

Effect of sintering condition on the grain growth of Cr₂O₃ doped UO₂ pellets

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1. Introduction

In order to enhance the nuclear fuel safety, it may be the essential solution to reduce the fission gas release (FGR) and to increase the resistance to PCI (pellet-cladding interaction). The recent study is focused on the modification of the microstructure of fuel pellet to increase fission gas retention and to reduce PCI phenomenon. PCI improvement can be achieved by enlarging the pellet grain size and enhancing the fuel deformation.

A large grain pellet can minimize fission gas release and deform easily at an elevated temperature. So, the recent development of nuclear fuel pellet materials is mainly focused on the large grain pellets. The various methods of fabrication processes for large grain UO₂ pellets have been investigated extensively. Those parameters include the additives, sintering temperature, sintering time, sintering atmosphere, and so on [1-3]. Among those, additive doping technology has been widely studied because this technology can significantly increase the grain size and is quite compatible with the industrial pellet fabrication process.

Cr-doped UO₂ pellet is one of the promising candidates for PCI remedy. It was shown that the grain size and softness of UO₂ pellets could be enhanced by doping Cr or Cr compound in UO₂. Various in-pile test results revealed that the PCI properties were enhanced considerably [4]. In the sintering process of Cr-doped UO₂ pellet, it was known that tight adjusting of sintering atmosphere is most important to achieve large grain pellet. The relevant research revealed that the doped Cr₂O₃ became liquid phase in optimized oxygen potential and that liquid phase promoted the grain growth. Recently, KAERI has shown that grain size of Cr-doped UO₂ pellet could be more enlarged by adjusting process parameters [5].

In this paper, we introduced a sintering process which can form a liquid phase for a large grain growth in Cr₂O₃ doped UO₂ pellet. The study on the effect of dwell time in H₂ atmosphere during sintering process on the grain structure of sintered pellet is also a part of this work.

2. Experimental

Cr₂O₃ and UO₂ powder mixture were prepared by blending two powders for 4h in a tumbling mixer. The contents of the Cr₂O₃ were determined to be 1500 ppm

in weight. The UO₂ powder used in this work was produced through the ADU (Ammonium Di-Uranate) process. The prepared Cr₂O₃ containing UO₂ powder mixtures were pressed into green pellets at 3 ton/cm². The green pellet were sintered at 1700 °C for 1h in pure for H₂. After that, the sample pellets were further sintered at the same temperature for 4h under the atmospheres of 3 vol%CO₂+H₂. In order to investigate the effect of dwell time in H₂ atmosphere during sintering process on the grain structure of sintered pellet, it changed to vary dwell time in H₂ atmosphere. The process names and sintering atmosphere in sintering process of Cr₂O₃-doped UO₂ pellets were listed in Table 1.

The sintered density of the UO₂ pellets was measured by the water immersion method. The pellets were sectioned axially, ground and polished. The polished pellets were thermally etched at 1290 °C in carbon dioxide gas in order to examine their grain boundaries. The grain structures were examined by an optical microscope and the grain size was determined by the linear intercept method.

3. Results

Cr₂O₃ doped UO₂ pellets were fabricated by two-step sintering process which was changed in sintering gas H₂ to 3vol%CO₂ during isothermal sintering. In two-step sintering process, the oxygen potential of H₂ gas at 1700 °C is low enough to reduce initially doped Cr₂O₃ and the solubility of metallic Cr in UO₂ is very low. It means that the amount of Cr that can form a liquid phase for grain growth increase. In the oxygen potential of 3vol%CO₂-H₂ gas at 1700 °C, the Cr is oxidized to Cr₂O₃ because the Cr₂O₃ is stable phase at this potential. In Cr-O phase diagram, Cr and Cr₂O₃ coexist in equilibrium up to the eutectic temperature of 1660 °C and a liquid phase is formed in mixture composition of Cr and Cr₂O₃ at sintering temperature [7].

Table 1. Sintering atmosphere applied in this study.

Process name	Sintering atmosphere	
	Dwell time(h)	
	H ₂	3%CO ₂ -H ₂
H-1	1	4
H-2	2	4
H-3	3	4
H-4	4	4

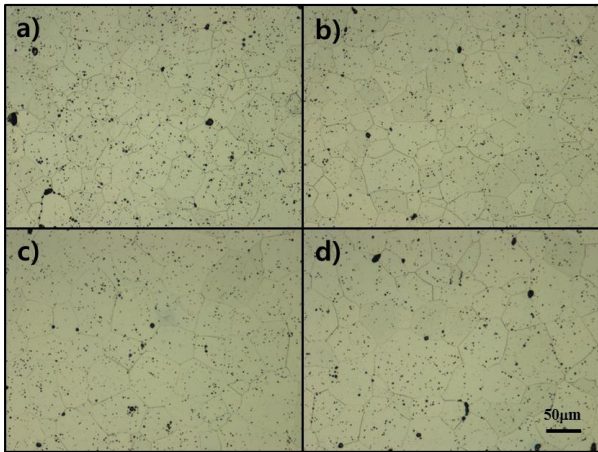


Fig. 1 Optical micrographs of sintered Cr_2O_3 -doped UO_2 pellets. Pellets were sintered by (a) H-1, (b) H-2, (c) H-3, (d) H-4 process, respectively.

Fig. 1 shows the grain structure evolution according to the sintering condition of the Cr_2O_3 doped UO_2 sintered pellets. The measured grain size H-1, H-2, H-3 and H-4 were 33, 36, 43 and $48\mu\text{m}$, respectively. Fig. 2 shows the variations of grain size as a function of dwell time in a hydrogen atmosphere during sintering process in Cr_2O_3 doped UO_2 pellets. As the dwell time increases in a hydrogen atmosphere, the grain size of sintered pellet is increased. This grain size variation reveals that the grain growth of the pellets was deeply influenced by the dwell time in a hydrogen atmosphere during sintering process. A sufficient period of time for the reduction of Cr_2O_3 into Cr is needed to obtain large grain size of pellet. It means that the rate of reduction of Cr_2O_3 into Cr is very slow in a hydrogen atmosphere at sintering temperature. It seems that the hydrogen slowly is diffused into the interior of the pellet due to densification of pellet.

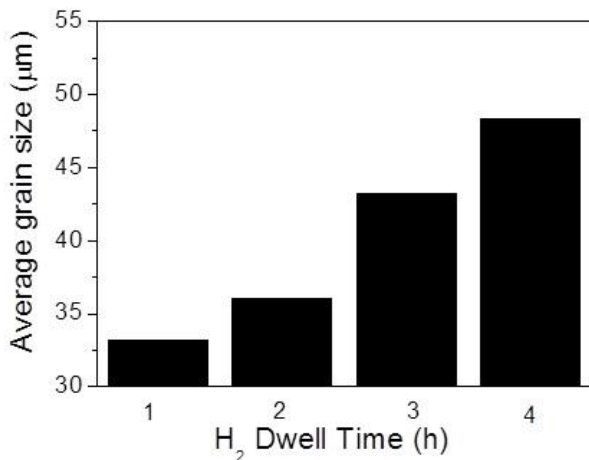


Fig. 2 Grain size variation as a function of H_2 dwell time during sintering process in Cr_2O_3 doped UO_2 pellets.

3. Conclusions

In this paper, Cr_2O_3 doped UO_2 pellets were fabricated by two-step sintering process. The grain growth of pellet is related to dwell time in a hydrogen atmosphere during sintering process. In order to obtain large grain in pellet, it is important to increase amount of Cr that can form a liquid phase for grain growth by increasing dwell time in a hydrogen atmosphere during sintering process.

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REFERENCES

- [1] J. B. Ainscough, B. W. Oldfield and J. O. Ware, *J.Nucl.Mater.*, 49, 117(1973).
- [2] R. N. Singh, *J.Nucl.Mater.*, 64, 174(1977).
- [3] K. W. Kang, J. H. Yang, J. H. Kim, Y. W. Rhee, D. J. Kim, K.S. Kim and K. W. Song, *J.Nucl.Sci.Tech.*, 45, 1150(2008).
- [4] K. W. Kang et al., *J.Nucl.Sci.Technol* 47(2010) 304-307.
- [5] L. Bougeois, Ph. Dehaut, C. Lemaignan, A. Hammou, *J.Nucl.Mater.*, 297(2001)313.
- [6] J. H. Yang et al., *J. Nucl. Mater.* 429(2012)25-33.
- [7] N. Y. Toker, L. S. Darken, A. Muan, *Metallurgical Transactions B* 22B (1991).