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UO_2 Kernel Preparation by M-EG Process and Its Irradiation Test

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1. Introduction

Kernels of KAERI TRISO fuels are prepared in the following steps: (1) preparation of a raw material solution(UN solution) by UO_3 (or U_3O_8) powder dissolution in the concentrated HNO_3; (2) broth preparation and physical property control by mixing UN, THFA, PVA, and H_2O; (3) preparation of spherical liquid gel droplets and dried-ADU gels in sequence through a reaction between uranyl ions and ammonia ions in a gelation column; (4) ageing, washing, and drying processes of ADU gel using AWD equipment; (5) UO_3 calcination by thermal decomposition of dried-ADU gel in the air; (6) fabrication of UO_2 kernel by reducing the UO_3 and sintering in the H_2 [1,2].

In this study, improved KAERI processes for UO_2 kernel preparation were presented. ADU gel washing procedure in AWD processes and the heating mode in sintering process were modified and the internal structures of UO_2 kernels are presented as a result. PIE results of the kernels from the irradiation test of KAERI TRISO fuels at HANARO were presented briefly. A block diagram of the material flow and experimental apparatus for UO_2 kernel fabrication are shown in Fig.1

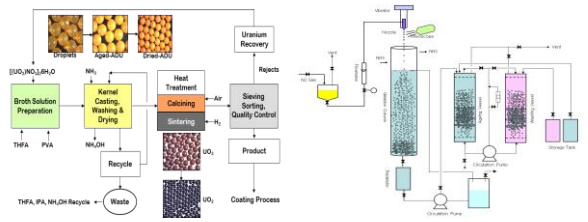

Fig.1. A block diagram and experimental apparatus.

2. Kernel Preparation and Irradiation Test

ADU gel formation and AWD treatment

Spherical liquid ADU gel droplets are made by adding proper amounts of THFA and PVA to UN (uranyl nitrate, $UO_2(NO_3)_2 \cdot xH_2O$) solution, and then by feeding it into a gelation column equipped with vibrating nozzle system. After that, it is aged in an ammonia solution to make ADU gel particles by chemical reaction of the uranyl ion (UO_2^{2+}) and the ammonium ion (NH_4^+) in the gelation column.

The spherical ADU gel particles go through aging-washing-drying processes and dried ADU gel particles are obtained. The unreacted THFA and reaction by-product, NH_4NO_3, were removed in washing step. Final pure ADU gel particles composed of uranium complex compound, $UO_3 \cdot xNH_3 \cdot yH_2O$, and PVA were obtained.

Thermal decomposition and UO_2 fabrication

ADU gel particles obtained from the process contain a trace of water, unremoved by-products and organic additives, and the PVA which is used for the sustainment of the spherical structure of the gel particles. In the thermal decomposition process of the ADU gel particles, the PVA, the unremoved water and the by-products, and decomposition vapors from the heating process in the air are removed and a phase change from ADU gel into UO_3 occurs at the same time.
The UO_3 particles are reduced into UO_2 completely and go through a thermal treating process in H_2 atmosphere. UO_2 internal structures are densificated in an elevated temperature in the same atmosphere as in the reduction process. In this sintering process, the internal structure and the density of the final UO_2 particle could be determined.

Internal Structure Improvement Experiment

For this study, spherical UO_2 particles were prepared with 9 different washing and sintering conditions. Other processes such as ageing, drying and calcining are carried out in the same conditions. The process conditions are briefly described in Table 1.

Table 1. UO_2 kernel preparation conditions.

	Washing Condition Change						
	Broth	Dropping	Ageing	Washing Solution	Drying	Calcining	Sintering
1	d = 1.19 ~1.20 μ = 80~100	F = 80~100 Q = const.	Room temp. Medium= NH_4OH	125ml, 5min., 3times	Vac.= 500 ~550 mmHg	500℃ Air	1600~ 1700 ℃ H_2
2				10min,			
3				15min,			
4				250ml, 5min, 3times			
5				10min.			
6				15min.			
7				375ml, 5min., 3times			
8				10min.			
9				15min.			

d : density(g/cm³), μ : viscosity(cP), F : frequency (Hz), Q : feeding rate (ml/min.)

Irradiation Test

The irradiation test samples were fabricated by mixing and compaction of graphite powder (mixed powder with natural + synthesis) with TRISO-coated particles made of the UO_2 kernels [3]. The irradiation test device, a non-instrumented capsule, is designed for an irradiation in the OR position at HANARO [4]. TRISO-coated fuel particles are about 900μm in diameter. Fig. 2 shows the typical photographs of UO_2 kernel, TRISO-coated particle, overcoated particles, test sample compact, and irradiation test device, respectively.

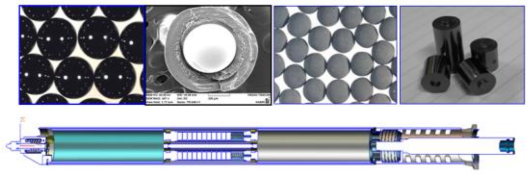

Fig.2. Photographs of kernel, TRISO, graphite compact, and irradiation test device.

3. Results

UO_2 kernels were prepared with the optimum experimental processes that consist of preparation of spherical liquid-ADU droplets by M-EG (modified-external gelation) method, AWD process, and thermal treatment process. Fig.3 shows the image of liquid-ADU droplets, dried-ADU gel particles, UO_3 particles, and spherical UO_2 kernels which were obtained from KAERI process.

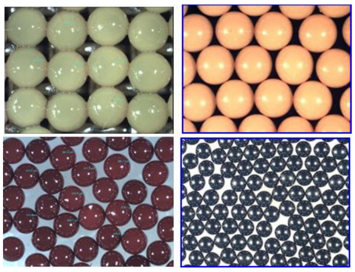

Fig.3. Photographs of liquid droplets, dried-ADU gel, UO_3 particles, and UO_2 kernel.

The internal structures of UO_2 kernels obtained from the original and the improved washing and sintering conditions are presented in Fig. 4 which shows microscopic images at the horizontal cross sections of final sintered-UO_2 kernels. The original sintering temperature was 1600℃ and it was increased to 1700℃ and their washing conditions were different. The state of internal structure of final UO_2 kernel was improved by changing experimental conditions. Pores and the darkish circle in center of the kernel were nearly eliminated, and the grain size distribution inside the kernel was improved.

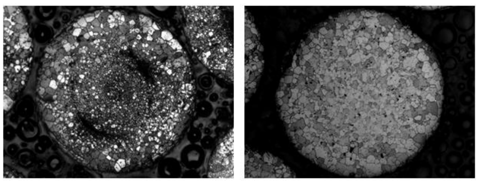

Fig.4. Cross section images of UO_2 kernel from washing and sintering condition change.

On the other hand, the irradiation test using KAERI test rod was started in August 2013 to March 2014 in the HANARO. The fuel is comprised of 480um diameter(average) LEU fuel kernels with an enrichment of 4.5wt%U-235, coated with TRISO coatings(i.e., a buffer layer, a layer of silicon carbide sandwitched between two pyrolytic carbon layers, IPyC and OPyC) to make up the 900um TRISO-coated fuel particles. Table 2 presents the general specifications of the UO_2 kernels that were irradiated at HANARO.

Table 2. UO_2 kernels properties.

Properties	Design value	Measured value	Remarks
Kernel			
- Diameter (μm)	480 ± 30	477.84	average
- Density (g/cm³)	10.65 ± 0.25	10.68	average
- U-235 enrich. (wt%)	4.5 ± 0.10	4.504	chemical analysis
- O/U ratio	2.00 ± 0.01	2.003	average
- Total uranium (wt%)	≥ 87.0	88.13	cal. value
- Sphericity (aspected ratio)	< 1.2	≤ 1.04	average

The maximum power of fuel compact is estimated to be 56W at 25.06 EFPD, and the maximum power of particle is 215.4 mW in rod. And, the maximum discharged burn-up is about 37,344 MWD/MTU(3.99 FIMA)[5].

For the post irradiation examination inspection of the irradiated TRISO fuels, the compact was cut with the low speed diamond cutter and the resin mounting and polishing works were carried out. Fig. 5 shows the SEM image of UO_2 kernel after HANARO irradiation test. The internal structures of irradiated compact were investigated by inspecting the grains, pores, and cracks in the kernel or coating layers. The kernels appeared to maintain their shapes, but grain sizes were decreased in general.

Also, Fig. 6 shows the EPMA results of UO_2 kernel after HANARO irradiation test. Analysis of the internal structures and fission products for the irradiated UO_2 kernel is in progress. The numbers and colors in these figures are qualitative and show relative intensities only, and more detail analyses will be progressed.

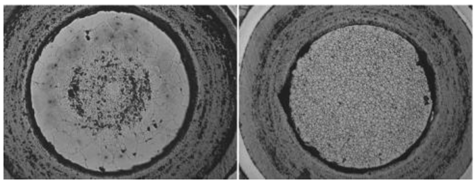

Fig.5. SEM photographs after irradiation test.

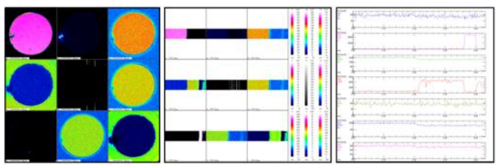

Fig.6. EPMA images of irradiated kernels.

4. Conclusions

This study presents;

(1) UO_2 kernel preparation using modified sol-gel process
(2) Internal structure improvement of UO_2 kernel through washing condition and sintering temperature control
(3) PIE results of the kernels in the irradiation test of KAERI TRISO fuels at HANARO.

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