Development of commercial manufacturing technology for large grain UO₂ pellets

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1. Introduction

To increase economic benefit and safety margins of nuclear plant, development of nuclear fuel for high burn-up and long-term cycle is important. Research on fuel pellets focuses on increasing the pellet density and grain size to increase the uranium contents and the high burn-up safety margins for LWRs.[1] Large grain pellet can reduce the corrosive fission gas release at high burn up. Large grain pellet also has viscoplasticity and viscoplastic soft pellets can lower the pressure to a cladding caused by a thermal expansion of a pellet at an elevated temperature during transient operations. Those advantages can provide room for additional power uprates and high burnup limits.

Large grain UO_2 pellets could be obtained by applying the additive doping technology [2-5]. However, introduction of foreign element in UO_2 fuel could alternate the in-reactor fuel performance. So in order to commercially use the doped UO_2 fuel in reactor, long-term verification of the pellet is required. Besides the using of additives, there are other methods to promote grain growth of the UO_2 pellet through the pretreatment of UO_2 and/or U_3O_8 component powders [6]. Because these methods do not change the fuel composition, technology can be more easily applied in the commercial.

Among these, KAERI had been developed the large grain UO₂ pellet fabrication process by optimizing the recycle process of defective UO₂ pellets. The characteristic of that technology is lowering the oxidation temperature of defective UO₂ pellet to obtain sinter active U_3O_8 powder. The powder properties of BET surface area and particle size of a recycled-U₃O₈ powder was enhanced by lowering the oxidation temperature. A low temperature oxidized U₃O₈ powder with a high BET surface area and a small particle size was effective in mitigating the density drop and in promoting a grain growth of UO₂ pellets. This developed process only requires lowering the oxidation temperature so it has good compatibility with conventional commercial UO₂ pellet fabrication process.

KEPCO NF has conducted various qualification tests for the KAERI developed large grain UO_2 pellet

technology to commercialize that during the last two years. To enlarge the lab-scale technology to mass production scale, KEPCO NF has developed various technologies, and optimized several process parameters.

In this study, the qualification test results of large grain UO_2 pellets fabrication which were performed on the mass product line of commercial UO_2 pellets in KEPCO NF is introduced. KEPCO NF developed commercial scale technologies are also presented.

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 for large grain pellets was prepared by oxidizing the defective UO_2 pellets at $350^{\circ}C$ in air. That temperature was chosen because the specific surface area (BET) increase of U_3O_8 powder was saturated at this temperature. For a comparison, conventional U_3O_8 powder was also prepared by oxidizing the UO_2 pellets at $450^{\circ}C$ in air. Oxidation of UO_2 pellet at low temperature requires much time. So, in order to recycle the defective UO_2 pellets for mass production scale, KEPCO NF developed new heat treatment equipment.

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 350 and 450°C were prepared. 8wt% of the respective U_3O_8 powder was added to the UO₂ powder. 0.3wt% of AZB as a pore former and 0.2wt% of ACRAWAX as a lubricant were also included in the mixed powder. In addition to this, 30ppm of Al₂O₃ 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 Al₂O₃ is to simulate that impurity. Mixing was carried out for ~1 hr in Nauta Mixer.

The mixed powders were pressed into green pellets. The green pellets were sintered at 1740°C for 3.3 h in a commercial sintering furnace. Pure hydrogen gas was blown. The flowing rate of H_2 gas was 5 Nl/hr

3. Qualification 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 of UO_2 (10.96 g/cm³). A pellet was sectioned longitudinally and polished to observe microstructure. For measuring grain size, thermal etching was performed at 1250°C for 1.5hr. 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.

Table 1 shows the summary of the qualification 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 around 2 times larger than the conventional one. The thermal stability of developed pellet was remarkably improved comparing to the conventional pellet. The other properties of the developed pellet satisfied well the specification requirements.

Property	Conventional UO ₂ pellet	Developed UO ₂ pellet	Specification requirements
Grain size(µm)	7~8	12~17	$\geq 5\mu m \text{ (target: } \geq 10 \ \mu m)$
Sintered density(%TD)	~95.8	~95.7	95-96.5%TD
Thermal stability(%TD)	~0.6	~0.2	$\leq 1.0\%$ 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)

Table 1. Summary of qualification test results

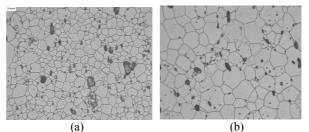


Fig.1. The grain morphologies of (a) conventionally prepared UO₂ pellet and (b) sintered by developed process

Fig. 2(b) shows the grain structure of UO_2 pellets obtained through the developed process. It clearly show that the developed large grain pellet not only has a large grain but also stable pore structure when we compare this figure with Fig. 2(a) which from conventional pellet.

4. Conclusions

KEPCO NF has successfully conducted the qualification test to apply the KAERI developed large grain pellet technology to the KEPCO NF's commercial UO₂ pellet production line. KEPCO NF evaluated the commercial manufacturability of the developed labscale process and developed optimization technology for mass production. From qualification test result, it was found that the pellet quality such as grain size or thermal stability was remarkably enhanced. In addition to this, the developed process could be adopted in the existing fabrication line without installation of any additional equipment. KEPCO NF has plan to use the developed process in the producing the large grain pellet for Advanced Korean Fuel ("HIPER"). Now additional reproducibility tests and process optimization are undergoing.

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