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Characteristics of radiocesium-bearing microparticles and Type B particles using multiple synchrotron radiation X-ray analyses

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Introduction: A large amount of radiocesium was emitted into environment by the Fukushima Nuclear Power Plant (FDNPP) accident in March, 2011. Adachi et al. (2013) reported glassy water-insoluble microparticles including radiocesium, called as radiocesium-bearing microparticles (CsMPs). The CsMP is spherical with 1-3 μm in diameter and the radioactivity ranges from 0.5 to 4 Bq. It has been suggested that the CsMP was mainly emitted from Unit 2 or Unit 3 of FDNPP based on the Cs-134/Cs-137 activity ratio in the samples. In contrast, Ono et al. (2017) reported new particles called as Type B particles emitted from Unit 1. Type B particles are in various shapes and the size is 50-300 μm with radioactivity ranging from 30 to 100 Bq. These differences may represent the difference of generating process or condition of each unit in the plant. Previous studies have reported chemical properties of radioactive particles in detail but the number of particles reported is small. In this study, we tried to understand radioactive particles systematically by analyzing a lot of particles separated using wet separation method.
Method: In this study, we collected 53 Type B particles and 13 CsMPs from road dusts, non-woven fabric cloths from Fukushima, and aerosol filters from Kanagawa Prefecture by a wet separation method. After measurement of radioactivity with a high-purity germanium semiconductor detector, scanning electron microscope and energy dispersive X-ray spectroscopy analyses were performed to confirm that separated particles were CsMPs or Type B particles. We investigated inner structure and calculated the volume and porosity of Type B particles by X-ray μ-computed tomography (CT). We determined Rb/Sr ratio by X-ray fluorescence (XRF) analysis. Redox condition of each unit was investigated by X-ray adsorption near edge structure (XANES) analysis for Uranium in particles.
Result: CT combined with XRF analysis showed the presence of many voids and iron particles in Type B particles. In addition, Cs-137 concentration of CsMPs were $\sim$10000 times higher than that of Type B particles, which suggests that Type B particles were formed by fuel melt. In contrast, CsMPs were formed by gas. Among Type B particles, spherical particles had higher Cs-137 concentration than non-spherical particles. Type B particles with larger porosity had higher Cs-137 radioactivity because of capturing a lot of volatile elements such as Cs and Rb within the particles. Moreover, four spherical particles had inclusions in their voids which are considered to be formed by rapid cooling of gaseous materials. XANES analysis showed the presence of U(IV) in a Type B particle, whereas U(VI) in other Type B particles and a CsMP. These results suggest that Type B particles and CsMPs are totally different in forming process and they have information of condition in Units.