Thermochemical investigation of calcium silicate insulation with cesium hydroxide

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Abstract

Experimental study using thermogravimetry and X-ray diffractometry was conducted to investigate chemical interaction between calcium silicate insulation and cesium hydroxide under steam and hydrogen atmospheres. The bulk use of calcium silicate insulation in nuclear power station is of concern when severe accident occurs that such interaction will be highly encountered. Our results suggested that chemical interaction occurred in temperature range of 575-730°C regardless of atmospheres and yielded formation of new compound, $CsAlSiO_4$.

Keywords: Cesium, Calcium silicate, Insulation, Thermochemistry

1. Introduction

Report of internal investigation in primary containment vessel (PCV) of Fukushima Daiichi Nuclear Power Station (1F) unit 2 by TEPCO indicated unexpected dose rate distribution of the lower 10 Gy/h at the pedestal, where most of debris should be existed, and the higher 70-80 Gy/h at the pedestal vicinity [1]. Deposits found in this vicinity were suspected as source of the high dose rate. Based on condition and location where these deposits existed, it is likely that formation of such deposits was originated by steam containing fission products such as Cs, leaked from, e.g., safety relief valves gasket. We further presume that the leakage steam directly hit the main steam piping thermal insulation (i.e. calcium silicate insulation or calsil), following that the adsorption of Cs on the calsil occurred. In this study we investigated interaction between the calsil and Cs under various atmospheres.

2. Experimental method

A calsil block, which is equivalent in quality to the thermal insulation used in 1F, was pulverized and mixed with cesium hydroxide monohydrate, $CsOH•H_2O$ with an agate mortar. Then, the mixed powders were pressed into pellets with diameter of 3 mm or 5 mm. TG-DTA, XRD and SEM/EDS were used to investigate reactions between calsil and Cs^+ at high temperature, associating phase changes, and morphologies and elemental distributions of end-products in Ar-5%H_2 and Ar-20%H_2O atmospheres.

3. Results and discussion

As shown in Figure 1, $CsAlSiO_4$ compound is present in the sample heated up to 730°C. Then some chemical interaction for the calsil mixed with $CsOH•H_2O$ in Ar-5%H_2 occurred in the temperature range of 575°C to 730°C. It was also found that a similar XRD pattern appeared for the sample heated in Ar-20%H_2O. Thus, our results suggested that formation of new compound occurred regardless of atmospheres. The thermodynamic calculations [2-4] were performed to check whether $CaSiO_3$ reacted with CsOH. As a result, it was found that such a reaction did not occur thermodynamically. However, if alumina is present, $CsAlSiO_4$ can be formed by the following reaction:

$$2CaSiO_3(s)+Al_2O_3(s)+2CsOH(c) \rightarrow 2CsAlSiO_4(s)+2CaO(s)+H_2O(g)$$

where s, c, and g are solid, condensed, and gaseous phase, respectively. Therefore, presence of impurity Al in the calsil might play an important role in such a chemical interaction with CsOH, which might be one of the causes for high dose rate reading at the pedestal vicinity.

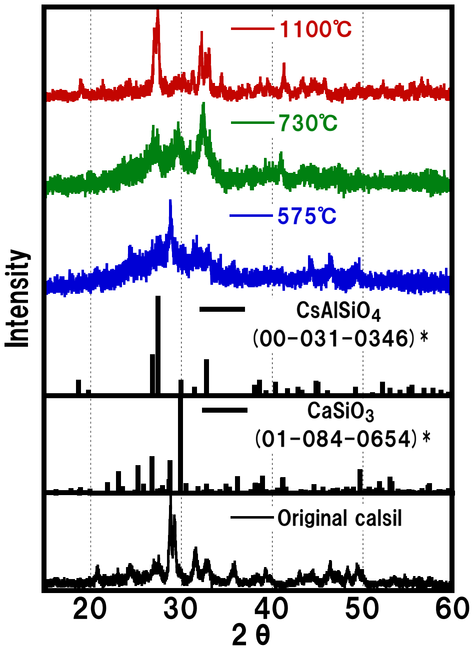

Figure 1 XRD patterns of the mixtures of calsil and $CsOH•H_2O$ heated at 575°C, 730°C and 1100°C in Ar-5%H_2 together with those of original calsil, $CsAlSiO_4$ and $CaSiO_3$ (*ICDD card number)

References

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