**Supplemental Material**

**Study of the thermal conductivity of a metal-coated multi-walled carbon nanotube using molecular dynamics atomistic simulations**

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During regular MD simulations such as equilibrium MD (EMD), non-equilibrium MD (NEMD), homogenous NEMD (HNEMD), the effects of energy exchange between electrons and atoms are completely ignored. This disregard would lead to erroneous outcomes and conclusions when the electronic contribution to the heat transport also exists in a material system [1].

Two-temperature model (TTM) coupled with NEMD which is based on inhomogeneous Langevin thermostat provides a mechanism to take the energy exchange between the electron gas and atoms into account. This method was proven to be effective when the electron-phonon coupling is pivotal such as the heat transport through a metal-semiconductor interface [1], shock-induced melting of Ni single crystal [2], thermal conductivity of SiC-reinforced aluminum metal matrix composite [3]. To incorporate energy transfer by the electronic subsystem in the model, the following heat diffusion equation is used [4].

*C­­e.ρe (∂Te/∂t) = ∇(κe∇Te) - gep(Te - Ta) + gsTa*  (1)

where *Ce* is the electronic specific heat, *ρe* is the electron density, *κe* is the electronic thermal conductivity, *gep* is the coupling constant for the electron-phonon interaction, and *gs* is the coupling parameter for electron stopping. These are all related to the material properties and may depend on the local temperature. The quasi one-dimensional structure is divided into finite number of cells. In each cell, *Ta* and *Te* are the average local temperatures of atomic subsystem (phononic temperature) and electronic subsystem (electronic temperature), respectively. This equation describes the timely evolution of electronic temperature at the end of each MD timestep where the total amount of energy loss by the atomic subsystem in each cell is taken as the input energy for the electronic subsystem in the same cell. This loss/gain in the energy would be balanced only when $g\_{ep}=\frac{3nk\_{B }γ\_{ep}}{m}$ and $g\_{s}=\frac{3nk\_{B }γ\_{s}}{m}$*,* where *kB* is Boltzmann’s constant (1.380648 × 10-23 J.K-1), *γep* and *γs* are the friction coefficients due to electron-phonon interactions and electron stopping, respectively, *m* is the mass of the atom and *n* is the number of atoms per unit volume. The results of the simulations are not presumed to be influenced by the size of the cell and the electronic contribution to the heat capacity can be assumed as being linearly proportional to the absolute temperature [5,6]: *Ce = σ T*  (2)

where *σ = 0.34π2D(εF)kB2* isSommerfield constant with *D(εF)* being the energy density of states evaluated at the Fermi energy [6]. Wiedmann-Franz Law is used to approximate the value of electron thermal conductivity [7]. Based on the assumption that the ratio of the thermal conductivity and electrical conductivity is a function of temperature, we have

$\frac{κ\_{e}}{σ}=\frac{π^{2}k\_{B}^{2}T}{3e^{2}}$ (3)

Using a constant value for the coupling parameter results in a good agreement with the experimental data [2], even though the electron-phonon coupling parameter depends on both the density and temperature. Therefore, for simplicity, we assume that the electron-phonon interaction is homogenous in the metal and the change of electron-phonon coupling strength near the boundary is ignored. Also, the electron stopping effect is considered as negligible, i.e. *gs= 0*. The value of *gep* is experimentally deduced from which *γep* is computed [8]. The parameters of the TTM-NEMD for Ni are listed in table 1.

Table 1- Input Parameters of the TTM model for Nickel

|  |  |
| --- | --- |
| parameter | Value (unit) |
| *Ce* | 21.82938×10-6 (eV/e-.K) |
| *ρe* | 0.18298 (e-/ Å3) |
| *κe* | 0.0575155 (eV/ps.Å.K) |
| *γep* | 5.575921 g/mol.ps |
| *gs* | 0 |
| *v0* | 0 |

The calculations to get the necessary input parameters for Ni follow,

1. **Electronic Specific Heat, Ce**: In order to find the electronic specific heat for Ni at 300K, we use *Ce = σ T.*

For Ni, σ=7.02mJ/mol.K2 [9], Thus, at 300K, $C\_{e}=7.02×10^{-3}\frac{J}{mol.K^{2}}×300 K×6.242×10^{18}\frac{eV}{J}×\frac{1 mol}{6.022×10^{23}e^{-}}= 21.82938 ×10-6 eV/e-.K$

1. **Electron density,**$ ρ\_{e}$**:** It is the number of electrons per unit volume. The density of pure Nickel is found to be 8.917 g/cm3 and the molar mass of pure nickel at room temperature is 58.6934 g/mol [10]. Therefore, the density of Ni in terms of atoms per cubic centimeter was calculated as follows,

$ρ ≈ \frac{8.917 ^{g}/\_{cm^{3}} × 6.022×10^{23} ^{atoms}/\_{mol}}{58.6934 ^{g}/\_{mol}}≈ 0.914893 ×10^{23} \frac{atoms}{cm^{3}}$

The number of valence electrons per each Ni atom is 2. Hence, the electron density $ρ\_{e}$, for Ni is calculated as follows:

 $ρ\_{e}≈ 0.914893 × 10^{23} ×2 \frac{e^{-}}{cm^{3}} × \frac{1}{10^{24}}\frac{cm^{3}}{A°^{3}} ≈ 0.18298 \frac{e^{-}}{A°^{3}}$

1. **Electron thermal conductivity, *κe*:** Wiedemann-Franz Law was used to approximate the value of electron thermal conductivity. Based on the assumption that the ratio of the thermal conductivity and electrical conductivity is a function of temperature, we have

$$\frac{κ\_{e}}{σT}=\frac{π^{2}k\_{B}^{2}}{3e^{2}}=L$$

 For Ni, electrical resistivity at 300K is 7×10-8 Ωm and Lorenz factor is 2.15×10-8 WΩ/K2 [7], therefore,

$$κ\_{e}=2.15×10^{-8}\frac{WΩ}{K^{2}}×\frac{1}{7×10^{-8}Ωm}×300K= 92.14 W/mK$$

 = 92.14 $\frac{J}{s.m.K}$×6.242×1018 $\frac{eV}{J}$×$\frac{m.s}{10^{22}ps.A^{o}}$

 = 0.0575155 $\frac{eV}{ps.A^{o}.K}$

1. **Electron-ion interaction coefficient, *γ*ep:** It has been experimentally found that *gep =* 3.6×1017 W/m3K [8]. *gep* is the coupling constant for the electron-phonon interaction and it is related to *γ*ep via the following formula: *gep* = $\frac{3nk\_{B }γ\_{ep}}{m}$, where *kB* is Boltzmann’s constant (1.380648 × 10-23 J.K-1), *m* is the mass of the Ni atom and *n* is the number of Ni atoms per unit volume. For Ni, density is 8.917 g/cm3.

$$n=\frac{8.917×10^{6}^{g}/\_{m^{3}}×6.022×10^{23}^{atoms}/\_{mol}}{58.6934^{g}/\_{mol}}= 0.914893×10^{29} atoms/m3$$

Therefore, ***γ*ep**$=\frac{3.6×10^{17}\frac{W}{m^{3}K}×58.6934\frac{g}{6.022×10^{23}}}{3×0.914893×10^{29}\frac{atoms}{m^{3}}×1.38065×10^{-23}\frac{J}{K}}= 5.575921 g/mol.ps$

**References**

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