Development of Polonium Surface Contamination Measure in Lead-Bismuth Eutectic Coolant

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Introduction

- Lead-Bismuth Eutectic (LBE)
  - advantageous characteristics such as a high boiling point, a low melting point, and chemically inert.
  - can be utilized as a coolant and/or target for innovative fast reactors and accelerator-driven sub-critical systems (ADS).

- Polonium issue
  - one of the problems to be solved in the use of LBE
Polonium-210

- produced by neutron capture of bismuth-209 in LBE
- a radioactive nuclide that emits 5.3 MeV alpha-rays with a 138-day half life
- can be a serious problem in inhalation or sticking to skins
Measure for polonium issue

- The shielding of alpha-ray is easy.
- Not a serious problem if the sealing of the primary loop is good.
- It can be a serious problem in the maintenance of the primary loop or in the coolant leak accident.
- Removal of polonium surface contamination is necessary before the maintenance or in the accident.
Polonium surface contamination removal

- Use of solvent
  - The area in the primary loop to be decontaminated can be large.
  - The amount of contaminated solvent can be also large.
  - The regulation of polonium contaminated liquid is strict.
  - It can be almost impossible to release polonium contaminated solvent to the environment.

- Other effective method should be developed.
Present study

1. Study of baking method for polonium contamination
2. Study of Method to analyze polonium density distribution by alpha-ray spectrum using unfolding method
1. Study of baking method for polonium contamination

- **Purpose**
  - Development of baking method for the removal of polonium surface contamination

- **Experiments**
  - Quartz glass plate
  - Stainless steel plate (preliminary)
Quartz glass baking experiment
-Polonium contamination of quartz glass plate -

- Neutron irradiated LBE was heated in an infrared furnace which was filled with argon gas (Temp. 900 °C).
- During the heating, a piece of quartz glass plate was set on the alumina crucible (Temp. 600 °C).
Quartz glass baking experiment
-Baking of contaminated quartz glass-

- The contaminated quartz glass plate was baked in a vacuum (2Pa).
- Baking temperature
  - 200 °C, 300 °C, 400 °C, 500 °C
- Baking time
  - 2min., 5min., 15 min., 60min., 180min.
- Repetition of baking
  - Max. 4 times
Quartz glass baking experiment  -Measurements-

- Before and after the each baking, the weight of stuck material on the glass and the alpha-ray from the material were measured.
- The effect of the baking was estimated.
  - Change of mass
  - Change of alpha-ray count
Quartz glass baking experiment -Results-

Fig. 1 Decrease of mass after baking

Fig. 2 Decrease of alpha-ray count after baking
Stainless steel baking experiment
-Polonium contamination of stainless steel -

- Neutron irradiated LBE in an alumina crucible was heated in a vacuum (610°C, 0.4 Pa).
- A 316SS plate was set above the LBE (230°C).
- Mass and alpha-ray count of adherent material on the stainless steel plate were measured.
Stainless steel baking experiment
-Baking of polonium contaminated stainless steel -

- The contaminated stainless steel plate was baked in a vacuum (0.4Pa).
- Baking temperature
  - 300°C, 400°C, 500°C, 600°C
- Baking time
  - 30 min.
- Mass and alpha-ray count were measured after the baking.
Stainless steel baking experiment -Results -

Fig. 3 Decrease of mass after baking

Fig. 4 Decrease of alpha-ray count after baking
Discussion for baking experiment

- Polonium on stainless steel was released at 500°C (0.4 Pa). Vapor pressure of Pb-Po at 500°C is 0.47 Pa (Buongiorno, 2003). This fact suggests that polonium on the stainless steel was Pb-Po.
- Polonium on quartz glass plate was released at 300°C (2 Pa). This fact suggests that polonium on the quartz glass was a simple substance of Po.
- The reason of the difference is under investigation. (Because of the difference of evaporation and adherent temperature? Because of the difference of material (quartz glass, stainless steel? )
- There is a possibility to remove polonium contamination without removing other non-radioactive material by the baking method.
2. Study of Method to analyze polonium density distribution by alpha-ray spectrum unfolding method

- **Purpose**
  - Development of a method to estimate polonium-210 density distribution in LBE metal by alpha-rays spectrum from the surface.
  - It can be used for investigation of polonium surface contamination.
Method of polonium distribution estimation from alpha-rays spectrum

Detector

Alpha-rays spectrum

Unfolding

Polonium distribution in lead-bismuth eutectic

Decrease of energy

Polonium-210
Unfolding calculation

\[ c_i = \sum_{j=1}^{n} A_{ij} x_j \quad (i = 1, \ldots, m) \]

\[ (j = 1, \ldots, n) \]

Constraint

\[ x_j \geq 0 \]

\[ m > n \]

\[ C_i : \text{Counting rate of } i\text{'th bin in alpha-rays spectrum} \]

\[ A_{ij} : \text{Response function for } j\text{'th layer} \]

\[ x : \text{Number of alpha-rays sources at } j\text{'th layer} \]

\[ m : \text{Number of energy bins of alpha-rays spectrum measurement} \]

\[ n : \text{Number of layers of LBE column} \]

\[ S_{ii} : \text{Error of } C_i \]

Optimum \( x \) is calculated to minimize \( J \).

\[ J = (Ax - c)^T S^{-2} (Ax - c) \]

UFO/Q code, which was developed for neutron spectrometry, was used.
Example of measured alpha-ray spectrum

$\alpha$-counts from Ingot2-5

T. Obara, INES-1(2004)
Calculation result: Polonium distribution (40 layers)
Discussion for polonium distribution analysis

- It was shown that vertical distribution of polonium from the surface could be calculated by the spectrum of alpha-rays from the surface.
- To estimate the distribution exactly, alpha-rays spectrum measurement with low errors is needed.
- It is difficult to estimate the distribution very close to the surface because of rapid change of the spectrum.
- It is expected to utilize the method for the study of polonium migration in metals and for the investigation of polonium surface contamination.
Conclusion

- The polonium contamination issue were discussed.
  - To develop an effective measure to remove polonium surface contamination is important.
  - Decontamination using solvent can not be effective.
Experiments to remove polonium contamination on quartz glass by baking method were performed.

- The polonium can be removed if the baking temperature is 300°C or higher in a vacuum (2 Pa).
- This fact suggests that the polonium is single substance.
Preliminary experiments to remove polonium contamination on stainless steel by baking method were performed.

- The polonium can be removed if the baking temperature is 500°C or higher in a vacuum (0.4 Pa).
- This fact suggests that the chemical form of the polonium is lead-polonide.
- It is needed develop an effective method by low baking temperature.
A method to estimate polonium density distribution in depth by spectrum of alpha-ray was developed using unfolding method.

- The method make it possible to investigate the polonium diffusion phenomena in metal.
- The method can be used for the investigation of the polonium surface contamination.