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Polonium formation in Pb–55.5Bi under proton irradiation

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Abstract

Lead–bismuth eutectic (LBE) has been proposed both as spallation target and as coolant in a future accelerator driven system (ADS). The safety analyses require knowledge concerning the elements formed during proton irradiation, and their volatility. Discs of lead–bismuth eutectic (LBE) were irradiated with protons having 71 and 590 MeV energy resp. to analyse Po-isotopes formation. The generation of the isotopes Po 206 and 208 was clearly indicated. The ratio $^{206}Po/^{208}Po$ is dependent on the proton energy and as well on the beam intensity. In parallel to the analytical examinations calculations were carried out using the FLUKA Monte Carlo code to compare the analytical results with the calculations. The evaporation behaviour of irradiated LBE discs was examined as well whereby volatile Hg was found, but no Po evaporation was detected under the chosen conditions.

1. Introduction

Lead and lead–bismuth eutectic (LBE) have been proposed both as spallation target and as coolant in a future accelerator driven system (ADS) [1,2]. At PSI, the development and construction of a 1 MW Pilot Experiment (referred to as 'Megapie') will use liquid lead bismuth (LBE) as target material [3]. The facility will be installed in the secondary induced neutron source (SINQ) area, operating with 590 MeV protons. Up to now the ferritic/martensitic steel T91 is foreseen to be used as window material. For testing the steel under Megapie relevant conditions (elevated temperature, flowing LBE, static load, proton beam irradiation) the liquid solid reaction (LiSoR) experiment was initiated [4,5]. LiSoR loop is installed in the IP1 bunker and is irradiated with 71 MeV.

The irradiation of LBE with high energetic protons generates different radionuclides which might play an important role concerning safety aspects. Therefore it is necessary to know which elements are formed during irradiation and if they are volatile under these conditions or if they may stay in the liquid eutectic. Especially the generation of Po isotopes is one of the important operational problems. Twenty five isotopes of polonium are known, with atomic masses ranging from 194 to 218. Po is very dangerous to handle in even milligram or microgram amounts and special equipment and strict control is necessary due to the complete absorption of the energy of the alpha particle into human tissues. Thus the Swiss authority for public health (BAG – Bundesamt für Gesundheit) addressed among other things the following questions:

- Amount and ratio of the Po-isotopes produced in LBE under irradiation with proton energies of 71 and 590 MeV.
- Evaporating behaviour of irradiated LBE at elevated temperature.

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In this report the experiments of irradiation of LBE with 71 and 590 MeV are reported, the results of the α- and γ-measurements are presented and a preliminary evaporating experiment is described. Additionally calculations by using the FLUKA code were carried out for the different irradiations performed and calculated and experimental data are compared.

2. Experimental

Impag AG (Switzerland) supplied the eutectic Pb–55.5Bi (44.8 wt% Pb and 55.2 wt% Bi) alloy which contained as less as only a few ppm of impurities: Ag 11.4, Fe 0.78, Ni 0.42, Sn 13.3, Cd 2.89, Al 0.3, Cu 9.8, Zn 0.2.

2.1. Irradiation

For our experiments, LBE was melted and filled into pre-heated glass tubes with a diameter of 16 mm. The cylinders of LBE were then isolated by carefully dismantling the glass container. Afterwards small discs were cut out of the rods with 2 mm in thickness and 16 mm in diameter. Each disc was labelled, weighed and measured.

The discs were placed into austenitic steel containers in which a bottom of T91 steel 1 mm in thickness was welded. The bottom is the so-called beam window through which the proton beam was entering. T91 is a martensitic 9Cr1Mo steel. The container was closed with a cap fixed tight to the container by using a tension ring. Fig. 1 shows the container with some LBE discs (∅16 mm).

In total two irradiation experiments were performed. In the first irradiation the proton energy was 71 MeV, the same energy used in the LiSoR experiments. Irradiation time was quite short but due to the high beam intensity the activation of LBE discs was sufficient. The second LBE irradiation experiment was performed with a proton energy of 590 MeV which is the energy foreseen for MEGAPIE. The parameters for each irradiation are summarised in Table 1.

Fig. 1. Austenitic steel container with a bottom of T91 steel constructed for irradiation of 16 mm ∅ LBE discs.

Table 1
Parameters chosen for the irradiation experiments

Experiment	No. 1	No. 2
Proton energy	71 MeV	590 MeV
Beam intensity	70 nA	1.5 nA
Beam profile	Vertical 2 mm, horizontal 4 mm	Gaussian distribution, FWHM 4.5 cm
Duration	14 s	8 h
Number of discs	12	15
Identification	No. 20–32	No. 1–15

All irradiated LBE discs were primarily investigated by means of γ-spectrometry. Additionally, more complex α-analyses were carried out on the discs having the highest dose rate. To distinguish between different Po-isotopes (e.g. ^{206}Po, ^{208}Po, ^{210}Po) in the irradiated LBE-discs by high-resolution α-spectroscopy, quantitative radiochemical separation methods had to be applied. The method used here is based on dissolution of the Pb–Bi alloy in concentrated nitric acid, wet etching and conversion of the nitrate into chloride medium by two consecutive evaporation steps. The Pb–Bi-chloride residue is then re-dissolved in concentrated HCl and from this solution Po is deposited spontaneously on silver discs, which are taken directly as a source for α-spectrometric measurement. The chemical recovery or yield is controlled by adding a known quantity of a ^{209}Po-tracer to the solution prior to the deposition step. Scoping experiments revealed that this isotope was not produced by the proton bombardment and that all generated (^{206}Po, ^{208}Po), naturally present (^{210}Po) and added spike isotopes (^{209}Po) remained α-spectrometrically resolved.

The irradiation was performed without using a moderator, i.e. the fast neutrons produced during the spallation process were not decelerated to thermal neutrons. Hence only spallation products generated by fast neutron reactions should be formed. It is very likely that the Po-isotopes ^{206}Po and ^{208}Po were produced in the presence of Bi under this condition, but the generation of ^{210}Po is not expected. During the irradiations neither the detection of the produced neutrons nor the shape of the beam profiles was recorded.

2.2. Evaporation experiment

A device was designed and constructed to perform the evaporating experiment on irradiated LBE discs. In

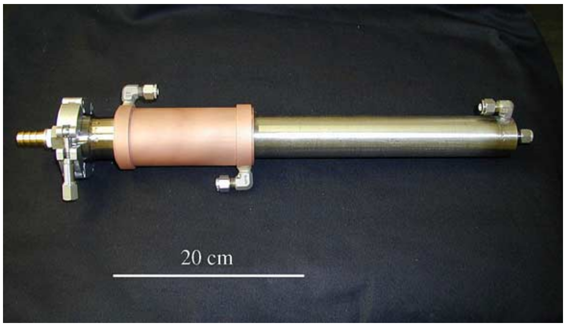

Fig. 2. Apparatus for investigating the evaporating behaviour of irradiated LBE.

principal, the system works as follows: In a hot part of a tube (heated by heating wires) the irradiated LBE is molten, in the cold part of the tube (cooled by a water-cooler coil) the volatile components are absorbed again. The tube is connected with an inlet and outlet system to control the gas flow. A thermocouple is placed in the hot part of the tube close to the ceramic container with the LBE disc. The tube is shown in Fig. 2.

The inner part of the tube was lined with a thin aluminium foil on which the evaporating products should be deposited. Additionally, an active carbon filter was placed in the outlet tube of the gas stream in order to collect mercury first of all. After each test, the aluminium foil and the active carbon filter was replaced and analysed in order to detect volatile deposits with α- and γ-spectroscopy.

For the tests only LBE discs irradiated with 590 MeV protons were selected. Three discs of this campaign (15 discs in total) were chosen for this experiment having nearly the highest over-all activity after irradiation (see also Fig. 3). In Table 2 the parameters for the three evaporation experiments are presented.

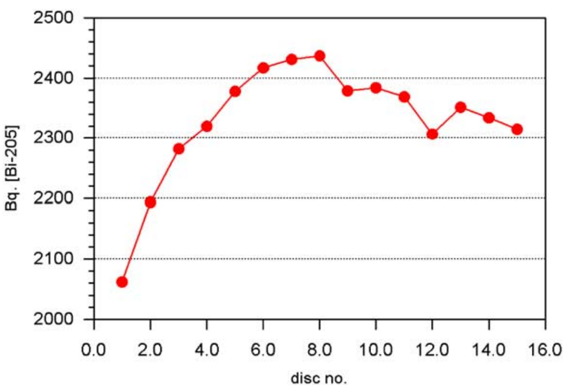

Fig. 3. The measured activity of the isotope Bi-205 versus the irradiated LBE discs.

Table 2
Experimental data for the evaporation experiments

Label of specimen	Temperature (°C)	Duration (h)	Gas flux Ar/2% H_2 (ml/min)
7	140	2	50
6	180	2	50
5	320	2	50

2.3. Calculations

The FLUKA Monte Carlo code [6] was used to simulate the experiment. The parameters from Table 1 concerning the sample geometry and the beam characteristics were used in the calculations. Production rates of the residual nuclei produced during irradiation were generated and written to a file. These production rates were then read and processed using the ORIHET3 code [7] which performs the buildup of the radionuclide inventory during the irradiation time.

3. Results and discussion

3.1. Irradiation with 71 MeV protons

The dose rate and α-signals were measured of each disc and as well of the irradiation container. The disc located next to the beam window (marked with '20') had the highest dose rate (40 mSv/h) compared to the other discs and was therefore selected for the quantitative radiochemical separation process. To show the homogeneity between the produced isotopes of Po the disc was cut into 4 pieces, whereby each specimen was separately analysed. The α- and γ-signals obtained from the 4 samples showed a large variation of the data among each other (see Table 3). This is most likely due to the fact that the beam did not hit the container in the centre during irradiation (i.e. the irradiation of the discs was not uniform). The alignment of the beam is 2 mm in vertical and 4 mm in horizontal direction. The analysis of the polonium isotopes 206 and 208 in the 4 pieces differs therefore as well, but the measured ratio of the isotopes 206–208 measured always more or less the same value. The average values obtained on the 4 quarters for Po-206 and Po-208 and the overall α-activity revealed for the 4 solutions is given in Table 3. Apart from tiny traces of naturally present ^{210}Po, this isotope was not detected because the generation is almost meaningless in absence of thermal neutrons.

The level of the beam intensity is dependent on the position of the target hence the amount of elements generated under irradiation varied as well due to the position. The energy shift is not as significant and therefore the ratio stayed nearly constant although a little

Table 3
Activity of ^{206}Po and ^{208}Po in the four quarters of LBE disc no. 20 and the overall activity of the 4 solutions

Solution	Activity on October 24, 01 (Bq/g)		Ratio	α-Spectrometry
	^{206}Po	^{208}Po	$^{206}Po/^{208}Po$	[Bq/g]
1. Quarter	0.0298	1.11E+1	3.72E+2	12 ± 3
2. Quarter	0.1191	4.17E+1	3.51E+2	44 ± 4
3. Quarter	1.3823	4.76E+2	3.44E+2	470 ± 30
4. Quarter	4.6413	1.54E+3	3.32E+2	1540 ± 70

drift can also be observed: less intensity leads to a higher value of Po-206 isotopes proportionally to Po-208, thus a higher ratio, whereas higher intensity produces a higher amount of Po-208 in the ratio $^{206}Po/^{208}Po$, thus a lower ratio.

Due to the off centre deviation of the beam during irradiation the total amount of the Po-isotopes could not be correctly calculated and here only the ratio $^{206}Po/^{208}Po$ revealed by the FLUKA code will be presented. The value determined is about 300 for the ratio $^{206}Po/^{208}Po$ which is in good agreement with the analytical results (compare Table 3).

3.2. Irradiation with 590 MeV protons

In total 15 Pb–Bi discs were irradiated in a steel container with 590 MeV proton beam for 8 h. Gamma spectra were recorded for each disc whereby the following isotopes were analysed: ^{177}Lu, ^{183}Re, ^{183}Ta, ^{185}Os, ^{188}Ir, ^{188}Pt, ^{189}Ir, ^{191}Pt, ^{195}Hg, ^{195}Pt, ^{196}Au, ^{198}Au, ^{199}Au, ^{201}Tl, ^{202}Tl, ^{203}Hg, ^{203}Pb, ^{205}Bi, ^{206}Bi, ^{206}Po, ^{207}Bi. The amount of the isotopes detected in each disc is different but the ratio of the intensity among each other remained constant. The measured activity for the isotope Bi-205 is given in Fig. 3 vs. the disc number. Disc number 8 yielded the highest over-all activity and was therefore selected to perform the quantitative radiochemical separation process for subsequent α-analysis. The result achieved is the following: The polonium isotopes 206 and 208 were clearly detected, the isotope 210 was not formed under these conditions. The values were revealed by analysing the disc of Po-isotopes: $^{206}Po = 6.64E+2$ Bq/g, $^{208}Po = 4.23$ Bq/g and hence the ratio for $^{206}Po/^{208}Po = 1.57E+2$. The ratio differs from the ratio obtained for the irradiation with 71 MeV protons. The capture cross-section of Bi is dependent on the energy of the neutrons, i.e. with different energies the ratio of the generated isotopes varies as well.

The following results were obtained from the FLUKA calculations: $^{206}Po = 2.66E+2$ Bq/g, $^{208}Po = 1.23$ Bq/g and hence the ratio for $^{206}Po/^{208}Po = 2.17E+2$. Thus, while the calculated activities are lower by almost a factor of 3, the ratio deviates about 30%. The activities of each disc were obtained as well and the allocation is the same as measured (see Fig. 3). Anyway the calculated activities are as well lower by almost a factor of 3. The analytical results obtained by α- and γ-spectroscopy are therefore in good agreement and the reason for the discrepancy in activity has to be searched in the FLUKA code which is currently under investigation.

3.3. Evaporation experiments

The evaporation experiment was performed with Pb–Bi discs irradiated with 590 MeV each having a comparable and a relatively high over-all activity. Three temperatures were chosen for investigating the evaporating behaviour of irradiated Pb–Bi:

- the lowest value, 140 °C, is just above the melting point of LBE,
- about 200 °C is the expected temperature of Pb–Bi in the LiSoR loop system,
- 320 °C is the highest temperature expected in the test section of LiSoR.

After 2 h of heating at the according temperature the Al-foil and the carbon filter was dismantled for α- and γ-spectroscopy in order to detect volatilised products. The results are summarised in Table 4.

With increasing temperature the amount of evaporated Hg-203 increases. It was shown that activated carbon is a material capable to absorb Hg. Metallic Hg has a quite high vapor pressure already at room temperature, at 100 °C it is about 0.3684 mbar. If Hg forms an intermetallic compound in LBE it is obvious that the vapor pressure of it is high or the compound is not stable. Further investigations are needed to analyse the evaporated Hg-species (metallic or in a compound).

The evaporation of Po was not observed neither in the Al-foil nor in the carbon filter. As reported in [6] evaporation of polonium from Pb–17Li is 1000 times smaller than expected in vacuum for an ideal solution at 523 K. It is known that Po evaporates out of a Pb–17Li melt in the form of an intermetallic compound [8]. The low vapor pressure of this polonide leads to a small evaporation rate of PbPo instead of polonium metal. For an exact estimation it is necessary to know the ratio between lead polonide and atomic polonium, which is difficult to predict. Anyway, further evaporation experiments should be performed with irradiated LBE by variation of the cover gas (Ar, Ar/H_2, ...), time,

Table 4
Results of the evaporation experiment of irradiated LBE

7	140 °C	Al-foil	Hg-203	<0.3 Bq/sample
		Carbon filter	Hg-203	<0.3 Bq/sample
6	180 °C	Al-foil	Hg-203	<0.3 Bq/sample
		Carbon filter	Hg-203	0.6 Bq/sample
5	320 °C	Al-foil	Hg-203	28 Bq/sample
		Carbon filter	Hg-203	46 Bq/sample

temperature and by using an appropriate absorber for Po.

4. Conclusions and outlook

Discs of lead–bismuth eutectic (LBE) were irradiated with protons having energies of 71 and 590 MeV resp. to analyse generated Po-isotopes. The generation of the isotopes Po 206 and 208 was clearly indicated. The ratio ^{206}Po/^{208}Po is dependent on the energy and as well on the intensity of the proton beam due to the different capture cross-sections for formation of different nuclides. As expected the isotopes ^{209}Po and ^{210}Po were not produced during the irradiation experiments most likely due to the fact that only fast neutrons were generated during the irradiation processes and that presence of thermal neutrons (that could produce these Po-isotopes) were negligible. The results of the calculations achieved with the FLUKA code concerning the ratio ^{206}Po/^{208}Po are in good agreement with the analytical results both at 71 and 590 MeV. Whereas the calculated amount of each Po-isotope (206 and 208) produced during the irradiation of 590 MeV and the calculated over-all activities are lower by almost a factor of 3. The reason for the discrepancy in the specific activities is currently under investigation.

The evaporation behaviour of irradiated LBE discs was examined as well whereby volatile Hg was found, but no Po evaporation was detected under the chosen conditions. Further investigation on irradiated LBE is necessary in order to examine the evaporation behaviour of different Po-isotopes at elevated temperature by variation of the experimental parameters and by using an appropriate absorber material.

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