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The impact of the number of walls on CNTs thermal conductivity – molecular dynamics approach

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Abstract: Improved thermal performance can be obtained by using die-attach adhesives e.g. epoxy with high conductivity fill materials such as diamond, copper, silver, alumina, aluminium nitride, berillium oxide etc.

Currently there is a lot of ongoing research in order to improve the basic thermal property of adhesives, which is mainly heat conductance. One of the most interesting idea is to take benefit of carbon nanotubes (CNT) promising properties to develop new composites with improved thermo-mechanical characteristics (in particular high thermal conductivity) with respect to traditional epoxy adhesives currently used in microelectronics industry.

To calculate the thermal conductivity of various types of CNTs molecular dynamics (MD) simulation was used (Materials Studio, Accelerys). The calculations was performed to find out the impact of CNT number of walls on their thermal conductivity.

Key words: carbon nanotube, molecular modelling, thermal conductivity,

1. INTRODUCTION

The modern society is heavy focus on devices which analyse lost of information form lots of sources like sensors, internet, keyboard, internal and external memory etc. The manufactures have to cope with wishes of consumer – the device should be: smaller, lighter, cheaper, has more functionality and more reliable. Due to those requirements heat flux in applications is increasing and new technologies and materials are developed.

Very high integration, miniaturization and reliability is possible to reach by the thermal management which plays the crucial role in electronic devices. There is a direct link between electronic component reliability and operating temperature. A rise in temperature from 75ºC to 125ºC can be expected in a five-fold increase in failure rate [1].

Nowadays, new packaging solutions are required to solve thermal management issues generated by the ongoing trend towards higher miniaturisation, higher speed and higher power dissipation in electronics devices. The primary thermal transport mechanisms and the commonly used heat removal techniques vary substantially from one packaging level to the next. First level of the packaging hierarchy is primarily concerned with conducting heat from the chip to the package surface and then into the printed wiring board. At this packaging level, reduction of the thermal resistance between the silicon die and the outer surface of the package is the most effective way to lower the chip temperature [1].

To assure very good heat dissipation from silicon dies the combination of a different approaches which would improve the thermal management techniques are conducted. Therefore a new materials for packaging are developed, and the numerical and experimental analysis of heat transfer in electronic components supported by the reliability tests are performed. In this paper the research on CNTs (Carbon NanoTubes) as a filler in epoxy thermally conductive composites (TCC) is presented.

Carbon nanotubes, discovered by Iijima in 1991 [2], have been one of the most explored material. CNTs are rolled graphene sheets of hexagonal array of the carbon atoms. Carbon nanotubes have a diameter from a few angstroms to tens of nanometers. This allotropic form of carbon has a very interesting physic properties: very large aspect ratio (up to 10 000) [3], high rigidity modulus (even 1 TPa) [4], high tensile strength (up to

60 GPa) [5] and low density. Moreover, the literature reports that the CNT's has a very high value of thermal conductivity (6600 W/mK for an individual SWNT – single-walled CNT and more then 3000 W/mK for an individual MWNT – multi-walled CNT) [6,7] and high electrical conductivity (10^6 S/m for SWNT and more then 10^5 S/m for MWNT). Such a good properties would make them suitable in polymer composites as a thermally and electrically conductive filler.

The heat conduction along the CNTs is assured by electrons and phonons. The thermal conductances of CNTs are usually reported as a sum of electron and phonon contribution. In case of TCC fillers, the phonon part seems to be more important than electron part due to the limitation in heat transfer through the thermal contacts between filler particles.

The very useful tool to predict the phonon part of thermal conductivity of CNT is molecular dynamics (MD) simulations which make possible research on molecular and atomic level. The calculations were performed to find out the impact of CNT number of walls on their phonon part of thermal conductivity.

2. MOLECULAR DYNAMICS (MD) SIMULATION

To predict the thermal conductivity of single SWNT and MWNT (Multi-Walled CNT) the nonequilibrium molecular dynamics (NEMD) method was used. NEMD was implemented in the commercial software Materials Studio. At the beginning forcefield COMPASS (Condensed-phase Optimized Molecular Potentials for Atomic Simulations Studies) was adjust to calculate interatomic chemical bonds and non-bonding potential energy.

In the Fig. 1 the schematic structure of CNT used in simulations are presented. The length of each tested CNTs was about 12 nm. CNTs were divided into equal parts. The set of atoms on left hand side end was named as "Cold" region and on the opposite end was named as "Hot" region.

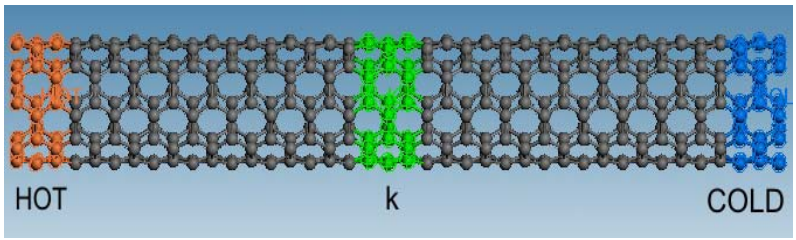

Fig. 1. Schematic structure of SWNT used in MD simulation

In this simulation, constant heat flux was imposed on the system and temperature gradient as result was calculated. The advantage of this method is fact that the heat flux which converges slowly is exactly known and need not be calculated. The temperature and its gradient converges rapidly in local regions. They were calculated over time and over particles in each region of CNT as well. The heat flux was applied by adding fixed quantity of kinetic energy ΔE_k in the "Hot" region and subtracting the same fixed quantity of kinetic energy ΔE_k in the "Cold" region. The method of applying of heat flux was proposed by Ikeshoji and Hafskjold [10]. In this algorithm the changes of kinetic energy in "Hot" and "Cold" regions is by scaling the velocities of each atom in both regions by the factor R and subtracting a small velocity v_{sub} from this scaled velocity. The values of R and v_{sub} was determinate to conserve the total angular momentum and they was calculated in every time step in the following way:

$$E_k = \frac{1}{2}\sum m v_i^2 \tag{1}$$

$$\vec{P} = \sum m\vec{v}_i \tag{2}$$

$$A = E_k + \frac{1}{2}\left(\frac{P}{N}\right)^2 \sum \frac{1}{m} - \left(\frac{\vec{P}}{N}\right)\sum \vec{v}_i \tag{3}$$

$$B = 2E_k - \left(\frac{\vec{P}}{N}\right)\sum \vec{v}_i \tag{4}$$

"Hot" region:

$$R = 1 + \left[-B + \left(B^2 + 4A\Delta E_k \right)^{\frac{1}{2}} \right] / 2A \tag{5}$$

$$v_{sub} = \frac{(R-1)P}{N\,m} \tag{6}$$

"Cold" region:

$$R = 1 + \left[-B + \left(B^2 - 4A\Delta E_k \right)^{\frac{1}{2}} \right] / 2A \tag{7}$$

$$v_{sub} = \frac{(R-1)P}{N\,m} \tag{8}$$

The new values of velocities after rescaling:

$$\vec{v}_i' = R\vec{v}_i - \vec{v}_{sub} \tag{9}$$

Where E_k is the kinetic energy of the "Hot" or "Cold" region before scaling, P is the total angular momentum of "Hot" or "Cold" region before scaling, N is the number of atoms in "Hot" or "Cold" region, m is the mass of atom, v_i is the velocity of atom i before scaling, v_i' is the velocity of atom i after scaling, A and B are the temporary factors used in velocity rescaling.

In all simulation 1 fs as a time step was set. At the beginning of simulation canonical ensemble (NVT) with Berendsen thermostat for 20 ps were conducted. This operation make the system stable at the set temperature as is showed in Fig. 2.

Next operation was using micro-canonical ensemble (NVE) for 80 000 fs. During this time i the velocities of atoms in "Hot" and "Cold" regions n every 1 fs was rescaled. Duo to this the heat flux in the CNT was imposed.

The final operation was averaging atomic velocities over 1 ps and using this averaging to calculate the temperature gradient in CNT molecular model. The temperature distribution along CNT was shown in Fig. 3.

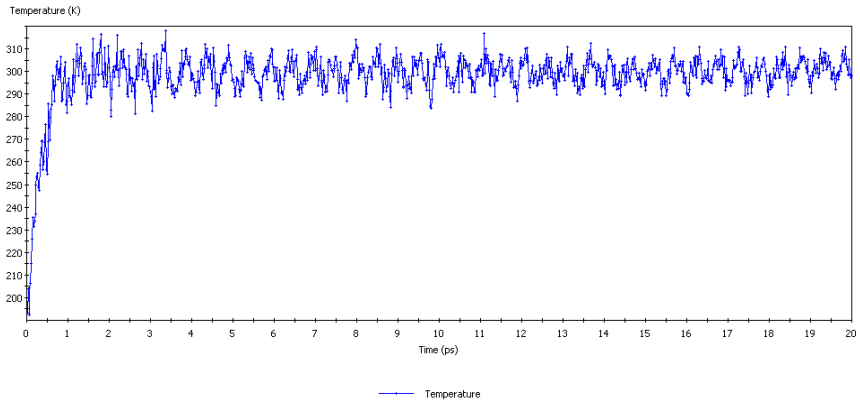

Fig. 2. Temperature of CNT during NVT with Berendsen thermostat

The temperature T_k of region k can be calculated as:

$$T_k = \frac{1}{3n_k k_B} \sum_{i=1}^{n_k} m_i v_i^2 \tag{10}$$

where n_k is amount of atoms in section k, m_i is the mass of i atom, v_i is the velocity of i atom and k_B is the Boltzmann constant.

With energy ΔE_k added in "Hot" region and subtracted in "Cold" region is possible to calculate heat flux as follows:

$$J = \frac{\Delta E_k}{A\Delta t} \tag{11}$$

where A is the cross section area of CNT, which is the ring with 3Å thickness, Δt is the time step in which the velocity of each atom in "Hot" region and "Cold" region was rescaling.

Thermal conductivity can be calculated from Fourier's law:

$$\lambda = -\frac{J}{\partial T / \partial z} \tag{12}$$

where $\partial T/\partial z$ is the temperature gradient along CNT which can be estimated by linear fitting from temperature distribution.

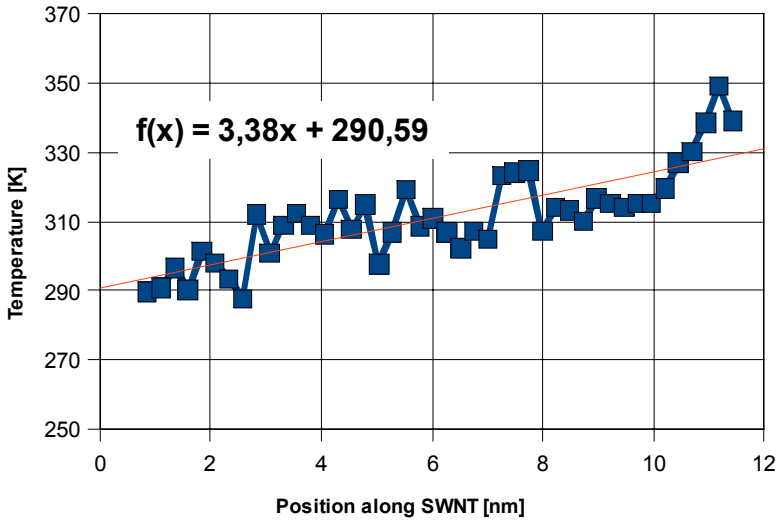

Fig. 3. Temperature distribution along the SWNT.

3. RESULTS AND DISCUSSION

The CNTs with (11,11) internal nanotube was generated. From second to fifth wallthe chiral vector was generated randomly by software.

At the beginning of simulation canonical ensemble (NVT) with Berendsen thermostat in 300 K for 20 ps were conducted. For each CNT the same energy ΔE_k equal $5.17 \cdot 10^{-22}$ J was applied. For the calculations of the cross-sections area for 3,4Å diameter of atoms was taken. The obtained simulation results are gathered in table 1 and presented in Fig. 4.

Table 1. Results of simulations

No. of walls	1	2	3	4	5
ΔE_k [J]	$5.17 \cdot 10^{-22}$	$5.17 \cdot 10^{-22}$	$5.17 \cdot 10^{-22}$	$5.17 \cdot 10^{-22}$	$5.17 \cdot 10^{-22}$
A [m^2]	$1.41 \cdot 10^{-18}$	$3.39 \cdot 10^{-18}$	$6.85 \cdot 10^{-18}$	$1.05 \cdot 10^{-17}$	$1.49 \cdot 10^{-17}$
J [W/m^2s]	$3.66 \cdot 10^{11}$	$1.53 \cdot 10^{11}$	$7.55 \cdot 10^{10}$	$4.91 \cdot 10^{10}$	$3.47 \cdot 10^{10}$
$\partial T/\partial z$ [K/m]	$3.38 \cdot 10^{9}$	$1.84 \cdot 10^{9}$	$1.03 \cdot 10^{9}$	$2.27 \cdot 10^{8}$	$3.12 \cdot 10^{8}$
λ [W/mK]	108	83	74	216	111

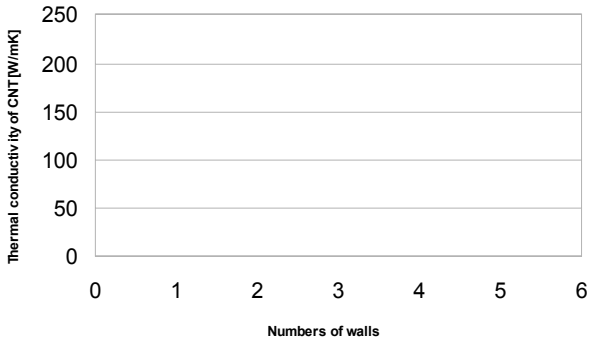

Fig. 4 The numbers of walls influence on thermal conductivity

Following the results in Fig.4 thermal conductivity is decreasing when the numbers of walls is increasing from one to three walls (from 108 to 73 W/mK). For the CNT with four walls the thermal conductivity has the highest value – 216 W/mK and for CNT with five walls – 111 W/mK.

The result obtained for four-walled CNT is quite unexpected. Such high value of thermal conductivity is probably result of too short time of simulation (80 ps) and the steady sate in the whole system was not reached.

4. CONCLUSIONS

The numbers of CNT walls influence on thermal conductivity was presented in this paper. The thermal conductivity of CNTs was obtained by using a commercial software Materials Studio (MS) ver. 4.4 Accelerys Inc. In this molecular dynamics simulations Ikeshoji and Hafskjold algorithm was used. It was implemented in Forcite module of MS by using PERL scripts.

The thermal conductivity for presented CNTs is much lower than presented in [6] and [7] because only the phonon part of thermal conductivity was taken into account. Nevertheless, the results obtained for SWNTs presented in [11,12] are in the same order of magnitude as our results.

For more complex systems as MWNTs longer time of simulation is required (hundreds or even thousands picoseconds).

5. ACKNOWLEDGEMENTS

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