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Thermal conductivity of uranium–plutonium oxide fuel for fast reactors

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Abstract

A new thermal conductivity correlation for fully dense uranium–plutonium oxide fuel for fast reactors was formulated for fuel pin thermal analysis under beginning of irradiation conditions. The data set used in correlating the equation was systematically selected to minimize experimental uncertainty. The electron conduction term for uranium dioxide formulated by Harding and Martin [J. Nucl. Mater. 166 (1989) 223] was adopted to compensate for so few high temperature measurements. The excellent predictability of the new correlation was validated by comparing the calculated with measured fuel center temperatures in an instrumented irradiation test in the experimental fast reactor JOYO for low oxygen-to-metal (O/M) ratio fuel up to 1850 K.

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1. Introduction

The objective of this work was to formulate a new thermal conductivity correlation, which is to be used for fuel pin thermal analysis under beginning of irradiation conditions, for uranium–plutonium oxide fuel in fast reactors.

Oxide fuels can be categorized into three types. The first is uranium dioxide, which is widely used in both thermal and fast reactors and has been well characterized. The second is the thermal reactor type mixed-oxide (TR-MOX) fuel, which contains plutonium up to 10 mass% in heavy metal and has gained attention by its emerging utilization in thermal reactors. An equation for the thermal conductivity of TR-MOX fuel was recently reported by Duriez et al. [1]. The third is the fast reactor type mixed-oxide (FR-MOX) fuel, which contains 15–30 mass% plutonium. Unfortunately, there have been few thermal conductivity experiments for FR-MOX fuel especially in the past decade.

Martin [2] comprehensively reviewed the thermal conductivity of oxide fuels and established the basic approaches to their characterizations. Philipponneau [3] reviewed Martin's work and developed an equation for FR-MOX fuel; their major difference is in the oxygen-to-metal (O/M) ratio dependence. Based on the fuel pin thermal analysis for the short-term irradiation experiments in the experimental fast reactor JOYO, Philipponneau's equation tends to underestimate the thermal conductivity. The temperature dependence, especially at higher temperatures, also affects the results of fuel pin thermal analysis. However, it is very difficult to clarify both dependencies because of so few thermal conductivity measurements.

In this work, the thermal conductivity of oxide fuels was investigated similarly to Martin's approach. A new correlation for FR-MOX fuel was formulated by implementing some unique concepts to compensate for the limited high temperature data. The extent of O/M ratio dependence predicted by the proposed thermal conductivity equation was validated with the results of JOYO irradiation experiments by the 'Integral Method', which verifies the thermal conductivity equation based on the predictability of temperature increase from the fuel surface to the fuel center.

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2. Previous studies and investigations of thermal conductivity

2.1. Basic formulation

The thermal conductivity of fully dense oxide fuel (k_0) in the as-fabricated condition is normally described by Eq. (1):

$$k_0 = k_{\text{Phonon}} + k_{\text{Electron}}, \tag{1}$$

which is the sum of the contributions by phonon (k_{Phonon}) and electron (k_{Electron}) conduction.

The phonon conduction term for oxide fuel is in general written as Eq. (2) and the electron conduction term for uranium dioxide, which was derived from Winter [4] in Eq. (3).

$$k_{\text{Phonon}} = \frac{1}{A + BT}, \tag{2}$$

$$k_{\text{Electron}} = \frac{C}{T^2} \exp\left(-\frac{D}{T}\right), \tag{3}$$

where T is the temperature, and A, B, C, and D are constants. Below 2000 K phonon conduction is dominant whereas at higher temperatures electron conduction becomes dominant.

The constant A comes from the scattering of phonons by lattice defects and constant B from phonon–phonon scattering processes. The electron–hole pairs (small polarons) migration behavior determines C and D. Philipponneau [3] revealed that A increases parabolically with deviation from stoichiometry. Bonnerot [5] observed that B slightly depends on plutonium content. However, both Martin and Philipponneau concluded that the effect of plutonium content could be neglected for FR-MOX fuel. The foregoing knowledge leads to the following fundamental correlation:

$$k_0 = \frac{1}{A_1 + A_2\sqrt{(2 - \text{O/M}) + A_3} + BT} + \frac{C}{T^2} \exp\left(-\frac{D}{T}\right), \tag{4}$$

where k_0 is the thermal conductivity of fully dense FR-MOX fuel in W/mK, T the temperature in K, O/M the oxygen-to-metal ratio, and A_1, A_2, A_3, B, C, and D are constants.

2.2. Experimental data in previous studies

The thermal diffusivity and conductivity data for oxide fuels have been accumulated in this work are from Bonnerot [5], VanCraeynest and Weilbacher [6], Fukushima et al. [7], Conway and Feith [8], Bates [9], Laskiewicz et al. [10], Lucuta et al. [11], Topliss et al. [12], Kosaka et al. [13], Hetzler et al. [14], Elbel and Schmidt [15], and Elbel and Vollath [16]. These data had been measured with various techniques: such as laser flash, modulated electron beam, and radial heat flow techniques for as-fabricated state specimens.

The diffusivity of FR-MOX fuel [5–7] was multiplied by the specific heat, as reported by Fink [17], to convert into the conductivity in this work. The thermal conductivity data (k) were then normalized to the fully dense state (k_0) by the following modified Loeb formula:

$$F = 1 - \alpha P, \tag{5}$$

where F is the porosity correction factor ($k = Fk_0$), P the volume fraction of porosity, and α is the coefficient. For $P < 0.1$ in uranium dioxide, IAEA [18] recommends $\alpha = 2.5$; the coefficient for high density uranium dioxide pellets is equivalent to mixed oxide [2]. Bakker et al. [19] showed that the contribution by radiation conduction in oxide fuel is negligible.

2.3. Effect of plutonium in stoichiometric oxide fuels

The temperature dependence of the thermal conductivity of nearly stoichiometric oxide fuels (O/M ratios: 1.995–2.004) are shown in Figs. 1–3. Fig. 1 is for uranium dioxide, Fig. 2 is for TR-MOX fuel and Fig. 3 is for FR-MOX fuel. The overlays of the Harding and Martin [20] and Delette and Charles [21] equations are included in Figs. 1 and 2. Fig. 2 also includes an overlay of Duriez's equation, and Fig. 3 includes the overlays of Harding's, Duriez's, and Philipponneau's equations.

In Figs. 1 and 2, Harding's equation appears to fit better than Delette's with the experimental data of both uranium dioxide and TR-MOX. Fig. 3 illustrates that the thermal conductivities measured for FR-MOX by several authors agree fairly well with each other as well as with the Philipponneau's and Duriez's equations.

Although previous studies have reported that 20% plutonium additions to uranium oxide lowers the thermal conductivity by 5% [2] or 8% [22], it is noteworthy that Harding's equation fit well even with stoichiometric FR-MOX fuel over the full temperature range as shown in Fig. 3. In practice, averaged residuals between experimental data in Fig. 3 and the calculated values over 700 K by Harding's equation results in 0.06 W/mK or 1.7% (147 points). This implies that the effect of plutonium addition is minor and can be neglected in the correlation.

2.4. Effect of deviation from stoichiometry in FR-MOX

Figs. 4 and 5 compare the effect of deviation from stoichiometry, i.e., O/M ratio dependence for hypostoichiometric fuels (O/M ratios: 1.96–1.99). They also show that the degradation, scatter, and difference among the authors increase with deviation from stoichiometry. This

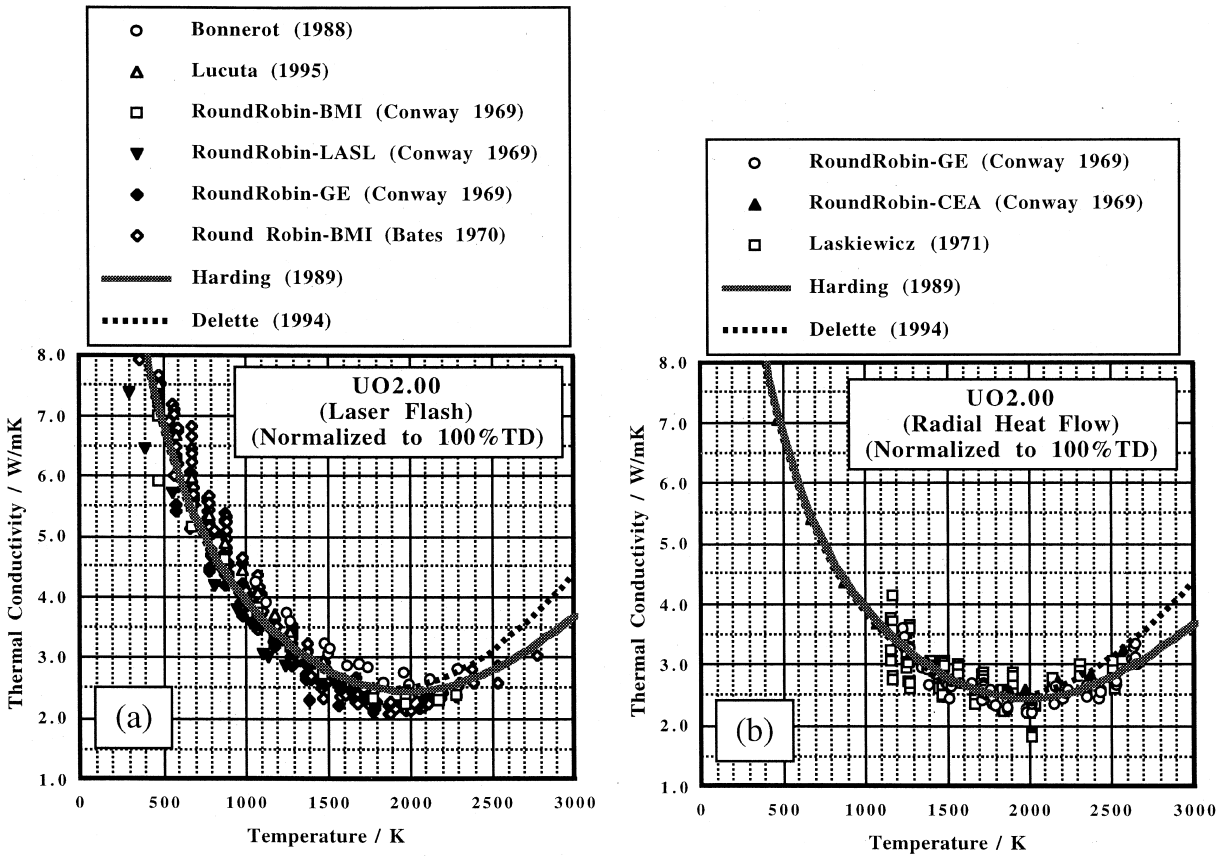

Fig. 1. Temperature dependence of thermal conductivity of uranium dioxide fuel (a) laser flash; (b) radial heat flow (CEA,GE).

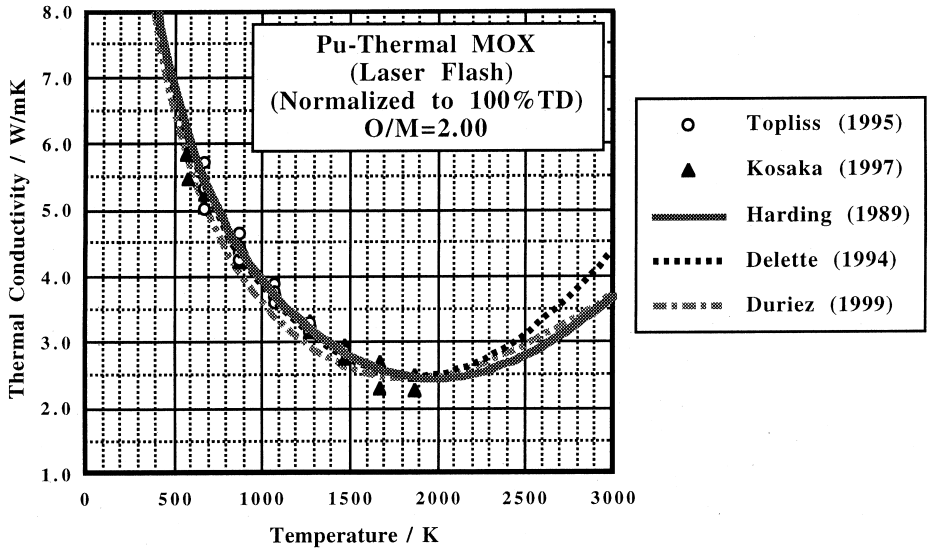

Fig. 2. Temperature dependence of thermal conductivity of stoichiometric, thermal reactor type uranium–plutonium oxide fuel.

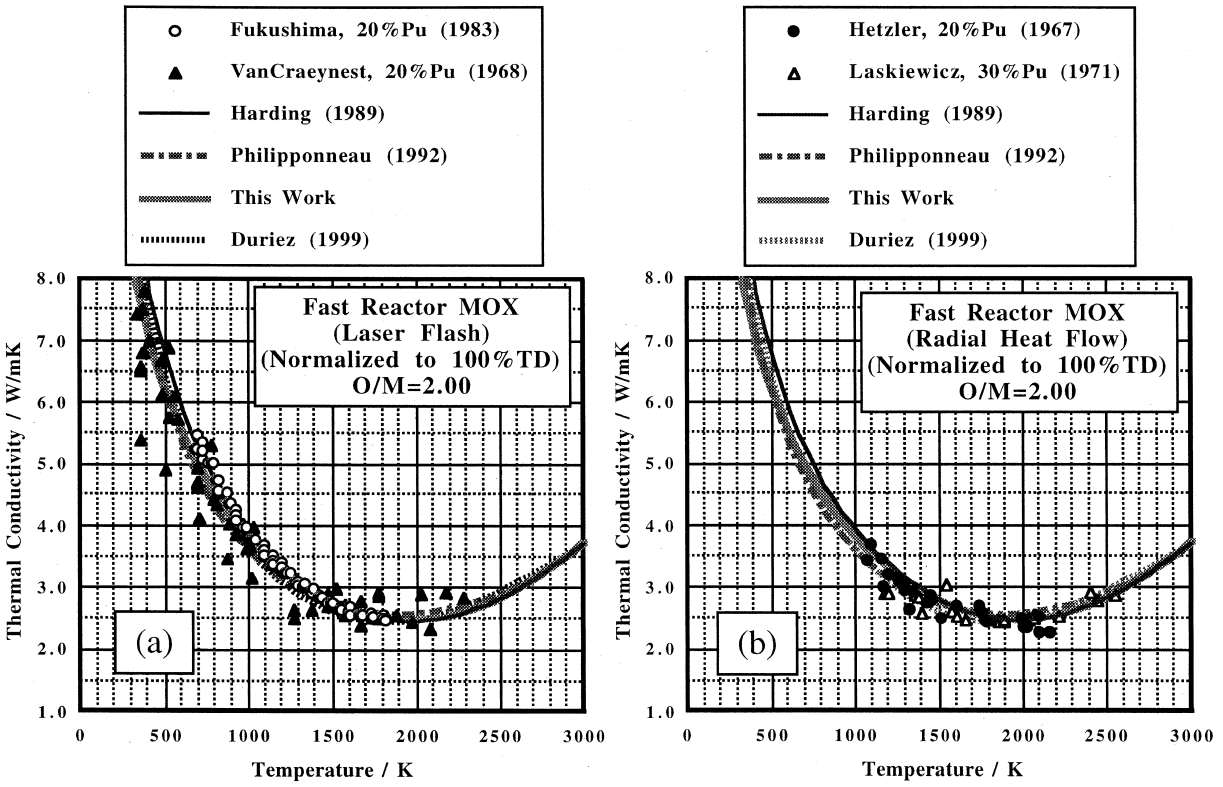

Fig. 3. Temperature dependence of thermal conductivity of stoichiometric, fast reactor type uranium–plutonium oxide fuel (a) laser flash (JAERI,CEA); (b) radial heat flow (GE).

phenomena seems to be caused by an 'O/M drift' phenomena (chemical instability) of the specimens as reported by Elbel and Vollath [16].

In addition, the correlations by Duriez et al. [1] and Philipponneau [3] are overlaid in Figs. 4 and 5 for comparison. The Philipponneau's equation (in Figs. 3–5), tends to overestimate the effect of the deviation from stoichiometry in comparison with the Duriez's equation.

3. Data set and proposed formulation

3.1. Data set

In selecting a data set to be used to formulate a reliable thermal conductivity equation, there were two basic criteria that had to be adhered to to assure reliability; the data should be reliable from the viewpoint of the respective authors and measurement techniques and the specimens should have a density greater than 93%TD.

Using this criteria, the data from VanCraeynest and Weilbacher [6], Fukushima et al. [7], Laskiewicz et al. [10], and Hetzler et al. [14] in Fig. 3 (O/M ratio = 2.00) and Hetzler et al. [14] and Elbel and Schmidt [15] in Fig. 5(a) (O/M ratio = 1.98) were selected as the data set of 221 points for formulation. The fuel parameters in this data set are 20–30 mass% plutonium, O/M ratios 1.98 and 2.00, and densities of 94.3–96.4%TD over the temperature range of 337–2552 K.

3.2. Proposed formulation

As described above, the data set covered the temperature range of 337–2552 K, but at temperatures higher than 2200 K, there are fewer data points compared to uranium dioxide data. This means that it is nearly impossible to determine the electron conduction term's constants C and D in Eq. (4) by the data set only. The low number of high temperature data can be compensated for by alternative methods.

Ronchi [23] measured heat capacity and thermal diffusivity of stoichiometric, high-density mixed-oxide fuel at temperatures from 1800 to 2600 K, and pointed out that thermal conductivity of mixed-oxide fuel is effectively equal to that of uranium oxide above 2400 K.

The Harding's equation for uranium dioxide (Fig. 3), fits very well for stoichiometric FR-MOX fuel up to

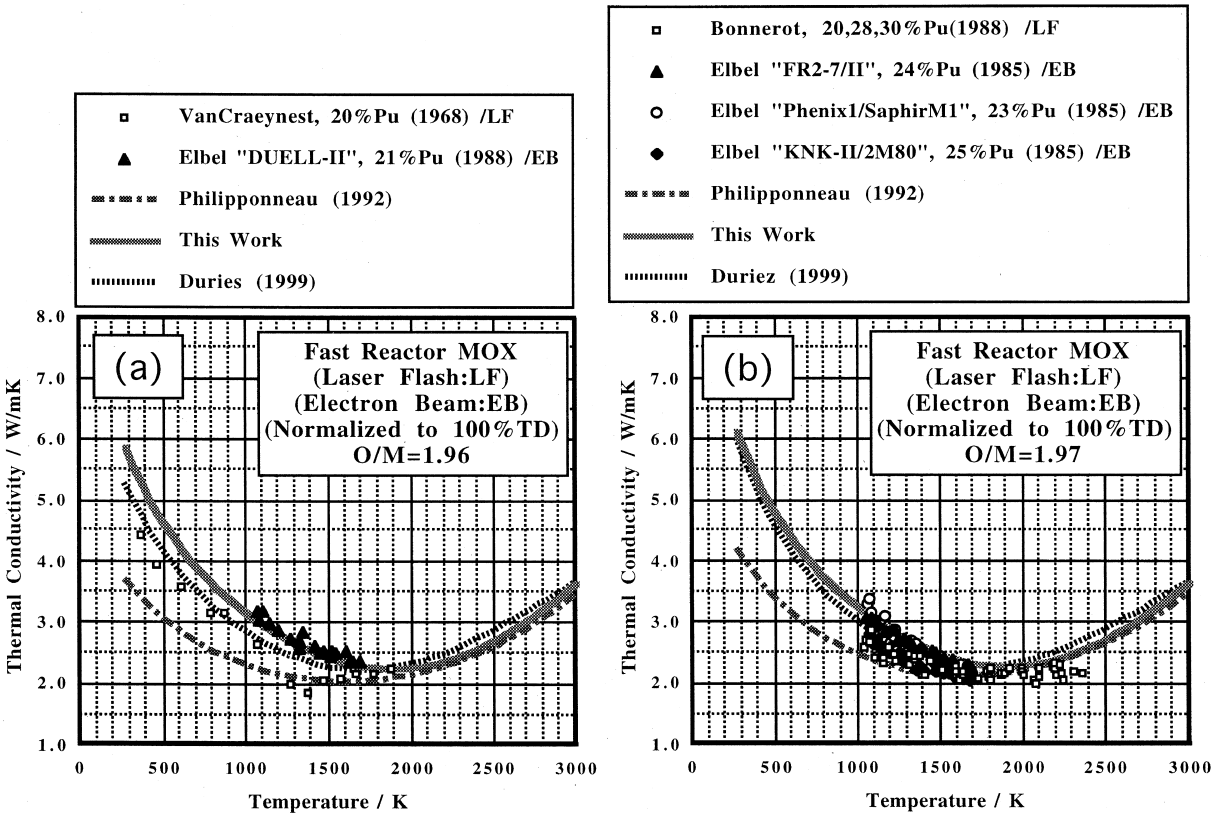

Fig. 4. O/M ratio dependence in fast reactor type uranium–plutonium oxide fuel (a) O/M ratio = 1.96; (b) O/M ratio = 1.97.

2500 K. This suggests that electron conduction term with the constants C and D in Harding's equation ($C = 4.715 \times 10^9$ and $D = 16361$ in Eqs. (3) and (4)) will be applicable to mixed-oxide fuels.

By subtracting the electron conduction contribution from each data, constants A_1, A_2, A_3, and B in Eq. (4) can be determined by the least square method, which leads to the following equation:

$$k_0 = \frac{1}{0.06059 + 0.2754\sqrt{2 - \mathrm{O/M}} + 2.011 \times 10^{-4} T} + \frac{4.715 \times 10^9}{T^2} \exp\left(-\frac{16361}{T}\right), \quad (6)$$

A_3 is so small (2.792×10^{-12}) that it was deleted from the equation. The standard deviation between the data set and the calculated values is 0.20 W/mK (absolute) or 6.2% (relative). This equation is also included in Figs. 3–5.

Figs. 4 and 5 also reveals that, in lower O/M ratio regions (<1.98), the equations need to be verified by more experimental measurements to determine the degree of O/M dependence.

4. Validation of formulation

4.1. Instrumented irradiation experiments in JOYO

Instrumented irradiation experiments using the Integral Method are desirable and favorable to avoid the effect of chemical instability, especially at beginning of irradiation conditions. This method verifies the thermal conductivity equation based on the predictability of temperature increase from the fuel surface to the fuel center. But, the fuel pin thermal and mechanical behavior must be modeled well enough to calculate a confident temperature profile.

The results of the INTA-2 test [24] in JOYO that irradiated low O/M ratios fuel pellets (O/M ratios: 1.95 and 1.96) were used for the validation. The irradiation conditions of the test fuel pins were recently reported by Sekine et al. [25,26]. The fuel center temperatures of 12 fuel pins were successfully measured. Each fuel pin was loaded with annular fuel pellets with tungsten–rhenium type thermocouple inserted through the central annular hole. The pins were clad with advanced austenitic steel (7.5 mm in outer diameter and 0.4 mm wall thickness) and filled with high purity helium gas,

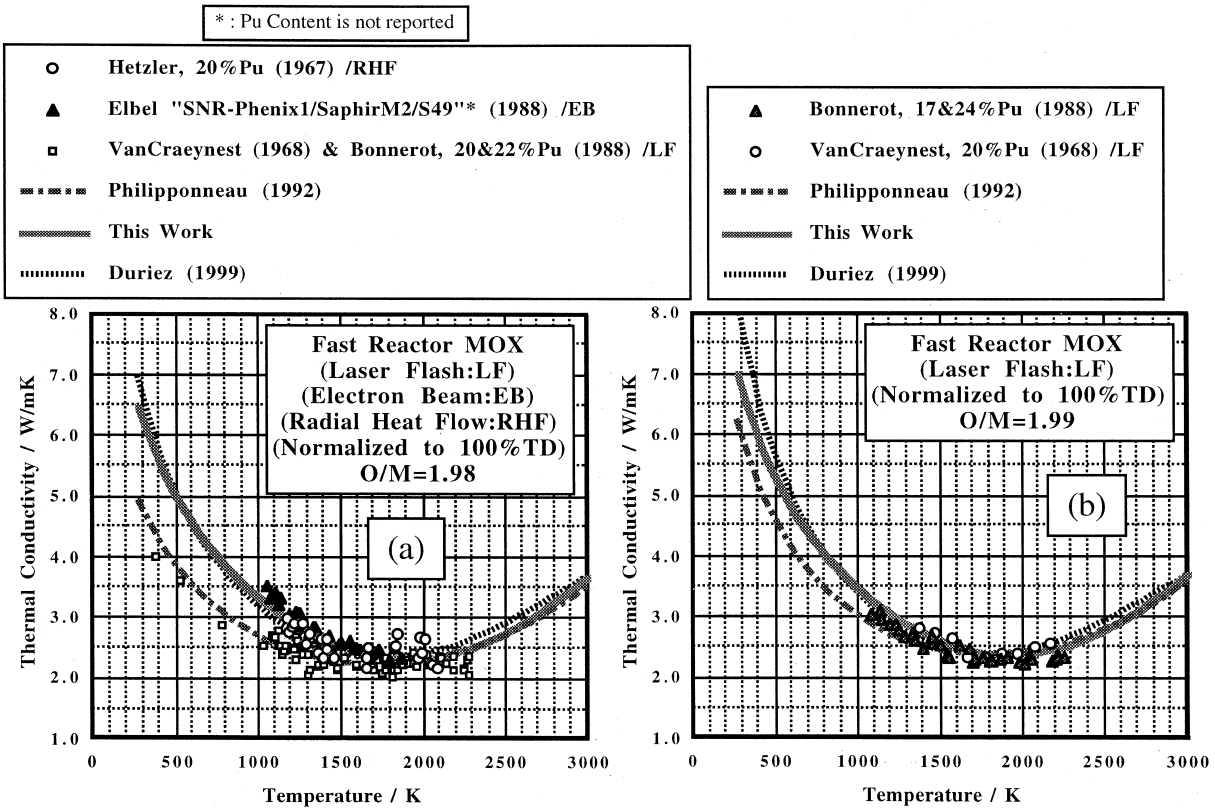

Fig. 5. O/M ratio dependence in fast reactor type uranium–plutonium oxide fuel (a) O/M ratio = 1.98; (b) O/M ratio = 1.99.

one fuel pin was loaded with a tag gas capsule, summarized in Table 1.

4.2. Procedure of thermal analysis

A one-dimensional temperature profile in a fuel pin was calculated using FORTRAN program that combines several models for fuel pin thermal and mechanical behaviors. The outline of the program is briefly described below and the detailed description may be found in Ref. [27].

Gap conductance was modeled by Ross and Stoute [28] as the sum of the contributions by gap gas, radiation, and fuel-to-cladding contact. The gap gas contribution is formulated in Eq. (7)

$$H_{\text{gas}} = \frac{k_{\text{mixed-gas}}}{R + TJD + GAP}, \tag{7}$$

where H_{gas} is the gap conductance by gap gas contribution, $k_{\text{mixed-gas}}$ the thermal conductivity of mixed gas, R the roughness of cladding and fuel pellet surfaces, TJD the temperature jump distance at cladding and fuel pellet surfaces, and GAP is the fuel-to-cladding gap at power. In practice, during lower power irradiation, both the radiation contribution relative to gap gas conductance and the fuel-to-cladding contact are negligible and can be neglected. Chantoin et al. [29] reported that the effect of surface roughness on gap conductance was also negligible; $R = 0$ in Eq. (7).

Fuel pellets are subdivided into 60 ring meshes, each mesh thermally expands by the thermal expansion coefficient [30] and the accumulated increments leads to gap closure (shrinkage). Fuel pellet relocation behavior, which largely influences the heat transfer between cladding and fuel pellet surfaces, has been investigated for solid fuel pelleted fuel pins at high power and correlated into Eq. (8) [31]

$$\Delta G = 0.111 R_{\text{ci}} G_0 - 45, \tag{8}$$

where ΔG is the diameter relocation in micrometer, R_{ci} the cladding inner diameter in mm, and G_0 is the as-fabricated diameter gap in micrometers. It is assumed that Eq. (8) is applicable for annular fuel pellets and at lower powers. The diameter increment of the relocation calculated by Eq. (8) is added to the fuel pellet outer diameter.

Calculations were conducted based on the initial maximum power (just after the reactor reached full

Table 1
Fuel pin specifications and irradiation conditions of instrumented irradiation experiments in JOYO

Test	No.	Pin ID	Fuel pellet outer diameter, lot mean (mm)	Fuel pellet inner diameter, lot mean (mm)	Fuel pellet O/M ratio, lot mean (–)	Fuel pellet density, lot mean (% TD)	Fuel pellet Pu content (Pu/(Pu + U)), lot mean (mass%)	As-fabricated diametral gap, PinAverage (μm)	Tag gas, Y: Yes, N: No	Linear heat rate at initial maximum power[a] (KW/m)	Cladding midwall temperature at initial maximum power (K)
INTA-2	1	I2201	6.54	1.90	1.96	91.4	18.9	155	N	21.9	825
INTA-2	2	I2202	6.54	1.90	1.96	91.4	18.9	155	N	30.2	768
INTA-2	3	I2203[b]	6.54	1.90	1.96	91.4	18.9	155	N	–	–
INTA-2	4	I2204	6.49	1.90	1.96	91.4	18.9	208	N	28.9	773
INTA-2	5	I2206	6.54	1.90	1.96	91.4	18.9	160	N	31.5	766
INTA-2	6	I2207	6.54	1.90	1.96	91.4	18.9	160	N	29.3	779
INTA-2	7	I2208	6.54	1.92	1.96	95.5	18.9	162	N	29.9	777
INTA-2	8	I2209	6.54	1.90	1.96	91.4	18.9	160	N	23.7	826
INTA-2	9	I2210	6.49	1.91	1.96	95.3	18.9	211	N	30.0	774
INTA-2	10	I2211	6.56	1.91	1.95	92.2	18.9	143	N	29.0	776
INTA-2	11	I2212	6.54	1.90	1.96	91.4	18.9	160	Y	29.0	776
INTA-2	12	I2213	6.52	1.90	1.95	92.0	18.9	183	N	29.1	775
INTA-2	13	I2214	6.60	1.92	1.96	92.4	18.9	104	N	30.0	776

[a] Fuel centerline temperature were measured at $X/L = 0.5$, 0.8 or 0.9.
[b] Thermocouple was misconnected.

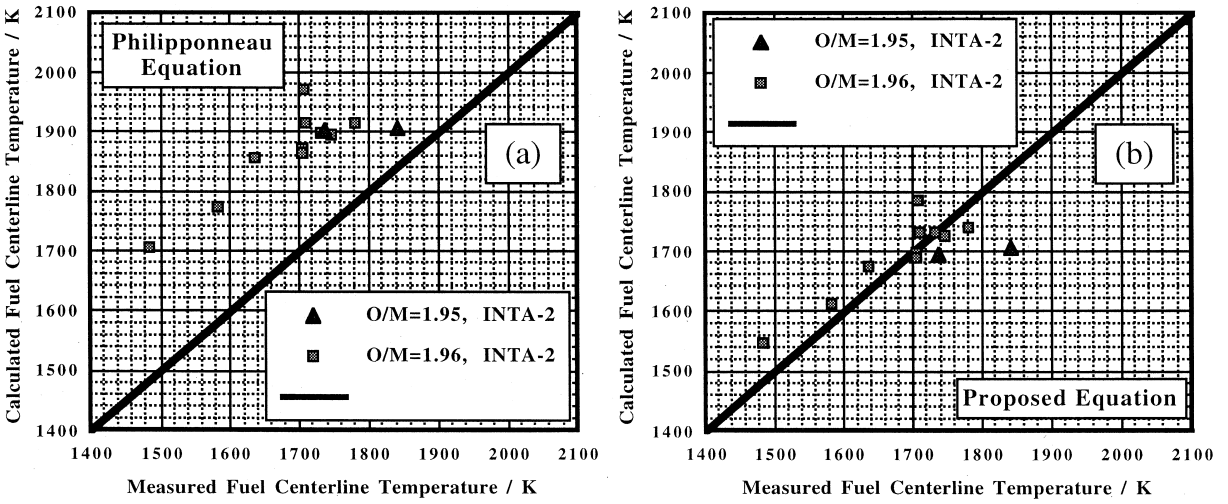

Fig. 6. Calculated and measured fuel center temperatures by Philipponneau's equation (a) and the proposed equation; (b) for instrumented irradiation experiments in JOYO.

power). Neither microstructure evolution as observed in the post-irradiation examinations nor oxygen redistribution were assumed. The effect of the fuel pellet porosity on thermal conductivity had been characterized [32]. The applicable coefficients, $\alpha = 2.5$ for high density fuel pellets (~95%TD) and $\alpha = 1.9$ for medium density fuel pellets (~92%TD), were used in Eq. (5).

4.3. Results of the analysis

Fig. 6 compares the measured and calculated fuel center temperatures using Philipponnueau's equation and the proposed equation (Eq. (6)). As noted in Section 1, Philipponneau's equation tends to overestimate the fuel center temperatures, whereas the proposed equation appears to predict very well. The averaged residuals are 177 K using Philipponneau's equation (in Fig. 6(a)) and −1 K using the proposed equation (in Fig. 6(b)).

The scatter in the predicted fuel center temperatures for the proposed equation should be examined to confirm the reliability of the Integral Method. The standard deviation (54 K) between measured and predicted fuel center temperatures is anticipated to include a large uncertainty induced by the relocation behavior. In the case where the relocation behavior is dominant, the uncertainties in the temperature increase in the fuel pin can be expressed as follows:

$$\sigma^2_{\text{total}} = \sigma^2_{\text{relocation}} + \sigma^2_{\text{others}}, \tag{9}$$

where σ_{total} is the standard deviation of total temperature increase in the fuel pin, $\sigma_{\text{relocation}}$ the standard deviation induced by the relocation behavior, and σ_{others} is the standard deviation induced by other factors. In cases where the diameter relocation increases or decreases 19 μm (the standard deviation in Eq. (8)), the averaged change in the fuel center temperature in 12 fuel pins is 44 K. With $\sigma_{\text{relocation}}$ as the averaged change ($\sigma_{\text{relocation}} =$ 44 K) and σ_{total} as standard deviation between measured and predicted fuel center temperature ($\sigma_{\text{total}} = 54$ K) in Eq. (9), then σ_{others} is equal to 31 K. Although the scatter of the relocation tends to be large, 12 measured data points should be statistically sufficient. This confirms that the effect of the relocation behavior is dominant and the ratio of σ_{others} to total temperature increase in fuel pins is less than 4%.

As described above, Eq. (6) is predictable for in-pile thermal performance and was verified by the Integral Method up to 1850 K for fuels with lower O/M ratios (1.95 and 1.96).

5. Discussion

In correlating the thermal conductivity of FR-MOX fuel, there have been two major problem areas that relate to the deficiency in measured data. One problem is when the O/M ratios is below 1.97 and the other is at temperatures above 2200 K.

Presently only instrumented irradiation experiments can determine the O/M ratio dependence of thermal conductivity. This is because fuel pellets in the fuel pin are sealed with very high purity inert gas, which prevents the O/M drift phenomena. As described in Section 4, the proposed equation (Eq. (6)) can predict well the thermal conductivity of lower O/M ratio (1.95 and 1.96) fuels up to 1850 K, even though the equation is based on the measurements of higher O/M ratios (1.98 and 2.00). This accuracy is the culmination of

sophisticated methods of experimentation and thermal analysis.

More experimental studies at higher temperatures (above 2200 K) are necessary to determine the temperature dependence of thermal conductivity in the future, particularly since mixed-oxide fuel still has larger uncertainties than uranium dioxide. A detailed comparison of stoichiometric oxide fuels in Figs. 1–3 shows that the thermal conductivity of both uranium dioxide and mixed dioxide are comparable. Ronchi [23] pointed out that the thermal conductivity of mixed dioxides can be effectively equal to that of uranium dioxide above 2400 K. As shown in Fig. 1, Harding's equation for uranium dioxide correlated well with experimental data and appears to be reliable up to 2700 K, because of the abundance of theoretical investigations and experimental measurements, including several other related physical properties. This means that the proposed equation would also correlate effectively at higher temperatures for mixed-oxide.

As discussed above, the proposed equation (Eq. (6)) correlates very well with the O/M ratio dependence and is applicable to typical FR-MOX fuel at high temperature.

6. Concluding summary

A new equation for the thermal conductivity of mixed-oxide fuel for fast reactors is as follows:

$$k_0 = \frac{1}{0.06059 + 0.2754\sqrt{2 - \mathrm{O/M}} + 2.011 \times 10^{-4} T} + \frac{4.715 \times 10^9}{T^2} \exp\left(-\frac{16361}{T}\right), \quad (10)$$

where k_0 is the thermal conductivity of fully dense FR-MOX fuel in W/mK, T the temperature in K, and O/M is the oxygen-to-metal ratio.

The data set (221 points) used to formulate the equation was selected based on the following criteria for minimizing experimental uncertainty; the data must be reliable from the viewpoint of the respective authors and the measurement techniques and specimens should have a density greater than 93%TD. The Harding's electron conduction term for uranium dioxide was adopted to compensate for the limited number of measurements at high temperatures.

The equation was validated for calculating fuel center temperatures by the Integral Method and demonstrated that it is also applicable for FR-MOX fuels with lower O/M ratios (1.95 and 1.96) up to 1850 K.

This new equation has been shown to predict well and is recommended for fuel pin thermal analysis with typical FR-MOX fuel pellets under beginning of irradiation conditions.

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