New synthesis method of simulated fuel containing high volatile Cs compounds

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Abstract: To systematically investigate the release behaviors of Cs and I from the fuel under accident conditions, we have developed a new synthesis technique of simulated fuel containing high volatile compound of CsI. The synthesized simulated-fuel has a similarity of microstructure with the real irradiated UO_2 fuel from the viewpoint of the grain size and the very fine dispersed CsI particles.
Keywords: Release behavior, CsI, Simulated nuclear fuel, Air-tight sintering

1. Introduction

The severe accident of the Fukushima-Daiichi Nuclear Power station recalled the importance of improved source term for the enhancement of Light Water Reactor safety. The more accurate evaluation of release behaviors of fission products, such as Cs and I which have a high environmental radiological impact, is crucial for the improvement of source term. Although such research requires the systematic experiments using a lot of irradiated fuels, it is hardly possible owing to the inconvenience and high cost for the treatment of irradiated fuels. In response to this issue, in particular for the chemical states of fission products in the fuel, the utilization of simulated nuclear fuel (SIMFUEL) in laboratory experiments as the substitute of irradiated fuels has been proposed [1]. However, it is difficult to synthesize SIMFUEL containing high volatile compounds containing Cs and/or I, due to their massive vaporization in the sintering process. Although the Spark Plasma Sintering (SPS) can achieve the fabrication of pellet containing Cs and I by a quick sintering at relatively low temperatures [2], the SIMFUEL with the microstructural similarity with the irradiated fuels is still unavailable due to limited grain growth.

Therefore, the objective of present study is to develop a new synthesis technique of SIMFUEL containing volatile Cs and I compounds having more similarity with the real irradiated fuel, namely CsI dispersion and grain growth of the matrix.

2. Synthesis method

The new synthesis method named as Air-Tight Sintering (ATS) is based on the sintering of the green pellet of SIMFUEL sealed in a metal crucible to suppress the vaporization of high volatile compounds in the sintering process. The Fe and Mo crucibles were used at the sintering temperatures of 1450 and1500 ºC, respectively. The ZrO_2 stabilized by 8 mol% Y_2O_3 (YSZ) was used as the surrogate of UO_2 because of its stable CaF_2 structure and expected similar chemical properties with UO_2 which strongly affects the redox of some fission products having a high affinity with Cs and /or I. The CsI was chosen as simulant of Cs and I volatile compounds.

The CsI powders (grain size: below 150 μm) were mixed in concentration of 2.0 or 5.0 mol% with YSZ powders (median grain size: about 1.5 μm) in an agate mortar. About 65 mg of the mixed powders were then pressed into a columnar pellet with the 3 mm diameter at about 200 MPa. After placing the green pellet into the Fe or Mo crucible, the crucible's lid was screwed on tightly in a glove box under a nitrogen atmosphere and then welded by electron beam in vacuum. Sintering was performed in an electrical resistance furnace under a reducing atmosphere of Ar-5% H_2. The heating rate was 10 ºC/min up to 800 ºC and then changed to 3 ºC/min up to target temperature (1450 or 1500 ºC). After holding for 2.5 or 5 or 10 hours and cooling down, the apparent density of the sintered pellets was obtained by geometrical method. The pellet samples were characterized by XRD and SEM-EDS, including the determination of grain size of samples by the line intercept method.

3. Synthesis results

The XRD results for all the sintered pellet samples showed only two phases: CsI and YSZ, indicating that CsI was successfully embedded into the sintered pellet. Microstructural analysis by SEM-EDS on fracture surface of samples confirmed the presence of very fine white CsI particles, which are homogeneously located on the grain boundaries in the pellet.

The SIMFULE sintered by ATS at 1500 ºC for 5 hours was compared with that by SPS [2] and the irradiated fuel [3] as shown in Table 1. The grain size of the sintered pellet by ATS is mainly in the range of 2 to 9 μm, giving an average value of 5.58 μm. The CsI particle size is typically 30-500 nm. All these clearly shows the SIMFULE by ATS is closer to irradiated fuel than that by SPS.

Table 1: Comparisons of SIMFUELs by ATS and SPS.

	Relative density/%	Grain size/μm	CsI particle size/nm
YSZ SIMFUEL by ATS	93.2%	5.58 ± 1.64	< 500
CeO_2 SIMFUEL by SPS	- (pellet was cracked)	< 1	2000-3000
Irradiated fuel	< 95%	5-10	< 10

4. Conclusions

By ATS, the CsI was successfully embedded in the sintered SIMFUEL pellet of YSZ, which has a good microstructural similarity with the irradiated fuel, including CsI dispersion and grain size. Therefore, by adding other semi- and low-volatile fission product elements, such as Mo and Ru, the SIMFUEL containing CsI synthesized by ATS can be provided to a series of systematic experiments of Cs and I behavior focusing on the chemical states of Cs and I compounds in a nuclear fuel.

Reference

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