A study of the fission gas release during the oxidation of the irradiated large grain UO_2 specimens

Dae Ho Kim, Yong Sik Yang, Jae Geon Bang, Sun Ki Kim, Ik Sung Lim, Kun Woo Song
Korea Atomic Energy Research Institute, 1045, Daedeok-daero, Yuseong-gu, Daejeon, 305-303, Korea
kdh@kaeri.re.kr

1. Introduction

Within an irradiated UO_2 fuel, the fission gas inventory can be divided into two major regions, the grain matrix and the boundaries. In a normal operation condition, most of the fission gases which remain in a fuel are accumulated and stabilized in gas bubbles in intra or inter-granular gas bubbles, and some of them (about 5~10%) are released to the rod free volume. But during an accident condition, such as a RIA, these stabilized gas bubbles act as an additional fission gas release inventory and according to their location, the fission gas release mechanisms and release times are different.

In our previous study, we tried to distinguish the fission gas inventory by a controlled oxidation technique[1].

In an oxidation environment, the grain boundary can be oxidized faster than the grain matrix. Due to the volume expansion, which is caused by the UO_2 to U_3O_8 phase change, a grain boundary separation can occur and the fission gases are released from the inter-granular bubbles instantaneously.

The final goal of this study is to measure the fission gas inventory of a large grain UO_2 pellet and find the grain size effect on the fission gas behavior and, at the current stage, several tests were performed to measure the fission gases which are contained in the grain boundary by a controlled oxidation technique.

In this paper, the oxidation test results are presented and detailed characteristics of the specimens which were irradiated up to 35MWd/kgU are summarized.

2. Specimen Preparation and Oxidation Test

2.1 Specimen preparation

The irradiated UO_2 specimens were prepared from the large grain UO_2 irradiation test rods which were irradiated at a research reactor HANARO up to 35MWd/kgU[2].

In the advanced PWR fuel development project, a large grain UO_2 pellet was developed by a U_3O_8 seed addition method. In order to confirm the newly developed UO_2 pellet's in-reactor performance, an irradiation test(six test rods) has been performed from 2003 at HANARO research reactor. In 2007, three test rods, which had been removed from the HANARO, were transported to PIE and extended irradiation test of remaining three rods will be finished at 2010.

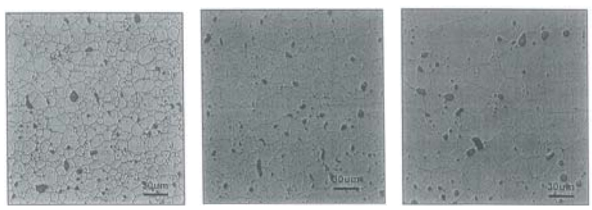
The removed test rods contain three kinds of pellets which have different grain size 8, 15, 23μm respectively.

The 8μm pellet was manufactured by commercial fabrication process but 15μm and 23μm pellets were manufactured by newly developed U_3O_8 seed addition method. Detailed pellet information is shown in the Table I.

Table 1. Composition of the specimens

Parameter	Powder Composition	Grain size	Sintering
Specimen 1	$88^{w/o}$ UO_2 powder + $12^{w/o}$ U_3O_8 powder	8 ㎛	Sintered (4hr)
Specimen 2	$88^{w/o}$ UO_2 powder + $7^{w/o}$ U_3O_8 powder + Additive $5^{w/o}$ U_3O_8 seed	15 ㎛	Sintered (4hr)
Specimen 3	$88^{w/o}$ UO_2 powder + $7^{w/o}$ U_3O_8 powder + Additive $5^{w/o}$ U_3O_8 seed	23 ㎛	Sintered (46hr)

The sintering temperature is same for all pellets but specimen 3's sintering time was larger than others. Comparison result of the grain size between each pellet before an irradiation is shown in a Fig. 1.



a) 8 ㎛ b) 15 ㎛ c) 23 ㎛
Fig. 1. Morphology of the before irradiation specimens

A few centimeters segments were prepared for the oxidation test. After the specimen cutting, each specimen weight is about 150mg. Fig. 2 shows the specimen preparation from the irradiated pellets.

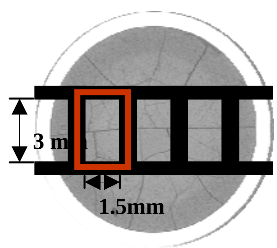


Fig. 2. Oxidation specimen of the irradiated pellet

2.2 Test Condition

The controlled oxidation test was performed by PIA(Post-irradiation Annealing) apparatus which was installed in a post irradiation examination facility[3]. The test temperature is 500℃ and its heating rate is 10℃/min. During the test, the specimen is swept by regulated gases(STD-Air and Helium). After five hour temperature holding at 500℃ the test was stopped. During the test, released ^{85}Kr was measured by gamma counter continuously. After the cooling period, the specimens were transported to the SEM and a microstructure observation was performed.

2.3 Test results

The ^{85}Kr release kinetics for a specimen 1, 2, 3 during an oxidation are given in Fig. 3 and 4. As shown in Fig. 3, specimen 1's(the smallest grain) release begins first and a bigger grain size specimen shows a late release of ^{85}Kr.

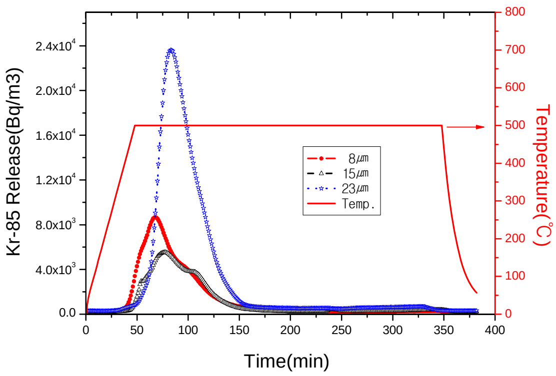


Fig. 3. ^{85}Kr release behavior during oxidation test

But, the total release quantity is not inverse-proportion or proportion to the grain size. Although the specimen 3 has the largest grain size, its ^{85}Kr release is the maximum. On the other hand, specimen 1(8μm) shows a larger release than specimen 2(15μm). These results can be confirmed in Fig. 4.

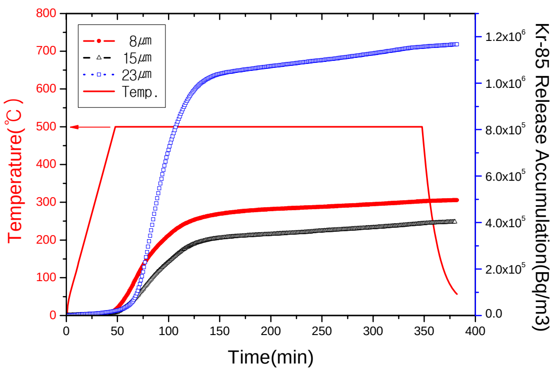


Fig. 4. Cumulative ^{85}Kr release results

In Fig. 4, the ^{85}Kr release from the 23μm specimen is 3 times larger than that 15μm specimen and specimen 1's release is slightly larger than specimen 2's one.

After the oxidation test, the SEM observation was performed to confirm a grain boundary separation. Fig. 5 shows the SEM image of each specimen of ×1000 and ×2000 magnifications respectively and this figure shows a grain boundary separation occurred by an oxidation for all specimens.

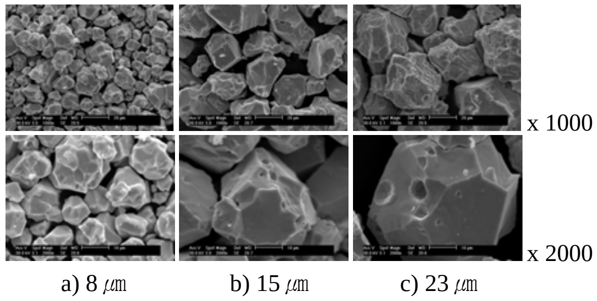


Fig. 5. SEM observation of the oxidized specimens

3. Conclusion

The controlled oxidation tests were performed by using the irradiated UO_2 pellets which were manufactured with different grain sizes.

Although the specimens' burnup are very similar, they show a different fission gas release behavior. Specimen 1, which has the smallest grain size, shows the fastest release start. But specimen 2, which has the largest grain size, shows the maximum fission gas release.

The SEM observation, which was performed after the oxidation test, shows that a grain boundary separation occurred for all the specimens and it is concluded that all the fission gases were released from the grain boundary.

At the current stage, the effect of the grain size on the fission gas inventory and the fission gas behavior is not clear. And more experiments such as an annealing test will be performed to investigate the effect of the grain size on the fission gas behavior.

REFERENCES

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