

Effect of Heating Rate on the Sinterability of UO_2 -(5~20wt%) CeO_2

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1. Introduction

A MOX(Mixed Oxide) pellet is considered as a major fuel for the next generation nuclear power reactors. It is generally known that the compositions, sintered density and microstructure of the MOX pellet are dominant factors for maintaining a longer cycle operation in nuclear power reactors [1].

In this work, CeO_2 powder being used as surrogate of PuO_2 , owing to its similar high temperature material properties, the effect of the heating rate on the $(\text{U,Ce})\text{O}_2$ pellet as a function of the CeO_2 content(5, 10, 15 & 20wt%) under an oxygen(CO_2) atmosphere is investigated. Heating rate is an important control parameter of the sintered density and microstructures[2].

2. Methods and Results

2.1 Experimental method

4 different additive contents of CeO_2 (UO_2 -5wt% CeO_2 , UO_2 -10wt% CeO_2 , UO_2 -15wt% CeO_2 and UO_2 -20wt% CeO_2) were prepared under the same preparation condition as follows. Each $(\text{U,Ce})\text{O}_2$ powder is mixed for 1 hr in a mixer. And the mixed powder is milled to minimize the particle size and to homogenize the powder mixture, 4 hrs in a Dynamic Mill(DM) device. The DM jar revolves at 25 rpm. A zirconia ball (dia. 8 mm) as milling media was loaded into the jar with 70% of the volume of the jar. A sample size is 50g of UO_2 - CeO_2 powder mixture. Green pellets were prepared by a hydraulic press with a compaction pressure of 300 MPa. These green pellets were sintered at 1500°C for 4 hrs under a CO_2 atmosphere with various heating rates(0.5~8°C/min). Sintered density and grain size of the $(\text{U,Ce})\text{O}_2$ pellets were determined by the immersion method with water and the linear intercept method, respectively. The shrinkage of these pellets was measured with a thermo-mechanical analyzer, TMA-92 device (Setaram, France).

2.2 Results and discussion

Fig. 1 and Fig. 2 show the sintered density and the grain size of the $(\text{U,Ce})\text{O}_2$ pellet for 4 different content of CeO_2 as a function of the heating rates under a CO_2 atmosphere, respectively. As shown in Fig. 1, the sintered density of the $(\text{U,Ce})\text{O}_2$ pellet decreased monotonically as the heating rate increased, regardless of CeO_2 content. Fig.

2 shows that the grain size of the $(\text{U,Ce})\text{O}_2$ pellet decreased with an increasing heating rate. But over the heating rate of 2°C/min, the grain size shows a saturated pattern. Also the grain sizes of these pellets decreased with increasing CeO_2 content under the same heating rate condition. As a result, a lower heating rate is desirable to create both a larger grain size and a higher sintered density of the $(\text{U,Ce})\text{O}_2$ pellet under these sintering conditions. However, the grain size of $(\text{U,Ce})\text{O}_2$ pellet is saturated above 15wt% of the content of CeO_2 . Fig. 3 shows the representative microstructures of the $(\text{U,Ce})\text{O}_2$ pellet with 4 different contents of CeO_2 as a function of the heating rates(0.5 and 2.0°C/min)

Fig. 4 shows densification curves of the $(\text{U,Ce})\text{O}_2$ pellet as a function of the heating rate. And also Table 1 lists the solid solution formation temperature, T_{SSF} , of the $(\text{U,Ce})\text{O}_2$ pellet. As shown in Fig. 4 and Table 1, starting point of T_{SSF} increased with increasing heating rate, but it decreased as the additive content of CeO_2 increased. Meanwhile, the ending point of T_{SSF} increased with an increasing both the additive CeO_2 content and the heating rate. From this result, it is thought that the wider temperature range of solid solution formation is required as the additive content of CeO_2 increased. But for above 15wt% of the CeO_2 content these densification curves show a saturated pattern.

3. Conclusion

Results of the experiments described in this work lead to the following conclusions :

- The sintered densities and grain sizes of $(\text{U,Ce})\text{O}_2$ pellet decreased with both an increasing heating rate and increasing additive content of CeO_2 . But over the heating rate of 2°C/min, the grain size of $(\text{U,Ce})\text{O}_2$ pellet is little affected by the heating rate and the additive CeO_2 content.
- The starting point of the solid solution formation increased with an increasing heating rate, but its starting point decreased as the additive content of CeO_2 increased. However, the ending point of the solid solution formation increased with increasing both the additive CeO_2 content and the heating rate.
- These results show a saturated pattern for above 15wt% of the CeO_2 content.

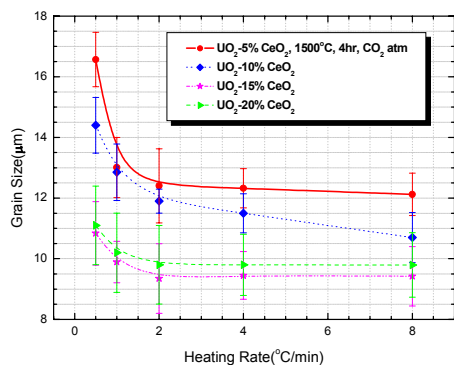


Fig. 1 The sintered density of the (U,Ce)O₂ with four different CeO₂ content as a function of the heating rate.

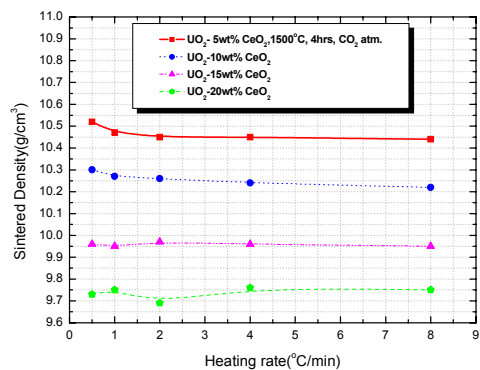


Fig. 2 The grain size of the (U,Ce)O₂ with four different CeO₂ content as a function of the heating rate.

Table 1. Solid solution formation temperature of (U,Ce)O₂ as a function of the heating rate.

Content	Solid Solution Formation Temperature (T _{SSF})							
	5(wt%CeO ₂)		10(wt%CeO ₂)		15(wt%CeO ₂)		20(wt%CeO ₂)	
	start	end	start	end	start	end	start	End
0.5	900	1150	850	1210	830	1250	810	1250
1	920	1200	870	1250	855	1270	845	1280
2	940	1230	920	1280	880	1290	865	1290
4	980	1270	950	1300	920	1320	900	1270
8	1010	1270	970	1300	935	1320	910	1305

REFERENCES

- [1] IAEA-TECDOC-584(91-021-02), February 1991
 [2] Randall M. German, "Sintering Theory and Practice," John Wiley & Sons, Inc., (1996) 171

Acknowledgement

This work has been carried out under the Nuclear R & D program by the Ministry of Science and Technology (MOST), Korea.

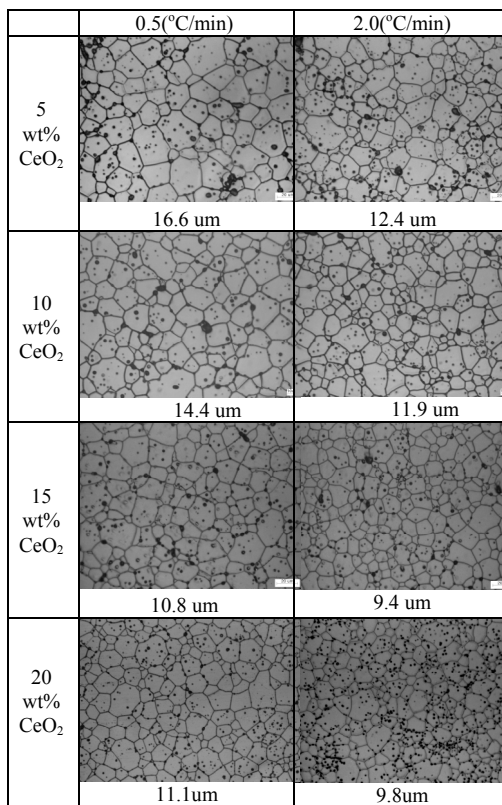


Fig. 3. Microstructures of the (U,Ce)O₂ with 4 different CeO₂ content as a function of the heating rate.

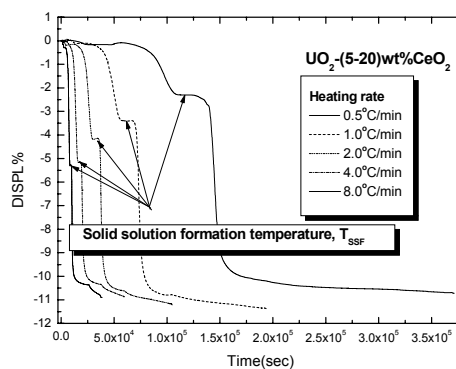


Fig. 4 The shrinkage of the (U,Ce)O₂ with four different CeO₂ content as a function of the heating rate