Comparison of radiocesium-bearing microparticles of different shape and elemental distribution by multiple synchrotoron radiation X-ray analyses

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Introduction: Adachi et al. (2013) reported glassy water-insoluble microparticles including radiocesium (Type-A particles) emitted by the Fukushima Dai-ichi Nuclear Power Plant (FDNPP) accident. It has been suggested that Type-A paritcle was mainly emitted from Unit 2 or 3 of FDNPP based on the $^{134}Cs/^{137}Cs$ activity ratio. In contrast, Ono et al. (2017) reported new radioactive particles (Type-B particles) emitted from Unit 1. In this study, we tried to understand these radioactive particles systematically by analyzing a large number of particles separated using a wet separation method (Miura et al., 2018).

Method: In this study, we collected 10 Type-A and 57 Type-B particles by the wet separation method. We investigated inner structure and calculated the volume and porosity of Type-B particles by X-ray μ-computed tomography (CT). We also determined rubidium (Rb) / strontium (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 (U) in particles.

Results and discussion: CT combined with XRF analysis showed the presence of many voids and iron particles in Type-B particles. In addition, specific ^{137}Cs concentration of Type-A particle was ~10000 times higher than that of Type-B particle. Among Type-B particles, spherical Type-B particles had higher ^{137}Cs concentration than non-spherical Type-B particles. Type-B particles with larger porosity had higher ^{137}Cs radioactivity because of capturing various volatile elements such as Cs and Rb within the particles. XANES analysis showed the presence of U(IV) in a Type-B particle, whereas U(VI) in other Type-B particles and a Type-A particle. These results suggest that Type-A and -B particles are totally different in terms of their formation process and phisico-chemical condition during the formation.