Fabrication and Out-of-pile Test of Large grain UO₂ pellets

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1. Introduction

In the development of a nuclear fuel pellet for LWR, there are two key features for UO_2 fuel performance improvements. The reduction of Fission gas release and Pellet-clad-interaction can enhance the nuclear fuel safety. It can be achieved with large grain sized UO_2 pellets, because a grain size enlargement is expected to enhance fuel plasticity at an elevated temperature of transient operation as well as the fission gas retention capability.

The various methods fabrication processes for large grain UO_2 pellets has been investigated extensively. Among those, additives doping technology has been widely studied due to its effectiveness at increasing grain size and compatibility with an industrial pellet fabrication process.

Korea Atomic Energy Research Institute (KAERI) has been developing the additive-doped large grain UO_2 pellets for PCI remedy [1,2]. For this purpose, KAERI has designed several additives system, developed fabrication processes for additives doped UO_2 pellets and investigated pellet properties for the last several years[3-5].

In this paper, we introduced the fabrication processes for large grain UO₂ pellet by using additives including MnO-Al₂O₃ and Cr₂O₃. The out-of-pile tests were evaluated for additives doped UO₂ pellets. A variety of out-of-pile tests such as creep, oxidation and microstructure analysis were performed for large grain UO₂ pellets.

2. Experimental

2.1. Fabrication of large grain UO₂ pellets

 Cr_2O_3 powder and mixture of MnO and Al_2O_3 powder were selected as additives. The composition of additives are 95MnO-5Al_2O_3(mol%). The contents of the Cr_2O_3 and MnO-Al_2O_3 additives were determined to be 1500ppm and 1000ppm (Mn+Al)/U in weight. These powders were mixed with UO₂, 8wt%-U₃O₈ and 5wt%pore former. The prepared additives containing UO₂ powder mixtures were pressed into green pellets at 3 ton/cm². The green pellets were sintered by using two sintering processes.

The Cr_2O_3 doped UO_2 pellets were sintered at 1700 °C for 4h in pure for H₂. After that, the sample pellets were further sintered at the same temperature

for 1h under the changed atmospheres of 3 vol%CO_2+H_2.

The MnO-Al₂O₃ doped UO₂ pellets were sintered at 1730° C for 4h in H₂. For comparison, pure undoped UO₂ pellets were also prepared.

2.2 Test

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 °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.

The compressive creep tests were performed under 60 MPa at 1450 °C. The tests were conducted in a mix gas of 10% hydrogen and 90% argon gas in order to maintain the constant stoichiometry of the specimens during the experiment.

The oxidation tests were performed at 380° C for 60 min in oxygen atmosphere. And then Microstructure of these samples was examined by optical microscope.

3. Results

Fig.1. Shows the grain structure of the MnO-Al₂O₃ doped UO₂ and Cr₂O₃ doped UO₂ pellets. For a comparison, a grain structure of undoped pure UO₂ pellet was also shown in this figure.



Fig. 1 Optical micrographs of (a) undoped UO_2 , (b) Mn-Al doped UO_2 and (c) Cr_2O_3 doped UO_2 pellet

The grain structure of un-doped UO₂ pellet shows typical UO₂ grain structures and its grain size was measured to be about 9 μ m. The grain sizes of the MnO-Al₂O₃ doped UO₂ and Cr₂O₃ doped UO₂ pellets were measured to be about 40 μ m and 48 μ m. These grain sizes are about 5 times larger than that of undoped UO₂ pellet. The grain sizes are greatly enlarged in MnO-Al₂O₃ and Cr₂O₃ doped UO₂ pellets.



Fig. 2 Compressive creep deformation curves of the developed UO_2 and undoped UO_2 pellet

Fig. 2 shows the compressive creep deformation curves of the large grain doped UO₂ pellets. Creep tests were performed under an initial applied stress of 60MPa and a temperature of 1450 °C. For a comparison, the deformation curve of the undoped UO₂ pellet is presented together. Deformation curves clearly show that the addition of MnO-Al₂O₃ and Cr₂O₃ additives increases the creep deformation of the UO₂ pellets considerably. For example, the deformation strains of the doped UO₂ pellets after 5h are about four to seven times larger than that of the undoped UO₂ pellet. This result reveals that not only the grain size of the UO₂ pellets but also the additives in the UO₂ pellets are important for the compressive creep deformation behavior of UO₂.



Fig. 3 Oxidation test at 380 $^\circ\!\!\!C$ for the developed UO2 in comparison to undoped UO2 pellet

Fig. 3 show the surface oxidation of the large grain doped UO₂ and undoped UO₂ pellets. Oxidation tests were performed at 380 °C for 60 min in oxygen atmosphere. For undoped UO₂, The oxidation proceeded deeply into the interior and many small intergranular cracking and spalling of oxidized grains occurred by fast oxygen diffusion into interior of UO₂. On the contrary, with the additives (MnO-Al₂O₃, Cr₂O₃) doped UO₂ pellets, the oxidation proceeded only thinly on the surface and only large cracks were formed on the surface. This results revealed that the oxidation resistance of UO₂ fuel is enhanced by the grain size enlargement obtained by additives (MnO-Al₂O₃, Cr₂O₃) doping.

3. Conclusions

In this paper, large grain UO_2 pellets were fabricated by the doping of MnO-Al₂O₃ and Cr₂O₃. These pellets have large grain size that are about five times large than that of undoped UO_2 pellet. Large grain pellets can enhance high temperature plasticity and oxidation resistance. Developed additives doped UO_2 pellets can be potential candidates for a PCI remedy.

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REFERENCES

[1] J. B. Ainscough, B. W. Oldfield and J. O. Ware, J.Nucl.Mater., 49, 117(1973).

[2] R. N. Singh, J.Nucl.Mater., 64, 174(1077).

[3] K. W. Kang , J. H. Yang, J. H. Kim, Y. W. Rhee, D. J. Kim, K.S. Kim and K. W. Song, J.Nucl.Sci.Tech., 45, 1150(2008).

[4] K. W. Kang et al., J.Nucl.Sci.Technol 47(2010) 304-307.

[5] L. Bougeois, Ph. Dehaudt, C. Lemaignan, A. Hammou, J.Nucl.Mater., 297(2001)313.

[6] J. H. Yang et al., J. Nucl. Mater. 429(2012)25-33.

[7] N. Y. Toker, L. S. Darken, A. Muan, Metallugical Transactions B 22B (1991).