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Effect of sintering atmosphere on grain boundary structure of Cr_2O_3 doped UO_2 pellets

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1. Introduction

Increasing the burn up may lead to a high duty and a faster and higher power variation such as a higher maximum power or load follow operation. In such operating conditions, the risk of a fuel failure is related to a pellet-clad-interaction (PCI) [1].

Recent development of advanced fuel pellet materials is mainly focused on the large grain pellet which can deform easily at an elevated temperature. There are various process parameters of sintering to increase the grain size of UO_2 pellets. Among those, the technology regarding the doping of additives has been studied widely because this technology can increase the grain size significantly and is quite compatible with an industrial pellet fabrication process. Cr-containing UO_2 pellet is one promising candidate for a PCI solution. It was shown that the grain size and softness of UO_2 pellets could be enhanced by doping Cr or Cr compound in UO_2. Various in-pile tests results revealed that the PCI properties were enhanced considerably [2-4].

In the sintering process of a Cr- doped UO_2 pellet, it was known that tight adjusting of the sintering atmosphere is most important to achieve a large grain pellet. The relevant research revealed that the doped Cr_2O_3 became liquid phase in optimized oxygen potential and that liquid phase promoted grain growth. Since the grain boundary(GB) structure depends on the chemical state of solute atoms in grain boundary, it is expected that the grain boundary shape may be changed depending on the oxygen potential of sintering atmosphere. To observe the effect of oxygen potential on grain boundary structure of the doped pellets, the sintering atmosphere was varied by using CO_2/H_2 gas mixture. The change of oxygen partial pressure during the isothermal sintering is expected to perturb the local concentration of dissolved Cr or liquid phase especially along the grain boundary, thus showing a variety in grain boundary structure and grain growth behavior.

2. Experimental

Cr_2O_3 and UO_2 powder mixtures were prepared by blending two powders for 12h in a tumbling mixer. The UO2 powder used in this work was produced through the ADU (Ammonium Di-Uranate) process. The prepared Cr_2O_3 containing UO_2 powder mixtures were pressed into green pellets with 3 ton/cm^2. The green pellets were sintered at 1700℃ for 10 h. The heating rate to sintering temperature was 300 K/h. The CO_2 and H_2 gas mixtures were used to control the oxygen potential during the sintering process.

The sintered density of the UO_2 pellets was measured by the water immersion method. The pellets were sectioned axially, ground and polished. The polished pellets were thermally etched at 1290℃ in carbon dioxide gas in order to examine their GBs. The grain structures were examined by an optical microscope and the grain size was determined by the linear intercept method. GB structures of the sintered pellets were observed by using scanning electron microscopy (SEM). The elemental composition variations along the grain boundaries or around precipitates were measured by using energy dispersive X-ray spectroscopy (EDS) and standardless quantitative data were obtained by the ZAF correction algorithm of the EDAX Genesis X-ray microanalysis software.

3. Results

Variations in grain size as function of Cr_2O_3 contents and sintering atmospheres of are shown in Fig. 1. The grain size for the pellets sintered in dry hydrogen is irrelevant to the Cr_2O_3. When the CO_2 ratio in the sintering gas was increased, the grain growth was occurred in doped pellets.

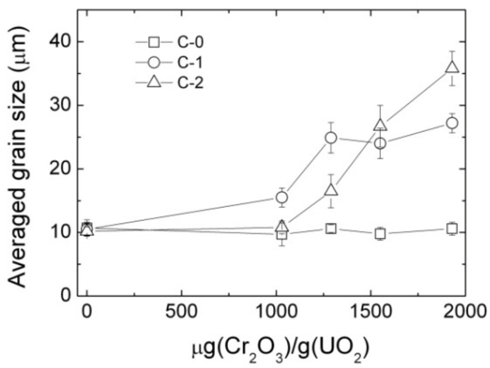

Fig. 1. Grain size variation as a function of doped Cr_2O_3 amounts and sintering atmospheres in Cr_2O_3 doped UO_2 pellets sintered by conventional process.

Detailed observation of GB structure and chemical analysis for Cr_2O_3 doped UO_2 pellets have been conducted by using by SEM and EDS. Fig. 2 shows a microstructure evolution of sintered pellets with an increase of oxygen potential of sintering atmosphere.

As the oxygen potential of sintering gas increased, the grain boundaries became undulated. Since the wiggled grain boundary means the increase of GB energy, formation of the undulated GB indicates that driving force for GB movement is getting higher as the oxygen potential increases. A second interesting feature is the preferential precipitation of Cr-rich phase in GBs and its shape change. Tiny dark precipitates were frequently observed in GBs as denoted by white arrows in Figs. 2(a), (b) and (d). When the oxygen potential is increased, the stitch-like line shape precipitates appeared in the GBs as shown by black arrows in Fig. 2(c) and (d). In addition to this, when the oxygen potential of sintering gas was increased further, the formation of a eutectic liquid phase at the triple junction and GBs were observed as shown by dark gray arrows in Figs. 2(d) and (e). The Cr/U ratios at the selected positions denoted by numbers in Figs. 2(b), (c) and (e) were depicted in Fig. 2(f). The grain boundary morphology was changed from normal flat interface to undulated interface with increasing of oxygen potential of sintering gas. This result might indicate that GB energy is changed with increase of oxygen potential and/or the Cr^{3+} concentration in GBs. Moreover, there is a possibility that grain growth behaviors are synergistically affected by change of interface energy.

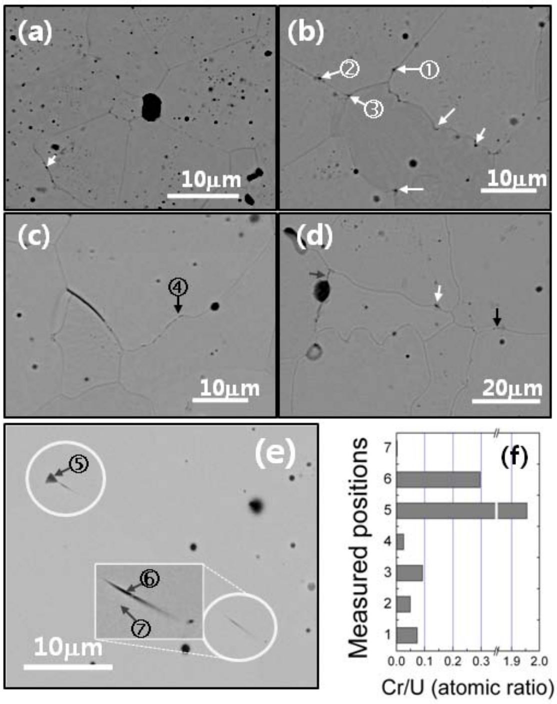

Fig. 2. Microstructure changes according to the increase of oxygen potential of sintering atmospheres in 1920ppm of Cr_2O_3 doped UO_2 pellets.

4. Summary

It was found that the grain interface structure were changed from a flat normal interface to undulated interface via partially faceted interface as the oxygen potential of sintering gas increased. This result indicates that partially faceted and undulated GBs could contribute to the increase of grain growth rate synergistically.

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