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Thermal Conductivity of a Simulated Fuel with Dissolved Fission Products[1]

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The thermal diffusivity of a simulated fuel with fission products forming a solid solution was measured using the laser-flash method in the temperature range from room temperature to 1673 K. The density and the grain size of the simulated fuel with the solid solutions used in the measurement were $10.49\,\mathrm{g \cdot cm^{-3}}$ (96.9% of theoretical density) at room temperature and $9.5\,\mu\mathrm{m}$, respectively. The diameter and thickness of the specimens were 10 and 1 mm, respectively. The thermal diffusivity decreased from $2.108\,\mathrm{m^2 \cdot s^{-1}}$ at room temperature to $0.626\,\mathrm{m^2 \cdot s^{-1}}$ at 1673 K. The thermal conductivity was calculated by combining the thermal diffusivity with the specific heat and density. The thermal conductivity of the simulated fuel with the dissolved fission products decreased from $4.973\,\mathrm{W \cdot m^{-1} \cdot K^{-1}}$ at 300 K to $2.02\,\mathrm{W \cdot m^{-1} \cdot K^{-1}}$ at 1673 K. The thermal conductivity of the simulated fuel was lower than that of UO_2 by 34.36% at 300 K and by 15.05% at 1673 K. The difference in the thermal conductivity between the simulated fuel and UO_2 was large at room temperature, and decreased with an increase in temperature.

KEY WORDS: laser-flash method; simulated fuel; solid solution; thermal conductivity; thermal diffusivity

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1. INTRODUCTION

The concept of the direct use of spent PWR fuel in CANDU reactors (DUPIC) is a dry processing technology to manufacture CANDU fuel from spent PWR fuel material without separating the fissile materials and fission products in the fuel. Spent PWR fuel typically contains 0.9 wt% fissile uranium and 0.6 wt% fissile plutonium, which exceeds the natural uranium fissile content of 0.71 wt%. The neutron economy of a CANDU reactor is sufficient enough to allow DUPIC fuel to be used in a CANDU reactor, which was originally designed for natural uranium fuel. The concept was proposed and termed DUPIC fuel cycle by the Korea Atomic Energy Research Institute (KAERI) and the Atomic Energy Canada Limited (AECL) with participation of U.S.A. in 1991 [1, 2]. The DUPIC fuel cycle offers several benefits to countries with both PWR and CANDU reactors: no need for spent PWR fuel disposal, savings on natural uranium resources for the fabrication of CANDU fuel, and an extended burnup of CANDU fuel by utilizing the DUPIC fuel.

The thermal properties of a nuclear fuel should be known to assess the behavior of the fuel elements at high temperature in a reactor. The main characteristic of a DUPIC fuel is its initial content of fission products as impurities. The thermal properties of a DUPIC fuel are expected to be different from a CANDU fuel because of the fission products. This causes adverse effects on the in-reactor fuel behavior, such as the thermal conductivity, thermal expansion, creep, fission gas release, and the swelling of the pellets. The thermal conductivity of a nuclear fuel is one of the most important properties because it affects the fuel operating temperature and maximum power of the nuclear power plant. The thermal conductivity, k, can be obtained from the thermal diffusivity, α, measured under transient conditions using the following equation because it is very difficult to measure the thermal conductivity under steady-state conditions at high temperatures (above 1500 K). The relation between the two properties is

$$k = \alpha c_p \rho, \tag{1}$$

where c_p is the specific heat at constant pressure and ρ is the density.

The importance of the thermal conductivity of UO_2 fuel for determining the fuel operating temperatures has led to numerous experimental and theoretical studies. The thermal conductivity, k, of irradiated UO_2 depends on the deviation from stoichiometry, x, the burnup, b, and the fractional porosity, p, as well as the temperature, T:

$$k = k(x, b, p, T), \tag{2}$$

Changes in the thermal conductivity occur during irradiation because of fission-gas bubble formation, pores, cracks, fission product build-up, and possible changes in the oxygen-to-uranium ratio (O/U). The dependency on the temperature and porosity has been studied extensively [3–5] and incorporated into computer codes used for in-pile fuel behavior analysis [6]. There are several studies on the effect of an impurity on the thermal conductivity of UO_2 [7, 8]. However, little work has been done on the effect of a burnup on the thermal conductivity because of the difficulty in dealing with high radioactive material. So, simulated spent fuel has usually been used to estimate the thermal diffusivity of an irradiated fuel. In the case of a DUPIC fuel, the direct measurement of the thermal properties is also very difficult in a laboratory due to its high level of radioactivity. As a part of the DUPIC fuel development program, the thermal properties have been investigated using a simulated fuel. Simulated fuel provides a convenient way to investigate the intrinsic fuel thermal properties. There are several studies on the thermal diffusivity of a simulated spent fuel. Lucuta et al. [9, 10] studied the thermal conductivity of a stoichiometric and a hyperstoichiometric simulated spent fuel with equivalent burnups of 1.5, 3, and 8 at%. They established the effects of the fission products of a simulated fuel. They reported that the thermal conductivity of a simulated spent fuel was lower than that of UO_2 and that a small increase in the O/U ratio (2.001) resulted in a slight decrease in the thermal conductivity. Each 1 at% burnup increase corresponds to a decrease in the thermal conductivity of about 6–9% at low temperatures (300 K) and 1–2% at high temperatures (1770 K). The thermal resistivity, i.e., the inverse of the thermal conductivity, increased linearly with the temperature and the burnup. However, it is difficult to distinguish between the effects of a solid precipitated fission product and the effects of a dissolved fission product on the thermal conductivity because they coexist in a simulated fuel. The dissolved fission products in UO_2 fuel reduce the thermal conductivity, and thus the precipitated fission products increase it.

In this paper, the thermal diffusivity of a simulated fuel with fission products forming a solid solution with UO_2 has been measured using the laser-flash apparatus in the temperature range from room temperature to 1673 K in order to investigate the effects of dissolved fission products in UO_2 on the thermal diffusivity. The thermal conductivity was calculated by combining the thermal diffusivity with the specific heat and density.

2. EXPERIMENTAL

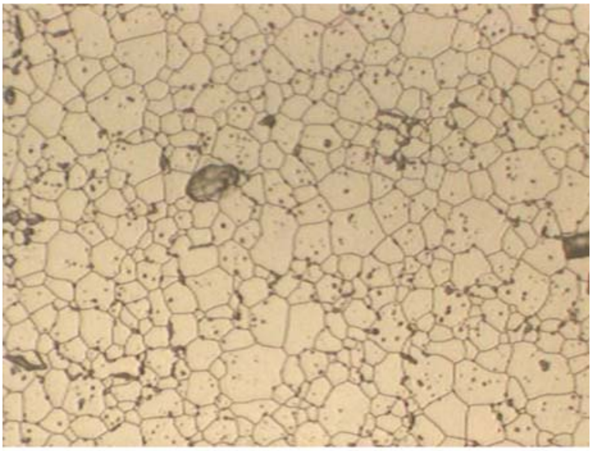
2.1. Sample Preparation and Characterization

Simulated spent fuel pellets with an equivalent burnup of 6 at% were used in this study. The specimens were fabricated by compaction and sintering the powder prepared by adding stable oxides as surrogates for the fission products into UO_2. The fission product composition of the irradiated fuel was determined by its initial enrichment and irradiation history. The ORIGEN (Oak Ridge Isotope Generation and Depletion) code was used to calculate the compositions of the fission products, which were added into the UO_2 powder. In this study only the fission products forming solid solutions were added into the UO_2 in order to confirm the effect of solid solutions on the thermal conductivity of a simulated fuel. Table I shows the contents of the fission products added into the UO_2 powder.

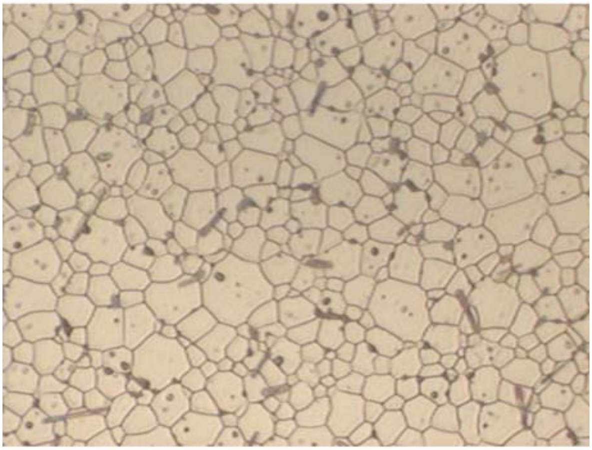
To prepare a simulated fuel, the mixed powder of UO_2 and the additives were pressed at 300 $MN \cdot m^{-2}$ into green pellets, and sintered at 1973 K for 4 h in a flowing 100% H_2 gas stream. Complete descriptions of the fabrication methods and characterization results have been provided in a previous publication [11]. The theoretical density of the simulated fuel was calculated by assuming that the fission products added to the UO_2 were fully formed solid solutions with UO_2. The density and the grain size of the simulated fuel with the solid solutions used in

Table I. Contents of Surrogates for Fission Products Added to UO_2 Powder

Fission Products	Simulated Fuel with Dissolved Fission Products (6 at%)	Simulated Fuel (3 at%) [10]	Forms in UO_2
Sr (SrO)	0.173	0.072	Solid solution
Y (Y_2O_3)	0.144	0.041	Solid solution
Zr (ZrO_2)	0.723	0.339	Solid solution
La (La_2O_3)	0.245	0.106	Solid solution
Ce (CeO_2)	0.477	0.285	Solid solution
Nd (Nd_2O_3)	0.816	0.460	Solid solution
Mo (MoO_3)	–	0.359	Metallic precipitate
Ru (RuO_2)	–	0.364	Metallic precipitate
Pd (PdO)	–	0.149	Metallic precipitate
Rh (Rh_2O_3)	–	0.028	Metallic precipitate
Ba ($BaCO_3$)	–	0.147	Oxide precipitate
Total	2.578	2.350	



(A) UO_2



(B) simulated fuel

Fig. 1. Optical microscope images of (A) UO_2 and (B) 6 at% burnup simulated fuel with dissolved fission products (500×).

the measurement were $10.49\,\mathrm{g \cdot cm^{-3}}$ (96.9% of theoretical density) and $9.5\,\mu\mathrm{m}$, respectively. It is also assumed that the specimens used in the experiment are stoichiometric because they are sintered in conditions of 100% H_2 at high temperature. The microstructures of the pellets are shown in Fig. 1. The UO_2 and the simulated fuel almost have the same microstructure and grain size.

2.2. Measurement of the Thermal Diffusivity

The thermal diffusivity of the simulated fuel with the dissolved solid solution in UO_2 was measured by the laser-flash method over the temperature range of 300–1673 K in a vacuum using a laser-flash apparatus (LFA 427, Netzsch). Disk samples with a 10 mm diameter and ~1 mm thickness were taken from the pellets for the thermal-diffusivity measurements.

3. RESULTS AND DISCUSSION

The thermal diffusivity of the simulated fuel is shown in Table II and Fig. 2 along with that of UO_2 [10] as a function of temperature for the purpose of a comparison. From the figure it is observed that the thermal conductivity of the simulated fuel and UO_2 decreases progressively with an increase in temperature. The thermal diffusivity of the simulated fuel with the solid solution decreases from $2.108\,\mathrm{m}^2\cdot\mathrm{s}^{-1}$ at 300 K to $0.626\,\mathrm{m}^2\cdot\mathrm{s}^{-1}$ at 1673 K. The effect of the additives is obvious as the results show a significant degradation of the thermal diffusivity of the simulated fuel with the dissolved fission products when compared to that of UO_2. The difference of the thermal diffusivity between the simulated fuel with the dissolved fission products and UO_2 is large at room temperature, and it decreases with an increase in temperature. This is a similar trend to the results of the simulated spent fuel measured by Lucuta et al. [10].

The thermal conductivity of the simulated fuel with the dissolved fission products in UO_2 was evaluated by multiplying the thermal diffusivity by the specific heat capacity and the density. The specific heat of the simulated fuels was measured by Verrall and Lucuta [12]. They acquired the results for three kinds of simulated fuels with equivalent burnups of 6 and 8 at% which were in close agreement with undoped UO_2. Fink [13] reviewed the available published data on the specific heat of UO_2 and recommended the best fitted equation. The specific heat capacity recommended by Fink was used in calculating the thermal conductivity of the simulated fuel. The density of the simulated fuel with the dissolved fission products measured by Kang et al. [14] was used in the calculation.

To consider the effect of the porosity we used the modified Loeb equation, i.e.,

$$k = k_{\mathrm{TD}}(1 - \beta P), \tag{3}$$

where P is the pore volume fraction, subscript TD refers to the theoretical density, and $\beta = 2.58\text{–}0.58 \times 10^{-3}\,\mathrm{T}$.

Table II. Thermal Diffusivity and Thermal Conductivity of Simulated Fuel

Temp. (K)	Specific Heat ($J \cdot kg^{-1} \cdot K^{-1}$)	Density ($kg \cdot m^{-3}$)	Diffusivity ($m^2 \cdot s^{-1}$)	Thermal Conductivity ($W \cdot m^{-1} \cdot K^{-1}$)				
				Measured	95% TD	UO_2 [10]	3 at% [10]	8 at% [10]
300	236.57	10,490.8	2.108	5.232	4.973	7.576	5.395	4.010
473	277.56	10,437.7	1.585	4.592	4.375	6.424	4.858	3.724
673	298.91	10,373.2	1.213	3.761	3.593	5.086	4.079	3.353
873	308.4	10,305.4	0.997	3.169	3.035	4.329	3.618	2.958
1073	312.62	10,234.4	0.857	2.742	2.633	3.492	3.062	2.654
1273	316.03	10,160.2	0.762	2.447	2.356	3.003	2.735	2.386
1473	322.3	10,082.8	0.692	2.249	2.171	2.715	2.442	2.168
1673	334.59	10,002.2	0.626	2.095	2.02	2.378	2.178	1.951

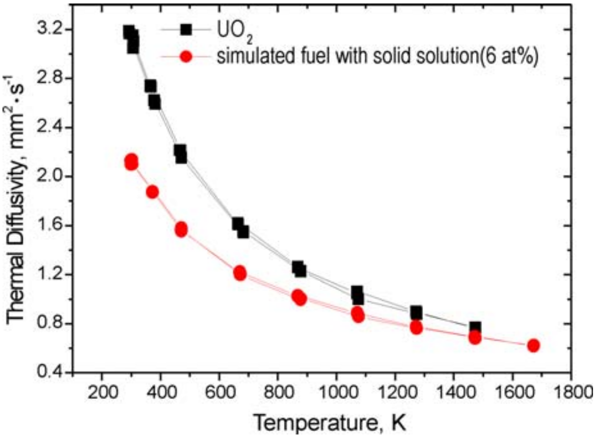


Fig. 2. Thermal diffusivity as a function of temperature for UO_2 and 6 at% burnup simulated fuel with dissolved fission products.

Therefore, the thermal conductivity normalized to 95% of TD, k_{95} is given by

$$k_{95} = k_M(1-0.05\beta)/(1-\beta P), \tag{4}$$

where the subscripts M and 95 denote the measured value and the value corresponding to 95% of the TD, respectively.

The thermal conductivity of the simulated fuel, normalized to 95% of the TD, is shown in Table II for various temperatures between 300 and 1673 K. It is plotted in Fig. 3 against temperature with the thermal conductivity of other simulated fuels and UO_2 which were measured by Lucuta et al. [10].

From the figure, as expected, it is observed that the thermal conductivity of the simulated fuel with the solid solution is lower than that of UO_2 and the difference decreases progressively with temperature. The thermal conductivity of the simulated fuel decreases from 4.973 $W \cdot m^{-1} \cdot K^{-1}$ at 300 K to 2.020 $W \cdot m^{-1} \cdot K^{-1}$ at 1673 K. At 300 K, the thermal conductivity of the simulated fuel is 65.64% that of UO_2 and at 1673 K it is 84.95% that of UO_2. Most of the difference in the thermal conductivity of the simulated fuel when compared with fresh UO_2 is due to the difference in the thermal diffusivity; the differences in density and specific heat had a small effect.

It is well known that heat in UO_2 is transferred by lattice vibrations and electrons. The electronic mobility is too slow to provide a significant

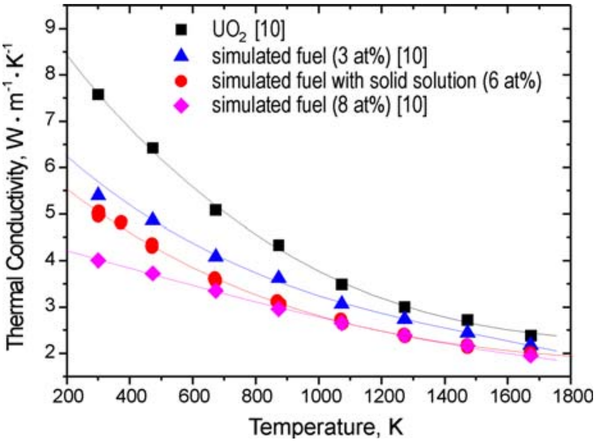


Fig. 3. Thermal conductivity of simulated fuels and UO_2 as a function of temperature.

contribution at temperatures lower than around 1800 K, and consequently a heat transfer mode by lattice vibrations is dominant. The degradation of the thermal conductivity of the simulated fuel is due to the additives that cause scattering of the phonons. At low temperature the effect of scattering of the phonons is high; however, heat transfer by electrons within UO_2 increases as the temperature increases. Thus, as mentioned above, the thermal conductivity of the simulated fuel with the solid solution is lower than that of UO_2 and the difference decreases progressively with temperature.

The thermal conductivity of the 6 at% burnup simulated fuel with the dissolved fission products is lower than that of the simulated fuel of a 3 at% burnup. It is higher than that of a 8 at% burnup below a temperature of 1000 K; however, it becomes the same above 1000 K. The total amounts of additives are 2.578 wt% for the 6 at% burnup simulated fuel, with the dissolved fission products, 2.35 wt% for the 3 at% burnup simulated fuel, and 6.27 wt% for the 8 at% burnup simulated fuel. Simulated fuel of the 6 at% burnup with the dissolved fission products used in this study incorporates the fission products that form solid solutions only, while the simulated fuels of the 3 at% burnup and 8 at% burnup have fission products that form metallic and oxide precipitates as well as solid solutions. Generally, since the metallic precipitates in the simulated fuels intuitively should increase the thermal conductivity, the reduction is caused primarily by the dissolved fission products. The temperature dependence of the thermal resistivity ($R = 1/k$) of the simulated fuels and UO_2 is shown in Fig. 4 as a function of temperature.

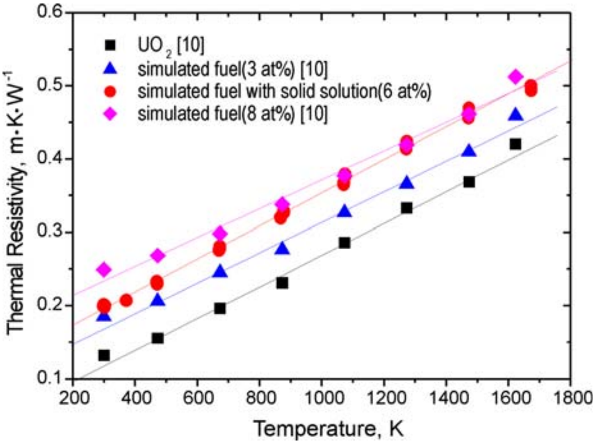


Fig. 4. Thermal resistivity of simulated fuels and UO_2 as a function of temperature.

The thermal resistivity of the simulated fuel with the dissolved fission products increases linearly with temperature up to 1673 K. This linearity is similar to the results obtained by Fukushima et al. [8] for single additive tests and by Lucuta et al. [9, 10] for simulated spent fuel. This linearity indicates that the thermal conductivity can be expressed as a function of temperature by using the following equation,

$$R = 1/k = A + BT = R_l + R_p, \tag{5}$$

where T is the absolute temperature, A and B are constants, R_l is the thermal resistivity caused by phonon–lattice defect interactions, or the lattice defect thermal resistivity, and R_p is the thermal resistivity caused by phonon–phonon interactions based on the Umklapp process, or the intrinsic lattice thermal resistivity.

The value of A and B were determined by fitting straight lines to the data. For the simulated fuel with the dissolved fission products, we found that A was 0.126 $(m \cdot K \cdot W^{-1})$ and B was 0.226×10^{-3}. A is 0.053 and B is 0.22×10^{-3} for UO_2, A is 0.101 and B is 0.219×10^{-3} for the 3 at% burnup simulated fuel and A is 0.181 and B is 0.216×10^{-3} for the 8 at% burnup simulated fuel [10]. The value of A for the simulated fuel with the dissolved fission products is higher than that for UO_2. It shows a similar trend of an increase with the burnup of the simulated fuel. The value of B shows that the slope of the resistivity plot slightly decreases as the burnup of the simulated fuel increases. However, the value of B of the simulated fuel with the dissolved fission products is higher than that of UO_2.

It is thought that since the simulated fuel used in this study has no metallic precipitates to increase the thermal conductivity, the slope of the thermal resistivity curve is higher than that of UO_2.

The thermal conductivity of the simulated fuel with the dissolved fission products can be expressed as a function of the temperature by using the following equation:

$$k = \frac{1}{0.126 + 0.226 \times 10^{-3} T} \ \mathrm{W \cdot m^{-1} \cdot K^{-1}} \tag{6}$$

4. CONCLUSIONS

The thermal diffusivity of a simulated fuel with fission products that form a solid solution in UO_2, was measured using the laser-flash method over the range of room temperature to 1673 K. Based on the work reported in this paper, we can draw the following conclusions:

1. The thermal diffusivity decreased from $2.108\,\mathrm{mm^2 \cdot s^{-1}}$ at room temperature to $0.626\,\mathrm{mm^2 \cdot s^{-1}}$ at 1673 K.
2. The thermal diffusivity of the simulated fuel was lower than that of UO_2. The difference of the thermal diffusivity between the simulated fuel and UO_2 was large at room temperature, and it decreased with an increase in temperature.
3. The thermal conductivity of the simulated fuel calculated by combining the thermal diffusivity with the specific heat and density was also lower than that of UO_2. Most of the difference was due to the difference in the simulated fuel thermal diffusivity because the specific heat and density of the simulated fuel are similar to that of UO_2.
4. The thermal resistivity of the simulated fuel increased linearly with temperature up to 1673 K and can be expressed as a function of temperature by using the following equation:

$$R = 1/k = 0.126 + 0.226 \times 10^{-3} T$$

5. The data measured and calculated in this study will be useful for the performance evaluation of in-reactor fuel behavior.

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REFERENCES

1. I. J. Hastings, P. G. Boczar, C. J. Allan, and M. Gacesa, *Proc. Sixth KAIF/KNS Annual Conf.*, Seoul, Korea (1991).
2. J. S. Lee, K. C. Song, M. S. Yang, K. S. Chun, B. W. Rhee, J. S. Hong, H. S. Park, and C. S. Rim, *Proc. Int. Conf. on Future Nuclear Systems: Emerging Fuel Cycles and Waste Disposal Options Global '93*, Seattle, Washington (1993).
3. G. J. Hyland, *J. Nucl. Mater.* **113**:125 (1983).
4. J. H. Harding and D. G. Martin, *J. Nucl. Mater.* **166**:223 (1989).
5. D. G. Martin, *J. Nucl. Mater.* **110**:73 (1982).
6. MATPRO – *A Handbook of Materials Properties for Use in the Analysis of Light Water Reactor Fuel Rod Behaviour*, TREE-NUREG-1005, EG&G Idaho, Inc. (1976)
7. M. Hirai, *J. Nucl. Mater.* **173**:247 (1990).
8. S. Fukushima, T. Ohmichi, A. Maeda, and M. Handa, *J. Nucl. Mater.* **114**:312 (1983).
9. P. G. Lucuta, H. Matzke, and R. A. Verrall, *J. Nucl. Mater.* **223**:51 (1995).
10. P. G. Lucuta, H. Matzke, R. A. Verrall, and H. A. Tasman, *J. Nucl. Mater.* **188**:198 (1992).
11. K. H. Kang, K. C. Song, J. S. Moon, H. S. Park, and M. S. Yang, *Met. Mater.* **6**:583 (2000).
12. R. A. Verrall and P. G. Lucuta, *J. Nucl. Mater.* **228**:251 (1996).
13. J. K. Fink, *J. Nucl. Mater.* **279**:1 (2000).
14. K. H. Kang, K. C. Song, M. S. Yang, S. H. Lee, J. B. Ko, and S. W. Kim, *Int. J. Thermophys.* **27**:161 (2006).