Mechanical and Thermal Properties of $(U,Pu)O_{2-x}$.

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Abstract. Mechanical and thermal properties of uranium-plutonium mixed oxide (MOX) pellets were evaluated by measured sound speeds of longitudinal and transverse waves in the pellets. Specimens with various porosities were prepared and the sound speeds were measured by the ultrasound pulse-echo method. The results showed that Young's modulus was lower in hypo-stoichiometric composition. In addition, Debye temperature was calculated with the measured sound speeds.

Key Words: MOX, sound speed, Young's modulus, heat capacity.

1. Introduction

Mechanical and thermal properties of MOX are necessary in designing nuclear fuels and simulating their irradiation behaviors in a reactor. Generally, it is understood that the properties are dependent on porosity, Pu content and O/M. Young's modulus is one of the most important properties. Young's modulus of UO_2 has been studied by various researchers. Porosity and temperature are particularly important factors that can decrease Young's modulus of UO_2 compared with other factors such as additives of fission products [1 – 2]. The porosity effect has been thoroughly investigated, and researchers concluded that the porosity, as an effect of density, can be well described by a linear function [3 – 5]. The effect of temperature on Young's modulus of UO_2 was reported for temperatures less than 2073 K [6]. The experimental data were extrapolated to the melting temperature and it was stated that the Young's modulus of UO_2 decreased to approximately 30% of its room temperature value. On the other hand, limited data of Young's modulus of MOX with 20% Pu content was reported [7, 8]. Therefore, this paper aims at evaluating the effect of porosity on Young's modulus of MOX with 20% Pu content. The sound speeds were measured by the ultrasound pulse-echo method [9]. The experimental data included porosity to evaluate its effect quantitatively. From the results, Young's modulus and Poisson's ratio were calculated referring to the literature method [10].

2. Experimental Methodology

Powders of MOX with 20% Pu content which was adjusted in nitrate solution and made into powders by microwave heating was utilized [11]. Celphere produced by AsahiKASEI was blended in the powders to control densities of sintered pellets. These powders were pressed into green pellets of about 5 mm diameter and 7 mm length. The pellets were heat-treated at 1073 K for dewaxing and sintered at 1923 K in an atmosphere of Ar/H_2 mixed gas with moisture. After sintering, the pellets were heat-treated and adjustment of the targeted O/M was made based on the oxygen potential data reported by Kato [12]. List of the specimens which includes porosity is shown in table 1.

TABLE 1: POROSITY, SOUND SPEEDS AND CALCULATED PROPERTIES OF THE SPECIMENS.

	No.1	No.2	No.3	No.4
Porosity	0.1439	0.0986	0.0715	0.0543
Longitudinal wave speed (m/s)	4380.7	4660.6	4838.2	5051.8
Transverse wave speed (m/s)	2407.0	2523.2	2557.3	2650.0
Young's modulus (GPa)	141	163	175	192
Poisson's ratio	0.284	0.293	0.306	0.310
Debye temperature (K)	386			

Parts of sintered pellets were ground by agate mortar for X-ray diffraction. The X-ray diffraction analysis confirmed that the homogenous specimens with controlled Pu content and O/M were obtained such that the lattice structures of the specimens were those of FCC single phase and the lattice parameters of the specimens agree well with Vegard's law in Pu [13] as shown in *FIG. 1*.

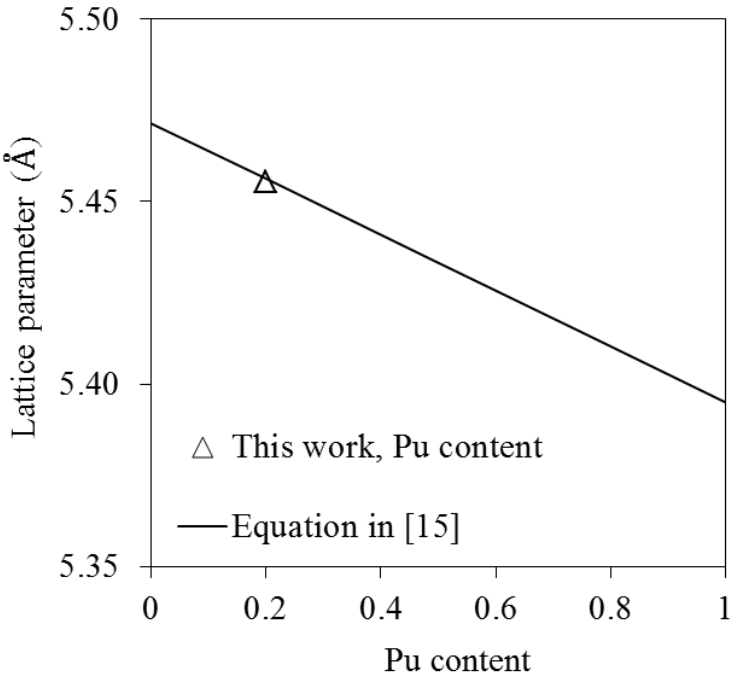

FIG. 1. Lattice parameter evaluated by XRD measurements and calculated by the equation in [15].

Sound speeds of longitudinal and transverse waves in the pellets were measured by the ultrasound pulse echo method with the ultrasonic flaw detector (KJTD Co., Ltd, HIS-3) using a frequency of 5 MHz at room temperature. Five data were measured at five different points on the end faces of the MOX pellet and averaged. The sound speeds in a CeO_2 pellet were measured as a standard before and after the MOX pellet studies.

3. Results and Discussion

The measured sound speeds are shown in table 1 together with porosity of the specimens. The effect of porosity to decrease the sound speeds for $\mathrm{Pu} = 20\%$ and $\mathrm{O/M} = 2.000$ was obtained from specimen as shown in *FIG. 2*.

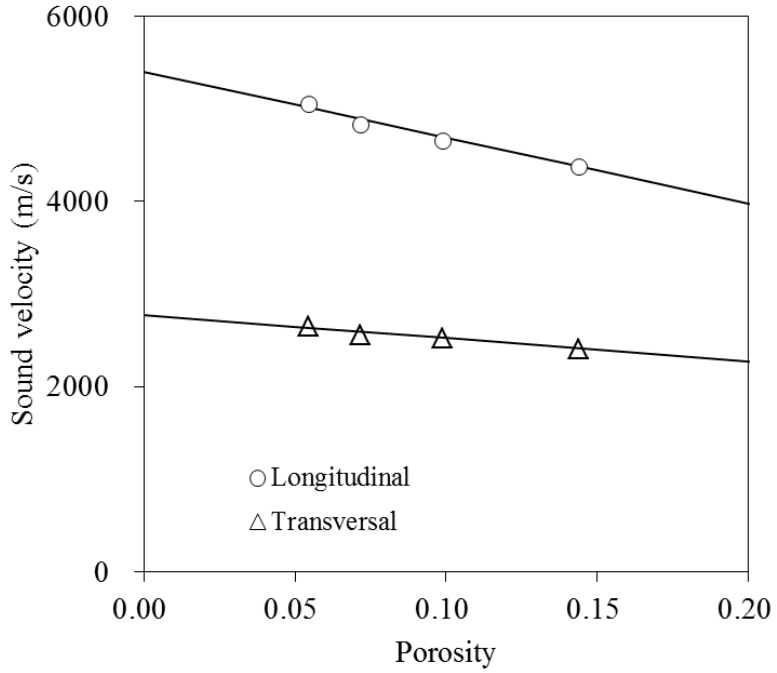

FIG. 2. Sound speeds of longitudinal and transversal waves in the specimens as a function of porosity and the linear fittings.

The effect of porosity was well fitted by linear function.

$$v_L(\mathrm{Pu} = 20\%, \mathrm{O/M} = 2.000) = 5399(1 - 1.3172p) \quad (1)$$

$$v_T(\mathrm{Pu} = 20\%, \mathrm{O/M} = 2.000) = 2774(1 - 0.8945p) \quad (2)$$

From the measured sound speeds, Young's modulus E and Poisson's ratio ν were calculated by

$$E = \rho {v_T}^2[(3{v_L}^2 - 4{v_T}^2)/({v_L}^2 - {v_T}^2)] \quad (3)$$

and

$$\nu = ({v_L}^2 - 2{v_T}^2)/2({v_L}^2 - {v_T}^2) \quad (4)$$

where ρ is density of the specimen. Calculated E and ν by equations are also shown in table 1. *FIG. 3* and *4* show the effect of porosity on Young's modulus and Poisson's ratio, respectively. The line was obtained by using the sound speeds calculated by equations (3) and (4). While the effect of porosity on Poisson's ratio is very slight, that on Young's modulus was significantly large. The effect of porosity on the Young's modulus was almost linear as discussed by Phani [14] and well agrees with the reported data by Nutt ($p = 0.127 \sim 0.228$) [7]. Young's modulus of the MOX for $p = 0.2$ was almost half of the porosity-free value indicating that the elasticity of MOX became significantly worse as the density decreased.

As a thermal property, the Debye temperature T_D was evaluated with the sound speeds of porosity-free values. They are defined as

$$T_D = (h/k_B)(9N/4\pi a^3)^{1/3}(1/v_L^3 + 2/v_T^3)^{-1/3} \quad (5)$$

where h is the Plank constant, k_B is the Boltzmann constant, N is the number of atoms in the unit cell and a is the lattice constant. The calculated T_D are shown in table 1. The Debye temperatures of $(U_{0.8}Pu_{0.2})O_2$ obtained in this work was 386 K. Although the literature data of MOX was not found, the value was on a good trend of those of UO_2 (379 K by Yamada [1]) and PuO_2 (415 K by Roof [15]).

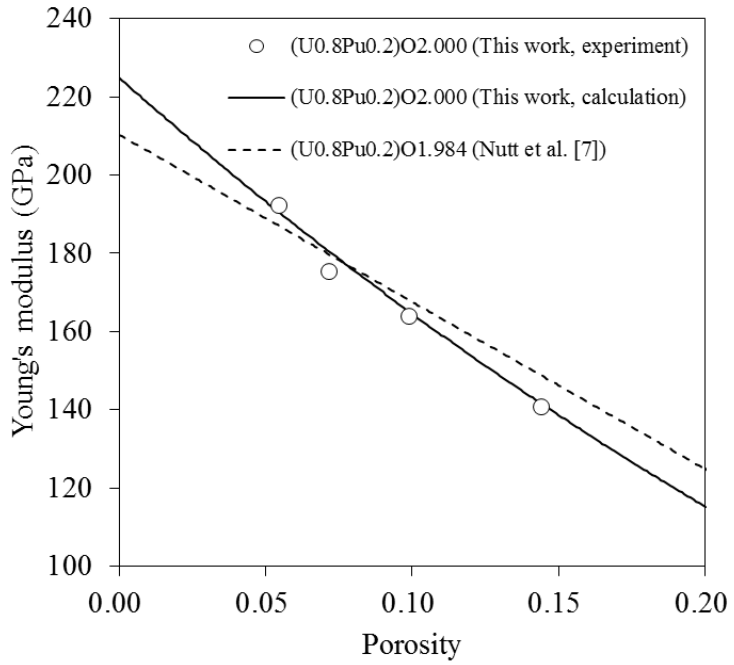

FIG. 3. Young's modulus of $(U_{0.8}Pu_{0.2})o_2$ as a function of porosity calculated by equation (3) together with literature data.

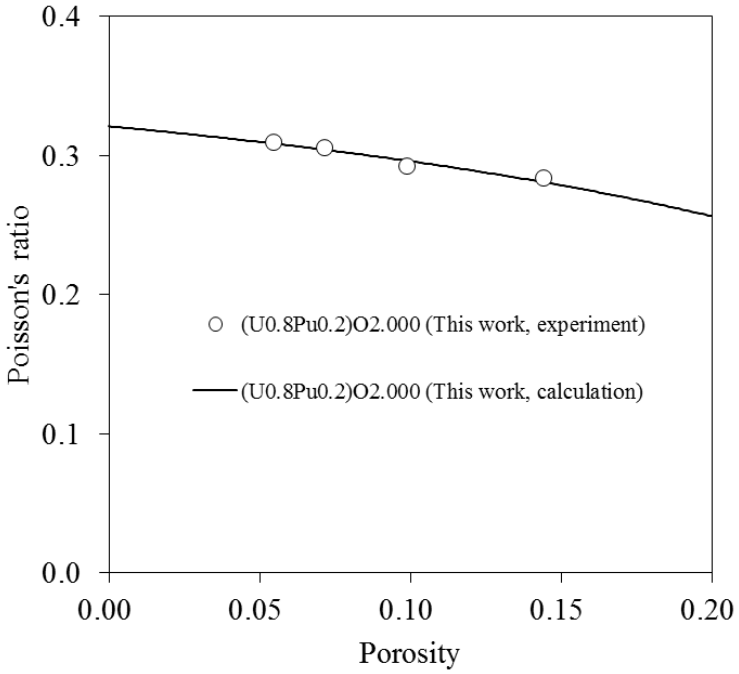

FIG. 4. Poisson's ratio of $(U_{0.8}Pu_{0.2})o_2$ as a function of porosity calculated by equation (4).

4. Conclusion

MOX pellets were prepared with different porosity and the sound speeds of longitudinal and transverse waves in the MOX pellets were measured. The effect of porosity to decrease the sound speeds was well described by a linear function and the fitting equations were obtained. Young's modulus evaluated with the sound speeds significantly decreased with increasing porosity and the values were in good agreement with the data available in the literature. Finally, the Debye temperature was calculated as a thermal property of MOX which also had a good agreement with the literature data.

5. Acknowledgement

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Reference

[1] Yamada, K., et al., "Study of the thermodynamic properties of $(U,Ce)O_2$", Journal of Nuclear Materials 247 (1997) 289-292.

[2] Kurosaki, K., et al., "Characterization of simulated burnup fuel by nanoindentation", Journal of Nuclear Materials 350 (2006) 203-207.

[3] Roque, V., et al., "Effects of the porosity in uranium dioxide on microacoustic and elastic properties", Journal of Nuclear Materials. 277 (2000) 211-216.

[4] Roque, V., et al., "Study by acoustic microscopy of irradiated and non-irradiated uranium dioxide", Journal of Nuclear Materials 275 (1999) 305-311.

[5] Hall, A.R., "Elastic moduli and internal friction of some uranium ceramics", Journal of Nuclear Materials 37 (1970) 314-323.

[6] Marlowe, M.O., "HIGH TEMPERATURE ISOTHERMAL ELASTIC MODULI OF UO2", Journal of Nuclear Materials 33 (1969) 242–244.

[7] Nutt, A.W., "Elastic and Anelastic Response of Polycrystalline UO_2-PuO_2", Journal of American Ceramic Society 53 (1970) 205-210.

[8] Padel , A., et al., "CONSTANTES ELASTIQUES DES CARBURES, NITRURES ET OXYDES D'URANIUM ET DE PLUTONIUM", Journal of Nuclear Materials 33 (1969) 40-51.

[9] Panakkal, J.P., "Ultrasonic velocity in sintered uranium dioxide pellets", Journal of Materials Science letters 3 (1984) 835-836.

[10] Kato, M., et al., "Thermal and Mechanical Properties of UO_2 and PuO_2", Proceedings of 13th Information Exchange Meeting, Seoul, Republic of Korea, September 2014.

[11] Koizumi, M., et al., "Development of a Process for Co-Conversion of Pu-U Nitrate Mixed Solutions to Mixed Oxide Powder Using Microwave Heating Method", Journal of Nuclear Science and Technology 20 [7] (1983) 529-536.

[12] Kato, M., et al., "Analysis of oxygen potential of $(U_{0.7}Pu_{0.3})O_{2\pm x}$ and $(U_{0.8}Pu_{0.2})O_{2\pm x}$ based on point defect chemistry", Journal of Nuclear Materials 389 (2009) 164-169.

[13] Kato, M., "Lattice parameters of $(U,Pu,Am,Np)O_{2-x}$", Journal of Nuclear Materials 385 (2009) 117-121.

[14] Phani, K.K., “Porosity dependence of ultrasonic velocity and elastic modulus in sintered uranium dioxide – a discussion”, et al., Journal of Materials Science letters 5 (1986) 427-430.

[15] Roof, J.R.B., “Simultaneous determination of X-ray Debye temperature and Grüneisen constant for actinide dioxides: PuO_2 and ThO_2”, Journal of Nuclear Materials 2 (1960) 39-42.