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Molecular dynamics calculations of heat conduction in actinide oxides under thermal gradient

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ABSTRACT

Thermal conductivities of UO_2, PuO_2 and $(U_{0.8},Pu_{0.2})O_2$ have been investigated by non-equilibrium molecular dynamics (NEMD) simulation between 300 K and 2000 K. The thermal conductivity was directly calculated by the temperature gradient on the system according to Fourier's law in NEMD simulation. The thermal conductivity obtained from the NEMD simulation decreases with a decrease of the supercell size, which means the phonon scattering occurs at the system boundaries in the micro-system. In addition, the present NEMD simulation, as well as previous EMD simulation studies, clearly shows that the Umklapp process causes the decrease of thermal conductivity at high temperatures. When comparison is made with literature data, the calculated results obtained from the relatively small supercell are in good agreement with the measured ones for the above actinide dioxides.

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1. Introduction

Uranium dioxide (UO_2) and mixed oxide (MOX: UO_2-PuO_2) nuclear fuels have been used in light water commercial reactors and fast breeder reactors (FBRs), respectively. During operation, there is an extremely large temperature gradient in the fuels. Especially, in the MOX fuel, the central pellet temperature is more than 2200 K, and the outside temperature is about 1000 K due to its low thermal conductivity under the high heating rate of the FBRs. This thermal environment is unique to the MOX nuclear fuel. It is well known that such a large temperature gradient accompanying irradiation causes oxygen and actinide redistributions and fuel restructuring (Olander, 1976). Also, these behaviors are deeply related to the alteration of thermophysical properties, e.g. thermal conductivity and melting point, from those of the unirradiated (fresh) fuel. In order to elucidate these phenomena for the safe management of nuclear fuels, post-irradiation examinations have been performed rather than out-of-pile examinations because it is difficult to reproduce the in-pile conditions of the fuels by the out-of-pile examinations. In addition to these examinations, it is of great importance to understand the various phenomena occurring inside the fuel under the in-pile conditions. In the Japanese sodium fast reactor "Monju", about 20–30 mol% plutonium enriched MOX fuels are used. Therefore, 20 mol% Pu content was set for the calculating thermal conductivity of MOX fuels in this study.

For these purposes, computer simulation techniques have been extensively developed so far. Among them, the molecular dynamics (MD) simulation is successfully used to evaluate the microscopic (atomic) behavior of fuels, e.g. irradiation and diffusion behaviors. In addition, the macroscopic physicochemical properties, e.g. specific heat, thermal conductivity, can be evaluated. In previous studies, the authors have discussed various thermal properties, e.g. thermal conductivity (Arima et al., 2006; Uchida et al., 2010), melting point (Arima et al., 2009) and atomic diffusion (Arima et al., 2010). However, most of these properties have been obtained by the equilibrium molecular dynamics (EMD) simulation, where "equilibrium" means the thermodynamic equilibrium. For example, the thermal conductivity can be defined as the time integration of the heat flux autocorrelation function in the Green-Kubo formulas for the EMD. Furthermore, even in the non-equilibrium MD (NEMD) simulation, which is sometimes called the perturbed MD simulation, given by Evans and Morris (Evans and Morris, 1990), the

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temperature is used as one controlled parameter for the whole system.

In the present study, the authors focused on the NEMD simulation that can give the temperature gradient to the system. Thus, the simulated system can be directly compared with the real one. As the first step to understand the complex phenomena caused by the thermal gradient in the oxide fuels, the thermal conductivities, which are easily calculated by Fourier's law, were evaluated for UO_2, PuO_2 and $(U_{0.8},Pu_{0.2})O_2$ in the temperature range from 300 K to 2000 K. In addition, verification of the NEMD approach adopted in this study was done from the technical side of the simulation.

2. Methodology

In MD calculations, the motions of particles are determined by the interatomic potential function. Here, the Busing-Ida type potential function with partially ionic charges is adopted. This potential function consists of Coulomb, short-range and van der Waals terms, and is given by,

$$U_{ij}(r) = \frac{z_i z_j e^2}{r} + f_0(b_i + b_j)\exp\left(\frac{a_i + a_j - r}{b_i + b_j}\right) - \frac{c_i c_j}{r^6} \quad (1)$$

where r is the distance between i and j ions, and z_i is the effective ionic charge of i ion. In this potential function, the ionic bonding of 67.5% was assumed in the present study. For example, the effective ionic charge of U is $+4 \times 0.675 = +2.7$. In Eq. (1), f_0 is an adjustable parameter ($= 4.186$ JÅ^{-1}mol^{-1}). Potential parameters, a_i, b_i, and c_i, are given for the i ion. In the present study, the potential parameters proposed by Inaba et al. (Inaba et al., 1999) and Arima et al. (Arima et al., 2005) were used (Table 1). These potential parameters were determined based on experimental literature data, e.g. their thermal expansions and bulk moduli. MD calculations were performed by using a customized MXDORTO program; the original program was developed by Kawamura and Hirao (Hirao and Kawamura, 1994). To give the temperature gradient to the simulated system, the momentum exchange algorithm was implemented in the customized program (Draghi and Avalos, 2003; Plathe, 1997). This momentum exchange algorithm is explained in the following.

In this algorithm, the supercell is long in the z direction (Fig. 1) and is divided into several thin slabs. The temperature of the slab, T_{slab}, is defined as

$$T_{slab} = \frac{1}{3N_{slab}k_B}\sum_{i=1}^{N_{slab}} m_i v_i^2 \quad (2)$$

where N_{slab} is the number of particles in the slab and k_B is the Boltzmann constant. To maintain the heat flux and the temperature gradient on the multi-component system, i.e. the oxides, the particle with the largest kinetic energy from the far left slab and that with lowest kinetic energy from the central slab are simultaneously selected, subsequently the energy and momentum of each particle is exchanged at an adequate frequency. Obviously, such an exchange keeps the total momentum constant as well as the total energy of the overall system. Therefore, assuming the elastic collision between two selected particles, the new velocity of particles of the central slab or far left slab, respectively, are given by

$$v_f^{new} = -v_f^{old} + 2\frac{m_f v_f^{old} + m_c v_c^{old}}{m_f + m_c} \quad (3)$$

$$v_c^{new} = -v_c^{old} + 2\frac{m_f v_f^{old} + m_c v_c^{old}}{m_f + m_c} \quad (4)$$

where m_c and m_f are the mass of particles selected in the central slab and far left slabs. As a consequence, the temperature of the central slab is the highest, and that of the far left slab is lowest. This is called the momentum exchange algorithm since $v_f^{new} = v_c^{old}$ and $v_c^{new} = v_f^{old}$ in the single component system, i.e. $m_f = m_c$ (Draghi and Avalos, 2003). The typical temperature gradient in the supercell is shown in Fig. 2. The heat flux in the z direction can be obtained, according to the expression

$$J_z = \frac{1}{2N\Delta t}\sum_{j=1}^{n_{ex}}\frac{1}{2A_j}m_c\left\{\left(v_c^{new}j\right)^2 - \left(v_c^{old}j\right)^2\right\} \quad (5)$$

Here, n_{ex} is the number of momentum exchanges, N is the total number of time steps during the simulation, and Δt is time per one step ($=2.0 \times 10^{-15}$ s). As can be seen from Fig. 1, the energy transported to the hot slab from the cold slab is flowing to both sides from the center. Therefore, the factor 2 in front of N indicates this effect. In this expression, A is the cross-sectional area of the x-y direction.

MD calculations were performed for the supercell which consisted of $(n_x, n_y, n_z) = (3, 3, 40–160)$ fluorite unit cells using the NPT ensemble with the Nose thermostat. The temperature was varied from 300 K to 2000 K, and the pressure was fixed at 0.1 MPa. Thermal conductivity (κ) was calculated from Fourier's law, Eq. (6), using the temperature gradient (dT/dz) and the heat flux (J_z) obtained from the exchange frequency of kinetic energy.

$$q_z = -\kappa\frac{dT}{dz} \quad (6)$$

In order to compare calculated and measured thermal conductivities, all thermal conductivities were corrected to the full dense oxide (porosity = 0), using the Brand-Neuer formula (Fink, 2000; Brandt and Neuer, 1976):

$$\kappa_0 = \frac{\kappa_p}{1 - \alpha p} \quad (7)$$

with α=2.6–0.5·[T(K)/1000], where κ_p is the thermal conductivity with porosity p and κ_0 is the thermal conductivity with $p = 0$.

Table 1
Parameters of the Busing-Ida type potential function used in the present simulation.

i	z_i	a_i (Å)	b_i (Å)	c_i (kJ$^{0.5}$ Å ^{3}mol$^{-0.5}$)	Reference
O^{2-}	−1.35	1.847	0.166	83.7	(Inaba et al., 1999)
U^{4+}	2.7	1.318	0.036	0	(Arima et al., 2005)
Pu^{4+}	2.7	1.272	0.0325	0	

3. Results and discussion

3.1. Exchange frequency dependence of thermal conductivity

In the momentum exchange algorithm, the heat flux and the temperature gradient are determined by the frequency of replacement of particle velocity. To examine the effect of exchange frequency on the thermal conductivity, it was varied from 0.002 to 0.01and the results are shown in Fig. 3. Here, Δn (unit in number of step) means the exchange interval of kinetic energy. As shown in this figure, accurate thermal conductivity values are obtained at high exchange frequencies. Meanwhile, at low exchange frequencies and low temperatures, thermal conductivity values are scattered. In the momentum exchange algorithm, the velocity exchange is taken place at every Δn steps, consequently, the whole of

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