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Effect of Sintering Conditions on the Grain Growth of an Al-doped UO_2 Pellet

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1. Introduction

In developing a nuclear fuel for PWR (Pressurized Water Reactor) at a high burnup and extended cycle, there are several issues which must be considered. One of them is the effect of an increase of the fuel rod internal pressure due to FGR (Fission Gas Release). The release of fission gases in a reactor fuel rod is an important performance-limiting factor. Another is an effect of PCI (Pellet-Clad Interaction). Cladding isolates fuel from the coolant and prevents a release of radionuclide and a contamination of the primary circuit. A high integrity of a cladding material under an irradiation is essential. PCI-induced failure (stress corrosion cracking, etc.) must be considered. The cause of the PCI phenomenon is various. A fuel pellet swelling is a major factor for PCI, which is mainly affected by a fission gas and a bubble swelling. It can be thought that the effect of an intrinsic thermal expansion of a UO_2 pellet is relatively small.

Several ways to prevent the negative influence of FGR have been suggested. A large-grain pellet has been considered to reduce FGR and gas swelling of cladding, because they are rate-controlled by gas atom diffusion from the grain interiors to the grain boundaries [1]. A large-grain pellet can be produced by controlling the powder treatment, sintering conditions, and by using additives. Also, a large-grain pellet can be fabricated by mixing many kinds of additives with a UO_2 powder. To obtain a large-grain pellet, Nb_2O_5, TiO_2, Cr_2O_3, Al_2O_3, SiO_2 and MgO have been chosen [2-6]. However, if the amount of additives is too large, it may lead to secondary problems.

According to ASTM designation C753-99, in the nuclear-grade sinterable uranium dioxide powder, the impurity content shall not exceed the individual element limit specified on a uranium weight basis. The maximum concentration limit of aluminum is restricted to 250 μg/gU.

In a previous work [7], a large-grain pellet was fabricated as a function of the aluminum content and oxygen potential within the permissible limit. In the result, the peak of the grain size was found at the solubility limit (~42 ppmAl [8]) of aluminum in UO_2. Also, it was shown that the most effective oxygen potential for the grain growth of the Al-doped UO_2 is about ΔG_o=~205 kJ/mol.

In the present work, the effect of the sintering temperature and time on the fabrication of an Al-doped large grain pellet was investigated. On the basis of previous results, the sintering test was conducted by using a doping content of 42 ppmAl and a sintering atmosphere of dry hydrogen.

2. Experimental and Results

Al-doped UO_2 sintered pellets were prepared as follows. $Al(NO_3)_3 \cdot 9H_2O$ (Aldrich, 98+%, aluminum nitrate nonahydrate) powders were dissolved in alcohol or distilled water. ADU-UO_2 (BNFL, Ammonium Diuranate) and Al-diluted water was mixed with 42 ppmAl. The mixture was dried under a flowing air condition for more than 48h. Dried powder mixture was compacted with a compaction pressure of 300 MPa and sintered in the various sintering conditions (temperature and time). The sintering tests were conducted in a flowing dry hydrogen atmosphere.

The density of the sintered pellet was measured by using an immersion method. The density-measured pellet was cut in the axial direction. And then a grinding and polishing process was performed. To observe the grain structure, a thermal etching for the polished samples was carried out at 1300 ºC for 2h in a flowing CO_2 atmosphere. The microstructure of the samples was observed by using OM (Optical Microscopy). The grain size of the sample was measured by using the linear intercept method.

The grain size data of the Al-doped UO_2 with various Al contents are shown in Figure 1. Although the Al content and the oxygen potential of the sintering atmosphere are very low, it shows that the grain sizes increase with an increasing Al content.

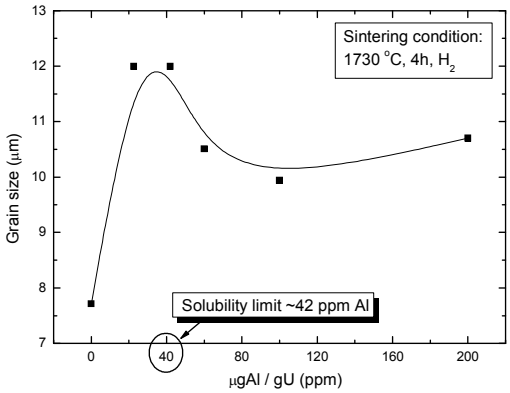

Figure 1. Grain sizes of Al-doped UO_2 pellets with various Al contents at 1730 ºC for 4 h in a flowing H_2.

It is remarkable that the peak of the grain size is found at a solubility limit of Al in UO_2 (~42 ppmAl [8]). It can be thought that the effect of dissolved Al in the

UO_2 matrix on the grain growth is more effective than that of the precipitate. In the range of more than 42 ppmAl, the grain size of the Al-doped UO_2 slowly increases.

Figure 2 shows the relationship between the sintering condition (temperature and time) and grain growth of the UO_2 and 42 ppmAl-doped UO_2 pellet.

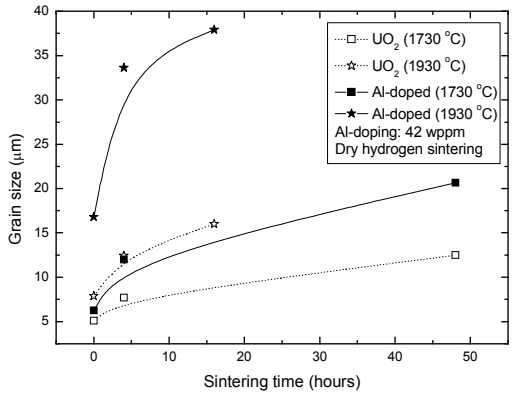

Figure 2. Relationships between sintering condition and grain size of UO_2 and 42 ppmAl-doped UO_2 pellet.

The grain sizes of the UO_2 and Al-doped UO_2 pellet gradually increase with an increasing sintering temperature and time. The effect of the sintering temperature and time on the grain growth of the Al-doped UO_2 pellet is larger than that of UO_2.

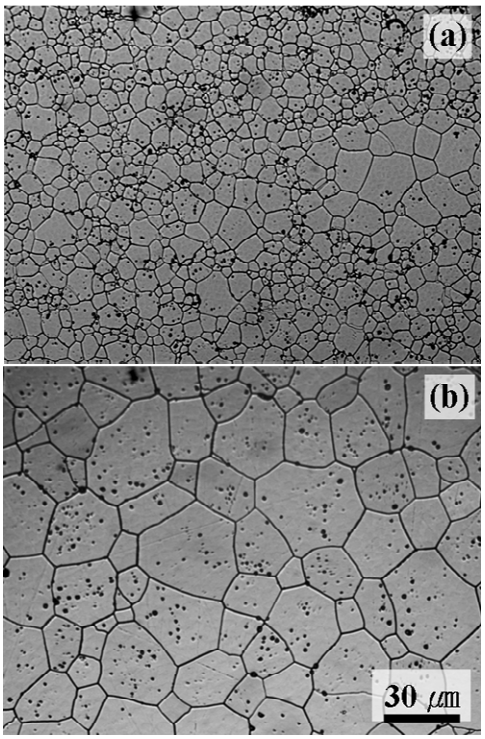

Figure 3. Optical microscopic image of the 42 ppmAl-doped large grain pellet (×200): (a) 1730 ºC, 4h, H_2 (typical sintering condition), (b) 1930 ºC, 16h, H_2.

From the results of the Al-doped UO_2, it can be thought that the grain sizes due to the change of the sintering temperature and time are saturated with 40~45 μm at 1730 ºC and 25~30 μm at 1930 ºC.

3. Conclusion

Effects of the sintering condition (temperature and time) on the fabrication of an Al-doped large-grain pellet were investigated by using small amounts of aluminum within the permissible limit.

As a result, the grain sizes of the UO_2 and Al-doped UO_2 pellet increased with an increasing sintering temperature and time. It was shown that the sintering conditions (temperature and time) have a larger effect on the grain growth of the Al-doped UO_2 pellet than that of UO_2.

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