Liquid phase sintering of Cr_2O_3-doped UO_2

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1. Introduction

Recently, the Cr_2O_3 doped UO_2 pellet has been intensively investigated in terms of fuel fabrication and irradiation tests [1]. It is supposed that the Cr_2O_3 doped UO_2 fuel is more resistant to PCI failure. The Cr_2O_3 doped UO_2 pellet is known to have a large grain size, compared to undoped UO_2 pellet [2]. The grain growth mechanism in the Cr_2O_3 doped UO_2 is changed depending on the Cr_2O_3 content and sintering gas. The liquid phase sintering has been reported in the specific gas environment [2]. In this study, we have investigated the liquid phase sintering in the Cr_2O_3-doped UO_2 using detailed ceramography.

2. Experimental method

The UO_2 powder used in this work was produced through the IDR process. Mixture of IDR-UO_2 and Cr_2O_3 powder was wet ball-milled in order to homogeneously disperse the additive. The green pellets used for sintering experiments were obtained by pressing the milled powder under the pressure of 3 ton/cm^2.

Sintering was performed at 1700°C for 4h in mixed gas of CO_2 and to H_2. The ratio of CO_2 to H_2 in the mixing gas was 1.6%. The heating and cooling rate was 5K/min.

The microstructure of polished section of sintered pellets was observed with an optical microscope. The detailed morphological observations were made by scanning electron microscope. Identification of liquid phases was performed using energy dispersive X-ray analysis.

3. Results

Fig. 1 shows the equilibrium oxygen potential of CO_2/H_2 mixing gas with temperature together with equilibrium Cr-Cr_2O_3 reaction line [3]. In this figure, the *R* denotes the volume ratio of CO_2/H_2 in %. The increase of *R* extends the limit of the Cr_2O_3 phase stable region to higher temperature. In addition, when the *R* is properly adjusted, Cr_2O_3 phase are reduced to Cr phase via CrO liquid phase as the temperature increases. The temperature where the liquid phase appears is very close to the conventional sintering temperature of UO_2. This fact makes the liquid phase sintering possible in the Cr_2O_3-doped UO_2 system.

Fig. 2 shows the microstructure of Cr-Cr_2O_3 obtained by annealing the Cr-Cr_2O_3 powder mixture at 1700°C for 4h in 1.6% CO_2-H_2 mixing gas. In this figure, the light color is the Cr-rich phase and dark color is Cr_2O3-rich phase. It clearly confirmed that the Cr-Cr_2O_3 system is eutectic and melted at 1700°C in 1.7% CO_2 containing H_2 atmosphere.

Fig. 3 shows the microstructure of 3wt% Cr_2O_3 doped UO_2 pellet. Fig. 3(a) shows the typical grain structure in which the very large grain size of UO_2 is observed. Fig. 3(b) shows a more detailed structure. There are three kinds of phases in grain boundary. EDAX work reveals that white, gray, dark gray phase are UO_2, Cr-rich, and Cr_2O_3-rich phase, respectively. It implies that large grain growth of UO_2 phase is achieved by mass transport via the can be eutectic liquid phase of Cr-Cr_2O_3 in the sintering process.

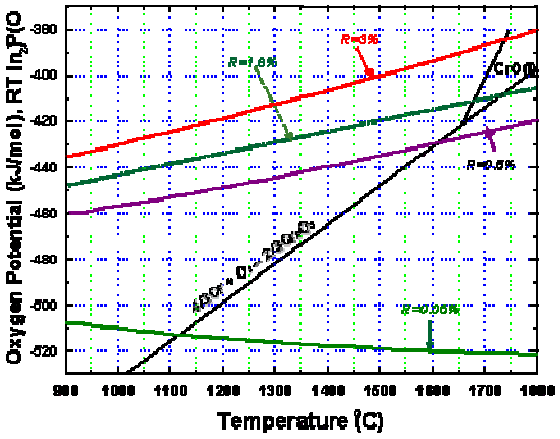


Fig. 1. Temperature dependence of oxygen potentials of sintering atmosphere and chromic oxide

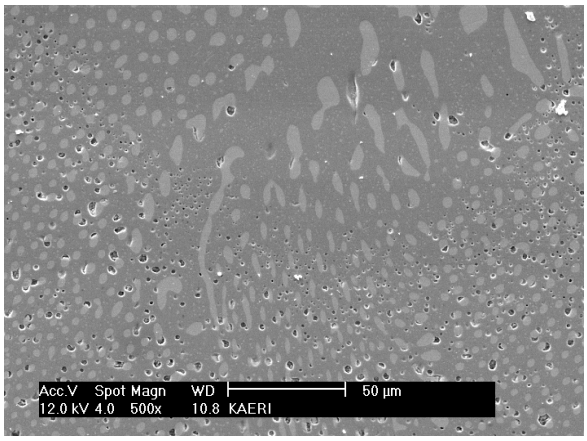
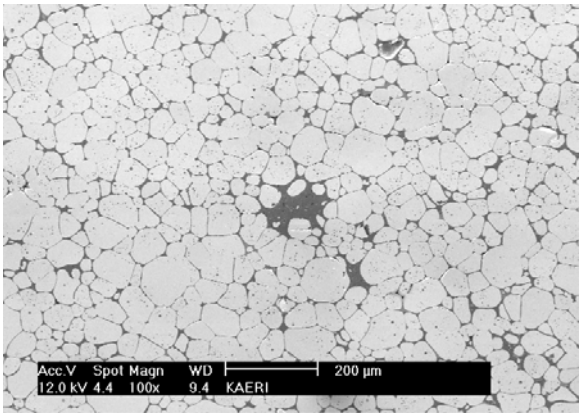
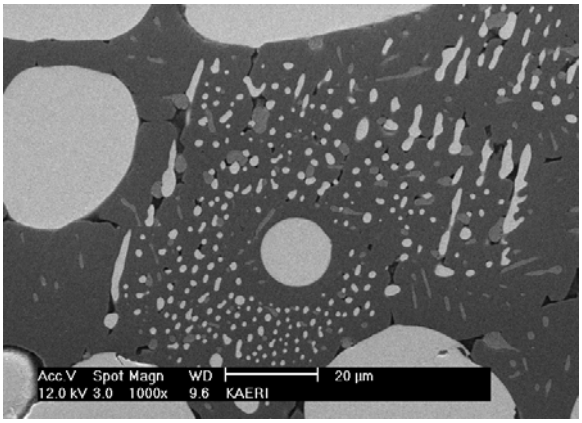


Fig. 2. SEM microstructure of Cr-Cr_2O_3 mixture obtained by annealing at 1700°C for 4h in 1.6% CO_2-H_2 mixing gas.



(a)



(b)

Fig. 3 SEM microstructure of 3wt%Cr_2O_3-doped UO2 pellet obtained by sintering at 1700℃ for 4h in 1.6% CO_2-H_2 mixing gas.

Fig. 4 shows SEM morphology of 0.5wt% Cr_2O_3-doped UO_2 pellet. The liquid phase is formed at along the grain corner of UO_2 phase.

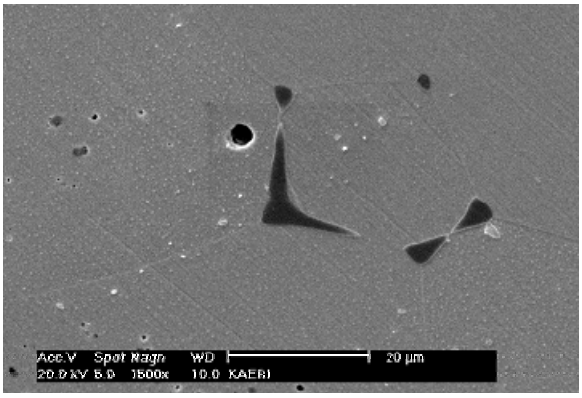


Fig. 3. SEM microstructure of 3wt%Cr_2O_3-doped UO_2 pellet obtained by sintering at 1700°C for 4h in 1.6% CO_2-H_2 mixing gas.

4. Conclusion

Liquid phase sintering in Cr_2O_3-doped UO_2 pellet has been confirmed through detailed ceramography. The large grain growth of UO_2 in Cr_2O_3 doped UO_2 system corresponds to the existence of Cr-Cr_2O_3 eutectic liquid phase at the sintering stage.

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