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Synchrotron radiation X-ray analyses of the radioactive single airborne particle emitted by the Fukushima nuclear accide

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The Fukushima Daiichi nuclear power plant (FDNPP) accident released radioactive materials into the air environment over the entire Northern Hemisphere in March 2011. In order to elucidate environmental transfer of the radioactive materials from the FDNPP accident, a large number of studies have been carried out until today. However, we still do not know the exact physical and chemical properties of the radioactive materials. Such knowledge is necessary to construct the numerical models to estimate the geographical distributions and to evaluate the human exposures during and after the FDNPP accident. Therefore, we studied the radioactive materials which were released in the air environment by a FDNPP accident based on the multiple SR (synchrotron radiation) X-ray analyses of the single airborne particles with strong radioactivity trapped in Tsukuba, Ibaraki Prefecture at the time of the FDNPP accident. The samples were the radioactive single particles collected on quartz fiber filter at the Meteorological Research Institute, Tsukuba using a high-volume aerosol sampler on March 14-15. We selected the radioactive single particles out of this filter using micromanipulator and transferred to the KaptonR tape on an acrylic plate for SR X-ray analyses.

SR experiments were performed at the beam line BL37XU of SPring-8. The monochromatic SR X-ray beams were focused to about 1 μm (horizontal) x 1 μm (vertical) by K-B mirror. Two excitation X-ray energies were selected depending on the target elements for analysis: i.e., 15.0 keV (low-energy mode) and 37.5 keV (high-energy mode). SR X-ray fluorescence (XRF) imaging was applied to obtain elemental distribution, and X-ray absorption near edge structure (XANES) analysis was used for chemical state analysis, and X-ray powder diffraction (XRD) analysis was carried out to obtain crystal structural information of the particles.

We have successfully analyzed three radioactive single airborne particles. XRF analysis has revealed the existence of Cs in all of them. We were able to detect various elements shown below depending on the excitation energy. In addition, XRF imaging shows that each element exhibited uniform distribution in the particles.

High-energy mode: Cs, Ba, Te, Sn, Mo, Zr, Rb, Zn, Fe

Low-energy mode: Fe, Mn, Cr, Zn, Ti

Each particle showed different chemical compositions. XANES analysis of Sn,Mo,Zn,Fe in the particles showed that these metallic elements existed in high oxidation states in glass matrix. Furthermore, XRD analysis shows that the particle was amorphous because no diffraction line was observed. These results suggest that the detected elements are components of materials constituting the reactor and fission products. It is presumed that the reactor materials including the nuclear fuel melted at a high temperature and quenched by releasing to the air environment as a glassy materials.

Keywords: Fukushima Daiichi nuclear power plant, Synchrotron radiation X-ray analysis, airborne particle, strong radioactive particle