Effect of grain boundary on lattice thermal conduction of tungsten revealed by molecular dynamics simulations

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ABSTRACT

Tungsten (W) and tungsten-based materials are being considered to be used in the divertor and the first wall as plasma facing materials (PFMs) in the fusion reactor, which requires them to withstand the heat loads. The influence of grain boundaries on the heat flux has been studied by the molecular dynamics (MD) simulations in this paper. Various grain boundaries have been constructed in simulation cells and their lattice thermal conductivities (LTCs) and thermal resistances have been calculated. It turns out that there exist sharp temperature drops across these grain boundaries, indicating that the LTCs near grain boundaries are much smaller than those inside the bulk tungsten. The grain boundary effect on the LTCs of polycrystalline W samples has been analyzed by the combination of MD and finite element results. The results may be potentially helpful for the design of the international thermonuclear experimental reactor (ITER) and the choice of PFMs.

1. Introduction

Tungsten (W) and tungsten-based materials are being considered to be used in the divertor and the first wall as plasma facing materials (PFMs) in the fusion reactor owing to their low sputtering yield, low tritium retention, no chemical erosion, good thermo-mechanical properties and high thermal conductivity [1–4]. However, melting, cracking and blistering might occur in W under such working condition [1,5–9]. These problems are correlated with radiation damage of W [1], which is closely related to lattice thermal conduction. This is because the damage in solids is caused by the displacements of atoms, which involves the lattice dynamics rather than electron dynamics. It is well known that the electron thermal conductivity takes up the main contribution to the total thermal conductivity in metals or alloys. However, the electron thermal conductivity has a little influence on the defect production if the electron thermal energies do not convert to the atomic kinetic energies. As both the radiation damage process and the phonon thermal conductivity are related with the lattice dynamics, understanding the phonon thermal conductivity is also helpful to account for the damage mechanisms.

Bai et al. [10] had recently described that grain boundaries (GBs) might have exceptional ability for materials to resist radiation damage, suggesting that polycrystalline W may be a better choice for PFMs. However, the PFMs must reliably withstand thermal cycles with heat fluxes of the order of 20 MW/m^2 up to several thousand times as required in the international thermonuclear experimental reactor (ITER) [11]. Under the impact of heat flux, the microstructure of W armor may be changed, accompanying with the degradation of the property. Therefore, it is important to know the role of GBs under the impact of heat flux, which motivates us to study the effect of GBs on lattice thermal conduction of W.

2. Simulation method

Molecular dynamics (MD) simulation has been a powerful method to research the microscopic processes of thermal conduction by phonons [12–18]. The lattice thermal conductivities (LTCs) can be calculated using either equilibrium MD (EMD) [13,18] or non-equilibrium MD (NEMD) [13–15,19]. The NEMD method, which calculates the temperature gradient (TG) of simulation system by maintaining the heat flux across a particular crystal plane, will be used in this work. The schematic representation of three-dimensional periodic simulation cell is shown in Fig. 1. The NEMD method has been summarized in Refs. [1,19]. The cell is divided into several slabs parallel to the yz plane. Kinetic energy is artificially transferred from the region at a quarter of the lateral dimension ($L_{x/4}$) to the region at three quarters of the lateral dimension ($L_{3x/4}$) and flows back due to thermal conduction. The heat flux is generated through exchanging the hottest atom in the region $L_{x/4}$ and the coldest atom in the region $L_{3x/4}$. The distribution of atomic kinetic energies is so wide that we can always find the hottest atom in the region $L_{x/4}$ to have more kinetic energy than

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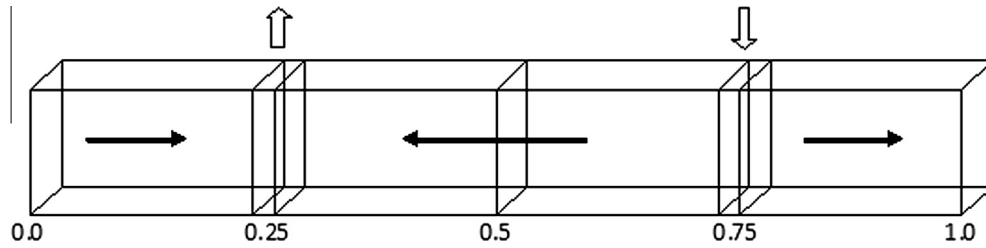
Fig. 1. Schematic representation of simulation cell with grain boundaries. The region at one quarter of the lateral dimension ($L_{x/4}$) is the cold region and the region at three quarters of the lateral dimension ($L_{3x/4}$) is the hot region. These black arrows denote the direction of thermal conduction in the simulation cell and the hollow arrows indicate the thermal conduction out of the simulation cell.

the coldest atom in the region $L_{3x/4}$. Therefore, the heat flux (J) formula can be given as following:

$$J = \frac{\Delta \varepsilon}{2AMt} \tag{1}$$

where t is time step and set as 1 femtosecond (fs) in this paper. And $\Delta \varepsilon$ is the kinetic energy difference between the coldest atom and the hottest atom, exchanged every M MD steps, M is set as 20 in this paper, which has no great influence on the LTC. In the formula (1), A is the cross-sectional area of simulation cell. The heat flux can be conducted in both directions of x, so the heat flux should be divided by the number 2. The TG along the x direction can be obtained by the curves between the temperature and the average abscissa values of these atoms (T–x curves). In the statistical mechanics, the slab temperature can be deduced by the velocity of micro-particles (v_i) of one slab.

$$T = \frac{1}{Nk_B}\sum_{i}^{N} m_i v_i^2 \tag{2}$$

where N is the atomic number of the slab, m_i is the mass of the atom i, and k_B is the Boltzmann constant. Therefore, the TG can be obtained from the computed T–x curves. Based on the Fourier heat transfer law, the formula of LTC (κ_x) along the x direction can be presented as [1,19],

$$\kappa_x = \frac{J_x}{\partial T/\partial x} \tag{3}$$

where J_x is the heat flux along the x direction, and $\partial T/\partial x$ is the slope of the T–x curve.

Before the calculation, the equilibrium cell parameter (a) at 300 K has been determined with the constant pressure-constant temperature (Nose-Parrinello-Rahman) ensemble simulation, which makes use of a third-order predictor–corrector algorithm to integrate the equations of motion. In the remainder all the temperatures are set to 300 K by the initial thermalization. The dimension of simulation cell is depended on the crystallographic orientation and the GB type. In order to eliminate the finite size effect [1,19], the dimensions of various simulation cells should be as close as possible and the length of the x direction (L_x) is set as about 1000a, the lengths of the y and z direction (L_y and L_z) are set as about 5a.

Finnis and Sinclair (F–S) [20] proposed an empirical N-body potential for W with conventional central pair-potential part and N-body potential part. The total potential energy of system is expressed as

$$E = \frac{1}{2}\sum_{i,j} V(r_{ij}) - A\sum_{i}\sqrt{\sum_{j(\neq i)} \varphi(r_{ij})} \tag{4}$$

Where $V(r_{ij})$ is the central pair-potential energy between atom i and atom j, and can be expressed as

$$V(r_{ij}) = \begin{cases} (r_{ij} - c)^2(c_0 + c_1 r_{ij} + c_2 r_{ij}^2) & r_{ij} \leqslant c \\ 0 & r_{ij} > c \end{cases} \tag{5}$$

And $\varphi(r_{ij})$ is the charge density of atom i derived from the contribution of extra-nuclear electron of atom j. It can be expressed as

$$\varphi(r_{ij}) = \begin{cases} (r_{ij} - d)^2 + \frac{\beta}{d}(r_{ij} - d)^3 & r_{ij} \leqslant d \\ 0 & r_{ij} > d \end{cases} \tag{6}$$

The six parameters β, c, c_0, c_1, c_2 and d are free for fitting experimental data and the crystal parameters. These parameters can be taken from Ref. [20].

In the remainder the constant volume-constant temperature ensemble (canonical ensemble, NVT) has been carried out and the periodic conditions are applied in the three coordinate axis directions. We chose a total simulation time of 0.6 ns, which is enough to achieve the steady state.

We have recently studied the thermal conduction of tungsten through computing the LTC with the same method [1]. The different influencing factors including temperature, heat flux, crystallographic orientation and GB have been analyzed. And we have found that GBs are thermal barrier regions with low LTC. Therefore lattice defects may be one of the important factors determining the thermal conduction of W. It is our purpose to research the effect of various GBs on the LTC of W.

3. Results and discussion

As shown in the Fig. 1, the simulation cell contains GB when x is equal to $0.0L_x$, $0.5L_x$ or L_x. We have constructed several types of GBs. The GB parameters are shown in Table 1. N_{tot} is the total number of atoms in the simulation cell. For the coincidence site lattice (CSL) grain boundary, the rotation axis is the y axis of the simulation cell. And for the twist boundary, the rotation axis is twist axis and is x axis of the simulation cell. GB 1, GB 5 and GB 7 are twin boundaries. GB 3 is a $\Sigma 5$ CSL grain boundary, and the LTCs of $\Sigma 5$ GB have been calculated and analyzed in Ref. [1]. GB 9 is a twist boundary, of which twist axis is [$\bar{1}00$] and twist angle is 36.9 degree. The dimensions of the left and right of these simulation cells are also listed in the Table 1.

The variation of heat charge (ε) of the system including GB 2 can be seen in Fig. 2. GB 2 is $\Sigma 3$ CSL grain boundary, which is the example of the calculation and analysis process. The linear relation between heat charge and time (t) is very good, which means that the heat flux remains unchanged. The slope of this curve ($\Delta\varepsilon/\Delta t$) can be derived to 2.20802×10^{-3} eV/fs. Therefore, the heat flux can be obtained.

$$J_{GB2} = \frac{2.20802 \times 10^{-3}}{2 \times 3\sqrt{3} \times 2\sqrt{6}a^2} \tag{7}$$

As shown in Fig. 3(b), the temperature profile (T–x cure) of the GB 2 case, averaged 10^5 MD steps. For comparison, we also present