NUMERICAL SIMULATION FOR THERMAL CONDUCTIVITY OF NANOGRAIN WITHIN THREE DIMENSIONS

by

Ya-Fen HAN*, Hai-Dong LIU, and Xue CHEN

School of Energy and Power Engineering, Northeast Electric Power University, Jilin, China

Original scientific paper
https://doi.org/10.2298/TSCI170816257H

In order to improve the accuracy of simulation, the lattice Boltzmann method was adopted to get the thermal conductivities of 3-D nanograins. For the wide application, the length of nanograins axis is between 1 nm to 9 nm, and the diameter ratio of gap to spherical segment is 0.2 to 0.9, 30 sets of results of numerical simulation were taken. Correlations were fitted from the results of numerical simulation by multiple linear regression analysis. Then, in the range of temperature between 294 K to 700 K, the temperature value was taken every 50 K. Then final fitted formula of thermal conductivity for nanograins was got by the binomial fitting method. The results of fitted formula agree well with the numerical results. The results show that the thermal conductivities decrease with the diameter of nanograins reducing within the 3-D spherical segment when the diameter ratio, δ, of the gap to spherical segment is fixed. The effective thermal conductivities would increase with the ratio, δ, increasing when the spherical segment diameter is fixed and the ratio is lower than 0.6. The thermal conductivities would remarkably decrease when the ratio is larger 0.6.

Key words: *nanograins, lattice boltzmann method, grain size, heat conduction, fitted*

Introduction

With the development of micron and nanotechnology, the researches on the thermal property of nanomaterials have attracted wide attention of researchers [1, 2]. Due to the limitations of nanoexperimental measurement techniques, theoretical predictions, and numerical simulations have become the primary means of the research for thermal properties of nanomaterials. When the solid materials are in nanoscale, the phonon ballistic transports play the dominant role, and the micro and nanoscale effects occur in the internal heat transfer [3-5]. So far the researchers have extensively researched on the nanowires and 1-D or 2-D nanofilms in the fields of the thermal properties of solid materials. The molecular dynamics and Monte Carlo methods are mainly used [6, 7].

In the fields of thermal science, the thermal insulation silica nanomaterials have become potential in the fields of aeronautics and astronautics and construction, due to the characteristics of light weight and low thermal conductivity [8, 9]. The nanograins constituting its solid skeleton are 3-D spherical structures. The solid grain sizes of the thermal insulation nanomaterial are generally 2-5 nanometer. So the Knudsen numbers are smaller (close to 0.1) and close to the phonon mean free path. The phonon heat transports are in the quasi ballistic

*Corresponding author; e-mail: hanyafen@126.com

transport region, which belong to the acoustic thick condition. The atom numbers are enormous in the solid grains, so numerous nodes are usually used in numerical calculation, and a large amount of computation time is needed to calculate the phonon scattering. In this case, the considerably high computational cost will be used by the molecular dynamics method [10]. Both the lattice Boltzmann method (LBM) and the Monte Carlo method have good computational accuracy for the simulation of phonon heat transport in the quasi ballistic transport region. But the Monte Carlo method also needs a great deal of computation time, for its calculation accuracy depends on the number of sampling besides the lattice density within the large size and high dimension of nanograins [11]. Considering the computational cost and accuracy, the LBM has more obvious computational advantages [12].

In LBM, complex macroscopic phenomena were replaced by regular microscopic particle motions. The LBM has the advantages of easy implementation and simple processing of boundary conditions. In fact, LBM has been widely applied to simulate the phonon heat transfer in solid material. Amona *et al*. [10] computed the thermal conductivity for in-plane and out of-plane directions at different surface scattering factors for the thin silicon film using LBM. Christensen and Graham [13] introduced a coupled lattice Boltzmann finite difference method to simulate the phonon heat transfer in a 2-D domain. The simulations of heat transfer in nanometerials of multidimensional structure were rarely found.

As aforementioned, high computational cost will be spent by numerical simulation when the scale of nanograin is in the subcontinuum. For facilitating engineering applications, it is important to obtain the correlation for calculating the thermal conductivities of nanograins.

In this study, the LBM was adopted to obtain numerical results of the heat transfer for 3-D nanograins of silica aerogel. The multiple linear regression analysis was utilized to get the fitted formulas calculating thermal conductivity of nanograins. The fitted formula showed the effects of geometrical sizes and temperature of nanograins on the thermal conductivities.

Physical and mathematical models

Physical model

The 3-D spherical segment model was established to demonstrate nanograins of silica aerogel, as shown in fig. 1. In the figure, d, is the diameter of spherical segment, A – the diameter of surface gap, and L – the length of axis. To facilitate the analysis, the dimensionless scale was taken as $\delta = A/d$. Given the boundary conditions: the constant temperature boundary was on the surface gap, and the left side was high temperature $T_1 = 301$ K; the right side was low temperature $T_2 = 300$ K; the spherical surface was adiabatic, and it is diffuse scattering boundary.

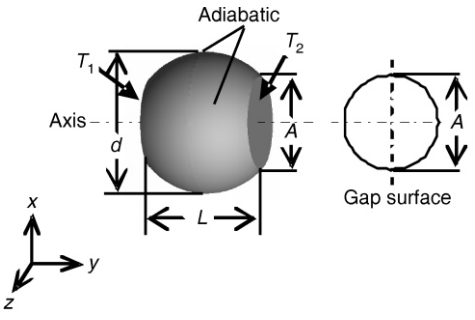

Figure 1. Geometry and boundary condition of spherical segment structure

The LBM

Boltzmann transport equation (BTE) was a general fundamental equation for treating energy transport problems:

$$\frac{\partial f}{\partial t} \quad v \quad f \quad a\frac{\partial f}{\partial v} \quad \frac{\partial f}{\partial t}_{c} \tag{1}$$

where f is the distribution function of particle, t – the time, and a – the acceleration.

The collision term in the right of eq. (1) was non-linear function. For simplicity, the re-

laxation time approximation was introduced. The BTE with relaxation time approximation was given:

$$\frac{\partial f}{\partial t} \quad v \quad f \quad \frac{f^{\mathrm{eq}} \quad f}{\tau} \tag{2}$$

where τ was the phonon relaxation time.

The BTE under the relaxation time approximation can be transformed to an equation on the phonon energy density formulation, and the lattice Boltzmann equation of each lattice point could be derived as [10]:

$$e_i(r \quad \Delta r, t \quad \Delta t) \quad (1 \quad \beta_i)e_i(r,t) \quad \beta_i e_i^{\mathrm{eq}}(r,t) \tag{3}$$

where $e_i(r, t)$ is the energy density distribution of discrete phonon, $e^{\mathrm{eq}}{}_i(r, t)$ – the energy density distribution of discrete equilibrium phonon, and β_i – the weight function.

The energy density distribution of discrete equilibrium phonon in lattice could be calculated by the formula:

$$e_i^{\mathrm{eq}}(r,t) \quad \frac{1}{m}\sum_{j \ 1}^{m} e_i(r,t) \tag{4}$$

where m is the total number of velocity direction in the lattice.

According to the lattice vibration energy and the calculations of states density and Debye frequency [11], the phonon energy density could be obtained:

$$e \quad \frac{9\eta k_b T^4}{T_D^3}\int_{0}^{T_D/T} \frac{x^3}{e^x \quad 1}\mathrm{d}x \tag{5}$$

where η is the number density of the wave vector, x – the dimensionless frequency ($\hbar\omega/k_bT$), ω – the frequency of phonons, $\hbar$ – the Planck constant divided by 2 , k_b [JK^{-1}] – the Boltzmann constant (=1.38 10^{-23}), and T – the temperature.

The heat flux in nanograins could be express:

$$q \quad \sum_{i \ 0}^{m} ve_i(r,t) \tag{6}$$

Based on the temperature and heat flux, the effective thermal conductivity could be solved through:

$$k_{\mathrm{eff}} \quad \frac{L \ q\mathrm{d}S}{\Delta T \ \mathrm{d}S} \tag{7}$$

where q is the steady heat flux through the media cross-section area dS at $y = 0$ boundary, and ΔT – the temperature difference of the left and right boundaries.

For the nanograins with 3-D structure, its numerical simulations were performed by the D3Q15 velocity model of LBM. The discretized velocity for D3Q15 model which was shown in fig. 2 was expressed:

$$c_i \quad \begin{cases} (0,0,0)v & i \quad 0 \\ (\ 1,1,0)v, (0,\ ,1,0)v, (0,0,\ 1)v & i \quad 1 \quad 6 \\ (\ 1,\ 1,\ 1)v & i \quad 7 \quad 14 \end{cases} \tag{8}$$

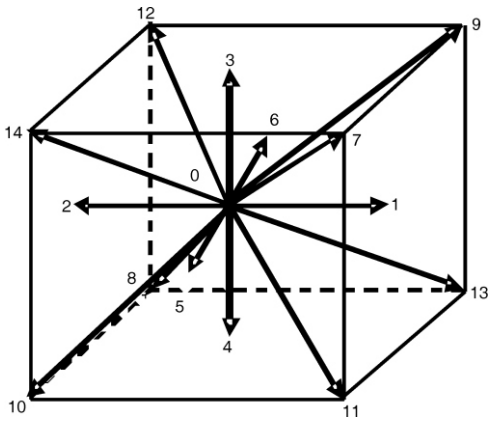

Figure 2. The D3Q15 model

The phonon energy transfer between lattices was restricted by the relation between the lattice distance Δr and time step Δt:

$$\Delta r \quad c_i \Delta t \tag{9}$$

Fitted by multiple linear regression analysis

The axis length of nanograin was defined as the characteristic scale. By investigating the results of numerical simulation, fig. 2, we could discover that nanograin thermal conductivities were affected by the length of axis and the ratio, δ, of diameter of gap to spherical segment. Besides, similar as literature [14], the basic form of fitted formula was structured:

$$\frac{k_{\text{bulk}}}{k_{\text{eff}}} \quad x \quad y\frac{\Lambda}{A} \quad z(\text{Kn})^{\theta} \tag{10}$$

where, x, y, z, and θ were the unknowns, and assumed,

$$\overline{Y} \quad \frac{k_{\text{bulk}}}{k_{\text{eff}}} \tag{11}$$

$$\overline{X}_1 \quad \frac{\Lambda}{A} \tag{12}$$

Substituting eqs. (11), (12) for eq. (10), we have:

$$\overline{Y} \quad x \quad y\overline{X}_1 \quad z(\text{Kn})^{\theta} \tag{13}$$

Considering the expression form of thermal conductivity for nanoscale thin film proposed by Majumdar [14], the value of θ should be in the field $U(1, \sigma)$. So the value of θ was respectively assumed to be 1.4, 1.3, 1.2, … , 0.6, and let:

$$\overline{X}_2 \quad (\text{Kn})^{\theta} \tag{14}$$

Then eq. (13) is changed to:

$$\overline{Y} \quad x \quad y\overline{X}_1 \quad z\overline{X}_2 \tag{15}$$

In multiple linear regression analysis, the y was fitted by n group values of (x_{1i}, x_{2i}, x_{3i}, ..., x_{mi}) (i = 1,2, ..., n) which were the observed value of the x_1, x_2, x_3, ..., x_{mi}.

The form of linear expression is assumed to be:

$$y \quad b_1 \quad b_2x_1 \quad b_3x_2 \quad \ldots\ldots \quad b_{m\ 1}x_m \tag{16}$$

where b_1, b_2,……, b_{m+1} were the regression coefficients. According to the least squares, the value of following eq. (17) should be the smallest.

$$\Phi \quad \sum_{i\ 1}^{n}[y_i \quad (b_1 \quad b_2x_1 \quad b_3x_2 \quad \ldots \quad b_{m\ 1}x_m)]^2 \tag{17}$$

So the regression coefficients should satisfy the following equation set:

$$(CC^T)\begin{matrix} b_1 \\ b_2 \\ b_3 \\ \vdots \\ b_{m\ 1} \end{matrix} \quad C\begin{matrix} y_1 \\ y_2 \\ y_3 \\ \vdots \\ y_n \end{matrix} \tag{18}$$

where

$$C\begin{matrix} 1 & 1 & 1 & \ldots & 1 \\ x_{11} & x_{12} & x_{13} & \ldots & x_{1n} \\ x_{21} & x_{22} & x_{23} & \ldots & x_{2n} \\ \vdots & \vdots & \vdots & \ldots & \vdots \\ x_{m1} & x_{m2} & x_{m3} & \ldots & x_{mn} \end{matrix} \tag{19}$$

The regression coefficients in eq. (18) could be solved by using the Cholesky separation method. When the length of nanograins axis was between 1 to 9 nm, and the diameter ratio of gap to spherical segment is 0.2 to 0.9, 30 sets of results of numerical simulation were taken. The values of x, y, and z in fitted eq. (15) were corresponding to the regression coefficients of equation set (18).

$$\begin{matrix} x & b_1 \\ y & b_2 \\ z & b_3 \end{matrix} \tag{20}$$

The numerical results were substituted in set of eq. (18), and then the regression coefficients of eq. (20) were got. These values were returned to fitted eq. (15), and compared with the numerical results without fitted computation. In the end, fitted computing results were better when the value of θ equaled to 0.8 by taken its different values close to 1. The x, y, and z were 1, 3, and 0.8 respectively.

Expression of thermal conductivity for spherical segment grains was obtain:

$$\frac{k_{\text{eff}}}{k_{\text{bulk}}} \quad \frac{1}{1 \quad 3\dfrac{\Lambda}{A} \quad 0.8(\text{Kn})^{0.8}} \tag{21}$$

where Kn is the Knudson number and it is defined as the ration of phonon mean free path and characteristic length of the system.

The phonon mean free path was used in the previous fitted formula, so it could be applied to computing the thermal conductivity of different materials with spherical segment structure. For silicon dioxide nanograins, the phonon mean free path was equal to 0.6 nm.

By observing the relationship between thermal conductivity and temperature of nanograins, fig. 3, the same law to bulk materials was revealed. In the range of temperature between 294 K to 700 K, and its value was taken every 50 K. Then final fitted formula of thermal conductivity for nanograins was got by the binomial fitting method. The eq. (22) showed the re-

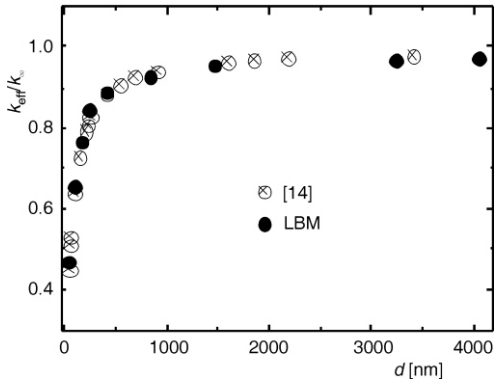

Figure 3. Comparison of normal thermal conductivity in Si films at 300 K

lationship of thermal conductivity, diameter of spherical segment gap, Knudsen number, and temperature of nanograins.

$$k_{\text{eff}} \quad \frac{1}{1 \quad 3\frac{\Lambda}{A} \quad 0.8(\text{Kn})^{0.8}} k_0 \qquad (22)$$
$$1.06\ \ 10^{\ 3}(T \quad T_0) \quad 6.19048\ \ 10^{\ 7}(T \quad T_0)^2$$

where k_0 is thermal conductivity of bulk material when 300 K and T_0 – the value equals to 300 K.

Equations (21) and (22) could be applied to non-metallic solid materials.

Program validation

In this paper, the normal phonon heat transports were numerically simulated in silicon film and the results were compared with the [14]. For silicon, the group velocity, mean free path and phonon relaxation time were 6400 m/s, 41 nm, and 6.53 ps, respectively.

The title name of *y* axis is film thickness and the vertical axis is the ratio of the film effective thermal conductivity to the bulk, and the title name of *x* axis is the thickness of silicon film in fig. 3. As can be seen from the diagram, the normal thermal conductivity would decrease with the decrease of the film thickness. The results from the developed LBM is reasonably matched with those of Majumdar [14], which are derived by using the Matthiessen rule for diffuse boundary scattering under the gray approximation.

Results and discussion

Results of numerical simulation

The phonon heat transfer was simulated in 3-D nanograins by LBM. For silica, the group velocity, mean free path and phonon relaxation time were 4100 m/s, 0.6 nm and 0.1463 ps, respectively.

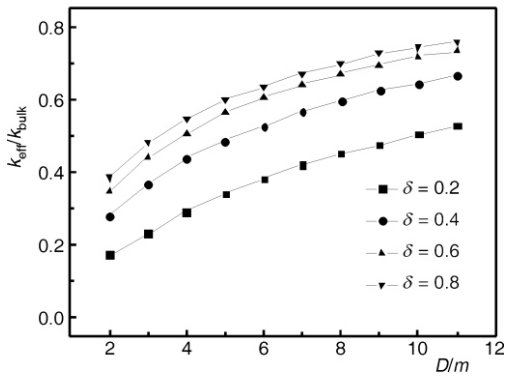

Figure 4. Effective thermal conductivities changing with the nanograin diameters at different δ

Figure 4 shows the thermal conductivity variation with the diameter of the spherical segment nanograin at the different δ, and the δ are 0.2, 0.4, 0.6, and 0.8, respectively. The nanograin thermal conductivities increased with the diameter increasing at all δ in fig. 4. This was because the phonon transports were in the diffusion-ballistic regions within the simulation scale. The regions would present boundary scattering effect. That was the phonon boundary scattering effect was enhanced and the additional thermal resistance was generated in the smaller size nanograins. Thus, the effective thermal conductivities become lower when the diameters of nanograin decrease.

In addition, the effective thermal conductivities increased with the δ increasing at same diameters of spherical segment in fig. 4. And the increasing intensities of the thermal conductivity become smaller and smaller with δ increasing. This was because that δ increasing causes the gap area of spherical segment increasing and axial size decreasing. Furthermore, the changings make the thermal conductivities increase. But the changing intensities will become weaker at lager δ. That was due to the effects of gap area of the spherical segment increase weaker than diameter decrease.

Results of fitted formula

Figure 5 indicates comparison of thermal conductivity results of numerical simulation and fitted eq. (21) when the temperature of nanograins was 300 K and the value of respectively was 0.3 and 0.7. We could find that thermal conductivities of nanograins would increase when the diameter and axis of spherical segment increased. The results of fitted formula were in better agreement with numerical simulation.

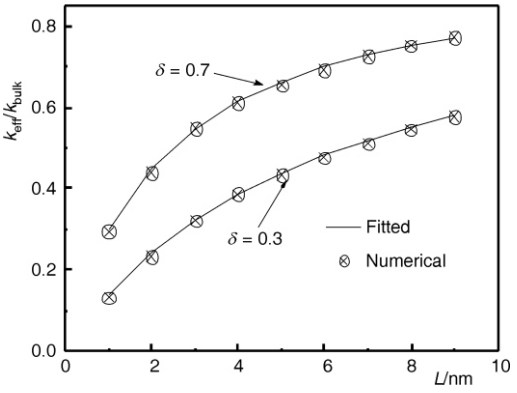

Figure 5. Fitted result of effective thermal conductivities of SiO_2 nanograin at 300 K

Figure 6 shows the effective thermal conductivities of the nanograin at different temperatures calculated from the fitted eq. (22), and its comparison with the numerical simulation results. As could be seen, the same variation trend of the thermal conductivity for nanograin to bulk material with temperature was revealed. The results of fitted formula are also in better agreement with the numerical results.

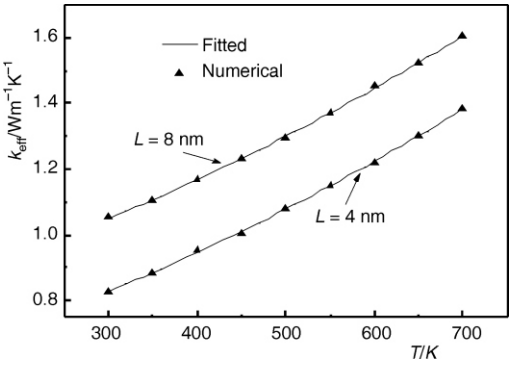

Figure 6. Fitted result of effective thermal conductivity of SiO_2 nanograin at different temperatures

Conclusion

The thermal conductivity of 3-D nanograins was fitted for numerical simulation results of LBM by multiple linear regression analysis in the work. The thermal conductivity of nanograins would rise with the increasing of diameter and axis of spherical segment. The effective thermal conductivities also increases with the diameter ratio, δ, of gap to spherical segment increasing when diameter of spherical segment was fixed. But the increasing degree would become lightly when the ratio was larger than 0.6. The results of fitted formula agree well with the numerical results. The variation of thermal conductivity of nanograins with temperature had the same law as the bulk materials. The variation of thermal conductivity of fitted results with temperature and scales from the fitted formula was consistent with the numerical results.

Acknowledgment

This work is supported by National Natural Science Foundation of China (Grant No. : 51606034), which are greatly appreciated.

Nomenclature

A – the force per unit mass
a – acceleration
d – the diameter of spherical segment, [m]
$\mathrm{d}S$ – the infinitesimal area vertical to direction of heat-flow, [m^2]
$e_i(r, t)$ – the energy density distribution of discrete phonon
f – the distribution function of particle
Kn – the Knudson number
k_0 – the thermal conductivity of bulk material when (T = 300 K) [$Wm^{-1}K^{-1}$]
k_{bulk} – the thermal conductivity of bulk material [$Wm^{-1}K^{-1}$]
k_b – the Boltzmann constant and (=1.38 10^{-23}), [JK^{-1}]
L – the axis length of spherical segment, [nm]
m – the total number of direction lattice point
q – the steady heat flux, [$Wm^{-2}K^{-1}$]
r – radius of spherical segment, [m]
Δr – the lattice distance, [nm]
S – surface
T_0 – the value equals to 300 K
T_1 – the left side temperature of the model, [K]
T_2 – the right side temperature of the model, [K]
ΔT – the temperature difference of the left and right boundaries, [K]
Δt – the time step, [s]
$\underline{v}$ – velocity
$\overline{X}_1$ – the value of vector
x – the dimensionless frequency, (x) $\hbar\omega / k_b^{**}T^{**}$

Greek symbols

η – the number density of the wave vector
θ – the parameter to be obtained
τ – the phonon relaxation time, [s]
ω – the frequency of phonons, [Hz]

Subscripts

eff – the effective value
c – collision of particle
eq – equilibrium of the discrete phonon direction of particle velocity
i – direction of particle velocity bul

References

[1] Bouhrour, A., *et al*., Thermomagnetic Convection of a Aagnetic Aanofluid Influenced by a Magnetic Field, *International Scientific Journal, 21* (2007), 3, pp. 1261-1274
[2] Fallah, K., *et al*., Mohammad Hossein Borghei, Simulation of Natural Convection Heat Transfer Using Nanofluid in a Concentric Annulus, *International Scientific Journal, 21* (2007), 3, pp. 1275-1286
[3] Yang, N., *et al*., Violation of Fourier's Law and Anomalous Heat Diffusion in Silicon Nanowires, *Nano Today, 5* (2010), 5, pp. 85-90
[4] Sellan, D. P., *et al*., Cross-Plane Phonon Transport in Thin Films, *Journal of Applied Physics, 108* (2010), 11, pp. 113524
[5] Tian, Z. T., *et al*., A Molecular Dynamics Study of Effective Thermal Conductivity in Nanocomposites, *International Journal of Heat and Mass Transfer, 61* (2013), June, pp. 577-582
[6] Randrianalisoa, J., *et al*., Monte Carlo Simulation of Cross-Plane Thermal Conductivity of Nanostructured Porous Silicon Films, *Journal of Applied Physics, 103* (2008), 5, pp. 053502
[7] Murakaw, A., *et al.*, An Investigation of Thermal Conductivity of Silicon as a Function of Isotope Concentration by Molecular Dynamics, *Journal of Crystal Growth, 26*7 (2004), 3-4, pp. 452-457
[8] Lu, Z. X., *et al*., Multi-Scale Simulation of the Ttensile Properties of Fiber-reinforced Silica Aerogel Composites, *Materials Science & Engineering A, 625* (2015), Feb., pp. 278-287
[9] Liu, H., *et al*., Investigation of the Effect of the Gas Permeation Induced by Pressure Gradient on Transient Heat Transfer in Silica Aerogel, *International Journal of Heat and Mass Transfer, 95* (2016), Apr., pp. 1026-1037
[10] Amon, C.H., *et al*., Modeling of Nanoscale Transport Phenomena: Application to Information Technology, *Physica A, 362* (2006), 1, pp. 36-41
[11] Han, Y.F., *et al*., Comparison of Lattice Boltzmann Method and Monte Carlo Method for Modeling Phonon Heat Conduction, *Journal of Harbin Institute of Technology (New Series), 20* (2013), 5, pp. 75-80

[12] Han, Y. F., *et al*., Modeling of Phonon Heat Transfer in Spherical Segment of Silica Aerogel Grains, *Physica B: Condensed Matter, 6* (2013), July, pp. 58-63
[13] Christensen, A., Graham, S., Multiscale Lattice Boltzmann Modeling of Phonon Transport in Crystalline Semiconductor Materials, *Numerical Heat Transfer Part B, 57* (2010), 2, pp. 89-109
[14] Majumdar, A., Microscale Heat Conduction in Dielectric Thin Films, *Heat Transfer, 115* (1993), 1, pp. 7-16

Paper submitted: August 16, 2017
Paper revised: October 29, 2017
Paper accepted: October 30, 2017