Investigation of Physical and Chemical Natures of Particulate Radioactive Matters Emitted from the Fukushima Nuclear Accident

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We have investigated particulate matters containing radionuclides emitted by the Fukushima Daiichi Nuclear Power Plant (FDNPP) Accident found in the environment. In this study, we summarize detailed physical and chemical natures of radioactive particles found by our investigations.
Radioactive particles were sampled from aerosols collected at two institutions in Tsukuba, the Meteorological Research Institute (MRI) and the National Institute of Advanced Industrial Science and Technology (AIST), in March 2011. The detailed sampling procedures were described elsewhere[1)]. We also sampled radioactive particles from a sediment of an outdoor pool and soils collected in the Fukushima Prefecture. They were subjected to a scanning electron microscopy with energy-dispersive X-ray spectroscopy and a gamma-ray spectrometry. The synchrotron radiation (SR) experiments were nondestructively carried out at BL37XU, SPring-8. The SR-X-ray microbeam was used as an analytical probe for chemical composition analysis by X-ray fluorescence spectroscopy (SR-μ-XRF), chemical state analysis by X-ray absorption near edge structure analysis (SR-μ-XANES) and crystal structure analysis by X-ray diffraction (SR-μ-XRD).
Analytical results suggest a presence of three groups (Groups A, B and C) of the radioactive particles having different physical and chemical natures as described below. Group A particles are almost spherical with diameters of 1~5 μm. They were first found in aerosols collected at MRI during March 14th and 15th[1)] 2011. In addition to radioactive Cs, various heavy elements (Rb, Zr, Sn, Ba etc.) are detected in common to Group A. These heavy elements could be derived from fission products of the nuclear fuel. The Group A particles sometimes contain U which might originate from the fuel[2)]. In addition to heavy elements from the fuel, they consist primarily of Si, Fe and Zn associated with construction materials of the reactor. They are insoluble in water and could have a long-term impact on the environment because they are Si-based glassy materials[2-4)]. It is highly possible that they were emitted from the reactor No.2 or 3 of the FDNPP. However it is difficult to assume a single process of preparation and emission for all Group A particles because their chemical composition is inhomogeneous.
Group B particles were found in soils of northwestern region of the FDNPP. It is pointed out that radionuclides emitted from the reactor No.1 fell on this region[3)]. In contrast to Group A, the Group B particles are large (>100 μm) and non-uniform shaped. While their matrix is the Si-based glass like Group A, there is a significant difference in chemical composition between Groups A and B. Several metal elements such as Fe, Mo, Sn and U were concentrated into micro region. Results of the SR-μ-XANES/XRD indicate the presence of non-glass phase in the concentrated region.
Group C particles were sampled from aerosols collected at AIST in March 30th 2011. Although size of these particles are similar to Group A, their shape is not spherical but rugged. In contrast to Groups A/B, Si is not a major component of the Group C particles. There is clear distinction in heavy elemental composition between Group C and Groups A/B. It is expected to that these differences in physical and chemical natures among three groups of radioactive particles were derived from the difference in process of preparation and emission. Therefore this study demonstrated the fact that particulate radioactive matters were emitted from the FDNPP into the environment several times through different processes.

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