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## I. INTRODUCTION

A knowledge of the production of polonium in fusion reactor blankets that use LiPb alloys as the breeder and coolant is of great importance in the overall evaluation of the safety of fusion reactors. The isotope of polonium which produces the greatest hazard is polonium 210. This isotope is an alpha emitter and has a half-life of 138 days. Because of the relatively short half-life, it does not pose a waste disposal problem; however, due to its high volatility and high biological hazard potential, it does pose a danger in the event of an accident in which coolant escapes confinement. As a part of the MARS reactor study the production of polonium 210 was considered and calculated for that reactor. This report, which is a series of extracts from the MARS report, is presented separately to illustrate the type of calculation and the levels of polonium 210 to be anticipated and to suggest a method of control. The report will first describe the calculational method which was used to determine the polonium 210 production rate; second, the results of the calculations will be given; and finally the blanket inventory will be determined.

## II. CALCULATION OF POLONIUM PRODUCTION RATE

### II.1 Introduction

The polonium produced in the MARS reactor arises from two principal reactions:

1.  $\text{Bi}^{209}(n,\gamma) \text{Bi}^{210} \xrightarrow{\beta^-} \text{Po}^{210}$ , and
2.  $\text{Pb}^{208}(n,\gamma) \text{Pb}^{209} \xrightarrow{\beta^-} \text{Bi}^{209}(n,\gamma) \text{Bi}^{210} \xrightarrow{\beta^-} \text{Po}^{210}$

These two reactions were considered separately. The calculation was also separated into beam and tube zones.

## II.2 First Wall Tube Zone Calculation

The first wall tube zone (closest to the plasma) was first considered. The first reaction results in  $3.53 \times 10^{15}$  atoms of  $\text{Po}^{210}$  produced per second. This is based on the assumption that a state of equilibrium has been reached, i.e. the  $\text{Bi}^{210}$  concentration has increased to its saturated level (this occurs several hours after irradiation begins). The number of atoms of Po produced/s is equal to  $5.54 \times 10^{-3}$  Ci of  $\text{Po}^{210}$  activity added per second in the entire tube zone. In other words, this is the amount of  $\text{Po}^{210}$  produced by the Bi impurity in LiPb in the tube zone.

The second reaction was split into two parts. If Part A of the reaction ( $\text{Pb}^{208}(n,\gamma) \text{Pb}^{209} \xrightarrow{\beta^-} \text{Bi}^{209}$ ) is calculated, information from the first reaction can be used to calculate Part B of the second reaction ( $\text{Bi}^{209}(n,\gamma) \text{Bi}^{210} \xrightarrow{\beta^-} \text{Po}^{210}$ ). In other words, if the amount of additional  $\text{Bi}^{209}$  that is produced is determined, we can determine the amount of polonium produced (by both reactions) by multiplying the initial production rate of  $\text{Bi}^{209}$  by one plus the ratio of  $\text{Bi}^{209}$  produced to  $\text{Bi}^{209}$  impurity initially present in the LiPb.

It was calculated that  $2.784 \times 10^{18}$  atoms of  $\text{Bi}^{209}$  were produced per second in the tube zone. Since there are  $1.056 \times 10^{26}$  atoms of  $\text{Bi}^{209}$  originally in the tube zone as an impurity in the LiPb, the equation for curies of  $\text{Po}^{210}$  produced per second is

$$\text{Ci}_{\text{Po}^{210}} \text{ added/s} = \text{Ci from Reaction 1} + \text{Ci from Reaction 1} \times$$

$$\frac{\# \text{ density of added Bi}^{209}}{\# \text{ density of Bi}^{209} \text{ initially}} \times \text{the fraction of Pb}^{209} \text{ being irradiated}$$

where

$$\# \text{ density of added Bi}^{209} = \text{Prod. rate of Bi}^{209} \times \text{time of irradiation} .$$

In the case of the tube zone, this yields

$$\text{Ci}_{\text{Po}^{210}} \text{ added/s} = 5.54 \times 10^{-3} \left[ 1 + \frac{2.784 \times 10^{18} t(0.136)}{1.056 \times 10^{26}} \right]$$

where  $t$  is the time of irradiation in seconds and  $(0.136)$  = fraction of  $\text{PbLi}$  being irradiated. This equation indicates that the length of time required for the  $\text{Po}^{210}$  production to double is 8.8 years.

### II.3 Beam Zone Calculation

In the rear sections of the blanket, designated the beam zone, reaction 1 produces  $2.567 \times 10^{14}$  atoms  $\text{Po}^{210}/\text{s}$  or  $4.4 \times 10^{-4}$  Ci of  $\text{Po}^{210}$  added/s. Using the same methodology as in the tube zone, we arrive at the equation

$$\text{Ci}_{\text{Po}^{210}} \text{ added/s} = 4 \times 10^{-4} + 4 \times 10^{-4} \left( \frac{4.916 \times 10^{18}}{1.5 \times 10^{26}} \right) (0.136)t$$

and the time required for the production of the  $\text{Po}^{210}$  to double was  $\sim 8$  years.

### II.4 Total Production Rate

1. The total (tube + beam zone) amount of activity produced from reaction 1 was  $6 \times 10^{-3}$  Ci of  $\text{Po}^{210}$  per second.
2. The total (tube + beam zone) amount of activity produced through reaction 2 could be expressed as a function of the amount produced from reaction 1, time of irradiation and the ratio of  $\text{Bi}^{209}$  produced to that present as an

impurity in the LiPb. This yielded  $\cong 6 \times 10^{-3}$  (t/8) Ci of Po added per second (t is the time of irradiation in years).

3. The total amount of polonium activity added per second throughout the entire blanket is

$$Ci_{Po} \text{ added/s} \cong 6 \times 10^{-3} + 6 \times 10^{-3} \times \frac{t(\text{years})}{8} .$$

### III. POLONIUM INVENTORY AND REMOVAL SYSTEM

The tritium recovery system (TRS) which recovers tritium by the vacuum degassing of droplets of the LiPb alloy can be used as the polonium cleanup system. The vapor pressure of Po is 2 torr at 500°C; therefore, the evaporation process will simultaneously remove Po from the coolant.

The Po decay and evaporation rate at steady state should be equal to the Po production rate which is  $6 \times 10^{-3}$  Ci/s,  $1.34 \times 10^{-6}$  g/s, as expressed by the relationship,

$$\frac{dm}{dt} + \frac{vnA}{4} = 1.34 \times 10^{-6} \text{ g/s} \quad (1)$$

in which  $dm/dt$  is the decay rate,  $m$  is the total mass of Po in the liquid metal,  $v$  is the molecular velocity for Po vapor,  $n$  is the particle density in vapor, and  $A$  is the surface area for evaporation. Since the half-life of Po is 138 days

$$\frac{dm}{dt} = m \left( \frac{0.693}{138 \times 3600 \times 24} \right) \text{ g/s} = m(5.9 \times 10^{-8}) \text{ g/s} . \quad (2)$$



The  $\text{Li}_{17}\text{Pb}_{83}$  flow rate to the TRS is  $1.76 \times 10^4$  kg/s, the droplet radius is 0.1 cm and the residence time is 1.0 s, so that the total surface area of evaporation is  $5.3 \times 10^7$   $\text{cm}^2$ .

The Po velocity can be calculated by

$$v = \sqrt{\frac{8 RT}{\pi M}} = 2.8 \times 10^4 \text{ cm/s} \quad (3)$$

in which R is the gas constant, T is the temperature, and M is the molecular weight.

The Po particle density per  $\text{cm}^3$  in the gas phase can be calculated by

$$Mn = \frac{MP}{RT} = \frac{Mx\gamma P_o}{RT} = \frac{M(m/M)}{(m_o/M_o)} \gamma P_o / RT \quad (4)$$

in which P is the partial pressure of Po in LiPb,  $P_o$  is the vapor pressure of pure Po,  $\gamma$  is the activity coefficient of Po in LiPb, x is the mole fraction of Po in the liquid phase, m is the Po mass, M is the Po molecular weight (= 210),  $m_o$  is the LiPb mass ( $= 1.46 \times 10^{10}$  g), and  $M_o$  is the LiPb molecular weight (= 173). By combining Eqs. (1)-(4) the following relation is obtained,

$$m[5.9 \times 10^{-8} + 9.0 \times 10^{-5} \gamma P_o] = 1.3 \times 10^{-6} \text{ g/s} .$$

If Po in LiPb is an ideal solution,  $\gamma = 1$ , then

$$m = 0.014 \text{ g}, \quad \text{or} \quad 64 \text{ curies} .$$

However, the activity coefficient of Po in bismuth has been measured<sup>(1)</sup> and

can be calculated by the relationship,

$$\log \gamma = - \frac{2728.3}{T} + 1.1176 .$$

If the activity of Po in LiPb is the same as that in Bi, then

$$\gamma = 3.9 \times 10^{-3} ,$$

and  $m = 3.6 \text{ g} ,$  or  $1.6 \times 10^4 \text{ curies} .$

Another mechanism which might retard the evaporation of Po is its rate of diffusion in the LiPb alloy droplets in the tritium removal system. For this calculation it is assumed that the diffusion of Po in the liquid LiPb alloy is similar to the self-diffusion of liquid Pb ( $4 \times 10^{-5} \text{ cm}^2/\text{s}$  at  $773^\circ\text{K}$ ); the radius of the droplets is 0.1 cm and the residence time is 1.0 s. Based upon the relationships<sup>(2)</sup> between  $(Dt/a^2)^{1/2}$ , and the rate of release from a sphere, approximately 20% of the Po would be released from the surface of the sphere if the release were principally controlled by the Po diffusion rate. The resulting concentration of Po in the liquid alloy at steady state, in which the quantity of Po evaporated in the TRS equals the Po generation rate, can be determined by the relationship,

$$\dot{v}(C_{IN} - C_{OUT}) = \dot{g} , \quad (9)$$

where  $\dot{v}$  = the flow rate to the TRS ( $1.76 \times 10^4$  kg/s)

$C_{IN}$  = the concentration of Po in the coolant entering the TRS

$C_{OUT}$  = the concentration of Po in the coolant leaving the TRS

$\dot{g}$  = the Po generation rate ( $1.3 \times 10^{-6}$  g/s).

Because  $C_{OUT}$  is 0.8  $C_{IN}$  the quantity

$$(C_{IN} - C_{OUT}) = 0.2(C_{IN}) ;$$

therefore, by use of Eq. (9)

$$C_{IN} = 3.8 \times 10^{-13} \text{ g Po/g alloy .}$$

The total quantity,  $m$ , of Po in the entire inventory,  $I$ , of coolant (14,600 tons) can be determined by the relationship

$$m = C_{IN} \times I .$$

Substitution of the proper numeric values indicates  $m = 5.5 \times 10^{-3}$  g Po.

This total mass of Po is less than that calculated for the evaporation of Po from an ideal solution. It can be assumed, therefore, that Po diffusion in the liquid alloy will not be the rate limiting process.

In conclusion, the Po inventory in LiPb is estimated to be between 64 and 16,000 curies depending on the activity coefficient. The TRS off-gas system will receive  $6 \times 10^{-3}$  Ci/s of Po, because the decay rate of Po is small compared to the evaporation rate.

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