Release Characteristics of Cesium from Green Pellet Fabricated with Spent Fuel Under Different Sintering Conditions

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1. Introduction

The dry process, known as DUPIC(Direct Use of spent PWR fuel in CANDU reactor), for fabricating fuel pellets from spent fuel as recycling technology has been well demonstrated by establishing an optimization process for fuel fabrication through a number of batch processes using typical PWR spent fuel. As considering a strategy for extending the burn-up in LWR fuel, experimental verification for analyzing the effect of spent fuel burn-up on fuel fabrication is necessary in some respects that one of key parameters influencing the fuel fabrication characteristic would amount of fission products contained as impurity elements in spent fuel. A high burn-up spent fuel has a higher amount of fission products compared with typical spent fuel irradiated in about 27,000 MWd/tU. A preliminary study showed that the sintered pellet density fabricated with a high burn-up fuel has a lower value than that of common fuel burn-ups of about 30,000 MWd/tU. To provide better understanding a remote fuel fabrication characteristic in an aspect of wide ranges of spent fuel generated from PWR reactor, the influence of fission products release on fabrication characteristics of the dry processed fuel with a high burn-up fuel of 65,000 MWd/tU were experimentally evaluated. It is expected that key fission product affecting fabrication characteristics in dry process is cesium isotope due to the boiling point of 670°C and the low dissociation temperature of its oxides(<700°C) [1,2]. This study focus to analyze the release characteristics of cesium from green pellets fabricated with a variation of compaction pressure under different sintering conditions using tubular furnace in IMEF M6 hot cell.

2. Experimental

The spent PWR fuel with the high burn-up of 65,000 MWd/tU(**K23-M03**) with short decay time in K23 assembly from Uljin # 2 in 2001 was used for fabricating the green pellets as raw materials for the study of release behavior of cesium during sintering step. Fuel pellets were fabricated in accordance with the optimized fuel fabrication process flow [3]. The process parameter of each step was established from both the pre-qualification test and qualification test. The optimized process conditions were applied for fabrication of the OREOX (Oxidation and Reduction of Oxide fuel) powder and green pellet. U_3O_8 powder oxidized at 500°C for 5 hrs was treated by the OREOX process which is composed of 3 cycles of oxidation at 450°C in air and a reduction at 700°C in 4%H_2/Ar. After milling and precompaction steps, green pellet was fabricated by the final compaction process with a pressure range from 88 MPa to 150 MPa. The typical sintering conditions in an Ar-4%H_2 atmosphere were summarized in Table 1.

Table 1. Sintering conditions for the evaluation of release behavior of cesium from green pellets.

Test No.	Compaction pre.(MPa)	Sintering conditions	Wt. (g)
1	88	1,650°C(1 step)	12.2
2	88	1,350°C(1 hr holding)→ Reheating to 1,650°C	12.1
3	120	1,650°C(1 step)	12.8
4	120	1,350°C(1 hr holding)→ Reheating to 1,650°C	12.3
5	102	1,650°C(1 step)	13.6

The gamma spectrometer system (GAMMA-X™ HPGE, Coaxial Photon Detector System, ORTEC Gamma Vision 32 GMX Series) was aligned to green pellet located in tubular furnace for obtaining gamma spectrum due to the release of cesium from green pellets. The cumulative release fraction of cesium during sintering step was calculated from a count rates measured during each test, compared with the initial count rate of cesium from green pellet before the test. Fig. 1 shows the schematic diagram of equipment for the fission products release evaluation.

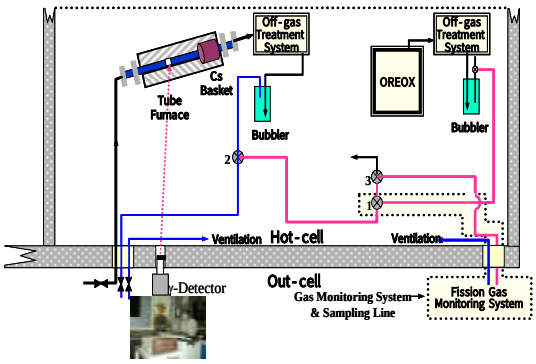

Fig.1. Schematic diagram of equipment for the fission products release evaluation.

3. Results and Discussion

2.1 Fabrication of green pellets

The green pellets were fabricated with using OREOX powder of K23-M03 spent fuel in a range of 80~ 160 MPa of compaction pressure, as shown in Fig. 2. The geometric density of green pellets increased with compaction pressure. But in high compaction pressure over 150MPa, some cracks on green pellet were identified. Therefore, the green pellets fabricated in a range of 80 ~ 120MPa were used for sintering experiments.

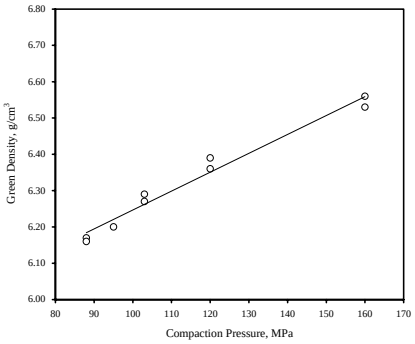

Fig.2. Geometric density of green pellets with compaction pressure.

2.2 Release Characteristics of Cesium form green pellets

Fig. 3 shows the release characteristics of cesium from green pellets under different sintering conditions, test No. 1 and 3 in Table 1. Also, the release characteristic of cesium from green pellets under Test No. 2 and 4 in Table 1 is illustrated in Fig. 4.

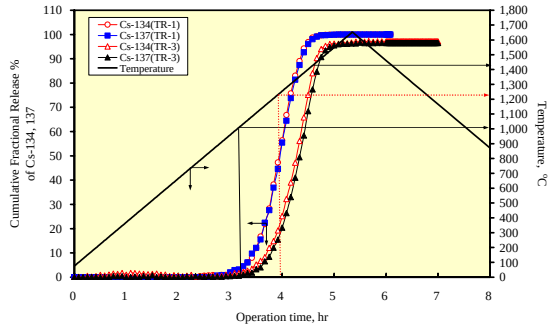

Fig.3. Cumulative release fraction of Cs-134 and Cs-137 from green pellets with sintering time(Test No. 1 & 3).

A difference between test No. 1 and 2 or test No. 3 and 4 is temperature history during sintering step. Cesium was released in a temperature range of 1,100°C to 1,450°C during sintering step The sintering process is generally divided into three stages : (a) Initial stage of sintering (Neck formation and growth), (b) Intermediate stage of sintering (Network of tubular pores), (c) Final stage of sintering (Pore shrinkage). The onset of densification for sintering occurs above 1,100 ~ 1,150°C [4]. It is clear that temperature range in cesium release from green pellet corresponds to intermediate sintering stage. It was reported that cesium is gaseous above 1,200°C, release of cesium occurs by axial migration in the in the grain boundary porosity in the central region of the fuel. Especially, a marked release of cesium on high burn-up fuel was found due to the pellet rim region where the micro-structure changes to a decrease in grain size and the formation of numerous small pores.

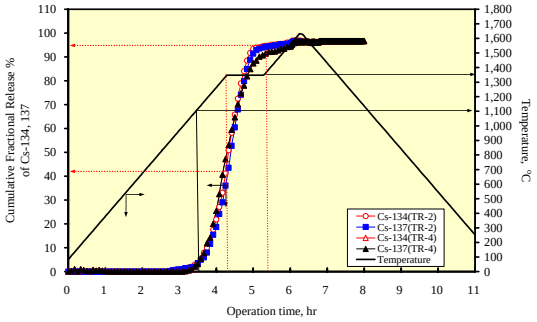

Fig.3. Cumulative release fraction of Cs-134 and Cs-137 from green pellets with sintering time(Test No. 2 & 4).

Based on these results, it is probably expected that gaseous cesium release during intermediate sintering stage affects the network formation of pores, followed a formation of residual pores on green pellet.

3. Conclusion

The release characteristics of cesium from green pellets fabricated with high burn-up spent fuel were evaluated for the analysis of effect of fission products release on the sintering process. Cesium as gaseous phase was released in a temperature range of 1,100°C to 1,450°C during sintering step. This temperature range corresponds to intermediate sintering stage. Therefore, it is expected that cesium release during this stage affects the network formation of pores.

Acknowledgement

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