Estimation of ^{210}Po Losses from the Solid ^{209}Bi Target Irradiated in a Thermal Neutron Flux

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Abstract

A brief summary of Po formation processes and release mechanisms from polonium-contained media is provided. Quantitative estimations of ^{210}Po losses from the solid ^{209}Bi target irradiated in the thermal neutron flux is given as an example. Other potential applications where this analysis could be directly applicable are mentioned.
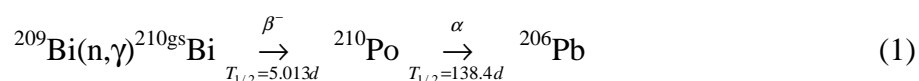
Introduction

The use of such heavy liquid metals as lead, bismuth and their alloy as a coolant in nuclear reactors (NR) and/or neutron production targets in the accelerator-driven systems (ADS) is connected with formation of alpha-radioactive isotopes of polonium under irradiation by neutrons or high energy protons. Due to a high radiotoxicity of polonium there is a rather strong limitation of its content in the environment (atmospheric air, water system, etc). Based on the nuclear safety standards one can conclude that in practice mainly polonium is the most dangerous among other nuclides characteristic for the applications as above.

In this context this work briefly summarizes the main processes how Po isotopes can be formed and, in particular, how they can be released from Po contained media. Detailed description of Po release processes including their quantitative evaluations is provided. Finally, we give a particular example on estimates of maximal ^{210}Po losses from the solid ^{209}Bi target irradiated in a thermal neutron flux at the high flux reactor of ILL Grenoble (France). We believe that a similar approach on Po formation and release estimates could be easily applicable for other applications such as ADS, neutron spallation sources, RNB production targets, etc., where either solid or molten metals containing Po will be present.

## Polonium formation and release processes

The main channel of formation of ^{210}Po in NR's with lead-bismuth coolant is due to the following reaction:

$$^{209}\text{Bi}(n,\gamma)^{210\text{gs}}\text{Bi} \underset{T_{1/2}=5.013d}{\overset{\beta^-}{\longrightarrow}} {}^{210}\text{Po} \underset{T_{1/2}=138.4d}{\overset{\alpha}{\longrightarrow}} {}^{206}\text{Pb} \tag{1}$$

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These NR's had been in long-term operation in Russian nuclear submarines (NS) and land-based prototypes of such NS. A specific α-activity of ^{210}Po in these facilities in some cases were found to be of the order of $\sim 3 \cdot 10^{11}$ Bq/kg.

In lead cooled NR like BREST-300 polonium is formed due to existence of bismuth admixture in lead (from $\sim 10^{-2}$ to $\sim 10^{-4}$ %), as well as due to the additional formation of ^{209}Bi through the following reaction:

$$^{208}Pb(n,\gamma)^{209}Pb \underset{T_{1/2}=3.25h}{\overset{\beta^-}{\rightarrow}} {}^{209}Bi \qquad (2)$$
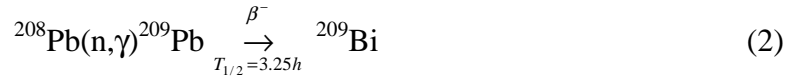
During a long-term NR operation at nominal power this reaction leads to the formation of ^{209}Bi in a considerable amount, and in this way actually accounts for the formation of the ^{210}Po in lead coolant. As a result, the activity of ^{210}Po in some cases can reach the levels of the order of $\sim 5 \cdot 10^7$ Bq/kg.

Besides ^{210gs}Bi as a source of formation of ^{210}Po there is also a metastable isotope ^{210m}Bi that is formed as a result of neutron capture reaction on ^{209}Bi and is a long-lived ($T_{1/2}=3 \cdot 10^6$ years) α-emitter. Its accumulation in the coolant is directly proportional to the energy production of NR. The concentration of ^{210m}Bi in the NR coolant of Russian NS's could be as high as $\sim 10^6$ Bq/kg.

Under high-energy proton irradiation of lead-bismuth ADS target some other α-activity polonium isotopes like ^{209}Po ($T_{1/2}=102$ years), ^{208}Po ($T_{1/2}=2.9$ years), etc., can be formed in a considerable amount.

Under normal operation, when the coolant or molten target circuit is pressurized, polonium is not of hazard. It may appear only during a depressurization of the circuit during repair works, refueling, inter-circuit leakage and/or emergency leakage of the coolant inside the working rooms. In this case the main reason of its danger is accounted for by α-activity aerosols of polonium in the work room air and surface contamination as a result of aerosol deposition, as well as a transfer of α-activity due to different contacts. That is why the treatment of radioactive polonium-contained coolant requires a rather high technology culture and taking of special measures to secure the radiation safety.

One of the particular features of the polonium is its high volatility [1,5]. Its maximal evaporation rate G_s^v in vacuum can be described by a modified Lengmuer equation using the Raul's law [6]:

$$G_{Po}^v = \{4.38 \cdot 10^{-3} \cdot a_t \cdot \gamma(T) \cdot P_{Po}(T) \cdot M_0 \cdot F\} / \sqrt{M \cdot T} \quad \text{, in Bq/s,} \qquad (3)$$

where
a_t is specific activity of Po inside polonium-contained media, in Bq/kg;
$P_{Po}(T)$ - pressure of saturated vapor of Po, in Pa;
M_0 - molecular mass of polonium-contained media, in g/mol;
F - evaporating surface, in m^2;
M - molecular mass of Po^{210}, in g/mol;

T - working temperature, in °K;

$\gamma(T)$ - chemical activity coefficient of Po in polonium-contained media.

Since 1946 the study of polonium distillation rate from bismuth media had been implemented in the Mound Laboratory relative to development of Po-Be neutron sources [1]. At that time it was found that polonium vapor pressure, measured in mm of mercury column, is considerably lower than that of saturated vapors of metallic polonium determined by Brooks, namely

$$\lg P_{Po}(T) = 7.2345 - 5377.8 / T . \tag{4}$$

This fact has confirmed that Po in vacuum has been presented as inter-metallic compound in the form like Bi-Po with a decreased volatility. The definition of Po activity coefficient has been entered as deviation of Po behavior from ideal solution of Po in the Bi sample. The following definition was proposed:

$$\gamma(T) = V_{P_o}(T) / P_{P_o}(T) , \tag{5}$$

where

$V_{Po}(T)$ is the volatility of Po from Bi sample at temperature T in °K (i.e. an observed polonium vapor pressure divided by the mole fraction of polonium in bismuth), in Pa;

$P_{P_o}(T)$ is saturated vapor pressure of metallic Po (see formula (4)), in Pa, at temperature T, in °K.

Engle in [1] has suggested the following equation to determine the polonium activity coefficient in bismuth:

$$\gamma(T) = 1.1176 - 2728.3/T \tag{6}$$

On the other hand, an observed polonium vapor pressure under diluted polonium solution in bismuth proposed in [1] can be described by the following equation:

$$V_{Po}(T) = \alpha(T) \cdot P_{Bi}(T), \tag{7}$$

where $\alpha(T)$ is a relative polonium volatility with respect to bismuth. In accordance with [1] $\alpha(T)$ can be determined from the following formula:

$$\lg \alpha(T) = -4.9828 - 18220 / (T + 1728). \tag{8}$$

In this case the dependencies $\gamma(T)$ and $\alpha(T)$ have been obtained for the temperature interval from ~450°C to ~850°C. Comparing (5) and (7) one can see that:

$$\gamma(T) \cdot P_{Po}(T) = \alpha(T) \cdot P_{Bi}(T) \tag{9}$$

However, under extrapolation of the equations (6) and (8) into the temperature region lower than 250°C the formula (9) is not valid due to uncertainties in determination of $\gamma(T)$, $\alpha(T)$, and $P_{Bi}(T)$. To be on the safe side, it was accepted in this paper that $\alpha(T) \cdot P_{Bi}(T)$ is higher than $\gamma(T) \cdot P_{Po}(T)$. We note that the formulas (6) and (7) are not dependant on polonium concentration in bismuth in a very wide range of concentrations (more than 4 orders of magnitude).

In the work of Tupper [2] and Feuerstein [3] it was also mentioned about less evaporation rate of polonium from Pb-Bi and Pb-Li eutectics in comparison with that of metallic polonium. Meanwhile, their results proved to be in a good agreement with those obtained from the formula (3) and by Russian specialists [6].

One more important feature has been found in [2,3,6], i.e. the polonium evaporation rate in gas atmosphere was considerably lower (about 3 orders of magnitude) than that in vacuum. The dependence of that effect versus gas pressure can be approximately described as follows:

$$G^{g}_{Po}(T) = G^{v}_{Po}(T) \cdot \beta(P), \qquad (10)$$

where
$$\beta(P) \approx 100/P, \text{ for } P \geq 10^5 \text{ Pa}. \qquad (11)$$

The above temperature dependence of polonium volatility allows to estimate potential polonium losses in different experiments related to the ^{209}Bi irradiation in a thermal neutron flux, and in particular where the target temperature did not exceed the melting temperature of Bi (271.4^{o}C).

Assessment of polonium formation and losses: the case study

The experiment was performed [7] at the high neutron flux reactor of ILL Grenoble. The thermal neutron capture cross section of ^{209}Bi for the ground state formation of ^{210gs}Bi was measured from the quantity of ^{210}Po formed via decay of ^{210gs}Bi (see equation (1) for details). A ^{209}Bi target was irradiated for 3 days in a thermal (Maxwell's distribution at 50°C) neutron flux of $2.3 \cdot 10^{13}$ n/cm^2/s. The target consisted of a pure ^{209}Bi (0.539 mg) metal deposited on the Ni backing disk (8 mm diameter and 0.125 mm thick) . The ^{209}Bi deposit thickness was 1.1±2% μm. The target was enclosed in Al cylindrical container (22 mm diameter, 62 mm height, and 1 mm thick). The container was filled with ~50% He and ~50% air gas at the atmospheric pressure. During the irradiation the outer surface of the container was maintained at 50 °C by the circulating water. The inner conditions of temperature and pressure were not well known but were estimated to be less than 250°C and 1.8 bar. In addition, no melting signs of ^{209}Bi have been observed after irradiation.

Taking into account a cross section of 24mb [8] for the neutron capture and the formation of ^{210gs}Bi, the mass m_{Po} of ^{210}Po formed during 3 days of irradiation and 84 days of cooling is given in Table I as a function of irradiation (+) and cooling time (–):

Table I. Accumulation of ^{210}Po versus irradiation and cooling time.

Time	+ 1 day	+ 2 days	+ 3 days	– 84 days
m_{Po}, ng	$1.73 \cdot 10^{-3}$	$6.7 \cdot 10^{-3}$	$1.47 \cdot 10^{-2}$	$5.25 \cdot 10^{-2}$

As the temperature in the target and the surrounding environment were not well known during the irradiation the calculations of maximal ^{210}Po loss by evaporation from Bi have been implemented for 3 temperatures: 250^{0}C (523^{0}K); 200^{0}C (473^{0}K), and 150^{0}C (423^{0}K). The temperature during the cooling was maintained at 20^0 C with a pressure of ~1 bar.

Calculations were made using formula (3) with replacement of $\gamma(T) \cdot P_{Po}(T)$ by $\alpha(T) \cdot P_{Bi}(T)$ and using formulas (10) and (11) as well. The bismuth saturated vapor pressure $P_{Bi}(T)$, in

mm of mercury, was determined using the following dependence obtained in [4] for the temperature range from 206^0**C** to 259^0**C**:

$$\lg P_{Bi}(T) = 7.213 - 8397 / T .\tag{12}$$

The results of calculations of the main parameters of interest are presented in Table II.

Table II. Main parameters obtained and used for final calculations.

Parameters	Target temperature		
	150^0C (423^0K)	200^0C (473^0K)	250^0C (523^0K)
$P_{Bi}(T)$, Pa	$3.08 \cdot 10^{-11}$	$3.86 \cdot 10^{-9}$	$1.89 \cdot 10^{-7}$
$\alpha(T)$	$3.08 \cdot 10^3$	$1.97 \cdot 10^3$	$1.29 \cdot 10^3$
$G_{Po}^g(T)$, Bq/s (at P=1.8 bar)	$3 \cdot 10^{-9}$	$1.96 \cdot 10^{-6}$	$2.1 \cdot 10^{-4}$
$\alpha(T) \cdot P_{Bi}(T)$, Pa	$0.95 \cdot 10^{-7}$	$7.70 \cdot 10^{-6}$	$2.44 \cdot 10^{-4}$

The estimated activity of ^{210}Po accumulated in the target after 1 and 3 days of irradiation, as well as after 84 days of cooling is presented in Table III.

Table III. Total and specific activities of ^{210}Po in the target (also see Table I for the masses).

Parameters	Irradiation (τ_0) and cooling time (τ_s) in days		
	$\tau_0=1$; $\tau_s=0$	$\tau_0=3$; $\tau_s=0$	$\tau_0=3$; $\tau_s=84$
Q_{Po} , Bq	$2.84 \cdot 10^2$	$2.44 \cdot 10^3$	$8.72 \cdot 10^3$
a_t, Bq/kg	$5.26 \cdot 10^8$	$4.52 \cdot 10^9$	$1.61 \cdot 10^{10}$

Finally, total loss of polonium from the target was determined from the equation:

$$Q^{vol}(T,\tau_0) = \int_0^{\tau_0} G_{Po}^g(T,\tau)d\tau\tag{13}$$

Assuming that the main loss of Po was occurred during irradiation at temperature T in ^{0}K during $\tau_0=3$ days, as well as introducing an average evaporation rate $\overline{G}_{Po}^g(T)$ within that time, one can obtain the equation for relative loss of polonium $\varepsilon(T)$ from the target:

$$\varepsilon(T) = (100 \cdot \overline{G}_{Po}^g(T) \cdot \tau_0)/Q_{Po}, \text{ in } \%,\tag{14}$$

where Q_{Po} is a total activity, in Bq, accumulated in the target during 3 days of irradiation and 84 days of cooling.

Average values of polonium evaporation rate and the predictions of the relative Po losses $\varepsilon(T)$ at different irradiation temperatures are presented in Table IV. Since we have used a maximal approach for Po loss estimates (see paragraph after formula (9)), the uncertainties for the final results are not indicated. In brief, from Table IV it is seen that Po losses due to the evaporation process are increasing with temperature but remain always well below 1% in the temperature range from 150^oC to 250^oC. Consequently, Po measurements in similar conditions cannot be influenced more than by a similar order of magnitude due to Po evaporation from the target.

Table IV. Total and relative losses of polonium from the target.

Parameters	Irradiation temperatures		
	150^0C (423^0K)	200^0C (473^0K)	250^0C (523^0K)
$\overline{G}_{Po}^{g}(T)$, Bq/s	$1.5 \cdot 10^{-9}$	$1.0 \cdot 10^{-6}$	$1.05 \cdot 10^{-4}$
$Q^{vol}(T,\tau_0)$, Bq	$3.9 \cdot 10^{-4}$	$2.59 \cdot 10^{-1}$	$2.72 \cdot 10^{1}$
$\varepsilon(T)$, %	$4.5 \cdot 10^{-6}$	$2.9 \cdot 10^{-3}$	$3.1 \cdot 10^{-1}$

Conclusions

Maximal Po losses were estimated in the case of the solid Bi target placed in a high thermal neutron flux for irradiation. We showed that not more than 1% of the created Po could be lost in the most penalizing scenario. This result is extremely important for the neutron capture cross section measurements, where alpha activity of ^{210}Po is an observable. A similar approach on Po formation and release estimates could be easily applicable for other applications such as ADS for nuclear waste transmutation, neutron spallation sources, RNB production targets, etc., where either solid or molten metals containing Po or other volatile substances are present.

Acknowledgements

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