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Thermo-mechanical properties of carbon nanotubes and applications in thermal management

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Abstract
Thanks to their very high thermal conductivity, high Young’s modulus and unique tensile strength, carbon nanotubes (CNTs) have become one of the most suitable nano additives for heat conductive materials. In this work, we present results obtained for the synthesis of heat conductive materials containing CNT based thermal greases, nanoliquids and lubricating oils. These synthesized heat conductive materials were applied to thermal management for high power electronic devices (CPUs, LEDs) and internal combustion engines. The simulation and experimental results on thermal greases for an Intel Pentium IV processor showed that the thermal conductivity of greases increases 1.4 times and the saturation temperature of the CPU decreased by 5 °C by using thermal grease containing 2 wt% CNTs. Nanoliquids containing CNT based distilled water/ethylene glycol were successfully applied in heat dissipation for an Intel Core i5 processor and a 450 W floodlight LED. The experimental results showed that the saturation temperature of the Intel Core i5 processor and the 450 W floodlight LED decreased by about 6 °C and 3.5 °C, respectively, when using nanoliquids containing 1 g l−1 of CNTs. The CNTs were also effectively utilized additive materials for the synthesis of lubricating oils to improve the thermal conductivity, heat dissipation efficiency and performance efficiency of engines. The experimental results show that the thermal conductivity of lubricating oils increased by 12.5%, the engine saved 15% fuel consumption, and the longevity of the lubricating oil increased up to 20 000 km by using 0.1% vol. CNTs in the lubricating oils. All above results have confirmed the tremendous application potential of heat conductive materials containing CNTs in thermal management for high power electronic devices, internal combustion engines and other high power apparatus.

Keywords: CNTs, thermal greases, nanofluids, nano lubricating oils, thermal management
Classification numbers: 5.11, 5.14

1. Introduction

Thermal management is widely recognized as being an important aspect of electronic devices such as computers and light emitting diodes (LEDs), since the device’s performance is significantly affected by the temperature of the chips. In
addition, the device’s lifetime can be decreased drastically because of large thermal stresses. The challenge for thermal management is to develop a high thermal conductivity medium that can accommodate temperature with increasing power density of the devices [1].

Carbon nanotubes (CNTs) are well-known nanomaterials with many excellent properties. CNTs are some of the most valuable materials, with high thermal conductivity of 2000 W m$^{-1}$ K$^{-1}$ compared with 419 W m$^{-1}$ K$^{-1}$ of Ag [2, 3]. This suggests an approach of applying CNTs in greases, liquids or lubricating oils for heat dissipation in computer processors, LEDs, engines, and other high power electronic devices [4–18]. In this paper, we present results on the fabrication of heat conductive materials containing CNT based thermal greases, nanofluids and lubricating oils, and application of these materials to thermal management in CPUs, LEDs and engines.

2. CNT based greases for thermal management in micro-processors

We developed two types of CNT based grease for heat dissipation in micro-processors:

A commercial silicon thermal paste (Stars) thermal grease for computers (Stars Company) containing CNTs, named CNTs/Stars.

An AS5 grease for computers (Arctic Silver Incorporated) containing CNTs, named CNTs/AS5.

The CNTs used in these greases were multiwall CNTs, produced using the thermal CVD technique with diameter and length in the range 15 nm–90 nm and several tens of $\mu$m, respectively [19]. The concentration of CNTs in this grease was in the range 0–7 wt%. The CNT thermal greases were pasted between the surface of the CPU and the heat sink as shown in figure 1. We chose a personal computer, Intel Pentium IV, 3.066 GHz, 512 MRAM, 80 GB Hard-disk and Window XP Service Pack 2 operating system, for the experiment. The temperature of the micro-processor was measured using SpeedFan 4.3.3 software and the micro-processor was pushed to operate at full load (100% speed of the micro-processor) by using Stress Prime 2004 ORTHOS software [10].

Figure 2 shows the measured temperature of the micro-processor as a function of working time in the case of CNTs/Stars thermal grease with different concentrations of CNTs: not using any thermal grease, and utilizing CNTs/Stars thermal grease with 0, 1, 2, 3, 5 and 7 wt% CNTs. It is clearly seen from figure 2 that without a thermal matching medium, the temperature of the micro-processor reaches 85 °C within 20 s and the computer was automatically shut down. This obviously confirmed the necessity of a thermal matching medium for the device. By adding CNTs (0, 1, 2, 3, 5 and 7 wt%) into the Stars thermal grease, the temperature of the micro-processor decreased gradually. For the case of 2 wt% CNTs/Stars grease, the temperature increase time and saturated temperature of the micro-processor were 200 s and 63 °C, respectively. Whereas these value are 75 s and 66 °C for the case of using commercial Stars thermal grease without any CNTs. The same effect was found for the case of using AS5 thermal grease instead of the Stars grease.

We developed a model for simulation of the measured data. The thermal dissipation system model can be expressed as shown in figure 3(a), where $I$ is heat-generating power of the micro-processor, $C_1$ is heat capacity of the micro-processor; $C_T$ is heat capacity of the thermal grease, $R_{la}$ is the heat resistance between the micro-processor and thermal grease layer, $R_{lb}$ is the heat resistance between the thermal grease layer and the cooling fan, $C_2$ is heat capacity of the cooling fan, $R_S$ is the heat resistance between the cooling fan and the environment, $U$ is the temperature of the environment. Since the heat capacity of the thermal grease is very much smaller than that of the micro-processor and the cooling fan,
the model in figure 3(a) can be simplified as shown in figure 3(b), where $R_1 = R_{1a} + R_{1b}$ is the heat resistance of the thermal grease layer, $U_1$ is the temperature of the microprocessor, and $U_2$ is the temperature of the cooling fan. Based on the model shown in figure 3(b), we have differential equations of thermal dissipation process as the following:

$$I_1 = (U_2 - U_1)/R_1,$$

$$C_1dU_1 = (I - I_1)dt,$$

$$I_2 = (U - U_2)/R_2,$$

$$C_2dU_2 = (I_1 - I_2)dt,$$

where $I_1$ is the heat current from the microprocessor to the cooling fan and $I_2$ is the heat current from the cooling fan to the environment. The micro-processor is an Intel Pentium 4 processor 524 (3.06 GHz). The dimensions of the microprocessor are $1.476\times1.476^\circ$, the mass of the microprocessor is 21.86 g, the heat-generating power is 84 W and the heat capacity is 15.6 J K$^{-1}$. The cooling fan consists of 7 cm$^2$ copper/aluminum and aluminum flake (see figure 1) and the total mass of the cooling fan is 220 g. In order to simplify the simulation process, we assumed that temperature of the aluminum flake is identical to the environment temperature and the heat resistance of the outside part of the aluminum flake is ignored. Hence, we determined the heat capacity of core fan was 89 J K$^{-1}$ and the heat resistance between the cooling fan and the environment was 0.43 K W$^{-1}$. Therefore we have $I = 84$ W, $U = 20^\circ C$, $C_1 = 15.6$ J K$^{-1}$, $C_2 = 89$ J K$^{-2}$ and $R_2 = 0.43$ K W$^{-1}$. At initial time, the temperatures of the micro-processor and the cooling fan were similar to the environment temperature, i.e. $U_1 = U_2 = U = 20^\circ C$. The simulation process was performed with different thermal greases with different $R_1$ values [10].

Figure 4(a) shows the simulation and experimental results of the micro-processor as function of operation time in the case of Stars thermal grease (a) and CNTs/Stars thermal grease (b) [10]. With permission from Elsevier.

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Figure 4(a) shows the simulation and experimental results of the micro-processor as function of operation time in the case of Stars thermal grease (a) and CNTs/Stars thermal grease (b) [10]. With permission from Elsevier.
thermal grease was 1.37 times higher than that of the STARS thermal grease alone [10].

We obtained similar results when using AS5 thermal grease instead of Stars grease. Figure 5(a) shows the simulation and experimental results of the micro-processor temperature as a function of working time when using the AS5 thermal grease without any CNTs. The results show that the heat resistance of the AS5 thermal grease layer and the thermal conductivity of the AS5 thermal grease were 0.027 K W\(^{-1}\) and 8.89 W m\(^{-1}\) K\(^{-1}\), respectively. Figure 5(b) shows the simulation and experimental results of the micro-processor temperature as functions of working time when using CNTs/AS5 thermal grease (2 wt% CNTs). The simulation results show that the heat resistance of the thermal grease layer and the thermal conductivity of CNTs/AS5 thermal grease were 0.015 K W\(^{-1}\) and 16.2 W m\(^{-1}\) K\(^{-1}\), respectively. This means that the thermal conductivity of the CNTs/AS5 thermal grease (2 wt% CNTs) was 1.82 times higher than that of AS5 thermal grease alone [10].

The above results confirm the advantage of CNTs as an excellent additive component in thermal greases for heat management in the micro-processors of PCs, LEDs and other high power electronic devices.

3. CNT based nanofluids for thermal management in micro-processors and LEDs

In addition to the CNT based thermal greases mentioned above, we also developed nanofluids containing CNTs for heat dissipation in CPUs and high power LEDs. A distilled water (DW) and distilled water/ethylen glycol (DW/EG) solution were used in our research as base fluids. The CNTs were functionalized with –COOH and –OH functional groups by chemical oxidation methods [13–16]. The existence of carboxyl (COOH) and hydroxyl (OH) functional groups bonded to the ends and sidewalls of the CNTs was proven by Raman and Fourier transform infrared (FTIR) spectra [15]. In order to disperse the functionalized CNTs in the liquids, we used the Tween-80 surfactant and an ultrasonic vibration treatment over 30 min. The functionalized CNTs were perfectly dispersed in the nanofluids with concentration from 0.1 to 1.3 g l\(^{-1}\). The dispersion of the CNTs in the nanofluids were proven by Malvern Zetasizer Nano ZS Instrument measurement [13, 15, 16]. The prepared CNT nanofluids showed good stability by using functionalized CNTs and the Tween-80 surfactant, due to a hydrophobic-to-hydrophilic conversion of the surface nature due to the generation of a hydroxyl group, and the Tween-80 surfactant provides lower surface tension of liquids and increases the immersion of CNTs [4, 20, 21].

Figure 6(a) shows a schematic view of the liquid cooling system for the computer processor using DW containing CNTs. The CNT nanofluid was pumped into the Cu substrate with 2 cm\(^3\) s\(^{-1}\) flow rate. The volume of the liquid tank was 500 ml. The environmental temperature was kept at 15 °C for all measurements. Figure 6(b) shows the measured result of the micro-processor temperature as a function of working time when using DW and CNTs/DW for heat dissipation. At initial time, the temperature of the micro-processor was about 16 °C. The saturated temperature of the processor reached 35 °C, 30 °C and 28 °C when using DW, 0.6 g CNTs/liter DW and 1 g CNTs/liter DW after 30 min working time, respectively. These results indicated that by mixing CNTs (1 g l\(^{-1}\)) in the DW, the saturation temperature of the CPU decreased by 7 °C, compared to the DW solution without any CNTs. A similar result was obtained when we replaced the DW solution with the DW/EG solution.

Figure 7(a) shows a schematic view of the liquid cooling system for the Intel Core i5 processor using CNT based

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**Figure 5.** The simulation and measured temperatures of the micro-processor as a function of operation time in the case of the AS5 thermal grease (a) and CNTs/AS5 thermal grease (b) [10]. With permission from Elsevier.
DW/EG solutions. The pump power consumption of the cooling system was 1.8 W. The dimensions and the power consumption of the two fans were $120 \times 120 \times 38 \text{ mm}^3$ and 3.6 W, respectively. The heat radiator was made of aluminum material, and the dimensions of the heat radiator were $150 \times 120 \times 25 \text{ mm}^3$. The environmental temperature was kept at $20^\circ C$ for all measurements by using an air conditioner.

Figure 7(b) shows the micro-processor measured temperature as a function of working time when using CNT based DW/EG solutions for thermal dissipation. At initial time, the temperature of the micro-processor was about $30^\circ C$. The saturation temperature of the micro-processor reached $57^\circ C$, $54^\circ C$ and $51^\circ C$ when using a DW/EG solution without CNTs, DW/EG solution with 0.5 g of CNTs/liter concentration, and an DW/EG solution with 1 g of CNTs/liter concentration after 350 s of working time, respectively. These results indicate that by mixing CNTs (1 g l$^{-1}$ concentration) in the DW/EG solution, we could decrease the saturation temperature of the CPU by 6$^\circ C$ compared to using DW/EG solutions without CNTs [15].

Other results were also obtained with the use of an appropriate CNT based liquid cooling configuration for heat dissipation of high power LED lights. Figure 8(a) shows the schematic view of the heat dissipation system for 450 W LED floodlights using CNT nanofluids. In figure 8(a), the aluminum heat sink was set to directly contact with nine LED chips; the tracks inside the aluminum heat sink was fabricated to allow fluid flows through it and to absorb heat from the
LED chips. The pump power consumption of the cooling system was 1.8 W. The dimensions and power consumption of the fan were $120\times 120\times 38$ mm$^3$ and 3.6 W, respectively. The heat radiator was made of aluminum material with $150\times 120\times 25$ mm$^3$ dimensions. The dimensions of aluminum heat sink and LED chip were $210\times 210\times 17$ mm$^3$ and $40\times 40\times 3$ mm$^3$, respectively. The power of the LED chip and power consumption of the LED floodlight were 50 W and 450 W, respectively. The environmental temperature was kept at 20 °C for all measurements. The temperature of the LED chip was directly measured by using an attached temperature sensor and WH7016E electronic digital temperature controller. Figure 8(b) shows the real images of the 450 W LED floodlight using CNT nano fluid for heat dissipation (b).

The experimental results of heat dissipation for the 450 W LED floodlight with different concentrations of CNTs in nano fluid are shown in figure 9(a). The temperature of the LED chip was 20 °C at initial time and then saturated after 40 min working time. When using DW/EG solution for heat dissipation, the saturation temperature of the LED chip was about 55 °C. The saturation temperature of the LED chip reached 53.7 °C, 52.5 °C, 51.9 °C and 50.6 °C when using nano fluids with 0.3 g l$^{-1}$, 0.5 g l$^{-1}$, 0.7 g l$^{-1}$, 1.0 g l$^{-1}$ and 1.2 g l$^{-1}$ of CNT concentration, respectively. These results indicate that by mixing CNT–OH with 1.2 g l$^{-1}$ concentration in the nano fluid, the saturated temperature of the LED chip decreased by 4.5 °C compared to the fluid without CNTs. According to the datasheet of the LED chip, if the operating temperature drops 10 °C, the lifetime of LED chips increase approximately two-fold. Thus, the lifetime of the LED can be estimated by the following expression

$$L = L_0 2^{\Delta t/10},$$  \hspace{1cm} (1)

where $L_0$, $L$, and $\Delta t$ indicate the basic-lifetime, the extra-lifetime, and the temperature reduction of the LED, respectively. Thus, the extra-lifetime percentage of the LED is determined by the expression

$$L(\%) = \frac{L - L_0}{L_0} \times 100\% = (2^{\Delta t/10} - 1) \times 100\%. \hspace{1cm} (2)$$

From expression (2), the extra-lifetime percentage of the LED was estimated as shown in figure 9(b). It clearly showed that the lifetime of the LED is further extended when the CNT concentration is increased. The extra-lifetime percentage of the LED reached to the value of 33% with 1.2 g l$^{-1}$ of CNTs concentration.

In addition to the 450 W LED floodlight, we also developed other configurations for a 100 W LED street light using CNT based liquids for heat management using the scheme shown in figure 10 (a). In this LED configuration, the aluminum heat sink was set in direct contact with the 100 W...
LED chip, and the tracks inside the aluminum substrate were fabricated to allow liquid to flow through the substrate and absorb heat from the LED chip. The pump power consumption of the cooling system was 1.8 W. The heat radiator was made of aluminum material, and the dimensions of the heat radiator were $100 \times 2000 \times 45$ mm$^3$. The temperature of the LED chip and radiator were directly measured by using an attached temperature sensor and an OMRON electronic digital temperature controller. The environmental temperature was 28 °C for all measurements. Figure 10(b) shows the real images of the 100 W LED street light using the CNT nano-fluid for heat dissipation.

Figure 11 shows the temperature of the 100 W LED street light as a function of operation time in the case of using
4. CNT based lubricating oils for engines

Among the above-mentioned results, the CNTs are considered as excellent candidates for improving the thermal properties of lubricants, which promises heat dissipation application for internal combustion engines. We have developed a fabrication process for the synthesis of lubricating oils containing CNTs, shown in figure 12(a). The CNT–OH was dispersed in polyanapholephin (PAO) and other standard additives with 0%–0.1% volume fraction of CNTs to obtain lubricating oils. The detailed process of the fabrication will be published elsewhere. Figure 12(b) is the real image of the synthesized lubricating oil containing CNTs.

The thermal conductivity of the CNT lubricating oils was estimated using an ECC thermal conductivity meter and compared with our computational model presented in reference [22]. In [22] we derived the expression for thermal conductivity enhancement in CNT suspensions, denoting the liquid molecule radii and CNT diameter to be \( r_l \) and \( r_{CNT} \), as well as the volume fraction of the nanoparticles as \( \varepsilon \) and the volume fraction of the liquid as \( 1 - \varepsilon \). The effective thermal conductivity of CNT, lubricating oils is expressed as

\[
\frac{k_{eff}}{k_l} = 1 + \frac{1}{3} \frac{k_{CNT} \varepsilon r_l}{k_l (1 - \varepsilon) r_{CNT}}.
\]

where subscripts 'l' and 'CNT' denote quantities corresponding to the base lubricating oils and CNTs, respectively. Equation (3) shows that the thermal conductivity of the lubricating oils strongly depends on diameter, volume fraction and the thermal conductivity of CNTs. In the calculation, the thermal conductivity of the nanotubes is taken as 1800 W m\(^{-1}\) K\(^{-1}\) and that of the base oils as 0.1448 W m\(^{-1}\) K\(^{-1}\); the effective radius of the lubricating oil molecule is 0.4 nm [22].

Figure 13(a) shows the dependence of the thermal conductivity of the CNT lubricating oils at different CNT concentrations. We chose the measurement at 25 °C and 85 °C because these are related to the temperature of the engine before and during working. It is seen that that thermal conductivity increases linearly with CNT content. The dashed lines in figure 13(a) show a simulated result that is very well suited with the measurement. The thermal conductivity of the lubricating oils with 0.1% CNT volume fraction is increased up to 12.5% compared to the original oil.

The fabricated lubricating oils with 0.1% CNT volume fraction were tested on different engines such as a T55 tank engine, a ZIL 131 engine, a URAL 375D engine and a small marine diesel engine. Figure 13(b) shows the measured data of the thermal conductivity of the CNT based lubricating oil
at 0, 0.05 and 0.1% wt. vol CNTs, measured at different working stretch road distances of the engine. The lubricating oil without CNT additive was measured up to 5000 km working of the engine. The lubricating oil with 0.05 and 0.1% wt. vol. CNTs can work up to 20 000 km without degradation of the engine performance. This indicates that the longevity of the CNT lubricating oil with 0.1% vol. CNTs can be used up to 20 000 km. The other experimental testing parameters also showed that the engine can save up to 15% fuel consumption by using 0.1% vol. CNTs in lubricating oils. The above results have confirmed the advantages of CNTs as an excellent additive component in lubricating oils for heat dissipation in internal combustion engines.

5. Conclusion

It is clearly seen that CNT based greases, liquids and lubricating oils have opened up many effective applications in thermal management for high power electronic devices such as CPUs, LEDs and other high power machines such as internal combustion engines. It is expected that the CNTs based greases, liquids and lubricating oils will be widely used in thermal management in the near future.

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References


Figure 13. The thermal conductivity of lubricating oils containing CNTs with different volume concentration measured at 25 °C and 85 °C (a) and the thermal conductivity of the CNT lubricating oils after working stretch road distances (b).