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Thermal conductivity of MWCNT/epoxy composites: The effects of length, alignment and functionalization

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ABSTRACT

Carbon nanotubes (CNTs) show great promise to improve composite electrical and thermal conductivity due to their exceptional high intrinsic conductance performance. In this research, long multi-walled carbon nanotubes (long-MWCNTs) and its thin sheet of entangled nanotubes were used to make composites to achieve higher electrical and thermal conductivity. Compared to short-MWCNT sheet/epoxy composites, at room temperature, long-MWCNT samples showed improved thermal conductivity up to 55 W/mK. The temperature dependence of thermal conductivity was in agreement with $\kappa \propto T^n$ ($n = 1.9$–2.3) below 150 K and saturated around room temperature due to Umklapp scattering. Samples with the improved CNT degree of alignment by mechanically stretching can enhance the room temperature thermal conductivity to over 100 W/mK. However, functionalization of CNTs to improve the interfacial bonding resulted in damaging the CNT walls and decreasing the electrical and thermal conductivity of the composites.

1. Introduction

Carbon nanotubes (CNTs) have high electrical and thermal conductivity and excellent mechanical properties [1]. The theoretical thermal conductivity of single-walled carbon nanotube (SWCNT) is predicted to reach over 6600 W/mK [2]. Experimental measurements on individual multi-walled carbon nanotube (MWCNT) and SWCNT exhibit high thermal conductivity over 3000 W/mK [3] and 3500 W/mK [4], respectively. However, the entangled network of carbon nanotube sheets only shows thermal conductivity of 20–30 W/mK at room temperature measured with the comparative methods [5,6] and close to 80 W/mK with the bolometric technique [7]. CNT alignment is an effective approach for improving their mechanical properties and their thermal and electrical conductivity [5,6,8–10] along the alignment direction. External magnetic or electric fields [6,9] and mechanical stretching [10] have been used to improve CNT alignment in CNT sheet materials.

For nanotube–polymer composites, dispersing either SWCNT or MWCNT slightly increases the thermal conductivity [11–13] compared to pristine polymers. Higher loading of carbon nanotubes in composites results in higher thermal conductivity, but its thermal conductivity value is far below its theoretical predictions based on the simple rule of mixture due to their large interface scattering and contact resistance. Alignment of CNTs in the composite also increases the thermal conductivity [5,8,14,15] along the alignment direction due to reduced number of interface and contact resistance. Similarly, graphene also shows a similar trend and thermal

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conductivity of single graphene sheets is estimated to be as high as 5300 W/mK [16]; however, graphene/epoxy composites showed reduced thermal conductivity to around 80 W/mK [17]. For the thermal transport in the polymer, thermal interface resistance (R_K) between nanotubes and the polymer matrix plays an important role [18–20] and the thermal conductivity improvement is limited by the large interface resistance.

For the electrical transport in the CNT networks and their composites, the intertube contact is the main reason for the reduction of electrical conductivity [21]. Minimizing the number of intertube contacts is important for achieving higher electrical conductivity. This also can be applied to the thermal transport. Therefore, to achieve higher electrical and thermal conductivities in the carbon nanotube networks and their composites, we need to focus on: (1) reducing the number of intertube contacts by using longer CNTs, (2) improving the degree of alignment, and (3) ensuring a good electrical/thermal interface in CNT–CNT and CNT–epoxy.

For this purpose, we used long-MWCNT and its networks to see the effects of CNT length and mechanically stretched these long-MWCNT sheets to achieve a higher degree of alignment to reduce the number of intertube contacts in the resultant composites. Long-MWCNT composite already showed great performance improvement [10] compared to previous CNT composites [22]. Although the mechanical properties of functionalized CNT composite were noticeably improved due to higher interfacial bonding between the nanotubes and epoxy matrix, the electrical conductivity of CNT sheet composite was decreased due to the wall damage [23]. Most of thermal conductivity measurements for CNT composite were performed at room temperature and temperature dependent measurements are limited to CNT [3,24,25] or SWCNT network [5]. This research was to further study the effects of CNT length, alignment and functionalization on both electrical and thermal conductivity and their temperature dependence.

2. Experimental

Two types of long-MWCNTs were used. CVD grown forest (long-MWCNT I) was purchased from Cnano (CA, USA) and long-MWCNT sheet (long-MWCNT II) made by floating catalyst technique was purchased from Nanocomp (NH, USA). For comparison purpose, short-MWCNTs from Cnano (Flotube 7000) were also used. EPON862 with epicure-W (both from Miller-Stephenson Chemical Company) was used for nanotube/epoxy composites. For the low concentration nanotube composite fabrication, the nanotubes were mixed with epoxy based on the target weight percentage, sonicated in a cooling bath and cured in a hot press for 2 h at 177 °C [5,9,22]. The resultant composites had a CNT content ranging from 2 wt.% to 10 wt.%. To increase the nanotube concentration, CNT sheets were used with diluted epoxy and cured in a hot press as described elsewhere [22]. The typical weight percent of nanotubes in the CNT sheet/epoxy composite was about 60 ± 5 wt.%.

A mechanical stretching approach was also used to improve the degree of nanotube alignment in the CNT sheets. Stripe of MWCNT sheet (Nanocomp) was clamped between tensile stage of Shimadzu (AGS-J, Shimadzu Scientific Inc., Japan) and gauge length was stretched with the 0.5 mm/min. Alignment degree was checked by polarized Raman (Invia microscope, Renishaw) using 785 nm excitation. Linear

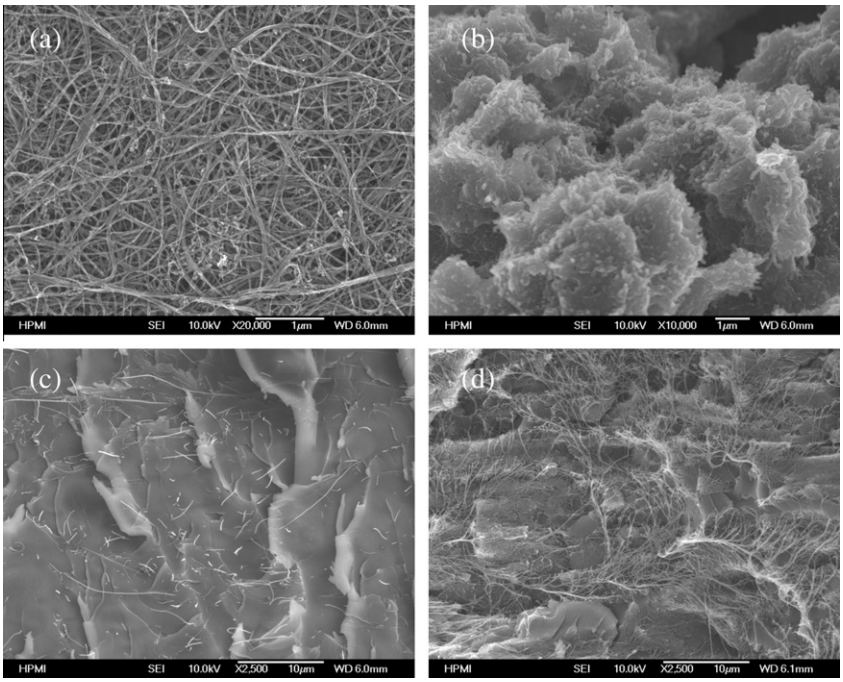

Fig. 1 – SEM images of (a) long-MWCNT sheet and fracture surfaces of (b) 10 wt.% short-MWCNT/epoxy composite, (c) 2 wt.% long-MWCNT I/epoxy composite and (d) 6.38 wt.% long-MWCNT II/epoxy composite; even at low magnification, both long-MWCNTs clearly showing over 10 μm length.

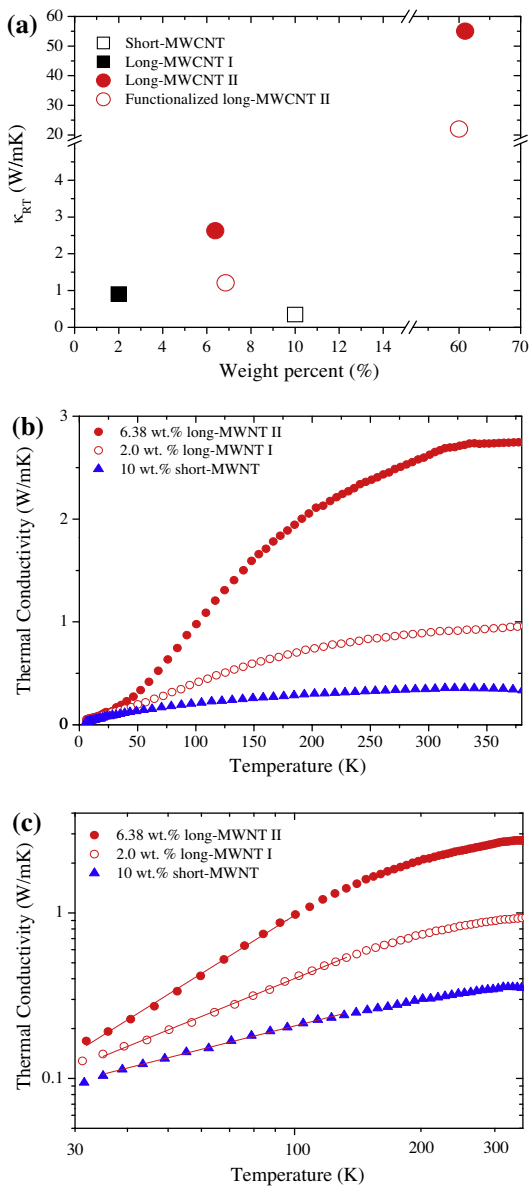

Fig. 2 – (a) Room temperature thermal conductivity (κ_{RT}) of different MWCNT and epoxy composite as a function of weight percentage. (b) Temperature dependence of thermal conductivity of composite with different nanotube length. In the case of short-MWCNT composite, the improvement is minimal compared to room temperature thermal conductivity of epoxy (0.2 W/mK). However, the improvement is significant in long-MWCNT composites. (c) log–log plot of thermal conductivity as a function of temperature. Long-MWCNT composite shows exponent close to 2.

polarizer was inserted to the detector side for VV configuration and sample was rotated to the laser polarized direction.

The in-plane electrical and thermal conductivities were measured using the physical property measurement system (PPMS, Quantum Design) with thermal transport option. A resistive heater and a temperature sensor were attached to one end of a sample through metal lead using thermally conductive silver epoxy, while the other end was attached to a cold foot and a second temperature sensor. At a given power setting of the heater, the temperature difference between the two sensors was used to calculate the in-plane thermal conductance and thermal conductivity was calculated by the dimension. High vacuum and radiation shields were used to minimize the heat loss from the heater [26]. Electrical conductivity was measured afterward with same two-probe contacts at the same temperature.

Nanotube/epoxy composites of a low concentration were cut into 7 mm × 7 mm squares with typical thickness ~1 mm. The thermal conductivity was measured using a two probe contact. CNT sheet/epoxy composites were measured in a similar manner, and the probe distance was less than 5 mm and the thickness was less than 100 μm.

Functionalization was performed by attaching an epoxide group as described in a previous paper [23].

3. Results and discussion

Fig. 1(a) shows SEM image of long-MWCNT sheet that was used in CNT sheet/epoxy composites. Fig. 1(b) shows the fracture surface images of 10 wt.% short-MWCNT/epoxy composite and (c) and (d) show 2.0 wt.% of long-MWCNT I/epoxy composite based on Cnano and 6.38 wt.% of long-MWCNT II (re-dispersion from Nanocomp CNT sheet), respectively. The length of long-MWCNTs from Cnano was longer than 10 μm, while the short-MWCNT was less than 1 μm. Nanocomp long-MWCNTs' length was more than 100 μm [27]. As seen in the SEM images, all the MWCNTs are high purity with minimal amount of amorphous carbon.

Fig. 2(a) shows the summary of room temperature thermal conductivity for short- and long-MWCNT/epoxy composites as a function of CNT concentration. Higher CNT weight percentage loading in long CNT/epoxy composites demonstrated higher room temperature thermal conductivity. By further increasing the weight percentage up to 60 wt.% using random long-MWCNT sheets, higher thermal conductivity up to 55 W/mK was achieved, which is nearly three times higher than our previously reported value of short SWCNT buckypaper/epoxy composites [5].

The long-MWCNT composites exhibited higher thermal conductivity than the short-MWCNT composites when the

Table 1 – Summary of CNT type and weight percentage of low concentration MWCNT composite.

	Manufacturer	Typical length	Typical diameter (nm)	Concentration (wt.%)
Short-MWCNT	Cnano	1 μm range	10–20	10
Long-MWCNT I	Cnano	5 mm from CVD	50–100	2.0
Long-MWCNT II[a]	Nanocomp	>100 μm range	<10	6.38

[a] Re-dispersion from sheet.

Table 2 – Room temperature two-probe electrical and thermal conductivity of CNT sheet/epoxy composites at its highest value.

CNT sheet for epoxy composite	Electrical conductivity (S/cm)	Thermal conductivity (W/mK)
Random short-MWCNT	100	6
Random long-MWCNT	640	55
25% stretched long-MWCNT[a]	1300	83
40% stretched long-MWCNT[a]	800	103
Functionalized long-MWCNT	160	22

[a] Along stretched direction.

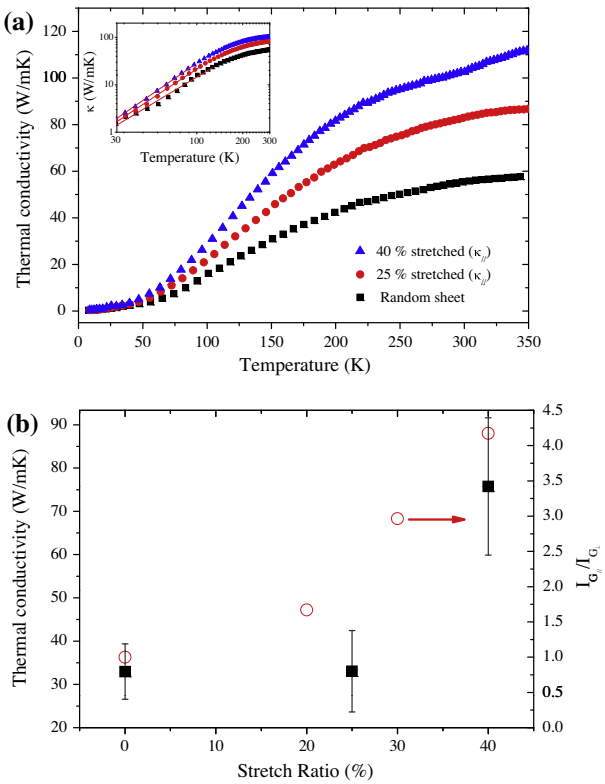

Fig. 3 – (a) Temperature dependence of thermal conductivity of random, 25% and 40% stretched long-MWCNT sheet/epoxy composite with the highest thermal conductivity. Inset is log–log plot of the thermal conductivity and the exponent is almost same. (b) Comparison of averaged room temperature thermal conductivity and Raman intensity ratio at parallel and perpendicular direction as a function of stretch ratio. Higher stretch ratio gives higher alignment degree and this leads larger intensity ratio and thermal conductivity.

weight percentages were the same. At room temperature, 10 wt.% short-MWCNT/epoxy composite showed thermal conductivity of 0.35 W/mK, which was nearly twice of the thermal conductivity of epoxy, 0.2 W/mK. However, the long-MWCNT composites showed 0.9 W/mK (Cnano) and 2.6 W/mK (Nanocomp) even at lower concentrations of 2.0 wt.% and 6.38 wt.%, respectively. The results were in agreement of the effects of length on the thermal conductivity in CNT composites [15] and summarized in Table 1.

Fig. 2(b) shows a temperature dependence of thermal conductivities of low-weight percentage nanotube/epoxy composites. They show a typical parabolic increase at the low temperature range, linear dependence at medium temperature range and saturated at room temperature. A larger radiation contribution showed an upturn of thermal conductivity at room temperature and was excluded from the data [25]. Because of the relatively large diameter of the MWCNTs (over 10 nm), their thermal conductivity was similar to a two dimensional system rather than one dimensional SWCNT and more acoustic phonon contribution dominated the thermal transport [28]. Therefore, $k(T)$ show T^2 dependence and similarly graphite also has $T^{2.3}$ dependence due to quadratic dispersion of out-of-plane phonon mode [28]. The exponents from the log–log plot (Fig. 2(c)) of thermal conductivity between 30 K and 130 K show 1.6, 1.0, and 0.6 from higher thermal conductivity sample to the lower one. The long-MWCNT and higher concentration sample (6.38 wt.%) had an exponent of 1.6, which were closer to the two dimensional system because more phonons were contributed from the nanotubes. These results show similar trends as carbon black filled epoxy composites since interface scattering and resistance are major dominating factors for thermal transport [29]. At a higher temperature range over 130 K, thermal conductivity of the CNT composites showed either linear or sublinear temperature dependence as previously observed [14], which comes from the start of Umklapp process from phonon–phonon scattering.

Even though long-MWCNT I and II are coming from different manufacturer and have different diameter, 50–100 nm for long-MWCNT I and 5–10 nm for long-MWCNT II, long-MWCNT composites show higher thermal conductivity compared to short-MWCNT composite.

To increase the nanotube concentration over 60 wt.% in a composite, CNT sheets were used as shown in Fig. 1(a). Using the random long-MWCNT sheets, the composite's highest room temperature thermal conductivity was over 55 W/mK as summarized in Fig. 2(a) and Table 2. To improve the in-plane thermal conductivity, CNT sheets were stretched mechanically and aligned. Fig. 3(a) shows the temperature dependence of the thermal conductivity of random, 25% and 40% stretch-induced aligned long-MWCNT sheet/epoxy composite samples with the highest room temperature thermal conductivity. The 40% stretched samples significantly improved the CNT alignment in the resultant sheets and their composites [10,23]. At room temperature, their thermal conductivity increased up to 83 W/mK (25% stretched) and 103 W/mK (40% stretched) along the alignment direction,

which was double when compared the random sample. Inset of Fig. 3(a) is the log–log plot of temperature dependence and they show similar quadratic temperature dependence ($\kappa \propto T^n$, $n = 1.9$–2.2).

Fig. 3(b) shows the averaged room temperature thermal conductivity over three different batches and typical Raman intensity ratio of G-band peak for parallel and perpendicular direction from the polarized Raman measurement as a function of stretch ratio. Typical thickness of three layer CNT sheet/epoxy composite is 75 μm, but thinnest composite we can achieve is down to 40 μm. High electrical and thermal conductivity of 25% stretched CNT sheet composite in Table 2 is due to its smaller thickness. However, averaged thermal conductivity remains to 33 W/mK until 25% stretch, and doubled to 75 W/mK at 40% stretched samples irrespective of sample batch.

High degree of CNT alignment showed much higher thermal conductance and conductivity along the aligned direction. The probe distance between hot and cold was less than 5 mm, which will reduce the radiation effects [24]. The saturation of thermal conductivity at higher temperatures confirmed the reduced radiation from the surface and onset of the Umklapp process. We previously reported the improved electrical and mechanical properties of long-MWCNT sheet/epoxy composites with the stretch-induced alignment [10]. However, thermal conductivity improvement at 40% stretched CNT sheet composite was limited by a factor of two, which was not as good as the electrical or mechanical improvements which showed an enhancement of greater than three times [10].

Thermal conductivity anisotropy of the resultant composites was also studied. Fig. 4 shows temperature dependence of thermal conductivity and electrical resistivity in both parallel and perpendicular direction. Thermal conductivity values along the perpendicular direction was lower than that of the parallel direction and the anisotropy ratio ($\kappa_{\parallel}/\kappa_{\perp}$) was 2.2 and 4.9 at room temperature in 25% and 40% stretched CNT sheet/epoxy composite, respectively. And this value is similar to the G-band Raman intensity ratio as well. Insets of Fig. 4(a) and (b) show the temperature dependence of anisotropy, which slightly increased over the temperature range because of more radiation contribution close to room temperature. However, the anisotropy of CNT sheet/epoxy composite thermal conductivity was much lower than that of a thin nanotube sheet [24] or magnetic field aligned CNT films [6]. This may come from two aspects: (1) more intertube contacts existed between aligned nanotubes in the CNT sheet composite because of pressure during fabrication procedure and dense packing of nanotubes and (2) the role of epoxy resin between the nanotubes as a thermal transfer medium compared to the empty space in thin sheet. For the thermal conductivity of CNT composites, the thermal interface between nanotubes and medium is important [17,30] and smaller thermal interfacial resistance (R_K) is crucial for higher thermal conductivity.

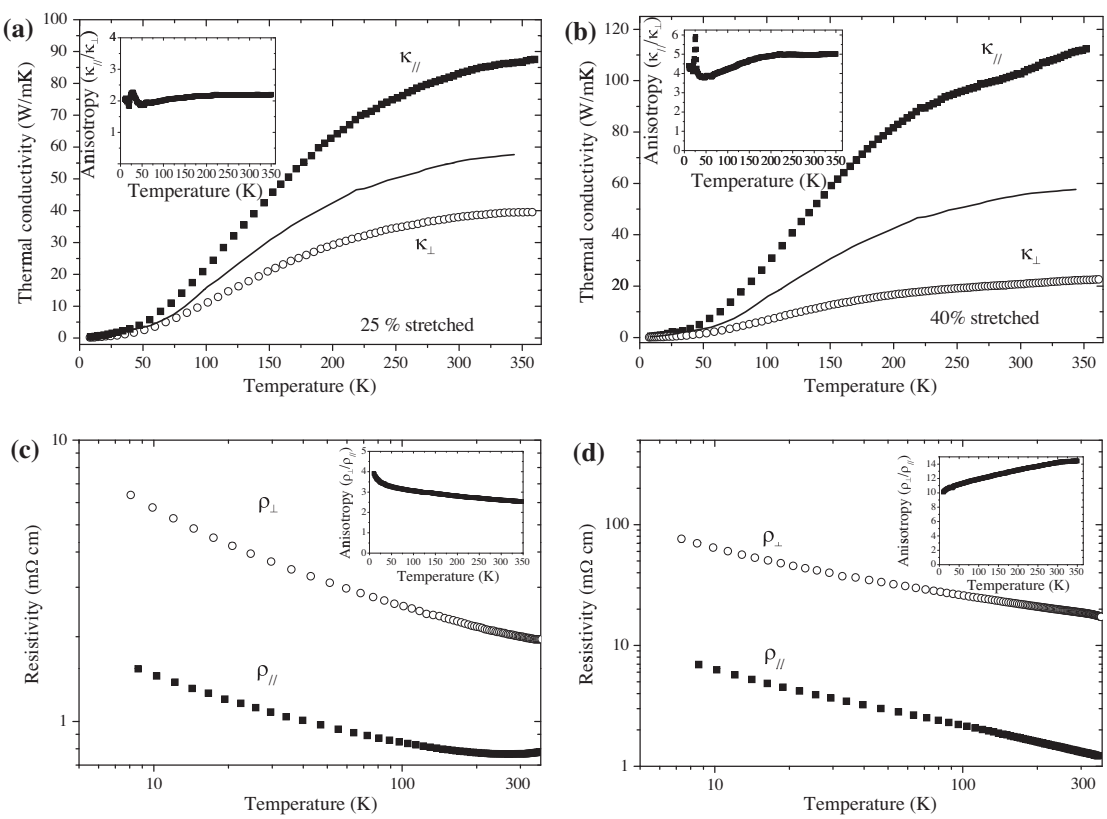

Fig. 4 – (a,b) Temperature dependence of thermal conductivity of 25% and 40% stretched long-MWCNT sheet composite in parallel and perpendicular direction. Line is the result of random sheet composite for comparison. Inset shows anisotropy of thermal conductivity. (c,d) Two-probe electrical resistivity of 25% and 40% stretched long-MWCNT sheet/epoxy composite as a function of temperature measured simultaneously. Inset shows anisotropy of electrical conductivity.

In the case of CNT sheet composites, the higher concentration of CNT introduces additional intertube contacts, so we should consider CNT–CNT thermal contact resistance as well as CNT–matrix interface. Simulation results [31] show that larger R_K and intertube contacts further reduce the anisotropy, which agreed with the observations in this study.

The temperature dependence of CNT sheet composites also followed similar trends as low CNT concentration composites described in the previous section. The thermal conductivity (κ) showed nearly a quadratic temperature dependence ($\kappa \propto T^n$, $n = 2.0$–2.3) at temperatures below 150 K, had a linear temperature dependence after that, and saturated at the high temperature ranges in both parallel and perpendicular direction of alignment. This exponent was from the two-dimensional nature of phonon distribution and the value is very close to the two dimensional system such as graphite.

All of the previous measurements on nanotube films [28,31] showed the exponent close to two. Single layer graphene on the substrate also has 2D properties and its exponent in the temperature dependence was also close to two [32]. Hence, even with over 60 wt.% CNT concentration, the resultant composites demonstrated typical in-plane thermal transport behaviors.

The Lorenz ratio, $\kappa/\sigma T$, is in the order of 2–5×10^{-6} WΩ/K^2 in the case of random CNT sheet/epoxy composites and 4–7×10^{-6} WΩ/K^2 in aligned CNT sheet/epoxy composites at various temperatures. These values were two orders higher than expected for electron transport and close to the previous measurements on carbon nanotube films [6,24,28]. Therefore, phonon contribution was dominant at all temperatures and was clearer in the case of the aligned CNT sheet composites.

Along with thermal conductivity, 2-probe electrical resistivity of the 25% and 40% stretched samples were measured simultaneously and shown in Fig. 4(c) and (d). As expected, the resistivity was lower along the aligned direction. The anisotropy ratios from electrical resistivity were 2.6 and 14.2 at room temperature. Electrical conductivity had a larger anisotropy than thermal conductivity, especially for higher aligned samples. Epoxy was slightly thermally conductive but electrically insulating, which also contributed to the higher electrical anisotropy. Table 2 summarizes all of the room temperature electrical and thermal conductivity values.

CNT functionalization is generally adopted to improve the mechanical properties of nanotube/epoxy composites [23,33]. Epoxidation of CNT improved the mechanical properties of composite, but peroxide acid treatment damaged the side walls and decreased the electrical conductivity [23,34]. As summarized in Fig. 2(a), the functionalization reduced the room temperature thermal conductivity because of (1) the defects generated on the nanotube wall during the oxidation procedure and (2) the increased interfacial thermal resistance (R_K). The thermal conductivity of functionalized long-MWCNT/epoxy composite was reduced compared to pristine

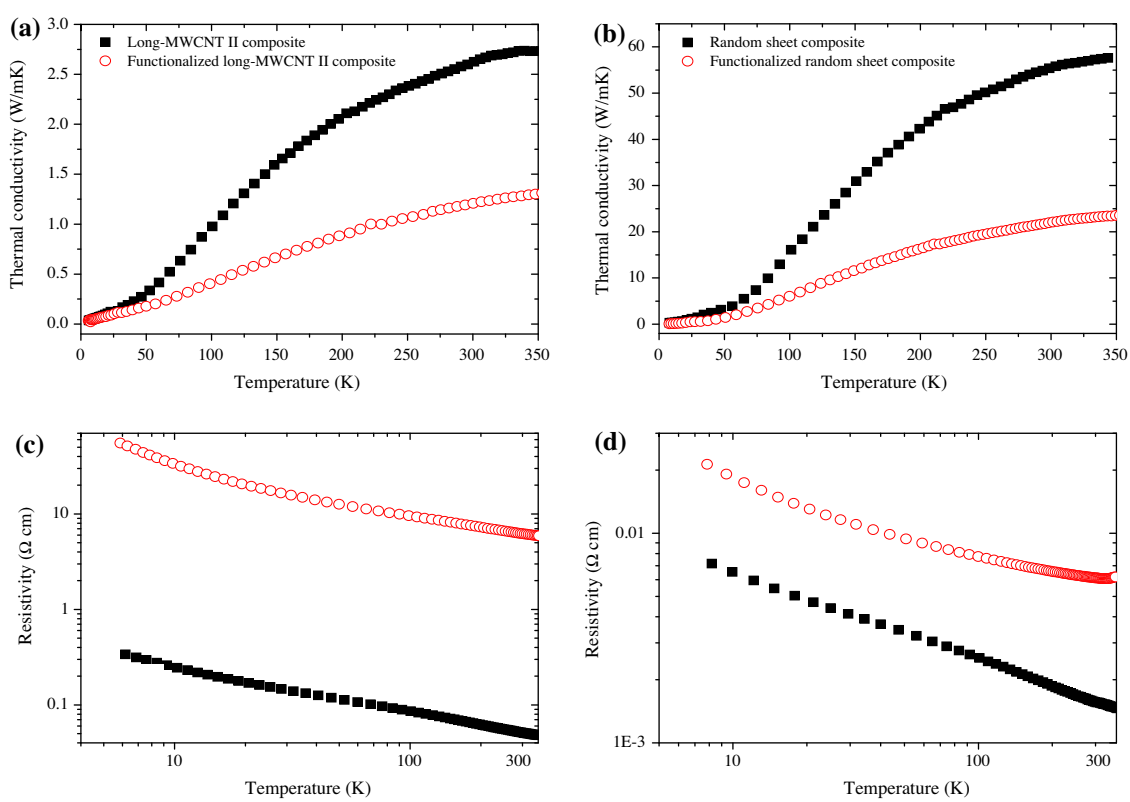

Fig. 5 – Temperature dependence of (a,b) thermal conductivity and (c,d) resistivity of pristine and functionalized long-MWCNT BP/epoxy composite based in low concentration and CNT sheet. Even though functionalization increased the interfacial bonding between SWCNT and epoxy, it decreased the electrical an termal conductivity because of the damage of SWCNT wall during the oxidation procedure.

long-MWCNT/epoxy composites. This trend was the same with higher concentration of CNT composites based on the sheets.

Fig. 5(a) shows the comparison of temperature dependence of thermal conductivity in pristine (6.38 wt.%) and functionalized long-MWCNT (6.85 wt.%) composites at low concentration. Even though at higher weight percentage of functionalized MWCNT composite, its room temperature thermal conductivity decreased from 2.6 W/mK to 1.2 W/mK by 54%. A high concentration composite using CNT sheet in Fig. 5(b) also shows similar trend and room temperature thermal conductivity decreased by 60% from 55 W/mK to 22 W/mK. However, the slope of temperature dependence was basically the same with an exponent of 2, which means that the same phonon transport mechanism exist between them.

The electrical conductivity also decreased after functionalization, as shown in Fig. 5(c) and (d). In the case of low concentration nanotube composite, room temperature electrical conductivity decreased by two orders of magnitude, however, that of high concentration CNT sheet composite decreased by 75%. From these results, we conclude that low concentration CNT composite is more sensitive to the electrical transport because it is more close to the percolation threshold. In the low dimensional structures, its electrical transport was more sensitive to the defects [35]. Therefore, thermal conductivity reduction after functionalization is less than the reduction of electrical conductivity.

4. Conclusion

The electrical and thermal conductivity of various MWCNT/epoxy composites at different concentrations were measured. The long-MWCNT composites showed a higher electrical and thermal conductivity than short-MWCNT composite samples. A higher concentration (60 wt.%) of long-MWCNT sheet/epoxy composites showed the highest thermal conductivity of 55 W/mK at room temperature, while the mechanically stretch-induced alignment further improved the thermal conductivity up to 103 W/mK for 40% stretched CNT sheet composites. Averaged thermal conductivity of 40% stretched CNT sheet composite gave twice that of the random composite. High concentration and alignment of long-MWCNT resulted in less intertube contact barriers and higher electrical and thermal conductivity in the resultant composites. The temperature dependence of high concentration CNT composite also confirms that the two dimensional phonon distributions for thermal transport were similar to the mechanism of graphite materials.

Functionalization was also attempted to further improve the CNT composite mechanical properties, but it noticeably decreased the thermal conductivity and even more the electrical conductivity because of the damage to the CNT structures. This also confirms the low dimensionality of CNTs and phonon mediated thermal transport mechanism in CNT.

Acknowledgments

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