

Characterization of ^{238}U , ^{232}Th , ^{40}K , and ^{226}Ra in NORM and Air Particulate Occurring from Phosphate Processing Facility

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1. Introduction

The naturally occurring radioactive materials (NORM) are widely spread throughout the environment, it occupies the largest part of public radioactivity exposure. According to report of the US National Academy of Sciences, 82% of the human radiation dose received annually comes from the natural radionuclides.

Radiation exposure doses from natural radionuclides is not significantly concerned level compared to those from artificial radionuclides which could be released into the environment by nuclear tests and power plant accident. It could make a serious risk problem to unspecific public when exposed to the natural radionuclides during long-term period at high activity concentration levels. Therefore, it is essential to make a professional and systematic approach for the proper handling and diminution.

Since concern regarding the natural occurring radioactivity has been growing over the last decade, 'Act on protective action guidelines against radiation in the natural environment' was implemented in 2012. The purpose of the legislation is to systematically manage the radiation exposure of public and worker related to the raw materials and by-products.

Airborne particulates is especially hazardous to the worker in the processing facility handling of NORM because they contains as well as toxic elements but also natural radioactivity from the ^{238}U , ^{232}Th , ^{40}K , ^{226}Ra , etc. Since it have been well-known that inhalation (as well as ingestion) cause much greater ionizing damage to the human tissue, the radiation exposure via inhalation of air particulates should be controlled and restrained.

In this study, radio-activities of ^{238}U , ^{232}Th , ^{40}K , and ^{226}Ra occurring in raw materials and by-products at phosphate processing facility were complementally determined using ED-XRF, γ -spectrometry and ICP-MS method. Although the degree of immersion in the human body varies depending on the particle size and density, the particle size is especially used as a key factor in assessing dose. Thus, size distribution characteristics of ^{238}U concentrations in airborne particulates collected at the phosphate processing facility were also evaluated.

2. Methods and Results

2.1 Apparatus and reagents

All of reagent was used with analytical grade. For the alkali-fusion of raw materials and by-product samples, lithium metaborate and lithium bromide (XRF scientific Limited) with automatic fusion machine (K2 Prime, Katanax, Canada) were used. For analysis of radioactivity, n-type HPGe gamma spectrometer (EG&G ORTEC, USA), ICP-MS (SPECTROMS, SPECTRO, Germany), and ED-XRF (XEPOS HE, SPECTRO, Germany) were complementally used.

Airborne particulates were collected by Non-viable Ambient Sizing Sampler (Anderson MARK III, USA). It consists of 8 stages ($>9\ \mu\text{m}$: 1 stage; $5.8\text{-}9\ \mu\text{m}$: 2 stage; $4.7\text{-}5.8\ \mu\text{m}$: 3 stage; $3.3\text{-}4.7\ \mu\text{m}$: 4 stage; $2.1\text{-}3.3\ \mu\text{m}$: 5 stage; $1.1\text{-}2.1\ \mu\text{m}$: 6 stage; $0.7\text{-}1.1\ \mu\text{m}$: 7 stage; and $0.4\text{-}0.7\ \mu\text{m}$: 8 stage)

2.2 Procedure

The analytical process of ^{238}U , ^{232}Th , ^{40}K , and ^{226}Ra in the raw material and by-product samples is shown in the Fig. 1. The solid samples were digested using alkali-fusion method and concentrated by iron hydroxide co-precipitation. The liquid sample such as phosphoric acid was directly concentrated by iron hydroxide co-precipitation. The co-precipitation was conducted by adding Fe-carrier and adjusting to the pH 7 with concentrated ammonia solution. After the precipitate was allowed to settle down by centrifuging, the supernatant was discarded and the precipitate was dissolved with 20 mL of 3 M HNO_3 .

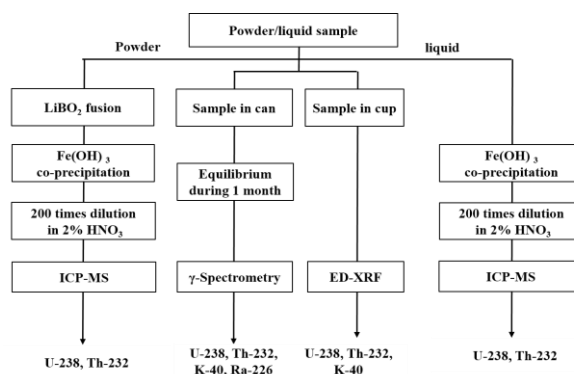


Fig. 1 Scheme of the analytical procedure of ^{238}U , ^{232}Th , ^{40}K , and ^{226}Ra

Finally, concentration of the uranium and thorium isotopes were measured by ICP-MS. For ED-XRF and γ -spectrometry, the samples regardless of physical form was directly measured after preparing in the specific containers. Contrary to ^{40}K nuclide which could be directly determined with the high gamma emission rate, indirect measurement is suitable for ^{238}U , ^{232}Th , and ^{226}Ra in γ -spectrometry due to their low emissions or peak interferences. Thus, supposed radioactive equilibrium, $^{234\text{m}}\text{Pa}$, ^{228}Ac , and ^{214}Bi were chosen as their indicators of ^{238}U , ^{232}Th , and ^{226}Ra , respectively.

The ^{238}U in the filter paper samples was extracted using hot plate refluxing procedure with 50 mL of aqua regia. Extraction was carried out at 180°C for 4 hours. Digested sample solution was filtered. Finally, solution was prepared for ICP-MS analysis after being dried and re-dissolved in 10 mL of 3 M HNO_3 .

3. Results and Discussions

3.1 ^{238}U , ^{232}Th , and ^{226}Ra activity in NORM samples

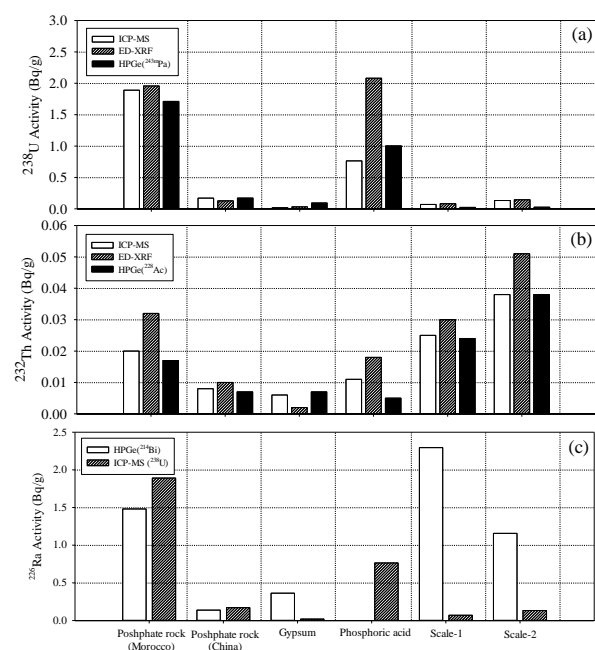


Fig. 2 ^{238}U , ^{232}Th , and ^{226}Ra activities in NORM samples of the phosphate rock process

The ^{238}U and ^{232}Th concentrations in the phosphate rocks (raw materials), phosphoric acid (product), gypsum and scales (by-products) were determined using three analytical methods (e.g., ICP-MS, ED-XRF, and γ -spectrometry). Based on the results of three methods, the activity concentrations of ^{238}U in the all of NORM samples were relatively higher than those of ^{232}Th nuclide. And the activity concentration of phosphate rock imported from the Morocco was exceeded over than regulation criteria of 1.0 Bq/g. Contrary to the phosphate rock samples, the radioactivity concentration

of ^{238}U (based on ICP-MS results) in the by-product samples were less than 0.1 Bq/g.

If ^{226}Ra and ^{238}U activity measured by γ -spectrometry and ICP-MS, respectively were compared, concentration ratios (γ -spectrometry/ICP-MS) for the phosphate rock samples were close to 1. However, those for scale, gypsum, and phosphoric acid samples were very different to raw materials. This difference was reflected the disequilibrium between ^{238}U and ^{226}Ra during the chemical reaction process. Because of hydrophilic properties of U, more than 90% of it was moved into the liquid phase of intermediate product. However, due to the ionic property of the Ra, almost of Ra has a tendency to concentrate in the phosphogypsum and scale.

3.2 ^{238}U concentrations of the airborne particulates

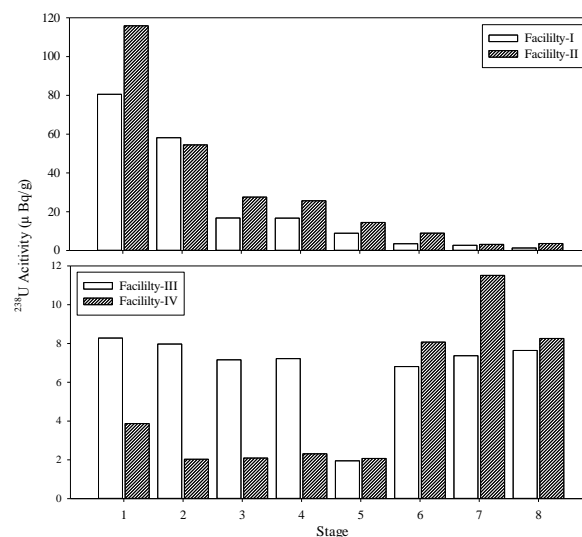


Fig. 3 ^{238}U activity in air particulates with particle sizes

In the Fig. 3, ^{238}U concentrations in size-fractioned airborne particulates collected at phosphate processing facility were presented. The concentration of ^{238}U in the all of size-fraction ranged from $1.29 \mu\text{Bq}/\text{m}^3$ to $115.89 \mu\text{Bq}/\text{m}^3$. The ^{238}U concentrations in the air particulates were highly varied with facility and process.

REFERENCES

- [1] International Atomic Energy Agency, Assessing the need for radiation protection measures in work involving minerals and raw materials, IAEA Technical Report Series No 49, Vienna, Austria, 2006.