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Preliminary analysis of polonium-210 contamination for China LEAd-based Research Reactor

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ABSTRACT

An important issue should be considered for liquid lead–bismuth eutectic (LBE) cooled reactors, which is the accumulation of radioactive ^{210}Po generated by neutron-irradiated LBE. It could cause problems during maintenance or refueling operations. Considering the safety of China LEAd-based Research Reactor (CLEAR-I), the activity of ^{210}Po in LBE coolant was calculated by VisualBUS system developed by FDS Team. The evaporation process from LBE to cover gas and migration to containing room and reactor building during normal operations were evaluated in this present work. According to preliminary analysis, there is little ^{210}Po contamination diffusing into the reactor building and its level in gaseous effluent was within the limits of NRC Regulation (10 CFR). It could not lead to problems during normal operations.

1. Introduction

With the development of nuclear energy, people are becoming more and more concern about the problem of radioactive waste disposal. Accelerator driven subcritical system (ADS) is considered as an effective method for nuclear waste transmutation (IAEA-TECDOC-985, 1997). In 2011, the Strategic Priority Research Program of “the Future Advanced Nuclear Fission Energy – ADS transmutation system” was carried out by the Chinese Academy of Sciences (Zhan et al., 2012). The Institute of Nuclear Energy Safety Technology/FDS team takes charge of the design and construction of the lead alloy cooled reactor and has finished the conceptual design of critical/sub-critical dual-models natural circulation lead alloy cooled reactor named China LEAd-based Research Reactor (CLEAR-I) (Wu et al., 2012, 2013; Bai et al., 2011).

Liquid lead–bismuth eutectic (LBE: 44.5%Pb–55.5%Bi by weight) is a promising material to be used in ADS as coolant, due to its nuclear, physical and chemical properties (Ohno et al., 2006). However, one of the most important issues is the generation and accumulation of polonium (^{210}Po) in LBE. ^{210}Po is a radioactive nuclide that emits 5.3 MeV alpha-rays with a 138.4-day half-life and it is highly volatile. The ^{210}Po would be generated mainly as a result of neutron capture of ^{209}Bi and subsequent beta decay as shown in Eq. (1). There are some well known research institutions such as the Paul Scherrer Institute (PSI, Switzerland), the Los Alamos National Laboratory (LANL, USA), the Tokyo Institute of Technology (Japan) and the Institute of Physics and Power Engineering (IPPE, Russia) are working on the issue of polonium (Amaya et al., 2009).

$$^{209}\mathrm{Bi}(n,\gamma)^{210}\mathrm{Bi}\ \xrightarrow[t_{1/2}=5d]{\beta^-}\ ^{210}\mathrm{Po}\ \xrightarrow[t_{1/2}=138.4d]{\alpha}\ ^{206}\mathrm{Pb} \tag{1}$$

We show below that most of the polonium is retained in the LBE coolant during normal operating conditions and without causing any danger if the coolant loop is sealed. However, polonium could accumulate in the cover gas due to its evaporation from LBE. Due to the normal leakage of the cover gas, polonium could diffuse outside the primary system to the containing room and reactor building, which may cause problems during maintenance or refueling operations.

Therefore, in this paper, we evaluate the distribution and migration of polonium contamination for CLEAR-I to discuss its safety. According to preliminary analysis, the ^{210}Po contamination level in gaseous effluent was found to be within the limits of NRC (Nuclear Regulation Commission) Regulation (10 CFR). And it can therefore be directly discharged into the environment during normal operations.

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2. Codes and methods

2.1. Calculation codes and data libraries

According to Eq. (1), the yield of polonium from LBE irradiated in a reactor can be calculated from the production of ^{210}Bi, the decay of ^{210}Bi to ^{210}Po, and the decay of ^{210}Po to ^{206}Pb. In order to carry out the above calculations, the neutron transport and material activation were calculated by VisualBUS (Wu et al., 2007, 2010) based on the use of the MCNP (X-5 Monte Carlo Team, 2003) transport code and the FISPACT-2007 (Forrest, 2007) inventory code. The neutron spectra of LBE coolant in various regions of the reactor were calculated using MCNP and the data library HENDL3.0 (Zou et al., 2010). These spectra are inputs to the inventory code FISPACT-2007 using the EASY-2007 data library to calculat the activities of the LBE coolant. The neutronics model of the reactor shown in Fig. 1 is modeled by MCAM (Wu, 2009; Zeng et al., 2006) (Monte Carlo Automatic Modeling Program for Radiation Transport Simulation) which was developed by FDS Team, China, as a bi-directional interface program between general commercial CAD systems and MC radiation transport codes (e.g. MCNP, TRIPOLI).

2.2. Calculation models for the migration of polonium

When generated by neutron activation in the lead-bismuth coolant, polonium rapidly forms lead polonide (i.e. PbPo) which is a chemically stable compound. This fundamental assumption has been directly confirmed at the Idaho National Laboratory (INL) (Amaya et al., 2009; Buongiorno et al., 2004). In accordance with Li et al. (1998), it is assumed that 99.8% of polonium is in fact in lead polonide form and only up to 0.2% present in LBE is in the unbound elementary form (i.e. pure Po) (Li et al., 1998; Buongiorno, 2001).

The polonium will evaporate from the hot LBE liquid into the cover gas (Argon) which is used to prevent the LBE coolant directly contacting air. The mole fractions of PbPo and Po in LBE coolant (calculated in Section 3) are extremely low. In this case, the LBE solution is approximately ideal, and Raoult's Law gives a sufficiently good estimate of the vapor pressure (Jolkkonen, 2007). Under a conservative assumption, we can assume that the activity coefficient (γ) in the Raoult's Law is one (i.e. $\gamma = 1$). So the PbPo and Po partial vapor pressure above the LBE bath can be evaluated as:

$$P_i = P_i^* \cdot x_i \tag{2}$$

Where P_i is the partial vapor pressure of PbPo or Po, in Pa, x_i is the PbPo or Po mole fraction in the LBE coolant and P_i^* is the pure PbPo or Po vapor pressure available from previous experimentations and can be computed from the following equations (Li et al., 1998; Buongiorno et al., 2003; Miura et al., 2007):

$$\log P_{\mathrm{Po}}[\mathrm{Pa}] = -\frac{5440}{T[\mathrm{K}]} + 9.46 \quad (368 \sim 604\ ^\circ\mathrm{C}) \tag{3}$$

$$\log P_{\mathrm{PbPo}}[\mathrm{Pa}] = -\frac{6790}{T[\mathrm{K}]} + 8.46 \quad (400 - 550\ ^\circ\mathrm{C}) \tag{4}$$

$$\log P_{\mathrm{PbPo}}[\mathrm{Pa}] = -\frac{7270}{T[\mathrm{K}]} + 9.06 \quad (640 - 850\ ^\circ\mathrm{C}) \tag{5}$$

Where log is the 10 base logarithm.

When the concentration of ^{210}Po in cover gas is in equilibrium during the normal operation of the reactor, the accumulating amount of ^{210}Po is considerable. Due to the normal leakage of argon, ^{210}Po could migrate to the containment room and reactor building from the cover gas. The containing room is used for protecting the control rod drive mechanism, in-vessel refueling system and containing the leaking radioactive cover gas. Reactor building is designed for protecting the reactor proper, primary loop and the second loop, and containing the leaking radioactive contamination. The configurations of containing room and reactor building are depicted in Fig. 2.

3. Results and discussion

It is assumed that the reactor runs continuously for 30 years with the full power ($\sim$10 MW) and without refueling. The neutron transport of the primary coolant was simulated by MCNP code and the neutron flux of different regions (labeled by these black border boxes) are shown in Fig. 3. Based on these neutron spectra, the activation of LBE was performed with the FISPACT2007 code. As Fig. 4 shows, the (n, γ) cross section of ^{209}Bi (ENDF/B-VII.1) has multiple resonances which create dips in the neutron flux spectra in Fig. 3.

According to our calculations, we found that the amounts of ^{210}Po could reach its maximum after 5 years operation as Fig. 5 shows. The maximum activity of ^{210}Po was approximately 1.4×10^{16} Bq and its specific activity in LBE $a_0 = 1.8 \times 10^{10}$ Bq/kg (760 tons of LBE).

The migration of ^{210}Po will be evaluated in the following subsections. Furthermore, in order to characterize the radiotoxicity of ^{210}Po, we would use of the concept of Derived Air Concentration (DAC) (Buongiorno et al., 2004; Larson, 2002). The DAC can be defined as the following: a worker inhaling air (which

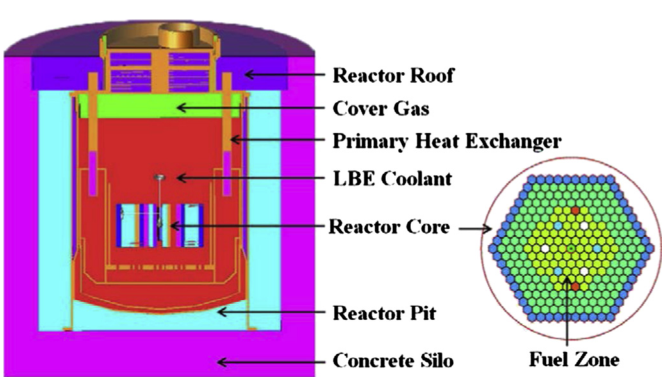

Fig. 1. 3D neutronics model of China LEAd-based Research Reactor.

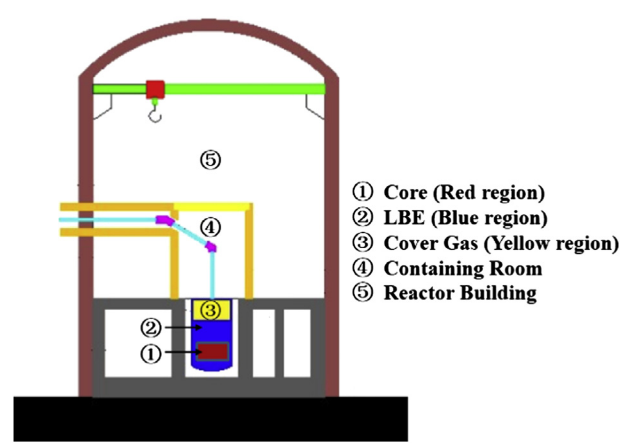

Fig. 2. The calculation model for the migration of the ^{210}Po.