Effect of Small Amounts of Aluminum Doping on the Grain Growth of a UO_2 Pellet

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1. Introduction

In the development of a nuclear fuel pellet for pressurized water reactor (PWR), there are two key parameters which must be significantly considered. One is the effect of an increase of the fuel rod internal pressure due to fission gas release (FGR). The release of fission gases, i.e. volatile fission products (xenon, krypton, etc.), in a reactor fuel rod is an important performance-limiting factor. A critical review of gas release mechanisms in UO_2 has been given by many investigators.

Another is an effect of the pellet-cladding interaction (PCI). 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 seriously considered. The cause of the PCI phenomenon is various. The fuel pellet swelling is a major factor for PCI, which is mainly affected by 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.

Especially, in order to develop a nuclear fuel pellet for PWR at high burnup, the FGR and PCI are considerable factors. Several ways to prevent the bad influence of FGR and PCI have been suggested.

A large-grain pellet has been considered to reduce FGR and gas swelling, because they are rate-controlled by gas atom diffusion from the grain interiors to the grain boundaries [1]. The large-grain pellet can be produced by controlling the powder treatment, sintering conditions, and by using additives.

The large-grain pellet can be fabricated by mixing many kinds of additives to 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 [2-8]. However, if the amount of the additives is too large, it may lead to secondary problems (formation of defect structure, change of radioactive characteristics, degradation of thermophysical properties of fuel materials, etc.).

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 the present work, the fabrication of a large-grain pellet was performed by using small amounts of aluminum within the permissible limit. Effects of the sintering condition (oxygen potential) on the fabrication of the Al-doped large-grain pellet were also investigated.

2. Methods and Results

2.1 Powder preparation and sintering

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 various contents. 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 at 2003 K for 4h in the various sintering conditions which were controlled by the oxygen potential by changing the ratio of CO_2 to H_2.

2.2 Grain size measurement of Al-doped UO_2 pellets

The density of the sintered pellet was measured by using an immersion method. Density-measured pellets were cut in an 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 1573 K 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, and the results are shown in Table 1.

Table 1. The measured grain size* of the Al-doped UO_2 pellets

Al content (μgAl/gU)	H_2	CO_2/H_2 0.5%	CO_2/H_2 1.5%	CO_2/H_2 3.0%
0	7.711	6.999	7.891	6.942
23	11.996	10.718	12.18	11.448
42	11.996	15.588	17.464	14.365
60	10.509	13.703	14.054	13.304
100	9.938	12.459	11.31	11.203
200	10.699	11.657	11.203	10.993

* The unit of the grain size is μm.

According to the results of the measured density of the pellets, the densities of the pellets were 95±0.5%T.D. despite various Al contents. Therefore, in this experiment, effects of the sintered density on the grain growth can be excluded. The grain size data of the Al-doped UO_2 are shown in Figure 1. Although the Al content is very low, it shows that the grain sizes definitely increase with an increasing Al content.

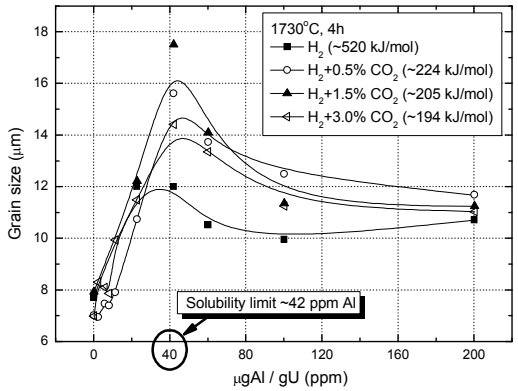

Figure 1. Grain sizes of Al-doped UO_2 pellets with various Al contents.

In this result, it is remarkable that the peak of the grain size was found at a solubility limit of Al in UO_2 (~42 ppmAl [9]). 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 and grain size of the Al-doped UO_2 pellet. The grain size of pellet sintered in an oxidized atmosphere is larger than that in a pure H_2 atmosphere.

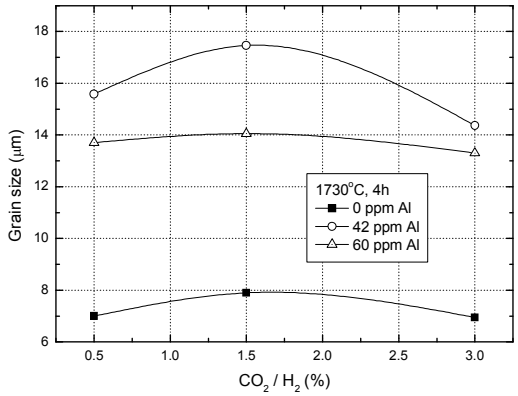

Figure 2. Relationships between sintering condition and grain size of Al-doped UO_2 pellet.

Effect of the oxygen potential on the grain size didn't linearly increase. A maximum grain size was found in the CO_2/H_2=1.5% atmosphere. That is to say, it was shown that the most effective oxygen potential on the grain growth of Al-doped UO_2 was about ΔG_o=~205 kJ/mol.

3. Conclusion

The fabrication of a large-grain pellet was performed by using small amounts of aluminum within the permissible limit. Effects of the sintering condition (oxygen potential) on the fabrication of the Al-doped large-grain pellet were also investigated.

As a result, the peak of the grain size was found at the solubility limit of Al in UO_2. Also, it was shown that the most effective oxygen potential on the grain growth of the Al-doped UO_2 is about ΔG_o=~205 kJ/mol.

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