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 Cs_2MoO_4 (*g*) with steam was better described by taking into account the participation of $Cs_2Mo_2O_7$.

Keywords Severe accident, CsOH, Cs_2MoO_4-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, Cs_2MoO_4, has been recognized in view of the Phebus test results [1].

During the whole course of the severe accident of BWR, the oxygen potential, $p(O_2)$ should be high enough for stabilization of Cs_2MoO_4 in the steam dome region. However, the effect of steam on the partitioning of Cs_2MoO_4 and CsOH is not yet clear. In the equilibria, higher molybdates of cesium such as $Cs_2Mo_2O_7$ play some role as we can postulate the reaction: $2Cs_2MoO_4(g) + H_2O(g) = 2CsOH(g) + Cs_2Mo_2O_7(g)$. Therefore, we performed detailed analyses of Cs-Mo-O-H system at high temperatures. We have constructed a thermodynamic model of Cs_2MoO_4-MoO_3 pseudo-binary system [2]. Also we have studied the vaporization and deposition of $Cs_2Mo_2O_7$ in both dry and humid atmosphere [3, 4]. Those studies form the basis to study the partitioning of Cs and Mo in the accident conditions.

Thermodynamic analysis

The thermodynamic calculation were taken from ref. [2] and JANAF database. As 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 Cs_2MoO_4 as long as the inventory is concerned. In a MELCOR analysis [5], there was 2×10^6 mol (~ 40,000 kg) of steam generated in the core region. Then, the average molar flow ratio of $H_2O(g)$/Cs was about 2,000. The molar flow ratio $H_2(g)/H_2O(g)$ 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 equilibria in the Cs-Mo-O-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 H_2/H_2O at 1 and 1/100, respectively. The relative importance of Cs_2MoO_4 and CsOH is not very sensitive to the H_2/H_2O 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 also to H_2MoO_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 $Cs_2Mo_2O_7(g)$ increases as the gas cools. The deposition of molybdates starts at ~1900K at 75bar and ~1550K at 3.5 bar.

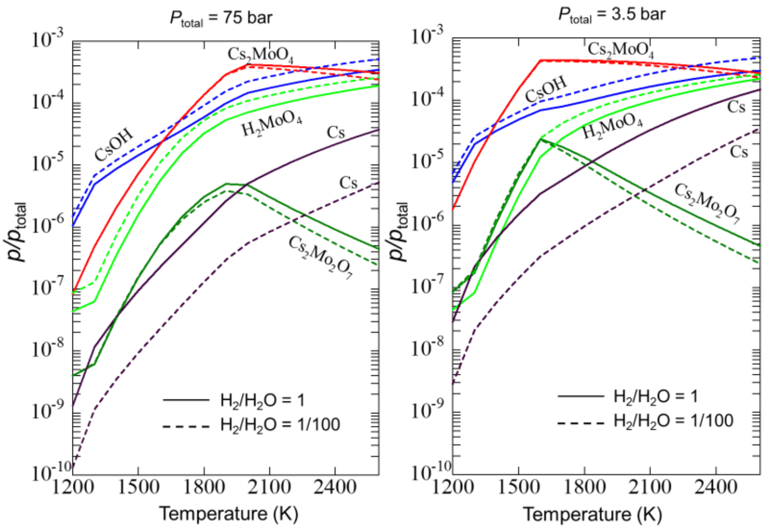

Figure 1. Partial pressure of species in core region with different molar ratio H_2/H_2O

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