Proceeding of ICNM - 2009
1st International Conference on Nanostructured Materials and
Nanocomposites (6 – 8 April 2009, Kottayam, India)

Published by : Applied Science Innovations Private Limited, India.
http://www.applied-science-innovations.com

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Thermal Transport Properties of Carbon Nanotubes

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Abstract : Because of their unique structure, novel properties and potential applications, carbon nanotubes (CNTs) have attracted significant attention since their discovery. The thermal properties of CNTs are of interest for basic science as well as for technological applications. In the present paper, our experimental studies of the thermal transport properties of CNTs in recent years were introduced. The measurement techniques and the obtained results of the thermal transport properties of one individual CNTs, CNT arrays, CNT nanostructures and CNT composites were present. The mechanism of heat transport in CNTs has been further discussed.

Introduction

Carbon nanotubes (CNTs) have attracted considerable attention since their discovery due to the wide potential applications. Theoretical and experimental investigations have demonstrated that the thermal conductivity of this new class of one-dimensional materials could exceed that of diamond and graphite which have been well known in nature to have superior thermal performance [1-4]. Several groups have measured the thermal properties of millimeter-sized CNT mats and packed carbon nanofibers. The obtained data were scattering and highly depended on the CNT preparation methods and the sample microstructures [5-8]. In recent years, we carried out a series of studies on the thermal transport properties of one individual CNTs, CNT arrays, CNT nanostructures and CNT composites. Here we present the measurement techniques and the obtained results and further discuss the mechanism of heat transport in CNT materials.

Thermal conductivity of one individual CNTs

The thermal conductivities of three different diameters of CNTs at room temperature are shown in Fig. 1. The CNTs made by an arc-discharge evaporation method were chosen as the test samples. A sample-attached T-type nanosensor was applied to measure the CNT thermal conductivity [4]. d_o and d_i represent the outer and inner diameter of the CNT. It is clearly seen from Fig. 1 that the thermal conductivity of a CNT at room temperature increases as its diameter decreases. The present result for a CNT with a diameter of 9.8 nm is 2069 W/(m·K). The diameter-dependent thermal conductivity indicates that the interactions of phonons and electrons between multiwalled layers affect the thermal conductivity. The thermal conductivity increases as the number of multiwalled layers decreases. A single-walled carbon nanotube is expected to have much higher thermal conductivity. Fig. 2 represents the temperature dependence of the thermal conductivity for a CNT with a diameter of 16.1 nm. The measured thermal conductivity increases with an increase in temperature and appears to have an asymptote near 320 K. This tendency is the same as those obtained previously [3], which is attributed to the onset of umklapp phonon scattering.

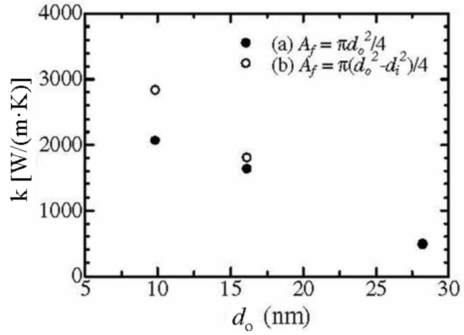

Fig.1 Diameter dependence of the thermal conductivity of a single CNT

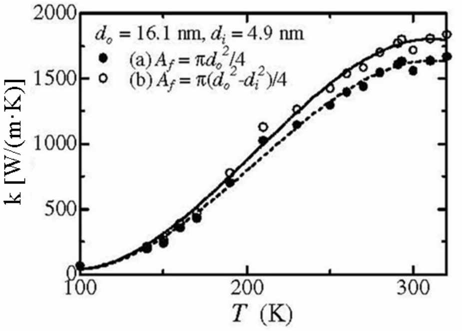

Fig.2 Temperature dependence of the thermal conductivity of a single CNT, d_o=16.1 nm

Thermal conductivity of CNT array

We measured the thermal diffusivity, α, of a CNT array in the temperature range of -55-200 °C by using a nanosecond laser flash (LF) technique. The sample was prepared using plasma enhanced hot filament chemical vapor deposition (PECVD) [9]. The CNT array is vertical to the glass substrate and its thickness is about 20 μm. The diameter of the CNTs is around 60-150 nm. Fig. 3 shows the dependence of α on the temperature. It is observed that α of the CNT array increases slightly with temperature in the -55~70 °C temperature range and exhibits no obvious change in the 75~200 °C temperature range. The tested CNT has very large room-temperature thermal diffusivity which is about 4.6 cm^2/s, indicating that CNTs possess excellent thermal transport properties. The mean thermal conductivity (k) of individual CNTs was estimated from the thermal diffusivity, specific heat (C_p), and density (ρ) by using the correlation of $k = \alpha \rho C_p$. Although α only varies moderately with the temperature, it is clearly seen from Fig. 4 that k of individual CNTs increases smoothly with the temperature increase, reaching about 750 W/(m·K) at room temperature. Fujii et al. reported a room-temperature thermal conductivity of about 500 W/(m·K) for one single CNT with a diameter of 28.2 nm[4]. It is worthy noting that the measurement in Ref. 4 only gives the lowest bound of intrinsic thermal conductivity of the tested CNT.

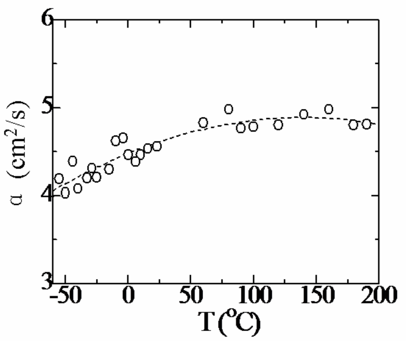

Fig. 3. Temperature dependent thermal diffusivity of the CNT array

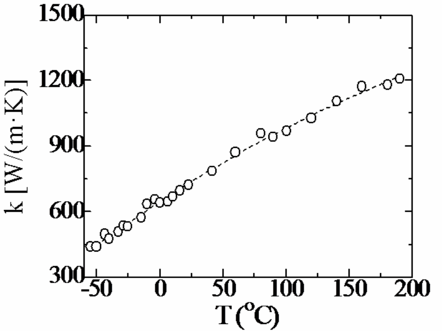

Fig. 4. Estimated mean thermal conductivity of individual CNTs on the temperature

Thermal conductivity of CNT nanostructures

A reversibly dispersible pellet consisting of self organized and highly aligned CNTs has been produced by heating and dehydrating an aqueous suspension containing hydrophilic CNTs [10]. The thermal diffusivity of the CNT pellet was measured by a LF technique. Fig. 5 presents the thermal diffusivity of the CNT pellet from room temperature to 700 °C. It is seen that the thermal diffusivity decreases with an increase in the temperature, from 3.63 mm^2/s at 27 °C to 2.59 mm^2/s at 700 °C. Fig. 6 represents the calculated thermal conductivity of this CNT pellet as a function of temperature. It is seen that the thermal conductivity increases with temperature and tends to reach a maximal value at about 800 °C. The measured thermal conductivity is low compared to the theoretical calculation and previous measurements. It is worth noting that in our measurement heat flows perpendicularly from the front surface to the rear surface of the test pellet. The thermal conductivity is along the direction vertical to CNT alignment direction. CNT walls have similar structures to graphene sheet and the thermal conductivity of CNTs shows greatly anisotropic behavior. Heat transports substantially quicker through axial direction than through radial direction. Furthermore, there exist numerous pores among different domains and surface-surface boundaries even in a well-aligned domain. These pores and boundaries increase the thermal resistance inside the CNT pellet. Therefore the obtained low thermal conductivity is reasonable.

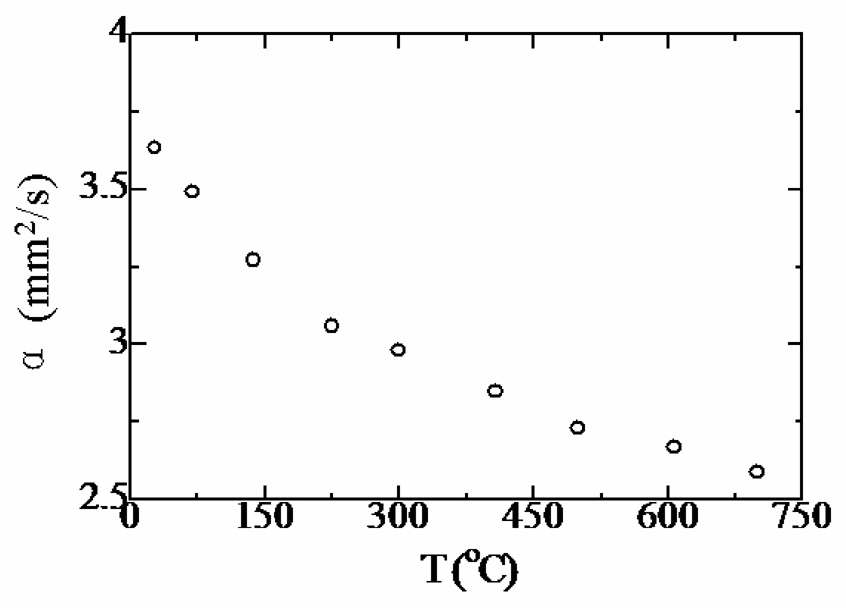

Figure (5) : Thermal diffusivity of the CNT pellet.

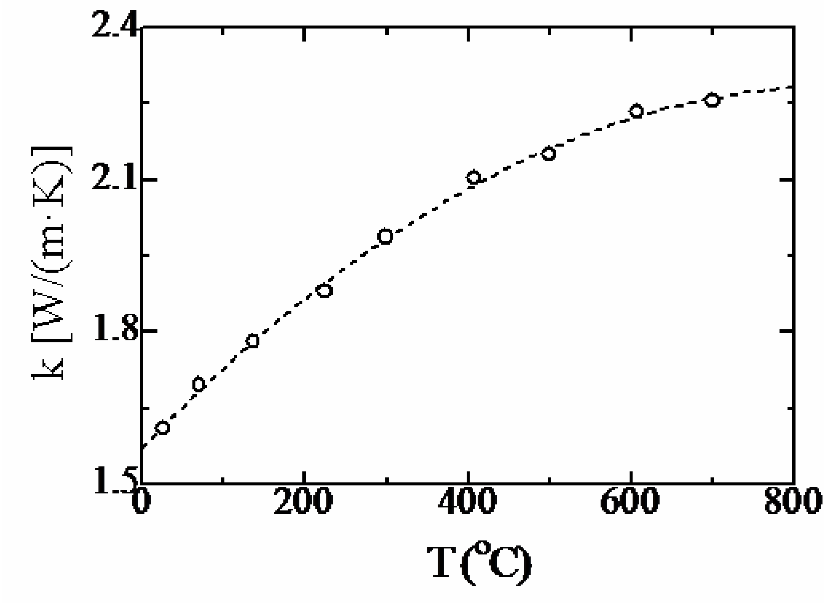

Figure (6) : Thermal conductivity of the CNT pellet.

Thermal conductivity of CNT nanocomposites

Carbon nanotubes (CNTs) are excellent candidates as dispersions for preparing thermal conductivity enhanced nanocomposites due to their very high thermal conductivity and very large aspect ratio. We prepared various CNT contained nanocomposites including nanofluids, phase change nanocomposties, thermal interface nanocomposites. Here we present an example on the thermal conductivity behaviors of ethylene glycol (EG) based nanofluids containing CNTs treated by the technique described in Ref. 11. Fig. 7 depicts the thermal conductivity enhancements of the CNT nanofluids as a function of the volume fraction of nanotubes. k and k_0 represent the thermal conductivities of the nanofluid and the base fluid, respectively. ϕ is the volume fraction of CNTs. Substantial increases in thermal conductivity are seen for all measured nanofluids, with thermal conductivity enhancement up to 17.5% observed for nanotube loading at 1.0 vol% in EG. The experimental data clearly indicate that the ratios of the enhancements increase monotonously with the volume fraction of CNTs. For all the measured volume fractions, the thermal conductivity enhancement ratios of distilled water (DW) based nanofluids are smaller than the corresponding values of EG based nanofluids. The volume fractions and the morphologies of CNTs play dominant roles on the thermal transport in the nanofluids. Fig. 8 depicts the thermal conductivity enhancement ratios of the nanofluids with different CNT loadings in EG as a function of ball milling times (*t*). Fig. 8 shows peak and valley values in the thermal conductivity enhancement with respect to the milling time for all the studied CNT loadings. For nanofluid at a volume fraction of 0.01, the thermal conductivity enhancements present a peak value of 27.5% and a valley value of 10.4% when the milling times are 10 h and 28 h respectively. The maximal enhancement is intriguingly more than two and half times as the minimal one. Interestingly, when further increased the milling time from 28 h to 38 h, $(k-k_0)/k_0$ increases from the valley value of 10.4% to 12.8%. Though the increment is not pronounced, it illustrates a difference tendency from that in the milling time range from 10 h to 28 h. The nanofluid with CNTs milled for 10 h has largest increment in thermal conductivity.

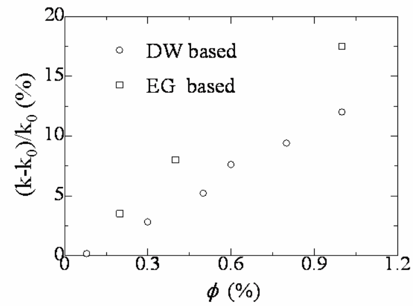

Fig. 7. Thermal conductivities of nanofluids as a function of nanotube loadings

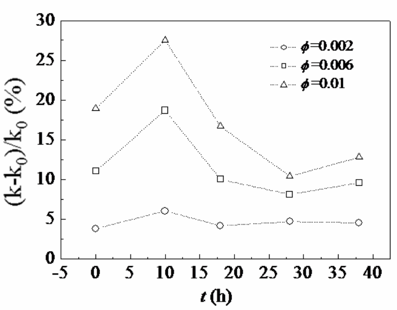

Fig. 8. Dependence of the thermal conductivity enhancement on the ball milling time

Acknowledgement

The following financial supports are greatly appreciated: National Science Foundation of China (50876058), Shanghai Educational Development Foundation and Shanghai Municipal Education Commission (07SG56), Innovation Program of Shanghai Municipal Education Commission (08ZZ99), and the Program for Professor of Special Appointment (Eastern Scholar) at Shanghai Institutions of Higher Learning.

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