## TOPICS Fukushima 26 Jan 2015 No. 58

## Radioactive cesium is adsorbed onto vermiculite and biotite. Mechanism of cesium adsorption has been clarified.

The research group of the Japan Atomic Energy Agency (JAEA) and collaborators have found that radioactive cesium tends to be adsorbed onto vermiculite (right photograph). Vermiculite is one of the clay minerals that commonly occurs in Fukushima Prefecture, and is well known as soil amendment material for gardening. It would be expected that the development of decontamination method based on these results contributes to the purification or volume reduction of contaminated soil in Fukushima.



The research group of Dr. Ryuhei Motokawa (Assistant Principal Researcher, JAEA), Dr. Tsuyoshi Yaita (Unit Manager, JAEA), Dr. Hitoshi Endo (Associate Professor, High Energy Accelerator Research Organization), Dr. Shingo Yokoyama (Principal Research Scientist, Central Research Institute of the Electric Power Industry), and Dr. Shotaro Nishitsuji (Assistant Professor, Graduate School of Science and Engineering, Yamagata University) has revealed that vermiculite adsorbs a large amount of cesium ions. They investigated the adsorption behavior of radioactive cesium and the structural changes in vermiculite clay with an increase in cesium concentration in dipping solution. As the result, they clarified that once one cesium ion adsorbed at a specific point of vermiculite clay interlayer, the cesium and other ions with similar chemical properties tend to occupy the neighboring site in the same interlayer space. Finally substantial cesium adsorbed collectively in the selective interlayer spaces. Furthermore, they elucidated the electrostatic interactions in the clay interlayer space in relation to the cesium adsorption. The direct bonding of substantial positive cesium with oxygen atoms of the clay sheet, locally compensates with the layer charge of the clay sheet. Thus, the attractive interaction between the clay sheets on the opposite interlayer is weakened. Consequently, the two clay layers are separated, and the other cesium ions could be potentially adsorbed onto the newly formed surface of the separated layer. Therefore, substantial cesium ions are adsorbed in the layer of vermiculite one after another like "Domino Toppling".

In order to investigate this mechanism at atomic/molecular scale, the research group employed a method called "Small Angle X-ray Scattering". In this method, a sample is irradiated by X-ray,

and the microscopic structure of the sample is determined by analyzing the intensity of the scattered X-ray as a function of the scattering angle. The spatial resolution of the measurements was between 0.1 and 100 nanometer (one billionth of 1 meter). As a result, it was found that the structure has changed as shown in the lower figure.

To date, there have been no reports where the structural change of clay minerals due to the cesium adsorption is clarified qualitatively. The present finding is expected to provide a useful insight into the environmental remediation of Fukushima Prefecture. The research group also succeeded in establishing a qualitative theoretical model to analyze the structure of clay minerals. It is expected that the model will contribute not only to nuclear energy field but also to the various fields of applied science, such as environmental science, analytical chemistry, materials science, and nanostructure science.



On the other hand, the research group of Dr. Toshihiro Kogure (Associate Professor, the University of Tokyo) and Dr. Tsuyoshi Yaita and their collaborators clarified that weathered biotite is one of the promising absorbents for radioactive cesium in real soil in Fukushina by using radiation recording medium, imaging plate (IP), and electron microscope (SEM/TEM) techniques.

The research group first collected soil contaminated with radioactive cesium at Iitate Village, Fukushima Prefecture. The soil particles were put on IP and the radiation from each particle was recorded. The right figures show the images observed by IP. The red to green color shows the strength of radioactivity, so we can understand the brief radioactive cesium concentration for each particle. On the basis of the



images, the particles including radioactive cesium were then transferred to the electron microscope, and the structures and chemical composition were investigated. As a result, they found the particles containing radioactive cesium into several types.

Electron microscope images of minute particles containing radioactivity (upper) and chemical composition of the minute particles determined by the X-ray emitted from the particles (below)



The upper figures show the SEM images of the three types of the particulates containing radioactive cesium and the lower figures display the characteristic X-ray spectra emitted from the particulates. From these results, the major chemical composition of each particulate was determined. For the three types of paticulates shown here, the left, middle, and right figures correspond to a weathered biotite, a composite particulate composed of inorganic and organic materials, and a composite particulates composed from various inorganic materials, respectively.

After slicing the particulates into thin sections, they further analyzed the detailed micro-structures inside of the particulates using electron microscope (TEM) with higher resolution. As a result, it became clear that a large amount of radioactive cesium would be immobilized on weathered biotite. In contrast to the previous laboratory scale experiments, it was found that cesium in real soil of Fukushima is not always concentrated in the surface or edge of mica particles but homogeneously distributed in this mineral. Actually, much weathered biotite occurred in Fukushima soil due to the long-time weathering of granite that commonly exists in the east part of Fukushima Prefecture. Thus, there is a high possibility that a large amount of radioactive cesium in soil of forests and rice fields are strongly trapped in this mineral.

As a next step, the research group is planning to specify the radioactive particles in soil of the other regions of Fukushima Prefecture. Using the collected samples of radioactive particles, they will continue investigating trapping states of cesium and its stability, and the possibility of decontamination by chemical treatment.

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