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EFFECTS OF DIFFERENT FACTORS ON THE HEAT CONDUCTION PROPERTIES OF CARBON NANOTUBES

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Abstract. The effects of structure, purity, and alignment on the thermal conductivity of carbon nanotube-based films and fibers were studied experimentally to understand the heat transport phenomena occurring in the nanostructured materials. The thermal conductivity of the macroscopic films and fibers was determined by employing the parallel thermal conductance technique. The effects of different factors on the heat conduction properties were investigated to evaluate the roles of bulk density and cross-sectional area in the thermal conductivity of the nanostructured materials. The results indicated that macroscopic films and fibers produced from carbon nanotubes can conduct heat very efficiently, depending on a variety of factors. The structure, purity, and alignment play fundamentally important roles in determining the heat conduction properties of carbon nanotube-based films and fibers. Macroscopic films and fibers produced from single-walled carbon nanotubes typically possess high heat conduction properties. The non-carbonaceous impurities negatively affect the heat conduction properties because of the low degree of bundle contact. Carbon nanotube-based films and fibers give rise to a power-law dependence of thermal conductivity with respect to temperature. The specific thermal conductivity decreases with increasing bulk density. Low bulk density can compensate for the adverse effect of poor alignment on specific thermal conductivity. A maximum specific thermal conductivity is achieved at room temperature, but Umklapp scattering occurs. The specific thermal conductivity of carbon nanotube-based fibers is significantly higher than that of carbon nanotube-based films because of the increased degree of bundle alignment.

Keywords: Carbon nanotubes; Material properties; Thermal conductivity; Thermal physics; Thermal properties; Phonon scattering

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1. Introduction

Carbon nanotubes, also called buckytubes, are nanoscale hollow tubes composed of carbon atoms. The cylindrical carbon molecules feature high aspect ratios, typically above 10^3, with lengths up to millimeters and diameters from about one nanometer up to tens of nanometers [1,2]. This unique one-dimensional structure and concomitant properties endow carbon nanotubes with special natures, rendering them with unlimited potential in nanotechnology-associated applications [1,2]. Carbon nanotubes are members of the fullerene family. Although the first fullerene molecules were discovered in 1985 [3], it was not until Sumio Iijima reported his findings in 1991 about needlelike carbon tubes in *Nature* that carbon nanotubes came to public awareness [4]. Since then, carbon nanotubes with various structures have been discovered [5,6]. According to the number of graphic shells, they are mainly categorized as single-walled and multi-walled carbon nanotubes. A single-walled carbon nanotube can be described as a long tube formed by wrapping a single graphene sheet into a cylinder with a diameter of about one nanometer, the ends of which are capped by fullerene cages. Multi-walled carbon nanotubes are concentrically aligned single-walled carbon nanotube assemblies with different diameters. The distance between adjacent shells is about 0.34 nanometer. Single-walled carbon nanotubes differ from multi-walled carbon nanotubes not only in their dimensions, but also in their corresponding properties.

Novel mechanical, electrical, and chemical properties absent in other materials have been discovered in carbon nanotubes [7,8]. As for thermal properties, carbon nanotubes outperform diamond as the best thermal conductor [9,10]. Carbon nanotubes can exhibit unique ability to conduct heat [9,10], referred to as heat conduction properties. Specifically, carbon nanotubes are a highly effective thermal conductor in the longitudinal direction. However, a thermal barrier is formed in the radial direction. At room temperature, the thermal conductivity of an individual single-walled carbon nanotube in the radial direction is about 1.52 W/(m·K) at room temperature [11]. In contrast, carbon nanotubes show superior heat conduction properties along the longitude directions, with the highest known elastic modulus and tensile strength among known materials. At room temperature, the thermal conductivity in the longitudinal direction is about 3500 W/(m·K) [12]. When macroscopic, ordered assemblies of single-walled carbon nanotubes are formed, the thermal conductivity of carbon nanotube-based films and fibers could reach up to around 1500 W/(m·K) at room temperature [13]. The heat conduction properties of carbon nanotube networks vary significantly, with a minimum of thermal conductivity less than 0.1 W/(m·K) [14]. The heat conduction properties depend on a variety of factors, for example, misalignment and impurities. Single-walled carbon nanotube are stable up to around 1000 K in air and around 3000 K in vacuum [15]. While various techniques have been developed to produce carbon nanotubes in sizable quantity, high yield, and purity, the study of heat transport phenomena involved in carbon nanotubes is still an active area of interest [16] because of the potential for applications in thermal management.

The thermal conductivity of carbon nanotubes depends heavily upon the crystallographic defects of cylindrical sidewalls. Phonons can scatter because of the crystallographic defects of cylindrical sidewalls. This will lead to the increased relaxation rate, thereby decreasing the thermal conductivity of carbon nanotubes associated with the reduced mean free path of phonons [17,18]. In single-walled carbon nanotubes, the mean free path of phonons varies from 50 nm to 1500 nm [19,20]. The crystallographic defects of cylindrical sidewalls will lead to a significant reduction in mean free path [21], for example, 4 nm or less [22]. The thermal conductivity of carbon nanotubes depends heavily upon the structure of the carbon nanotubes. The thermal conductance of multi-walled carbon nanotubes is significantly higher than the sum of that of each individual shell because of the inter-wall interactions [23]. The thermal conductivity of a multi-walled carbon nanotube is lower than that of a single-walled carbon nanotube with an identical diameter configuration [24] because of an increase in cross-sectional area. Applications of carbon nanotubes are aimed to make use of their unique heat conduction properties to solve problems at the nanoscale.

Bulk carbon nanotubes can be used as composite fibers in polymers, but the bulk structure will reduce the ability to conduct heat, causing a decrease in thermal conductivity. The thermal conductivity of carbon nanotube-based fibers is comparable to that of common metals [25]. Through chemical modification, however, the ability to conduct heat is comparable to or higher than that of highly conductive metals [26], for example, copper. The thermal conductivity varies significantly depending upon the density and cross-sectional area of the bulk material. The bulk carbon nanotube material contains pores [27,28]. Consequently, the thermal conductivity of the non-compact bulk material is much lower than that of the skeletal material, since the bulk volume is inclusive of the void fraction. The skeletal portion of the bulk material is often referred to as the "matrix" or "frame". However, there is considerable uncertainty in determination of the density of the bulk material. Consequently, the effects of structure, purity, and alignment on the thermal conductivity of carbon nanotube-based films and fibers are still poorly understood. Little research has been conducted to determine which parameters are important for high thermal conductivity.

This study relates to the heat conduction properties of carbon nanotube-based films and fibers. Carbon nanotube-based films and fibers were produced, and the parallel thermal conductance technique was employed to determine the thermal conductivity of carbon nanotubes. The effects of structure, purity, and alignment on the thermal conductivity of carbon nanotube-based films and fibers were investigated to understand the characteristics of thermal transport in the nanostructured materials. The effects of different factors on the heat conduction properties were investigated to evaluate the roles of bulk density and cross-sectional area in the thermal conductivity of the nanostructured materials. The objective is to gain insight into the fundamental characteristics of thermal transport in carbon nanotubes. Particular emphasis is placed on the dependence of thermal conductivity on carbon nanotube structure, purity, and alignment, with an attempt to improve the heat conduction properties for carbon nanotube-based films and fibers.

2. Experimental methods

Preparation of carbon nanotube-based films and fibers. Carbon nanotube-based films were produced by spinning continuously onto a single rotational winder of a spinning machine. The preparation process of carbon nanotube-based films is depicted schematically in Figure 1. A dense film was laid down with aligned carbon nanotube bundles. The dense film was sprayed with acetone to further condense the carbon nanotube networks. By preparing a dense film in such a manner, the carbon nanotubes were highly aligned [29], although the surface tension effect is significant in the extrusion process [30]. A laser was used to cut the carbon nanotube-based film into small pieces for the measurement of thermal conductivity. Carbon nanotube-based fibers were produced by stretching the dense film with two rotational winders of the spinning machine. The two rotational winders operate with only a small difference in rotation rate.

Thermal conductivity measurement methods. To determine the thermal conductivity, the parallel thermal conductance technique was employed [31,32], since the diameter of the needle-like samples was very small. Fourier's Law was used to compute the thermal conductivity. This steady-state method has been carried out to measure the thermal conductivity of boron nitride nanotube sheets [33], and carbon nanotube sheets [34] and yarns [35,36]. The configuration of the parallel thermal conductance technique is depicted schematically in Figure 2. A preliminary measurement of thermal conductance was performed with respect to the sample holder itself so as to determine the background or base-line heat conduction and losses associated with the sample stage. The sample was attached and the thermal conductance of the system was measured. The parallel thermal conductance can be obtained by subtraction. In this method, all conductance factors arising from the thermal contacts, sample, and thermal radiation from the sample accounted for. The radiative heat losses were caused primarily by the thermal radiation from the hot surface of the heater. However, such heat losses were already included in the baseline. Therefore, a correction factor of 0.5 was introduced into the method [31,32] to account for the radiative heat losses.

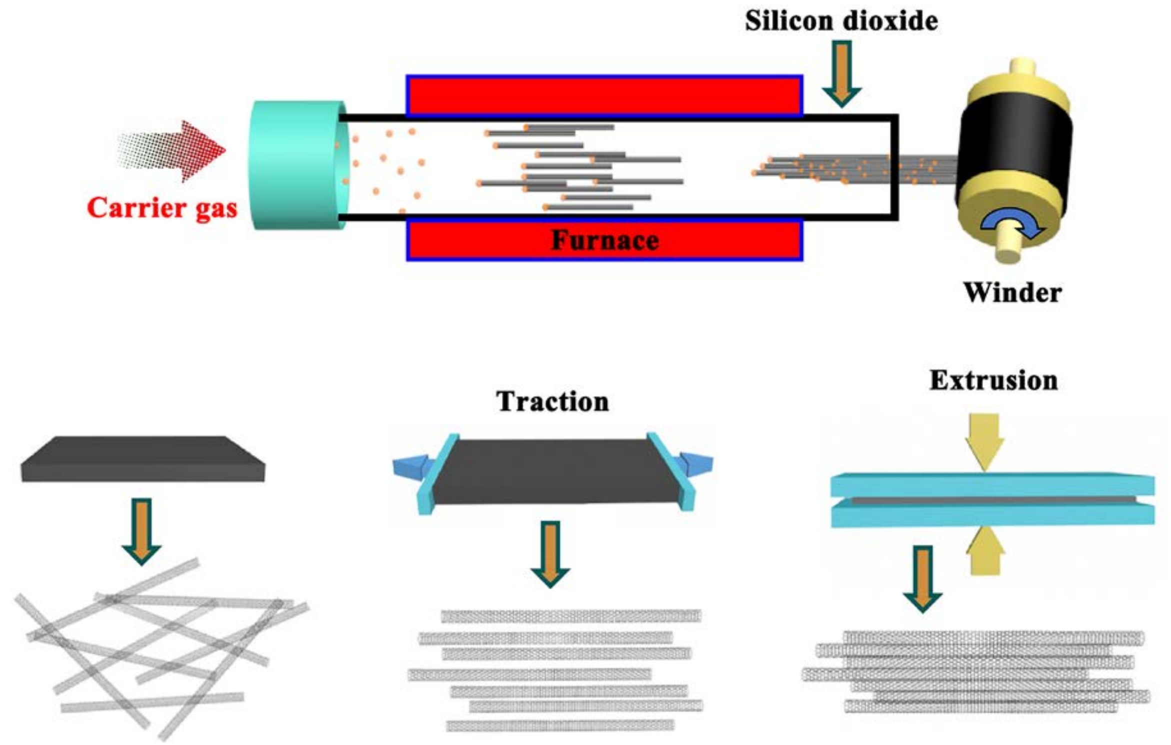

Fig. 1. Schematic illustration of the preparation process of carbon nanotube-based films. A dense film was laid down with aligned carbon nanotube bundles

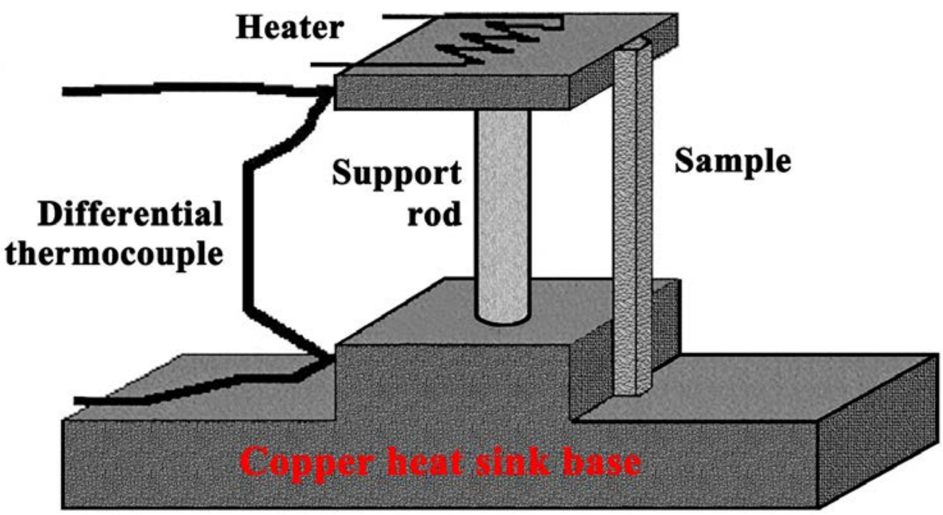

Fig. 2. Schematic illustration of the configuration of the parallel thermal conductance technique. The baseline was measured with the sample holder itself and then the sample was attached to perform the measurement of parallel thermal conductance in which the baseline was subtracted

Linear mass density measurement methods. The linear mass density of the carbon nanotube-based films was measured by using a gravimetric method [37]. In contrast, the linear mass density of the carbon nanotube-based fibers was measured by using a vibroscope method [37,38]. The carbon nanotube-based fibers were tensioned between two hard points of a mechanical vibroscope. Mechanical vibration was induced, and the fundamental frequency was measured. The specific thermal conductivity was defined as the thermal conductivity normalized by bulk density. The bulk density was determined in terms of the cross-sectional area and the linear mass density.

3. Results and discussion

Material characterization. The macroscopic fibers produced from carbon nanotubes are characterized by scanning electron microscopy. The surface morphology is analyzed with scanning electron microscope SU3800, Hitachi High-Tech Corporation. Scanning electron microscopy images are presented in Fig. 3 for the carbon nanotube fibers prepared by the method described above. The carbon nanotubes are highly aligned. However, the carbon nanotube-based fiber material contains a relatively small amount of short, deformed carbon nanotubes. The carbon nanotubes are of the order of several hundred microns.

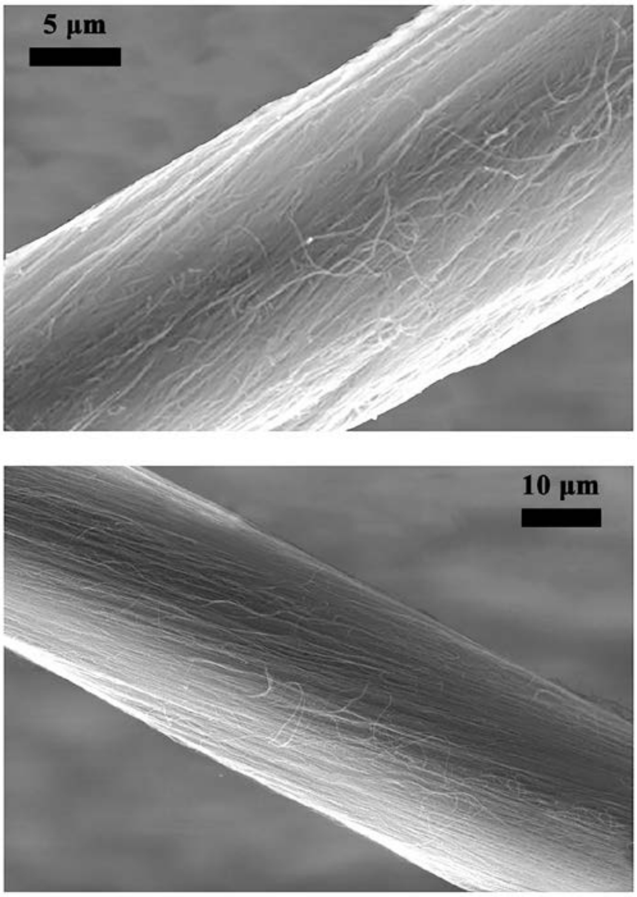

Fig. 3. Scanning electron microscopy images of the carbon nanotube fibers prepared by the method described in detail above

Four samples are prepared with different structure, purity, and alignment in order to investigate the effects of various factors on the thermal conductivity. Three structural parameters are selected in terms of alignment, purity, and the number of walls. The structural parameter space for the selection of the carbon nanotube-based materials is illustrated in Fig. 4 in the style of the design of measurement experiments. For type A, the carbon nanotube-based films or fibers are composed mostly of multi-walled carbon nanotubes. In addition, there is a small amount of short deformed carbon nanotubes. For type B, a mixture of carbon nanotubes is produced. More specifically, the carbon nanotube-based films or fibers are composed of single-walled and multi-walled carbon nanotubes. In addition, there is a small amount of amorphous carbon. For type C, the carbon nanotube-based films or fibers are composed mostly of single-walled carbon nanotubes. In addition, there is a small amount of non-carbonaceous impurities. For type D, the carbon nanotube-based films or fibers are composed of single-walled carbon nanotubes with very high purity. The understanding of which parameters are important for high thermal conductivity is made possible through the comparison of heat conduction properties between the carbon nanotube-based materials.

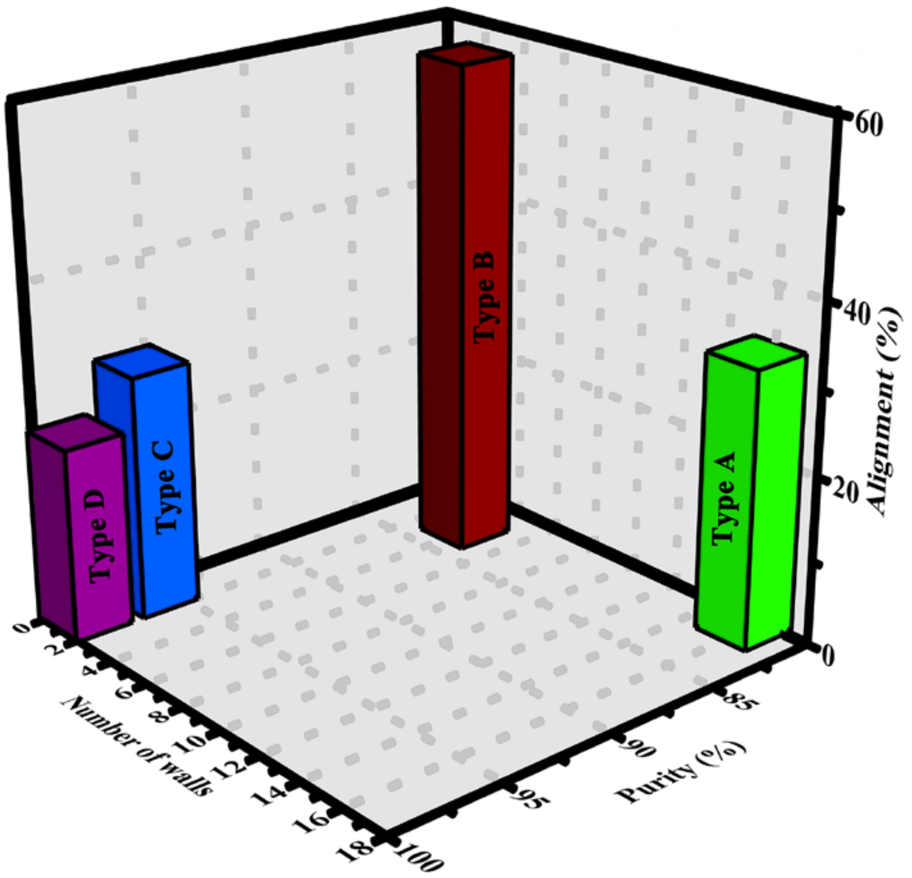

Fig. 4. Structural parameter space for the selection of the carbon nanotube-based materials in the style of the design of measurement experiments

Thermal conductivity of carbon nanotube-based films. The effects of structure, purity, and alignment on the specific thermal conductivity of carbon nanotube-based films are illustrated in Fig. 5 at different temperatures. The specific thermal conductivity of the A-type film is lower than that of the other-type films. Multi-walled carbon nanotubes generally have low thermal conductivity. The A-type film is composed mostly of multi-walled carbon nanotubes. Therefore, the specific thermal conductivity is lower than that of the film composed of single-walled carbon nanotubes. The specific thermal conductivity of the C-type film is higher than that of the A-type film, since the structure is different from each other. The structural variation will lead to the difference in thermal conductivity. However, the specific thermal conductivity of the C-type film is much lower than that of the D-type film. This is because the presence of non-carbonaceous impurities reduces the degree of bundle contact between single-walled carbon nanotubes, which increases the thermal resistance at the interface junctions and degrades the thermal performance of the C-type film.

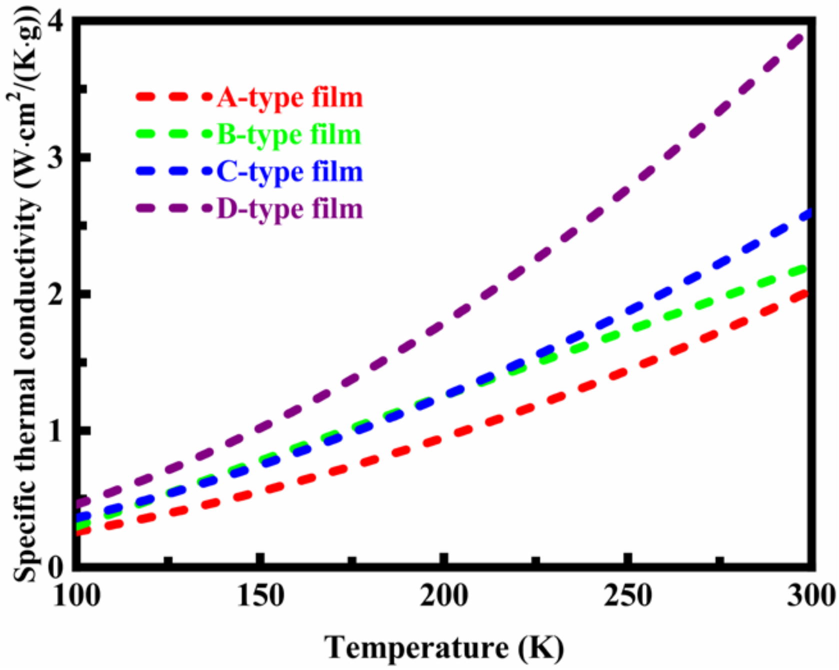

Fig. 5. Specific thermal conductivity of the carbon nanotube-based films at different temperatures. There exist significant differences in terms of structure, purity, and alignment between the films

While the specific thermal conductivity increases with temperature, the effect of temperature is different between the B-type film and the other-type films. The specific thermal conductivity of the B-type film has a linear relationship with temperature. For the other-type films, there is an exponential increase in the specific thermal conductivity of the nanostructured material. The effect of temperature on thermal conductivity is illustrated in Figure 6 for the carbon nanotube-based films. The thermal conductivity of all the carbon nanotube-based films presents a power-law dependence with respect to temperature, except that of the B-type film. The power law index is about 1.87, 1.80, and 1.96 for the thermal conductivity of the A, C, and D type films, respectively. In contrast, the temperature index is 0.99 for the thermal conductivity of the B-type film. The temperature index is small, which reduces the dimensionality of the propagation of phonons [39,40] because of an increase in the degree of bundle alignment and contact.

The effect of bulk density on the specific thermal conductivity at room temperature is illustrated in Figure 7 for the carbon nanotube-based films. At room temperature, the highest specific thermal conductivity is achieved for the D-type film. In addition, the power law index is largest, although the degree of bundle alignment is very low, which reduces the mechanical stiffness of the carbon nanotube-based film. At room temperature, the lowest specific thermal conductivity is obtained for the A-type film. A tentative explanation could be made for the distinctive phenomenon. The specific thermal conductivity of the nanostructured material decreases with increasing bulk density. The low bulk density of the D-type film compensates for the adverse effect of poor alignment on the specific thermal conductivity of the nanostructured material. Therefore, the bulk density may be controlled to produce carbon nanotube-based films with high thermal conductivity.

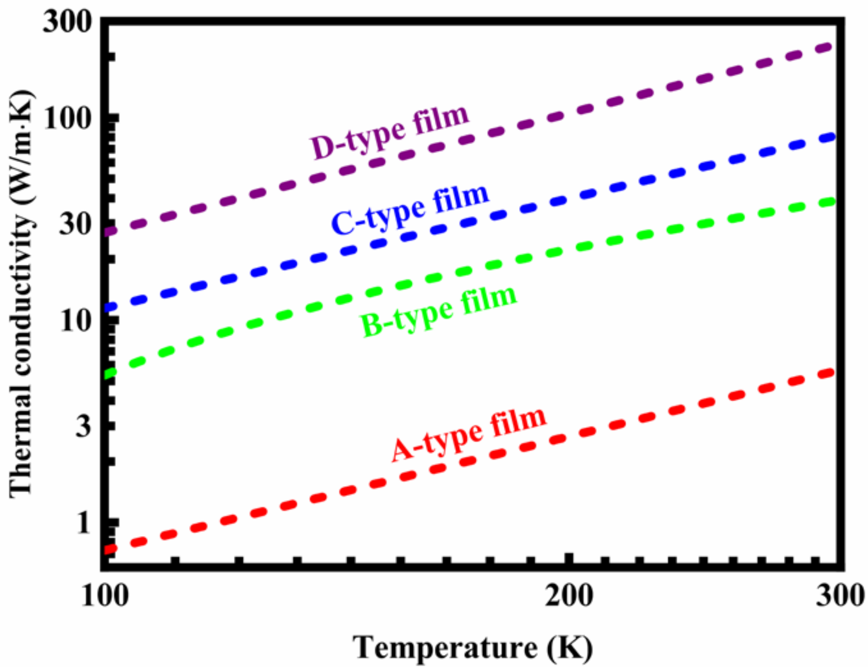

Fig. 6. Thermal conductivity of the carbon nanotube-based films at different temperatures. The thermal conductivity is indicated with points plotted on a logarithmic coordinate system

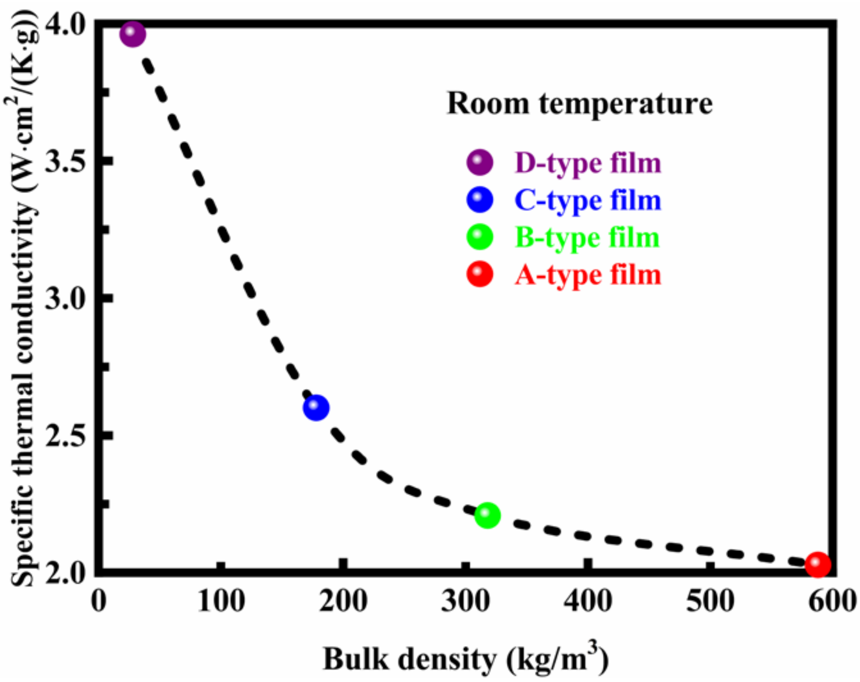

Fig. 7. Effect of bulk density on the specific thermal conductivity of the carbon nanotube-based films at room temperature

Thermal conductivity of carbon nanotube-based fibers. The effects of structure, purity, and alignment on the specific thermal conductivity of carbon nanotube-based fibers are illustrated in Fig. 8 at different temperatures. The specific thermal conductivity of the carbon nanotube-based fibers is around 10 $W \cdot cm^2/(K \cdot g)$ at room temperature, which is comparable to or higher than that of pristine and chemically modified carbon nanotube-based fibers [25,26]. A maximum specific thermal conductivity is obtained at room temperature, which appears as a peak in the dependence of temperature in Figure 8, as determined previously [26,40]. The maximum specific thermal conductivity represented by the peaks indicates the occurrence of

Umklapp scattering. The Umklapp scattering is the dominant process for thermal resistivity, which limits the specific thermal conductivity. High thermal conductivity of carbon fibers is often associated with a high modulus of elasticity [41,42]. Therefore, the carbon nanotube-based fibers have sufficient strength to be used as a reinforcement for composite materials. Clearly, the specific thermal conductivity of the carbon nanotube-based fibers is significantly higher than that of the carbon nanotube-based films. This is caused by the improved degree of bundle alignment for the carbon nanotube-based fibers. A high degree of bundle alignment will lead to an increase in thermal conductivity because of the increased interfacial area between the carbon nanotubes.

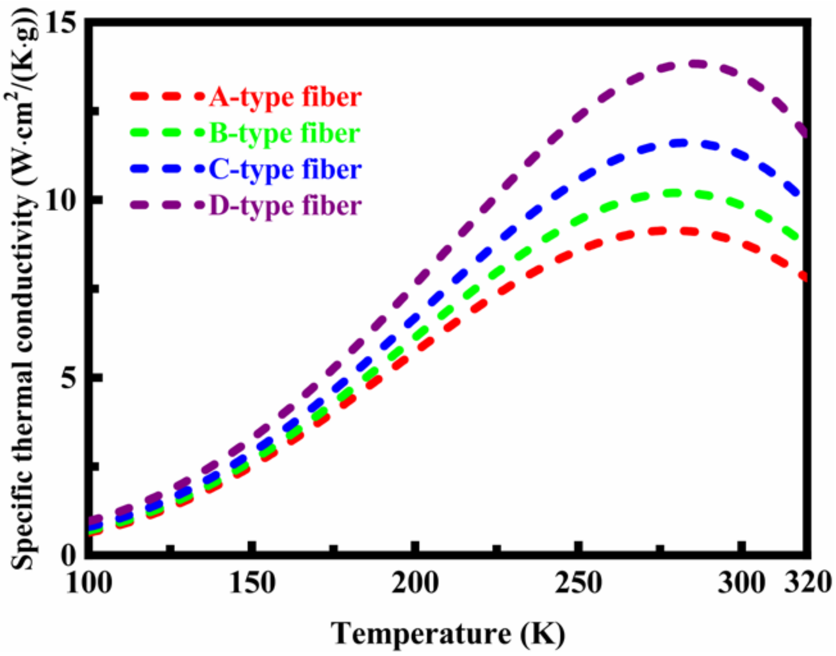

Fig. 8. Specific thermal conductivity of the carbon nanotube-based fibers at different temperatures. There exist significant differences in terms of structure, purity, and alignment between the carbon nanotube-based fibers

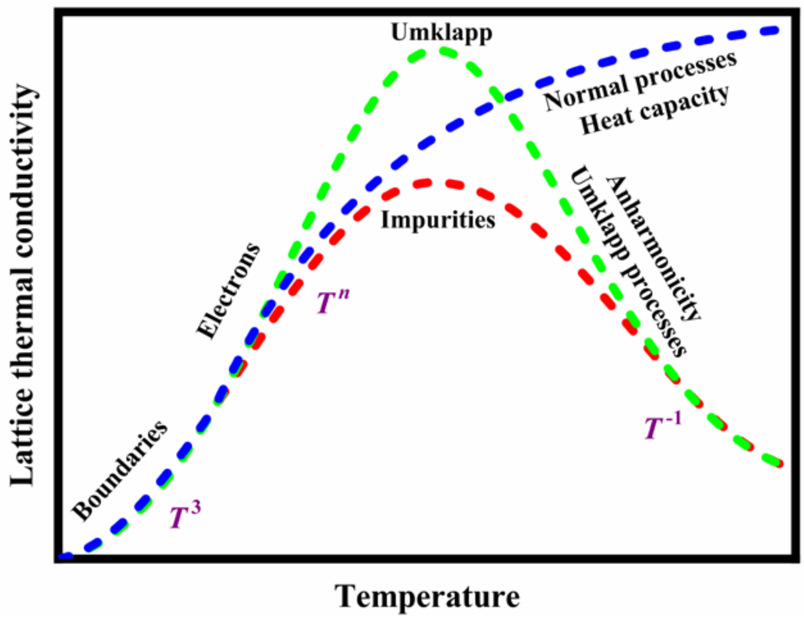

Fig. 9. Schematic illustration of the various mechanisms responsible for the temperature dependence of lattice thermal conductivity

It is important to understand the physics behind the observed behavior of the specific thermal conductivity, for example, the maximum specific thermal conductivity caused by the occurrence of Umklapp scattering. The various mechanisms responsible for the temperature dependence of lattice thermal conductivity are illustrated in Fig. 9. Assuming that the Boltzmann transport equation is valid and the relaxation time and phonon group velocity are independent of frequency, the lattice thermal conductivity k can be written as $k = \frac{1}{3}Cv_g\Lambda = \frac{1}{3}Cv_g^2\tau$, where C is the heat capacity, v_g is the phonon group velocity, Λ is the phonon mean free path, and τ is the relaxation time. Note that this assumption breaks down when the scale of the physical system is comparable to or smaller than the wavelength of the phonons responsible for thermal transport. Although the above model is fairly crude, the expression for the lattice thermal conductivity is actually a surprisingly good approximation. Very often, the above equation is used to estimate the phonon mean free path on the basis of experimental results for the other parameters in the equation. If the lattice vibrations are entirely the normal modes, the temperature dependence of lattice thermal conductivity is determined by the specific heat is therefore proportional to the cube of temperature [43,44], as illustrated in Fig. 9. A heat flow will transport without decaying or scattering, which implies that the lattice thermal conductivity be infinite as harmonic oscillations. In fact, anharmonicity persists along with the normal processes. The major mechanism of anharmonicity is called the Umklapp processes, as indicated in Fig. 9. While the normal processes conserve the total phonon momentum, the Umklapp processes change the phonon momentum, causing the scattering of phonons [43,44]. The Umklapp processes increase with increasing temperature. From the above equation, since the velocity of sound is independent of temperature, the lattice thermal conductivity depends upon the phonon mean free path and the heat capacity. At low temperatures, the Umklapp processes are reduced, and the phonon mean free path may reach several micrometers, being comparable with the scale of the physical system. The phonon mean free path thus becomes constant at the grain boundaries. Therefore, the lattice thermal conductivity depends upon the heat capacity that varies as the cube of temperature, as illustrated in Fig. 9. At high temperatures, the heat capacity is almost constant, and the phonon mean free path is inversely proportional to temperature. In this case, the Umklapp processes predominate the lattice thermal conductivity so that the lattice thermal conductivity is inversely proportional to temperature, as illustrated in Fig. 9. This dependency originates from the temperature dependency of the probability for the Umklapp processes to occur.

The effect of cross-sectional area on the specific thermal conductivity at room temperature is illustrated in Figure 10 for the carbon nanotube-based films and fibers. Some previous data are also included for comparison. These previous data are available in the literature [26,35,36,45-48]. The carbon nanotube-based fibers have higher specific thermal conductivity than the carbon nanotube-based films. Under the same cross-sectional area conditions, the specific thermal conductivity of the carbon nanotube-based films and fibers is higher than that previously reported in the literature. The carbon nanotubes are of the order of several hundred microns, which is significantly greater than the phonon mean free path. As the length of carbon nanotubes increases, the effect of temperature on thermal conductivity becomes more pronounced [12,48]. The length of the carbon nanotubes contained in the carbon nanotube-based films and fibers is much greater than that of the carbon nanotubes used in the literature. Consequently, the carbon nanotube-based films and fibers are very efficient at conducting heat and typically have higher specific thermal conductivity in comparison to the previous data.

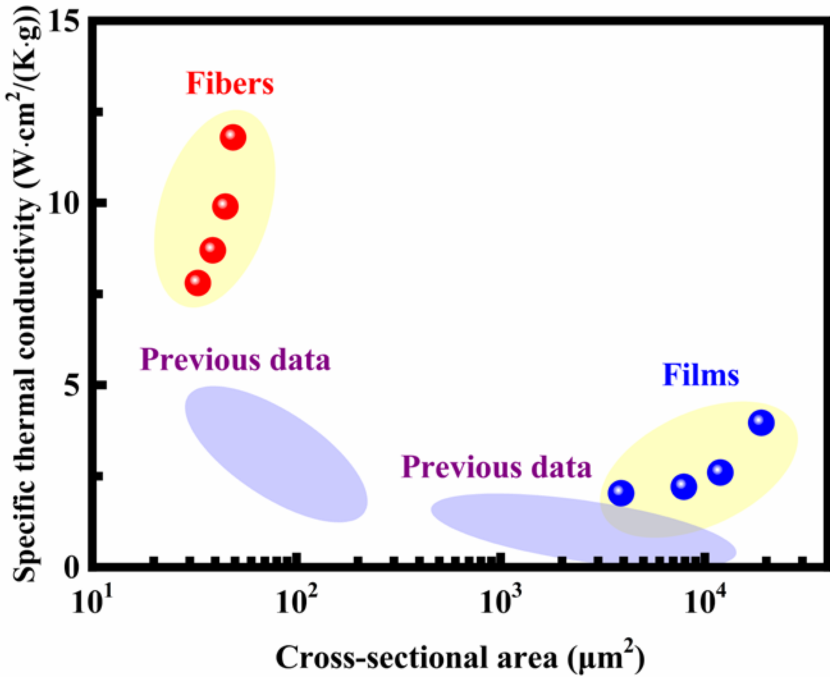

Fig. 10. Effect of cross-sectional area on the specific thermal conductivity of the carbon nanotube-based films and fibers at room temperature. Some previous data are also included for comparison

4. Conclusions

Carbon nanotube-based films and fibers were produced, and the parallel thermal conductance technique was employed to determine the thermal conductivity of carbon nanotubes. The effects of structure, purity, and alignment on the heat conduction properties of carbon nanotube-based films and fibers were investigated to understand the characteristics of thermal transport in the nanostructured materials. The major conclusions are summarized as follows:

- Macroscopic films and fibers produced from multi-walled carbon nanotubes generally have low thermal conductivity.
- The presence of non-carbonaceous impurities reduces the degree of bundle contact between carbon nanotubes, thereby increasing the thermal resistance at the interface junctions and degrading the thermal performance.
- The thermal conductivity presents a power-law dependence with respect to temperature or has a linear relationship with temperature.
- The specific thermal conductivity decreases with increasing bulk density. Low bulk density can compensate for the adverse effect of poor alignment on specific thermal conductivity.
- A maximum specific thermal conductivity is obtained at room temperature because of the occurrence of Umklapp scattering.
- The specific thermal conductivity of carbon nanotube-based fibers is significantly higher than that of carbon nanotube-based films. The improved thermal properties are caused by the increased degree of bundle alignment.
- The prepared carbon nanotube films and fibers can conduct heat very efficiently because of the increased length of the carbon nanotubes.

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