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Research Paper

A New Modified Hamilton-Crosser and Nan Models for Thermal Conductivity of Different Lengths Carbon Nanotubes Water-based Nanofluids

Mansoor Farbod, Ameneh Ahangarpour

Physic Department, Faculty of Science, Shahid Chamran University of Ahvaz, Ahvaz, I.R. Iran, Emails: farbod_m@scu.ac.ir, a.ahangarpour@scu.ac.ir

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Corresponding author: Mansoor Farbod (farbod_m@scu.ac.ir)

Abstract. In order to investigate the shape effect of nanoadditives on thermal conductivity of nanofluids, different length carbon nanotubes (CNTs) are made and using a two-step method, different nanofluids are prepared. The CNTs are cut into different lengths by functionalization at different refluxing times of 1, 2 and 4 hours. To probe the effect of aspect ratio of CNTs on the obtained experimental data, modified Hamilton-Crosser and Nan models are developed. It is found that the original Hamilton-Crosser and Nan models could not predict the experimental thermal conductivities. By replacing $n = 6 + xL/D$ instead of the shape factor of n=6 in the Hamilton- Crosser, where L and D were length and diameter of CNTs and also by replacing φ (xL/D) instead of φ (volume fraction) in the Nan model, the prediction of modified equations show very good accordance with the experimental data which means the shape of nanoadditives has high impact on nanofluids' properties.

Keywords: Hamilton-Crosser model, Nan model, Model modifications, Thermal conductivity.

1. Introduction

Nanofluids, which were introduced by Choi [1] in 1995, are a homogenous mixture of nanoparticles and a base fluid. They are practical engineering materials which have higher heat transfer performance compared to the usual fluids such as water, ethylene glycol and oil. Researchers have used different nanoparticles such as metal nanoparticles (Al, Cu, Ag, Au, Fe and so on), nonmetal nanoparticles (Al_2O_3, CuO, TiO_2, SiC and so on) and carbon nanotubes (CNTs) to prepare nanofluids. Among them, the CNTs were more attractive because of their high thermal conductivity (~3000 W/m.K), high aspect ratio and unique electrical, optical and mechanical properties [2-3]. Experimental results have shown that there is a strong relationship between the thermal conductivity of nanofluids and different parameters such as nanoparticle volume fraction, nanoparticle size, nanoparticle shape, thermal conductivity of nanoparticle and base fluid, temperature, PH and surfactant [4-10]. For example, Lyu et.al [11] have investigated the effects of ultrasonication time on the stability and thermophysical properties of MWCNT-water nanofluid, over a different range of temperatures and solid concentrations. Also, the effects of the addition of MWCNTs on the heat transfer and pumping power have been studied. It was found that the nanofluid is a good heat transfer fluid, with a negligible penalty in pumping power.
The experimental results have been interpreted and analyzed by different mechanism such as nanoparticles Brownian motion, nanoparticles aggregation, formation of liquid layer around nanoparticles on the nanoparticle/liquid interface, chemistry of surface and convection [12-18]. Based on these mechanisms, different models have been proposed to predict the experimental data. Some of these models have been either proposed to predict certain nanofluids or considered some of effective parameters on thermal conductivity of nanofluids. Most of these models have modified the classical models such as Maxwell [19], Hamilton-Crosser [20] and Jeffry [21] which had been used to estimate the thermal conductivity of fluids containing millimeter and micrometer size particles, to predict the thermal conductivity of nanofluids. Because the classical models just had considered some limited parameters such as thermal conductivity of base fluid, thermal conductivity of particles, particle volume fraction and particle shape factors, they had not been able to predict the thermal conductivity of nanofluids in their initial form and needed some modifications. Different researchers have tried to modify the classical models by introducing some new considerations [22-25]. For example, Yang et.al [26] developed a model for the effective thermal conductivity of nanofluid in axial and radial direction of a finite length cylindrical nanoparticle in a fluid medium by considering the effect of interfacial layer. The results of investigation the effect of particle's shape namely length-to-diameter ratio of particle on the thermal conductivity of nanofluids containing cylindrical nanoparticle showed that the thermal conductivity of these nanofluids increases firstly and then trends to be stable with the increasing of length-to-diameter ratio of additive particles. This model was in good agreement with the available experimental data for nanofluids with different types and length-to-diameter ratios. Also, in another paper Yang et.al [27] proposed a new thermal conductivity model for nanorod-based nanofluids. The model assumed a nanorod with layer is split apart in axial and radial direction. In this model, the allocation proportion of heat conduction in axial and radial directions was depended on the

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aspect ratio of the nanorod. The model showed a better precision for nanorod-based nanofluids. In addition, some studies were focused on the effect of the magnetic field on the ferrofluids which are considered a subclass of nanofluids. Mousavi et.al [28] developed a structural model to predict in-field thermal conductivity of ferrofluid at low and high magnetic fields. In this model the aggregation of the magnetic particles with increasing the magnetic field strength was considered. The model was employed to predict the thermal conductivities of the Fe_3O_4-kerosene and Fe_3O_4-ethylene glycol ferrofluids using the experimental data and the agreement was good. Also, Bunoiu et.al [29] determined the effective thermal conductivity for a kerosene-based ferrofluid with magnetite particles, both in absence and presence of a static magnetic field. They found that by applying a static magnetic field, the effective thermal conductivity was increased. In addition, they computed the effective specific heat and the effective thermal diffusivity both in the absence and presence of the static magnetic field and found that these parameters can be controlled by the magnetic field. Amani et.al [30] have investigated the applicability of an ANN and genetic algorithms for modeling and multiobjective optimization of the thermal conductivity and viscosity of water-based spinel-type $MnFe_2O_4$ nanofluid under magnetic field. They obtained the optimal conditions, i.e., maximum thermal conductivity and minimum nanofluid viscosity by this modeling.

In this work the experimental data for thermal conductivity of carbon nanotubes water based nanofluids have been examined using the classical models. In order to predict the results, the classical models were modified by introducing a correction coefficient and aspect ratio of carbon nanotubes.

2. Experiments and Models

In the previous work done by the authors [31], thermal conductivity of water-based carbon nanontubes' nanofluids were measured at different temperatures with different concentrations of carbon nanotubes. Thermal conductivity of samples was measured by using KD2-Pro thermal analyzer (Decagon devices, Inc., USA). The maximum error of measurement was ±5%. It is important to notice that the KD2-Pro has the ability to show the quality of the measurement by an error factor. If the error is less than 0.01, the thermal conductivity value is reliable. To ensure a precise measurement, all measurements were repeated 10 times and the average value was reported. In order to have stable suspensions, functionalized carbon nanotubes were used. The functionalization process was done by refluxing carbon nanotubes in the mixture of sulfuric and nitric acids with the ratio of 3:1 and refluxing time of 1, 2 and 4 hours. It was found that the lengths of carbon nanotubes were decreased by functionalization and also revealed that the longer the refluxing time the shorter the length of CNTs. The micrometer lengths of CNTs were decreased to 203, 171 and 135 nm after 1, 2 and 4 hours refluxing time. Figure 1 shows the SEM images of pristine and the refluxed CNTs. In this way, the authors were able to prepare different length CNTs by which different nanofluids were prepared. The thermal conductivities of such nanofluids were measured and the data were compared with some classical models. Figure 2 shows the experimental data measured at 40 °C against CNTs volume fraction and those predicted by classical models using same data. In this figure, K_{eff}/K_{bf} is the ratio of thermal conductivity of nanofluids and that of the base fluid which was deionized water.

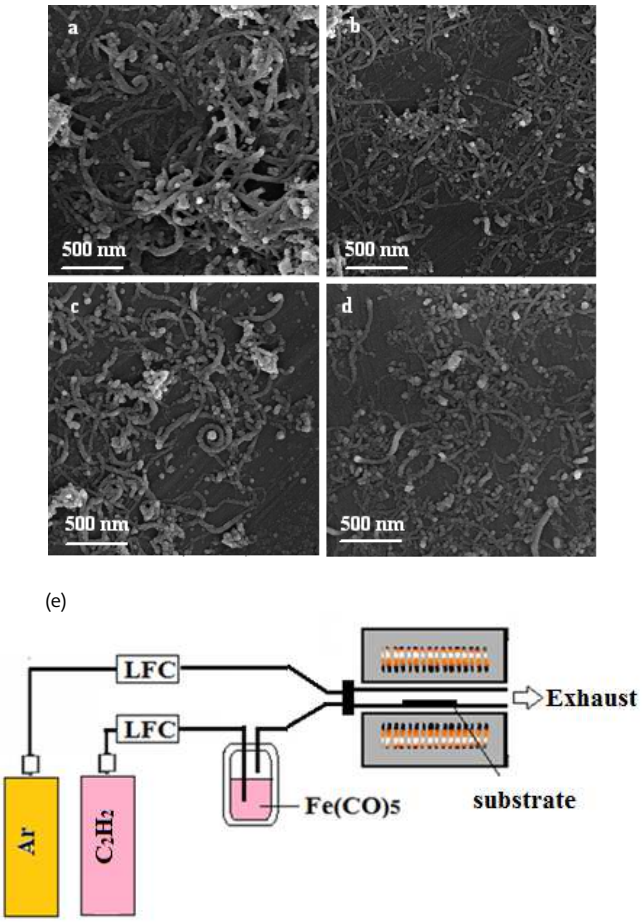

Fig. 1. SEM images of a) pristine CNTs and functionalized MWCNTs at, b) 1, c) 2, d) 4 hours refluxing times, e) a view of the system used in the chemical vapor deposition method for synthesis of CNTs.

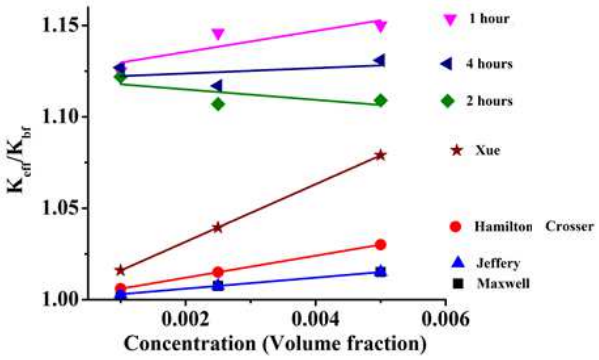

Fig. 2. Comparison of K_{eff}/K_{bf} nanofluids prepared using different time functionalized CNTs and the prediction of some classical models using the same data.

It should be noted that the K_{eff}/K_{bf} was also calculated based on Nan model, but because the prediction of the thermal conductivity ratio was much higher than those plotted in Fig 2, its curve was not shown in this figure. The predicted values of thermal conductivity ratio by Nan model for carbon nanotube volume percentages of 0.1, 0.25 and 0.5 were 2.27, 5.31, and 9.64 respectively.

As can be observed, the predicted values of classical models are quite far from the experimental data. The first model that predicted the thermal conductivity was the Maxwell model [19]. This model had considered a highly diluted suspension containing noninteracting spherical particles as:

$$k_{eff} = \frac{k_p + 2k_f + 2(k_p - k_f)\varphi}{k_p + 2k_f - (k_p - k_f)\varphi} k_f \tag{1}$$

where K_{eff}, K_p and K_f are the thermal conductivities of the suspension, spherical particles and base fluid respectively and ϕ is the volume concentration of the suspended particles. This model was successful to predict the thermal conductivity of suspensions containing millimeter and micrometer sized particles with low volume concentration. But, in the case of nanometer particles, the intersection resistance of particles is a parameter that should be considered, because such a resistance can reduce the thermal conductivity enhancement compared to the case of millimeter and micrometer particles [32].

Some models suggested different corrections to the Maxwell model. The Hamilton-Crosser model was in fact a modified Maxwell model to consider the particles' geometry by introducing an empirical shape factor of n as follows:

$$k_{eff} = \frac{k_p + (n-1)k_f - (n-1)(k_f - k_p)\varphi}{k_p + (n-1)k_f + (k_f - k_p)\varphi} k_f \tag{2}$$

where n is 3 for spherical particles and is 6 for cylindrical particles.

Further investigations on the particle geometry effects were done and found that the particle geometry has a key role in the thermal conductivity enhancement of nanofluids. It was shown that the stretched particles due to having longer contacts with each other compared to the spherical particles can enhance the thermal conductivity and so the heat transfer ability of nanofluid [33].

The Nan model predicted the thermal conductivity of a suspension containing carbon nanotubes [34] as:

$$k_{eff} = \frac{3 + \varphi\left(\frac{k_c}{k_f}\right)}{3 - 2\varphi} k_f \tag{3}$$

For $k_p >> k_f$, this equation is simplified as:

$$k_{eff} = k_f + \frac{1}{3}\varphi k_c \tag{4}$$

where k_{eff}, k_c and k_f are the thermal conductivities of the suspension, carbon nanotubes and base fluid respectively and ϕ is the volume concentration of carbon nanotubes. Because the Hamilton-Crosser and Nan models have parameters to consider the effect of cylindrical particles and carbon nanotubes, they were used to predict the thermal conductivity of our samples. It is realized that their predicted values had significant differences with our experimental data (Fig. 2). So the possibility of using the correction coefficient in these equations was investigated. According to our previous work [31] the length of carbon nanotubes was decreased with increasing the refluxing time so the correction coefficient was chosen somehow to be related to the length of carbon nanotubes. Since the length of carbon nanotubes is a dimensional quantity, the aspect ratio (the ratio of length to diameter) of CNTs is introduced as a dimensionless parameter. In the following, the corrections are applied to Hamilton-Crosser and Nan models. It is to be mentioned that both models did not consider the temperature dependence of thermal conductivity in their models, so the results of thermal conductivity at 40 °C were used. In order to achieve a completed equation for thermal conductivity of nanofluids, it is necessary to consider a temperature dependence parameter in the equations.

3. Results and Discussion

3.1 Suggested corrections on Hamilton- Crosser model

In the Hamilton-Crosser model, the shape factor of n had been considered 6 for cylindrical particles. In order to correct this model, a modified shape factor of $n = 6 + x\frac{L}{D}$ was considered, where x is the correction coefficient and $\frac{L}{D}$ is the aspect ratio of CNTs (L and D were the lengths and diameter of CNTs respectively). Since the aspect ratio is constant for CNTs in each sample, the different values of x were tested. The effective thermal conductivities (K_{eff}/K_f) were computed for x = 1, 2, 3, 4, 5 and compared with the experimental data. In order to calculate the thermal conductivities with different models, the thermal conductivity of CNTs and deionized water are chosen 3000 W/m.K [2] and 0.58 W/m.K respectively. The thermal conductivity of deionized water was measured by our team using KD2-Pro thermal properties Analyzer. Also, the volume concentrations of CNTs (ϕ) were 0.001, 0.0025 and 0.005 and the CNTs' diameter was 10 nm. As mentioned, the lengths of CNTs were 203, 171 and 135 nm for 1, 2 and 4 hours refluxing time, according to [31]. The related curves are shown in Fig 3.

As can be observed the modified Hamilton-Crosser model for $x = 1, 2$ is well matched to the experimental data. The accordance for $x = 1$ with volume fraction of 0.005 and for $x = 2$ and 3 with volume fraction of 0.0025 and for $x = 4$ and 5 with volume fraction of 0.001 is remarkable. This means that for complicated nanoadditives, some parameters such as shape factor needed to be modified to predict the experimental data correctly and also other parameters such as volume fraction are very important.

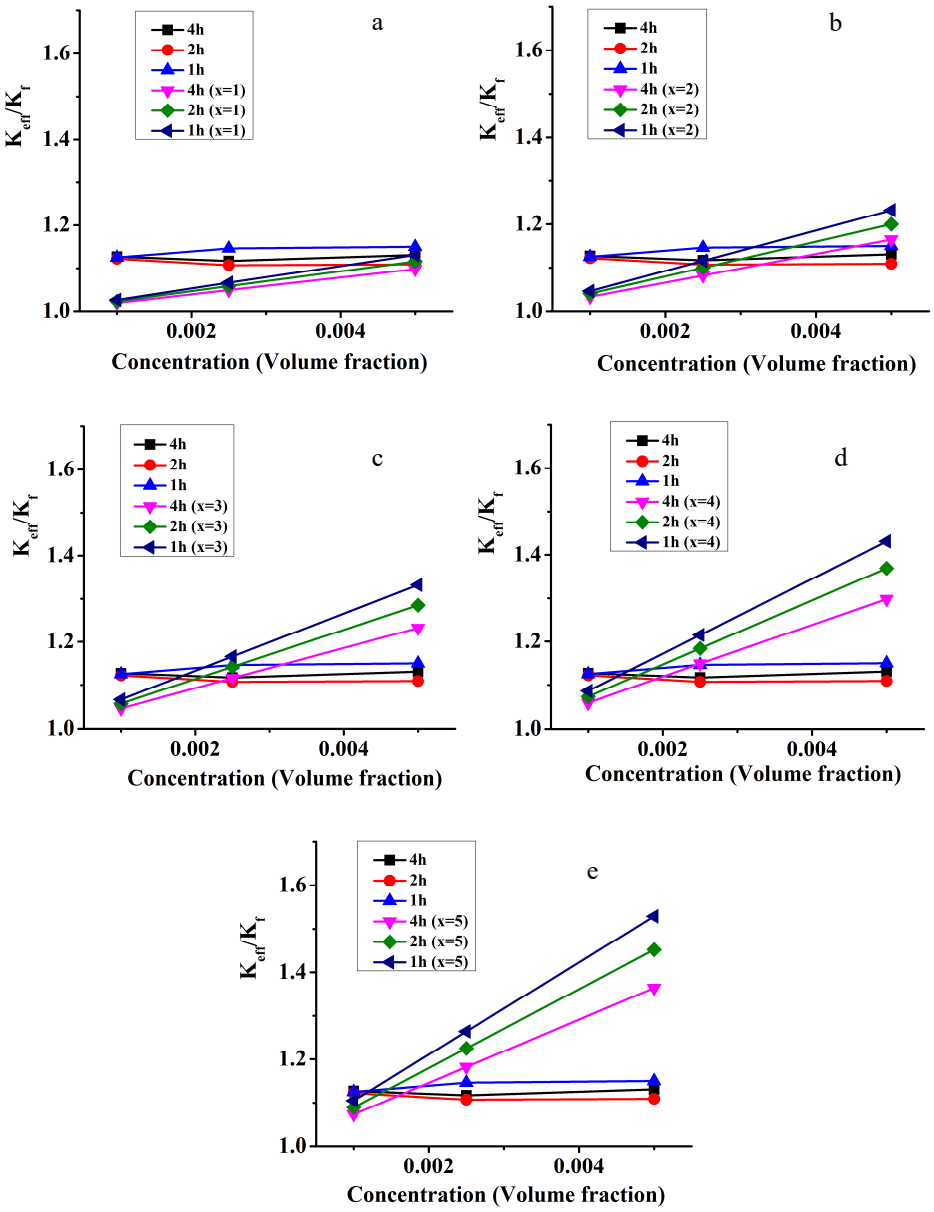

Fig. 3. Comparing the effective thermal conductivities of nanofluids containing different length CNTs with the corrected Hamilton-Crosser model a) $x = 1$, b) $x = 2$, c) $x = 3$, d) $x = 4$ and e) $x = 5$.

3.2 Suggested corrections on the Nan model

As mentioned above, the Nan model was presented to predict the thermal conductivity of nanofluids containing carbon nanotubes. In order to modify this model one should know that because the parameters K_p and K_f were fixed and known for CNTs, the only parameter which can be modified would be ϕ (the volume fraction of CNTs), so ϕ was replaced by $\varphi\left(x\frac{D}{L}\right)$, where x was the correction coefficient, D and L were the length and diameters of CNTs. It should be noted that the $\left(x\frac{L}{D}\right)$ term was also tried instead of $\left(x\frac{D}{L}\right)$, but no improvement was observed in the results. The suggested equation is:

$$k_{eff}=\frac{3+\varphi\left(x\frac{D}{L}\right)\left(\frac{k_c}{k_f}\right)}{3-2\varphi\left(x\frac{D}{L}\right)}k_f \tag{5}$$

Since $\frac{D}{L}$ is constant for CNTs in each sample, the different values of x were tested. The thermal conductivity enhancements were computed for x = 0.2, 0.3, 0.4, 0.5 and 1 and compared with the experimental data. The values of K_f, K_c, ϕ and D were as before. The related curves are shown in Fig 4.

Here again the accordance of the modified Nan's equation with the experimental data is excellent for x=0.3. This means that the introducing the CNTs parameters $\frac{D}{L}$ is necessary in prediction of the experimental data. It is to be noticed that Nan et al [34] had realized that their prediction was much higher than the experimental data but they had mentioned there was still much room for further enhancement of thermal conductivity of CNTs based nanofluids. It is also mentionable that in our modified model by increasing the ratio of $\frac{D}{L}$ or by decreasing the length of CNTs, the effective thermal conductivity increases which is in agreement with the experimental data [31].

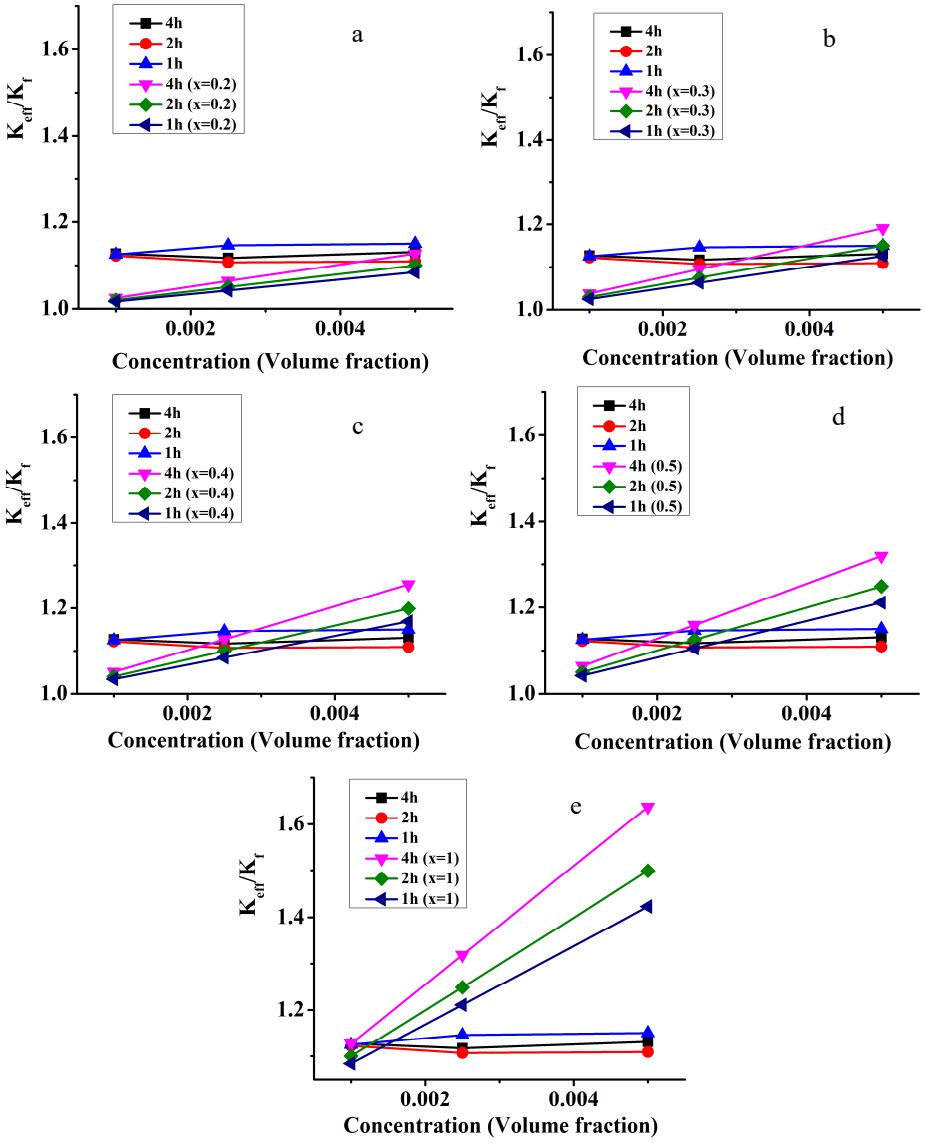

Fig. 4. Comparing the effective thermal conductivities of nanofluids containing different length CNTs with the corrected Nan model a) x = 0.2, b) x = 0.3, c) x = 0.4, d) x = 0.5 and e) x = 1.

4. Conclusion

The classical Hamilton- Crosser and Nan models were modified to predict the thermal conductivities of CNTs water based nanofluids. By replacing $n = 6 + x\frac{L}{D}$ instead of n=6 in Hamilton- Crosser, where L and D were length and diameter of CNTs it was found that the modified equation could predict the experimental data. Also by replacing $\varphi\left(x\frac{D}{L}\right)$ instead of ϕ (volume fraction) in the Nan model, the prediction of modified equation had very good accordance with the experimental data. Such corrections suggest that the shape of complicated nanoadditives has high impact on the nanofluid's properties. Also, one should notice that in order to achieve a comprehensive equation, it is necessary to look for an accurate equation to consider the temperature dependence of nanofluids.

Author Contributions

M. Farbod planned the scheme, initiated the project and suggested the experiments; A. Ahangarpour conducted the experiments and analyzed the empirical results. The manuscript was written through the contribution of all authors. All authors discussed the results, reviewed and approved the final version of the manuscript.

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Conflict of Interest

The authors declared no potential conflicts of interest with respect to the research, authorship and publication of this article.

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Data Availability Statements

The datasets generated and/or analyzed during the current study are available from the corresponding author on reasonable request.

Nomenclature

K_{eff}	Thermal conductivity of the suspension [W/m.K]	Φ	The volume concentration of the suspended particles
K_p	Thermal conductivity of the particles [W/m.K]	x	The correction coefficient
K_f	Thermal conductivity of the base fluid [W/m.K]	L	The lengths of carbon nanotubes [nm]
K_c	Thermal conductivity of carbon nanotubes [W/m.K]	D	The diameter of carbon nanotubes [nm]

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ORCID iD

Mansoor Farbod https://orcid.org/0000-0002-9774-705X
Ameneh Ahangarpour https://orcid.org/0000-0001-8239-7440

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