

Study on revaporization of deposited CsI from stainless steel 304 surface in high-temperature steam atmosphere

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We experimentally investigated the revaporization behavior of deposited CsI on the surface of structural material such as stainless steel in a subsequent high-temperature steam flow (i.e., reflood of the reactor at severe accident). The experimental results showed that the deposited CsI at 850K could be vaporized, leaving the formation of Cs₂CrO₄ on that once-deposited surface and consequently giving an increase of iodine release observed below 400K.

Keywords: Cesium iodide, revaporization, high-temperature steam, Cs₂CrO₄

1. Introduction

The volatile radioactive fission products (FP) such as iodine and cesium are released from degraded fuel in the event of nuclear severe accident (SA) to which they are transported to the reactor coolant system (RCS) and the containment building, as had been demonstrated in the Phebus-FP experiments [1]. During the transport, Cs and I can react and significantly deposit in the RCS as CsI [1]. In the case of reactor reflood where excess steam can be generated, the stability of deposit is likely to be affected such as revaporization. Therefore, we performed an experimental study using a thermal gradient tube (1000-400K) to investigate the possible mechanism of gaseous I formation due to CsI revaporization from the surface of RCS material, stainless steel 304, in a high-temperature steam atmosphere.

2. Experimental method

The experimental setup called TeRRa was set the same as the previous study [2] where CsI powder (Wako, 99.9 wt%) in alumina crucible was vaporized in a resistance furnace at the axial location having 1050K (max. 1273K). Before the vaporization started, Ar-20%H₂O had been fed to the thermal gradient tube (TGT) made of stainless steel 304 (SUS304) with temperature 1000-400K to achieve pre-oxidized condition. In the TGT, 33 sampling coupons SUS304 were placed at specific axial locations to represent the temperature of interest. After 2-hour vaporization under Ar-20%H₂O and followed by the cooling process, the crucible was removed and the filter placed at the TGT outlet was exchanged. Next, the heating process was repeated under the same mixed gas flow and sustained for 1.25 hours. After which, each part of TGT, and the filter were taken and dissolved into NaOH solution for the measurement of Cs and I deposit amount by ICP-MS. Separately, the sampling coupons were analyzed using SEM/EDS, XRD, and Raman spectrometer.

3. Results and discussion

The Cs and I distribution in the TGT are presented in Figure 1. Previous results [2] showed the initial deposition has a peak deposition at 850K. This initial deposition was further confirmed to be unstable and tend to vaporize, in which it decreased significantly (46% of Cs and 51% of I deposit). Following the revaporization, another deposition occurred at 800-750K where consisted of mainly CsI. Additionally, we measured a fractional increase of gaseous I downstream (<400K). This was resulted after the revaporization at 850K (dashed line) where a part of CsI was decomposed and had a reaction that producing gaseous I so that a lesser amount of I deposit remained (i.e. Cs/I>1). The Raman spectroscopy result on coupon No. 10 (Figure 2) where such Cs/I>1 observed (Figure 1, 850K), XRD, and element mapping (not shown here) confirmed the formation of cesium chromate (Cs₂CrO₄). Therefore, the gain of gaseous I which is obtained by the following possible reaction: $8\text{CsI}(c) + 2\text{Cr}_2\text{O}_3(s) + 5\text{O}_2(g) = 4\text{Cs}_2\text{CrO}_4(s) + 4\text{I}_2(g)$, can prevail during the revaporization by subsequent high-temperature steam flow.

References

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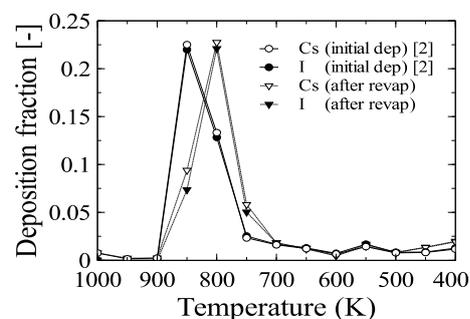


Figure 1. Deposition fraction of Cs and I in TGT at initial deposition [2] and after revaporization test

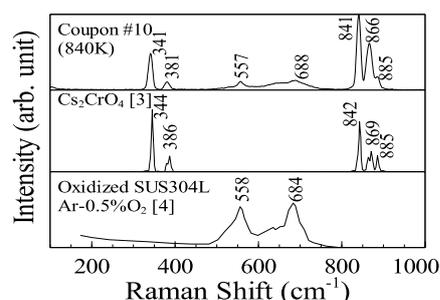


Figure 2. Raman spectrum of deposit on sampling coupon No. 10