

Discovering U particle with Zr from CsMP emitted from Unit 1 of FDNPP using synchrotron radiation X-ray analyses

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Introduction: A large amount of radiocesium (Cs) 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 (Type-A particles). Based on the ¹³⁴Cs/¹³⁷Cs ratio calculated by Nishihara et al. (2012), Type-A particle is thought to be emitted from Unit 2 or 3 of FDNPP (Higaki et al., 2017; Miura et al., 2018). Abe et al. (2014) detected uranium (U) in Type-A particle by synchrotron radiation measurement. In addition, Ochiai et al. (2018) reported that Type-A particle has UO₂ with zirconium (Zr). In contrast, Ono et al. (2017) reported new particles called as Type-B particles emitted from Unit 1. Type-B particle is different from Type-A particle in size, shape, Cs concentration and so on. We measured some Type-A and Type-B particles to clarify chemical condition of U by synchrotron radiation measurement. Difference in chemical condition of U in each particle may represent the difference of generating process or condition of each reactor unit at the accident.

Method: In this study, we collected Type-A and B particles from road dusts and non-woven fabric cloths from Fukushima 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 Type-A or Type-B particles. X-ray fluorescence (XRF) mapping and X-ray adsorption near edge structure (XANES) measurement were performed at BL37XU (SPring-8, Hyogo). Bent Crystal Laue Analyzer (BCLA) was used to detect U peaks, because energy of U L α emission is close to energies of rubidium K α and strontium K α emissions.

Results: We could detect U in Type-A and B particles using BCLA which decreased background contribution about one tenth. U-enriched area in the Type-B particle was about several microns, which is much bigger than U nano-particle in Type-A particle reported by Ochiai et al. (2018). This difference in size possibly suggested that U particle in Type-B particle was derived from melt in contrast to that in Type-A particle possibly produced from vapor. XANES measurement showed the presence of U(IV) in Type-B particle but also U(VI) in Type-A and B particles, which suggested U(VI)O₂ fuel was oxidized possibly due to its exposure to air at the surface of the particles. In the spots of detecting U, we also detected Zr which is used for fuel cladding tube. It is needed to identify chemical-states of Zr and U as a future study.

Keywords: FDNPP accident, Cesium-bearing microparticle, Synchrotron radiation X-ray analysis