial surface enrichment occurs within hours or days in a solid sample.

A detailed study of the segregation process and its kinetics may be of importance with respect to target and core handling and decommissioning.



Time dependence



Typical for a diffusion controlled Process

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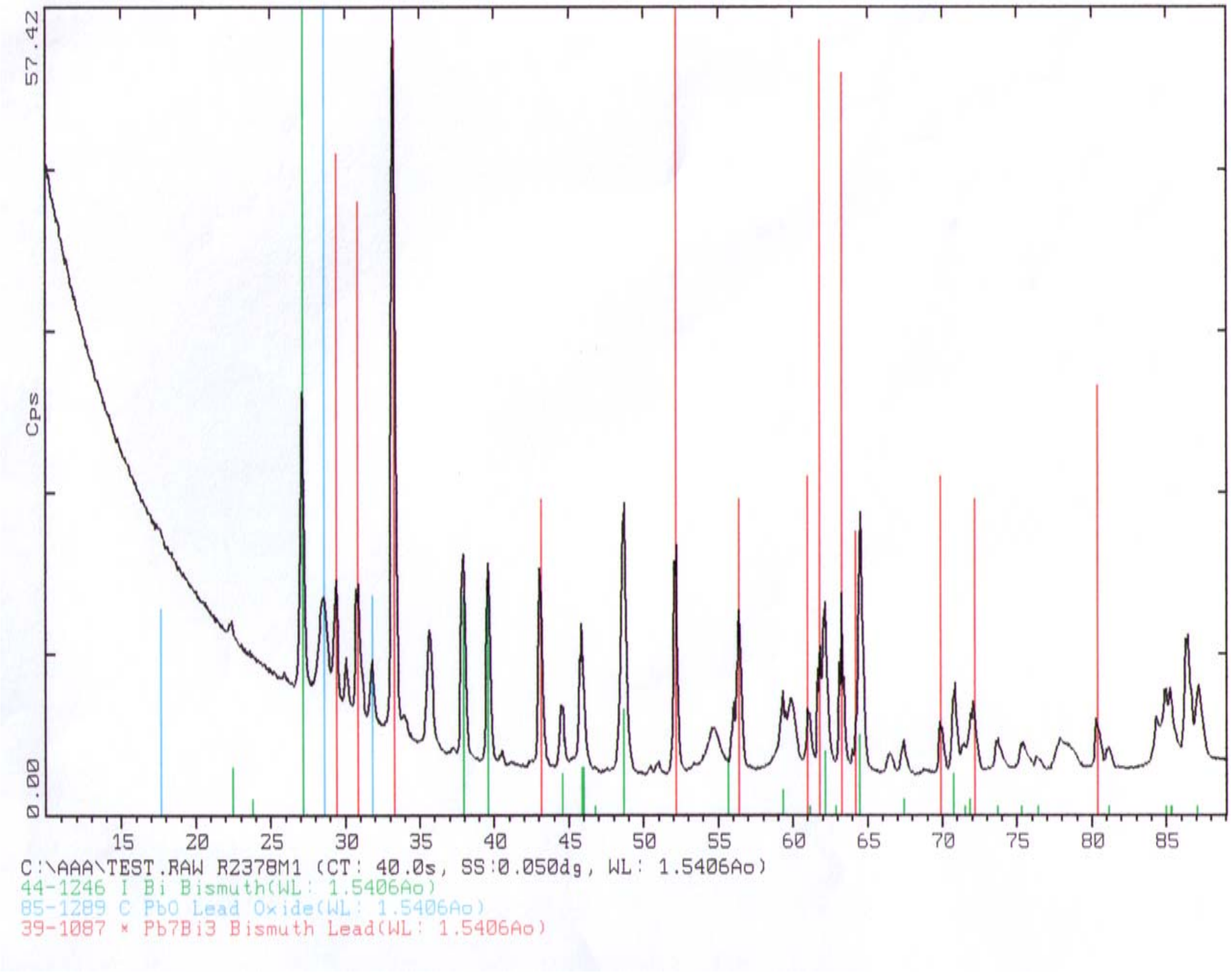
Driving force: Po bound in oxide phase?



Release studies in vacuum for windowless target design

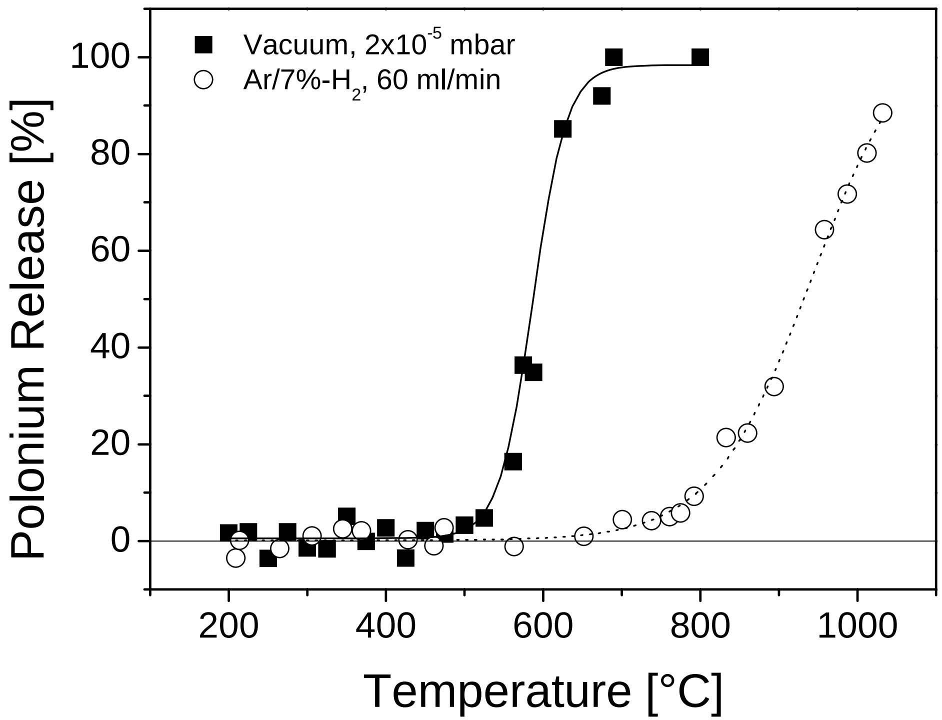
A procedure was devised to homogenize the Po concentration in LBE samples by melting. Analysis of α-activity is the done by dissolving part of the sample in concentrated HNO_3, evaporation to dryness, solution in diluted HNO_3 and subsequent liquid scintillation counting.

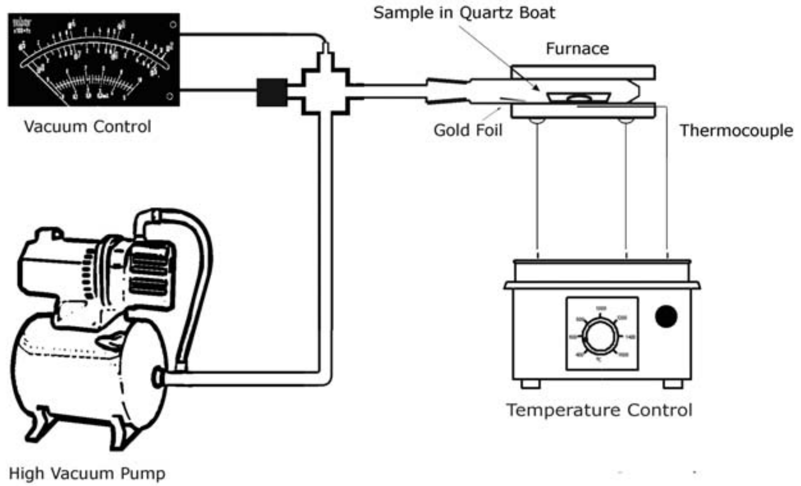
Advantage of this method: It makes us independent from ^{206}Po supply of CERN

Disadvantage: Analytics takes more time and effort than γ-spectrometry that can be used with ^{206}Po

First Results of Vacuum Release

As expected, much faster release in vacuum (2×10^{-5} mbar) is observed in 1h isochronous measurements, compared to Ar/7%-H_2 at ambient pressure





Detailed studies at various pressures including time dependent studies at different temperatures are planned to develop a mathematical description of Po evaporation behavior

Further studies required: Other Mechanisms of Po transport

At the presence of hydrogen and/or water and additional agitation, a process has been observed that leads to increased vapor phase concentrations of Po

The process seems to be more or less temperature independent

Is it really simply formation of H_2Po and its evaporation?

The mechanism of this process and its relevance for a spallation system needs to be clarified

Aerosol transport may play a role (has been observed in many loop systems, MEGAPIE experience will be valuable, not limited to LBE nor Po)

Development of Containment Strategies

❑MEGAPIE: evaporation based on the complete inventory is compatible with safe operation and accident scenarios (probably for long-term irradiation (ADS), reduction of the inventory is desirable)

❑Bonding of volatiles before they leave the condensed phase

Metal absorbers, salt melts, CeBi,

❑Deliberate evaporation and bonding to catcher/filter systems in the gas phase

Megapie-experience: Ag/Pd-absorbers for Hg and possibly halogens

Efficiency has still to be evaluated in PIE