# THE EFFECTS OF UNCERTAINLY OF NANOLAYER PROPERIES ON THE HEAT TRANSFER THROUGH NANOFLUIDS

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### **ABSTRACT**

Nanofluids which are suspension of nanoparticles in conventional heat transfer fluids attracted researches on different heat transfer applications while they enhance thermal transport properties in comparison with conventional base fluids. Recently, utilizing these new fluids is growing increasingly, but the ambiguities of their thermal-physical properties cause to do not involve efficiently in the industrial design to date. The recognized important parameters effecting on properties of nanofluids include the volume fraction of the nanoparticles, temperature, nanoparticle size, nanolayer (thickness and properties), thermal conductivity of the base fluid, PH of the nanofluid, and the thermal conductivity of the nanoparticles. However, there is a distinct lack of enough investigation and reported research on the nanolayer thickness and its properties. In this work, the effect of uncertainty of the nanolayer thickness and properties on effective thermal conductivity and effective viscosity of nanofluids and as a result on heat transfer is discussed in details. The results show that the uncertainties can cause 20% error in calculation of Nusselts number and 24% for Reynolds number. Therefore, more investigation needs to be done into properties of nanolayer in order to identify them accurately.

### INTRODUCTION

Conventional heat transfer fluids like water, engine oil and ethylene glycol have limitations on heat transport. On the other hand rapid development of technology and generating enormous amount of heat in new heat transfer systems such as micro electromechanical machines and high efficiency heat exchangers require an enhanced heat transfer fluid. The main factor in the efficiency of thermal transport of a heat transfer fluid is the thermal conductivity. However, conventional heat

transfer fluids have poor thermal properties comparing with solids. A way to improve thermal conductivity of conventional fluids is dispersing solid particles in them.

The idea of dispersing micrometer- or millimetre-sized solid particles in fluids can be traced back to Maxwell theoretical work in 1873. Numerous theoretical and experimental studies have been done to increase thermal conductivity properties of fluids by dispersing millimetre or micrometer sized particles in fluids. Although adding these solid particles may improve thermal conductivity properties of conventional fluids, they could cause stability, rheological, sedimentation, clogging and pressure drop problems.

S. Choi [1] proposed using nanofluids, which are solid-liquid composite materials consisting of nanometer sized solid particles, fibers, rods or tubes suspended in different base fluids. Thermal-physical properties of fluid play a vital role in order to calculate heat transfer in thermal systems. Numerous studies have been done for calculating effective thermal conductivity and viscosity of nanofluids as key factors in order to introduce nanofluids into industrial design and applications. Volume fraction of the nanoparticles, temperature, nanoparticle size, nanolayer, thermal conductivity of the base fluid, PH of the nanofluid, and the thermal conductivity of the nanoparticles have been introduced by several authors as important parameters effecting on properties nanofluids[2,3].

In this paper, it is discussed the thermal-physical properties of nanolayer in thermal conductivity and viscosity of nanofluids as well as the influences of them on heat transfer.

### **NOMENCLATURE**

k	[W/ mK]	Thermal conductivity
q	$[W/m^2]$	Heat flox
$\bar{\rho}$	$[kg/m^3]$	Density
d	[m]	Diameter
S	[m]	Thickness of the surfactant adsorption monolayer
β	[1/K]	Coefficient of volume expansion
$T_s$	[°C]	Temperature of the surface
$C_p$	[kJ/kg.°C]	Specific heat at constant pressure
$V^{'}$	[m/s]	Fluid velocity
r	[m]	Radius
t	[m]	Thickness
$\varphi$	[-]	Particle volume fraction
μ	[kg/m s]	Fluid dynamic viscosity
$\mu_r$	[-]	Ratio of viscosity(nanofluid to base fluid)
H	[m]	inter-particle spacing
$N_A$	[mol-1]	Avogadro's constant
$V_{B}$	[m/s]	Brownian velocity
g	$[m^2/s]$	Gravitational acceleration
$\stackrel{\circ}{L}c$	[m]	characteristic length of the geometry
$T_{\infty}$	[°C]	temperature of the fluid
h	[W/ m2.°C]	convection heat transfer coefficient
$\varphi_h$	[-]	Hydrodynamic volume fraction
9	[-]	Thermal conductivity ratio
Ψ	[-]	particle sphericity
δ	[-]	ratio of the radius of the outer interface to the inner
		interface of the nanolayer
τ	[-]	Particle shape factor
$arphi_e$	[-]	Equivalent volume fraction
$M_{\rm w}$	[kg/kmol]	Molecular weight of liquid
Pr	[-]	Prandtl number
Subscript	S	
1		Nanolayer
f		Fluid
p		Particle
eff		Effective
pe		Equivalent nanoparticle

### SOLID-LIQUID INTERFACIAL LAYER PHYSICS

Many theoretical analysis and molecular simulation have been done to investigate properties of liquid in solid-liquid interfaces. Probing structure of these interfaces was difficult and the theoretical analyses were not verified experimentally. Henderson and van Swol [4] analysed the properties of a fluid in presence of a hard wall. In their research, theoretical analysis has been done and the results of molecular dynamic simulation of hard sphere-fluid bounded by a pair of planar wall have been used. They predicted density oscillation of molecules close to the solid liquid interface from the simulation results. They also discussed the presence of layering of fluid molecules in the interface of planar wall and fluid.

Thompson and Robbins [5] worked on epitaxial order of fluid near solids. They showed the degree of slip on solid is directly related to wall-fluid interaction. They indicated that at large interactions substantial epitaxial ordering happen and the first or two fluid layers become locked to the wall. In addition, as

wall-fluid interactions increased further, the density of the first layer increased to that of the solid.

Huisman et al. [6] investigated structure of solid-liquid interface with a synchrotron X-ray diffraction method. This method can be used because of deep penetration of x-rays in matter. The specular reflectivity was measured in Ga/Diamond (111)-2x1 interface. They reported exponentially decaying density oscillation in the Ga/Diamond interface. In the experiment, liquid gallium was super cooled so the layering could be consequence of local freezing.

In 1998, Huisman and van der Veen [7] introduced a model for the density profile in the solid-liquid interface. Fig.1 shows electron density distribution of the solid-liquid interface. The solid line in the graph is electron density distribution in Ga/Diamond (111)-2x1 interface which calculated from their model, and the dashed line curves are the solid and liquid distributions. As it's shown in the graph and the schematic, model for interface structure of Ga atoms closed to solid surface forming as solid like layer with high electron density. The Ga atoms structure close to diamond surface is Ga2 dimer, which is a stable solid phase of Ga at low temperature and ambient pressure.

Doerr et al. [8] studied thin liquid hexane films on silicon with specular and off-specular X-ray scattering. Their experimental results show one solid-liquid interfacial layer extended to 4nm from the interface. They concluded that the ordering of an interfacial layer in solid-liquid interface is independent of liquid film thickness.

Yu et al. [9] studied interfacial properties of thin liquid film of TEHOS (tetrakis(2-ethylhexoxy)silane) on silicon (111) substrate with X-ray reflectivity. They showed that three electron density oscillations near the interface with a period of  $\sim 1$ nm, which is consistent with molecular density.

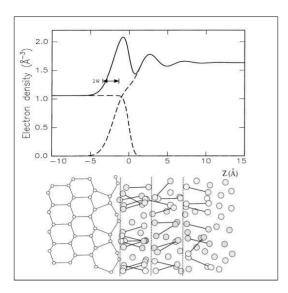
In 2000 Yu et al. [10] studied interface layering of TEHOS as a normal liquid at room temperature which was higher than freezing point. Samples of various thicknesses had been tested and density oscillations of a period of 1nm independent of film thicknesses reported.

Yu et al. [11] used synchrotron X-rays to study solid-liquid interface of three different liquids on silicon substrates. They studies ultrathin (45-90 Å) and thick (5000 Å) liquid films. They found that the liquid molecules are form 3-6 layers at the interface with plane close to molecular dimensions.

According to above mentioned studies there is no doubt in presence of liquid ordering in the solid-liquid interfaces. However, there are no certain models for predicting the interfacial layer properties.

### NANOFLUIDS EFFECTIVE THERMAL CONDUCTIVITY

After Choi [1] dispersed nano-sized particle in conventional fluids, many experimental studies have been done to determine effective thermal conductivity of nanofluids.



**Figure 1** Electron density distribution of the solid-liquid interface [7]

Maxwell [12] model which calculates effective thermal conductivity of fluids with suspended particles is base of some of the models for calculating effective thermal conductivity of nanofluids. This model takes into account particle and base fluid thermal conductivities and particle volume fraction:

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

There are several models have been developed without considering nanolayer effects which haven't been discussed in this study.

Independent experimental results show effective thermal conductivity of nanofluids is an order of magnitude larger than calculated amount from developed models. Many studies have been done to find factors effecting enhancement of effective thermal conductivity of nanofluids.

Eastman et al. [13] indicated dramatic enhancement in thermal conductivity of nanofluids is because of increasing surface area to volume ratio, and one can improve this enhancement by decreasing particle size. They also compared experimental results with theoretical predictions and recounted weaknesses of Hamilton-Crosser model regarding the particle size in predicting of effective thermal conductivity of nanofluids.

Keblinski et al. [14] explored possible answers for the anomalous effective thermal conductivity enhancement, Brownian motion of particles, interfacial nanolayer, ballistic conduction and particle clustering. In case of Brownian motion the analysis showed movement of nanoparticles could not transport significant amount of heat, and concluded there should be other reasons for such enhancement. They did analysis and simulate which demonstrated nanolayer play an important role in effective thermal conductivity of nanofluids,

because this liquid layer is more ordered than bulk liquid. The existence of this layer also increases effective volume fraction of nanoparticles. In their analysis, thermal conductivity of nanolayer assumed to be equal to  $k_p$  to estimate upper limit of nanolayer effect on enhancement of effective thermal conductivity. Therefore, indicated that if a double effective volume fraction is required for an amount of enhancement, there should be a nanolayer with thickness of 10nm, which is larger than experimental and simulation data for liquid layering on solid surfaces. Consequently, they concluded forming nanolayer couldn't have such big effect on effective thermal conductivity which was shown in experimental results. In addition, they concluded ballistic conductivity and particle clustering do not support such enhancement in effective thermal conductivity.

Xue [15] derived a model for predicting effective thermal conductivity of nanofluids based on Maxwell and average polarization theories. He considered particle and nanolayer as a "complex particle", and derived an equation for this complex particle. Weakness of his analysis is that he selected nanolayer thickness and thermal conductivity to fit experimental results, and he didn't explain any reason for these selections.

Yu et al. [16] renovated Maxwell model, and consider effects of interfacial layer on effective thermal conductivity of nanofluids. They used theory of formation of layered structure of liquid molecules on solid surfaces, and proposed this solid-like nanolayer around nanoparticle play a key role in enhancing thermal conductivity of nanofluids which acts as a thermal bridge. Nanoparticle and surrounded nanolayer assumed an equivalent nanoparticle, and defined increased volume concentration as below:

$$\varphi_e = \frac{4}{3}\pi(r_p + t_l)^3 n = \frac{4}{3}\pi r_p^3 n (1 + \frac{t_l}{r_p})^3 = \varphi(1 + \gamma)^3$$
 (2)

$$\gamma = \frac{t_l}{r_p} \tag{3}$$

where n is particle number per unit volume. Based on effective medium theory of Schwartz et al. (1995) they calculated equivalent nanoparticle thermal conductivity as below:

$$k_{pe} = \frac{\left[2(1-\vartheta) + (1+\gamma)^3(1+2\vartheta)\right]\vartheta}{-(1-\vartheta) + (1+\gamma)^3(1+2\vartheta)} k_p \tag{4}$$

$$\vartheta = \frac{k_l}{k_R} \tag{5}$$

And Maxwell model for thermal conductivity of nanofluid was modified to:

$$k_{eff} = \frac{k_{pe} + 2k_f + 2(k_{pe} - k_f)(1 + \gamma)^3 \varphi}{k_{pe} + 2k_f - (k_{pe} - k_f)(1 + \gamma)^3 \varphi} k_f \tag{6}$$

Therefore, they compared their model with classic Maxwell model for Cu-EG nanofluid, and concluded effects of

nanolayer is significant when nanoparticles are small below 5nm  $(r_n \sim t_l)$ , and their modified model results will reduce to original Maxwell equation when  $r_p \gg t_l$ . So the effects of the thickness and thermal conductivity of the nanolayer on thermal conductivity enhancement for a nanofluid were discussed. They concluded thermal conductivity enhancement is strongly dependent on the thickness of the nanolayer, but is almost invariant to the thermal conductivity of the nanolaver when  $k_1 > 10k_f$ , and nanolayer thickness is crucial to thermal conductivity enhancement. They assumed intermediate thermal conductivity between nanoparticle and base fluid thermal conductivities for nanolayer, and nanolayer conductivity is considered constant over nanolayer. Drawbacks to their model are nanolayer thickness and thermal conductivity amount, which has been adjusted for matching experimental data.

Yu & Choi [17] renovated Hamilton-Crosser model to include solid-liquid interface effects on effective thermal conductivity for non-spherical particles. Particles are assumed elliptical as well as particle and nanolayer as a complex particle. Equivalent thermal conductivity and volume fraction calculated for these elliptical particles. In order to analyse the model, they selected an unknown in their equation to fit a 2nm nanolayer thickness. Their investigation included k<sub>1</sub> equal to  $10k_f$ ,  $100k_f$  and  $k_p$ . Therefore, they founded in contrast with nanofluids with spherical particles, when k<sub>1</sub>=10k<sub>f</sub>, the effective thermal conductivity is less than the case of  $k_1=k_f$ . This is because of the nanolayer increases sphericity and reduces empirical shape factor which result to reduce effective thermal conductivity of nanofluids. However, if the conductivity is large enough, the effective conductivity will increase. They claim their model is in good agreement with experiment, but is unable to predict effective thermal conductivity in nonlinear behaviour.

Xue et al. [18] did a molecular dynamic simulation on a simple (mono-atomic) liquid to discover effects of ordered interfacial liquid layer on thermal transport. They proved this interfacial layer doesn't have any significant effect on thermal transport of nanofluid. Nevertheless, they stated that this result is for simple liquids, and maybe in more complex liquids effects of ordered layer are more significant.

Xue & Xu [19] assumed nanoparticle and nanolayer as a complex particle. They solved temperature distribution equation in the complex nanoparticle, and presented a model for effective thermal conductivity of this complex nanoparticle. They used Bruggmen's effective media theory for two phases composite to derive effective thermal conductivity of nanofluids:

$$(1 - \varphi_e) \frac{\kappa_{eff} - \kappa_f}{2\kappa_{eff} + \kappa_f} + \varphi_e \frac{(\kappa_{eff} - \kappa_l)(2\kappa_l + \kappa_p) - \alpha(\kappa_p - \kappa_l)(2\kappa_l + \kappa_{eff})}{(2\kappa_{eff} + \kappa_l)(2\kappa_l + \kappa_p) + 2\alpha(\kappa_p - \kappa_l)(\kappa_l - \kappa_{eff})} = 0$$
 (7)

$$\alpha = \left(\frac{r_p}{r_n + t_l}\right)^3 \tag{8}$$

They also compared results from above equation with experimental data, again drawback of their model is, they selected nanolayer thickness equal to 3nm, and nanolayer thermal conductivity 5W m<sup>-1</sup> K<sup>-1</sup> in A<sub>2</sub>O<sub>3</sub>-Water, 10 W m<sup>-1</sup> K<sup>-1</sup> in CuO-EG and 1.2 W m<sup>-1</sup> K<sup>-1</sup> in CuO -Water nanofluids to fit experimental results. In order to select nanolayer thickness equal to 3nm, they refer to Yu et al. [10] which concluded thickness of liquid layering on solids is several nano meters.

Xie et al. [20] discussed role of particle size, nanolayer thickness, volume fraction and thermal conductivity ratio of particle to base fluid on enhanced thermal conductivity. They derived a model by the approach of contribution of nanoparticle, base fluid and nanolayer in thermal conductivity of nanofluid as well as following the study of Lu [21] as

$$\frac{\kappa_{eff} - \kappa_f}{\kappa_f} = 3\theta \varphi_e + \frac{3\theta^2 \varphi_e^2}{1 - \theta \varphi_e} \tag{9}$$

$$\theta = \frac{\vartheta_{lf}[(1+\gamma)^3 - (\vartheta_{pl}/\vartheta_{fl})]}{(1+\gamma)^3 + 2\vartheta_{lf}\vartheta_{pl}}$$
(10)

Where 
$$\vartheta_{lf} = \frac{k_l - k_f}{k_l + 2k_f}$$
,  $\vartheta_{pl} = \frac{k_p - k_l}{k_p + 2k_l}$  and  $\vartheta_{fl} = \frac{k_f - k_l}{k_f + 2k_l}$ .

They assumed thermal conductivity of nanolayer in intermediate physical state between nanoparticle and base fluid with a leaner distribution by using Yu & Choi [20] as

$$k_l = \frac{k_f M^2}{(M-\gamma) \ln(1+M) + \gamma M} \tag{11}$$

$$M = \varepsilon_n(1+\gamma) - 1 \tag{12}$$

$$\varepsilon_{\rm p} = k_{\rm p} / k_{\rm f} \tag{13}$$

They also concluded that with decreasing nanoparticle size, effective thermal conductivity will increase, and the reason is contribution of nanolayer in small size nanoparticles. Specific surface areas (SSA) definition has been used to describe effects of nanolayer on effective thermal conductivity. Therefore, they indicated in micro sized particles SSA is so small and effects of nanolayer formed on the surface is negligible, whereas SSA for nanoparticles is large, so effects of nanolayer could not be neglected. Consequently, the impact of nanolayer would be more effective when the particle is small whilst the nanolayer is thick. In their analyses nanolayer thickness has been selected as 2nm, which is a drawback of their work.

Yajie et al. [22] built up a model for effective thermal conductivity of nanofluid; they assumed linear intermediate thermal conductivity for nanolayer, nanoparticle and nanolayer as a complex nanoparticle and four kinds of heat transfer in nanofluid: base fluid, nanoparticle, nanolayer and micro convection. They used Lu and Song (1996) model for heat conduction in suspension fluid and finally derived an equation for effective thermal conductivity as

$$k_{eff} = k_f \left[ 1 + F(Pe) + 3\theta \varphi_e + \frac{3\theta^2 \varphi_e^2}{1 - \theta \varphi_e} \right]$$
 (14)

$$\theta = \frac{\vartheta_{lf}[(1+\gamma)^3 - (\vartheta_{pl}/\vartheta_{fl})]}{(1+\gamma)^3 + 2\vartheta_{lf}\vartheta_{nl}}$$
(15)

They assumed nanolayer thickness 2nm based on the theory of magnitude of liquid layering on solids as nano-meters.

Leong et al. [23] developed a model for effective thermal conductivity of nanofluids. Proposed model takes into account volume fraction, thickness, thermal conductivity of the interfacial layer and particle as

$$k_{eff} = \left\{ k_f \frac{\varphi \epsilon(k_p - \epsilon k_f) [2\delta_1^3 - \delta^3 + 1] + (k_p + 2\epsilon k_f) \delta_1^3 [\varphi \delta^3 (\epsilon - 1) + 1]}{\delta_1^3 (k_p + 2\epsilon k_f) - (k_p - \epsilon k_f) \varphi [\delta_1^3 + \delta^3 - 1]} \right\}$$
(16)

In comparing the model with experimental data, they used 1nm for nanolayer thickness and  $k_i$ = (2~3)  $k_f$ .

Feng et al. [24] proposed a model for effective thermal conductivity of nanofluids by considering nanolayer and nanoparticle aggregation effects. They used Yu and Choi's model (Eq. 14) for effective thermal conductivity of non-aggregated particles, and proposed a model for effective thermal conductivity of clusters,  $k_{\rm agg}$  as

$$k_{agg} = \left[ \left( 1 - \frac{3}{2} \varphi_e \right) k_f + \frac{3k_f}{\omega} \varphi_e \left[ \frac{1}{\omega} ln \frac{(r_p + t_l)}{(r_p + t_l)(1 - \omega)} - 1 \right] \right] \quad (17)$$

$$\omega = 1 - \frac{k_f}{k_{pe}} \tag{18}$$

$$k_{\text{eff}} = (1 - \varphi_{\text{e}})k_{\text{non-agg}} + \varphi_{\text{e}}k_{\text{agg}}$$
 (19)

Table 1 Most common expression for thermal conductivity of nanofluids

Model	Remarks	Researcher/year
$k_{Maxwell} = \frac{k_p + 2k_f + 2(k_p - k_f)\emptyset}{k_p + 2k_f - (k_p - k_f)\emptyset} k_f$	Only Particle and fluid thermal conductivity and volume fraction have been considered.	Maxwell (1873)
$k_{eff} = \frac{k_{pe} + 2k_f + 2(k_{pe} - k_f)(1 + \gamma)^3 \varphi}{k_{pe} + 2k_f - (k_{pe} - k_f)(1 + \gamma)^3 \varphi} k_f$	Nanolayer thickness and its thermal conductivity have been considered.	Yu et al. (2003)
$(1 - \varphi_e) \frac{\kappa_{eff} - \kappa_f}{2\kappa_{eff} + \kappa_f} + \varphi_e \frac{(\kappa_{eff} - \kappa_l)(2\kappa_l + \kappa_p) - \alpha(\kappa_p - \kappa_l)(2\kappa_l + \kappa_{eff})}{(2\kappa_{eff} + \kappa_l)(2\kappa_l + \kappa_p) + 2\alpha(\kappa_p - \kappa_l)(\kappa_l - \kappa_{eff})} = 0$	Nanolayer thickness and its thermal conductivity have been considered.	Xue & Xu (2005)
$\frac{\kappa_{eff} - \kappa_f}{\kappa_f} = 3\theta \varphi_e + \frac{3\theta^2 \varphi_e^2}{1 - \theta \varphi_e}$ $\theta = \frac{\vartheta_{lf} [(1 + \gamma)^3 - (\vartheta_{pl} / \vartheta_{fl})]}{(1 + \gamma)^3 + 2\vartheta_{lf} \vartheta_{pl}}$	Role of particle size, nanolayer thickness, volume fraction and thermal conductivity ratio of particle to base fluid on enhanced thermal conductivity have been considered.	Xie et al. (2005)
$k_{eff} = k_f \left[ 1 + F(Pe) + 3\theta \varphi_e + \frac{3\theta^2 \varphi_e^2}{1 - \theta \varphi_e} \right]$	Nanolayer thickness and its thermal conductivity have been considered.	Yajie et al (2005)
$ k_{eff} = \begin{cases} k_{eff} = \\ k_f \frac{\varphi \epsilon(k_p - \epsilon k_f)[2\delta_1^3 - \delta^3 + 1] + (k_p + 2\epsilon k_f)\delta_1^3[\varphi \delta^3(\epsilon - 1) + 1]}{\delta_1^3(k_p + 2\epsilon k_f) - (k_p - \epsilon k_f)\varphi[\delta_1^3 + \delta^3 - 1]} \end{cases} $	This model takes into account volume fraction, thickness and thermal conductivity of the interfacial layer.	Leong et al. (2006)
$\begin{aligned} k_{eff} &= (1 - \varphi_e) k_{non-agg} + \varphi_e k_{agg} \\ k_{agg} &= \left[ \left( 1 - \frac{3}{2} \varphi_e \right) k_f + \frac{3k_f}{\omega} \varphi_e \left[ \frac{1}{\omega} ln \frac{(r_p + t_l)}{(r_p + t_l)(1 - \omega)} - 1 \right] \right] \end{aligned}$	Effects of nanolayer and nanoparticle aggregation have been considered.	Feng et al. (2007)
$\frac{\left(1 - \frac{\varphi}{\alpha}\right) \frac{k_{eff} - k_f}{2k_{eff} + k_f}}{\frac{\varphi}{\alpha} \frac{(k_{eff} - k_l)(2k_l + k_p) - \alpha(k_p - k_l)(2k_l + k_{eff})}{(2k_e + k_l)(2k_l + k_p) + 2\alpha(k_p - k_l)(k_l - k_{eff})}} = 0$	Effects of nanolayer thickness and its thermal conductivity have been considered.	Ghosh & Mukherjee (2013)

In order to compare the model with experimental data, they selected nanolayer thickness and thermal conductivity of 2nm

and 3kf respectively. The reason behind this choice for nanolayer thickness was using Hashimito et al. [25] model for

electron density profile at the interface and Li et al. [26] model which used  $t_l = \sqrt{2\pi}\sigma$  for determining interfacial layer thickness, where  $\sigma$  has value between 0.4 to 0.6nm, so interfacial nanolayer thickness was expected to be 1 and 2nm. Molecular dynamic simulation also performed by Xue et al. [18] which confirmed interfacial layer thickness is of the order of magnitude of a few atomic distances. Reason for using kl=3kf was several authors [20, 24], which considered kl equal to 2 or 3 kf. So they used kl=3kf in their calculations.

Ghosh & Mukherjee [27] considered effects of nanolayer on effective thermal conductivity of nanofluids and developed an expression for effective thermal conductivity of nanofluids. They used Langmuir formula of monolayer adsorption of molecule which offered by Wang et al. [28] as

$$t_l = \frac{1}{\sqrt{3}} \left[ \frac{4M_W}{\rho_f N_A} \right]^{1/3} \tag{20}$$

By assuming linear intermediate thermal conductivity for nanolayer between particle and fluid thermal conductivities, and solving heat flow rate in spherical particle with interfacial layer, they proposed an for thermal conductivity of nanolayer as

$$k_l = \frac{1}{r_p(r_p + t_l) \left[ c \ln\left(1 + \frac{t_l}{r_p}\right) + \frac{Dt_l}{r_p(r_p + t_l)} - \frac{E}{\lambda} \ln\left(1 - \frac{\lambda t_l}{k_p}\right) \right]}$$
(21)

$$\lambda = \frac{k_p - k_f}{t_l} \tag{22}$$

Where C, D, and E are functions of  $\lambda$ ,  $k_p$  and  $r_p$ . They used Xue and Xu [19] expression for thermal conductivity of equivalent particle and Bruggeman's effective media theory, and then offered their model as

$$\left(1 - \frac{\varphi}{\alpha}\right) \frac{k_{eff} - k_f}{2k_{eff} + k_f} + \frac{\varphi}{\alpha} \frac{(k_{eff} - k_l)(2k_l + k_p) - \alpha(k_p - k_l)(2k_l + k_{eff})}{(2k_l + k_p) + 2\alpha(k_p - k_l)(k_l - k_{eff})} = 0 \tag{23}$$

$$\alpha = \left(\frac{r_p}{r_p + t_l}\right)^3 \tag{24}$$

Table 1 summarize the most common expression for thermal conductivity of nanofluids. They are function of volume fraction, thermal conductivity of base fluid and particle. However, some of them include the effects of nanolayer.

### NANOFLUID EFFECTIVE VISCOSITY

In heat transfer flow applications, viscosity is as critical as thermal conductivity, because it's expected from nanoparticles to increase nanofluid thermal conductivity without increasing pressure drop which is dependent strongly to nanofluid viscosity. Therefore, recognizing factors effecting nanofluid viscosity and developing a model for predicting viscosity of nanofluid is an important issue in nanofluid applications. Consequently, in order to predict the flow and heat transfer rates in the convective nanofluids, it is necessary to consider the viscosity and correlation between viscosity and temperature.

Investigation of the rheological behaviour of fluid with dispersions could be traced back to Einstein's analysis of infinitely dilute suspensions of hard spheres in 1906 [29]. The most of the existing models derived from Einstein work.

However, the model does not consider particles interactions and is valid for low particle volume concentration about 2%.

$$\mu_r = \frac{\mu_{nf}}{\mu_f} = (1 + 2.5\varphi) \tag{25}$$

In 1952 Brikman extended Einstein's formula to volume concentration up to 4% [29] as

$$\frac{\mu_{nf}}{\mu_f} = \frac{1}{(1-\varphi)^{2.5}} \tag{26}$$

Frankei and Acrivos [30] proposed the following equation

$$\frac{\mu_{nf}}{\mu_f} = \frac{9}{8} \left[ \frac{(\varphi/\varphi_m)^{1/3}}{1 - (\varphi/\varphi_m)^{1/3}} \right] \tag{27}$$

where  $\phi_m$  is maximum attainable volume fraction that must be determined experimentally.

Lundgren (1972) proposed a model under the form of Taylor series [29] as

$$\frac{\mu_{nf}}{\mu_f} = \left[1 + 2.5\varphi + \frac{25}{4}\varphi^2 + O(\varphi^3)\right]$$
 (28)

Batchelor [31] include effects of Brownian motion on viscosity of dispersion as

$$\frac{\mu_{nf}}{\mu_f} = (1 + 2.5\varphi + 6.5\varphi^2) \tag{29}$$

Graham [32] considered inter-particle spacing on viscosity of dispersion, and then developed his model as

$$\frac{\mu_{nf}}{\mu_f} = 1 + 2.5\varphi + 4.5 \left[ 1 / \left( \frac{H}{r_p} \right) \left( 2 + \frac{H}{r_p} \right) \left( 1 + \frac{H}{r_p} \right)^2 \right]$$
(30)

where H is inter-particle spacing.

In 2006 Guo et al. considered the effect of particle diameter on viscosity, and developed Batchelor's model for low concentrations [33]:

$$\frac{\mu_{nf}}{\mu_f} = (1 + 2.5\varphi + 6.5\varphi^2) (1 + 350\varphi/r_p)$$
 (31)

Avsec and Oblak [34] offered shear viscosity in their study as

$$\mu_r = \frac{\mu_{nf}}{\mu_f} = 1 + (2.5\varphi_e) + (2.5\varphi_e)^2 + (2.5\varphi_e)^3 + (2.5\varphi_e)^4 + \cdots$$
(32)

They used Ward equation, and then included that nanolayer affected by changing volume concentration with effective volume concentration, which was named renewed Ward equation.

Nguyen et al. [29] discussed effects of temperature, particle size and concentration on nanofluid viscosity. They measured Al<sub>2</sub>O<sub>3</sub>-Water viscosity for two different particle sizes, 36 and 47nm, in temperature range of room condition to nearly 75°C. They proposed two correlations for determination of viscosity

ratio in Al<sub>2</sub>O<sub>3</sub>-Water, with Particle sizes 36 and 47nm respectively as

$$\mu_r = \frac{\mu_{nf}}{\mu_f} = (1 + 0.025\varphi + 0.015\varphi^2) \tag{33}$$

$$\mu_r = \frac{\mu_{nf}}{\mu_f} = 0.904 \ e^{0.1483\varphi} \tag{34}$$

Lee et al. [35] performed some experimental analyses for understanding the behaviour of  $Al_2O_3$ -Water nanofluid in very low volume concentrations (0.01-0.3 vol. %). In the experiments, thermal conductivity and viscosity of this nanofluid were measured. The experimental data has been compared with available models and previous experimental results. Oscillation type viscometer was used to measure viscosity as a function of temperature and volume concentration, and the experimental results showed nonlinear behaviour of nanofluid against volume concentration. They stated this behaviour implies that there are some particle—particle interactions which invalidate Einstein's model.

Murshed et al. [36] studied thermal conductivity and viscosity of nanofluids theoretically and experimentally. They stated the classical models by that time could not predict enhanced thermal conductivity because effects of particle size, distribution and interfacial layer were not included. So they assumed nanofluids include three component particles, liquid and interfacial layer to include effects of the interfacial layer on thermal conductivity and viscosity. In order to calculate the thickness of nanolayer Hashimoto et al. model employed. They used nanolayer thickness of 1nm and 2nm for spherical and carbon nanotubes respectively. Comparisons between their experimental results and available models showed the models under predict the shear viscosity. They concluded that the clusters and surface adsorption could be the reason of this difference, and these two factors can increase hydraulic diameter of particles and result in higher viscosity. They also stated the nature of the particle surface, ionic strength of the base fluid, surfactants, pH values, inter-particle potentials such as repulsive (electric double layer force) and attractive (van der Waals force) forces may play significant role to alter the viscosity of nanofluids

Masoumi et al. [37] introduced a model for calculating nanofluids effective viscosity in which Brownian motion is considered.

$$\mu_{eff} = \mu_f + \mu_{app} \tag{35}$$

 $\mu_{app}$  is apparent viscosity and shows effects of nanoparticles on the viscosity of nanofluids,

$$\mu_{eff} = \mu_f + \frac{\rho_p V_B d_p^2}{72CH} \tag{36}$$

Where C is correction factor and was determined from experimental data associated with Al<sub>2</sub>O<sub>3</sub>-Water nanofluid. Limitation of using the equation is  $\varphi < -b/a$ . However, the effect of solid-liquid interface was not considered in their model.

Hosseini et al. [38] presented a model for predicting viscosity of nanofluids. In their empirical model, nanofluid viscosity is a function of viscosity of the base liquid, particle volume fraction, particle size, properties of the surfactant layer, and temperature as

$$\frac{\mu_{nf}}{\mu_f} = exp\left[m + \alpha\left(\frac{T}{T_0}\right) + \beta(\varphi_h) + \gamma\left(\frac{d_p}{1+s}\right)\right]$$
(37)

where  $\varphi_h$  is hydrodynamic volume fraction.

$$\varphi_h = \varphi \left[ \frac{d_p + 2s}{d_p} \right]^3 \tag{38}$$

And m is a factor that depends on the properties of the system (like the solid nanoparticles, the base fluid, and their interactions), while  $\alpha$ ,  $\beta$ , and  $\gamma$  are empirical constants which determined from experimental data.

Yang et al. [33] studied effects of surfactant monolayer and interfacial nanolayer on nanofluid viscosity. They calculated equivalent volume concentration of particles by adding these two layer thicknesses to nanoparticle radios, put new volume concentration in Einstein model.

$$\varphi_{new} = \varphi \left[ \frac{r_p + t_l + s}{r_p} \right]^3 \tag{39}$$

$$S = \frac{1}{\sqrt{3}} \left[ \frac{4M_W}{\rho_f N_A} \right]^{1/3} \tag{40}$$

For nanolayer thickness they used Hashimoto et al. model and put nanolayer thickness 1nm.

$$\mu_r = \frac{\mu_{nf}}{\mu_f} = (1 + 2.5 \left[ \frac{r_p + t_l + s}{r_p} \right]^3 \varphi)$$
 (41)

Table 2 summarize the most common expression for viscosity of nanofluids.

## EFFECTS OF NANOLAYER PROPERTIES ON NANOFLUID'S PROPERTIES AND HEAT TRANSFER

In this section the effect of nanolayer thickness as well as thermal conductivity of nanolayer on effective thermal conductivity and viscosity of nanofluids, and consequently on heat transfer is discussed.

Thermal conductivity ratio of Yu & Choi model [20] is calculated for  $Al_2O_3$ -Water nanofluid, different nanolayer thickness (1 and 2nm), volume fraction range of 1 – 6%, nanoparticle size of 10nm, and different nanolayer thermal conductivities. As it is shown in Fig.2a for 6% volume fraction, effective thermal conductivity enhancement changes from 23% to 33% in different nanolayer thicknesses.

Same analyses have been done on model presented by Xue & Xu [19] for  $Al_2O_3$ -Water nanofluid; they took nanolayer thickness and thermal conductivity of nanolayer 3nm and 5 Wm<sup>-1</sup> K<sup>-1</sup> respectively. As it is shown in Fig. 2b, the thermal conductivity enhanced from 28% to 54% in 6% volume fraction of nanoparticles.

Another model which has been chosen for performing these analyses is Xie et al. [20] model. They assumed thermal conductivity of nanolayer in intermediate physical state between nanoparticle and base fluid with a linear distribution. In terms of different nanolayer thicknesses which has been

shown in Fig. 3a, in the case of 6% volume fraction of  $Al_2O_3$  in water, effective thermal conductivity enhancement increase from 20% to 44%.

Table 2 Most common expression for viscosity of nanofluids

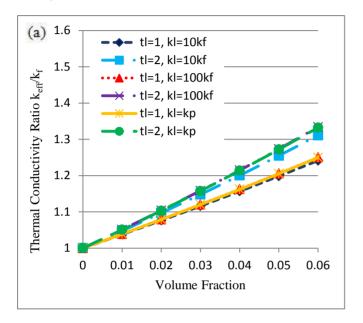
Model	Remarks	Researcher/year
$\frac{\mu_{nf}}{\mu_f} = (1 + 2.5\varphi)$	Valid for low volume concentration (less than 2%)	Einstein(1906)
$\frac{\mu_{nf}}{\mu_f} = \frac{1}{(1-\varphi)^{2.5}}$	Valid for volume concentration up to 4%	Brikman (1952)
$\frac{\mu_{nf}}{\mu_f} = \frac{9}{8} \left[ \frac{(\varphi/\varphi_m)^{1/3}}{1 - (\varphi/\varphi_m)^{1/3}} \right]$	Limited when volume concentration approaches $\phi_m$	Frankei and Acrivos (1967)
$\frac{\mu_{nf}}{\mu_f} = \left[1 + 2.5\varphi + \frac{25}{4}\varphi^2 + O(\varphi^3)\right]$	Proposed based on Taylor series. Applicable for spherical particle in dilute systems.	Lundgren (1972)
$\frac{\mu_{nf}}{\mu_f} = (1 + 2.5\varphi + 6.5\varphi^2)$	Effects of Brownian motion on viscosity has been considered	Bachelor(1977)
$\frac{\mu_{nf}}{\mu_f} = 1 + 2.5\varphi + 4.5 \left[ 1 / \left( \frac{H}{r_p} \right) \left( 2 + \frac{H}{r_p} \right) \left( 1 + \frac{H}{r_p} \right)^2 \right]$	Inter-particle spacing has been considered	Graham(1981)
$\frac{\mu_{nf}}{\mu_f} = (1 + 2.5\varphi + 6.5\varphi^2)(1 + 350\varphi/r_p)$	Effect of particle diameter on viscosity has been considered	Guo et al. (2006)
$\frac{\mu_{nf}}{\mu_f} = 1 + (2.5\varphi_e) + (2.5\varphi_e)^2 + (2.5\varphi_e)^3 + (2.5\varphi_e)^4 + \cdots$	Nanolayer effects entered in the viscosity equation by Modifying Ward equation and replacing volume concentration with equivalent volume fraction.	Avsec and Oblak (2007)
$\frac{\mu_{nf}}{\mu_f} = (1 + 0.025\varphi + 0.015\varphi^2), \ r_p = 36$	Empirical models for Al <sub>2</sub> O <sub>3</sub> -Water nanofluid.	Nguyen et al. (2008)
$\frac{\mu_{nf}}{\mu_f} = 0.904 \ e^{0.1483\varphi} \ , \ r_p = 47$		
$\mu_{eff} = \mu_f + \frac{\rho_p V_B d_p^2}{72CH}$	Effects of nanoparticles properties on the viscosity have been considered.	Masoumi (2009)
$\frac{\mu_{nf}}{\mu_f} = exp\left[m + \alpha\left(\frac{T}{T_0}\right) + \beta(\varphi_h) + \gamma\left(\frac{d_p}{1+s}\right)\right]$	The empirical model is a function of viscosity of the base liquid, particle volume fraction, particle size, properties of the surfactant layer, and temperature.	Hosseini et al. (2010)
$\mu_r = \frac{\mu_{nf}}{\mu_f} = (1 + 2.5 \left[ \frac{r_p + t_l + s}{r_p} \right]^3 \varphi)$	Effects of surfactant monolayer and interfacial nanolayer on nanofluid viscosity have been considered.	Yang et al. (2012)

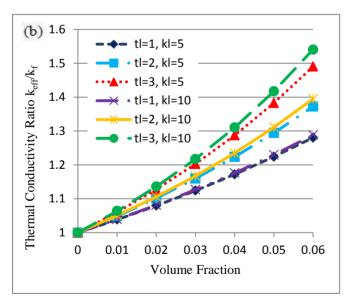
The result for Feng et al. [24] model is shown in Fig. 3b which thermal conductivity enhancement have 7% increase.

As it is indicated in Fig. 4a, viscosity of  $Al_2O_3$ -Water nanofluid is calculated for 10nm spherical size particles, volume fraction range 1 to 6 % by using Avsec and Oblak [34]

model. In 6% volume fraction, viscosity enhances 21% for nanolayer thickness equal to 0.5nm whereas it's 48% when nanolayer thickness is 3nm.

From the results of these analysis which has been done on several effective thermal conductivity models, it can conclude that the nanolayer properties like thickness and thermal conductivity impact on calculated effective thermal conductivity of nanofluids so more studies is necessary to develop models for this factors.



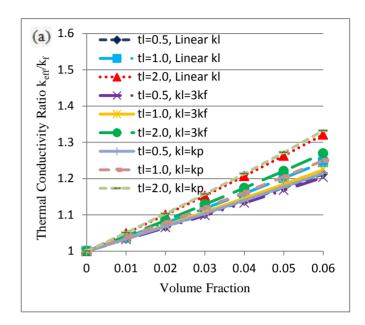


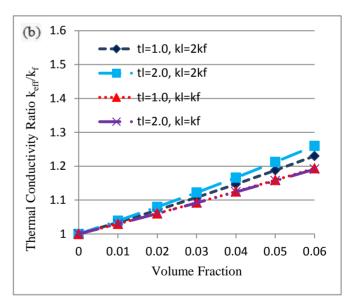
**Figure 2** (a) Thermal conductivity ratio of  $Al_2O_3$ -Water nanofluid, according to Yu & Choi [20] model ( $k_f = 0.604$ ,  $k_p = 46$ ,  $r_p$ =10). (b) Thermal conductivity ratio of  $Al_2O_3$ -Water nanofluid, according to Xue & Xu [19] model ( $k_f = 0.604$ ,  $k_p = 46$ ,  $r_p$ =10)

The mathematic analysis was done for Al<sub>2</sub>O<sub>3</sub>-EG nanofluid, which indicates nanolayer thickness is 19% of the

In terms of nanolayer thickness in most of studies a value has been selected or assumed by authors. Tillman & Hill [39] tried to derive an equation for determination of nanolayer thickness.

A mathematical procedure was developed to determine nanolayer thickness for any thermal conductivity profile. Thermal conductivity in nanolayer was assumed to be known, then by solving thermal conductivity equation in solid-liquid interface, nanolayer thickness derived.

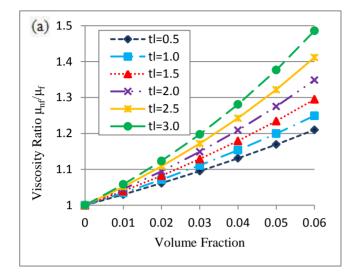


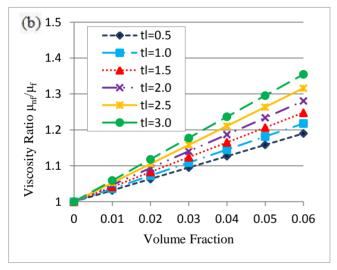


**Figure 3** (a) Thermal conductivity ratio of  $Al_2O_3$ -Water nanofluid, according to Xie et al. [20] model ( $k_f = 0.604$ ,  $k_p = 46$ ,  $r_p = 10$ ). (b) Thermal conductivity ratio of  $Al_2O_3$ -Water nanofluid, according to Feng et al. [24] model ( $k_f = 0.604$ ,  $k_p = 46$ ,  $r_p = 10$ ).

nanoparticle radius. Results for CuO-H<sub>2</sub>O with same analysis showed ratio of the radius of the outer interface to the inner

interface of the nanolayer  $,\delta$  ,equal to 1.22. They examed their model for  $k_{\rm f} < 1 W \ m^{\text{-}1} \ K^{\text{-}1},$  and various  $k_{\rm p},$  then concluded nanolayer thickness for all nanofluids are in the range of 19% to 22% of nanoparticle radius. Effects of bonding between nanoparticle and base fluid were not considered in their model. There is also no evidence for accuracy of the equation which was used for thermal conductivity of nanolayer.





**Figure 4** (a) Viscosity ratio of Al<sub>2</sub>O<sub>3</sub>-Water nanofluid, according to Avsec and Oblak [34] model. (b) Viscosity ratio of Al<sub>2</sub>O<sub>3</sub>-Water nanofluid, according to Yang et al. [33] model

In the case of forced convection, Nusselt and Reynolds numbers and in natural convection, Grashof or Reyleigh numbers are dimensionless numbers which have been using for designing engineering systems. They will be influenced by effective thermal conductivity and viscosity of the heat transfer fluids.

$$Nu = \frac{hL_c}{k_{eff}} \tag{42}$$

$$Re = \frac{\rho_{nf}VL_c}{\mu_{nf}} \tag{43}$$

$$Gr_{L} = \frac{g\beta \rho_{nf}^{2} (T_{s} - T_{\infty}) L_{c}^{3}}{\mu_{nf}^{2}}$$
(44)

$$Ra_L = Gr_L Pr (45)$$

Ranges of effective thermal conductivity enhancement and viscosity ratio for nanofluids are 1.20–1.44 and 1.19–1.48 respectively. By using these two extremes for calculating each dimensionless number, they will vary in a wide range.

There will be 20% difference in calculated Nusselt number, 24% in Reynolds number, 54% in Grashof number and 49% in Rayleigh number when two extremes of  $k_{eff}$  and  $\mu_{nf}$  are using in their formulas.

### CONCLUSION

It is clear understanding that the nanolayer is one of the key factors which must be considered for evaluation of the effective thermal conductivity and effective viscosity for nanofluids. Unfortunately, most of the available models for effective thermal conductivity and effective viscosity for nanofluids do not include the nanolayer. On the other hand, the ones which count the nanolayer are not accurate for predictions of unknown values. Therefore, these uncertainties can produce at least 20% difference in calculation of Nusselts number as well as 24% in Reynolds number, 54% in Grashof number and 49% in Rayleigh number. Consequently, the authors can conclude that the existing models for thermal conductivity and viscosity of nanofluids cause error in design of thermal systems by using nanofluids; therefore, more investigation is necessary in this area.

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