

## Overview of insoluble radioactive cesium particles emitted from the Fukushima Dai-ichi Nuclear Power Station

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In the early stage of the Fukushima Dai-ichi Nuclear Power Station (F1NPS) accident, number of spot type contamination has been observed in computed autoradiography (Kashimura 2013, Shibata 2013, Satou 2014). It's means presence of radioactive particles, however, insoluble cesium particle was overlooked because cesium, which is dominant radioactive element in the accident, becomes ionized in the environment. Adachi et al. (2013) showed presence of cesium (Cs)-bearing particles within air dust sample collected at Tsukuba, 170 km south from the Fukushima site, in midnight of 14 to morning of 15 March 2011. These particles were micrometer order small particles and Cs was could be detectable as element using an energy dispersive X-ray spectroscopy (EDX). However, other radioactive elements such as Co-60, Ru-103 and uranium, which were dominant element of radioactive particles delivered from Chernobyl accident, could not detected.

Abe et al. (2014) employed a synchrotron radiation (SR)-micro( $\mu$ )-X-ray analysis to the Cs-bearing particles, and they were concluded that (1) contained elements derived from nuclear fission processes and from nuclear reactor and fuel materials; (2) were amorphous; (3) were highly oxidized; and (4) consisted of glassy spherules formed from a molten mixture of nuclear fuel and reactor material. In addition, Satou et al. (2016) and Yamaguchi et al. (2016) disclosed that silicate is main component of Cs-bearing particles.

Satou et al. (2015) discovered two types of radioactive particles from soil samples collected in the vicinity of the F1NPS. These particles were remained in the natural environment more than four years, silicate is main component in common of each group particles. Group A particles were very similar to Cs-bearing particles reported by Adachi et al. except particle shape. On the other hand, group B is big particles found in north area from the F1NPS, and the strongest particles contained 20 kBq of Cs-137 within a particle. Radioactive ratio of Cs-134/Cs-137 of group A and B is completely different. Group B particles shown 0.92 (mean value) of Cs ratio, and specific radioactivity are much lowers than group A particles. In contrast, activity ratio in group A particles shown 1.0 (mean value), and it was consistent with previous studies by Adachi (2013). The location of soil samples, which was containing group B particles, has been contaminated with radioactive materials from Unit 1 with hydrogen explosion on 12 March (Satou et al. 2014, Chino et al. 2016). More than 300  $\mu$ m of diameter particles has been transported from the Unit 1 of F1NPS. This result shown that the insoluble radioactive cesium particles are emitted from not only Units 2 and/or 3 on 15 March but also Unit 1 on 12 March.

The insoluble radioactive Cs particles were spread widely, and it is require to evaluation for particulate percentage of contribution in total emitted radioactive cesium, and long term monitoring of these behaviors.