Isotopic signature and nano-scale texture of cesium-rich micro-particles: Release of uranium and fission products from the Fukushima Daiichi Nuclear Power Plant

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Highly radioactive cesium-rich microparticles (CsMPs) released from the Fukushima Daiichi Nuclear Power Plant (FDNPP) are nano-scale chemical fingerprints of the 2011 tragedy. U, Cs, Ba, Rb, K, and Ca isotopic ratios were determined on three CsMPs (35–780 Bq) collected within ~10 km from the FDNPP to determine the CsMPs' origin and mechanism of formation. Apart from crystalline Fe-pollucite, $CsFeSi_2O_6 \cdot nH_2O$, CsMPs are comprised mainly of Zn–Fe-oxide nanoparticles in a SiO_2 glass matrix (up to ~30 w% of Cs and ~1 wt% of U mainly associated with Zn–Fe-oxide). The $^{235}U/^{238}U$ values in two CsMPs; 0.030 (±0.005) and 0.029 (±0.003), are consistent with that of enriched nuclear fuel; but with a higher than average burnup estimated by a ORIGEN code and lower than non-irradiated fuel suggesting non-uniform volatilization of U from melted fuels with different levels of burnup, followed by sorption onto Zn–Fe-oxides. The nano-scale texture and isotopic analyses provide a partial record of the chemical reactions that occurred in the fuel during meltdown. Also, the CsMPs are identified as an important transportation medium for the release of radionuclides as a respirable form.