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Fundamental study of polonium contamination by neutron irradiated lead–bismuth eutectic

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Abstract

As a fundamental study of polonium contamination by neutron irradiated LBE, it was investigated to remove polonium surface contamination by baking method. The baking experiments were performed using quartz glass plates contaminated by material evaporated from neutron irradiated LBE liquid. The contaminated quartz glass plates were baked in vacuum (2 Pa) at various temperatures. The experimental results clearly show that polonium evaporated from LBE can be removed by baking samples at temperatures 300 °C and above. It is of note that the decrease in the weight of deposited materials baked at 300 °C differed from that observed at 400 °C or higher temperatures. At temperature of 300 °C, no change in weight was observed. The mass of polonium in the LBE samples was so small that no weight change could be observed by release of polonium. Thus, it might show that only the polonium among the adherent materials was removed by baking at 300 °C without removing other adhered material. The method is rather simple, so it is easy to apply the method for practical application. One of the expected applications may be the removal of polonium contamination in a primary loop before maintenance work of the loop. Also it shows that this method can be used to avoid the release of polonium from contaminated material, in case of an accident, by keeping the contaminated material at low temperature.

1. Introduction

Lead–Bismuth Eutectic (LBE) has a lot of advantages as coolant and target of accelerator driven sub-critical systems. It has a low melting point temperature, high boiling point temperature and it is chemically inert. This characteristic is also favorable as coolant of a fast reactor. A lot of research activity has been performed for utilization of LBE in nuclear systems [1–5]. One of the major problems to be solved is corrosion and erosion of metallic materials by LBE. Another problem is polonium contamination by neutron irradiated LBE [6–9]. By neutron capture of bismuth-209, polonium-210 is produced in LBE. Polonium-210 is a radioactive nuclide, which emits 5.3 MeV α-rays with 138 days half life. Special attention should therefore be paid in view of radioactive hazard. One of the important problems of this issue is polonium surface contamination from neutron irradiated LBE. Polonium-210 will be evaporated due to neutron irradiated LBE and it will stick on various material surfaces. A simple decontamination method can be important for the maintenance work of primary LBE loops and in LBE coolant leakage accidents.

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Experiments have been started to develop a method to remove the polonium surface contamination removal by baking method [11]. In this paper, the present status of the experiment is described.

2. Experiment for removal of polonium surface contamination by baking method

Some previous work shows that there is a possibility that polonium can be volatile if the temperature is above about 300 °C [10]. This fact suggests that the polonium sticking on a material surface can be released from the material if the material temperature becomes high. The possibility to remove polonium surface contamination by baking the contaminated material in vacuum condition is investigated. In the experiment, polonium was evaporated from neutron irradiated LBE and deposited on quartz glass plate. It was tried to remove the polonium on quartz glass plate by baking the glass. The reason why quartz glass plates were used for the experiment was that quartz glass plate was often used for experimental apparatus and that it was easy to investigate the contamination on the surface. The experimental procedure was the following:

LBE samples were prepared in alumina pots. The inner radius of the pots was 24 mm and the depth was 19 mm. Total weight of lead and bismuth was 5 g for each pot. The weight fraction of bismuth was 55.5%. Thermal neutron irradiation was performed to the LBE samples. For the irradiation, JRR-4, a research reactor in Japan Atomic Energy Research Institute, was used. The irradiation was performed at thermal neutron beam facility in JRR-4. Thermal neutron fluence in the irradiation was about 1.6×10^{13} cm^{-2}. After cooling of the LBE samples, they were used for the experiments. Polonium density in LBE was calculated from the thermal neutron fluence. Estimated specific radioactivity of polonium-210 in LBE was about 30 Bq g^{-1} when the experiment was performed.

Polonium from the LBE samples was deposited on quartz glass plates by the following way. A piece of quartz glass plate was put on the alumina pot that contained the neutron irradiated LBE. The pot and the glass plate were placed in an infrared furnace. Air in the furnace was removed and the furnace was filled with argon gas to avoid the chemical reaction of LBE with air. The sample was heated in the furnace. The maximum temperature of LBE surface was about 900 °C during the heating. The temperature of quartz glass surface was about 600 °C. The temperatures were measured by thermocouples. After the heating, the sample was cooled by natural cooling to room temperature. By the heating and the cooling, material evaporated from the LBE stuck on the surface of the quartz glass plate. The mass of stuck material was between 20 and 50 mg. The error of mass measurement was less than 1 mg. α-Ray from the stuck material was also measured using semiconductor α-ray spectrometer. Alpha-ray count from the stuck material was from $1 \times 10^{+5}$ to $2 \times 10^{+5}$ for 20 h measurement. So the statistical error of α-ray count was less than 1%.

Baking experiment was performed using the polonium contaminated quartz glass plates. After the measurements, the quartz glass plates with the material evaporated from LBE were placed in the infrared furnace again and were baked. Vacuum condition was kept during the baking experiment. The vacuum pressure was 2 Pa. To know the effect of baking temperature and baking time, baking was performed at various temperatures and time. The temperature was 200, 300, 400, and 500 °C. Baking time was 2, 5, 15, 60, and 180 min. To know the effect of repetition of baking, baking was repeated four times in the maximum. The results of repetition of baking were compared with the results of baking in long time. If there is no difference between them, it can be possible to predict the effect of baking in very long time from the results of short baking time. After the baking, the mass of stuck material on the quartz glass plates and α-ray count from the material were measured using same method as before. Measured count rate was between 10^3 and 10^5 for 20 h measurement. So the statistic error was less than 1% to the initial count rate. If the α-ray count decreases, it means that polonium stuck on the quartz glass surface was removed by the baking operation. The amount of polonium in the stuck material is so little that decrease of mass cannot be observed by the removal of polonium. If the decrease of mass is observed, it means that stuck material except polonium is removed by the baking. The change of mass of stuck material and α-ray count from the material was measured by the comparison of mass and α-ray count before and after the baking respectively. By the comparison, both the effect of removal of polonium and the effect of removal of other non-radioactive material were investigated. As a preliminary experiment, a quartz glass plate without deposited material was baked. No change of mass was observed by the baking.

3. Experimental result

The results of baking experiment are shown in Figs. 1–3. The statistic error of α-ray count were less than 1%, so no error bars are written in the figures. Fig. 1 shows the result of baking at the temperature of 200 °C. The figure shows the change of mass of stuck material and the change of α-ray count by baking. Baking time was 5, 15, and 60 min. Repetition of baking was 4 times, 2 times, and 4 times respectively. The figure shows the following fact. There was no decrease of mass by the baking. No effect was observed by long time baking. No change was observed by repetition of baking. No change

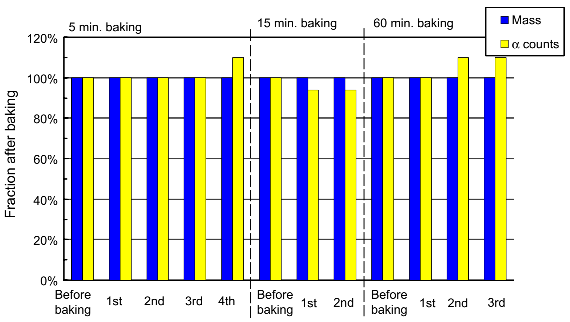

Fig. 1. Fraction of mass and α-ray count after baking (200 °C).

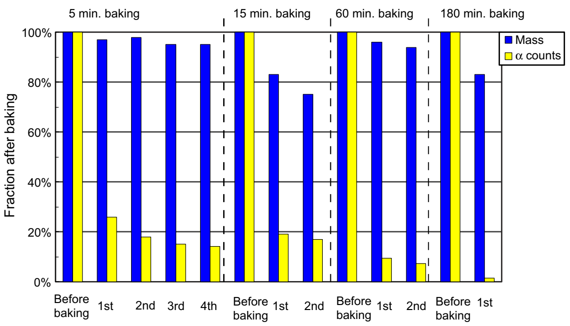

Fig. 2. Fraction of mass and α-ray count after baking (300 °C).

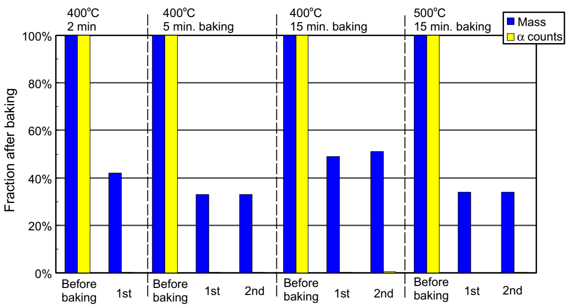

Fig. 3. Fraction of mass and α-ray count after baking (400 °C and 500 °C).

of α-ray count was observed by the baking except fourth trial of 5 min baking, second and third trial of 60 min baking. In the cases, the increases of α-ray count were observed. The increases were more than statistic error. The changes of α-ray spectrum were observed also in the cases. No effect was observed by longer baking and by repetition of baking too. This means that polonium contamination cannot be removed by the baking at 200 °C and that other non-radioactive material cannot be removed also.

Fig. 2 shows the result of baking at 300 °C. The time of baking was 5, 15, 60, and 180 min. The repetition of baking was 4 times, 2 times, and 2 times respectively. Repetition of baking was not performed in 180 min baking. By the baking the mass of stuck material decreased. But the decrease was not so much. By the baking for 15 min, the decrease of mass was approximately 20%. The decrease of mass by the baking for 5 min was small. By the repetition of baking four times, the decrease of mass was less than 10%. On the other hand, α-ray count decreased much by the baking. By baking for 5 min, the decrease of α-ray count after the baking was more than 80%. By the repetition of the baking, the α-ray count was decreased further. Long baking time resulted in large decrease of α-ray count. By the baking for 180 min, the decrease of α-ray count was more than 95%. This means that most of all polonium on the quartz glass plate was removed by the baking. The facts show that it is possible to remove polonium contamination on the quartz glass plate by baking in 300 °C. The decease of stuck material mass was less than 20%. This means that the removal of stuck material except polonium on the quartz glass is not so much by the baking.

Fig. 3 shows the result of baking at 400 °C and 500 °C. Baking time was 2, 5, and 15 min for the baking at 400 °C and was 15 min for the baking at 500 °C. Repetition of baking was 2 times in the maximum. The figure shows the following. The decease of stuck material mass was about 60% both in 400 °C and 500 °C. The repetition of the baking had little effect on the decrease of mass of stuck material. Alpha-ray count became almost zero after the first baking trial. This means that almost all polonium was removed by the baking.

Fig. 4 shows the relationship between baking temperature and decrease of mass after baking. The figure shows the following. If the baking temperature is 200 °C, the decrease of mass was little. If the baking temperature is 300 °C, the decrease of mass is about 20% in the maximum. If the baking temperature is 400 °C or above, the decrease of mass by the baking is about 60%.

Fig. 5 shows the relationship between baking temperature and decrease of α-ray count. The figure shows the

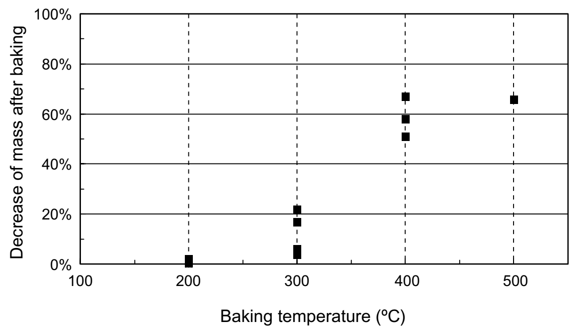

Fig. 4. Relationship between baking temperature and decrease of mass after baking.

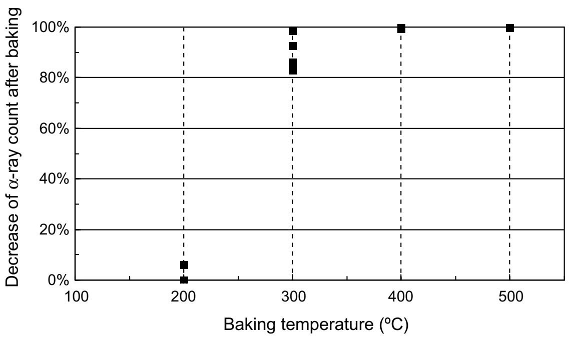

Fig. 5. Relationship between baking temperature and decrease of α-ray count.

following. If baking temperature is 200 °C, α-ray count does not change by baking. If baking temperature is 300 °C, the decrease of α-ray count is more than 80%. If baking temperature is 400 °C or above, the decrease of α-ray count is almost 100%. The change of the α-ray count shows the amount of removed polonium from quartz glass plate surface. The fact shows that polonium contamination can be removed effectively by the baking in 300 °C or higher temperature.

4. Discussion

By the baking experiments, an interesting characteristic was observed that was similar to the results obtained by Abakumov et al. [10]. In Abakumov's experiment, pure polonium became volatile when the temperature became above 300 °C. But it is difficult to discuss the phenomena in detail by comparing the baking experiment results against Abakumov's results, because the experimental condition was so different from the baking experiment.

To understand the phenomena, it is necessary to know the elements and the chemical forms of the stuck material. For the good understanding of stuck material, it can be also important to know the temperature of the evaporation from the LBE and the temperature when the material sticks on the quartz glass plate. In the experiment, it was difficult to set the temperature of LBE and the temperature of the quartz glass independently during the heating. The evaporated material was stuck on the glass by heating the LBE and the glass to high temperature and by the natural cooling to room temperature. So the temperature of the evaporation and the stick could not be observed. For the detail investigation of the phenomena, it is necessary to perform the experiment by setting the temperature of them independently. The LBE temperature should be set to the temperature of LBE in the expected system, and the stick temperature should be set to the materials in the system. And the analysis of the stuck material should be performed to know the elements and the chemical forms. The reason also can be clear why only a few percent of polonium was deposited on the quartz glass by doing the experiments with setting the temperatures independently.

A special furnace may be needed to perform the experiment. Quartz glass plates were used for surface contaminated material in the experiment. Experiments using metallic materials are also needed to confirm the effect of baking of polonium contaminated metallic materials. The experiments should be performed for the metallic materials that are expected to use in expected system.

When increase of α-ray count was observed in the baking at 200 °C, the change of α-ray spectrum was also observed. This suggests the change of polonium distribution in the material. Further study is needed to make clear the phenomena.

The contamination removal by baking is rather simple method, so its application is expected to be easy. The method can be effective for the material with complex geometry also. One of the applications of the method can be polonium contamination removal in LBE primary loop of nuclear system before maintenance of equipments of the loop, for example pipes, valves, pumps and so on. Before the maintenance, LBE in the loop should be drained. After the drain, polonium contamination can be stuck on the surface of the material that is inside of primary loop with other non-radioactive materials from LBE. To remove the contamination, the temperature of primary loop is kept more than 300 °C in vacuum condition. After that, inert gas, such as argon, is circulated in the primary loop with keeping the loop high temperature. By the heating polonium stuck on the surface inside the primary loop will be removed. At the exit of the gas, a cold trap is set. The temperature of the cold trap is kept lower than 200 °C. Removed polonium will be captured by the cold trap. Maintenance of primary loop can be done after all of the polonium in the primary loop is removed by the method. By this method, it will be possible to avoid the radiation hazard of polonium during maintenance of primary loop.

The experimental result suggests that the possibility of evaporation of polonium from contaminated material is low, if the material temperature is kept low. So it can be possible to avoid the contamination from polonium contaminated material by keeping the material temperature low. This can be effective to minimize polonium radioactive hazard in LBE coolant leakage accident. To know the possibility of the methods, it is needed to know the relationship between evaporation temperature, polonium stick temperature, baking temperature, and the contaminated material in more detail.

5. Conclusion

Experiment was performed to investigate the possibility to remove polonium which is evaporated from neutron irradiated LBE and which sticks on quartz glass plate by baking the glass in vacuum condition. The experiment demonstrated the following fact. If the baking temperature is 400 °C or higher, the polonium can be removed within 5 min by baking. In this case, other non-radioactive materials are removed altogether. If the baking temperature is 200 °C or lower, the polonium cannot be removed. If the baking temperature is about 300 °C, the polonium can be removed without removing other non-radioactive materials. This phenomenon can be useful for practical application of polonium contamination removal. The baking method is so simple that it can be applied to remove polonium contamination even if the contaminated material geometry is complicated. It may be possible to apply the removal of polonium contamination in LBE primary loop before maintenance of the loop. It can be also effective to avoid polonium release from polonium contaminated material by keeping the contaminated material temperature low. To know the phenomena in detail, further experiments are needed to investigate the relationship between polonium evaporation temperature from LBE, material temperature when polonium stick occurs, elements and chemical forms of stuck material, and baking temperature. In the present experiment, quartz glass plates were used for contaminated material. To make clear the possibility of baking method, it is needed to perform experiments using various metallic materials.

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