1-4 Thermodynamic Estimation of Fuel Debris Characteristics

— Thermodynamic Evaluation of High-Temperature-Reaction Products between Molten Core and Concrete —

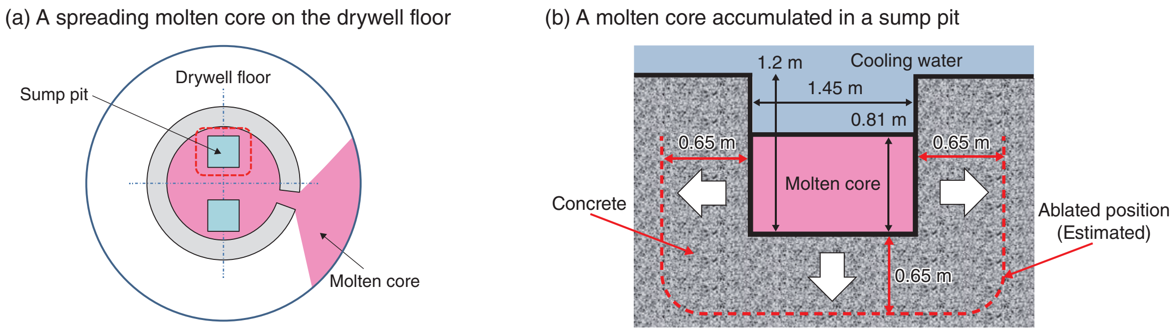

Fig.1-7 Image of a spreading molten core on the drywell floor of a containment vessel
(a) The molten core spreading on the concrete floor of the containment vessel melts and ablates concretes through its heat.
(b) Its temperature decreases with the mixture of concrete components and concrete ablation, then finishes.

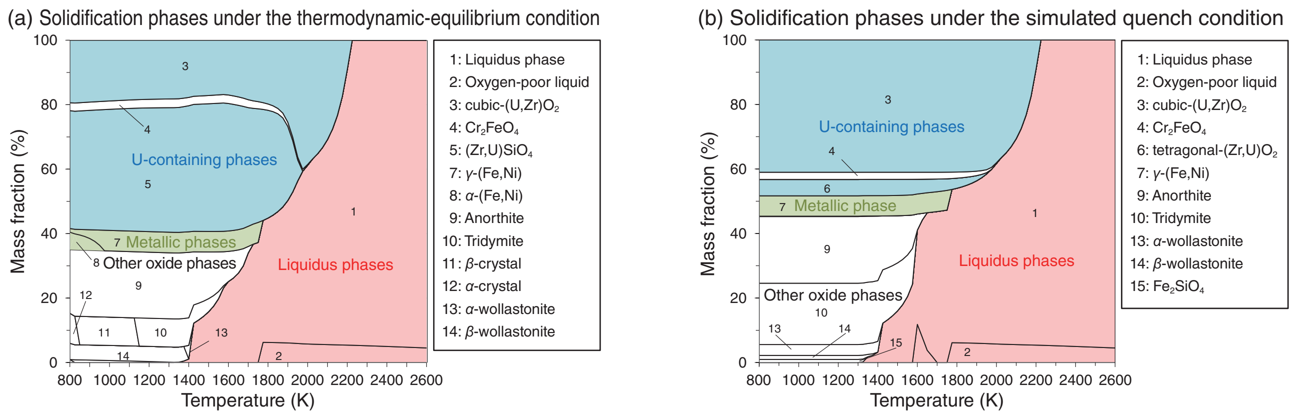

Fig.1-8 Thermodynamically estimated solidification phases of mixed molten-core–concrete material
(a) The oxide phases containing uranium (U) and zirconium (Zr), which have high liquidus temperatures, solidify during the cooling process of the liquidus phases composed of molten core and concrete. Metallic and oxide phases composed of concrete components then solidify. (b) The formation of a tetragonal solid solution of U and Zr in the place of zirconium silicate was predicted.

The TEPCO estimated that molten-core material penetrated the bottom of the pressure vessel and spread on the concrete floor in the containment vessel during the Fukushima Daiichi NPS (1F) accident, as shown in Fig.1-7. Core material at temperatures above 2000 °C melts and ablates the concrete. The liquidus temperature of the molten core decreased and the ratio of solid phases increased due to mixture of concrete components, and concrete ablation is then completed. The solidified material is called fuel debris and consists of complex materials composed of uranium (U) dioxide fuel, structural materials such as iron and zirconium (Zr), and concrete components. Estimation of the chemical phases and properties of fuel debris is important for fuel debris removal. We have evaluated phases using the experimental and computational approaches. This paper shows the part of our results obtained by the computational approach.

In this study, FactSage6.4 was used as thermodynamic-equilibrium software and NUCLEA was used as a thermodynamic database. The initial condition was decided with analytical results for concrete picked up from the 1F building and simulation results of the 1F-accident progression.

Under the thermodynamic-equilibrium condition, the solidification phases in regions under long-term cooling, such as the inside of molten materials, can be estimated. In this condition, U is expected to form a cubic solid solution with Zr ($(U,Zr)O_2$) and zirconium silicate ($(Zr,U)SiO_4$), as shown in Fig.1-8(a). The formation of a metallic phase made of stainless steel and oxide phases composed of concrete components were also estimated. It seems that $(U,Zr)O_2$ and $(Zr,U)SiO_4$ solidify in a low-liquidus-temperature oxide liquid composed of concrete components during the cooling process.

On the contrary, in regions under the quench condition simulated with the Scheil model, such as near the boundary of the cooling water, we estimate that most of the U solidifies into cubic $(U,Zr)O_2$ and tetragonal Zr rich solid solution ($(Zr,U)O_2$) forms, as shown in Fig.1-8(b).

In conclusion, it was estimated that main phases of fuel debris are $(U,Zr)O_2$ and $(Zr,U)SiO_4$ and that the formation behavior depends on the cooling conditions. These results, together with the actual fuel debris characteristics, will be useful for fuel debris removal and clarification of the progression of the 1F accident. We will estimate the fuel debris characteristics in more detail using experimental data and computational prediction.

Reference

Kitagaki, T. et al., Thermodynamic Evaluation of the Solidification Phase of Molten Core-Concrete under Estimated Fukushima Daiichi Nuclear Power Plant Accident Conditions, Journal of Nuclear Materials, vol.486, 2017, p.206-215.