

## Speciation of Uranium Released to the Environment with Cesium-Rich Microparticles from the Fukushima Daiichi Nuclear Power Plant

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Trace U was released from the Fukushima Daiichi Nuclear Power Plant (FDNPP) during the meltdowns. The speciation of U records the meltdown processes and property of debris, and also determines the environmental impacts. This study summarizes various types of U derived from the FDNPP focusing on the speciation to provide the updated knowledge on the behaviour of nuclear fuels during the meltdowns at the FDNPP.

At least three types of U occurrence have been identified at the present; *(i)* trace amounts of U associated with Fe–Zn-oxide nanoparticles embedded in SiO<sub>2</sub> matrix within CsMP; *(ii)* uraninite nanocrystals without detectable impurities, which were embedded within the Fe-oxide; and *(iii)* eutectic U–Zr-oxide nanoparticles. All types of U were found along with cesium-rich microparticles.

The type *(i)* particles may be formed by volatilization of the partially oxidized fuels with low burnups and the volatilized U adsorbed onto Fe–Zn-oxide nanoparticles based on the <sup>235</sup>U/<sup>238</sup>U isotope ratio greater than the average ratio calculated from the burnup.

Uraninite nanoparticles of type *(ii)* exhibited euhedral octahedral shape and revealed epitaxial growth of the encapsulating magnetite. The solidification of uraninite nanocrystals would first proceed within the Fe oxides melts as its cools, followed by the epitaxial growth of magnetite over uraninite nanocrystals.

The U–Zr-oxide nanoparticles of type *(iii)* likely formed from a U–Zr-oxide melt, which would have resulted from the interaction of molten fuel and ZrO<sub>2</sub> that had previously been produced through the reaction of zircalloy and steam at high temperature > ~1500 K. The cubic structure indicates rapid cooling of the liquid U–Zr-oxides droplets.