高濃度放射性セシウム含有微粒子（Cesium-rich microparticle, CsMP）の本質的解明に基づく炉内の情報と環境影響
(5) 福島第一原発から CsMP とともに環境中に放出されたウランの化学状態

Radioactive cesium-rich microparticle (CsMP): A window into the damaged reactors and its environmental impacts

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

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我々は初めて福島原発から放出されたウラン酸化物の原子スケール解析の結果を報告した。本発表では高濃度放射性セシウム含有微粒子(CsMP)に付随して環境中に放出されたウランの化学状態についてこれまでに発表してきた知見を総括し、原発内部に残されているウラン酸化物の形成過程、性状把握に寄与する。

キーワード：福島第一原子力発電所，ウラン酸化物、放射性セシウム含有微粒子

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_2 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 $^{235}U/^{238}U$ 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_2 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.

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