Effect of resintering in Cr-doped UO_2 pellets

Jang soo Oh, Jae Ho Yang, Ik Hui Nam, Jong Hun Kim, Dong-Joo Kim, Young Woo Rhee, Keon Sik Kim*

Korea Atomic Energy Research Institute,
(150-1 Dukjin-Dong), 1045 Daedeokdaero, Yuseong-gu, Daejeon-si 305-353, Korea
jangsoo@kaeri.re.kr

1. Introduction

Many researchers are focusing on the development of large-grain UO_2 pellets as they can reduce corrosive fission gas release and thus enhance fuel reliability. It is known that an additive doping technology is effective on the grain growth of UO_2 pellets. Among them, Cr_2O_3-doped UO_2 pellet is one of the promising candidates. To increase the grain size effectively, it is important to control the additive content and sintering atmosphere. Relevant research on the Cr_2O_3 doped UO_2 system revealed that the doped Cr_2O_3 formed a liquid phase under optimized oxygen potential, and those liquid phases promoted the grain growth [1].

In-reactor densification of UO_2 became of major concern in 1972 when previously unknown fuel rod deformations had been observed in several reactor plants [2]. The in-reactor densification of UO_2 pellets has resulted in a shortening of the fuel column, the formation of gaps between pellets in the fuel column and between the pellets and the cladding. The damage imposed operating constraints on some reactors [3].
Evaluation of the in-reactor densification of the fuel pellets is important. The in-reactor densification of fuel pellets is commonly estimated using a resintering test, because In-pile tests are rather prolonged, complex, and expensive.

In this study, we investigated the effect of resintering in Cr_2O_3 doped UO_2 pellets. The test pellets obtained by sintering in a controlled wet atmosphere were resintered in dry hydrogen. The results showed that the grain size as well as pellet density were increased after the resintering. In particular, the grain size was greatly increased after the resintering, and the increase of grain size was discussed in terms of solubility of Cr in UO_2.

2. Experimental

The starting materials were UO_2 powder produced through the ADU (Ammonium Di-Uranate) process. Cr_2O_3-doped UO_2 was prepared by using Cr_2O_3 powders and UO_2 powers. These powders were mixed for 12h using tumbling mixer. The contents of the Cr_2O_3 were cintrolled to be 1200 and 1500ppm in weight.

The prepared Cr_2O_3 containing UO_2 powder mixtures were pressed into green pellets at 3 ton/cm^2. The green pellets were sintered at 1700^{o}C for 6h in flowing mix gas of H_2 and CO_2. In step-wise sintering process, the oxygen potential of sintering gas was increased gradually to control the Cr dissolution rate.

A resintering test of sintered pellets was performed at 1700^{o}C for 24h in flowing H_2. The density of sintered and resinterd UO_2 pellets was measured by a pycnometer. The pellets were sectioned axially, ground and polished. The polished pellets were thermally etched at 1290^{o}C in carbon dioxide 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

Fig. 1 shows pore structure in Cr_2O_3-doped UO_2 pellets (μg(Cr)/g(U)=1200) after (a) sintering and (b) resintering. Fig. 1(a) shows that most of the pore consists of small things, and partially large pores are distributed. The number of pores greatly decreased by resintering in dry H_2 (Fig.1 b). The pore is transformed into lattice vacancies. The vacancies generated in this event migrate to the nearby pores, or to the grain boundaries. Vacancy migration to the grain boundary results in densification of the fuel.

However, in case of Cr_2O_3-doped UO_2 pellets (μg(Cr)/g(U)=1500), pore structure of sintered and resintered pellets is similar(Fig.1 c and d). It seems that because Cr inhibits the vacancy migration with increasing the Cr contents in UO_2 pellets, densification hardly occurred.

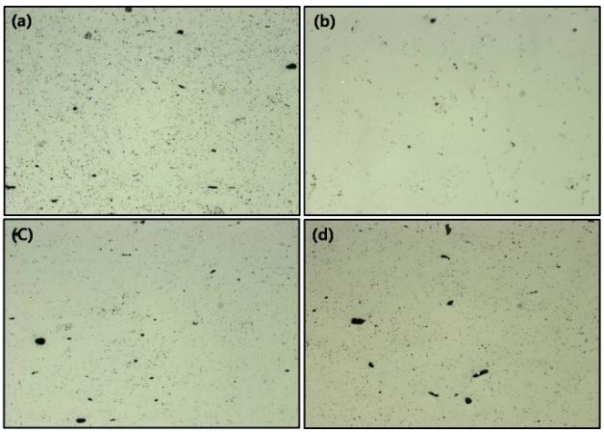

Fig. 1 Pore structure of Cr_2O_3-doped UO_2 pellets after sintering and resintering test

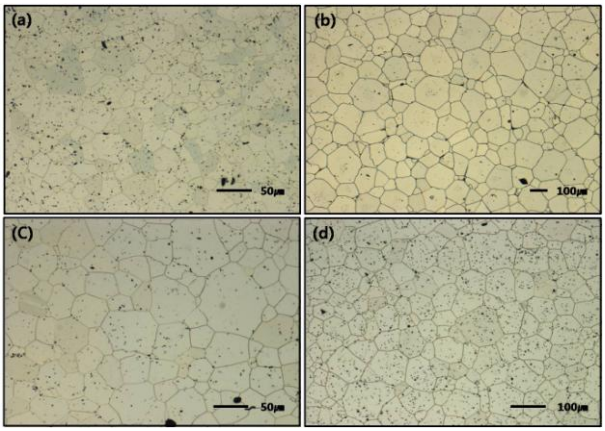

Fig. 2 Grain structure of Cr_2O_3-doped UO_2 pellets after sintering and resintering test

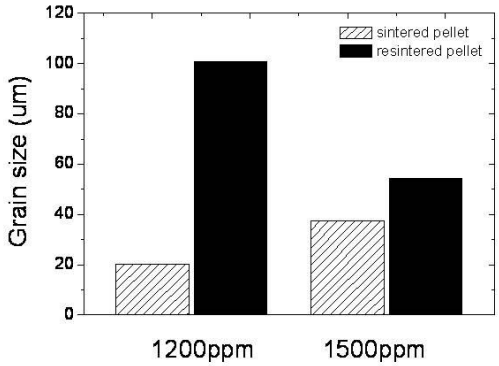

Fig. 3 Comparison of grain size change in Cr_2O_3 doped UO_2 pellets after a resintering test

Fig. 2 and 3 show the change of grain size before and after the resintering test in Cr_2O_3 doped UO_2 pellets. It shows that the grain size was greatly enlarged after resintering. In the case of 1200ppm of the Cr_2O_3 doped UO_2, the grain size was increased to about 5-times larger after resintering in dry hydrogen. Since the normal grain growth curve as a function of time shows a parabolic shape, this increase of grain size after resintering is abnormal.

It is known that the Cr solubility in UO_2 is decreased with a decrease in oxygen potential of the annealing gas. Since the sintering proceeded under wet hydrogen and resintering was under dry hydrogen, it is expected that a part of the dissolved Cr in the sintered UO_2 is precipitated or segregated during the resintering process. Therefore, it is considered that segregated or precipitated Cr plays an important role in the grain growth of pellets during the resintering process.

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