<u>Original Research Article</u> Calculation the Thermal Conductivity of Nanofluids containing Aligned Ultralong Single Walled Carbon Nanotubes

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15 ABSTRACT

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The thermal conductivity of carbon nanotubes (CNTs) depends on their length and the diameter. At room temperature, the thermal conductivity of CNTs increases as its length increases as well as its diameter decreases. Aligned long single-walled carbon nanotubes (AL-SWCNTs) are expected to be an ideal candidate for heat transfer materials owing to their small diameter, very long length and high thermal conductivity. In this work, we propose a theory model for thermal conductivity of AL-SWCNTs in nanofluids. The calculation results showed that the thermal conductivity enhancement of AL-SWCNTs nanofluids was about 18.5 times higher than that of MWCNTs nanofluids. The calculation results have confirmed the advantage of the AL-SWCNTs as excellent additive for nanofluids.

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18 Keywords: Aligned long SWCNTs, additive, nanofluid, thermal conducvitity

19 20 **1. INTRODUCTION**

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22 Since the discovery of the carbon nanotube (CNT) in 1991 [1] there have been extensive studies of its physical and chemical properties. Carbon nanotubes (CNTs) have attracted much attention because of 23 24 their unique structure and remarkable mechanical, thermal and electrical properties [2-6]. Single-walled 25 carbon nanotubes (SWNTs) are nanometer-diameter cylinders consisting of a single graphene sheet 26 wrapped up to form a tube [7]. Yu [8] and Pop [9] and their coworkers measured the thermal conductivity of SWCNTs, and found that the thermal conductivity is near 3500 W m⁻¹K⁻¹ at room temperature for an 27 SWCNT. Fujii et al [10] found in the experiment that the thermal conductivity of a carbon nanotube at 28 29 room temperature increases as its diameter decreases. Mingo et al [11] found that the thermal 30 conductivity of CNTs increases as its length increases, and thermal conductivity of CNTs reached about 31 4000 W $m^{-1}K^{-1}$ at 316 K with the tube length reaching one meter. However, long CNTs in composite are 32 usually crooked; this reduces the effective thermal conductivity of CNTs [20]. Therefore, aligned long 33 single-walled carbon (AL-SWCNTs) become the most ideal material for heat transfer owing to their small 34 diameter, large length, unique thermal conductivity, and high orientation for heat transfer [21].

In recent years, the problem of heat dissipation with features and strengthening functions of products has become more significant. Many approaches can improve the cooling system performance. The most feasible one is to enhance the heat transfer (dissipation) performance through the working fluid without modifying the mechanical designs or key components of the system. Recent studies have shown that the thermal conductivity of the suspension which contains suspended metallic or nonmetallic nanoparticles can be much higher than that of the base fluid, and it was called as "nanofluid"[12]. On this basis, adding certain kinds of nanomaterials into base fluid is considered to be a novel approach to enhance the 42 thermal conductivity in heat transfer medium [13]. Results showed that the thermal conductivity 43 enhancements of nanofluids could be influenced by multifaceted factors including the volume fraction of 44 nanoparticles, the tested temperature, thermal conductivity of the base fluid, nanoparticle size, 45 pretreatment process, and the additives of the fluids [14, 15].

46 CNTs have been used as additives in liquids to increase the thermal conductivity, one of the most 47 important issues in industry [16]. Owing to their very high thermal conductivity (above 2000 W/m.K 48 compared to thermal conductivity of Ag 419 W/m.K) [17], CNTs become ones of the most suitable nano 49 additives to fabricate the nanofluids for thermal dissipation in many industrial and consumer products [18, 50 19]. Experimental results of Hwang et al. indicated that the thermal conductivity of MWCNTs nanofluids 51 increase about 28% with 1% volume concentration of MWCNTs in ethylene glycol [24]. Lifei Chen et al. 52 also indicated that the thermal conductivity of MWCNTs nanofluids was about 10% with 1% volume 53 concentration of MWCNTs in distilled water [25].

As mentioned above, AL-SWCNTs own small diameter, large length, unique thermal conductivity, and high orientation for heat transfer, therefore AL-SWCNTs are most ideal additive for nanofluids. However so far there are no experiment and theories on AL-SWCNTs nanofluids. In this paper, we present the first calculation results on thermal conductivity enhancements of several AL-SWCNTs nanofluids.

59 2. CALCULATION METHOD

61 There are some more models for predicting the thermal conductivity of CNT-nanofluids [26]. Traditional 62 composite models, such as Maxwell [27], Hamilton-Crosser [28], Jeffrey [29], Davis [30], etc., were 63 proved to be inadequate, as they grossly under predicted the experimental data. The Yingsong Zheng 64 model [31] uses the average length efficiency of CNTs for calculations; however it is difficult to determine the average length efficiency in fact. In the Rashmi Walvekar model [32], the calculation results are very 65 high compared to the experimental results. In the S.U.S Choi model [33], an empirical parameter α must 66 67 be used for calculations. Therefore, it still has to be based on experimental results to select the value of the empirical parameter α in order to achieve accurate calculations. Similarly, the Seved Masoud 68 Hosseini model [34] must use three empirical parameters for calculations. The Venkata Sastry model [35] 69 70 has an important face for calculations because of the randomness of the orientation of each CNT and the 71 length of the CNT participating in percolation. On the other hand, the Venkata Sastry model must use the interfacial thermal conductance, which is an empirical parameter with wide ranging values, so it still has to 72 73 be based on experimental results in order to select the value of interfacial thermal conductance for 74 calculations. 75

In 2004, a model for thermal conductivity of nanofluids is derived from Hemanth et al, which is given for nanoparticle suspensions [22]. This model assumed that there are two parallel paths of heat flow through the suspension, one through the liquid particles and the other through the nanoparticles. In this model, the effective thermal conductivity is expressed as [22]:

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$$k_{eff} = k_m \left[1 + \frac{k_p \varepsilon r_m}{k_m (1 - \varepsilon) r_p} \right]$$
(1)

where k, r denote the thermal conductivity and radii; ε denote the volume fraction of the nanoparticles.
Subscripts "*m*" and "p" denote quantities corresponding to the liquid medium and solid nano particles,
respectively.

85 In 2008, Sarit K Das et al [23] used the expression (1) for thermal conductivity enhancement in carbon 86 nanotubes suspensions denoting the liquid molecule radii and CNTs diameter to be r_l and r_s as well as 87 volume fraction of the nanoparticles as ε and the volume fraction of the liquid as $(1 - \varepsilon)$. The effective 88 thermal conductivity of CNTs nanofluids is expressed as [23]:

$$k_{eff} = k_l \left[1 + \frac{k_s \mathcal{E} r_l}{k_l (1 - \mathcal{E}) r_s} \right]$$
⁽²⁾

90 where, subscripts "*I*" and "*s*", denote quantities corresponding to the liquid medium and carbon 91 nanotubes, respectively.

92 However, calculated results of the Sarit K Das el al. are higher than the experimental results. This is 93 because the shape of CNTs are cylindrical rather than spherical, and CNTs are very good thermal 94 conductors along the tube but good insulators laterally to the tube axis. Therefore, we developed a 95 modified model for accurately predicting the CNT-nanofluids' thermal conductivity, which takes into 96 consideration cylindrical shape as well as good thermal conductors along the tube of CNTs.

97 As we already know, CNTs are very good thermal conductors along the tube, but good insulators laterally 98 to the tube axis. On the other hand, CNTs disperse in nanofluids in all direction randomly. Therefore, by 99 calculation we obtained the effective thermal conductivity of CNTs (k_{eff-CNT}) by the following equation [26]:

$$k_{eff-CNT} = \frac{1}{2} k_{CNT}$$
(3)

101 In modified model, we replace the thermal conductivity of CNTs (k_{CNT}) with the effective thermal 102 conductivity of CNTs (k_{eff-CNT}) for all calculations. On the other hand, when using cylindrical shape of 103 CNTs in the model, we also obtained the effective thermal conductivity of CNT-nanofluids as follow [26]:

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$$\frac{k_{eff}}{k_l} = 1 + \frac{2}{3} \frac{k_{eff-CNT} \varepsilon r_l}{k_l (1-\varepsilon) r_{CNT}}$$
(4)

From equation (3) and equation (4) we obtained the effective thermal conductivity of CNT-nanofluids as
 follow [26]:

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$$\frac{k_{eff}}{k_l} = 1 + \frac{1}{3} \frac{k_{CNT} \varepsilon r_l}{k_l (1 - \varepsilon) r_{CNT}}$$
(5)

where, subscripts "*l*" and "*CNT*", denote quantities corresponding to the liquid medium and carbon nanotubes, respectively. The our modified model was compared to some experimental data of several other research groups, and the results show that modified model has correctly predicted the trends observed in experimental data. The modified model was published in Physics of Fluids in March 2015 [26].



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Figure 1. The effective length of some types CNTs

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Equation (5) shows that: the thermal conductivity of CNTs nanofluids strongly depend on diameter, 115 volume fraction, and thermal conductivity of CNTs. As we know, thermal conductivity of SWCNTs higher 116 117 than that of MWCNTs, and thermal conductivity of a carbon nanotube increases as its diameter decreases as well as its length increases [10, 11]. In other hand, the thermal conductivity of CNTs 118 119 nanofluids also depend on effective length of CNTs (as figure 1) [20]. Therefore, AL-SWCNTs are the 120 best additives for nanofluids. According to the dependence of thermal conductivity of SWCNTs on its 121 length and diameter, (which were reported by Yu et al [8], Fujii et al [10] and Mingo et al [11]) and the model of Sarit K Das et al (2008) [23], we calculated thermal conductivity enhancement of AL-SWCNTs 122 123 nanofluids by using our modified model.

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125 3. RESULTS AND DISCUSSION

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127 **3.1. Thermal conductivity of AL-SWCNTs based distilled water**128

129 The diameters of SWCNTs in our calculation were 3nm, 9.8 nm, 16.1 nm, and 28.2 nm, respectively; the lengths of SWCNTs were in the range of 2.76 - 3.70 µm. Table 1 shows the dependence of thermal 130 conductivity of SWCNTs on its diameters at room temperature. In the calculation, thermal conductivity of 131 132 distilled water (DW) and the radius of water molecule were 0.6 W/mK and 0.1 nm, respectively. The volume concentrations of SWCNTs in DW were in the range of 0.1 % – 1 % (equivalent to ε = 0.001 -133 0.01). Because the volume concentration of SWCNTs in nanofluids is very small, therefore the correlation 134 between the volume concentration of SWCNTs and thermal conductivity of nano fluids in equation (5) is 135 136 linear. Furthermore the influence of SWCNTs structure (including diameter and length) to the thermal 137 conductivity of the nanofluids is important issue for discussion. Therefore, we focus investigated the dependent of nanofluid's thermal conductivity on SWCNTs structure. Figure 2 shows the dependence of 138 thermal conductivity enhancement of AL-SWCNTs nanofluids on diameter and volume concentration of 139 140 AL-SWCNTs.

141Table 1. The dependence of thermal conductivity of SWCNTs on its diameters at room142temperature

Diameter of SWCNTs	Thermal conductivity of SWCNTs	References
3 nm	2800 W/mK	[8]
9.8 nm	2100 W/mK	[10]
16.1 nm	1600 W/mK	[10]
28.2 nm	500 W/mK	[10]

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Figure 2 showed that the thermal conductivity enhancement of SWCNTs based DW increased as volume 144 concentration of SWCNTs increased. At 1% of volume concentration, the thermal conductivity 145 enhancements of SWCNTs based DW were 104.7%, 24.1%, 11.2%, and 2% corresponding to diameters 146 of SWCNTs were 3 nm, 9.8 nm, 16.1 nm, and 28.2 nm, respectively. These results indicated that thermal 147 conductivity of SWCNTs based DW increased as diameter of SWCNTs decreased. In order to predict the 148 effect of AL-SWCNTs length to the thermal conductivity of AL-SWCNT based DW, we choose 3 nm of the 149 150 AL-SWCNTs diameter, and from 3 µm to 200 µm of the AL-SWCNTs lengths. Table 2 shows the 151 dependence of thermal conductivity of AL-SWCNTs on its length at room temperature. Figure 3 shows 152 the dependence of thermal conductivity enhancement of AL-SWCNTs nanofluids on length and volume 153 concentration of AL-SWCNTs.

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Figure 2. The dependence of thermal conductivity enhancement of AL-SWCNTs based DW on its
 diameter

158	Table 2. The de	pendence of thermal	conductivity of AL	-SWCNTs on its leng	gth at room temp	perature

Length of AL-SWCNTs	Thermal conductivity of AL-SWCNTs	References	
~ 3 µm	2800 W/mK	[8], [11]	
10 µm	3000 W/mK	[11]	
20 µm	3600 W/mK	[11]	
30 µm	3900 W/mK	[11]	
40 µm	4100 W/mK	[11]	
80 µm	4600 W/mK	[11]	
130 µm	5000 W/mK	[11]	
170 µm	5000 W/mK	[11]	
200 µm	5000 W/mK	[11]	

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Figure 3 show that the thermal conductivity enhancement of AL-SWCNTs based DW increased as concentration of AL-SWCNTs increased. At 1% of volume concentration, the thermal conductivity enhancements of AL-SWCNTs based DW were 104.6%, 112.2%, 134.7%, 145.9%, 153.4%, 172.1% and 187.1% corresponding to the lengths of AL-SWCNTs were 3 µm, 10 µm, 20 µm, 30 µm, 40 µm, 80 µm 164 and 130 µm, respectively. These results indicated that thermal conductivity of AL-SWCNTs based DW 165 increased as length of SWCNTs increased. However, thermal conductivity enhancement of AL-SWCNTs 166 based DW reached saturated value at 187.1% when length of AL-SWCNTs reached 130 µm. 167 Experimental data of Hwang et al (2006) [24] and Lifei Chen et al (2008) [25] indicated that the thermal 168 conductivity enhancement of MWCNTs based DW was about 10% as volume concentration of MWCNTs 169 was 1%. Therefore, thermal conductivity enhancement of AL-SWCNTs nanofluids was 18.7 times higher 170 than that of MWCNTs nanofluids at 1% of CNTs volume concentration.



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Figure 3. The dependence of thermal conductivity enhancement of AL-SWCNTs based DW on its length and volume concentration

174 **3.2. Thermal conductivity of AL-SWCNTs based ethylene glycol**

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Similarly, we calculated the thermal conductivity enhancement of AL-SWCNTs based ethylene glycol (EG) with the diameter and length of AL-SWCNTs were 3 nm and 3 μ m - 200 μ m, respectively. In the calculation, thermal conductivity of EG and the effective radius of EG molecule were 0.26 W/mK and 0.12 nm, respectively. The volume concentrations of AL-SWCNTs in EG were in the range of 0.1 % – 1 %. Figure 4 shows the dependence of thermal conductivity enhancement of AL-SWCNTs based EG on the length and volume concentration of AL-SWCNTs.

182 Figure 4 showed that the thermal conductivity enhancement of AL-SWCNTs based EG increased as 183 concentration of AL-SWCNTs increased. At 1% of volume concentration, the thermal conductivity 184 enhancements of AL-SWCNTs based EG were 290.1%, 310.8%, 373.0%, 404.0%, 424.8%, 476.6% and 185 518.0% corresponding to the lengths of AL-SWCNTs were 3 µm, 10 µm, 20 µm, 30 µm, 40 µm, 80 µm 186 and 130 µm, respectively. These results indicated that thermal conductivity of AL-SWCNTs based EG increased as length of SWCNTs increased. However, thermal conductivity enhancement of AL-SWCNTs 187 based EG reached saturated value at 518% when length of AL-SWCNTs reached 130 µm. Experimental 188 data of Hwang et al (2006) [24] indicated that the thermal conductivity enhancement of MWCNTs based 189 EG was about 28% when volume concentration of MWCNTs was 1%. Therefore, thermal conductivity 190 enhancement of AL-SWCNTs based EG was 18.5 times higher than that of MWCNTs based EG at 1% of 191 192 CNTs volume concentration.





Figure 4. The dependence of thermal conductivity enhancement of AL-SWCNTs based EG on its length and volume concentration

196 4. CONCLUSION

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198 The thermal conductivity enhancements of AL-SWCNTs nanofluids were calculated and compared to that of MWCNTs nanofluids. The thermal conductivity enhancement of AL-SWCNTs nanofluids reached 199 200 saturated values when length of AL-SWCNTs reached 130 µm. At 1% of CNTs volume concentration and 201 130 µm of CNTs length, the thermal conductivity enhancements of AL-SWCNTs based DW and AL-202 SWCNTs based EG were 187.1% and 777%, respectively. These results showed that the thermal 203 conductivity enhancement of AL-SWCNTs nanofluids was about 18.5 times higher than that of MWCNTs 204 nanofluids. The calculation results have confirmed the advantage of the AL-SWCNTs as excellent additive for nanofluids. We expected to hear about the experimental results on unique thermal 205 conductivity of AL-SWCNTs nanofluids in near future. With unique thermal conductivity, the AL-SWCNTs 206 207 nanofluids will open up the potential application in heat dissipation for high power electronic devices, 208 engines and other heat transfer systems. 209

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216 **COMPETING INTERESTS**

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218 Authors have declared that no competing interests exist.

219 **AUTHORS' CONTRIBUTIONS** 220

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222 Author Thang and Author Hong designed the study, performed the statistical analysis, wrote the protocol, and wrote the first draft of the manuscript. Author Minh managed the analyses of the study. All authors 223 224 read and approved the final manuscript.

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