Thermal properties of MnO-Al_2O_3 doped UO_2 fuel pellets

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1. Introduction

Improving fuel performance to reduce fission gas release and increase resistance to PCI (pellet cladding interaction) is a technical challenge. Both goals can be achieved with large grain microstructures, since grain size enlargement is expected to enhance fuel plasticity as well as fission gas retention capability. Many researchers have investigated the effects of the processing parameters on the grain size of UO_2 pellets. Those parameters include the additives, a higher sintering temperature, a longer sintering time, the sintering atmosphere, and so on[1-3]. 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. The oxide additives (such as Nb_2O_5, TiO_2, MgO, SiO_2 and the corundum type of M_2O_3 with M= Al, Cr, etc.) have been known to promote a grain growth of UO_2[4]. Doped cations in the UO_2 matrix or grain boundary enhance the chemical mobility of the U cation thereby promoting the sintering ability and the grain growth. The solubility of doped cations and their valence state are determined directly by the thermodynamic state of the sintering conditions.

Thermal properties are one of the most important properties of nuclear fuel. Thermal diffusivity and thermal expansion of MnO-Al_2O_3 doped UO_2 fuel pellets is unknown. This paper describes the thermal diffusivity and thermal expansion of MnO-Al_2O_3 doped UO_2 pellets with a large grain size.

2. Experimental

MnO and Al_2O_3 powders were mixed and ball milled for 24h in a jar containing zirconia balls and alcohol. After the ball mill, the suspensions of powder mixture were dried in air. The compositions of additives are $90MnO$-$10Al_2O_3$ and $95MnO$-$5Al_2O_3$(mole%), respectively. The prepared MnO-Al_2O_3 additives were added to UO_2 powder, and these powder mixtures (UO_2-MnO-Al_2O_3) were then mixed for 2hr in a tumbling mixer. The UO_2 powder used in this work was produced through the ADU(Ammonium Di-Uranate) process. The content of the additives was controlled to be 1000 ppm (Mn+Al)/U in weight. Final powder mixtures were pressed into green pellets at 3 ton/cm^2. The green pellets were sintered at 1730℃ for 4 h in flowing H_2 gas. For a comparison, pure un-doped UO_2 pellets were also prepared under the same sintering conditions. The density of the sintered pellets was measured by a water immersion method. The sintered pellets were sectioned axially, ground and polished. The polished pellets were thermally etched at 1290℃ in carbon dioxide gas in order to examine the grain boundaries. The grain structures were examined by an optical microscope. The grain size was determined by a linear intercept method.

3. Results

Fig.1 shows that microstructure of MnO-Al_2O_3, 1000ppm doped UO_2 pellets by an optical microscope. The average grain size of $90MnO$-$10Al_2O_3$ and $95MnO$-$5Al_2O_3$(mole%), 1000ppm doped UO_2 pellet was measured to be 43 ㎛ and 51 ㎛, respectively. These sizes were much larger than that of the un-doped UO_2 pellet.

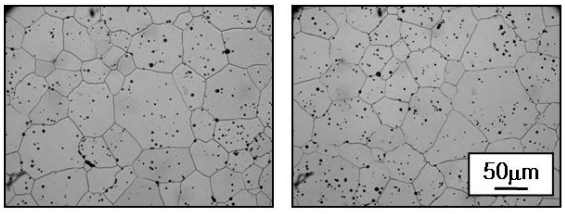

Fig.1 Microstructure of the MnO-Al_2O_3, 1000ppm doped UO_2.
a) $90MnO$-$10Al_2O_3$, b) $95MnO$-$5Al_2O_3$ (mole%)

Fig.2 shows the schematic diagrams of the thermal diffusivity measurement apparatus. The thermal diffusivity of the pellets was measured by the laser-flash method with a Laser-flash apparatus (Netzsch, LFA427). The thermal diffusivity was determined from the rear-surface temperature rise to reach half its maximum value, after the front surface of the sample was heated by the laser beam at various temperatures.

$$\alpha = \frac{\omega}{\pi^2}\frac{L^2}{t_{1/2}}$$

where L is the sample thickness.

Fig.3 shows the variation of the thermal diffusivity as a function of temperature. The measured thermal diffusivity of $MnO\text{-}Al_2O_3$ doped UO_2 pellets with a large grain size is similar to that of un-doped UO_2 tested in KAERI.

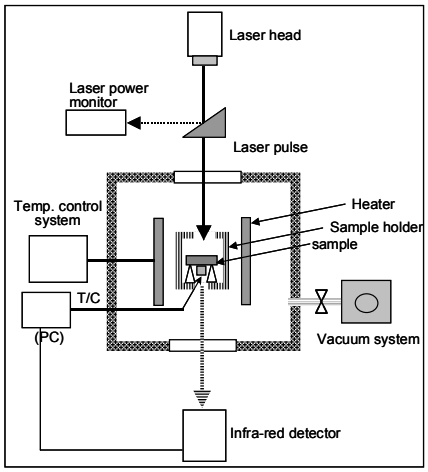

Fig.2. Schematic diagram of the thermal diffusivity measurement apparatus

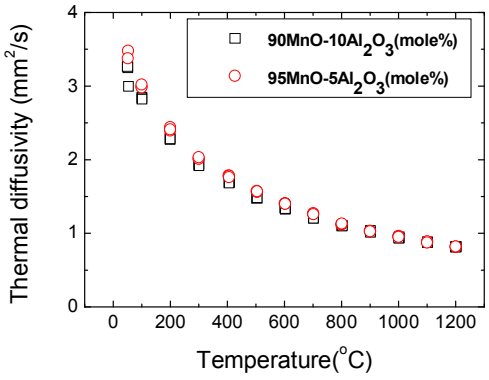

Fig.3. Thermal diffusivity of $MnO\text{-}Al_2O_3$ doped UO_2

Fig.4 shows the variation of the thermal expansion as a function of temperature. The thermal expansion behavior of $MnO\text{-}Al_2O_3$ doped UO_2 pellets was studied by using a push rod type dilatometer (Netzsch, Dil402C). The measured thermal expansion of $MnO\text{-}Al_2O_3$ doped UO_2 pellets is similar to that of un-doped UO_2.

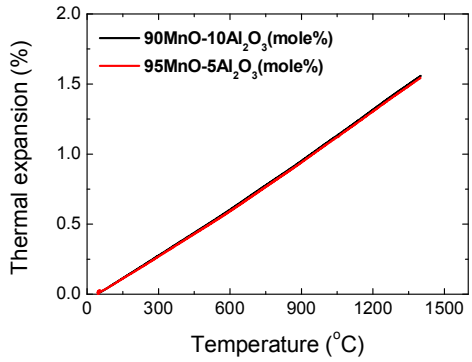

Fig.4. Thermal expansion of $MnO\text{-}Al_2O_3$ doped UO_2

4. Conclusions

The grain growth of $MnO\text{-}Al_2O_3$ doped UO_2 pellet was the largest at 1000 ppm, $95MnO\text{-}5Al_2O_3$(mole%). The average grain size is measured to be 51 ㎛. This size was much larger than that of the un-doped UO_2 pellet. The measured thermal diffusivity and thermal expansion of $MnO\text{-}Al_2O_3$ doped UO_2 pellets with a large grain size is similar to that of un-doped UO_2

Acknowledgements

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REFERENCES

[1] J.B.Ainscough, B.W.Oldfield and J.O.Ware, "Isothermal grain growth kinetics in sintered UO_2 pellets,"J.Nucl.Mater.,49, 117(1973)

[2] R.N.Singh, "Isothermal grain growth kinetics in sintered UO_2 pellets," J.Nucl. Mater., 64,174(1977)

[3] K.W.Kang, J.H.Yang, J.H.Kim, Y.W. Rhee, D.J.Kim, K.S.Kim and K.W.Song, "Improvement of UO_2 pellet properties by controlling the powder morphology of recycled U_3O_8 powder," J.Nucl.Sci. Tech., 45, 1150(2008)

[4] K.W.Song, K.S.Kim, K.W.Kang, and Y.H.Jung, "Effects of Nb_2O_5 and oxygen potential on sintering behavior of UO_2 fuel pellets,"J.Kor.Nucl.Soc., 31, 335(1999)

[5] I.H.Jung, Y.B.Kang, S.A.Decterov, and A.D.Pelton, "Thermodynamic evaluation and optimization of the $MnO\text{-}Al_2O_3$ and $MnO\text{-}Al_2O_3\text{-}SiO_2$ systems and applications to inclusion engineering," Metallurgical and Materials Transactions B, **35**,259(2004)