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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 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

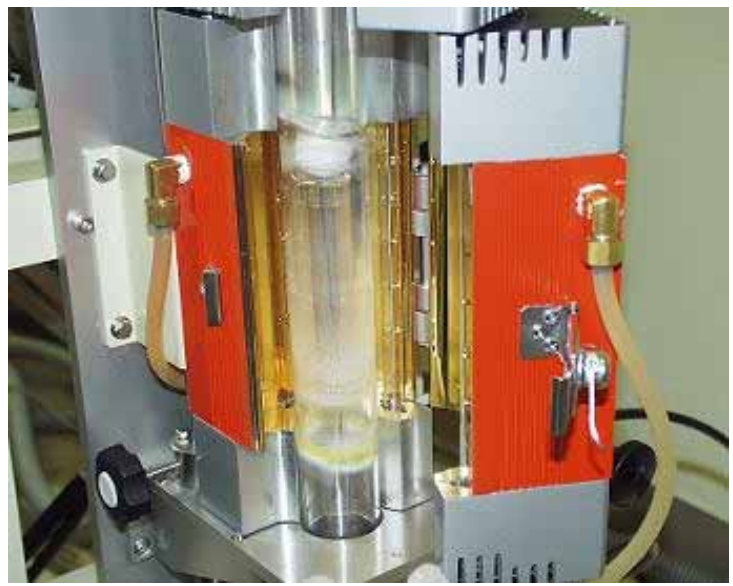
1. Study of baking method for polonium contamination
2. Study of Method to analyze polonium density distribution by alpha-ray spectrum using unfolding method

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℃).
- During the heating, a piece of quartz glass plate was set on the alumina crucible (Temp. 600℃).

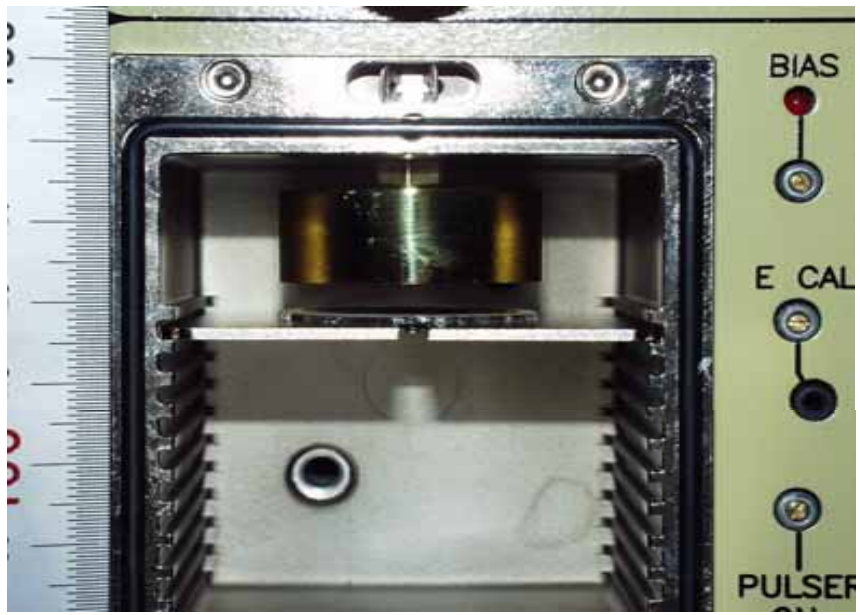


Quartz glass baking experiment
-Baking of contaminated quartz glass-

- The contaminated quartz glass plate was baked in a vacuum (2Pa).
- Baking temperature
  - 200℃, 300℃, 400℃, 500℃
- 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-

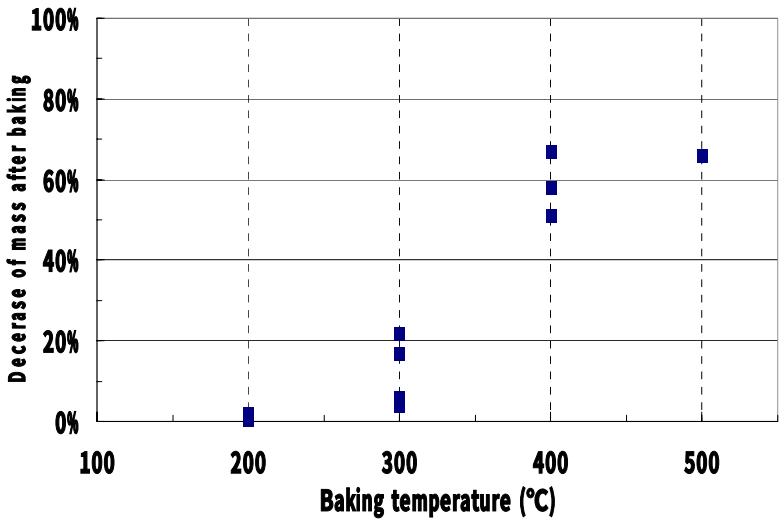


Fig. 1 Decrease of mass after baking

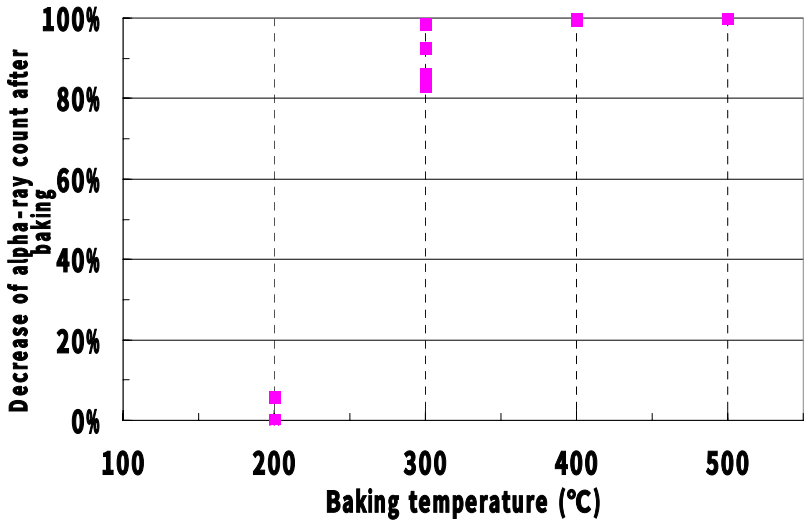
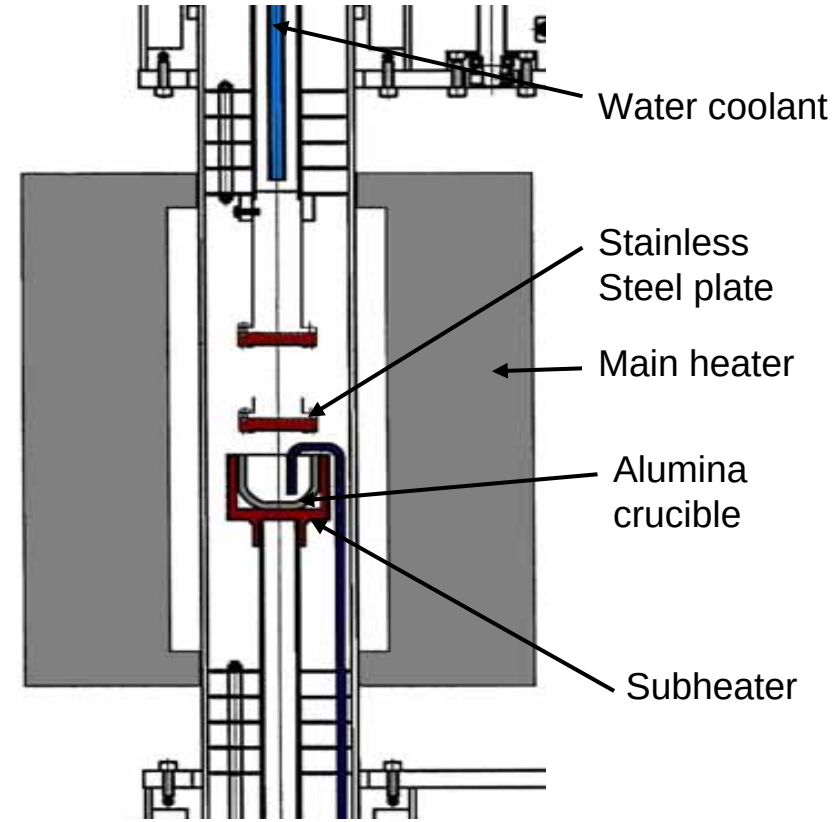


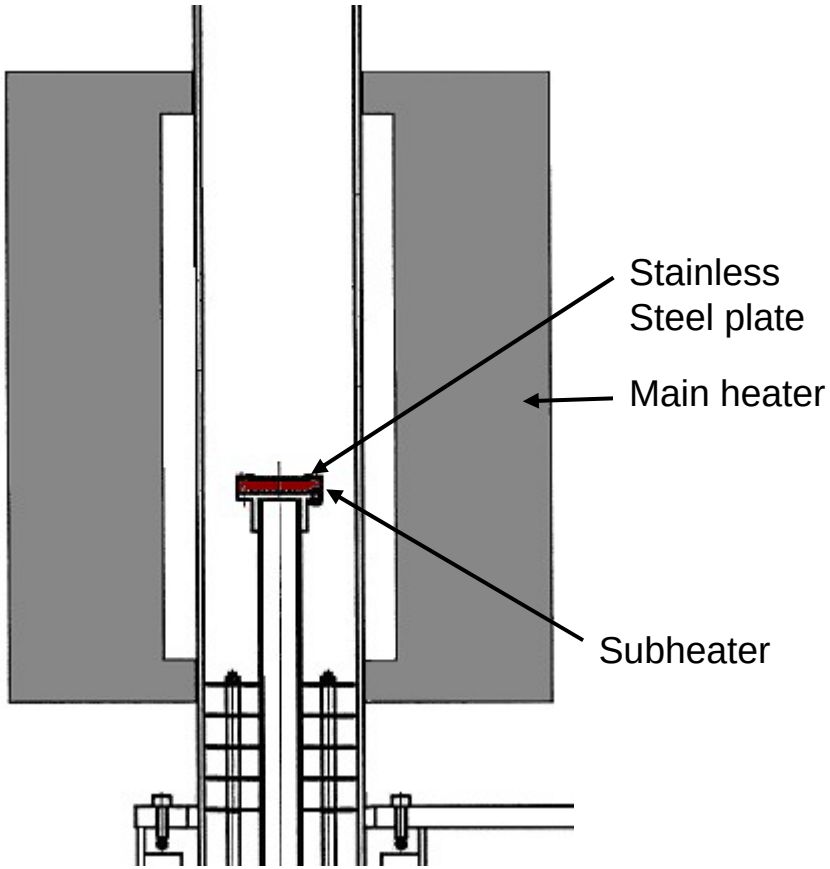
Fig. 2 Decrease of alpha-ray count after baking

Stainless steel baking experiment
-Polonium contamination of stainless steel -



- Neutron irradiated LBE in an alumina crucible was heated in a vacuum (610ºC, 0.4Pa).
- A 316SS plate was set above the LBE (230ºC).
- 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
  - 300ºC, 400ºC, 500ºC, 600ºC
- Baking time
  - 30 min.
- Mass and alpha-ray count were measured after the baking.

Stainless steel baking experiment -Results -

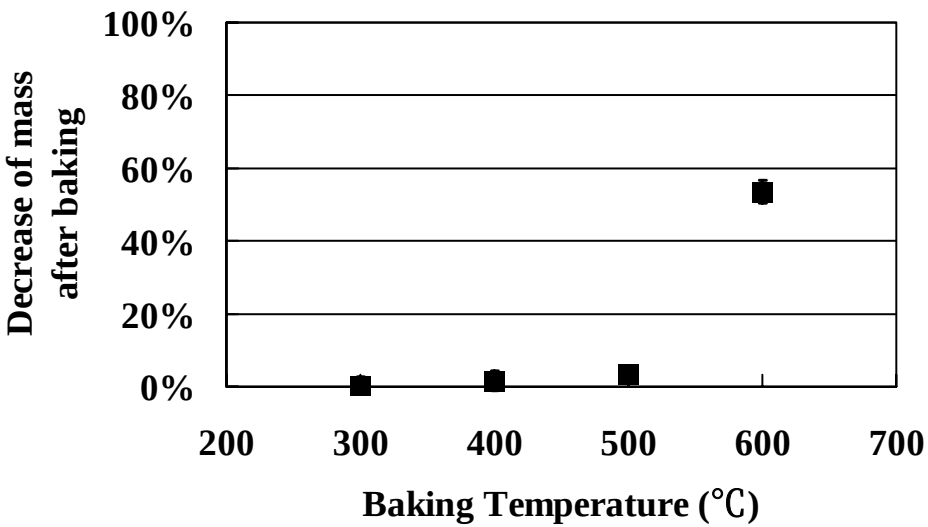


Fig. 3 Decrease of mass after baking

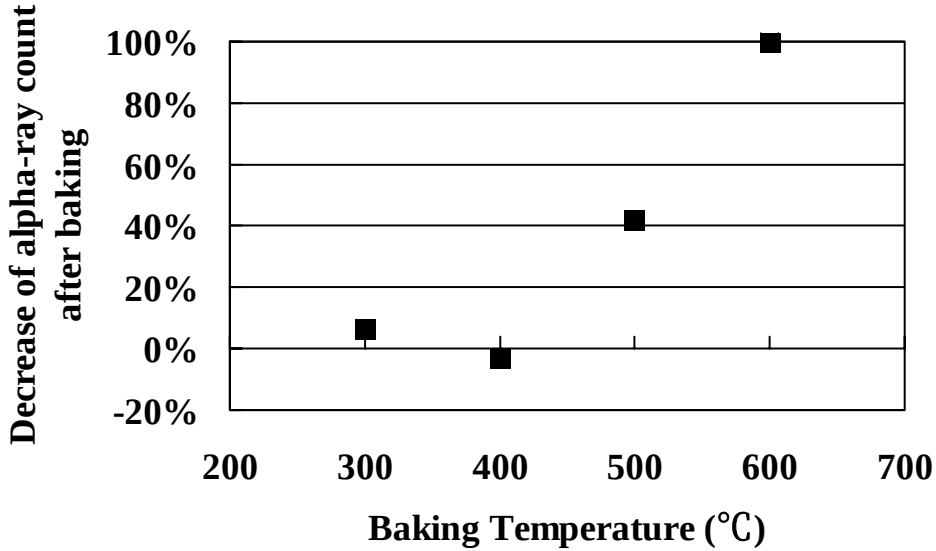


Fig. 4 Decrease of alpha-ray count after baking

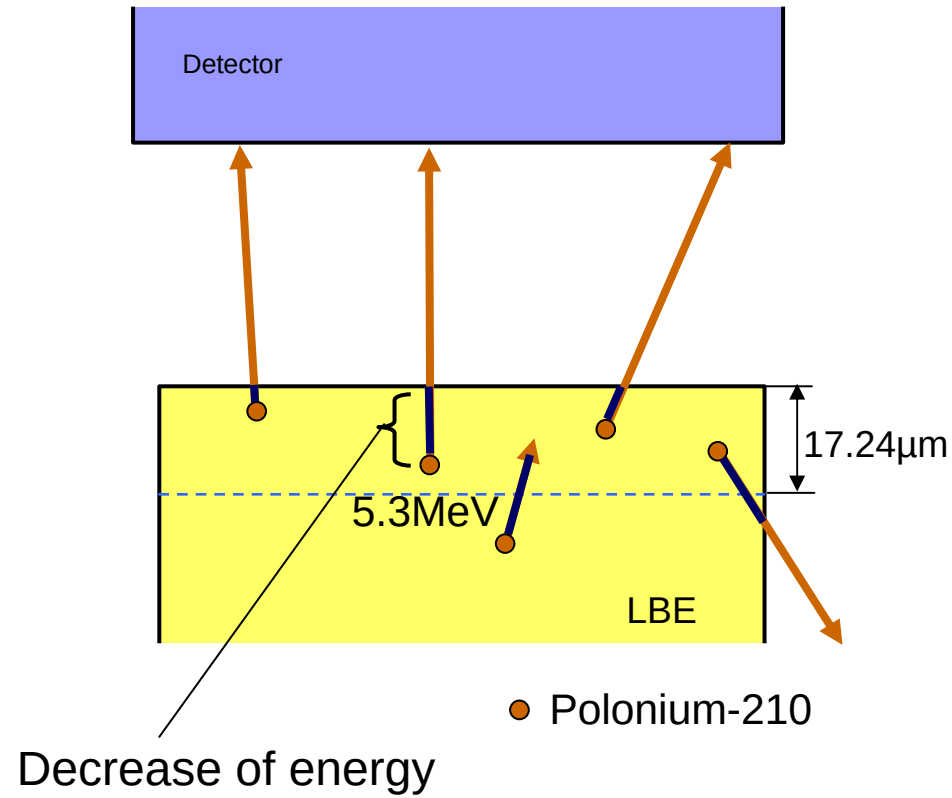
Discussion for baking experiment

- Polonium on stainless steel was released at 500ºC (0.4Pa). Vapor pressure of Pb-Po at 500ºC is 0.47Pa (Buongiorno, 2003). This fact suggests that polonium on the stainless steel was Pb-Po.
- Polonium on quartz glass plate was released at 300ºC (2Pa) . 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



Alpha-rays spectrum

Unfolding

Polonium distribution in lead-bismuth eutectic

Unfolding calculation

$$c_i = \sum_{j=1}^{n} A_{ij} x_j \qquad (i = 1, \ldots, m)$$

$$(j = 1, \ldots, n)$$

$$(m > n)$$

Constraint

$$x_j \geq 0$$

C_i : Counting rate of i'th bin in alpha-rays spectrum

A_{ij} : Response function for j'th layer

x_j : Number of alpha-rays sources at j'th layer

m : Number of energy bins of alpha-rays spectrum measurement

n : Number of layers of LBE column

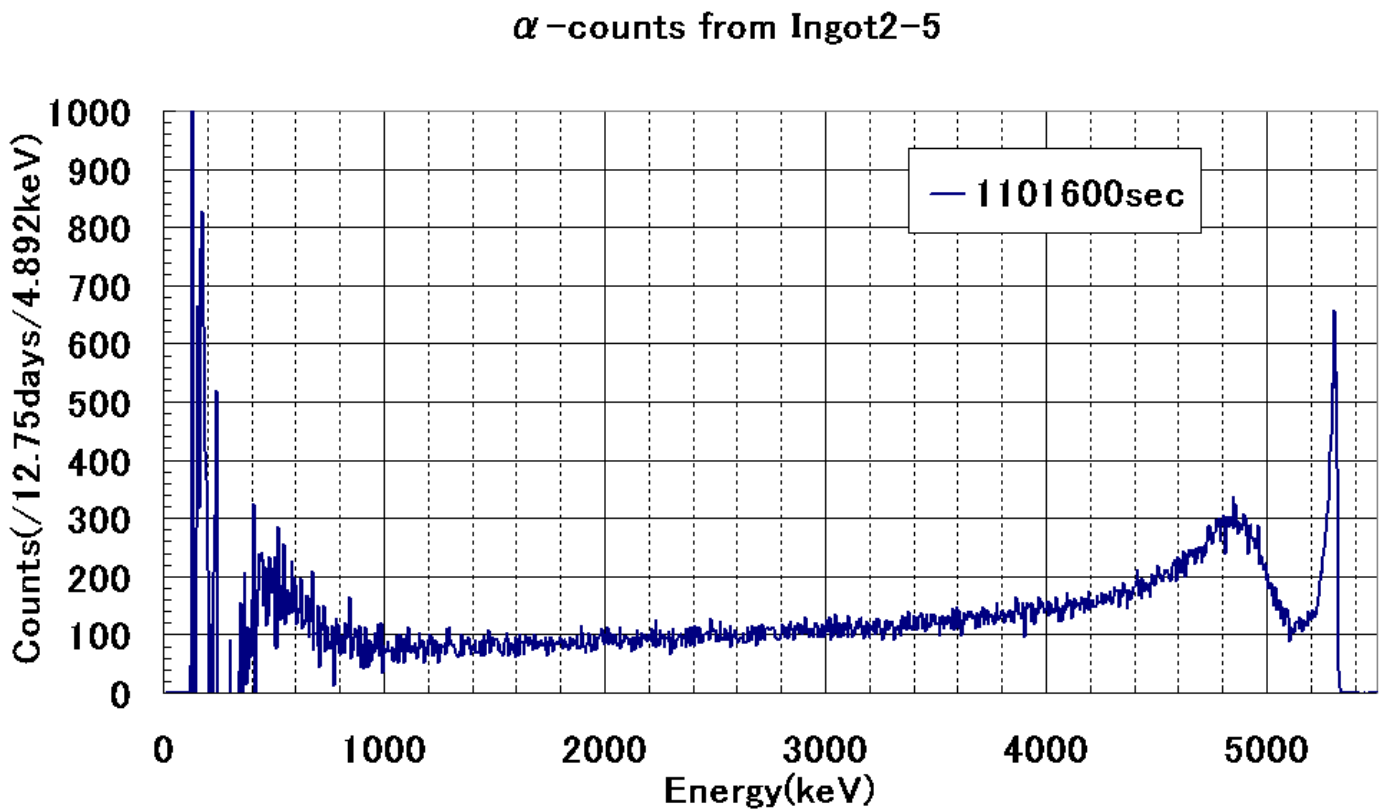
S_{ii} : Error of C_i

$$J = \left(\mathbf{Ax} - \mathbf{c}\right)^T \mathbf{S}^{-2} \left(\mathbf{Ax} - \mathbf{c}\right)$$

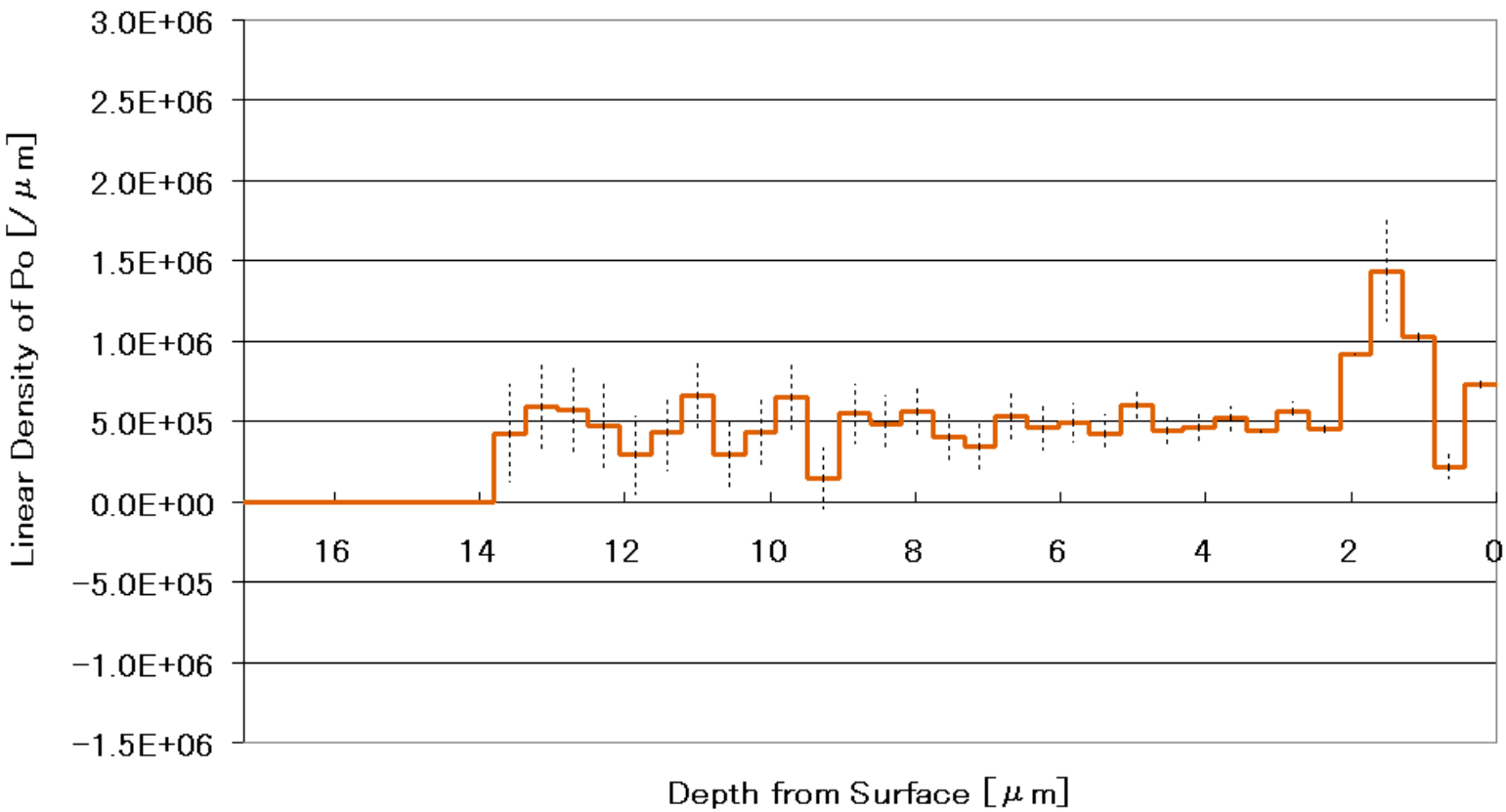
Optimum **x** is calculated to minimize *J.*

UFO/Q code, which was developed for neutron spectrometry, was used.

Example of measured alpha-ray spectrum



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 300ºC 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 500ºC 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.