The influence of porosity on thermal conductivity of low-density uranium oxide

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Abstract. In uranium-plutonium mixed oxide (MOX) fuel fabrication, the mitigation of specifications on fuel design is considered from a viewpoint of improvement of economic efficiency. The density of UO_2 pellets was controlled and the thermal conductivities of specimens were measured. The thermal conductivities decrease monotonically with increasing porosity in porosity range from 0.08 to 0.15. On the other hand, significant change of the thermal conductivities were not observed in porosity range from 0.15 to 0.20.

Key Words: thermal conductivity, porosity, density, uranium oxide.

1. Introduction

In uranium-plutonium mixed oxide (MOX) fuel fabrication, the mitigation of specifications on fuel design is considered from a viewpoint of improvement of economic efficiency. The pellet density of the MOX fuel for fast reactors is one of the important specifications in the fuel design and it fluctuates with changes in the properties of raw material powders. As the one of the mitigations of specifications, the tolerance expansion of the density specification is considered. In this consideration, it is necessary to confirm the applicability of the porosity correction equation to the thermal conductivity in a low density region. The relation among a porosity (p), a density (d) and a theoretical density (d_{th}) is described as follows: $p=1-(d/d_{th})$.

In this study, UO_2 pellet was adopted as test specimen for the following reasons. The same porosity correction equation can be applied to the thermal conductivities of UO_2 and MOX **[1]**. The stability of the oxygen to metal (O/M) ratio of UO_2 pellets in the thermal conductivity measurement is superior to that of MOX pellets. The specimens were prepared by a conventional powder metallurgy process and the densities of the specimens were adjusted in the wide range by using crystalline cellulose. The thermal conductivities of these specimens were measured and the applicability of the porosity correction equation to the thermal conductivity in a low density region was evaluated.

2. Experimental

2.1 Preparation of specimens

The specimens used in this study were UO_2 pellets fabricated by the conventional powder metallurgy process. The suitable amount of crystalline cellulose with average grain size of 20 μm was added to the raw powders of UO_2 to control the density of specimens. These powders were blended and were cold-pressed at 4.5 t/cm^2. The compacts were sintered at 1923 K for

3 h under an atmosphere of Ar-5%H_2 mixed gas to make the O/M adjusted to 2.00. ***Table 1*** shows the main characteristics of the specimens.

TABLE 1 MAIN CHARACTERISTICS OF THE SPECIMENS

Specimen No.	Composition	Theoretical density (%TD)	Porosity	Diameter (mm)	Thickness (mm)
UO2-1	UO_2	92.0	0.080	5.51	1.17
UO2-2		90.8	0.092	5.59	1.32
UO2-3		90.6	0.094	5.50	1.45
UO2-4		86.3	0.137	5.50	1.41
UO2-5		81.9	0.181	5.54	1.35
UO2-6		80.4	0.196	5.53	1.36
UO2-9		87.7	0.123	5.61	1.44
UO2-10		84.1	0.159	5.61	1.56

A part of these sintered pellet was crushed and the lattice parameters of them were measured by an X-ray diffractometer (RINT-1100, Rigaku Co. Ltd.). ***FIG.1*** shows the result of X-ray diffraction measurement. From the results of peak patterns and lattice parameters, the O/M of specimens were confirmed to be almost 2.00.

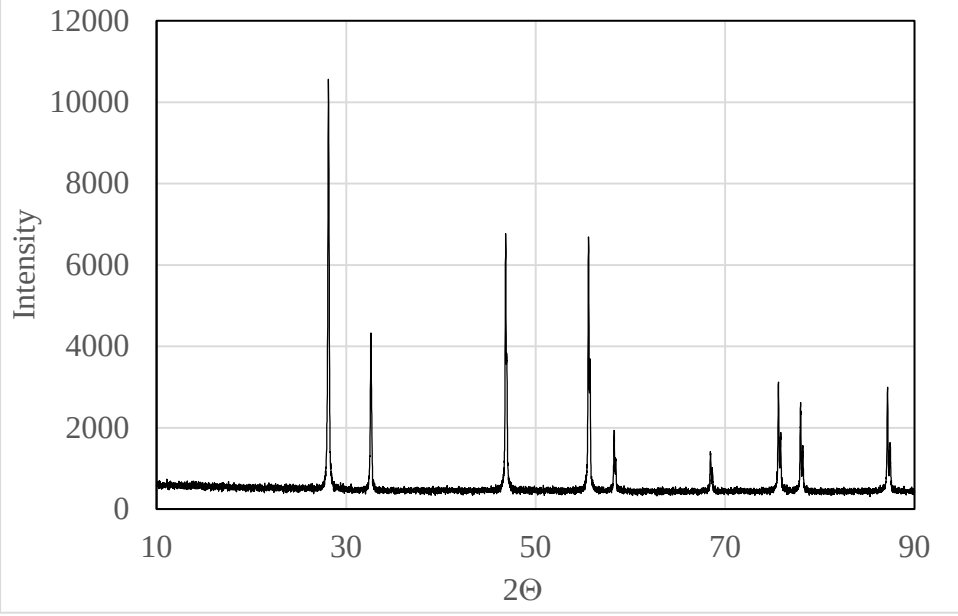

FIG.1 RESULT OF X-RAY DIFFRACTION MEASUREMENT

2.2 Thermal diffusivity measurements and thermal conductivity calculation

The thermal conductivity $\lambda(T)$ was obtained from thermal diffusivity $\alpha(T)$, density $\rho(T)$ and heat capacity $Cp(T)$ by the following equation:

$\lambda = \alpha(T) \cdot \beta(T) \cdot Cp(T)$.

The thermal diffusivities of specimens were measured at temperature range from 650 to 1530 K with a laser flash apparatus (TC-7000UVH, ULVAC-Riko Co. Ltd.). Details of this apparatus have been described elsewhere **[2]**. The recorded data were analysed by the curve-fitting method **[3]**, and thermal diffusivities were derived. Measurements were performed three times at each temperature, and thermal diffusivity was obtained from the average of them.

The densities of specimens at room temperature were measured by the immersion method. The change in density with thermal expansion during the measurement was corrected by the equation reviewed by Carbajo et al. **[1]**. The heat capacities of specimens were calculated by the equation reviewed by Carbajo et al. **[1]**

3. Results and discussion

3.1 Preparation of specimens

The thermal conductivities of specimens are shown in ***FIG.2*** as a function of temperature and density. As shown in ***FIG.2***, the thermal conductivities decreased with increasing temperature and decreasing density. ***FIG.3*** shows the thermal conductivities of specimens as a function of porosity. The porosity p was defined from the theoretical density ρ_{th} and the real density ρ of a specimen as follows:

$p = \frac{\rho_{th}-\rho}{\rho_{th}} = 1 - \left(\frac{\rho}{\rho_{th}}\right)$.

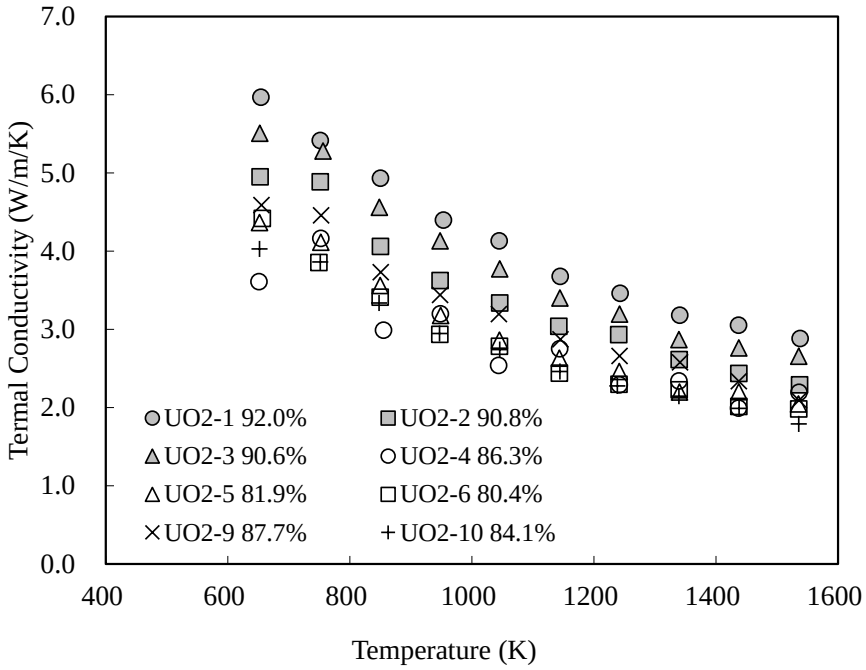

FIG.2 THERMAL CONDUCTIVITIES AS A FUNCTION OF TEMPERATURE

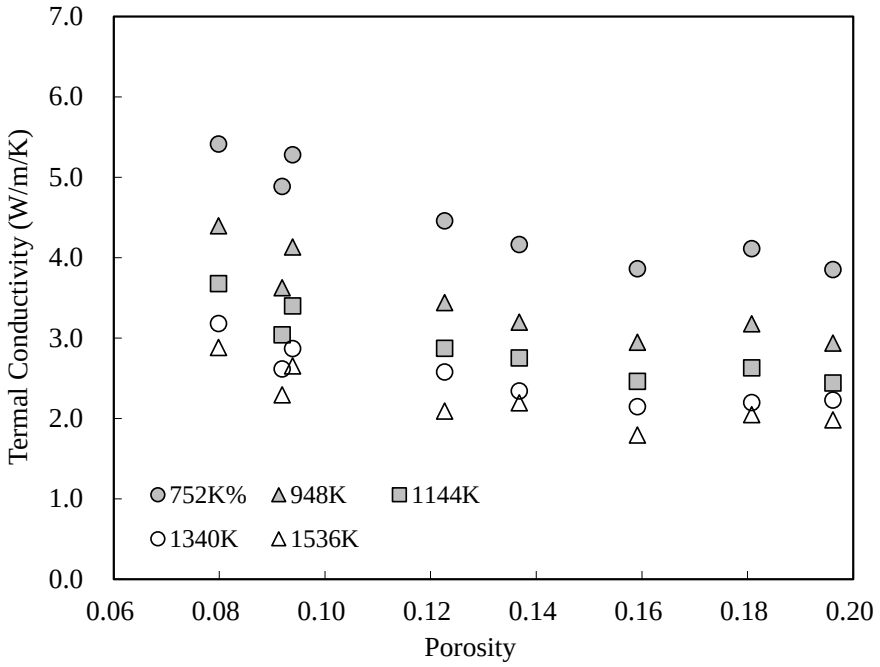

FIG.3 THERMAL CONDUCTIVITIES AS A FUNCTION OF POROSITY

The thermal conductivities decrease monotonically with increasing porosity in porosity range from 0.08 to 0.15. On the other hand, significant change of the thermal conductivities were not observed in porosity range from 0.15 to 0.20.

There have been very few report that the porosity effect on the thermal conductivities of UO_2 and MOX fuel were evaluated experimentally, only Craeynest et al. examined this effect experimentally [**4**]. In their study, the porosity effect on the thermal conductivities were examined in porosity range from 0.05 to 0.28, but the change in the dependency of thermal conductivity was not clearly shown within porosity range from 0.15 to 0.20 because the experimental data in those range was a little.

In order to derive the thermal conductivity λ_0 of a specimen of theoretical density from the thermal conductivity λ of a real specimen of porosity p, a lot of researchers have been using the following porosity correction equations given by Loeb **[5]** and Maxwell and Eucken **[6, 7]**.

$\lambda = F \cdot \lambda_0$,

$F = 1 - \alpha \cdot p$, Loeb equation

$F = \frac{(1-p)}{(1+\beta \cdot p)}$. Maxwell–Eucken equation

Here, F is the porosity correction factor and the values of α and β are correction coefficients of each equations.

Several researchers evaluated this effect theoretically **[8-10]** by using numerical analysis etc. They evaluated α between 1.0 to 4.0 and evaluated β between from 0.5 to 2.6.

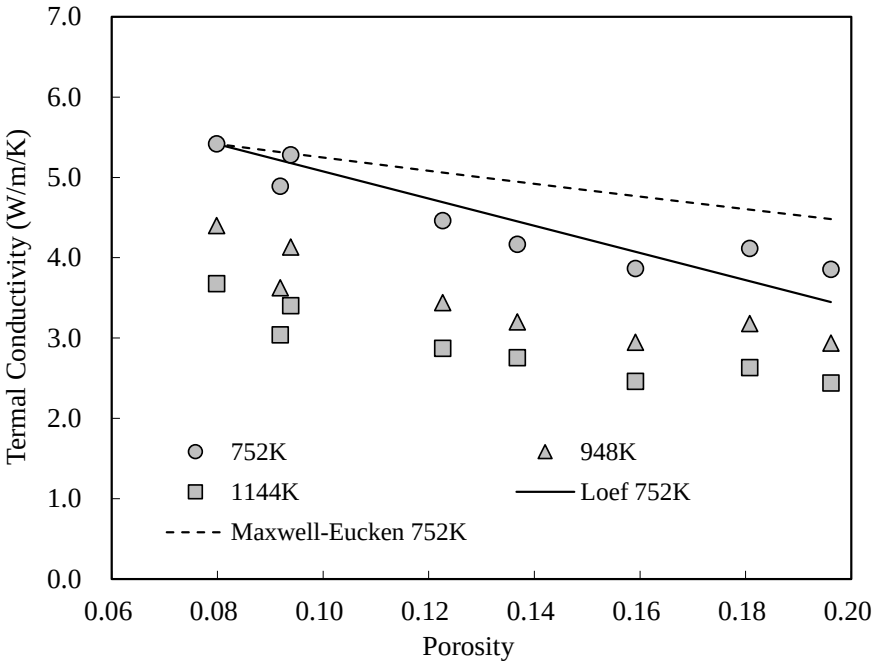

FIG.4 THERMAL CONDUCTIVITIES AS A FUNCTION OF POROSITY

FIG.4 shows the comparison between experimental results and calculation results obtained from the Loeb equation ($\alpha = 2.5$) and Maxwell-Eucken equation ($\beta = 0.5$) at 752 K. The values of α and β were referred from one of the values that the researchers of measuring the thermal conductivity of oxide fuel used. From the results of ***FIG.4***, the agreement between experimental result and theoretical result of Loef equation using $\alpha = 2.5$ is fairly good in the range from in the range from 0.08 to 0.15, but is not so good in the case of theoretical results of Maxwell–Eucken equation using $\beta = 0.5$. In porosity range from 0.15 to 0.20, the influence of porosity on thermal conductivity cannot be expressed by conventional equations. In order to evaluate the influence of porosity on thermal conductivity of low-density uranium oxide, it is necessary to establish other equation.

3. Conclusions

The UO_2 specimens were prepared by a conventional powder metallurgy process and the densities were adjusted in the wide range by using crystalline cellulose. The thermal conductivities of these specimens were measured. The thermal conductivities decrease monotonically with increasing porosity in porosity range from 0.08 to 0.15. On the other hand, significant change of the thermal conductivities were not observed in porosity range from 0.15 to 0.20. The agreement between experimental and theoretical results of Loef equation using $\alpha = 2.5$ is fairly good in porosity range from 0.08 to 0.15 but the influence of porosity on thermal conductivity cannot be expressed by conventional equations in porosity range from 0.15 to 0.20.

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