Partitioning of Cs and Mo in steam environment

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Abstract The effect of steam on the partitioning of Cs and Mo was calculated at temperature ranging from 1200-2600 K was calculated at total pressures of 75 and 3.5 bar, which correspond to those before and after RPV depressurization. The reaction of $\text{Cs}_2\text{MoO}_4 (g)$ with steam was better described by taking into account the participation of $\text{Cs}_2\text{Mo}_2\text{O}_7$.

Keywords Severe accident, CsOH, $\text{Cs}_2\text{MoO}_2\text{MoO}_3$, thermodynamics

Introduction

Cesium is one of the key fission products in the consequence analysis of severe accident of the Light Water Reactor. Traditionally, cesium has been considered to form mainly CsOH, while some Cs forms CsI to an extent corresponding to the iodine inventory. Recently, however, the importance of cesium molybdate, $\text{Cs}_2\text{MoO}_4$, has been recognized in view of the Phebus test results.

Thermodynamic analysis

The thermodynamic calculation were taken from ref. [2] and JANAF database. An example, Fukushima Daiichi Unit-1 (1F) contained 1140 mol-Cs/core (154kg-Cs/core). Molybdenum inventory was about 1850 mol/core (180kg-Mo/core). Therefore, there is enough Mo to form $\text{Cs}_2\text{MoO}_4$ as long as the inventory is concerned. In a MELCOR analysis [5], there was $2 \times 10^4$ mol (~ 40,000 kg) of steam generated in the core region. Then, the average molar flow ratio of $\text{H}_2\text{O}(g)/\text{Cs}$ was about 2,000. The molar flow ratio $\text{H}_2(g)/\text{H}_2\text{O}(g)$ would typically range from 1/100 to 1 due to the analysis. Referring to the MELCOR time-to-failure model, $T$ (fuel) < 2600 K was assumed in the calculation. The calculations were done at the total pressure of 75 bar and 3.5 bar, which represent the RPV pressure before and after the depressurization. Free energy minimizer ChemSage [6] was used for the calculation. Iodine was excluded in this analysis in order to focus on the equilibrium in the Cs-Mo-H system.

Results

Fig. 1 shows the partial pressure of different species at in the total pressures at 75 bar and 3.5 bar. The solid and dash lines correspond to the molar ratio of $\text{H}_2\text{O}/\text{H}_2$ at 1 and 1/100, respectively. The relative importance of $\text{Cs}_2\text{MoO}_4$ and CsOH is not very sensitive to the $\text{H}_2/\text{H}_2\text{O}$ ratio. On the other hand, the total pressure has a significant effect: CsOH$(g)$ becomes more predominant at lower temperature regions with increasing pressure. Although Mo is preferentially partitioned to $\text{H}_2\text{MoO}_4$ at the damaged core region, it will be transferred to cesium molybdates as the gas phase cools. Importance of Cs$(g)$ decreases rapidly, but that of $\text{Cs}_2\text{Mo}_2\text{O}_7(g)$ increases as the gas cools. The deposition of molybdates starts at ~1900K at 75 bar and ~1550K at 3.5 bar.

References