

# High-temperature gas-solid interaction of calcium silicate insulation with cesium hydroxide

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## Abstract

Here we report an experimental study to investigate cesium retention on calcium silicate insulation following the gas-solid chemical interaction at high temperature. This study used cesium hydroxide and was performed at 800°C under the oxidizing atmosphere. The experimental results showed that water-insoluble CsAlSiO<sub>4</sub> and dicalcium silicate/larnite (Ca<sub>2</sub>SiO<sub>4</sub>) were formed, which evidences that the insulation material could retain Cs in a stable form.

**Keywords:** Cesium, Calcium silicate insulation, Gas-solid chemical interaction, CsAlSiO<sub>4</sub>

## 1. Introduction

The investigation in primary containment vessel (PCV) of Fukushima Daiichi NPP (1F) unit 2 reported by TEPCO indicated uneven dose rate distribution of the lower 10 Gy/h at the pedestal, where most of the debris should have existed, and the higher 70-80 Gy/h at the pedestal vicinity [1]. Deposits found in this vicinity were suspected as a source of the high dose rate. Considering the condition and location where these deposits existed, it is likely that the formation of such deposits was originated by steam containing fission products such as Cs, leaked from, e.g., safety relief valves gasket. We presumed that the leaked steam had an exposure to the thermal insulation of main steam piping (i.e. calcium silicate insulation or calsil), and that it caused adsorption of Cs on the calsil. In the previous study [2], we investigated the interaction between calsil and cesium hydroxide (CsOH) using TG-DTA—in which the initial forms of both materials were solid—and obtained fundamental information such as chemical forms of Cs in calsil after the interaction. However, it should be noted that in the previous study the respective materials were in contact before the heating test—inducing high probability to interact—while in the present study those materials were separated in different locations, and then the vaporized cesium was transported with the gas flow. Therefore, the objective of this study was to prove and examine the applicability of previously obtained results, whether or not the interaction would occur in the gas-solid system.

## 2. Experimental method

A calsil block, which is equivalent in quality to the thermal insulation used in 1F, was prepared on Ni crucible and then placed downstream in the horizontal furnace, while the CsOH was located upstream and transported by steam flow. The gas-solid interaction tests were conducted at 800°C (i.e., the suggested temperature of chemical interaction for calsil and cesium [2]) under Ar-4%H<sub>2</sub>-20%H<sub>2</sub>O atmosphere with a flow rate of 100 cm<sup>3</sup>/min and a pressure of 0.1 MPaG. During the quenching process, the atmosphere condition was kept in Ar-5%H<sub>2</sub> to prevent further condensation on the sample.

## 3. Results and discussion

The XRD patterns, as presented in Fig. 1, are for calsil after the interaction followed by the dissolution process (the detail of dissolution is explained in Ref. [2]), along with the XRD pattern of TG-DTA sample S12 (i.e., calsil mixed with CsOH; temperature up to 1100°C under Ar-4%H<sub>2</sub>-20%H<sub>2</sub>O) [2]. It was found in this gas-solid system that water-insoluble CsAlSiO<sub>4</sub> and larnite (Ca<sub>2</sub>SiO<sub>4</sub>) were formed as in the previous experiments using TG-DTA [2]. This result corroborates the predicted reaction in the TG-DTA where interaction between calsil and CsOH at a temperature of 800°C will lead to the formation of CsAlSiO<sub>4</sub> and Ca<sub>2</sub>SiO<sub>4</sub>. This also indicates that calsil could retain Cs into this stable and water-insoluble form at high temperature.

## References

- [1] TEPCO, 2017, Progress report No. 5.
- [2] M. Rizaal, K. Nakajima, T. Saito, M. Osaka, K. Okamoto, J Nucl Sci Technol 2020 [in press].

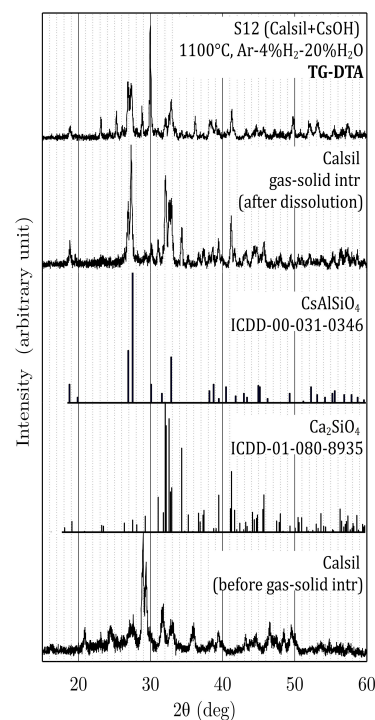


Figure 1 XRD patterns of calsil after the interaction with CsOH, and sample S12 from Ref[2]