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A Thermal Conductivity Model for LWR MOX Fuel and Its Verification Using In-pile Data

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Abstract

The MOX fuel for LWR is fabricated either by direct mechanical blending of UO_2 and PuO_2 or by two stage mixing. Hence Pu-rich particles, whose Pu concentrations are higher than pellet average one and whose size distribution depends on a specific fabrication method, are inevitably dispersed in MOX pellet. Due to the inhomogeneous microstructure of MOX fuel, the thermal conductivity of LWR MOX fuel scatters from 80 to 100 % of UO_2 fuel. This paper describes a mechanistic thermal conductivity model for MOX fuel by considering this inhomogeneous microstructure and presents an explanation for the wide scattering of measured MOX fuel' s thermal conductivity. The developed model has been incorporated into a KAERI' s fuel performance code, COSMOS, and then evaluated using the measured in-pile data for MOX fuel. The database used for verification consists of homogeneous MOX fuel at beginning-of-life and inhomogeneous MOX fuel at high burnup. The COSMOS code predicts the thermal behavior of MOX fuel well except for the irradiation test accompanying substantial fission gas release. The over-prediction with substantial fission gas release seems to suggest the need for the introduction of a recovery factor to a term that considers the burnup effect on thermal conductivity.

Key Words : fuel performance, thermal conductivity, MOX fuel, inhomogeneity, COSMOS

1. Introduction

Thermal performance is prerequisite to most aspects of fuel behavior. Not only is it extremely important from a safety point of view, particularly in the avoidance of fuel melting and loss of geometry, but also many of the material properties of interest like fuel creep and fission gas release are exponentially dependent on temperature [1].

The utilization of MOX (mixed oxide) fuel in a commercial reactor can reduce the accumulation of fissile plutonium from reprocessing and ex-weapon grade plutonium from the decommissioning of military devices. The slightly lower thermal

conductivity of MOX fuel compared to UO_2 leads to different central fuel temperatures and consequently to different fission gas releases for the same linear heat generation ratings. Therefore, it is indispensable to develop a more accurate model for temperature prediction in MOX fuel, which is determined from thermal conductivity [2].

Until now, several researchers have published results on the thermal conductivities of MOX fuels by experimental measurements and / or theoretical analyses. For example, Gibby [3] measured the thermal conductivity of MOX fuel up to a Pu content of 30 % and fitted by linear regression to $1/(A+B \cdot T)$, which showed the decrease of thermal conductivity of MOX with increasing Pu contents. On the contrary, the analytical thermal conductivity of MOX fuel was proposed by the available measured data by that time [4,5]. These models showed a sensitive dependence on the stoichiometry, while they were not influenced by Pu contents ranging from about 10 to 30%. Recently, CEA [6] measured the thermal conductivity of un-irradiated MOX fuel, which showed no significant dependence on Pu content from 3 to 15% and a decrease with O/M ratio. It is, however, generally accepted that the thermal conductivity of MOX is slightly less than that of UO_2, even though the content of Pu is less than 10% in the $(U,Pu)O_2$ fuel.

These relevant data on the thermal conductivity of MOX does not make universal thermal conductivity available, especially in commercially used Pu contents owing to the considerable scattering of thermal conductivities.

In the present paper, a new thermal conductivity model is proposed for MOX fuel considering its microstructural inhomogeneity. The newly developed thermal conductivity model has been incorporated into a KAERI' s fuel performance code, COSMOS [7], and then evaluated by using the measured temperature from MOX irradiation test.

2. Methodology for the Thermal Conductivity of MOX

2.1. Heterogeneity of MOX

It is recognized that MOX fuel has an in-reactor behavior almost identical to UO_2 fuel. The MOX fuel' s thermal conductivity assessed through in-pile irradiation, however, has been observed with wide scattering when compared with UO_2 thermal conductivity. In general, the thermal conductivity is lower in MOX than in UO_2 fuel following the mixing of plutonium into the UO_2 matrix. Some MOX fuel has a very homogeneously distributed plutonium throughout the pellet, whereas the inhomogeneous MOX has agglomerates of plutonium rich zones.

It is not obvious how much the inhomogeneity of plutonium-rich agglomerates degrades the thermal conductivity of MOX. For early-in-life MOX fuel, the inhomogeneous MOX could have higher thermal conductivity than the homogeneous MOX owing to the following combination of theoretical backgrounds:

- A random distribution of the particles results in a higher conductivity than a regular distribution.
- A medium containing different sized particles also has a higher conductivity than one containing equal sized particles.

However, thermal conductivity of MOX fuels at the beginning-of-life would change with irradiation. The inhomogeneous MOX has been observed with much localized fissioning in the plutonium-rich zone in which the local burnup is two or three times higher than the matrix and consequently, the microstructure becomes very porous. The concentrated local burnup and porous microstructure can lead to a more dramatic

Table 1. Area Fraction of the Phases and Distribution of the Input Plutonium in Different Types of MOX Fuels [9]

Fuel type	MIMAS-AUC	MIMAS-ADU	SBR
Area fraction of phase			
Matrix (%)	75.4	46.7	98-99
Pu-rich spot (%)	24.6	11.1	1-2
Coating around UO_2 particles (%)	-	42.2	-
Distribution of input plutonium			
Matrix (%)	39	15	96
Pu-rich spot (%)	61	39	4
Coating around UO_2 particles (%)	-	46	-

degradation of thermal conductivity in the inhomogeneous MOX than in the homogeneous MOX fuel.

To develop a mechanistic thermal conductivity model, it is assumed that a significant scattering of the thermal conductivity results from the unique microstructure of MOX fuel.

Since sufficient data related to the MOX microstructure for un-irradiated and irradiated fuel are not available, only two experimental results are used to characterize the inhomogeneous MOX microstructure. One of international MOX research program revealed the Pu contents in the Pu agglomerates had a distribution ranging from 1 to 20 wt % and the size of agglomerates in un-irradiated MOX fuel had a log-normal type distribution as shown in Fig. 1. For irradiated fuel, Siemens-KWU [8] published data on the microstructure of two MOX fuels produced by the Optimized Co-milling (OCOM) process. The OCOM 15 fuel contained 34 vol. % of MOX agglomerates with a nominal PuO_2 concentration of 15 wt. %, whereas the OCOM 30 fuel contained 17 vol. % of MOX agglomerates with a nominal PuO_2 concentration of 30 wt. %. They reported similarities between MOX agglomerates and the high burnup microstructure in the periphery of conventional UO_2 fuel. From these relevant data, the Pu agglomerates can be characterized by twice or three times higher burnup than the matrix and a porosity of about 20~30 % in high burnup MOX fuel.

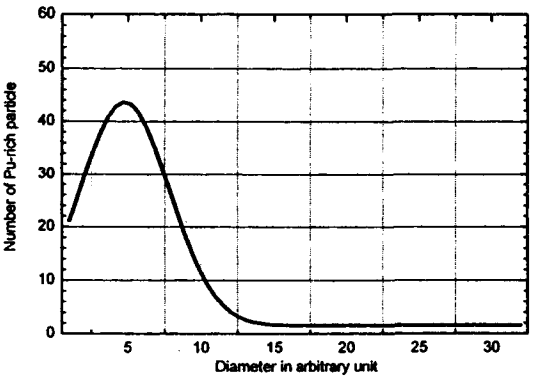

Fig. 1. Inhomogeneous Pu-rich Particle Distribution

Since all information is not available to characterize the microstructure of homogeneous and inhomogeneous MOX fuels, the general properties in Table 1 [9] are used to determine the distribution of area fraction and plutonium contents of the plutonium rich agglomerates in the MOX pellet.

2.2. Unified thermal Conductivity of MOX Fuel

Thermal conductivity for UO_2 or MOX fuel can be described in the general form

$$k = \frac{1}{(A + \alpha \cdot BU + B \cdot T)} + C \cdot e^{D \cdot T}$$

where $\alpha \cdot BU$ is the term that considers the thermal conductivity degradation with burnup. In the hyperbolic term, A and B result from the lattice contribution through a phonon-defect and the phonon-phonon scattering, respectively. On the other hand, the exponential term is caused by the electronic conduction which becomes dominant for temperatures higher than 1900 K.

To estimate in-pile thermal behaviors, the thermal conductivity has been measured in-pile and out-of-pile. In addition, many thermal conductivity models have been proposed for UO_2 and MOX fuel. For example, Halden's thermal conductivity [10] fitted with in-pile temperature data is expressed by

$$k = \frac{f_{mox}}{0.1149 + 0.0035 \cdot BU + \left(2.475 \times 10^{-4} - 7.921 \times 10^{-7} \cdot BU\right) \cdot TC} + 0.0132 \cdot e^{0.00188 \cdot TC}$$

where BU is burnup in MWd/kgOX and TC is temperature in degrees Celsius. The parameter of f_{mox} defines the ratio of MOX thermal conductivity to that of UO_2. The value of f_{mox} for UO_2 is equal to unity and the value of 0.92 is empirically recommended for MOX fuel.

The burnup degradation phenomenon is induced by the introduction of defects by irradiation into a previously almost perfect matrix. Furthermore, the thermal conductivity steeply decreases across the Pu-rich agglomerates due to higher local fissioning which leads to porous microstructures like rim structure in the high burnup UO_2 fuel. Accordingly, the thermal conductivity of irradiated MOX fuel is mainly dependent on the precise characteristics of Pu-rich spots.

The effect of the Pu-rich agglomerates on the thermal conductivity can be estimated by the assumption that MOX fuel consists of a matrix including some part of plutonium fed from the manufacturing stage and Pu-rich agglomerates. The matrix can contain a relatively large amount of Pu contents in the homogeneous microstructural MOX fuel, whereas the Pu contents in the matrix decrease in the inhomogeneous MOX fuel. The dependence of thermal conductivity on Pu-rich agglomerates can be considered for both homogeneous and inhomogeneous MOX fuel as follows.

The generally applicable thermal conductivity for two-phase material is used to assess the thermal conductivity of MOX fuel containing heterogeneity. The thermal conductivity of MOX fuel can be estimated from the combination of thermal conductivities for matrix and Pu-rich agglomerates [11]:

$$k_{matrix} = \frac{1}{A_{matrix} + B_{matrix} \cdot T}$$

$$k_{MOX} = k_{matrix} \cdot \left\{ 1 - a \cdot P_{PuR}^{\frac{2}{3}} \cdot \left[1 - \frac{1}{1 + \frac{1}{a} \cdot P_{PuR}^{\frac{1}{3}} \cdot \left(\frac{k_{matrix}}{k_{PuR}} - 1 \right)} \right] \right\}$$

where

k_{matrix} = thermal conductivity of a matrix in which a fraction of Pu contents are included (W/m-K)

k_{MOX} = integral thermal conductivity of heterogenous MOX fuel (W/m-K)

k_{Pur} = thermal conductivity of Pu-rich particles (W/m-K)

P_{PuR} = volumetric fraction of Pu-rich particles

a = anisotropy factor (a=1 means isotropic pore distribution).

Table 2. Values for the Increment Factor, ΔB_{MOX}

		UO_2	MOX
Molecular weight, W	g	~270	~271
Lattice parameter, a_0	å	5.470	5.470 - 0.074q
Melting temperature, T_m	℃	2,840 - 3.2BU	2,840 - 500q - 3.2BU

** BU : burnup in MWd/kgHM

The thermal conductivity of the matrix is derived from the Halden' s model and some relevant information on MOX fuel. It has been reported by laser flash method that the thermal conductivity of MOX pellet is influenced mainly by the stoichiometry that results in the significant reduction of thermal conductivity for non-stoichiometric fuel. However, the effect of stoichiometry measured at out-of pile seems ambiguous under irradiation since the thermal behavior of different stoichiometric fuels shows almost the same or sometimes the opposite of what is expected from the thermal conductivity observed at out-of-pile.

Considering the increment induced by the Pu addition, the phonon-defect term can be given by the expression

$$A_{mairix} = 0.1149+0.0035 \cdot BU+\Delta A_{MOX}$$

where ΔA_{MOX} is the increment of a constant A following the plutonium mixed in the matrix of MOX fuel. The Pu addition effect on the thermal behavior is determined in order to accommodate the lower thermal conductivity of MOX fuel up to 80 % of the value of UO_2. Therefore, the increment of A, is assumed to be proportional to the plutonium contents:

$$\Delta A_{MOX} = \beta \cdot q$$

The quantity of q is determined from the fraction of plutonium distributed in the matrix not in Pu-rich agglomerates.

Moreover, the newly suggested Pu addition factor would be expected to cover the thermal conductivity reduction through a possible modification of the stoichiometry in the range, 1.990 to 1.995 as soon as irradiation starts [12]. The lattice resistivity theory suggests the phonon-phonon term of

$$B_{matrix}=(2.475\times10^{-4} - 7.29\times10^{-7} \cdot BU) \cdot \Delta B_{MOX}$$

where ΔB_{MOX} indicates the increment due to plutonium distributed in the matrix and is given by

$$\Delta B_{MOX} = \left(\frac{W_{MOX}(q)}{W_{UO_2}}\right)\left(\frac{a_{0,MOX}(q)}{a_{0,UO_2}}\right)\left(\frac{T_{m,UO_2}(BU)}{T_{m,MOX}(q,BU)}\right)$$

The parameters of ΔB_{MOX} are summarized in Table 2. The difference of all parameters between UO_2 and MOX are not so sufficient that the change of the phonon-phonon term is not influenced significantly by the plutonium contents.

Phillipponneau's model [5] is selected for the local thermal conductivity for Pu-rich particles, which is expressed by

$$k_{Pu}=\left(\frac{1}{1.528\sqrt{x}+0.00931-0.1055+0.44\cdot BU+2.885\times10^{-4}\cdot TK}+76.38\times10^{-12}\cdot TK^3\right)\cdot f(p)$$

where

$f(p)$ = correction factor for porosity

$$\left(=\frac{1}{0.864}\cdot\frac{1-p}{1+2\cdot p}\right)$$

p = porosity

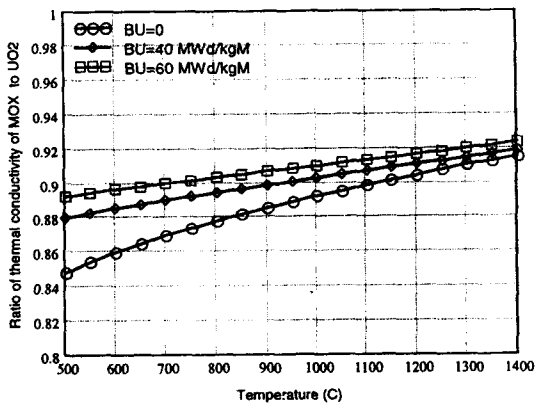

Fig. 2. Ratio of Homogeneous MOX to UO_2 Thermal Conductivity as a Function of Temperature

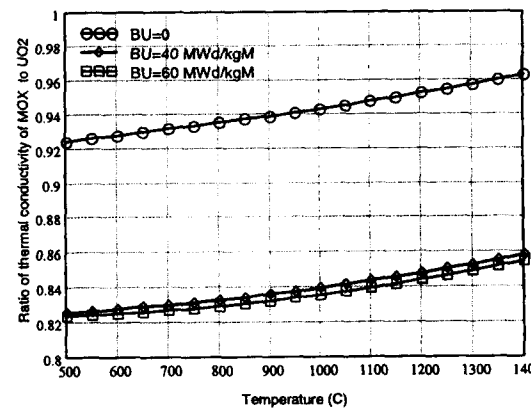

Fig. 3. Ratio of Inhomogeneous MOX to UO_2 Thermal Conductivity as a Function of Temperature

BU = burnup in MWd/kgHM not MWd/kgOX

x = stoichiometry

TK = temperature in Kelvin

Fig. 2 shows the ratio of homogeneous MOX to UO_2 thermal conductivity for the plutonium contents of ~7% which is the typical contents in the LWR MOX fuel. The thermal conductivity in the homogeneous MOX is estimated assuming the same degree of burnup degradation effect. The ratio of the thermal conductivity of MOX to UO_2 fuel increases with temperature, whereas the burnup makes the thermal conductivity increase significantly, especially in the lower temperature region. The thermal conductivity of MOX fuel ranges from 85 to 92% of UO_2 at the beginning-of-life.

Since little relevant data on Pu-rich particles are available, one of the international MOX program and Siemens-KWU [8] results are used only to characterize the microstructure of un-irradiated and irradiated fuel. The ratio of thermal conductivity of inhomogeneous MOX to UO_2 is plotted in Fig. 3 as a function of burnup and temperature. The fuel characteristics are based on those of MIMAS-AUC fuel. The newly proposed model indicates the reduction in MOX thermal conductivity ranging from 4 to 8 % compared to UO_2 fuel before irradiation. However, the ratio of MOX to UO_2 thermal conductivity decreases to ~82% to 86%, which indicates the higher burnup degradation effect in MOX fuel than UO_2 at the high burnup of 40 and 60 MWd/kgHM.

For both homogeneous and inhomogeneous MOX fuel, the difference between MOX and UO_2 thermal conductivity decreases with temperature. It should be noticed that the burnup effect on the thermal conductivity is more significant in inhomogeneous MOX than in homogenous MOX fuel. This is caused by assuming that the burnup is three times as high in the Pu-rich agglomerates as in the matrix. If the burnup in the agglomerate is lower, the burnup effect in a inhomogeneous MOX fuel would be reduced.

The reduction of MOX fuel' s thermal conductivity in Figs. 2 and 3 is comparable to ENIGMA' s [13] and Siemen' s [14] estimation, which indicate 8% less than that of standard UO_2 fuel and a relative decrease of 4~5 %,

respectively. However, it is slightly more conservative than BN' s results [15] which show the 4% reduction of thermal conductivity for 10% Pu/(Pu+U) fuel. The present characteristics are adequate for MOX fuel for verification of the developed model although they are not sufficient to accommodate all features of MOX fuel. It is needed more examinations for irradiated MOX fuel to clarify the properties of Pu-rich agglomerates.

3. Benchmark Calculation and Discussion

3.1. COSMOS Code

The developed thermal conductivity model has been incorporated into KAERI' s fuel performance code, COSMOS [7] and then verified using the data measured in the MOX irradiation test.

Since the estimation of radial power is required to get the accurate thermal behavior, a subroutine that calculates the radial power distribution for MOX fuel rods has been developed as a function of burnup and radial position based on a neutron physics calculation. The subroutine gives the radial power density for each radial ring when a pellet is divided into an arbitrary number of radial rings with equal volume and it has been incorporated into the COSMOS. The code is also equipped with a mechanistic fission gas release model for MOX fuel. Using the concept of an equivalent cell [16], the model considerers the uneven distribution of Pu within the UO_2 matrix and a number of Pu-rich particles that could lead to a non-uniform fission rate and fission gas distribution across the fuel pellet. The model was verified by the experimental data obtained from the FIGARO program, which consisted of the base irradiation of MOX fuels in the BEZNAU-1 PWR and the subsequent irradiation of four fabricated fuel segments in the Halden reactor. The calculated gas releases show good agreement with the measured ones. In addition, the developed analysis indicates that the microstructure of MOX fuel used in the FIGARO program is such that it has produced little difference in terms of gas release compared with UO_2 fuel [15].

Furthermore, the following in-pile phenomena have been also substantially upgraded continuously:

- Pellet-cladding mechanical interaction
- User-friendly input and output system with graphic interface
- Relocation at the beginning-of-life, densification, and swelling
- Cladding creep
- Cladding oxidation

3.2. Comparison with Measured Temperature

3.2.1. Homogeneous MOX Fuel at the Beginning-of-life

The developed thermal conductivity model is verified using the beginning-of-life thermal behavior of two homogeneous MOX fuel rods. The irradiated fuel rods have the typical PWR fuel specification. The fuel centerline temperature from the expansion thermometer (Fig. 4 (a)) and fuel thermocouple (Fig. 4 (b)) are given as a function of linear heating rate. The power corresponds to rod average linear heating rate in the expansion thermometer, and the local heating rate at the tip elevation for the fuel thermocouple, respectively. In the figure, the fuel surface temperatures are also included to check the effect of gap on thermal behavior.

The calculated centerline temperature shows very good agreement with the measured

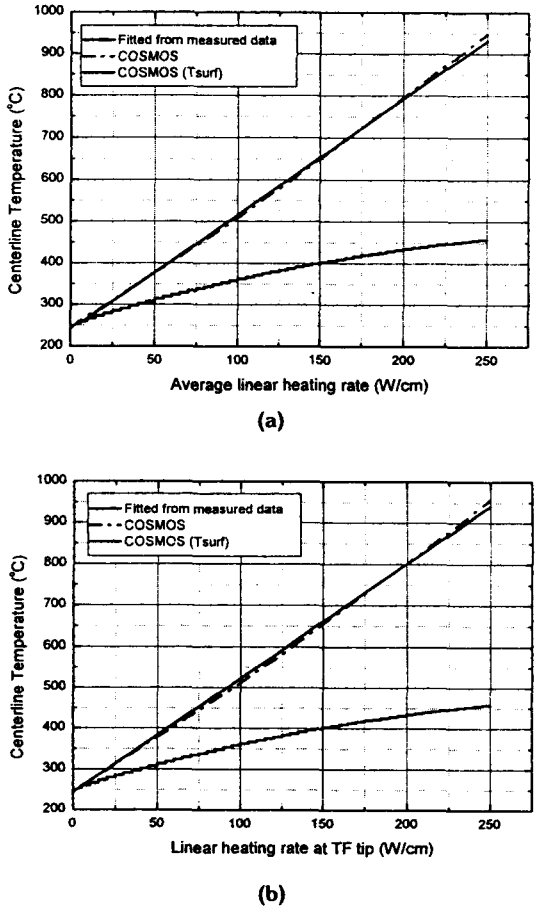

Fig. 4. Comparison of Measured Temperatures Using (a) Expansion Thermometer and (b) Fuel Thermocouple with the COSMOS Prediction for Homogeneous MOX Fuel

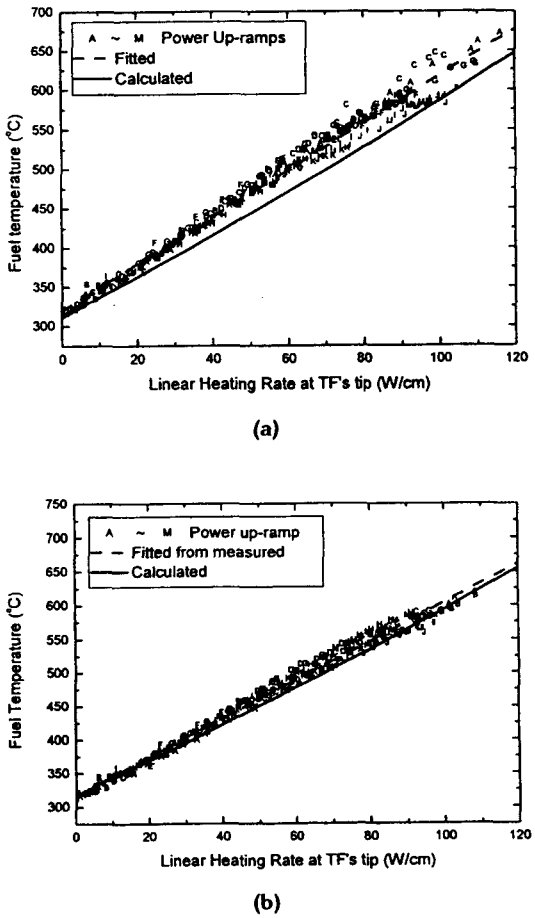

Fig. 5. Comparison of Measured Temperatures at Fuel Thermocouple' s Tip with the COSMOS Prediction for Inhomogeneous MOX (a) Rod 1 and (b) Rod 2

temperature. The fuel temperature increases linearly with linear heating rate. The fuel surface temperature is saturated since the gap width decreases with increasing the power in the fuel.

This calculation suggests that the developed thermal conductivity model well predicts the beginning-of-life thermal behavior in the homogeneous MOX fuel. It should be also noted that the COSMOS code simulates properly the thermal behavior with partly hollow pellets in which a thermocouple is inserted (Fig. 4 (b)).

3.2.2. Inhomogeneous MOX Fuel without Fission Gas Release

Two inhomogeneous MOX fuel rods were selected for re-irradiation test with instrumentation after base-irradiation up to the high burnup of ~50 MWd/kgOX. The instrumented MOX rods were irradiated under simulated PWR conditions. The

temperature was maintained below some threshold value to avoid the fission gas release. Two rods were equipped with a thermocouple in the center hole and except for the hole, the other part was solid. As shown in Fig. 5, the temperature for Rod 1 is slightly under-predicted whereas calculation of Rod 2 is consistent with the measured centerline temperature. The under-prediction for Rod 1 implies the possibility that the burnup effect in the present thermal conductivity model may not be considered enough to yield good agreement with measured temperature. In addition, it is noteworthy that the COSMOS code accurately predicts the in-pile behavior for re-instrumented MOX fuel base-irradiated in a commercial reactor.

3.2.3. Inhomogeneous MOX Fuel Tested for the Onset of Fission Gas Release

The main objective of the FIGARO program [15,17] was to evaluate the thermal behavior of MOX fuel at a burnup of about 50 MWd/kgHM and to determine whether the fission gas release threshold for MOX fuel was different from that for UO_2 fuel.

The fuel pellets were fabricated by the MIMAS-AUC process. A micronised master blend of UO_2 and PuO_2 powders were mixed with depleted UO_2 powder to reach the required Pu concentration. The main difference between two fuel rods was the grain size in the UO_2 matrix: 11 and 8μm, respectively. Two fuel rods were irradiated during five cycles at moderate power in Bezanu-1 PWR. Four segments, two from each rod, were cut at the same position for both rods and re-fabricated with a pressure transducer at the bottom and fuel thermocouple in the center hole at the top of fuel stack.

Fig. 6 shows the difference between the measured and the COSMOS calculated

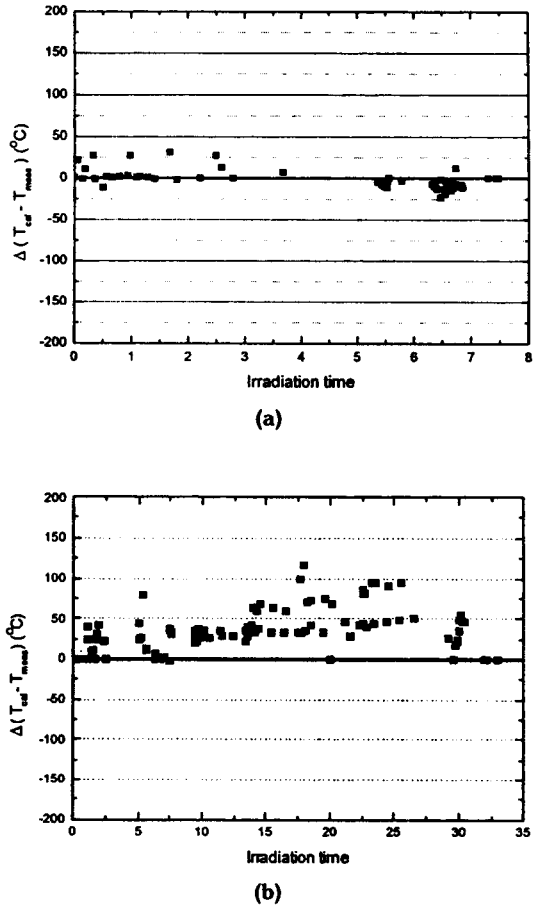

Fig. 6. Difference Between the Calculated and Measured Centerline Fuel Temperature for (a) MOX1 and (b) MOX2

temperatures for MOX1 and MOX2. During the calculation, the integral in-pile performance, including fission gas release and gap pressure, was also compared with the measured values. The comparison showed a good agreement between the measured and predicted values.

As shown in Fig. 6 (a), the COSMOS predicted the measured temperature within ±30°C without significant bias in one direction during irradiation time. On the other hand, COSMOS substantially

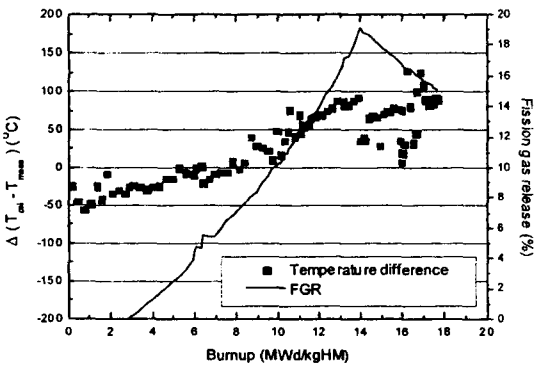

Fig. 7. Comparison of Calculated Fuel Centerline Temperature with the Measured Data

over-predicted the measured temperature for MOX2 in Fig. 6 (b). This over-estimation implies that thermal conductivity degradation is over estimated than that actually taking place in the irradiated fuel rods. That is, the irradiated pellets could have saturated thermal conductivity and/or exhibit recovered thermal conductivity at high burnup due to some fission gas release from irradiation damage. However, the saturation effect alone is not sufficient to explain the over-prediction for MOX2 fuel because both MOX1 and MOX2 fuel have almost the same burnup after base-irradiation. Therefore, the recovery of thermal conductivity seems to be more reasonable to give an explanation of the over prediction of temperature in MOX2 since the MOX2 rod experienced more fission gas release than that of the MOX1.

3.2.4. Inhomogeneous MOX Fuel with Significant Fission Gas Release

The developed thermal conductivity model was applied to verify the recent in-pile data obtained from the irradiation of MOX in the Halden reactor. The fuel rods were manufactured by MIMAS-AUC process. The main purpose of the test is to study thermal and fission gas release behavior of both solid and hollow MOX fuels.

The calculated fuel centerline temperature for solid fuel is compared with the measured values as shown in Fig. 7. The difference in the beginning of irradiation is insignificant between the measured and calculated temperature by COSMOS. However, COSMOS is under a bias towards over-prediction with significant fission gas release. As mentioned in 3.2.3, this over-estimation seems to be caused by the recovery of thermal conductivity after a substantial fission gas release occurred. Hence, it is needed that the recovery effect on thermal conductivity is included in the COSMOS code to get more accurate thermal performance of fuel with significant fission gas release.

4. Conclusions

A new mechanistic thermal conductivity model for MOX fuel was developed by considering its inhomogeneous microstructure. The general thermal conductivity model applicable to two-phase materials such as porous nuclear fuel was used to take into account the Pu-rich particles in the UO_2 matrix including the PuO_2 in solid solution. The area fraction and the distribution of plutonium input during the manufacturing of Pu-rich particles were determined from open literature information. The inhomogeneous MOX fuel rods irradiated to high burnup were characterized by FIGARO and Siemens-KWU results. The proposed model estimates that the MOX thermal conductivity with the Pu content of commercial LWR fuel ranges from 4 to 8% less than that of UO_2, which depends on the temperature and burnup. The developed thermal conductivity model was incorporated into a fuel performance code, COSMOS, and then verified

by the MOX in-pile database. A comparison of predicted centerline temperatures with the measured values shows reasonable agreement together with satisfactory results of the fission gas release and gap pressure when the amount of fission gas release is not sufficient to recover the irradiation damages. Consequently, it indicates the need for the introduction of the recovery factor in the burnup effect of thermal conductivity to analyze more realistically after significant fission gas release.

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