Dynamics of radioactive microparticles released from the Fukushima nuclear plant: heating and dissolution characteristics

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A significant amount of radiocesium was released due to the Fukushima Daiichi Nuclear Power Plant accident in March 2011, causing radioactive contamination in east Japan. Most of the radiocesium is believed to have been released in a gaseous state, descended with rainwater and sorbed mainly by clay minerals in soil. On the other hand, spherical microparticles substantially consisting of silicate glass (radiocesium-bearing microparticle, CsMP) were also released from the damaged reactors. CsMPs are generally spherules of a few microns in diameter and contain Cl, K, Fe, Zn, Mn, Rb, Sn, and Cs in silicate glass. Since CsMPs have very high radioactivity per particle compared with soil particles sorbing radiocesium, the influence of radiation on their vicinity is a concern. However, the knowledge about dynamics of CsMPs in the environment is still limited and required urgently. Here, we report heating and dissolution characteristics of CsMPs.

Incineration of radioactive waste including CsMPs is under consideration for volume reduction. Therefore, we collected CsMPs and heated in the air in order to investigate their thermal behavior. As a result of heating treatment, the radioactivity of CsMPs gradually decreased from 600 ˚C and almost disappeared when heated up to 1000 ˚C. Scanning electron microscope (SEM) examination revealed that the morphology of CsMPs did not change even after heating at 1000 ˚C, but Cl and alkaline elements such as Cs and K were diffused away from CsMPs. The internal structure of CsMPs after heating was also investigated using transmission electron microscopy (TEM). Fe, Zn and Sn originally dissolved in the glass matrix were crystallized to oxide particles inside the CsMPs. Furthermore, when CsMPs were heated together with weathered granite soil collected in Fukushima, the radiocesium released from CsMPs was adsorbed by soil particles. These results of heating experiments suggested that highly radioactive particles such as CsMPs do not remain in the waste incinerated at a sufficiently high temperature.

Although CsMPs have been reported as “insoluble” particles thus far, they should be gradually dissolved in the environment because they substantially consist of silicate glass. We conducted dissolution experiments using CsMPs collected from the environment and investigated the dissolution behavior and lifetime of CsMPs. CsMPs were incubated in three kind of solutions, namely pure water, artificial seawater, and hydrochloric acid, under various temperature conditions, and the dissolution rate of CsMPs was estimated from the decrease in radioactivity. Furthermore, the activation energy for dissolution was calculated from the Arrhenius plot, resulting in 65 and 88 kJ/mol for pure water (pH is ~5.2 due to absorption of carbon dioxide in the atmosphere) and seawater, respectively. The calculated dissolution rate at 13 ˚C (approximate average temperature in Fukushima) is ~10 times larger in seawater than in pure water, and CsMPs are expected to be completely dissolved in seawater within ~10 years. This suggests that most of the CsMPs that fell into the sea immediately after the accident have already been disappeared. SEM observation of the CsMPs before and after dissolution in pure water revealed that their size was decreased and their spherical shapes were considerably altered without maintaining geometry. In the case of the CsMPs dissolved in seawater, plate-like secondary minerals rich in Fe and Mg were precipitated around the CsMPs to form a crust, and glass matrix was dissolved inside the crust. In addition, when CsMPs were incubated in hydrochloric acid with pH adjusted to 3, the radioactivity decreased considerably at the beginning, but ceased to change with time. A layer mostly consisting of SiO_2 was formed on the surface of the CsMPs, which may become a passive layer inhibiting dissolution.

Keywords: microparticle, silicate glass, cesium, thermal characteristics, dissolution, Fukushima nuclear plant accident

