Effect of Heating Rate on the Sinterability of (U, Ce)O$_2$[CeO$_2$; 5~15wt%]

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1. Introduction

MOX(Mixed Oxide) pellet(UO$_2$-PuO$_2$) 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 (U,Ce)O$_2$ pellet as a function of the CeO$_2$ content(5, 10 & 15wt%) 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

3 different additive content of CeO$_2$(UO$_2$-5wt%CeO$_2$, UO$_2$-10wt% CeO$_2$ and UO$_2$-15wt%CeO$_2$) were prepared under the same preparation condition as follows. Each (U,Ce)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. Zirconia ball (dia. 8 mm) as milling media was loaded into the jar with 70% of the volume of the jar. 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 (U,Ce)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 (U,Ce)O$_2$ pellet for 3 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 (U,Ce)O$_2$ pellet decreased monotonically as the heating rate increased, regardless of CeO$_2$ content. Fig. 2 shows that the grain size of the (U,Ce)O$_2$ pellet decreased with an increasing heating rate. But above 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 (U,Ce)O$_2$ pellet under these sintering conditions. Fig. 3 shows the representative microstructures of the (U,Ce)O$_2$ pellet with 3 different content of CeO$_2$ as a function of the heating rates(0.5 and 2.0^{o}C/min)

Fig. 4 shows densification curves of the (U,Ce)O$_2$ pellet as a function of the heating rate. And also Table 1 lists the solid solution formation temperature, T$_{SSF}$, of the (U,Ce)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.

3. Conclusion

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

- The sintered densities and grain sizes of (U, Ce)O$_2$ pellet decreased with both an increasing heating rate and increasing additive content of CeO$_2$. But the above the heating rate of 2°C/min, the grain size of (U,Ce)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.

REFERENCES

[1] IAEA-TECDOC-584(91-021-02), February 1991
[2] Randall M. German, "Sintering Theory and Practice," John Wiley & Sons, Inc., (1996) 171
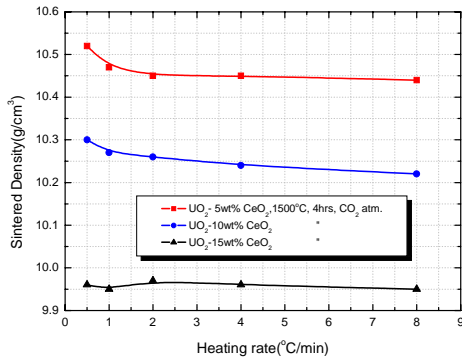
Fig. 1 The sintered density of the $(U,Ce)O_2$ with three different CeO_2 content as a function of the heating rate.
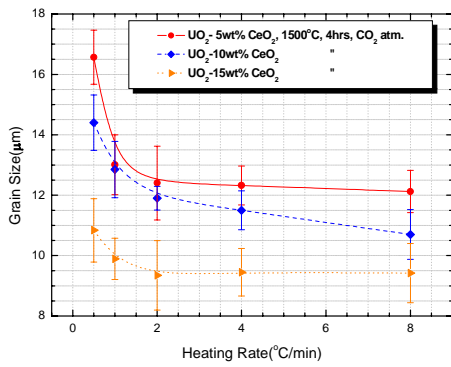
Fig. 2 The grain size of the $(U,Ce)O_2$ with three different CeO_2 content as a function of the heating rate.
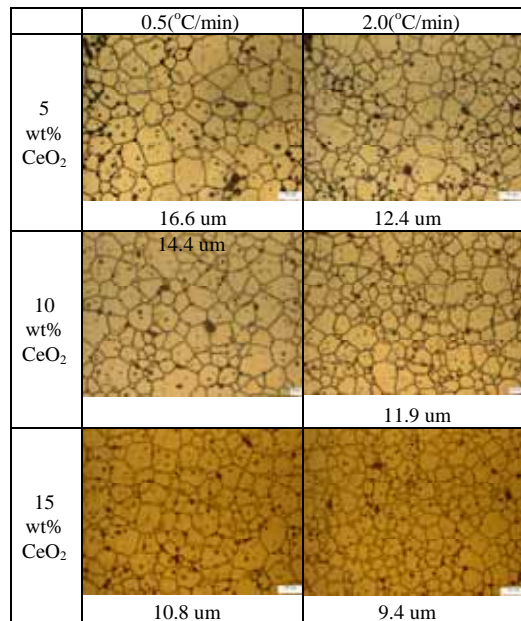

Fig. 3. Microstructures of the $(U,Ce)O_2$ with 3 different CeO_2 content as a function of the heating rate.
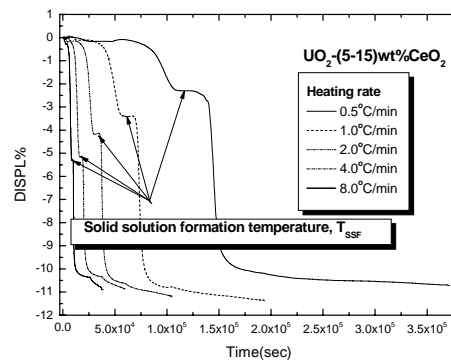

Fig. 4 The shrinkage of the $(U,Ce)O_2$ with three different CeO_2 content as a function of the heating rate

Table 1. Solid solution formation temperature of $(U,Ce)O_2$ as a function of the heating rate.

Content heating rate	5(wt%CeO_2)		10(wt%CeO_2)		15(wt%CeO_2)	
	Solid Solution Formation Temperature(T_{SSF})					
	start	end	start	end	start	end
0.5	900	1150	850	1210	830	1250
1	920	1200	870	1250	845	1280
2	940	1230	920	1280	865	1290
4	980	1270	950	1300	900	1270
8	1010	1270	970	1300	935	1320