Dynamic phenomena recorded in geochemical and environmental materials

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Chemically durable materials that occur in nature give novel opportunities to investigate dynamics of elements of interest at the time of formation. The chemical reactions recorded in the materials are typically at sub-micron scale, and thus, the microanalytical techniques frequently play a key role for the investigation. In this presentation, two examples, which demonstrate the usefulness of microanalysis for the better understanding the novel phenomena of elemental dynamics, will be reviewed; lead in the Archean zircon at the atomic-scale and radionuclides in the cesium-rich microparticles derived from Fukushima Daiichi Nuclear Power Plant.

We reported, for the first time, a direct, atomic-scale characterization of Pb in zircon (4.4 –3.1 Ga) from the early Archean Yilgarn craton in Australia using high-resolution HAADF-STEM. Two forms of Pb have been identified: Pb concentrated at ~3 atom% as a nanoscale patch in zircon structure, and Pb concentrated within the amorphous domain created by fission fragment damage. The first result suggests that the Pb atoms directly substitute for Zr^{4+} in the zircon structure, and the latter observation demonstrates that Pb diffusion can occur through amorphous regions created by radiation damage, although volume diffusion is typically considered to be the dominant mechanism for Pb diffusion. Beyond the first percolation point, i.e., when the amorphous domains overlap and form a fully interconnected network of amorphous domains, there is a new pathway for the diffusion of Pb that is faster than volume diffusion through crystalline zircon.

The nuclear disaster at the Fukushima Daiichi Nuclear Power Plant (FDNPP) in March 2011 caused partial meltdowns of three reactors. During the meltdowns, a type of condensed particle, a caesium-rich micro-particle (CsMP), formed inside the reactors via unknown processes. Here we report the chemical and physical processes of CsMP formation inside the reactors during the meltdowns based on atomicresolution electron microscopy of CsMPs discovered near the FDNPP. All of the CsMPs (with sizes of 2.0–3.4 μm) comprise SiO_2 glass matrices and ~10-nm-sized Zn–Fe-oxide nanoparticles associated with a wide range of Cs concentrations (1.1–19 wt% Cs as Cs_2O). Trace amounts of U are also associated with the Zn–Fe oxides. The nano-texture in the CsMPs records multiple reaction-process steps during meltdown in the severe FDNPP accident: Melted fuel (molten core)-concrete interactions (MCCIs), incorporating various airborne fission product nanoparticles, including CsOH and CsCl, proceeded via SiO_2 condensation over aggregates of Zn-Fe oxide nanoparticles originating from the failure of the reactor pressure vessels. Still, CsMPs provide a mechanism by which volatile and low-volatility radionuclides such as U can reach the environment and should be considered in the migration model of Cs and radionuclides in the current environment surrounding the FDNPP.

References

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