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H-C Model to Predict the Thermal Conductivity of Nanofluids

Bangming Gu

Institute of Applied Physics, Zhejiang Wanli University, 8 Qianhu South Road,
Ningbo 315101, China

Abstract: Using transient hot wire method, the effective thermal conductivity of carbon nanotubes (CNTs), silver nanowires and copper nanowires based nanofluids were measured. Taking “n” as fitting parameter, the Hamilton-Crosser model can fit well with the experimental results, which shows that shape factor should play a great role in the thermo-physical properties of the nanofluids.

Key words: Nanofluids, hamilton-crosser model, effective thermal conductivity1

INTRODUCTION

The poor thermo-physical properties of conventional heat transfer fluids such as water, ethylene glycol can no longer meet the cooling demands in many high-technology industries. For example, microprocessors become smaller but more powerful resulting in an ever-increasing thermal load. Besides, improving the efficiency of existing heat transfer process, in automotive systems for instance, is of great interest, which could lead to smaller heat exchangers for cooling and reduce the weight of the vehicle. Therefore, when Choi conceived the concept of nanofluids (NFs) in 1995, it has triggered considerable interest for its potential to enhance the heat transfer rate. Nanofluids are prepared by dispersing nano-particles or fibers in carrier fluids. Tremendous Thermal Conductivity (TC) enhancement has been observed by many groups. But, unfortunately, the experimentally observed anomalous TC enhancements cannot be predicted by conventional Maxwell's theory (Maxwell, 1881) and Hamilton and Crosser (H-C) model (Hamilton and Crosser, 1962). Lots of mechanisms were then proposed by researchers to explain the thermal transport properties of nanofluids. Those mechanisms include: Brownian motion and induced micro-convection; the shape, surface-area-to-volume ratio and size of nanoparticles; the interfacial conduction between the liquid and solid layer; clustering of nanoparticles; percolating networks easily formed by high aspect ratio fillers. But these models show large discrepancies among each other, which greatly restrict their applicability.

As a matter of fact, in the past few decades, it is reckoned that more than 300 research groups and companies are attracted in nanofluids research worldwide. Consequently, lots kinds of nanofluids have been configured out and their thermal conductivities have been

measured by different research groups. Many different kinds of nanoparticles including metallic particles/non-metallic particles/nano-fibers have been adopted as additives of nanofluids. The base fluids commonly used are water, acetone, decene, engine oil, ethylene glycol, poly oil and so on. Logically speaking, under such extensive research background, the mechanism of thermal-physical properties of nanofluids should have been understood clearly. But in fact, there is no reliable theory to predict the anomalous thermal conductivity of nanofluids until now.

As mentioned above, although many possible factors have been considered, including Brownian-motion, liquid-solid interface layer and surface charge state, so far there are no general mechanisms to rule the strange behavior of nanofluids including the highly improved effective thermal conductivity. The various theoretical models provided by different research groups usually have the following features: very complicated in order to integrate more parameters; semi-empirical equations containing one or more unknown parameters; only be applicable to a very small number of experimental results, all of which greatly hampered the application of nanofluids in heat transfer area.

Under the above background, this article is not intended to explore a new heat transfer mechanism behind the nanofluids and not to propose a new theoretical model. Based on the experimental data, we try to find out the most important factor affecting the heat transfer behavior of the nanofluids and use a simple, common the model to describe the thermal conductivity properties of nanofluids.

EXPERIMENTAL RESULTS

In this study, the TC of three different nanofluids systems with different concentrations was investigated.

The multi-walled Carbon Nano-tubes (CNTs) used in this study, made by chemical steam deposition with 95% purity, were provided by Zhejiang University. The properties are: 10-15 nm in diameter, 10-20 μm in length, thermal conductivity is about 2000 W mk⁻¹. Copper nanowires and silver nanowires are all commercially purchased from Nanjing XFNNNO Materials Tech Co., Ltd, China. The average diameter of silver nanowires and copper nano-wires is 60nm and 100-200 nm; the length are 20-30 and 0.8-6 μm, respectively. The terms CNTs-NFs, Ag-NFs and Cu-NFs were used to distinguish the three prepared nanofluids. Distill-ed water was used as the base fluid.

All the nanofluids were prepared though the so-called two-step method and polyvinyl pyrrolidone (PVP) were adopted as surfactants. The PVP has a molecular weight of 40,000, is of analytical grade and obtained from Shanghai Chemical Reagents Company. For all the samples, the nanofluids were ultrasonically oscillated at 50% amplitude using a 130 w, 20 kHz ultrasonic processor for 20 min and followed by 10 min stirring using a magnetic stirrer to obtain uniform dispersion of nanoparticles in the base fluids. The sonication was performed in an ice bath to maintain a constant temperature in the suspension,

Nanofluids with five concentrations: from 0.06-0.2% by volume were produced in this research. The nanoparticles were characterized using Scanning Electron Microscope (SEM). Thermal conductivities of all the water-based nanofluids were measured by using a transient hot wire method reported elsewhere (Gu and Wang, 2011; Gu *et al.*, 2013). In this experiment, two platinum hot wires of diameter 50 μm were used in order to eliminate the end effect. A 15 μm thick insulation layer was coated on the wire surface to minimize the leakage of electrical current from the electrodes to the surrounding fluid. To keep temperature constant, the cells were immersed in a electro thermostat DHP 3000 (WanHua Laboratory Instrument, China), the temperature fluctuant of which is smaller than 0.5°C. This hot wire serves as a heating element, through electrical resistance heating and as a thermometer simultaneously. The same heating currents were applied to both wires to compensate the end effects. By measuring the temperature dependent change in the electrical resistance of the platinum wire, the thermal conductivity can be calculated from the relationship between the electrical and the thermal conductivity. The experimental setup was calibrated by comparing the measured values of thermal conductivity for de-ionized water and ethylene glycole against literature values. The literature values of the base fluids were reproduced within an uncertainty of 2.0%. The

thermal conductivity of the fluid was measured after the nanofluid was settled for more than 30 min to eliminate the effects caused by oscillation. All the measurements were repeated at least five times to ascertain the accuracy of the experimental results.

SEM was used to acquire the morphology of the nanoparticles. The scanning electron micrograph of CNTs, Ag and Cu nanowires were showed in Fig. 1a-c, respectively. The electron micrograph indicates that the CNTs were not only aggregated but entangled Fig. 1a. Copper nanowires also showed aggregated and entangled Fig. 1c, while the silver nanowires were typically straight Fig. 1b.

The effective thermal conductivity of the CNTs-NFs, Ag-NFs and Cu-NFs were measured by transient hot wire method. Figure 2 depicts the TC enhancements of the three suspensions as a function of the volume fraction of nanoparticles (at 25°C). The ordinates is thermal conductivity enhancement ratio $\Delta\lambda/\lambda_0$, where $\Delta\lambda = \lambda_e - \lambda_0$. λ_e and λ_0 represent the effective thermal conductivities of the nanofluids and the base fluid, respectively. The x-coordinate represents the volume fraction (Φ) of the nanofluids.

MODEL DISCUSSION

Various theories have been developed to predict the thermal conductivity behavior of the nanofluids. In the present research, the H-C model [Hamilton, R.L. and Crosser, O.K., 1962] is employed:

$$\frac{\lambda_e}{\lambda_0} = \frac{\lambda_p / \lambda_0 + (n-1) - (n-1)\phi(1 - \lambda_p / \lambda_0)}{\lambda_p / \lambda_0 + (n-1) + \phi(1 - \lambda_p / \lambda_0)}$$

where, n is the particle shape factor, λ_p is the thermal conductivity of the nanoparticles. In Hamilton-Crosser' study, n should be a fixed value for a given shape. That is: for spherical particle n = 3 and for cylindrical particle n = 6. In order to use Hamilton-Crosser (H-C) model to fit the experimental thermal conductivity data, the parameter "n" is no longer fixed as 3 or 6 in this study, contrarily, the n is used as a fitting parameter. Figure 3. shows the comparison between the experimental results with calculated values using the H-C model with different 'n' values.

Using the parameter "n" in Hamilton-Crosser model to fit different experimental data is based on the following consideration:

- **Firstly:** Numerous theoretical model have been put forward to predict the thermal transport properties of

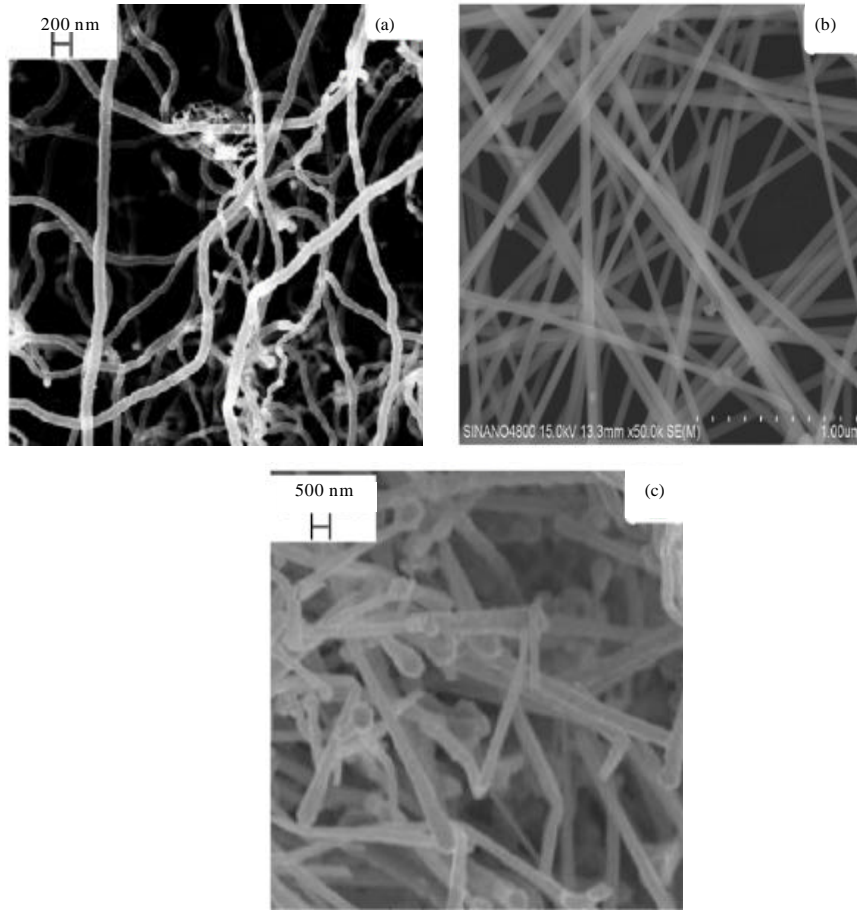


Fig. 1(a-c): SEM image of (a) MWCNTs (b) Ag and (c) Cu

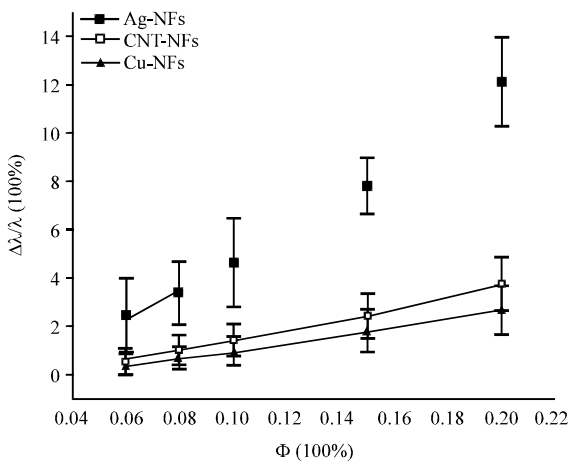


Fig. 2: Thermal conductivities of three water-based nanofluids vs., volume fraction at 298 k

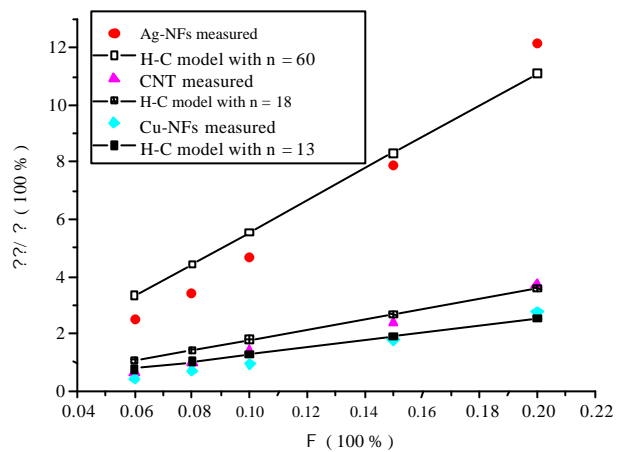


Fig. 3: Comparison between the experimental results with calculated values

the nanofluids. Those carefully constructed model do can be used to fit some experimental data. However, the

models usually have the following features: Very complicated in order to integrate more parameters;

semi-empirical equations containing one or more unknown parameters; only be applicable to a very small number of experimental results. For example, the Ganguly model (Ganguly *et al.*, 2004) includes the effects of particle size, nanolayer, particle movements, interactions and surface chemistry of nanoparticles and so on. Although the complicated model do can be perfectly fitted to certain experimental results by adjusting some empirical parameters but it is still not applicable to other nanofluids. So, why do not we use a simple formula? Indeed, despite numerous experimental and theoretical studies, a unique or integrated theory to establish the behavior of enhancement of thermal conductivity in nanofluids remains elusive.

The theoretical and experimental studies in recent years have shown the classical nature of thermal conduction in nanofluids. Indeed, all the published nanofluids data, with the exception of a few sets, fall between the upper and lower HS bounds. The publications in recent years have shown that the experimentally observed anomalous TC enhancement in spherical nanoparticle suspensions is due to aggregation of nanoparticles. That is to say, if the nanoparticles form a linear chainlike agglomeration, then the thermal conductivity enhancement can exceed that of the classical Maxwell or Hamilton-Crosser model prediction through percolation, however, most of the data still fall between the upper and lower HS bounds. That is the first reason that the classical H-C model is adopted here.

- **Secondly:** Although the classical Hamilton-Crosser model failed to predict the thermal conductivity of nanofluids, one of its important feature is that H-C model includes the particle shape effect. In other words, it can predict the effective thermal conductivity of suspensions containing spherical and non-spherical particles

Recent studies (Venkata Sastry *et al.*, 2008) have shown that the shape factor is a critical parameter for nanofluids TC enhancement, the enhancement value increased with the aspect ratio of nanoparticles increasing. Thus, the conventional H-C model cannot be readily applied to CNT-like nanofluids (nanofillers with different aspect ratios) because of their lack of accuracy of the particle size and length of the tube, thereupon, these models deviate far from the experimental data. It is believed, however, that in the modeling of the thermal conductivity for nanofluids with high aspect ratio, the shape factor should be of great importance.

In Hamilton-Crosser model, n should be a fixed value for a given shape. That is: for spherical particle $n = 3$ and

for cylindrical particle $n = 6$. Whereas, the experimental results demonstrated that the thermal conductivity varies with the nanotube's/cylinder's length, why do not we make some corresponding change?

Besides, experimental results have showed that for some nanoparticles the thermal conductivity differs greatly along the radial and axial direction. For example, thermal conductivity of CNT is highly anisotropic but the Brownian motion of the particles make them randomly positioned in the nanofluids. Experimental results demonstrated that when the particles are randomly aligned, no significant change in TC enhancement was observed, while when the particles are highly ordered-aligned in direction of the temperature gradient-great thermal conductivity enhancement was observed. Theoretical study by Kumar, S. also has similar conclusion (Kumar, 2005).

That is to say, not only the morphology of the cylinder (the aspect ratio) but the alignment of the particles affect the thermal properties of the nanofluids. We should make some amendments to the H-C model to fit the experimental result. As a result, to account for the experimental results, the 'n' is taken as fitting parameter, instead of a fixed value ($n = 6$) for cylindrical particle.

- **Thirdly:** Till now, the particle size and shape, the length-to-diameter ratio (the aspect ratio), the volume fraction, the ordered structure of the liquid at the solid-liquid interfaces, the interfacial resistance, the Brownian motion of the nanoparticles enabling the formation of different clusters, percolating structures/networks formed by nanoparticles with high aspect ratio all of the above factors or mechanism may be responsible for the unique enhancement TC of nanofluids. Various theoretic approach have been proposed by different research group taking into account one or some of the above factors. It is interesting to find that all the above factors are related to the particle shape effect. They associated with the shape under the following ways

For brownian motion: Although some controversies exist regarding the contribution of Brownian motion of nanoparticles to the effective thermal conductivity of nanofluids. But no matter how much the thermal motion of nanoparticles can affect the thermal conductivity of nanofluids, the Brownian velocity ($v = \sqrt{18k_B T / \pi \rho d^3}$) should vary with the effective particle dimension. So different aggregation structure with different effective diameter/aspect ratio resulting in different Brownian velocity and will contribute in various degrees to the TC of the system. On the other hand, the aggregation is

formed by the collision of the nanoparticles, which is directly caused by the Brownian motion of nanoparticles.

For the particle concentration: Individual spherical particles are likely to form dimmers, trimmers, which can be assumed as bricks and build some other types of clusters. Individual cylindrical particles can also accumulate into enormous structures of different sizes. Cluster size and size distribution change with the particle concentration, as a consequence, changing the effective aspect ratio as well.

For the molecular-level layer: In liquid-particle mixtures, the liquid molecules close to a particle surface are known to form layered structures and behave much like a solid. In (Choi *et al.*, 2001) Choi proved that such an organized solid-like structure of a liquid at the interface is a governing factor in heat conduction. The ordered nanolayer of liquid molecules, in fact, can be treated as the shape change of the nanoparticles.

For the thermal resistance: The interfacial resistances in nano-composites are thought to be one of the most important factors that contribute to the large discrepancies among the experimental values of the thermal conductivities in the literature. The thermal resistance at the solid-liquid interface can be expressed by the Kapitza interfacial Resistance. Different clusters have different contact areas with the surrounding liquids, resulting in different thermal resistance. In other words the interfacial resistance value changes with the shape of the nanoparticles.

In sum, the alterations of all the above-mentioned factors are equal to change of the particles' morphology, which can be characterized by the parameter "n" in Hamilton-Crosser model. To integrate all the above factors in a simple way, the 'n' is taken as a fitting parameter, instead of a fixed value ($n = 6$) for cylindrical particle.

- **Lastly:** Consideration here is not designed to provide a unique or integrated theory that can be applied to all kinds of nanofluids. But, by taking 'n' as a fitting parameter, instead of a fixed one, the Hamilton-Crosser model could be a simple, convenient equation applicable to some certain nanofluids. As a matter of fact, due to different experimental conditions, it is almost impossible to use a standard formula summarizing all of the experimental results (the experimental data differ greatly from or even contradictory with each other). The most important

work for the researchers now is not to provide new kinds of nanofluids or to put forward a complex formula. We should focus on resolving the discrepancies among different data sets presented in the literature by various groups or on confirming what's the leading factor caused the unrepeatability of the data. Taking 'n' as a fitting parameter can emphasize the key factor of the aspect ratio in the thermal conductivity enhancement of the nanofluids including non-spherical particles. The shape factor 'n' as a whole not only represents the aspect ratio; in a physical sense it also signifies the role of percolation as a mechanism of thermal transport in a nanotubes/nanowires/nanofibers-based nanofluid

From the point of view of practical application, a complicated equation cannot be conveniently used. The manufacturers care only about how to produce nanofluids that dramatically enhance the thermal conductivities of conventional heat transfer fluids. In this case, compared with those complicated models, a simple formula, which highlights the key factor in the thermal conductivity enhancement of the nanofluids, may be more convenient to be used. The experimental results in this study could provide an avenue for nanofluids' application. The poor heat transfer rates of conventional heat transfer fluids can be increased by dispersing nanoparticles with high aspect ratio in the liquid. In this case, the most important point is not to choose solids with highest thermal conductivity but to make the nanotubes/nanowires/nanofibers "straight" (not entangled) to have larger effective aspect ratio.

CONCLUSION

Particle size and shape, the aspect ratio, the volume fraction, the solid-liquid interfaces, the interfacial resistance, the Brownian motion... all of the above factors or mechanism may be responsible for the unique enhancement TC of nanofluids. It is interesting to find that all the above factors are related to the particle shape effect. In other words, the alterations of all the above-mentioned factors are equal to change of the parameter "n" in Hamilton-Crosser model (n itself depends on the shape of the dispersed particles). The thermal conductivities of MWCNTs-based, silver nanowires-based, copper nanowires-based nanofluids were measured by transient hot wire method-we proved that shape factor should be a key role in the thermal properties of nanofluids.

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