Mitigation of Cr evaporation in Cr-doped UO_2 pellets by controlling the sintering atmosphere

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1. Introduction

The technical issues of high burn up fuel are to reduce the fission gas release and improve the pellet-cladding interaction. In order to mitigate these issues, the recent development of fuel pellet for LWRs is mainly focused on the large grain UO_2 pellets, because a large grain pellet is expected to enhance fuel plasticity at an elevated temperature of transient operation as well as the fission gas retention capability[1-2].

Cr_2O_3-doped UO_2 pellet is one of the promising candidate for the high burn up fuel. 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 [3-4].

In conventional sintering process of Cr_2O_3-doped UO_2 pellets, the oxygen potential of sintering is that the Cr_2O_3 forms liquid phase of CrO near sintering temperature. In that process, it was found that considerable amount of Cr was evaporated during the sintering. Evaporated Cr can react with the refractory materials in a sintering furnace and deteriorate the robustness of furnace. Cr released to environments may be harmful.

In this paper, advanced sintering process which can reduce the evaporation of Cr during the sintering has been investigated. The effect of oxygen potential change at isothermal sintering stage on the pellet properties was examined. Amount of evaporated Cr from a pellet after the sintering was analyzed by using ICP-AES.

2. Experimental

Cr_2O_3 and UO_2 powder mixture were prepared by turbula mixing for 12h and then passed through sieves. The contents of the Cr_2O_3 were determined to be 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 by using two sintering process. First, the power compacts were sintered at 1700^{o}C for 6h in H_2+1.6%CO_2 during the whole sintering process. Second, the green pellets were heated up to 1700^{o}C in pure H_2 and then held for 1h. After that, the sample pellet was further sintered at the same temperature for 5h under the changed atmospheres H_2+5%CO_2.

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^{o}C in carbon dioxide gas 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. Average remaining amount of Cr in sintered Cr-doped UO_2 pellet was analyzed by using ICP-AES method.

3. Results

Fig. 1 shows comparison of conventional and advanced sintering process. Conventional sintering process was maintained in H_2+1.6%CO_2 during whole sintering. The pellet in this process was exposed to CrO liquid phase region for a long time at sintering temperature. However, Advanced sintering process quickly passed liquid phase region by changing H_2+1.6%CO_2 to H_2+5%CO_2 at sintering temperature.

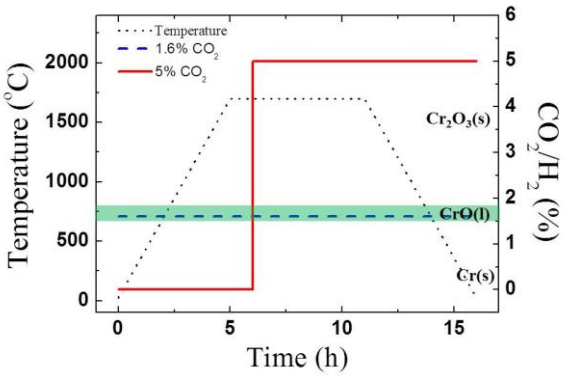

Fig. 1 Compare conventional and advanced sintering process.

Fig. 2 shows remaining amount of Cr in Cr-doped UO_2 pellets. In the case of a mixture of UO_2 and 1500ppm Cr_2O_3, the initial amount of Cr to U ratio is calculated to 1164ppm. Average amount of Cr in sintered Cr-doped UO_2 pellet decreased by 225ppm in conventional process and by 99ppm in advanced process. The results reveal that average amount of Cr evaporation in sintered Cr-doped UO_2 pellet was decreased by 66% in H_2+1.6%CO_2 than H_2+5%CO_2. It is considered that advanced sintering process considerably reduces exposure CrO(l) and lower possibility for sustainment of liquid phase.

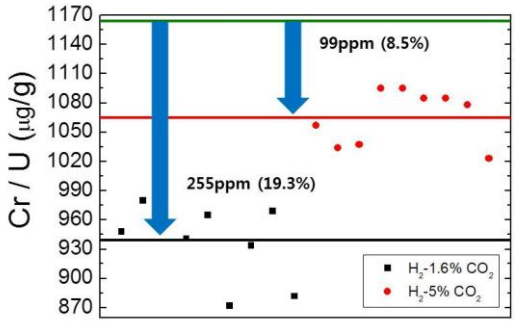

Fig. 2 Comparison of remaining amount of Cr in Cr_2O_3-doped UO_2 pellets sintered in H_2+1.6%CO_2 and H_2+5%CO_2.

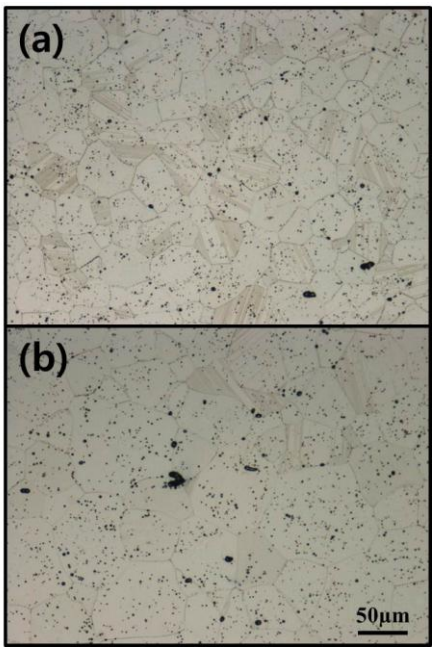

Fig. 3 Grain structure of Cr_2O_3-doped UO_2 pellets sintered in (a) H_2+1.6%CO_2 and (b) H_2+5%CO_2.

Fig.3 shows grain structure of Cr_2O_3-doped UO_2 pellet. The grain size of the advanced pellets was measured to be 48μm. This grain size is about 60% larger than that of conventional pellets. In advanced sintering process, the samples were heated up to 1700°C in pure H_2 and then held for 1h in order to suppress the solubility of Cr in UO_2. The oxygen potential of H_2 at sintering temperature is low enough to reduce initially doped Cr_2O_3 and the solubility of metallic Cr in UO_2 is very low. It can solve the problem that effect of grain growth on the pellets is decreased by reducing amount of liquid phase due to the solubility Cr in UO_2.

4. Conclusions

Advanced sintering process to mitigate the Cr evaporation during the sintering of Cr_2O_3-doped UO_2 pellet was investigated. The oxygen potential change at sintering temperature was considered to reduce Cr evaporation from the pellet.

As a result, average amount of Cr evaporation in sintered Cr-doped UO_2 pellet was decreased, and the grain growth of the pellets was more enhanced than conventional sintering process.

Advanced sintering process has advantage of mitigating of Cr evaporation from the pellets during sintering and enhancing grain growth of the pellets.

ACKNOWLEDGEMENT

This work was supported by the National Research Foundation of Korea (NRF) grant funded by the Korea govemment (MSIP) (No. 201300720)

REFERENCES

[1] Very High Burn-ups in Light Water Reactors, OECD NEA report NAEA No. 6224, (2006).
[2] K.W. Song, Y.H. Jeong, K.S. Kim, J.G. Bang, T.H. Chun, H.K. Kim and K.N. Song, “High Burn-up Fuel Technology in Korea”, Nucl. Eng. and Tech. 40 (2008) 21-36
[3] C. Delafoy, P. Dewes, T. Miles, AREVA NP Cr_2O_3-doped fuel development for BWRs, in : proceedings of International LWR fuel performance Meeting, San Francisco, USA, (2007)
[4] C. Nonon, JC. Menard, S. Lansiart, J. Noirot, S. Martin, GM. Decroix, O. Rabouille, C. Delafoy, B. Petitprez, PCI behavior of chromium oxide doped fuel, in : Proceedings of the International Seminar on PCI with Water Reactor Fuel, Aixen-Provence, France, (2004)