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Effects of Al_2O_3-NiO on the Grain Growth and Thermal Properties of UO_2 Pellets

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1. Introduction

As a method to enhance economy and safety of nuclear plant , development of nuclear fuel for high burnup and long term irradiation cycle is actively pursued in various fields [1]. To optimize the fuel properties many studies have been performed, focusing on the effects of processing parameters on the grain sizes of UO_2 pellets. Among those, the doping technology is preferred because of its easy compatibility with commercial pellet fabrication and significant effect o grain growth [2,3].

In this study, Al_2O_3-NiO mixture was prepared and added to commercially produced UO_2 powder in KEPCO NF. The effect of Al_2O_3-NiO on the properties of UO_2 pellet has been examined. In addition, the KEPCO NF developed commercial technology is also introduced.

2. Manufacturing Process

2.1 Preparation of the UO_2 and U_3O_8 powders.

UO_2 powder produced by commercial Dry Conversion process in KEPCO NF was used in this study. The U_3O_8 powder added for manufacturing of UO_2 pellets was prepared by oxidizing the defective UO_2 pellets at temperature under 350°C and the grinding sludge at 300°C in air respectively. The temperature was chosen because the specific surface area (BET) of U_3O_8 powder was maximized at that temperature [4-7]. For a reference sample, conventionally produced U_3O_8 powder was also prepared by oxidizing the UO_2 pellets at 450°C in air. Due to long oxidation of UO_2 pellet at low temperature and recycling the defective UO_2 pellets for mass production scale, KEPCO NF developed new heat treatment equipment for oxidation was introduced.

2.2 Pelletizing

The effect of the U_3O_8 powders on the pellet properties in accordance with oxidation temperature was evaluated. Two kinds of U_3O_8 powders oxidized at temperature under 350 and 450°C were prepared. 9wt% of the respective U_3O_8 powder was added to the UO_2 powder. About 0.3wt% of AZB as a pore former and about 0.25wt% of ACRAWAX as a lubricant were also included in the mixed powder. In addition to this, the additive mixture (30ppm Al_2O_3 and 30ppm NiO) was added to powder mixtures. During the fabrication process, very small amount of unintentional impurities could be included in the fuel powder mixture. The addition of Al_2O_3 and NiO intends to simulate that impurity addition within specification limit. Mixing was carried out for ~1 hr in Nauta Mixer.

The mixed powders were pressed into green pellets. The green pellets were sintered at 1720°C for ~3 hr in a commercial sintering furnace. Pure hydrogen gas was blown. The flowing rate of H_2 gas was 5 N ㎥/hr.

3. Test Results

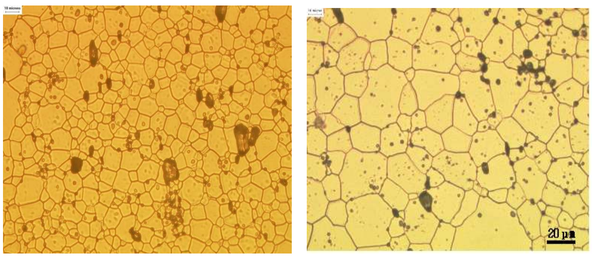
The characteristics of the oxidized powders such as specific surface area (BET), O/U ratio and mean particle size were measured. After the sintering process, all test specimens were grinded in order to investigate the surface integrity of the pellets. Sintered density was measured using immersion method. In all cases the densities were reported as a percentage of the theoretical density(TD) of UO_2 (10.96 g/cm^3). A pellet was sectioned longitudinally and polished to observe microstructure. For measuring grain size, thermal etching was performed at 1250°C for 1.5 hr. Grain size was determined by a linear intercept method. In addition, resintering density change and other characteristics like creep, thermal conductivity, thermal expansion etc. also were measured. The thermal behavior such as diffusivity, specific heat, thermal expansion and thermal conductivity of conventional and doped pellets are almost the same. The results of high temperature compressive deformation test showed that the strain of doped pellets was little bit smaller than the conventional samples.

Table 1 shows the summary of the test results. The pellet properties of developed UO_2 pellet, conventional UO_2 pellet and the specification requirement of UO_2 pellet were denoted in this table. As shown in the table, grain size of the developed UO_2 pellet was about 2 times larger than the conventional one. The thermal stability of developed pellet was remarkably improved compared to the conventional pellet. The other properties of the developed pellet satisfied well the specification requirements.

Fig. 1(b) shows the grain structure of UO_2 pellet obtained through the additive of Al_2O_3 and NiO. It clearly shows that the doped pellet not only has large grains but also stable pore structure compared to those of the conventional pellet in Fig. 2(a).

Table 1. Summary of test results

Property	Conventional UO_2 pellet	Developed UO_2 pellet	Specification requirements
Grain size(㎛)	7~8	~17	≥ 5㎛ (target: ≥ 10 ㎛)
Sintered density(%TD)	~95.8	~95.7	95-96.5%TD
Thermal stability(%TD)	~0.6	~0.2	≤ 1.0%TD
Enrichment, O/U ratio,	pass	pass	Specification limit
Uranium & Impurity, Hydrogen content	pass	pass	Specification limit
Dimensions, Surface finish	pass	pass	Specification limit
Microstructure (Pore size distribution & Morphology)	pass	pass	Specification limit (for information)
Thermal properties (Creep, thermal expansion, specific heat, thermal diffusivity)	equivalent		For information



(a) (b)

Fig.1. Grain morphologies of (a) conventional undoped UO_2 pellet and (b) Al_2O_3 and NiO doped UO_2 pellet

4. Conclusions

Large-grained UO_2 pellets were successfully developed through the micro-doping of Al_2O_3 and NiO mixture without any additional change in commercial UO2 production. The maximum average grain size of UO_2 pellets containing 60 ppm of $50Al_2O_3$-50NiO(mol%) was measured to be about 17 ㎛, and it is about 2 times larger than that of conventional undoped UO_2 pellets. Based on the results of the thermal properties, the doped pellets are considered equivalency to the commercial pellets. Now additional reproducibility tests and process optimization in mass production line are undergoing.

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