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Multiple synchrotron radiation X-ray analyses of radioactive microparticles emitted from the Fukushima Nuclear Accident

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The Fukushima Daiichi Nuclear Power Plant (FDNPP) accident occurred in 2011 is the largest nuclear incident since the 1986 Chernobyl disaster and has been rated at the maximum level of 7 on the International Nuclear Event Scale. Large amounts of radioactive materials were released into the environment during the accident. Although more than 4 years have passed since the accident, the radioactive materials emitted from the FDNPP have been detectable in the environment. However, little is known about the physical and chemical natures of radioactive materials released during the early stages of the accident. We previously found spherical aerosol microparticles (~2 μm, diameter) containing radioactive Cs in aerosol samples collected on March 14th and 15th, 2011, in Tsukuba[1)]. These radioactive microparticles have been commonly called "Cs-balls" . Synchrotron radiation (SR) X-ray microbeam analysis revealed the detailed chemical nature of the Cs-balls in a nondestructive manner, resulting in better understanding of what occurred in the plant during the early stages of the accident[2)]. SR-μ-X-ray fluorescence (XRF) analysis of the Cs-balls detected U as a nuclear fuel and various heavy elements derived from fission products (FPs) of the fuel. In addition, SR-μ-X-ray absorption near-edge structure (XANES) and SR-μ-X-ray diffraction (XRD) analysis revealed that the Cs-balls are glassy materials.

This study aims to demonstrate a hypothesis that water-insoluble radioactive materials similar to the Cs-ball had been extensively emitted to the environment from the FDNPP. We then focused on sediments in a swimming pool near FDNPP to find out radioactive microparticles. Total six spherical radioactive microparticles were separated from a sediment collected at an outdoor swimming pool located in Namie Town, Fukushima Prefecture, Japan. The SR experiments were carried out at BL37XU, a hard X-ray undulator beamline of SPring-8, Japan Synchrotron Radiation Research Institute (JASRI).

Gamma-ray spectra of the six particles detected both ^{134}Cs and ^{137}Cs in each particle with activity ratios of ~1 (decay corrected as of March 2011). It is thus confirmed that these particles are radioactive ones derived from the FDNPP accident. SR-μ-XRF analysis of the particles detected the following 13 heavy elements: Mn, Fe, Zn, Rb, Zr, Mo, Ag, Sn, Sb, Te, Cs, Ba and Pb. In addition, U was detected in one of the particles, which was further confirmed by U L_3-edge XANES analysis. We conclude that the U fuel, FPs, and components of the reactors are very likely the sources of the elements identified within the six radioactive microparticles, although further investigation will be needed to confirm their sources. We assume that, because these elements could have originated from multiple sources, they were melted together during the accident and eventually formed spherical microparticles. SR-μ-XANES spectra of Fe, Zn, Mo, and Sn K-edges for the individual particles revealed that they were present at high oxidation states, i.e., Fe^{3+}, Zn^{2+}, Mo^{6+}, and Sn^{4+} in the glass matrix, confirmed by SR-μ-X-ray diffraction analysis. These chemical natures of the six radioactive microparticles separated from the sediment collected in Fukushima Prefecture are in good agreement with those of the Cs-ball, the radioactive aerosol particle collected in Tsukuba after the FDNPP accident, revealed in our previous investigation[2)]. This study thus suggested a high possibility that water-insoluble glassy radioactive microparticles containing U fuel and FPs were emitted from the FDNPP and fell to a widespread area over the East Japan. In addition, we concluded that these radioactive microparticles in the glassy state may remain in the environment longer than those emitted as water-soluble aerosol particles containing the radioactive Cs.

1) K. Adachi et al.: *Scientific Reports* **3**, 2554 (2013)
2) Y. Abe et al.: *Analytical Chemistry* **87**, 8521-8525 (2014).

Keywords: Fukushima Daiichi Nuclear Power Plant accident, Radioactive material, Synchrotron radiation X-ray analysis, aerosol