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Technical Note

Experimental verification of thermal decomposition of lead polonide

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Abstract

The chemical form of polonium in lead–bismuth eutectic (LBE) is an important issue, considering the problem of polonium contamination in nuclear systems that use LBE as a coolant and/or an irradiation target. It has been thought that polonium exists as lead polonide in LBE. Polonium forms compounds with several metals, some of which decompose at high temperatures. Thermal decomposition of lead polonide was not confirmed experimentally, but the temperature of decomposition was foreseen to be around 600 °C. In this paper, the thermal decomposition of lead polonide and its decomposition temperature were confirmed using neutron-irradiated LBE. Neutron-irradiated LBE ingots containing polonium-210 were heated at temperatures of 550 ± 10 °C or 630 ± 10 °C in a vacuum. Polonium, lead and bismuth evaporated from the LBE ingots, and were deposited onto the surface of type 316 stainless steel (316SS) plates at various controlled temperatures between 220 ± 20 °C and 450 ± 20 °C. After heating, the number of alpha particles emitted from polonium-210 deposited on 316SS plates was measured. The experimental results showed a clear difference in the alpha particle count rate, which indicated that lead polonide decomposed at a temperature between 550 ± 10 °C and 630 ± 10 °C.

1. Introduction

Lead–bismuth eutectic (LBE) is a candidate coolant and irradiation target for the next generation of fast breeder reactors and accelerator-driven sub-critical systems (Gromov et al., 1994; Uchida et al., 2003; Zaki and Sekimoto, 1991). The thermal characteristics of LBE, such as its low melting point, high boiling point, and chemical inertness with air and water, make it suitable for utilization in nuclear systems. But there are some problems associated with utilizing LBE in nuclear systems, including the fact that such systems produce polonium-210. Namely, bismuth-209 changes to the bismuth-210 ground state or meta-stable state after neutron capture; the bismuth-210 ground state then decays to polonium-210 with a 5.0-day half-life. Polonium-210 is an alpha emitter of 5.3 MeV with a 138.4-day half-life, the alpha decay yield of which is almost 100%. The radioactive toxicity of polonium-210 is very high due to its alpha particle emissions. The (N,γ) reaction and subsequent decay reactions of bismuth-209 are shown in Fig. 1.

Elemental polonium at high temperature and its compounds such as polonium hydride show large volatility. The vapor pressure of elemental polonium was shown in the following equation (Abakumov and Ershova, 1974).

$$\log P_{\mathrm{Po}}(T) = 9.457 - \frac{5440}{T} \quad (368 - 604\ ^{\circ}\mathrm{C}), \tag{1}$$

where P is vapor pressure in units of Pa, and T is absolute temperature, K. According to a paper by Buongiorno et al., polonium hydride is a reaction product in the presence of humidity or hydrogen, thus its partial pressure is dependent on the concentrations and partial pressures of the reactants (Buongiorno et al., 2003). The Gibbs free energy variation of polonium hydride formation is measured and reported in the paper, thus it can be used to calculate partial pressure or concentration of polonium hydride if the gas phase temperature and conditions are known.

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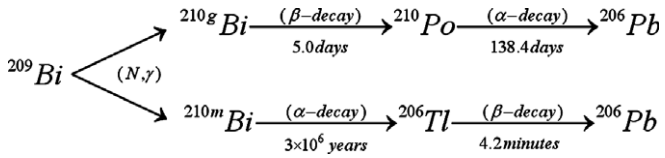

Fig. 1. (N,γ) reaction and subsequent decay reactions of bismuth-209.

Polonium-210 produced in neutron-irradiated LBE will diffuse into cover gas and onto material surfaces in the primary loop such as the heat exchanger and reactor vessel. The polonium-210 can be kept in the primary loop during nominal operation of a nuclear system if the primary loop is well sealed. However, the presence of polonium-210 puts workers at risk of a radioactive hazard. Workers should avoid direct contact with polonium-contaminated material surfaces and should not inhale aerosols of polonium-210 while performing maintenance on or decommissioning the primary loop. When an accident involving the rupture of the primary loop seal occurs, the neutron-irradiated LBE comes into contact with air and polonium-210 is released out of the primary loop. In such an accident, polonium-210 and its aerosol will contaminate a large surface area and the atmospheric space outside of the primary loop, resulting in a radioactive hazard to workers.

The transportation of polonium-210 depends on the chemical form of polonium-210 in LBE. It has been thought that polonium exists as lead polonide in LBE (Buongiorno et al., 2003; Loewen and Auman, 2004). Polonium forms compounds with several metals, such as silver, zinc, platinum, nickel and lead, some of which decompose at high temperatures. The thermal decomposition of lead polonide has not been experimentally confirmed, but it has been estimated that lead polonide will decompose to elemental polonium at around 600 °C (Goode, 1952). The vapor pressure of lead polonide is much smaller than that of elemental polonium shown as the following equations (Abakumov and Ershova, 1974; Buongiorno et al., 2003).

$$\log P_{\mathrm{PbPo}}(T) = 9.06 \pm 0.07 - \frac{7270 \pm 80}{T} (650 - 850\,^{\circ}\mathrm{C}), \quad (2)$$

$$\log P_{\mathrm{PbPo}}(T) = 8.46 \pm 1.26 - \frac{6790 \pm 1840}{T} (400 - 550\,^{\circ}\mathrm{C}). \quad (3)$$

Therefore, the thermal decomposition of lead polonide is a key issue, considering the potential release of polonium-210 from nuclear systems using LBE. In this study, experiments were performed to confirm the thermal decomposition of lead polonide using neutron-irradiated LBE ingots.

2. Experimental verification of thermal decomposition of lead polonide

A previous experimental result was used in the present experiments as a criterion to confirm the thermal decomposition of lead polonide. In the previous experiment, removal of elemental polonium from quartz glass surfaces, on which evaporated materials from neutron-irradiated LBE ingots had been deposited, was observed after heating at 300 °C in approximately 2 Pa (Miura et al., 2006; Obara et al., 2005). According to Eq. (1), the vapor pressure of elemental polonium at 300 °C is close to the system pressure of 2 Pa. Therefore, the polonium on the quartz glass surfaces would evaporate at 300 °C. From this previous experiment, it was expected that elemental polonium could not deposit onto material surfaces heated over 300 °C in a vacuum around 2 Pa. Hence, it would be possible to determine whether the chemical form of polonium-210 that evaporated from neutron-irradiated LBE and then was deposited onto material surfaces heated over 300 °C in a vacuum was elemental polonium or not. We heated neutron-irradiated LBE ingots and deposited the evaporated materials from the ingots containing polonium-210 to type 316 stainless steel (316SS) plates in a vacuum. The relationships between the alpha particle count rates measured from the 316SS plates and the temperatures, both of the neutron-irradiated LBE ingots and of the 316SS plates at heating, were investigated to confirm the thermal decomposition of lead polonide.

2.1. Preparation of neutron-irradiated LBE ingots

LBE ingots, each consisting of 2.225 g lead and 2.775 g bismuth, were prepared and placed in alumina crucibles. The fineness of the metals was 99.9999%. The LBE ingots in the crucibles were contained in two polyethylene cases and were irradiated by thermal neutrons to produce polonium-210 at neutron beam facility JRR-4 of the Japan Atomic Energy Agency (JAEA). Before the neutron irradiation, some gold wires whose length and weight were approximately 5 mm and 5 mg, respectively, were set on both sides of the cases to estimate the fluence of thermal neutrons. After the thermal neutron irradiation, the radioactivity of the gold wires was measured, and the fluence of thermal neutrons was calculated to be approximately 10^{13} n/cm^2. The specific radioactivity of polonium-210 produced in the LBE ingots was calculated from the fluence of thermal neutrons and the neutron capture cross section of bismuth-209. After several months of radioactive cooling, the specific radioactivity of polonium-210 in the ingots was calculated to be between 20 Bq/g and 40 Bq/g, at which point they were used in the experiments.

2.2. Experimental apparatus

The experiments made use of an electric furnace that had a quartz glass tube surrounded by a heater (main heater) (Fig. 2). The tube radius and height were 40 mm and 300 mm, respectively. All experiments were performed in the tube using two additional heaters: sub-heater 1, designed for the deposition of evaporated materials from a melted neutron-irradiated LBE ingot; and sub-heater 2, designed for heating a neutron-irradiated LBE ingot in an alumina crucible. Sub-heater 1 was connected to a water-cooling system. The temperature of each heater